

Cloud fraction responses to aerosols in marine low clouds using explainable machine learning: satellite observations and global climate modelling

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Abstract

Aerosol–cloud interactions (ACI) remain a primary uncertainty in the climate system. Marine low clouds (MLCs) are particularly important in this context because they cover large parts of the ocean surface and exert a strong net cooling effect by reflecting incoming solar radiation. Aerosols can modify these clouds by changing cloud droplet number concentration (N_d), cloud optical properties, precipitation formation, liquid water path (LWP), and cloud fraction (CF). Among them, aerosol-induced changes in CF, the CF adjustment, are potentially among the largest but least constrained components of the effective radiative forcing due to ACI. Estimating and understanding this adjustment remains challenging because aerosol, cloud, and meteorological conditions strongly covary in large-scale observations, while in models the simulated CF response may further depend on configuration choices, including cloud-cover parameterizations and prescribed constraints on N_d .

This thesis addresses these challenges by establishing an explainable machine-learning (ML) framework across near-global oceans. This framework combines region-specific eXtreme Gradient Boosting (XGBoost) models with the SHapley Additive exPlanations (SHAP) approach, using N_d as a proxy for cloud-active aerosols. Within each $5^\circ \times 5^\circ$ geographical window aggregated from the original data grids, CF is predicted by one XGBoost model from N_d alongside a set of thermodynamic and dynamical meteorological predictors, and SHAP values are used to quantify the CF responses while accounting for meteorological covariability. This thesis combines two complementary perspectives: an observation-based analysis using Moderate Resolution Imaging Spectroradiometer (MODIS) satellite retrievals and ERA5 reanalysis data, and a model-based analysis using output from the ICOSahedral Nonhydrostatic (ICON) atmosphere model coupled with the Hamburg Aerosol Module (HAM) (ICON-HAM). The overall aim is to quantify and physically interpret the CF response to aerosols in both MODIS and ICON-HAM, examining the role of meteorological conditions and model configuration choices, respectively.

The observation-based analysis reveals a near-globally positive $\ln N_d$ -CF sensitivity, strongest in stratocumulus-to-cumulus transition regions off the western continental coasts and in the Southern Hemispheric midlatitudes, and weakest in the tropics. Beyond direct sensitivities, this framework allows for the explicit quantification of the meteorological influence on the $\ln N_d$ -CF sensitivity using an interaction index (IAI) derived from SHAP interaction values. Thermodynamic variables, especially estimated inversion strength (EIS) and sea surface temperature (SST), play a larger role than dynamical variables both in controlling CF itself and in modulating the N_d -CF relationship. The IAI patterns suggest that higher EIS amplifies the $\ln N_d$ -CF sensitivity in the midlatitudes, while higher SST does so in stratocumulus-to-cumulus transition regions.

The model-based analysis applies the same explainable ML framework to six ICON-HAM simulations, combining two cloud-cover schemes—CC-RH, which depends only on relative humidity (RH), and CC-RH-LWC, which additionally depends on liquid water content (LWC)—with three prescribed lower bounds for N_d ($N_{d,\min} = 10, 40$ and 100 cm^{-3}). Compared with the MODIS-based results, ICON-HAM shows a more nonlinear and saturating $\ln N_d$ -CF relationship, and thus the SHAP-based sensitivity is extended using piecewise linear regression. The diagnosed CF sensitivity is found to be jointly shaped by the cloud-cover scheme and $N_{d,\min}$ via a “headroom” effect: higher $N_{d,\min}$ suppresses autoconversion and increases LWP; in CC-RH-LWC, this increases mean-state CF because CF depends explicitly on cloud water content. The elevated baseline leaves less room for further aerosol-induced CF increases, limiting the contribution of $\ln N_d$ to the CF prediction and weakening the diagnosed $\ln N_d$ -CF sensitivity. Thus, the expectation that CC-RH-LWC exhibits stronger sensitivities than CC-RH holds only at low $N_{d,\min}$ and reverses at higher values.

Overall, this thesis shows that the CF response to aerosols can be interpreted more meaningfully when meteorological covariability is taken into account and when the background cloud state is considered. In observations, this state dependence appears as regional meteorological modulation and spatial variations across cloud regimes; in climate models, it is further shaped by configuration choices that change the simulated mean-state CF. These findings advance the observational constraint on the CF response, identify different behaviours of the $\ln N_d$ -CF relationship in MODIS and ICON-HAM, and demonstrate how specific model configuration aspects affect the simulated response. Together, they suggest that meaningful model-observation comparisons require careful attention to the background state in which each estimate is derived.

Zusammenfassung¹

Aerosol-Wolken-Wechselwirkungen (ACI) bleiben eine zentrale Unsicherheit im Klimasystem. Marine tiefe Wolken (MLCs) sind in diesem Zusammenhang besonders wichtig, da sie große Teile der Ozeanoberfläche bedecken und durch die Reflexion einfallender Sonnenstrahlung einen starken kühlenden Nettoeffekt ausüben. Aerosole können diese Wolken verändern, indem sie die Wolkentröpfchenanzahlkonzentration (N_d), die optischen Wolkeneigenschaften, die Niederschlagsbildung, den Flüssigwasserpfad (LWP) und den Wolkenbedeckungsgrad (CF) beeinflussen. Unter diesen Prozessen gehören aerosolinduzierte Änderungen von CF, die sogenannte CF-Anpassung, potenziell zu den größten, aber am wenigsten eingeschränkten Komponenten des effektiven Strahlungsantriebs durch ACI. Die Abschätzung und das Verständnis dieser Anpassung bleiben jedoch schwierig, da Aerosol-, Wolken- und meteorologische Bedingungen in großskaligen Beobachtungen stark kovariieren, während die simulierte CF-Antwort in Modellen zusätzlich von Konfigurationsentscheidungen abhängen kann, einschließlich Wolkenbedeckungsparametrisierungen und vorgeschriebener Beschränkungen für N_d .

Diese Arbeit begegnet diesen Herausforderungen durch die Entwicklung eines erklärbaren Machine-Learning-(ML)-Frameworks über den nahezu globalen Ozeanen. Dieses Framework kombiniert regionsspezifische eXtreme-Gradient-Boosting-(XGBoost)-Modelle mit dem SHapley-Additive-exPlanations-(SHAP)-Ansatz und verwendet N_d als Proxy für wolkenaktive Aerosole. Innerhalb jedes geografischen $5^\circ \times 5^\circ$ Fensters, aggregiert aus den ursprünglichen Datengittern, wird CF durch ein XGBoost-Modell aus N_d zusammen mit einer Reihe thermodynamischer und dynamischer meteorologischer Prädiktoren vorhergesagt. SHAP-Werte werden anschließend verwendet, um die CF-Antworten zu quantifizieren und dabei meteorologische Kovariabilität zu berücksichtigen. Diese Arbeit verbindet zwei komplementäre Perspektiven: eine beobachtungsbasierte Analyse mit

¹Diese Zusammenfassung wurde mithilfe generativer KI-Werkzeuge aus dem englischen Original übersetzt und anschließend vom Autor inhaltlich überprüft.

Satellitenretrievals des Moderate Resolution Imaging Spectroradiometer (MODIS) und ERA5-Reanalysedaten sowie eine modellbasierte Analyse mit Ausgaben des ICOsahedral Nonhydrostatic (ICON) Atmosphärenmodells, gekoppelt mit dem Hamburg Aerosol Module (HAM) (ICON-HAM). Das übergeordnete Ziel ist es, die CF-Antwort auf Aerosole sowohl in MODIS als auch in ICON-HAM zu quantifizieren und physikalisch zu interpretieren, wobei jeweils die Rolle meteorologischer Bedingungen und von Modellkonfigurationsentscheidungen untersucht wird.

Die beobachtungsbasierte Analyse zeigt eine nahezu global positive $\ln N_d$ -CF-Sensitivität, mit den stärksten Werten in den Stratocumulus-zu-Cumulus-Übergangsregionen vor den westlichen Kontinentalrändern und in den mittleren Breiten der Südhemisphäre, und den schwächsten Werten in den Tropen. Über direkte Sensitivitäten hinaus erlaubt dieses Framework die explizite Quantifizierung des meteorologischen Einflusses auf die $\ln N_d$ -CF-Sensitivität mithilfe eines Interaktionsindex (IAI), der aus SHAP-Interaktionswerten abgeleitet wird. Thermodynamische Variablen, insbesondere die geschätzte Inversionsstärke (EIS) und die Meeresoberflächentemperatur (SST), spielen eine größere Rolle als dynamische Variablen, sowohl für die Kontrolle von CF selbst als auch für die Modulation der N_d -CF-Beziehung. Die IAI-Muster deuten darauf hin, dass höhere EIS die $\ln N_d$ -CF-Sensitivität in den mittleren Breiten verstärkt, während höhere SST dies in den Stratocumulus-zu-Cumulus-Übergangsregionen bewirkt.

Die modellbasierte Analyse wendet dasselbe erklärbare ML-Framework auf sechs ICON-HAM-Simulationen an. Diese kombinieren zwei Wolkenbedeckungsparametrisierungen—CC-RH, das nur von der relativen Feuchte (RH) abhängt, und CC-RH-LWC, das zusätzlich vom Flüssigwassergehalt (LWC) abhängt—mit drei vorgeschriebenen unteren Grenzen für N_d ($N_{d,\min} = 10, 40$ und 100 cm^{-3}). Im Vergleich zu den MODIS-basierten Ergebnissen zeigt ICON-HAM eine stärker nichtlineare und sättigende $\ln N_d$ -CF-Beziehung; daher wird die SHAP-basierte Sensitivitätsanalyse durch stückweise lineare Regression erweitert. Die diagnostizierte CF-Sensitivität wird gemeinsam durch das Wolkenbedeckungsschema und $N_{d,\min}$ über einen sogenannten “Headroom”-Effekt geprägt: Ein höheres $N_{d,\min}$ unterdrückt die Autokonversion und erhöht den LWP; in CC-RH-LWC erhöht dies den mittleren CF-Zustand, da CF explizit vom Wolkenwassergehalt abhängt. Dieser erhöhte Ausgangszustand lässt weniger Raum für weitere aerosolinduzierte CF-Zunahmen, wodurch der Beitrag von $\ln N_d$ zur CF-Vorhersage begrenzt und die diagnostizierte $\ln N_d$ -CF-Sensitivität abgeschwächt wird. Damit gilt die Erwartung, dass CC-RH-LWC stärkere Sensitivitäten als CC-RH zeigt, nur bei niedrigem $N_{d,\min}$ und kehrt sich bei höheren Werten um.

Insgesamt zeigt diese Arbeit, dass die CF-Antwort auf Aerosole sinnvoller interpretiert werden kann, wenn meteorologische Kovariabilität berücksichtigt und der Hintergrundzustand der Wolken einbezogen wird. In Beobachtungen zeigt sich diese Zustandsabhängigkeit als regionale meteorologische Modulation und als räumliche Unterschiede zwischen Wolkenregimen; in Klimamodellen wird sie zusätzlich durch Konfigurationsentscheidungen geprägt, die den simulierten mittleren CF-Zustand verändern. Diese Ergebnisse verbessern die beobachtungsbasierte Einschränkung der CF-Antwort, identifizieren unterschiedliche Verhaltensweisen der $\ln N_d$ -CF-Beziehung in MODIS und ICON-HAM und zeigen, wie spezifische Aspekte der Modellkonfiguration die simulierte Antwort beeinflussen. Zusammen deuten sie darauf hin, dass aussagekräftige Modell-Beobachtungs-Vergleiche eine sorgfältige Berücksichtigung des Hintergrundzustands erfordern, in dem die jeweilige Abschätzung abgeleitet wird.

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Karlsruhe, May 2026

Declaration on the use of generative AI tools

In the preparation of this dissertation, generative artificial intelligence (AI) tools were used as supporting tools for brainstorming, structural refinement, logical consistency checks, language and grammar correction. Notably, the German abstract was translated from the original English abstract with AI assistance.

Generative AI tools were not used to generate original scientific results, perform data analyses, create figures, or replace the author's scientific interpretation. Neither the original scientific results nor the thesis text were generated directly by AI. All AI-assisted suggestions were critically reviewed, checked, and revised by the author. The author takes full responsibility for the content, accuracy, originality, and scientific integrity of this dissertation.

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List of Acronyms

ACI	aerosol–cloud interactions
BLH	boundary-layer height
CCN	cloud condensation nuclei
CF	cloud fraction
CRE	cloud radiative effect
CTT	cloud-top temperature
ECMWF	European Centre for Medium-Range Weather Forecasts
EIS	estimated inversion strength
ERA5	fifth generation ECMWF atmospheric reanalysis
ERF	effective radiative forcing
ERFaci	effective radiative forcing due to aerosol-cloud interactions
GCM	global climate model
HAM	Hamburg Aerosol Module
IAI	interaction index
ICON	ICOsahedral Nonhydrostatic
ICON-HAM	ICON coupled with HAM
LHF	latent heat flux
LWC	liquid water content
LWP	liquid water path
MBL	marine boundary layer
ML	machine learning
MLC	marine low cloud
MODIS	MODerate Resolution Imaging Spectroradiometer

List of Acronyms

PD	present-day
PI	preindustrial
PLR	piecewise linear regression
PR	precipitation rate
RH	relative humidity
SH	specific humidity
SHAP	SHapley Additive exPlanations
SHF	sensible heat flux
SST	sea surface temperature
TOA	top of atmosphere
XGBoost	eXtreme Gradient Boosting

List of Symbols

α_c	cloud albedo	--
N_a	aerosol number concentration	cm^{-3}
N_d	cloud droplet number concentration	cm^{-3}
$N_{d,\min}$	prescribed lower bound for N_d	cm^{-3}
ω	vertical velocity	Pa s^{-1}
r_e	cloud droplet effective radius	μm
τ_c	cloud optical depth	--
u	zonal (eastward) wind	m s^{-1}
v	meridional (northward) wind	m s^{-1}

Chapter 1

Introduction

1.1 Earth's energy budget and the role of clouds

The Earth's weather and climate are fundamentally determined by the exchange of energy between the Earth system and space, a process dominated by radiative fluxes at the top of atmosphere (TOA). At TOA, incoming solar (shortwave) radiation is partly reflected back to space by gases, clouds, aerosols, and the surface, with the remainder absorbed by the Earth's atmosphere and surface. At the same time, the Earth-atmosphere system emits thermal (longwave) radiation to space (Trenberth et al., 2009; Forster et al., 2021). Energy entering the TOA is redistributed by internal energy flux, including non-radiative latent and sensible heat (Fig. 1.1), and drives atmospheric dynamics and the hydrological cycle (Stephens et al., 2012).

The difference between absorbed shortwave and emitted longwave sets the TOA energy balance and thus the global net energy gain or loss. If the system absorbs more energy than it emits, it accumulates energy and warms; if it emits more than it absorbs, it loses energy and cools. In a long-term and global equilibrium, the incoming shortwave radiation should be nearly balanced by the sum of reflected shortwave and outgoing longwave radiation. However, observations in recent decades show a persistent positive imbalance, indicating a continuous accumulation of energy in the Earth system (Loeb et al., 2021; von Schuckmann et al., 2023). This imbalance is primarily driven by anthropogenic radiative forcing, especially increasing greenhouse gas concentrations, which perturb the radiative equilibrium (Forster et al., 2021). As the system responds to this forcing, clouds and aerosols can either amplify or mitigate the resulting energy imbalance through feedbacks and radiative effects (Bellouin et al., 2020).

Clouds play a central role in Earth's radiative balance because they modulate both incoming shortwave and outgoing longwave fluxes through two compet-

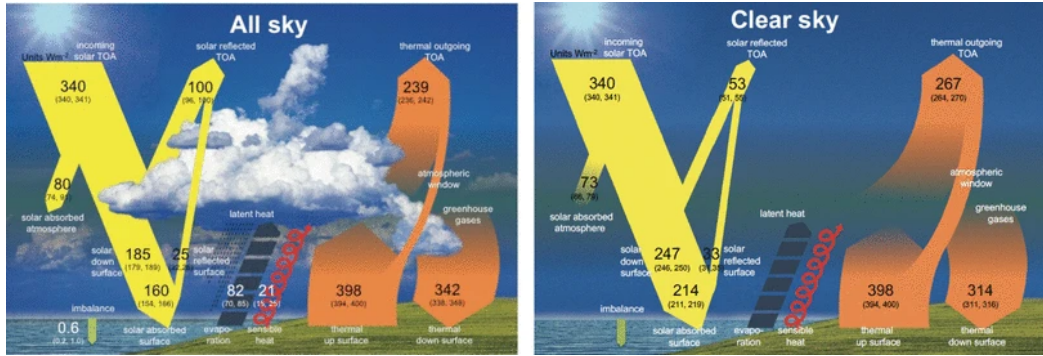


Figure 1.1: Schematic representation of the Earth’s present-day (early 21st century) global mean energy budget. Numbers indicate the globally-averaged best estimated magnitudes of each individual energy balance component in W m^{-2} , with their uncertainty ranges in parentheses. Reproduced from Wild et al. (2019).

ing effects. By reflecting solar radiation back to space, clouds increase planetary albedo and exert a cooling effect; by absorbing and re-emitting terrestrial thermal radiation, they reduce outgoing longwave radiation and contribute to warming (Ramanathan et al., 1989). This contrast between all-sky (with clouds) and clear-sky (without clouds) conditions is illustrated in Fig. 1.1. The cloud radiative effect (CRE) is defined as the difference between all-sky and clear-sky TOA radiation, and it depends on cloud amount, phase, type, altitude, optical thickness and microphysical properties (L’Ecuyer et al., 2019). Clouds sit at the interface between radiation, dynamics and the hydrological cycle, and even modest changes in their properties can considerably perturb the TOA radiative balance and the resulting climate response (Stephens et al., 2012; Zelinka et al., 2020). Accordingly, cloud feedbacks, which are defined as changes in the CRE response to surface warming, remain the dominant source of uncertainty in climate sensitivity estimates across climate models (Sherwood et al., 2020; Zelinka et al., 2014), making understanding of cloud processes and their responses to perturbations critical for projecting future climate change.

The influence of clouds on Earth’s radiative budget varies substantially across cloud regimes, because different clouds contribute to the shortwave albedo effect and the longwave greenhouse effect in distinct manners. A basic contrast is between high-level and low-level clouds, whose radiative influences are opposite because of their contrasting altitude and temperature. High clouds (e.g. thin cirrus) are often optically thin, allowing the majority of incoming solar radiation to pass through. At the same time, due to their high altitude and the resulting low cloud-top temperature (CTT), they absorb upwelling thermal radiation from the

warmer surface and re-emit it at much colder temperatures, reducing the longwave radiation lost to space. Therefore, while high clouds have a minimal influence on incoming shortwave radiation, they effectively trap outgoing longwave radiation, exerting a net warming effect (Hartmann et al., 1992; Chen et al., 2000).

In contrast, low-level clouds, particularly marine stratocumulus and shallow cumulus, have the opposite radiative effect. Due to their higher liquid water path (LWP) and more numerous and smaller cloud droplets, low clouds are typically optically thicker (higher cloud optical depth (τ_c)) than high clouds. This results in a higher cloud albedo (α_c) that effectively reflects incoming shortwave radiation, producing strong shortwave cooling. Meanwhile, their CTTs are relatively close to the surface temperature, yielding little thermal contrast and thus a weak longwave greenhouse effect (Hartmann et al., 1992; Boucher et al., 2013). Therefore, low-level clouds typically exert a net cooling influence on the global mean radiation budget.

Marine low clouds (MLCs), which are normally in liquid phase, comprise one of the most radiatively important regimes and a primary contributor to the Earth’s energy budget among all low-level clouds. This is because they are typically highly reflective relative to the dark ocean surface beneath and cover a vast spatial extent of the global oceans (Klein and Hartmann, 1993; Wood, 2012). MLCs are mainly composed of stratocumulus in the eastern subtropical ocean basins and shallow cumulus in the trade wind regions, spanning large expanses of the subtropics, tropics and midlatitudes (Bony et al., 2006; Scott et al., 2020). Even small changes in their coverage, optical depth, or microphysical properties can produce substantial changes in the TOA radiative balance. These clouds are tightly coupled to boundary-layer thermodynamics and dynamics, making them sensitive to local environmental conditions and to changes in the background climate state (Bretherton, 2015; Sherwood et al., 2020). Consequently, MLCs are widely regarded as one of the main sources of uncertainty in cloud feedbacks and climate sensitivity (Bony and Dufresne, 2005; Sherwood et al., 2020; Zelinka et al., 2020).

1.2 Marine low clouds: formation, development and controls

1.2.1 Cloud sources

1.2.1.1 Lifting mechanisms

Cloud formation typically begins when air is lifted and cools. As an air parcel ascends, it expands and cools adiabatically. Consequently, its capacity to hold water

vapour decreases (i.e. saturation vapour pressure drops), so relative humidity (RH) rises until it reaches saturation and eventually a supersaturation ($RH > 100\%$) can develop. At this stage, water vapour can condense onto aerosol particles and cloud droplets can form (Lohmann et al., 2016). Should no suitable aerosol particles be present in the atmosphere, condensation of water vapour onto ions—the next viable surface—would require a RH of roughly 420 %, a supersaturation nearly impossible to achieve in the natural atmosphere (Mason, 1960; Gordon et al., 2023).

In the troposphere, the initial ascent can be triggered primarily by four broad mechanisms (Lohmann et al., 2016): (i) convection driven by surface heating; (ii) large-scale convergence near the surface; (iii) synoptic-scale lifting along fronts (e.g. in extratropical cyclones); and (iv) orographic lifting (e.g. mountain ridges). In the marine boundary layer (MBL), where MLCs form, mechanism (iv) is generally not relevant. Compared to deep convective clouds, lifting for MLC formation is typically weaker but more persistent. Mechanism (i), namely turbulent eddies and shallow convective motions driven by surface heating and moisture fluxes, continuously generates small-scale updrafts and adiabatic cooling. This is an important trigger across most MLC regimes, particularly for stratocumulus and trade-wind cumulus (Wood, 2012; Bretherton, 2015). Mechanism (ii), mesoscale and larger-scale convergence, can also play a role, particularly for trade-wind cumulus organization and during transitions between cloud regimes (Bretherton and Blossey, 2017; Narenpitak et al., 2021; Saffin et al., 2023). Mechanism (iii), frontal lifting associated with extratropical cyclones, is relevant in the midlatitudes, where extensive and persistent shallow cloud decks are often observed in the cold sectors of trailing cold fronts during cold-air outbreaks (Norris and Klein, 2000; Field and Wood, 2007; Wood, 2012).

Once MLCs are established, cloud-top longwave radiative cooling becomes a critical process for many MLCs, especially for stratocumulus. Longwave emission from the cloud top cools the surrounding air and destabilizes the upper boundary layer, generating buoyancy that drives turbulent overturning. This turbulence helps maintain continued mixing and saturation within the boundary layer, thereby supporting persistent cloudiness. This cooling at the cloud top also sharpens the temperature jump across the boundary-layer top, reinforcing the capping inversion and enhancing convective instability (Wood, 2012; Bretherton, 2015; Lohmann et al., 2016; Quaas and Gryspeerdt, 2022).

1.2.1.2 Droplet activation

The lifting mechanisms described above explain how air parcels are brought toward saturation through adiabatic cooling. Once saturation is achieved, further cooling can produce a small supersaturation. The relevant process then shifts from the large-scale dynamics governing the motion of air parcels to the microphysical scale, where aerosol particles can activate into cloud droplets. However, supersaturation alone is insufficient for cloud formation, because water vapour requires suitable surfaces to condense onto. In a typical atmosphere, homogeneous nucleation (formation from pure water) is energetically unlikely; instead, atmospheric aerosol particles provide the most suitable pre-existing surfaces for heterogeneous nucleation by acting as cloud condensation nuclei (CCN), allowing droplets to form at supersaturations typically below 1 % (Wallace and Hobbs, 2006; Seinfeld and Pandis, 2016).

Whether a given aerosol particle activates into a cloud droplet is governed by Köhler theory (Köhler, 1936), which describes the competition between two physical effects that determine the equilibrium saturation ratio over an aqueous solution droplet as a function of its wet radius (Fig. 1.2, left). In other words, the Köhler curve itself is essentially given by the size-dependent equilibrium saturation ratio $S(r)$. The two competing components are as follows:

- **The Kelvin (curvature) effect:** A curved droplet surface has higher surface free energy than a flat surface, meaning water molecules are less tightly bound and thus the equilibrium vapour pressure over a droplet is higher. For smaller droplets, higher curvature increases the equilibrium vapour pressure, making them more prone to evaporate unless the environment is sufficiently supersaturated. As the droplet size increases, curvature decreases and the equilibrium vapour pressure approaches that of a flat surface, making growth possible at lower supersaturation.
- **The Raoult (solute) effect:** Soluble substances dissolved in a droplet reduce the mole fraction of water at the surface, decreasing the number of water molecules available to escape from the surface into the vapour phase. As a result, the equilibrium saturation ratio over a solution droplet is lower than over a pure-water droplet of the same size, so a solution droplet can be in equilibrium at a lower RH. For smaller droplets, the solute is more concentrated within a smaller volume, amplifying the reduction in the equilibrium vapour pressure. As the droplet grows, the solute becomes diluted and the equilibrium vapour pressure rises back toward that of pure water.

As an air parcel rises and cools and supersaturation is reached, a droplet grows along its Köhler curve and may surpass a critical point (the peak of the Köhler curve as shown in Fig. 1.2), which reflects the combination of the Kelvin and Raoult effects. This critical point on the curve corresponds to a so-called critical or activation saturation ratio (S_{act}) and a critical radius (r_{act}); once both are exceeded, the droplet becomes effectively activated and undergoes runaway growth spontaneously, whereas unactivated droplets remain in stable equilibrium as hygroscopically grown haze droplets (Lohmann, 2009; Seinfeld and Pandis, 2016). Figure 1.2 (left) also illustrates that smaller dry radii shift the Köhler curve such that the peak is moved towards a higher critical supersaturation ($S_{\text{act}} - 1$), meaning that smaller particles require higher ambient supersaturations to activate.

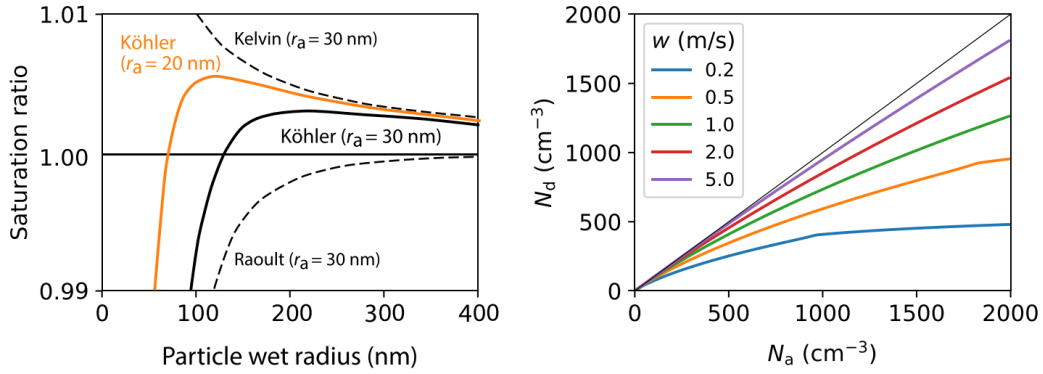


Figure 1.2: Köhler activation schematic. Left: Kelvin, Raoult, and combined Köhler curves for different aerosol dry radii (r_a). Right: the dependence of activated droplet number concentration (cloud droplet number concentration (N_d)) on aerosol number concentration (aerosol number concentration (N_a)) for different updraft velocities w (reproduced from Quaas and Gryspeerdt, 2022, Fig. 12.6 with permission from Elsevier).

In practice, the number of activated droplets and thus the final N_d is not solely a function of aerosol properties (e.g. N_a , size distribution and hygroscopicity), but also depends on dynamical conditions, mainly intensity of upward motions. Stronger vertical velocities (ω) can drive the environmental saturation ratio S_{env} to higher peak values through enhanced adiabatic cooling, activating a larger fraction of the available CCN population, including smaller and less soluble particles that require higher critical supersaturations. As shown in Fig. 1.2 (right), N_d increases sublinearly with N_a and is systematically higher for stronger updrafts (higher vertical wind speeds). In pristine marine environments, N_d is often “aerosol-limited” due to low background CCN concentrations, and therefore small increases

in N_a can lead to relatively substantial increases in N_d . Conversely, in more polluted areas, N_d tends to be “updraught-limited” (i.e. more limited by the peak supersaturation). In this regime, additional aerosols cannot activate because water vapour is depleted quickly by the large droplet population. The microphysical interplay between aerosol availability and vertical motion serves as the foundation for aerosol–cloud interactions (ACI) pathways discussed in Sects. 1.3.2 and 1.3.3, including the response of cloud optical properties to aerosol perturbations, and subsequent cloud adjustments.

1.2.1.3 Condensational growth

Figure 1.3 illustrates in more detail what happens after a droplet is activated (i.e., once the critical point at S_{act} and r_{act} is exceeded): the droplet enters a regime of unstable condensational growth. Water vapour molecules diffuse toward the droplet surface driven by the positive difference between S_{env} and $S(r)$ along the Köhler curve. As long as $S_{env} > S(r)$, the droplet grows spontaneously, while if $S_{env} < S(r)$ it evaporates and shrinks. In an ascending air parcel, supersaturation is produced by adiabatic cooling, but it is simultaneously balanced by (i) expansion due to decreasing pressure with height, which reduces the actual vapour pressure; (ii) loss of water vapour from gas phase to condensation, directly reducing the actual vapour pressure; and (iii) latent heat release due to the phase change from vapour to liquid, which warms the air parcel and raises the saturation vapour pressure. These three processes collectively reduce RH, so the supersaturation peaks shortly above cloud base and then declines as droplets continue to grow and the vapour loss rate increases (Quaas and Gryspeerdt, 2022). During the condensational growth, the continued growth progressively dilutes the solute and $S(r)$ approaches unity (Seinfeld and Pandis, 2016).

A defining characteristic of this diffusional growth is that the growth rate in terms of radius is inversely proportional to the droplet radius, $\frac{dr}{dt} \propto \frac{1}{r}$. Therefore, smaller activated droplets grow faster than larger ones, tending to narrow the droplet size distribution during the early stage of cloud formation (Lohmann et al., 2016). Condensational growth drives the initial increase in MLC liquid water mass, but by itself it is relatively slow to produce precipitation-sized droplets within the typical lifetime of MLCs—warm-rain formation usually requires collision–coalescence (Wallace and Hobbs, 2006; Seinfeld and Pandis, 2016).

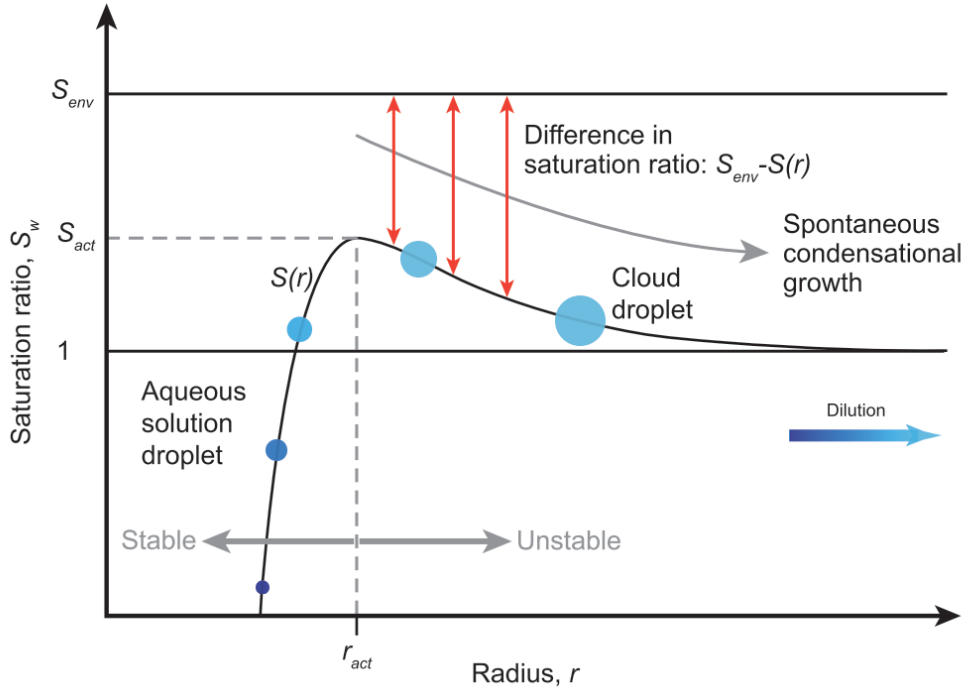


Figure 1.3: Schematic of condensational growth after Köhler activation at a given environmental saturation ratio S_{env} , driven by the difference between S_{env} and the equilibrium saturation ratio $S(r)$ on the Köhler curve; the peak defines the critical saturation ratio S_{act} and critical radius r_{act} for activation. (reproduced from Lohmann et al., 2016, Fig. 8.2 with permission from Cambridge University Press through PLSclear).

1.2.2 Cloud sinks

1.2.2.1 Collision and coalescence

In the environment where MLCs form, temperatures are normally above freezing, and mechanical collection, namely collision–coalescence, is the primary mechanism for droplet growth and warm-rain formation. This process relies on differential terminal fall velocities of different drop sizes: larger cloud droplets fall faster than smaller ones, collide with them, and merge into progressively larger drops that can eventually precipitate. As shown in Fig. 1.4, the droplet growth undergoes a shift in efficiency as they enlarge: growth by condensation (curve (a), blue) is efficient for smaller droplets, but it decelerates as droplets grow as discussed in Sect. 1.2.1. During this stage when the condensational growth dominates, growth by mechanical collection (curve (b), red) is initially negligible. However, as the

collector droplet approaches a radius of approximately $20\text{ }\mu\text{m}$ and continues to enlarge, growth by collision–coalescence accelerates and becomes the dominant mechanism (Wallace and Hobbs, 2006). This transition is broadly consistent with the often-cited threshold for cloud droplet effective radius (r_e) of approximately $14\text{ }\mu\text{m}$ (Fan et al., 2016; Rosenfeld et al., 2019, 2023), reflecting the nonlinear overtaking of collection over condensational growth.

Collision efficiency increases as the size difference between the collector (i.e. larger) droplet and the smaller droplet it encounters decreases. Upon contact, whether the droplets actually merge is set by the coalescence efficiency (i.e. whether they coalesce or rebound). The product of collision efficiency and coalescence efficiency defines the overall collection efficiency, which governs the rate of growth by collision-coalescence (Lohmann et al., 2016; Kreidenweis et al., 2019). This process is inherently self-accelerating: each successful coalescence event produces a larger drop with even faster gravitational settling and higher collection efficiency, further enhancing its ability to collect relatively large droplets that have grown through differential growth, ultimately yielding drizzle and raindrops that sediment out of the cloud.

This helps explain systematic regime differences: in remote marine conditions where aerosol loadings and thus N_d are low, available water vapour is distributed among fewer droplets, favouring larger mean droplet sizes and often with broader size spectra for a similar cloud water content, more readily initiating collision–coalescence than in more polluted continental regions (Mülmenstädt and Feingold, 2018; Kreidenweis et al., 2019). The resulting contrast in droplet size and spectral width strongly influences both cloud optical properties and precipitation efficiency—in continental or more polluted clouds, higher N_d keeps droplets smaller that are less likely to reach the size of efficient collection, thereby delaying or suppressing warm rain. Once precipitation does develop in cleaner air, aerosol scavenging by drizzle and rain can further reduce N_d , reinforcing the tendency toward fewer, larger droplets so that droplets collide and coalesce more efficiently. This creates a positive feedback loop that can amplify the contrast between clean and polluted clouds or between precipitating and non-precipitating clouds, which can be manifested in the transition between open- and closed-cell clouds (Jing and Suzuki, 2018; Rosenfeld et al., 2023). Since precipitation can thin or break up clouds, collision–coalescence represents an important sink pathway in the context of MLC ACI through which aerosol-driven cloud microphysical changes can influence cloud macrophysical properties (Sects. 1.3.2 and 1.3.3).

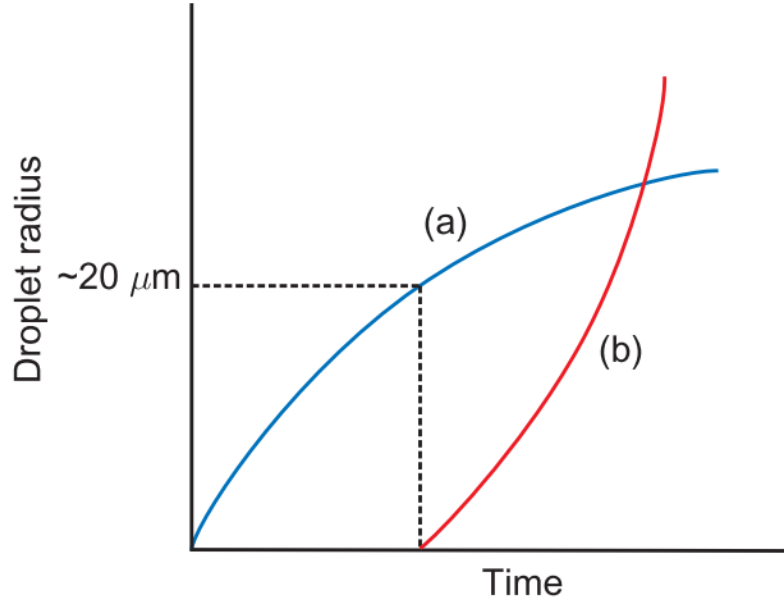


Figure 1.4: Schematic curves of droplet growth by (a) condensation from the vapour phase (blue) and (b) collection of droplets via collision-coalescence (red). Reproduced from Wallace and Hobbs (2006, Fig. 6.15) with permission from Elsevier.

1.2.2.2 Precipitation: autoconversion and accretion

In bulk microphysical descriptions of warm-rain formation in atmospheric models, which use averaged quantities rather than explicitly resolving the droplet size distribution, collision-coalescence is commonly parameterized by two processes:

- **Autoconversion:** the initial phase of droplet growth, where cloud droplets collide and coalesce with one another to form embryonic drizzle drops or raindrops. It represents the onset of warm-rain formation and the autoconversion rate depends mainly on liquid water content (LWC) and N_d . This stage is particularly sensitive to aerosol perturbations, because an increase in N_d reduces the mean droplet size and suppresses autoconversion (Khairoutdinov and Kogan, 2000; Wood, 2012).
- **Accretion:** the subsequent collection of the remaining population of smaller droplets by already-formed rain or drizzle drops falling through the cloud. It is responsible for the rapid intensification of precipitation once rain formation has begun. In mature precipitating MLCs, accretion typically dominates the total rain production rate and depends on both the availability of cloud

water and the existing rain water content

In almost all global climate models (GCMs), aerosol-induced cloud adjustments commonly reduce the autoconversion rate, i.e. they suppress the initial conversion of cloud droplets into drizzle (Quaas and Gryspeerdt, 2022). This is because autoconversion depends not only on the cloud liquid mass but also on N_d , and is therefore directly sensitive to aerosol-induced changes in droplet populations. Accretion, by contrast, is formulated as a function of cloud water and rainwater masses alone and is not directly affected by aerosol perturbations (Wood et al., 2009; Michibata and Takemura, 2015; Quaas and Gryspeerdt, 2022; Michibata et al., 2025). For this reason, autoconversion is generally regarded as the sole microphysical pathway through which aerosols can suppress precipitation and thereby potentially prolong cloud lifetime and modify cloud adjustments.

1.2.2.3 Evaporation and entrainment

While warm-precipitation serves as an important vertical sink for MLCs, cloud droplets can also shrink or disappear through evaporation when exposed to subsaturated ambient air and return to the vapour phase. This is particularly relevant in non-precipitating or thin stratocumulus. Evaporation occurs commonly at cloud edges where cloudy air mixes with drier environmental air. This mixing can be described by *entrainment*—the incorporation of surrounding environmental air into an air parcel and the associated modification of its properties—and *detrainment*, the reverse process where air leaves the air parcel (Lohmann et al., 2016). Mixing with entrained subsaturated air causes cloud droplets to evaporate until the mixed air becomes saturated, reducing the cloud water mass and thinning the cloud layer.

The rate of evaporation depends on the degree of subsaturation and on the droplet size. For a given LWC, a higher N_d implies smaller droplets with higher surface-area-to-volume ratios, facilitating more rapid evaporation. Nevertheless, the influence of N_d on the rate and spatial distribution of cloud evaporation can also be understood from the perspective of vapour diffusion: in the context of condensational growth as discussed in Sect. 1.2.1, the droplet growth rate scales inversely with droplet radius, $\frac{dr}{dt} \propto \frac{1}{r}$ ($\frac{dr}{dt} > 0$). This inverse size dependence remains valid for evaporation (i.e. negative growth, $\frac{dr}{dt} < 0$) under subsaturated conditions. Therefore, clouds with higher N_d and correspondingly smaller droplets evaporate more rapidly (Quaas and Gryspeerdt, 2022).

Evaporation cools the surrounding air through latent heat consumption, and modifies buoyancy and turbulent motions. In MLCs, evaporation is particularly important where cloud air mixes with unsaturated environmental air along cloud

boundaries: in more convective clouds, lateral entrainment and detrainment are more prominent, whereas in more stratiform clouds cloud-top entrainment dominates (Wood, 2012; Bretherton, 2015; Lohmann et al., 2016).

Cloud-top entrainment is therefore a key sink mechanism for MLCs, since most of them are not strongly convective. It refers to the incorporation of warm, dry free-tropospheric air across the capping inversion into the cloud-topped MBL. As illustrated in Fig. 1.5, entrainment occurs primarily in the entrainment interfacial layer, which is a thin turbulent zone, typically a few tens of meters thick just above the cloud. Entrainment is coupled with two radiative cooling mechanisms at the cloud top.

- **Longwave cooling:** The primary driver is the emission of strong longwave radiation from the cloud top to space, denoted as IR (inferred) cooling in Fig. 1.5. This cooling produces negative buoyancy and turbulent eddies, facilitating the downward transport of warm, dry free-tropospheric air across the inversion and into the boundary layer (Lohmann, 2009; Mellado, 2017).
- **Evaporative cooling:** The entrained air enhances the evaporation of cloud droplets to restore local thermodynamic equilibrium. The associated latent heat consumption further amplifies negative buoyancy locally, reinforcing downward turbulent mixing. The interaction between longwave cooling, evaporation, and turbulent eddies thus forms a positive feedback that continuously reduces cloud liquid water mass and thins the cloud (Bretherton, 2015; Mellado, 2017).

It is worth noting that the cloud-top longwave cooling plays a dual role: it acts as a maintenance mechanism by driving the turbulent overturning that brings the moisture up from below as discussed in Sect. 1.2.1. At the same time, it acts as a cloud-thinning and dissipation mechanism through entrainment. Because aerosol perturbations modify N_d and droplet size, they can alter the rate of evaporation and entrainment, providing a pathway for cloud adjustments (see Sect. 1.3.3).

1.2.3 Boundary-layer thermodynamic and dynamical controls

The formation of MLCs, including the microphysical processes described above, does not operate in isolation from its environment. Their formation, persistence and macrophysical properties are embedded in and modulated by the large-scale thermodynamic and dynamical state of the MBL, which regulates the supply of moisture and heat from below, the strength of turbulent mixing within MBL, and the entrainment of warm and dry air from the free troposphere. Figure 1.6

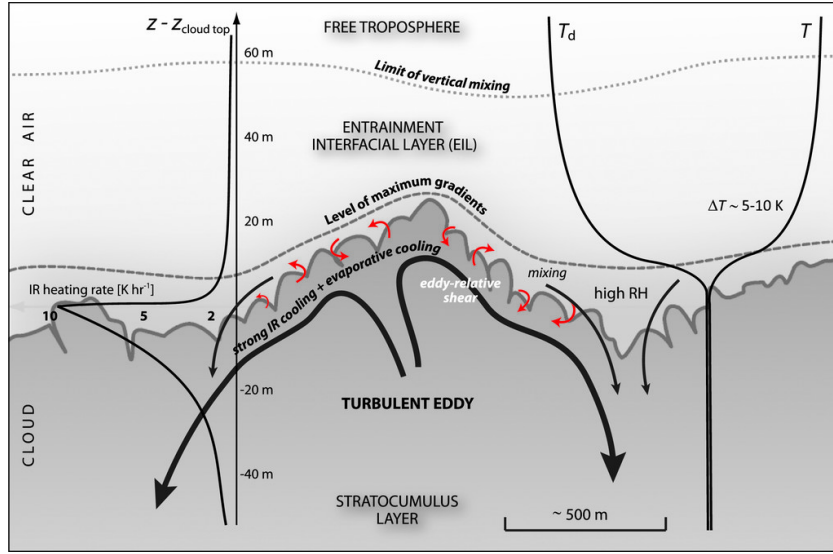


Figure 1.5: Schematic of cloud-top entrainment of warm, dry free-tropospheric air into marine stratocumulus, highlighting the interactions between cloud-top inferred (longwave) cooling, evaporative cooling, and the resulting turbulent eddies within the entrainment interfacial layer. Reproduced from Wood (2012) with permission from the American Meteorological Society.

summarizes several of the key large-scale meteorological controls and their expected influences on MLC regimes. Understanding these environmental controls is important not only to understand why and where MLCs form, but also in the context of ACI because some of the factors covary with both cloud properties and aerosol loading, creating meteorological confounders that tangle and complicate the observational quantification of ACI.

1.2.3.1 Thermodynamic stability and the capping inversion

A major control on the structure and persistence of MLCs is the thermodynamic stability near and above the MBL top. A strong capping temperature inversion, which separates the MBL from the free troposphere, suppresses deep vertical mixing across the cloud top, confines moisture within the boundary layer and favours a shallow, well-mixed boundary layer conducive to extensive low-cloud decks such as stratiform clouds and stratocumulus (Klein and Hartmann, 1993; Bretherton et al., 2013; Wood, 2012). Conversely, a weaker inversion reduces this energetic barrier to cloud-top entrainment, allowing more warm, dry free-tropospheric air to enter the MBL and thereby increasing entrainment drying (Scott et al., 2020; Myers et al., 2023). The estimated inversion strength (EIS) is

a refined and widely used metric of lower-tropospheric stability that explicitly accounts for the moist-adiabatic temperature profile (Wood and Bretherton, 2006), and it is a fundamental indicator of conditions favourable to MLCs. As shown in Fig. 1.6, stronger EIS is associated with increased low-cloud cover, a relationship that is well documented across studies (e.g. Myers and Norris, 2015; McCoy et al., 2017b; Andersen et al., 2023; Jia et al., 2024). Inherently related to this is the temperature structure of the lower free troposphere, for example, temperatures at 700 hPa and 850 hPa, which helps characterize the thermal stratification of the lower troposphere and the inversion strength.

1.2.3.2 Moisture, surface fluxes, and boundary-layer structure

The moisture state of the MBL and the lower troposphere, replenished by the turbulent exchange of heat and moisture at the ocean surface, is central to MLC evolution and maintenance, because these factors regulate boundary-layer buoyancy and moisture availability and thereby strongly influence cloud properties. Sea surface temperature (SST) sets an important lower-boundary condition for this coupling and can serve as an important surrogate for many processes (Qu et al., 2015b). There are two possible pathways for how SST influences MLCs: (i) warmer SST increases the sea-air temperature and moisture gradient, driving stronger latent heat flux (LHF) and evaporation into the boundary layer. The additional moisture and heat enhance buoyancy and thus upward turbulent mixing that reaches the cloud top, coupled with cloud-top radiative cooling. This intensifies cloud-top entrainment, leading to cloud thinning and dissipation (Rieck et al., 2012; Qu et al., 2015b). (ii) As SST increases, the moisture content of the boundary layer generally increases more rapidly with temperature than that of the subsiding free troposphere above, enhancing the humidity contrast between the MBL and the overlying free troposphere. As a result, entrainment of free-tropospheric air more efficiently dries the cloud layer and reduce cloud amount (van der Dussen et al., 2015; Qu et al., 2015b). Together, SST influences not only sensible heat flux (SHF) and LHF near the ocean and associated moisture and heat supply, but also the efficiency of entrainment drying at cloud top (Wood, 2012; Bretherton and Blossey, 2014; Bretherton, 2015). The impact of SST is schematically reflected in Fig. 1.6.

At the same time, the moisture content at multiple levels, represented for instance by RH and specific humidity (SH) both within the boundary layer (e.g. RH at 950 hPa) and in the free troposphere (e.g. RH at 700 hPa), determines the vertical moisture distribution and the degree of subsaturation above the cloud top, regulating the intensity of evaporation and entrainment drying. For example, as

Fig. 1.6 shows, elevated free-tropospheric moisture weakens entrainment drying, thereby moistening the MBL and facilitating greater cloudiness (van der Dussen et al., 2015; Myers and Norris, 2016). Additionally, boundary-layer height (BLH) provides a proxy for the depth of the turbulent layer development in which MLCs reside, closely linked to cloud structure and the degree of coupling to the surface. In this sense, these meteorological factors regarding humidity, SST, turbulent fluxes and BLH together define the local thermodynamic environment in which MLCs form and develop.

1.2.3.3 Large-scale dynamical controls

In addition to the thermodynamic conditions, MLCs are shaped by large-scale dynamics. This can be described by ω and horizontal winds, including zonal (eastward) wind (u) and meridional (northward) wind (v). A key controlling factor is subsidence, which regulates many aspects of MLC organization, including cloud-top entrainment, BLH, and the transition between more stratiform and more broken cloud regimes (Wood, 2012; Bretherton, 2015). In pressure coordinates, subsidence is commonly represented by ω , for which positive values indicate downward motions. The most relevant pressure levels in the context of MLCs are typically 700 hPa and 850 hPa for ω .

The relationship between subsidence and MLC cloudiness is nuanced and needs careful interpretation. In a climatological sense, persistent stratocumulus decks are typically found in regions of strong large-scale subsidence, such as the subtropical eastern ocean basins, where strong subsidence often co-occurs with cold SSTs and strong lower-tropospheric stability. Strong subsidence adiabatically warms the free troposphere and thus builds and maintains a strong capping inversion (i.e. higher EIS), favouring a shallower well-mixed boundary layer and stratiform clouds and preventing the transition into more convective regimes (Wood, 2012; Klein et al., 2017; Myers et al., 2023). However, this climatological co-occurrence does not imply that stronger subsidence itself necessarily increases cloudiness. As illustrated in Fig. 1.6, considering the isolated effect of subsidence at a fixed EIS, stronger subsidence tends to compress the boundary layer and push down the MLC top without proportionally lowering the cloud base, producing a shallower cloud layer with lower LWP and making it more vulnerable to entrainment erosion and hence generally associated with less and thinner MLCs (Myers and Norris, 2013; Bretherton, 2015). Conversely, weaker subsidence allows the MBL to deepen more freely, supporting a rising and hence cooler cloud top. At lower temperatures the saturation mixing ratio decreases, favouring more cloud condensation that

outpaces entrainment drying and promotes more persistent stratocumulus (Myers and Norris, 2013; Blossey et al., 2013). During equatorward advection, a weakening of subsidence and the associated rise in cloud-top height can initially thicken the cloud deck and delay the stratocumulus-to-cumulus transition (van der Dussen et al., 2016). However, if subsidence remains weak for long enough to deepen the boundary layer substantially, it may eventually lead to boundary-layer decoupling, cutting off the surface moisture sources and inhibiting the exchange of moisture and heat between the increasingly stratified subcloud and cloud layers. This may in turn favour cloud thinning or dissipation and accelerate the transition to more broken regimes (Wood and Bretherton, 2004; Sandu and Stevens, 2011). However, this should be viewed in the broader climatological covarying context rather than from an isolation or controlled perspective, because it typically co-occurs with increasing SSTs and decreasing EIS (Klein et al., 2017; Myers et al., 2023). Negative ω (i.e. large-scale ascent) acts in the opposite sense when present by favouring deepening and moistening of the boundary layer and promoting more convective clouds.

Horizontal winds provide additional large-scale dynamical controls. Near the surface, wind speed directly influences the shear-driven mixing and thereby the emission and transport of sea-spray aerosols and the intensity of SHF and LHF, and the turbulent exchange of heat and moisture between the ocean and the atmosphere, affecting the coupling between the surface and the MBL (Wood, 2012; Bretherton et al., 2013; Brueck et al., 2015). Stronger horizontal cold-air advection increases the temperature gradient between the ocean surface and the overlying air, which destabilizes their interface. This further strengthens upward buoyancy fluxes and thereby favours greater cloudiness (Fig. 1.6) (Norris and Iacobellis, 2005). Winds at lower-tropospheric levels influence the advection of temperature, humidity, and aerosols. They also influence the transport of air masses into and out of MLCs, the thermodynamic history of air masses from a Lagrangian perspective, their exposure to different chemical and physical regimes, and the organization of cloud fields on synoptic to mesoscale scales (Wood, 2012; Bretherton, 2015). In this way, integrating these dynamical components together with thermodynamic factors defines the large-scale meteorological regime in which aerosol perturbations act and therefore forms the basis of the predictor framework used later in the machine-learning analysis.

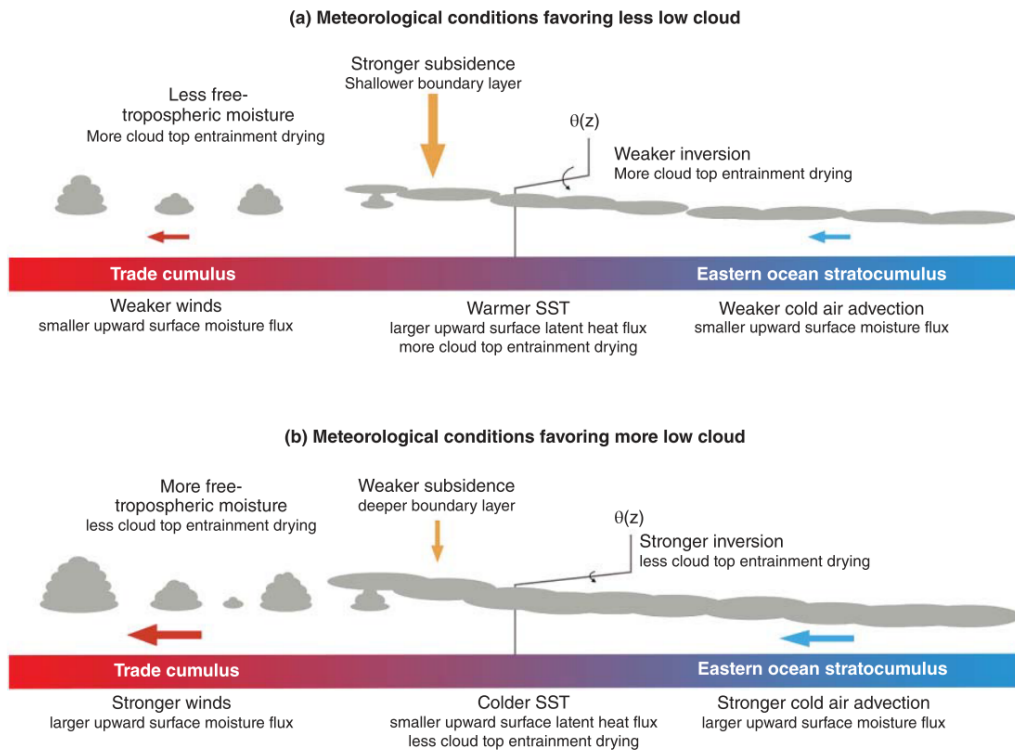


Figure 1.6: Schematic illustration of the responses of subtropical stratocumulus and trade cumulus to changes in individual large-scale meteorological factors, showing conditions favouring less (top) and more (bottom) low cloud. Reproduced from Myers et al. (2023) with permission from John Wiley & Sons; originally published in Scott et al. (2020) with permission from American Meteorological Society.

1.3 Aerosol-cloud interactions in marine low clouds

1.3.1 Radiative significance and climate motivation

Beyond their natural variability and sensitivity to environmental conditions, clouds are also modified by perturbations from atmospheric aerosols. Aerosols can influence the climate system directly by scattering and absorbing radiation, or more complexly, by altering cloud properties. The former is referred to as aerosol-radiation interactions and the latter is termed aerosol-cloud interactions (ACI). Despite decades of research, ACI remains one of the largest sources of uncertainty in estimates of anthropogenic radiative forcing and its translation into anthropogenic surface temperature change, and in model projections of future climate, complicating the interpretation of observed climate change (Bellouin et al., 2020; Forster

et al., 2021).

The standard metric for quantifying the impact of a forcing agent (i.e. climate driver) on the Earth’s radiative budget is the effective radiative forcing (ERF), defined as the change in net TOA radiative flux accounting for adjustments that occur independently of surface temperature (e.g. changes in atmospheric temperatures, water vapour and clouds) (Sherwood et al., 2015; Bellouin et al., 2020; Forster et al., 2021). Within this framework, ERF_{aci} denotes the ERF specifically arising from ACI. Figure 1.7 illustrates that aerosols collectively exert the largest negative ERF among all anthropogenic forcing agents since the preindustrial (PI) era (1750–2019), with effective radiative forcing due to aerosol-cloud interactions (ERF_{aci}) contributing the dominant cooling effect estimated at -0.84 [-1.45 to -0.25] W m^{-2} . Notably, while the ERF from aerosol-radiation interactions is relatively well-constrained, ERF_{aci} carries the greatest uncertainty among all anthropogenic forcing agents considered here (Forster et al., 2021).

These forcing agents drive changes in the Earth’s energy balance and thereby the historical evolution of global-mean surface temperature since the PI. As shown in Fig. 1.8, greenhouse gases such as CO_2 and methane contribute the majority of the observed warming, ACI offsets a substantial fraction of this warming. Importantly, the uncertainty associated with ACI dominates the overall uncertainty in the anthropogenic surface temperature change, accounting for more than 60 % of the total uncertainty in Fig. 1.8 (Im et al., 2026).

As presented in Sect. 1.1, MLCs are among the most important contributors to global shortwave cooling and associated uncertainty in the climate system. Such clouds are not only sensitive to local environmental conditions but are also particularly susceptible to aerosol perturbations for three primary reasons. First, MLCs are spatially extensive and frequently occur in vast and coherent regimes across substantial fractions of subtropical, tropical and midlatitude oceans, such that aerosol-induced changes can be integrated to scales that are impactful for the regional and global radiative energy budget (Warren et al., 1988; Wood et al., 2015). Second, they often form in relatively pristine maritime environments characterized by low background aerosol loadings, where increases in aerosols can produce disproportionately large relative changes in cloud microphysics and radiative properties (Platnick and Twomey, 1994; Carslaw et al., 2013). Third, MLCs span a diverse range of optical depth: while persistent stratocumulus and mid-latitude cloud decks tend to have relatively high optical depth and hence exert strong cooling, a substantial subset—comprising broken trade-wind cumulus and transitional cloud scenes—is relatively optically thinner (Turner et al., 2007; Leahy

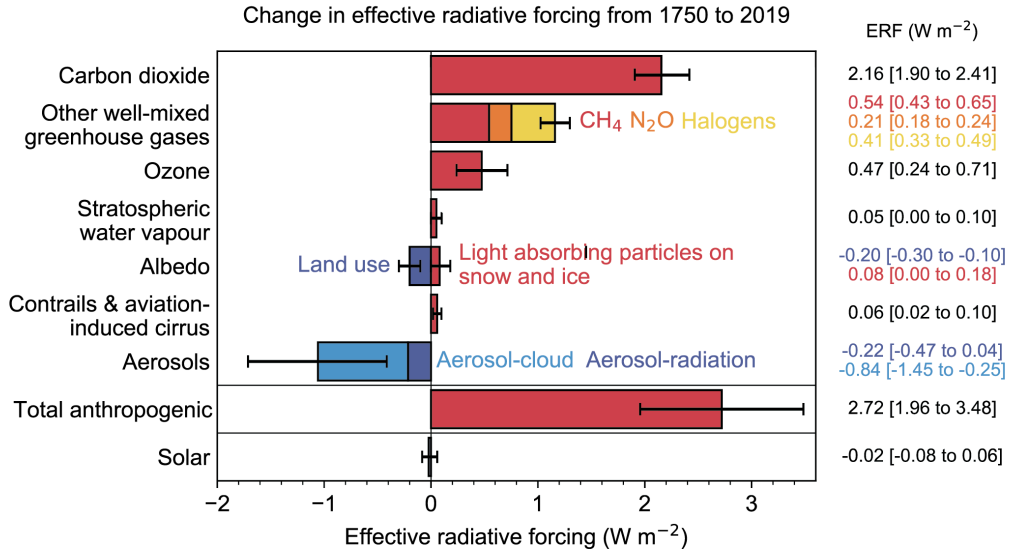


Figure 1.7: Change in effective radiative forcing from 1750 to 2019 for major contributing forcing agents. Bars show assessed best estimates with uncertainty ranges (W m^{-2}), including the aerosol contribution decomposed into aerosol-cloud and aerosol-radiation interactions. Reproduced from IPCC AR6 WG1 Chapter 7 (Fig. 7.6; Forster et al., 2021).

et al., 2012). Because α_c is less saturated in these optically thinner regimes, aerosol perturbations can more efficiently translate into changes in cloud reflectance. Together, it is essential to improve quantification of MLC response to aerosols and to better understand the underlying physical processes. The following subsection therefore details the fundamental physical mechanisms of ACI, beginning with the basic processes of MLC formation and the pathways through which aerosols modify and complexify them.

1.3.2 Microphysical pathway: the Twomey effect

Having introduced the radiative significance and climatic relevance of ACI, the following two subsections describe the physical mechanisms through which aerosols modify cloud properties, beginning at the microphysical scale. The most direct and fundamental pathway through which aerosols influence the Earth's radiative budget via clouds is the microphysical response—the change in cloud optical properties caused by aerosol-induced changes in N_d . Aerosol particles can act as CCN, and an increase in available CCN generally leads to an increase in N_d , provided that suitable thermodynamic conditions for activation are met (Ghan et al., 1998;

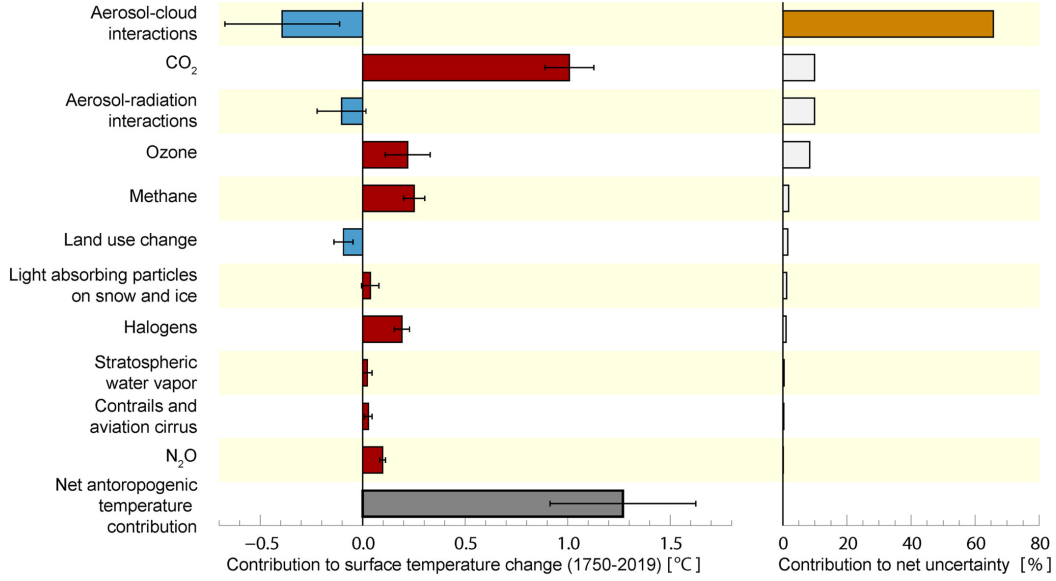


Figure 1.8: Contributions of major forcing agents to global-mean surface temperature change from 1750 to 2019 (left) and the percentage contribution to the net uncertainty in that change (right) Rows are ordered by decreasing relative contributions to uncertainty. Reproduced from Im et al. (2026), based on numbers from IPCC AR6 WG1 Chapter 7 (Forster et al., 2021).

Reutter et al., 2009; Fan et al., 2016). This is widely known as the *Twomey effect*.

Essentially a geometric phenomenon, the Twomey effect describes the relationship between cloud surface area and cloud brightness. For a given amount of cloud water, distributing the same amount of water over a larger population of droplets results in smaller droplets on average. This reduces r_e , thereby increasing the total surface area and the cloud becomes more reflective to incoming solar radiation i.e. higher τ_c and α_c . This is schematically illustrated by Fig. 1.9. In the cleaner environment, fewer aerosol particles activate into fewer and larger droplets, while in the polluted environment, more aerosols lead to more numerous and smaller droplets, making the cloud brighter.

The sensitivity of α_c to relative changes in N_d can be expressed as the cloud albedo susceptibility following Platnick and Twomey (1994) and Ackerman et al. (2000):

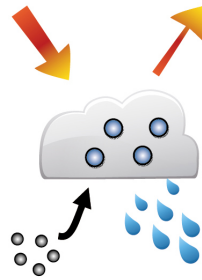
$$\frac{\partial \alpha_c}{\partial \ln N_d} = \frac{1}{3} \alpha_c (1 - \alpha_c) \quad (1.1)$$

which shows that α_c increases monotonically with N_d . It is expressed as a partial derivative so that other quantities, particularly LWP, are held constant (Quaas

et al., 2020). This formulation implies that the α_c sensitivity also depends on the background α_c itself. Specifically, the factor $\alpha_c(1 - \alpha_c)$ makes explicit that the sensitivity is maximized at intermediate α_c values. Physically, very bright clouds have little room to brighten further ($\alpha_c \rightarrow 1$, so $1 - \alpha_c \rightarrow 0$), while very thin clouds have little reflectance to begin with ($\alpha_c \rightarrow 0$). In both limits, $\alpha_c(1 - \alpha_c) \rightarrow 0$, and additional aerosol perturbations exert little Twomey effect. This is particularly relevant for MLCs, because although extensive stratocumulus decks can be near the high-albedo limit, broken trade cumulus and transitional MLCs frequently fall within the intermediate regime where the Twomey brightening is expected to be pronounced (Platnick and Twomey, 1994; Wood, 2012; Leahy et al., 2012).

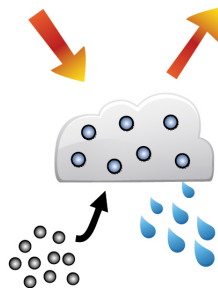
Aerosol-cloud interactions

(a)



Aerosols serve as cloud condensation nuclei upon which liquid droplets can form.

(b)



More aerosols result in a larger concentration of smaller droplets, leading to a brighter cloud. However there are many other possible aerosol-cloud-precipitation processes which may amplify or dampen this effect.

Figure 1.9: Schematic representation of aerosol–cloud interactions. (a) and (b) represent a marine low cloud under the clean and polluted condition, respectively (FAQ 7.2, Fig. 2; Boucher et al., 2013).

While this microphysical brightening pathway is conceptually straightforward

and well agreed to exert a net cooling effect on the climate system (Twomey, 1977; Boucher et al., 2013), its quantitative magnitude remains uncertain (Bellouin et al., 2020; Forster et al., 2021). This is because aerosol activation and the resulting N_d depend not solely on aerosol abundance, but also on aerosol composition and size distribution, updraft velocity, and the thermodynamic state of the cloud-forming environment, as discussed in Sect. 1.2.1 (Lohmann and Feichter, 2005; Seinfeld and Pandis, 2016; Lohmann et al., 2016). Moreover, at climate-relevant large scales, the quantification of the Twomey effect is complicated by the covariability of aerosol particles and updraft velocity, as well as by droplet sink processes (Gryspeerdt et al., 2017; Quaas et al., 2020).

The Twomey effect denotes the instantaneous microphysical response to an aerosol perturbation, prior to any subsequent changes in precipitation formation, entrainment, and in macrophysical properties LWP and cloud fraction (CF). The LWP and CF responses go beyond the instantaneous radiative effect itself and are generally referred to as cloud adjustments, which are discussed next.

1.3.3 Macrophysical pathway: cloud adjustments

Beyond the instantaneous microphysical brightening described by the Twomey effect, aerosol perturbations can also induce subsequent dynamical and other microphysical responses that alter cloud macrophysical properties, most importantly LWP and CF, and ultimately modify the CRE beyond the instantaneous Twomey effect. These are collectively referred to as cloud adjustments. These adjustments occur as the cloud system reacts to the modified droplet population and can either amplify or buffer the initial cooling exerted by the Twomey effect (Sherwood et al., 2015; Bellouin et al., 2020; Forster et al., 2021).

The primary cloud adjustment pathway is commonly associated with the cloud lifetime effect proposed by Albrecht (1989). As discussed in Sect. 1.2.2.2, drizzle formation depends inversely on N_d : aerosol-induced increases in N_d produce smaller droplets that are less efficient at initiating collision-coalescence, thereby suppressing drizzle and warm rain formation, allowing liquid water to accumulate within the cloud, leading to an increase in LWP and prolonging the cloud's lifetime, which results in a greater CF (Albrecht, 1989; Christensen et al., 2020). In this sense, clouds can become thicker and more spatially extensive, and hence this macrophysical response reinforces the initial Twomey effect to an additional negative radiative effect.

Smaller droplets can actually lead to a reduction in cloudiness. Smaller droplets have a higher surface-area-to-volume ratio and therefore evaporate more rapidly,

cooling the surrounding air, generating negative buoyancy that drives further turbulent mixing and entrainment of dry free-tropospheric air, ultimately causing cloud thinning or breakup (Jiang et al., 2006; Xue and Feingold, 2006; Small et al., 2009). Additionally, smaller droplets sediment more slowly, keeping more liquid water near the cloud top, enhancing the evaporative and radiative cooling at the cloud top, further increasing the entrainment (Ackerman et al., 2004; Bretherton et al., 2007; Michibata et al., 2016). As a result, the macrophysical pathway of ACI is inherently more complex than the Twomey effect, because multiple processes can amplify or dampen one another.

In more strongly precipitating cloud regimes, drizzle suppression can favour greater cloud amount and persistence, while in weakly precipitating or non-precipitating clouds, entrainment-related processes may dominate and lead to cloud thinning or dissipation (Xue and Feingold, 2006; Seifert et al., 2015; Christensen et al., 2020). Likewise, in transitional MLCs, aerosol perturbations may modify not only drizzle efficiency but also the timing of transitions between more overcast stratocumulus and more broken cumulus states, usually delaying the breakup of extensive cloud decks (Goren et al., 2019; Christensen et al., 2020; Diamond et al., 2022). Therefore, the magnitude and even the sign of cloud adjustments depend strongly on cloud regime and meteorological conditions, making them difficult to disentangle from meteorological covariability, retrieval errors, and modelling uncertainties. For this reason, cloud adjustments remain more uncertain than the Twomey effect and are a major source of uncertainty in aerosol effective radiative forcing (Bellouin et al., 2020; Forster et al., 2021). These challenges, and the specific motivation for focusing on the CF adjustment in this thesis, are discussed in the next section.

1.4 Cloud fraction adjustment: current state and challenges

Among the various components of ACI introduced above, the CF adjustment is a particularly important focus of this thesis. Because CF modulates the total area over which clouds exert their shortwave cooling effect, even modest fractional changes in cloud coverage translate into large radiative changes at the TOA (Zelinka et al., 2014; Rosenfeld et al., 2019). While earlier assessments often attributed a larger portion of the ERF_{aci} to the Twomey effect or to LWP adjustments (Zelinka et al., 2014; Ghan et al., 2016; Smith et al., 2020), more recent observational studies increasingly suggest that the CF adjustment may be the dominant contributor to the overall cooling effect (Bender et al., 2019; Chen et al., 2022b; Yuan et al., 2023;

Wall et al., 2023; Chen et al., 2024; Jia et al., 2024). At the same time, however, it remains one of the most difficult aerosol–cloud responses to quantify globally and robustly from observations, because underlying physical mechanisms depend on cloud regime, meteorological conditions, and the interplay between precipitation suppression, entrainment, and boundary-layer thermodynamics and because cloud and aerosol retrievals are themselves subject to cloud heterogeneity and scene-dependent satellite retrieval artefacts (Bellouin et al., 2020; Forster et al., 2021). This thesis therefore places the CF adjustment at its centre, examining it from both observational and modelling perspectives and using explainable machine learning (ML) as the common methodological thread. The following subsections briefly review the current state of knowledge and the key challenges in quantifying the CF adjustment from each perspective.

1.4.1 Observational quantification and challenges

Observational constraints on the CF adjustment broadly involve two complementary approaches. One approach uses opportunistic experiments (also known as natural experiments), such as ship tracks, volcanic eruptions and industrial emission changes. These provide a relatively clean causal signal because the aerosol perturbation source is well-defined and causality can be more confidently inferred (Coakley et al., 1987; Toll et al., 2019; Christensen et al., 2022). However, such cases are inherently limited in spatial or temporal representativeness. Ship tracks typically sample a narrow range of cloud regimes and often occur on sub-grid scales and volcanic events are episodic and geographically specific. Furthermore, opportunistic experiments are subject to selection bias, because signals such as ship tracks are typically only detectable when clouds are already present and in a state conducive to producing a visible perturbation, for example under particular LWP, precipitation, or stability conditions. In addition, for many opportunistic experiments, defining the appropriate unperturbed counterfactual remains challenging. These cases are therefore highly informative, but cannot by themselves straightforwardly constrain the global behaviour of the CF adjustment (Christensen et al., 2022).

The other approach is to use large-scale satellite observations, which provide the only way to observe aerosol–MLC interactions at the global scale and over climatologically meaningful periods. This is attractive because it enables near-global assessments and is therefore more suitable for constraining GCMs. A number of studies have used satellite data to explicitly or implicitly infer relationships between aerosol proxies and CF across different ocean basins and cloud regimes,

for example through meteorological stratification, mediation by N_d , cloud–aerosol pairing strategies, or more general statistical frameworks including ML (Quaas et al., 2008; Su et al., 2010; Chen et al., 2014; Gryspeerdt et al., 2016; Christensen et al., 2017; Andersen et al., 2017; Wall et al., 2023; Andersen et al., 2023; Jia et al., 2024). These studies have generally supported a positive aerosol–CF relationship in MLCs, especially in stratocumulus-to-cumulus transition regions, but they also show considerable spread in magnitude (Gryspeerdt et al., 2016; Andersen et al., 2017; Fuchs et al., 2018; Rosenfeld et al., 2019; Wall et al., 2022; Jia et al., 2024).

Among the available aerosol proxies, N_d is particularly suitable for observational ACI studies because it responds more directly to the availability of CCNs and is more closely linked to cloud microphysical and radiative properties than column aerosol optical depth, which is also sensitive to aerosol hygroscopic swelling and vertical structure (e.g. Gryspeerdt et al., 2017; Grosvenor et al., 2018). It is therefore widely used as a proxy for aerosol perturbations in observational studies of ACI.

The central challenge in such large-scale observational analyses is the covariability of aerosol, cloud, and meteorological factors. Disentangling the influence of aerosol perturbations on CF from the effects of meteorology is particularly difficult, because the same environmental conditions that favour cloud development also affect aerosol abundance, aerosol hygroscopic swelling, retrieval quality, and the derived N_d (Andersen et al., 2016; Gryspeerdt et al., 2019; Bellouin et al., 2020). Failing to account for these meteorological covariates can significantly bias the inferred aerosol signal, either artificially inflating or weakening the estimated CF sensitivity (Gryspeerdt et al., 2016). In addition, satellite retrieval artefacts can produce non-causal positive relationships. For example, N_d retrievals tend to be less reliable in heterogeneous, broken-cloud regimes, and N_d and CF derived from the same satellite scene are not fully independent, which can bias their apparent relationship high (Loeb and Schuster, 2008; Grosvenor et al., 2018; Arola et al., 2022; Jia et al., 2024). These issues, detailed in Sect. 3.2, imply that simple correlation-based estimates are inadequate.

These limitations motivate the application of advanced statistical and ML methods that can account for the influence and covariability of meteorology to large-scale observations. In this thesis, an explainable ML framework is used for this purpose. Rather than relying only on direct correlations between aerosol and cloud properties, such an approach learns the joint relationship between CF and both an aerosol proxy and environmental predictors.

1.4.2 Model-based quantification and challenges

From the modelling perspective, the standard approach to quantifying the CF adjustment is to compare paired simulations, most commonly between PI and present-day (PD) conditions, in which aerosol or precursor emissions are varied while other boundary conditions are held fixed, and its contribution is then diagnosed as part of the aerosol ERF (Lohmann and Feichter, 2005; Ghan, 2013; Boucher et al., 2013; Neubauer et al., 2019; Smith et al., 2020; Zelinka et al., 2023). Within this general framework, the exact model setups can vary, for example in whether the ocean is fully coupled or SSTs are prescribed, whether a single forcing agent is perturbed in isolation or all aerosol emissions are changed simultaneously (Ghan, 2013; Smith et al., 2020), and whether the large-scale circulation is freely evolving or nudged toward reanalysis data (Kooperman et al., 2012; Zhang et al., 2014). In addition, some studies use more idealized perturbation experiments in which, for example, simplified or artificial changes are imposed on aerosol, CCN, or cloud-relevant quantities (Gettelman et al., 2013; Gettelman, 2015). Despite these differences, these approaches share the same underlying logic: the aerosol influence is inferred from the difference between a perturbed simulation and a corresponding reference state.

GCM-based estimates generally support a positive CF response to aerosol perturbations, but the magnitude differs. In many GCMs the contribution of the CF adjustment to the total aerosol forcing is relatively weak compared with the Twomey effect and the LWP adjustment (Zelinka et al., 2014; Ghan et al., 2016; Smith et al., 2020; Zelinka et al., 2023). By contrast, recent observational studies increasingly suggest that aerosol-induced CF changes may be dominant for the forcing associated with MLCs (Chen et al., 2022b; Wall et al., 2023; Yuan et al., 2023; Chen et al., 2024), indicating that the CF response may be underestimated in GCMs (Wang et al., 2025).

A major difficulty in modelling the CF adjustment is that the key processes involved, including droplet activation, autoconversion, entrainment, and boundary-layer turbulence, occur on subgrid scales that are unresolved by typical GCM coarse grids and therefore must be parameterized. Furthermore, cloud cover itself is not a directly prognosed quantity in most GCMs, but is usually diagnosed from grid-mean RH and, in some cases, also LWC, which are modulated by the representation of the subgrid-scale processes. As a result, the simulated CF response depends not only on aerosol emissions and activation, but also on structural model choices, such as how cloud cover, autoconversion, and related boundary-layer processes are represented (Bellouin et al., 2020; Fons et al., 2024; Jia et al., 2026), meaning that

both the magnitude and the regime dependence of the CF adjustment can vary substantially across models and model configurations (Gryspeerd et al., 2020; Wang et al., 2025). The representation of these processes is introduced in Sect. 2.2.

These issues also complicate model–observation comparison. Observational estimates of the CF response are usually inferred from PD states, whereas models have often been evaluated through diagnostics from paired perturbation experiments. The two approaches therefore do not constrain the CF response exactly in the same context, which complicates a direct comparison. In this thesis, the model-based analysis complements the observational perspective by applying a consistent explainable ML framework to GCM output, in order to examine how models represent the relationship between aerosols and CF.

1.5 Aims, research questions and outline

As outlined in the previous sections, the CF response of MLCs to aerosol perturbations is radiatively important but remains difficult to constrain. The main challenges lie in: (i) from the observational perspective, the strong covariability of aerosol, cloud, and meteorological fields, which complicates the isolation of the aerosol-related signal; and (ii) from the modelling perspective, discrepancies between observation-based and model-based estimates, arising from incomplete representation of relevant subgrid processes in GCMs and the influence of model configuration choices. Against this background, this thesis aims to improve the quantification and physical understanding of how CF in MLCs responds to aerosol perturbations in both observations and models, and to compare these two perspectives. To tackle the challenges, fill the knowledge gaps and achieve this aim, two overarching research questions (RQs) are raised, and the thesis is therefore organized into an observation-based part and a model-based part:

RQ 1: How does MLC CF respond to aerosol perturbations globally, and how is this response influenced by meteorological conditions?

RQ 2: How does the CF response to aerosols emerge in a GCM, and how do model configuration choices affect this response?

To address these two overarching research questions, this thesis pursues two main objectives: (i) to use an explainable ML framework to quantify the relationship between N_d and CF from near-global satellite observations while explicitly accounting for meteorological covariability; and (ii) to apply the same framework to GCM output, enabling a consistent comparison with the observation-based results

and an assessment of how model-configuration choices influence the simulated CF response. To this end, the explainable ML framework is first established using near-global MODerate Resolution Imaging Spectroradiometer (MODIS) observations and fifth generation European Centre for Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis (ERA5) reanalysis meteorology, and is then applied to the advanced GCM ICOSahedral Nonhydrostatic (ICON)-Hamburg Aerosol Module (HAM):

Study 1: Observation-based CF sensitivity and its meteorological controls

The first part of the thesis addresses the observational quantification of aerosol-induced CF changes. Although large-scale satellite studies generally support a positive aerosol–CF relationship in MLCs, substantial spread remains in the reported magnitude, and the role of meteorological covariability remains a central challenge. Simple correlation analyses between aerosol proxies and CF are susceptible to confounding and do not readily characterize how the N_d –CF relationship itself varies with the ambient environment. Against this background, the following specific research questions are addressed in this part:

RQ 1.1: What are the magnitude and direction of MLC CF sensitivity to N_d (aerosol proxy) while accounting for meteorological factors?

RQ 1.2: Which meteorological variables most strongly control MLC CF, and how do meteorological variables modulate the relationship between N_d and CF?

These questions motivate the following **guiding hypotheses** regarding the physical behaviour of the system based on the physical understanding of MLC controls and ACI outlined in the preceding sections: (i) the CF response in MLCs to aerosol perturbations is positive but modulated—reduced or amplified—by ambient meteorology; (ii) CF sensitivities to $\ln N_d$ and meteorological variables, as well as the meteorological modulation of the $\ln N_d$ –CF sensitivity, exhibit distinct regional patterns.

To address these questions, the first study establishes regional eXtreme Gradient Boosting (XGBoost) models trained on nine years (2011–2019) of daily MODIS satellite products and ERA5 reanalysis data over near-global oceans, using N_d as a proxy for cloud-active aerosol. Regional XGBoost models predict CF from N_d and a set of thermodynamic and dynamical meteorological predictors. SHapley Additive exPlanations (SHAP) values are then used to explain the regional XGBoost

models and to quantify the sensitivity of CF to each predictor, and SHAP interaction values are used to characterize how meteorological conditions modulate the $\ln N_d$ –CF relationship. The data, explainable ML framework setup, and the results are presented in Chap. 3.

Study 2: Model-based CF sensitivity and the role of model configuration choices

The second part of the thesis extends the explainable ML framework established in Study 1 to ICON-HAM simulations to enable a consistent comparison between observation-based and model-based estimates of the CF response. In addition to the meteorological and statistical challenges also present in the observational case, model-based estimates may further be subject to structural choices in the parameterized representation of cloud cover and in values of prescribed lower bound for N_d ($N_{d,\min}$), both of which can modulate the pathway through which aerosol perturbations influence the diagnosed CF. In light of these challenges, the following specific research questions are addressed in this part:

RQ 2.1: To what extent can the explainable ML framework developed for observations be transferred consistently to ICON-HAM output, and how do the resulting model-based $\ln N_d$ –CF sensitivities compare with satellite-based estimates?

RQ 2.2: How do model configuration choices, specifically the cloud-cover parameterization schemes and different values of $N_{d,\min}$, shape the simulated $\ln N_d$ –CF relationship?

These questions motivate the following **guiding hypotheses** regarding the behaviour of the simulated system and its dependence on model configuration: (i) the $\ln N_d$ –CF relationships diagnosed from ICON-HAM differ quantitatively, and possibly even qualitatively, from the MODIS-based relationships; and (ii) the RH- and LWC-dependent cloud-cover parameterization scheme (CC-RH-LWC) exhibits larger $\ln N_d$ –CF sensitivities than the RH-only scheme (CC-RH), with $N_{d,\min}$ jointly modulating these sensitivities differently in each scheme.

To address these questions, the same XGBoost–SHAP framework established in Study 1 is applied to the ICON-HAM output, and the diagnosed $\ln N_d$ –CF sensitivities are compared with the observational benchmark from Chapter 3. The explainable ML framework is extended with a piecewise linear regression (PLR) approach to accommodate the stronger nonlinearity of the $\ln N_d$ –CF relationship in ICON-HAM. Furthermore, the analysis also investigates the role of differences

in the cloud-cover parameterization and in $N_{d,min}$. Six AMIP (atmospheric-only) simulations are conducted that combine the two cloud-cover schemes (CC-RH and CC-RH-LWC) with three different values of $N_{d,min}$ (10, 40, and 100 cm^{-3}). The data, ICON-HAM and experimental setup, and results are presented in Chapter 4.

Thesis outline

The structure of this thesis is summarized schematically in Fig. 1.10. Chapter 1 (this chapter) introduces the state of the art and the scientific basis, including the Earth’s energy budget, the role of MLCs in the climate system, ACI and the CF adjustment, and the observational and modelling challenges in constraining the CF response to aerosol perturbations.

Chapter 2 establishes the methodological framework used throughout the thesis. It provides details on the MODIS satellite products and ERA5 reanalysis data, the retrieval of key variables such as N_d , the representation of main variables and processes related to the CF adjustment in ICON-HAM, and an overview of the ML methods in atmospheric sciences with a technical focus on XGBoost and SHAP.

Chapter 3 presents the observation-based analysis (Study 1), which addresses **RQ 1** through the specific sub-questions **RQ 1.1** and **RQ 1.2**. Chapter 4 presents the ICON-HAM-based analysis (Study 2), which addresses **RQ 2** through **RQ 2.1** and **RQ 2.2**. Finally, Chapter 5 provides a concluding synthesis and discussion. It integrates the findings from both studies, examines their implications for constraining the CF adjustment, and outlines directions for future research.

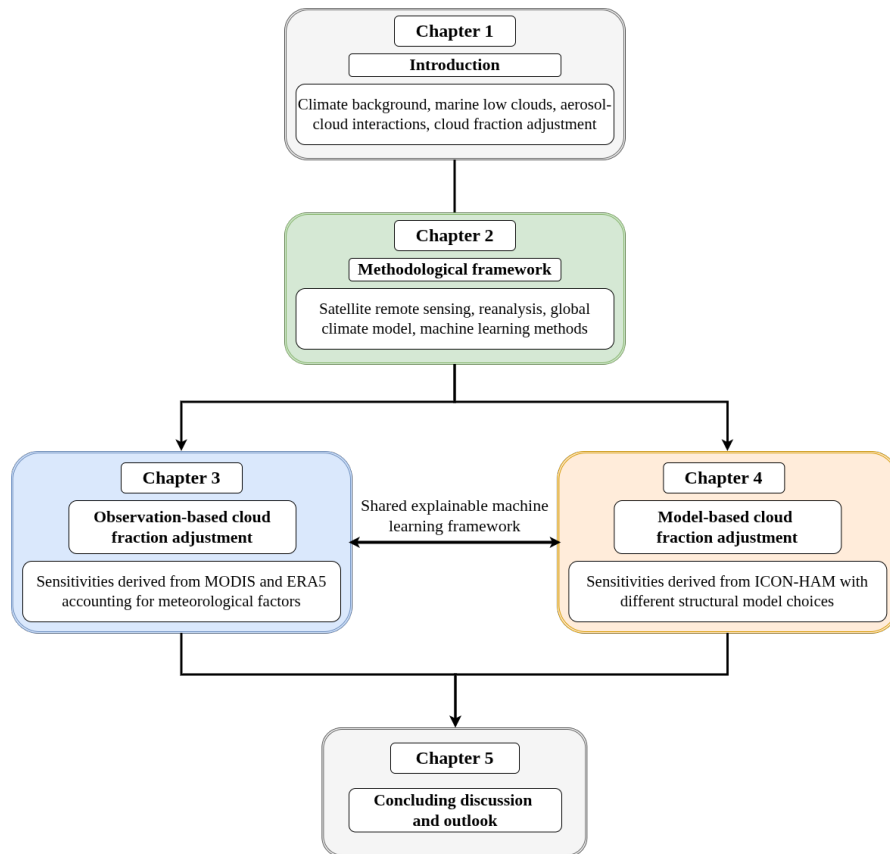


Figure 1.10: Schematic overview of the thesis structure. Both studies (Chaps. 3 and 4) share the methodological introduction in Chap. 2 and are linked by the consistent application of the explainable ML framework to satellite observations and GCM output, respectively. Their findings are synthesized in Chap. 5.

Chapter 2

Data and methods for quantifying cloud fraction adjustments

2.1 Satellite remote sensing and reanalysis

This thesis utilizes two complementary datasets: satellite remote sensing data from MODerate Resolution Imaging Spectroradiometer (MODIS; Platnick et al. 2017), providing global cloud macrophysical and microphysical properties, and meteorological fields from the ERA5, which characterize the large-scale dynamical and thermodynamic environment. The following subsections describe these data sources in turn, from the broader satellite context to the specific products and retrieval algorithms.

2.1.1 Spaceborne radiometry with MODIS

Satellite remote sensing refers to the measurement of the atmospheric and surface properties by detecting electromagnetic radiation emitted, scattered or reflected by the Earth system. It is typically performed from a distance using sensors mounted on spacecraft. Satellite observations provide indispensable information on MLC ACI since they are the only observing system that simultaneously captures both macrophysical and microphysical MLC properties at the global scale and over climatologically meaningful time periods, particularly over vast and remote oceanic regions where in situ measurements are sparse (Minnett et al., 2019; Stier et al., 2024; Im et al., 2026). Neither field campaigns—which deliver process-level detail but are inherently limited in spatial extent and duration—nor ground-based remote sensing sites—which build long time series but sample at fixed locations—can provide spatially extensive and frequently repeated measurements offered by

satellites over much of the global ocean. In this regard, satellite remote sensing is necessary for constraining the cloud fraction adjustment and is complementary to laboratory and field experiments (Rosenfeld et al., 2023; Gordon et al., 2023; Chen et al., 2024; Li et al., 2025).

Spaceborne sensors can be broadly classified into two categories according to their measurement principle. *Active* sensors, such as the 94 GHz cloud profiling radar aboard CloudSat (Stephens et al., 2008), the lidar aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) (Winker et al., 2009), and the combined lidar and radar instruments on the Earth Cloud, Aerosol and Radiation Explorer (EarthCARE) (Wehr et al., 2023). These active sensors emit their own radiation pulses and detect the backscattered return signal, enabling vertically resolved profiling of cloud and aerosol layers with relatively high detail, but only along narrow nadir tracks with more complex and expensive instrumentation (Stephens et al., 2002; Li et al., 2022a; Cairo et al., 2024).

Passive sensors, by contrast, detect naturally available radiation, either reflected and scattered solar radiation or emitted thermal radiation. They therefore infer cloud properties by measuring radiances across the solar and thermal spectra without emitting any signal of their own (Stephens and Kummerow, 2007; Rosenfeld et al., 2014; Bühl et al., 2017). Compared with active sensors, passive sensors trade vertical profiling for broad swaths, near-global coverage and high revisit frequency, making them well suited to the climatological and statistical studies of global ACI (Stubenrauch et al., 2013; Rosenfeld et al., 2014, 2023).

Satellite orbits provide a fundamental trade-off between temporal continuity and spatial coverage and resolution. Geostationary satellites maintain a fixed view of a given domain and provide continuous high-frequency monitoring, but from an altitude of approximately 36 000 km, resulting in coarser spatial resolution, and only a constellation of such satellites can achieve near-global coverage (Seinfeld et al., 2016; Letu et al., 2020). Polar-orbiting satellites, by contrast, circle the Earth at much lower altitudes (typically 700 km to 800 km), and therefore achieve finer spatial resolution and near-global coverage at the cost of less frequent (near-daily) spatial sampling at any given location (Kidder and Haar, 1995; Stubenrauch et al., 2013).

MODIS is a passive sensor on a polar-orbiting platform with a sun-synchronous 705 km orbit height (Stephens et al., 2002; King et al., 2003). It is essential for building the large and geographically diverse cloud-scene populations required for robust ACI analysis (Platnick et al., 2017; Forster et al., 2021). MODIS measures in 36 spectral bands/channels spanning a wide spectral range from 0.4 μm to 14.4 μm

with a swath width of approximately 2330 km and nadir resolutions of 250 m, 500 m, or 1 km depending on the band (King et al., 2003; Levy et al., 2013; Platnick et al., 2017; Angal et al., 2023). The near-global daily coverage by successive orbital swaths are illustrated in Fig. 2.1.

MODIS is flown on two platforms: *Terra* with a descending equatorial overpass at approximately 10:30 local solar time, and *Aqua* with an ascending equatorial overpass at approximately 13:30 local solar time (King et al., 2003; Platnick et al., 2003). The corresponding global daytime overpass times in UTC are shown in Fig. 2.1, which also highlights the swath patterns of these polar-orbiting platforms. While *Aqua* is part of the A-Train constellation, a coordinated series of satellites enabling near-simultaneous multi-sensor sampling (Stephens et al., 2002), *Terra* flies independently. The observational part of this thesis uses data exclusively from *Terra*, ensuring temporally consistent daytime sampling from a single overpass time for both cloud retrievals and the derived N_d . This choice is also physically relevant because MLCs over many marine stratocumulus regions tend to be closer to their morning maximum in coverage and thickness at 10:30 than 13:30 (Rozendaal et al., 1995; Wood et al., 2002; Wood, 2012). Furthermore, this captures the cloud state set by nighttime processes, which have been suggested to strongly shape the CF response to aerosol (Pugsley et al., 2025). The specific Level-3 cloud products retrieved from *Terra* and the derivation of N_d from them are described in Sect. 2.1.2 and Sect. 2.1.3, respectively.

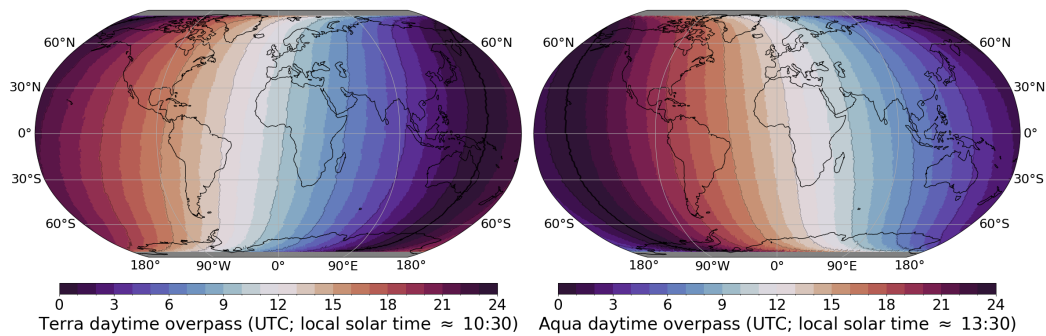


Figure 2.1: Global distribution of daytime MODIS overpass times in UTC for *Terra* (left) and *Aqua* (right). The diagonal stripes indicate individual orbital swaths and illustrate the near-global daily coverage of the polar-orbiting platforms.

2.1.2 MODIS Level-3 cloud products

MODIS data products are organized into a processing hierarchy of different levels. Level-0 and Level-1 products contain raw and calibrated radiances, respectively.

Level-2 products provide pixel-level geophysical retrievals at a 1 km nadir resolution, generated for each orbital swath. Level-3 products are derived by aggregating Level-2 retrievals onto a regular $1^\circ \times 1^\circ$ global grid, providing daily, monthly, and statistical summaries that are more suitable for large-scale statistical analyses (King et al., 2003; Platnick et al., 2017). This thesis uses the Terra Level-3 daily gridded product (MOD08_D3, Collection 6.1) for the period 2011–2019, spanning 60° N to 60° S.

MODIS cloud products are derived using established cloud-mask, cloud-phase, and cloud-property retrieval algorithms. The observational part of this thesis (Chap. 3) focuses specifically on single-layer warm cloud products, which target MLCs and exclude overlying ice clouds, multi-layer scenes and deep convective clouds. The single-layer and liquid conditions are identified using “1L” (single-layer) and “Liquid” designations, which rely on algorithmic discrimination applied at the Level-2 retrieval stage. The MODIS cloud thermodynamic-phase algorithm uses infrared and shortwave-infrared bands to distinguish liquid and ice phases, while multi-layer cloud detection additionally exploits water-vapour absorption bands to identify overlapping layers (Wind et al., 2010; Platnick et al., 2017; Marchant et al., 2020). To obtain a more conservative warm-phase dataset, an additional threshold of $CTT > 268\text{K}$ is applied to exclude cold-liquid and mixed-phase clouds, which also helps ensure more reliable N_d retrievals (Sect. 2.1.3).

The primary MODIS products used in this thesis are summarized below:

- **Cloud fraction:** Level-3 CF product (Cloud_Retrieval_Fraction_1L_Liquid) is defined as the fraction of pixels identified as cloudy within each $1^\circ \times 1^\circ$ grid cell. Pixel-level cloud detection is performed by the MODIS cloud mask algorithm, a Level-2 product that applies a series of spectral and spatial threshold tests across multiple channels, assigning each pixel a confidence of clear, probably clear, uncertain, or cloudy (Ackerman et al., 1998; Frey et al., 2008). CF serves as the predictand of the machine-learning framework in Chap. 3.
- **Cloud optical properties:** τ_c (Cloud_Optical_Thickness_1L_Liquid_Mean) and r_e (Cloud_Effective_Radius_1L_Liquid_Mean) are retrieved using a bispectral method that jointly derives both quantities from two channels: a non-absorbing visible or near-infrared channel whose reflectance is primarily sensitive to τ_c , and a water-absorbing shortwave-infrared channel whose reflectance is primarily sensitive to r_e (Nakajima and King, 1990; Platnick et al., 2017). τ_c and r_e form the foundation for the derivation of N_d (Sect. 2.1.3 and Sect. 3.2.1).

- **Cloud-top products:** The main cloud-top variable used in this thesis is CTT. It is retrieved from the thermal infrared radiation emitted by clouds, with the observed TOA radiance converted to temperature based on cloud altitude (Menzel et al., 2008; Baum et al., 2012). CTT is used both to further restrict the dataset to MLCs and to improve the reliability of N_d retrievals (Sect. 2.1.3 and Chap. 3).
- **Satellite viewing geometry:** Illumination and viewing conditions can influence retrieval quality. Therefore, filtering based on solar zenith angle and satellite zenith angle can help reduce geometrical biases and retrieval artefacts in N_d (Sect. 2.1.3 and Chap. 3).

Because MOD08_D3 variables are aggregated daily on a $1^\circ \times 1^\circ$ grid, they are well suited for collocation with large-scale meteorological fields from ERA5 described in Sect. 2.1.4.

2.1.3 Retrieval of Cloud Droplet Number Concentration

MODIS does not measure N_d directly; instead, it is derived from the retrieved τ_c and r_e . This derivation is fundamentally based on the adiabatic assumption, under which LWC is assumed to increase monotonically from the cloud base to the cloud top within a vertically homogeneous, adiabatic layer with constant N_d , following Quaas et al. (2006):

$$N_d = \alpha \tau_c^{0.5} r_e^{-2.5} \quad (2.1)$$

This is the formula used to calculate N_d in this thesis, where $\alpha = 1.37 \times 10^{-5} \text{ m}^{-0.5}$. This coefficient is obtained by treating several parameters as constants together with physical constants (Brenguier et al., 2000; Quaas et al., 2006). Specifically, the density of liquid water ρ_w , extinction efficiency Q_{ext} , adiabatic condensation rate c_w and the droplet spectral-shape factor k which relates volume-mean radius to effective radius. Eq. (2.1) can therefore also be expressed in the more explicit form (Grosvenor et al., 2018; Gryspeerd et al., 2022):

$$N_d = \frac{\sqrt{5}}{2\pi k} \left(\frac{f_{\text{ad}} c_w \tau_c}{Q_{\text{ext}} \rho_w r_e^5} \right)^{1/2} \quad (2.2)$$

where f_{ad} is the sub-adiabatic fraction, describing the reduction in the condensation rate due to mixing with subsaturated air. In our study, $f_{\text{ad}} = 1$ is used (i.e. fully adiabatic). This approach provides a practical way to estimate N_d from satellite

retrievals of τ_c and r_e . Grosvenor et al. (2018) further discussed the physical assumptions, uncertainties and best practices for satellite N_d retrievals. Gryspeerdt et al. (2022) showed that the estimated N_d depends heavily on the applied sampling and filtering strategy. In the observational part of this thesis (Chap. 3), several filtering conditions are applied to obtain more reliable retrievals, including improved validity of the adiabatic assumption and higher-quality τ_c and r_e retrievals. These filters are summarized by Gryspeerdt et al. (2022) and originate mainly from Quaas et al. (2006) and Grosvenor et al. (2018).

Beyond the basic requirement of using single-layer liquid-phase MODIS products with CTT > 268 K, the following criteria are used:

- **Optical thickness:** Only samples with $\tau_c > 4$ are retained because optically thin clouds are more susceptible to retrieval biases and may not be sufficiently thick for liquid water to be related to τ_c and r_e through the underlying adiabatic assumption (Nakajima and King, 1990; Quaas et al., 2006).
- **Droplet size:** Only cases with $r_e > 4 \mu\text{m}$ are kept, since very small retrieved droplet sizes are less likely for liquid-phase MLCs and can be associated with problematic retrievals (Nakajima and King, 1990; Quaas et al., 2006).
- **Viewing geometry:** Cases with solar zenith exceeding 65° and satellite zenith exceeding 55° are excluded because large illumination and viewing angles increase 3D radiative effects, shadowing, and multiple scattering (Maddux et al., 2010; Grosvenor et al., 2018).

The threshold of CF > 0.9 in Gryspeerdt et al. (2022) is not applied here, although it ensures more homogeneous and adiabatic clouds. This is because this thesis explicitly analyzes N_d –CF relationship, restricting to only CF > 0.9 would exclude a large part of the CF variability and bias the results towards near-overcast scenes.

A caveat in this thesis is that N_d is computed using $1^\circ \times 1^\circ$ Level-3 mean τ_c and r_e . Due to the nonlinearity of Eq. (2.1), calculating N_d from grid-box mean values rather than underlying pixel-level joint histograms may introduce additional biases. This limitation should be kept in mind when interpreting the results, and the potential biases are acknowledged and discussed in Sect. 3.2.3.3. This bias is expected to be small relative to the large-scale dynamical variability captured by the machine-learning framework, but quantifying its impact on N_d –CF sensitivities is left for future work. N_d retrieval biases and their effects on the quantification of CF response to aerosols are also discussed in Sect. 3.2.1 and Sect. 3.2.3.3.

2.1.4 ERA5 meteorological reanalysis

Large-scale thermodynamic and dynamical factors covary with aerosol loading and cloud properties. To account for this, this thesis uses meteorological fields from ERA5 to provide the necessary environmental context discussed in Sect. 1.2.3, since many key controls on MLCs, such as EIS and vertical motion, are important yet cannot be directly obtained from satellite observations alone. ERA5 therefore complements MODIS retrievals by supplying dynamically and thermodynamically relevant variables on a continuous spatiotemporal grid, enabling a joint analysis of cloud properties and their meteorological environment (Hersbach et al., 2020).

Reanalysis provides a spatially and temporally complete description of the global atmosphere by assimilating a broad range of historical observations, including radiosondes, aircraft, satellite data and surface stations, into a numerical weather prediction model, thereby producing meteorological fields that are physically consistent with one another. In this way, ERA5 provides global meteorological fields at hourly temporal resolution and $0.25^\circ \times 0.25^\circ$ horizontal spatial resolution from 1940 to present. It contains both single-level variables, which represent surface or near-surface conditions, and pressure-level variables, which describe the three-dimensional state of the atmosphere across 137 native model vertical levels, corresponding to 37 pressure levels from 1000 hPa to 1 hPa (Hersbach et al., 2020).

A critical technical requirement in this thesis is the harmonization of ERA5 datasets with $1^\circ \times 1^\circ$ Level-3 MODIS products, because the satellite data represent a daytime snapshot near 10:30 AM local solar time. Therefore, the native $0.25^\circ \times 0.25^\circ$ ERA5 grids are regridded to $1^\circ \times 1^\circ$ using bilinear interpolation. Furthermore, the hourly ERA5 data are collocated to match the UTC overpass times of Terra MODIS (illustrated in Fig. 2.1a) for each grid cell. In this way, we obtain daily datasets where the meteorological state represents the environment in which the clouds are observed. This common gridded framework facilitates a direct comparison between cloud properties and meteorological predictors at the large scale considered in this thesis.

As with any reanalysis product, ERA5 does not directly observe the atmosphere but represents the best estimate of the atmospheric state given the underlying model, data assimilation system, and available observations. Therefore, uncertainties remain, particularly for variables that are more subject to model-dependent biases or are indirectly constrained by observations. In the context of the MBL, for example, partly parameterized boundary-layer and convective processes can introduce additional uncertainty (Belmonte Rivas and Stoffelen, 2019; Hersbach et al., 2020). Nevertheless, ERA5 provides a state-of-the-art, physically coherent

meteorological dataset which are validated against independent observations. Its combination of practically high resolution, a multi-decadal record, and global coverage makes it well suited for the large-scale statistical analyses in this thesis.

2.2 Global climate modelling

In this thesis, GCMs refer to the dynamical models which numerically resolve the Navier-Stokes equations, which simulate global climate at resolutions from 10 km to 100 km; using such GCMs to simulate clouds and ACI is not easy due to the variety of spatial and temporal scales (Quaas and Gryspeerdt, 2022). Variations in model configurations and assumptions across studies, e.g. how they treat oxidant evolution, can lead to variations in the CF responses to aerosols. However, GCMs are important for the research of ACI on large-scales and offer a complementary global perspective on ACI that is not accessible from satellite remote sensing. GCMs can provide a physically consistent framework in which clouds, aerosols, radiation, and meteorology interact simultaneously. They provide a spatiotemporally representative framework that allows for the exploration of different climate states and the quantification of radiative forcing and future projections across various scenarios. In addition, they make it possible to examine ACI and the CF adjustment within a controlled modelling framework and to test how different process representations affect the simulated cloud response.

Unlike satellite observations, which are subject to retrieval uncertainties arising from assumptions such as plane-parallel geometry and the adiabatic cloud model, GCMs have uncertainties arising from the model formulation itself, including sensitivity to initial conditions and internal variability, uncertain input data and parameters, structural representations or omissions of key processes, parameterizations of unresolved subgrid-scale aerosol and cloud processes such as autoconversion, droplet activation and turbulence, coarse spatial resolution that cannot resolve individual cloud structures, and biases in the simulated large-scale meteorological environment (Carslaw et al., 2013; Mülmenstädt and Feingold, 2018; Gordon et al., 2023). All of these can propagate into the simulated CF adjustment signal and the large-scale meteorological environment of clouds. In the following, some parameterizations that are important for the representation of ACI and the CF adjustment are introduced.

2.2.1 Parameterization of cloud cover

In GCMs, the Navier-Stokes equations are solved at the model grid scale, while processes occurring below this scale (e.g. turbulence, convection, and cloud micro-physics) cannot be expressed and resolved by the fundamental equations; parameterizations are needed which refer to the empirical representation of unresolved subgrid-scale processes as functions of the resolved model variables on the grid scale. In the context of MLCs, many relevant processes and their sensitivity to aerosols are subgrid and thus parameterizations are needed.

To investigate the CF adjustment in GCMs, it is first necessary to consider how CF is represented in these models. In the real world, clouds form when RH reaches or exceeds 100 %, leading to saturation. However, in GCMs, clouds cannot be binarily represented as cloudy or clear like a cloud mask because their horizontal resolution is coarse and thus the grid-box mean vertical motions are rarely strong enough to saturate an entire grid cell. For this reason, GCMs must use fractional cloud coverage, cloud cover, or CF; these terms are used interchangeably in this thesis.

This is achieved by assuming subgrid-scale variability of humidity, often utilizing the total-water specific humidity (q_t). In this way, the physical basis is that even if the grid-cell mean total-water specific humidity ($\bar{q}_t$) is below the grid-mean saturation specific humidity ($\bar{q}_s$), part of subgrid-scale regions can locally exceed saturation. A common way to represent the fluctuations of q_t within a grid cell is by assuming a probability density function of q_t , denoted as $\mathcal{P}(q_t)$. As an illustrative example, if a uniform distribution of width $2\Delta q$ centered at ($\bar{q}_t$) is assumed, the CF is diagnosed as the part of the distribution exceeding the saturation-specific humidity q_s (Quaas and Gryspeerdt, 2022):

$$\text{CF} = \int_{q_s}^{\infty} \mathcal{P}(q_t) dq_t = \frac{\bar{q}_t + \Delta q - \bar{q}_s}{2\Delta q}. \quad (2.3)$$

Again, in this framework, cloud forms before the grid-box mean state itself becomes fully saturated. Furthermore, this framework inherently creates a threshold for cloud formation. Clouds begin to form once the most humid subgrid-scale region inside the grid cell exceeds saturation i.e. $\bar{q}_t + \Delta q > \bar{q}_s$, corresponding to $\text{CF} > 0$ according to Eq. 2.3. Since $\frac{\bar{q}_t}{\bar{q}_s}$ defines the grid-mean RH ($\overline{\text{RH}}$), the onset condition becomes $\overline{\text{RH}} > 1 - \frac{\Delta q}{\bar{q}_s}$. By parameterizing the distribution width as $\Delta q = \gamma \bar{q}_s$ with a scaling parameter γ , the critical RH at which clouds first trigger is $1 - \gamma$ (Quaas and Gryspeerdt, 2022).

In this sense, we can see that the CF can be more practically parameterized as a

function of grid-mean RH together with a critical threshold, instead of explicitly formulating CF in terms of $\bar{q}_t$, $\bar{q}_t$ and an assumed subgrid-scale probability density function of moisture. This kind of parameterization scheme diagnoses CF using only humidity without microphysical information, so the CF response to aerosol perturbations is only indirectly through humidity changes, tending to mute this response. This kind of schemes have already been applied in many GCMs such as UKESM and ECHAM (Wang et al., 2025). The one used in ECHAM, which is one of the cloud-cover parameterization schemes that we examined in Chap. 4, called CC-RH (Eq. 4.1 therein) (Sundqvist et al., 1989), uses this approach with the critical grid-mean RH specified empirically as a function of height. While another alternative class of schemes parameterizes liquid cloud cover in terms of LWC additionally, which is referred to as CC-RH-LWC (Eq. 4.2). As will be explored in Chap. 4, this difference is directly relevant for ACI studies because it allows aerosol-induced changes in autoconversion and LWP to feed back directly onto CF, and it influences how changes in droplet number and precipitation formation can translate into changes in cloud cover.

2.2.2 Parameterization of microphysics

Cloud microphysical processes in atmospheric models are commonly represented either with spectral-bin schemes or with bulk schemes (Khain et al., 2015; Morrison et al., 2020). Spectral-bin schemes explicitly discretize the droplet size distribution into numerous size bins and resolve the transfer between them due to the condensation and collision-coalescence growth, which operates closer to the underlying physical processes, but is too computationally expensive at global scales (Lohmann and Feichter, 2005; Khain et al., 2015). So they are used mainly in process models such as large-eddy simulations. By contrast, bulk schemes represent the size distribution analytically and categorize hydrometeor classes by size and phase, e.g. cloud liquid and rain, and simulate selected moments of their generalized size distributions rather than tracking individual size bins (Lohmann and Feichter, 2005; Morrison et al., 2020).

Within bulk microphysics, schemes are generally divided into single-moment and two-moment approaches based on how many prognostic variables they predict (Seifert and Beheng, 2006). Single-moment schemes typically predict only the specific mass of a hydrometeor class (e.g. total cloud liquid water), whereas two-moment schemes additionally prognose number concentration (Morrison et al., 2005; Morrison and Gettelman, 2008). To represent the underlying droplet population, these schemes assume a specific form for the droplet size distribution e.g.

a Gamma distribution. The model then predicts the corresponding moments by solving continuity equations that include sources and sinks such as phase changes and sedimentation (Quaas and Gryspeerdt, 2022). This allows N_d to vary independently of the mass and to respond explicitly to aerosol activation and to feed back on subsequent microphysical processes such as autoconversion, therefore crucial for relatively realistic simulations of ACI such as the Twomey effect and the cloud lifetime effect.

Droplet activation also needs to be parameterized. It is important because it provides the main pathway by which the aerosol population influences N_d . Cloud droplets are activated from the available CCN depending on the updraft velocity and aerosol size distribution and composition, with the cloud-scale (subgrid) vertical velocity typically estimated from the turbulence kinetic energy (Quaas and Gryspeerdt, 2022). An alternative approach relies on simpler, direct empirical fits between measured N_a or CCN concentrations and N_d (e.g. Jones et al., 1994; Boucher and Lohmann, 1995). Another alternative uses empirical fits to explicit air parcel model simulations, which account more physically for the aerosol size distribution, composition, and updraft conditions (Ghan et al., 2011).

2.2.3 Autoconversion and the cloud lifetime effect

As introduced in Sect. 1.2.2.2, autoconversion is essential when investigating aerosol-MLC interactions. In most GCMs, cloud adjustments, particularly the cloud lifetime effect, are parameterized by modifying the autoconversion process (i.e. through precipitation suppression) (Quaas and Gryspeerdt, 2022; Stier et al., 2024). Specifically, because the autoconversion of cloud droplets into raindrops is usually formulated to depend inversely on N_d , aerosol-induced increase in N_d can suppress drizzle formation and increase LWP in GCMs (Kreidenweis et al., 2019). A widely used example of this is the empirical parameterization by Khairoutdinov and Kogan (2000), originally derived from large-eddy simulations of marine stratocumulus. This formulation expresses the time rate of change in the cloud liquid water mixing ratio q_l due to autoconversion (i.e. autoconversion rate, in $\text{kg kg}^{-1} \text{s}^{-1}$) as:

$$\left. \frac{\partial q_l}{\partial t} \right|_{\text{au}} = 1350 q_l^{2.47} N_d^{-1.79}, \quad (2.4)$$

This formulation mathematically shows the highly nonlinear dependency on available liquid water and the strong inverse relationship with droplet number, and it is also used in the GCM part of this thesis (Chap. 4). In this sense, aerosol

perturbations are given a direct pathway to modify the LWP adjustment, whereas their pathway for the CF is indirect (Bellouin et al., 2020; Quaas and Gryspeerdt, 2022), because CF must arise as a secondary response to changes in humidity, or additionally in cloud water content, resulting from the suppressed precipitation and mediated by the cloud-cover parameterization.

It is worth noting that aerosol effects on warm clouds are often reduced to those processes that can be parameterized most readily, for example, the lifetime effect as implemented in GCMs is largely a modelling construct shaped by available parameterizations rather than being directly rooted in process-scale observations (Mülmenstädt and Feingold, 2018). Moreover, this abstraction gives aerosol a clear route to modify cloud water and precipitation, but represents only part of the broader cloud adjustments, and other potentially important processes such as cloud-top entrainment and mixing receive less attention, partly because they are difficult to parameterize (Mülmenstädt and Feingold, 2018). In most GCMs, entrainment is not explicitly represented, but only implicitly via turbulence and shallow convection schemes (Quaas and Gryspeerdt, 2022). This is one reason why many models tend to show overly strong positive LWP responses to aerosol perturbations: autoconversion depends strongly on N_d , but lacks the representation of small-scale feedbacks such as evaporation-entrainment feedbacks (Toll et al., 2019; Quaas and Gryspeerdt, 2022; Stier et al., 2024).

2.2.4 ICON-HAM as the main GCM

The global modelling framework used in this thesis is the ICON atmosphere model coupled to HAM. The overall coupled model configuration largely follows Salzmann et al. (2022), but uses the more recently developed ICON version 2.6.4. Although comprehensive documentation for this version has not yet been published, its general configuration is very similar to that described by Giorgetta et al. (2018) for ICON-A (icon-aes-1.3.00), an ICON atmosphere model in a configuration that uses the Max Planck Institute physics package. ICON-A and ICON2.6.4 are successors of the ECHAM6 (Stevens et al., 2013). The same HAM version 2.3 as in Salzmann et al. (2022) is used, which was originally developed by Stier et al. (2005) and later upgraded to version 2 by Zhang et al. (2012a). ICON-HAM in this thesis refers to ICON2.6.4-HAM2.3 hereafter, which as a whole was built based on ECHAM6.3-HAM2.3, following a long development lineage originating from the ECHAM-HAM by Stier et al. (2005). Thus, many aerosol and cloud process representations were inherited or further adapted within the ICON framework. While the ECHAM-HAM family has been widely used and evaluated in ACI

research (Lohmann et al., 2007; Neubauer et al., 2019; Tegen et al., 2019), ICON-HAM provides a more modern nonhydrostatic dynamical core on an icosahedral grid (Giorgetta et al., 2018), designed to overcome the computational bottlenecks and scalability limits of traditional spectral grids on modern supercomputing architectures but retaining the well-established HAM aerosol and cloud parameterizations at the same time. A general description of ICON-HAM is also provided in Sect. 4.2.1.1.

ICON-HAM uses specific parameterization schemes relevant to ACI that align with the theoretical concepts introduced in the previous sections:

- **Fractional cloud cover:** The default scheme is the CC-RH scheme which diagnoses CF as a function of grid-mean RH with a height-dependent critical threshold (CC-RH; Sundqvist et al., 1989; Salzmann et al., 2022), consistent with the RH-based probability density function framework discussed in Sect. 2.2.1. This default scheme is compared to an alternative one that explicitly couples cloud cover to LWC (CC-RH-LWC), introduced and discussed in more detail in Chap. 4. Different formulations of cloud-cover parameterization are of particular importance for this thesis because they influence how efficiently changes in N_d are translated into changes in CF.
- **Cloud microphysics, activation, and precipitation:** The representation of microphysics in the ECHAM-HAM / ICON-HAM model lineage is originally built on the earlier single-moment stratiform cloud scheme established by Lohmann and Roeckner (1996). To enable aerosol-cloud coupling, this foundation was extended to a two-moment cloud microphysics scheme with N_d treated prognostically following Lohmann et al. (2007) and subsequently refined by Lohmann and Hoose (2009) and Lohmann and Neubauer (2018). This two-moment microphysics scheme represents a broad range of warm and cold cloud microphysical processes, including condensation and evaporation, freezing and melting, deposition and sublimation, autoconversion and accretion, and the sedimentation of ice (Salzmann et al., 2022). Droplet activation is computed using the Abdul-Razzak and Ghan (2000) parameterization based on the Köhler theory, with aerosol information provided prognostically by HAM, as in Lohmann and Neubauer (2018) and Tegen et al. (2019). Precipitation formation is represented by the Khairoutdinov and Kogan (2000) autoconversion scheme (Eq. (2.4)), which carries the strong inverse dependence on N_d discussed in Sect. 2.2.3.

The simulations analyzed later in Chap. 4 compare the two cloud-cover schemes

(CC-RH and CC-RH-LWC), while the underlying cloud microphysics treatment is held constant. An additional model configuration choice examined there is the $N_{d,\min}$. In GCMs, $N_{d,\min}$ is commonly imposed to prevent unrealistically low N_d . However, it can also influence the simulated CF response by artificially constraining the clean-cloud regime, as will be shown in Chap. 4. The default value of $N_{d,\min}$ used in ECHAM-HAM and ICON-HAM is 40 cm^{-3} (Hoose et al., 2009; Neubauer et al., 2019). Details on the specific ICON-HAM setup, experiment design, and resulting sensitivities are discussed in detail in Chap. 4.

2.2.5 Summary of data sources

This thesis combines three primary datasets, including satellite observations, re-analysis, and global modelling data to investigate the CF adjustment from complementary perspectives. Terra-MODIS and ERA5 products provide the basis for the observational analysis in Chap. 3, while ICON-HAM model output forms the basis of the model-based analysis in Chap. 4. Table 2.1 summarizes the data sources used in this thesis and their respective roles.

Table 2.1: Overview of the primary datasets used in this thesis, including their types, main variables, application chapters, and purposes.

Data source	Type	Main variables	Used in	Purpose
Terra-MODIS MOD08_D3, Collection 6.1 (Platnick et al., 2017)	Satellite remote sensing	Cloud fraction, cloud optical and cloud-top properties; derived cloud droplet number concentration	Chap. 3	Observational analy- sis of CF adjustment
ERA5 (Hersbach et al., 2020)	Reanalysis	Thermodynamic, dy- namical, humidity, boundary-layer, and surface-flux variables	Chap. 3	Meteorological predictors for the ob- servational analysis
ICON-HAM (Salzmann et al., 2022)	Global cli- mate model	Simulated cloud, precipitation, and large-scale meteorolog- ical variables	Chap. 4	Model-based analysis of CF adjustment and sensitivity to model configuration choices

2.3 Machine Learning Techniques

2.3.1 Overview of machine learning concepts and methods

ML refers to a broad class of algorithms designed to extract relationships and perform tasks by learning directly from data rather than relying on explicit and physics-based programming for this given task (Beucler et al., 2023). These algorithms can be categorized by the availability of target data during training: In supervised learning, the ML model is trained on labelled input–output pairs to learn a mapping from features to a target variable (Hastie et al., 2009). In contrast, unsupervised learning extracts structure and patterns from unlabelled data, for example, through clustering, dimensionality reduction, or generative adversarial networks (Beucler et al., 2023; Molina et al., 2023). Within supervised learning, tasks are commonly divided based on the target variable into regression, which predicts continuous numerical values, and classification, which assigns samples to discrete categories or classes.

A wide range of algorithms with varying interpretability and complexity can be used. At the simpler end, linear models assume a linear relationship between features and the target. They are computationally inexpensive and straightforward to interpret, but their flexibility is limited when the underlying relationships are strongly nonlinear (Hastie et al., 2009; Schiller et al., 2025), which is common in atmospheric processes (Lai et al., 2025). To model such nonlinearities, decision trees can be used, which partition the feature space through a sequence of hierarchical conditional rules (Quinlan, 1986, 1987). Despite the relative interpretability of decision trees due to relatively low complexity, individual decision trees are unstable and prone to high variance and thus overfitting (Breiman, 1996; Jiang et al., 2024). Therefore, they are frequently integrated into ensemble learning methods, which combine multiple base learners to produce a single and more robust predictive model (Dietterich, 2000). One of the simplest examples is random forests, which average over predictions from many trees trained on random subsets of data and features, reducing variance and overfitting (Breiman, 2001). One important and widely used family of ensemble learning is boosting techniques, in which models iteratively construct sequential trees, with each new tree designed to correct the errors of the previous ones (Friedman, 2001; Chen and Guestrin, 2016). At the highest tier of complexity are neural networks. They have nonlinear function approximators organized into layers of interconnected nodes to capture highly complex, high-dimensional relationships but require large training datasets and are computationally expensive (LeCun et al., 2015; Goodfellow et al., 2016).

Building on basic neural networks, more advanced deep learning architectures have also emerged. Convolutional neural networks apply spatially aware filter operations (convolutions) to extract local features and are therefore particularly suited for spatially structured data, such as satellite imagery (LeCun et al., 2015; Reichstein et al., 2019). Recurrent neural networks and transformers are designed for sequential and long-range dependencies, and have been applied to time series prediction in weather and climate (Vaswani et al., 2017; Bi et al., 2023; Kim et al., 2025). Generative models such as variational autoencoders and generative adversarial networks learn to approximate the underlying data distribution and have been applied to stochastic downscaling and probabilistic prediction in weather and climate (Kingma and Welling, 2013; Ravuri et al., 2021). Physics-informed ML integrates physical constraints directly into the model architecture or loss function, improving generalization and physical consistency (Raissi et al., 2019; Karniadakis et al., 2021).

As model complexity and flexibility increase, so does its susceptibility to overfitting. The most common and basic practice to prevent overfitting is to split data into training, validation and unseen test sets. Overfitting then can be mitigated through (i) hyperparameters tuning, which are model configuration choices that are not learnt directly from the data but specified beforehand e.g. tree depth, number of trees, and learning rate. The validation set is used iteratively during training to monitor model development and performance and optimize hyperparameters. (ii) Regularization, which penalize overly complex solutions, for example, through feature or column subsampling, or by adding penalty terms to the loss function (e.g. L1 or L2 regularisation) that discourage large parameter values. (iii) Early stopping, in which training is halted once predictive performance on validation data no longer improves (Ying, 2019; Beucler et al., 2023). Ultimately, the test set is strictly reserved for the final, unbiased evaluation of the fully trained model. In geoscientific applications, preventing overfitting is critical because data often exhibit strong spatial and temporal dependence and can otherwise lead to overly optimistic model performance (Karpatne et al., 2019; Beucler et al., 2023).

Because many atmospheric applications require not only accurate prediction but also scientific understanding, interpretability should be considered when selecting ML methods. From linear regression to neural networks, the complexity and flexibility increase, while the interpretability decreases (Jiang et al., 2024; Schiller et al., 2025), meaning that more complex models usually need additional explainability tools to understand how individual feature contribute to the model prediction (McGovern et al., 2019; Molnar, 2025), which motivates the use of

methods discussed in Sect. 2.3.3.

2.3.2 Applications in atmospheric sciences

ML has become increasingly prominent across atmospheric and climate sciences, owing to the rapid growth of observational and model data and by its ability to extract complex, nonlinear relationships from high-dimensional data (Reichstein et al., 2019; Karpatne et al., 2019; Bracco et al., 2025). As illustrated schematically in Fig. 2.2, these applications can be organized into three broad domains—observation, theory and computation—and their interactions with many of the most impactful ones sitting at the intersections between them.

In the observation domain, ML is used to reconstruct incomplete records, classify patterns, and extend observational datasets in time, space, and observable quantities, both for satellite and in situ measurements (Bracco et al., 2025). Common paradigms include the automated detection and classification of cloud regimes and mesoscale organization from satellite imagery using convolutional neural networks (Denby, 2020; Rasp et al., 2020; Marais et al., 2020), statistical downscaling or super-resolution of coarse observations (Vandal et al., 2017; Stengel et al., 2020), and the retrieval of cloud properties from multispectral satellite observations (Min et al., 2020; Beucler et al., 2023). In the theoretical domain, ML supports the data-driven subgrid parameterizations of unresolved processes such as convection and microphysics that are too costly to resolve explicitly in GCMs (Rasp et al., 2018; Gentine et al., 2018), as well as reduced-order models that capture complex dynamical systems with lower-dimensional representations (Brunton et al., 2020; Bracco et al., 2025). In the computational domain, ML is increasingly used for data-driven weather forecasting that approaches or matches the skill of operational numerical prediction systems (Bi et al., 2023; Lam et al., 2023), and to support climate modelling by providing computationally efficient emulators (Watson-Parris et al., 2022; Van Katwyk et al., 2026), and also for bias correction by learning and correcting systematic errors in both weather and climate models (Watson, 2019; Bonavita and Laloyaux, 2020; Watt-Meyer et al., 2021).

Many of the most impactful applications of ML lie at the intersections of these three domains. In these overlaps, observational data, physical understanding, and computational tools are combined to address problems that cannot be tackled as effectively from a single perspective. For instance, the overlap between observation and computation supports data assimilation, while the overlap between theory and observation facilitates physics-guided model development and validation, helping ensure that the learnt patterns remain consistent with known physics. In

general, hybrid and physics-informed approaches combine physical constraints with data-driven flexibility, improving generalization while preserving physical consistency (Karniadakis et al., 2021; Reichstein et al., 2019; Bracco et al., 2025). However, the role of ML is not the same in weather and climate applications. As discussed by Watson-Parris (2021), weather forecasting is an initial value problem, sitting at the overlap between the observation and computation domains in Fig. 2.2, where ML models are trained on reanalysis data to interpolate within a well-sampled distribution of atmospheric states. In this setting, success is typically evaluated by short-term predictive scores. On the contrary, climate projection is essentially a boundary value problem, which is more constrained by external conditions and forcings, falling between the theory and computation domains. It requires extrapolation to unobserved conditions, making long-term statistics, process understanding, and the representation of forced responses and interactions far more important (Watson-Parris, 2021; Beucler et al., 2023; Lai et al., 2025). Therefore, ML in climate science emphasizes physical consistency, robustness across regimes, and interpretability. Accordingly, recent reviews highlight that ML in climate science is moving beyond prediction alone, towards approaches that also improve physical understanding, integrate diverse data sources, and support more efficient representation of complex processes (Reichstein et al., 2019; Beucler et al., 2023; Bracco et al., 2025).

In this context, the methodology used in this thesis can be placed primarily at the interface between the observation and theory domains. It uses ML to quantify relationships between CF and N_d from observations, reanalysis and a GCM, aiming to extract physically meaningful insights rather than prediction alone. This makes explainable ML particularly relevant, as it provides tools to relate what the model has learnt back to physical mechanisms, which is discussed in the following section.

2.3.3 Explainable Machine learning

As ML models grow in complexity in climate science, understanding how their predictions are made becomes more important. The black box challenge has motivated the development of explainable ML, which refers to a set of methods that make model behaviour more transparent and interpretable to humans (Guidotti et al., 2018; McGovern et al., 2019; Barredo Arrieta et al., 2020). In general, these methods can be classified along many dimensions, e.g., whether they provide *global* or *local* explanations, whether they are *model-agnostic* or *model-specific*, and whether they are *ante-hoc* (interpretable) or *post-hoc* (explainable) (Linardatos et al., 2021; Molnar, 2025). The last two largely overlap, so here we primarily introduce the

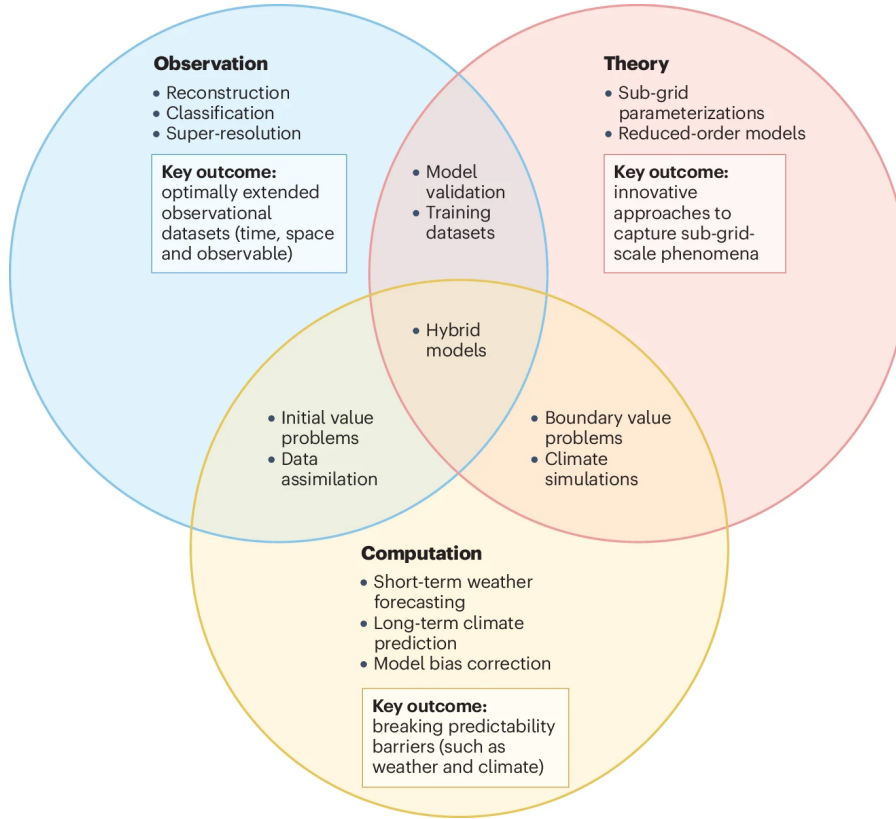


Figure 2.2: Schematic overview of three broad domains in which machine learning has contributed to climate science: observations, theory, and computation, together with their interactions. Major research topics in which ML has brought substantial improvements are highlighted within each domain and their overlaps. Reproduced from Bracco et al. (2025) with permission from Springer Nature.

global vs. local and model-agnostic vs. model-specific distinctions.

Global methods characterize the overall model behaviour, for example through feature importance rankings, partial dependence plots, accumulated local effects, or surrogate models, whereas local methods explain individual predictions by quantifying how specific input features contribute to a single model output (Lundberg et al., 2018; Molnar, 2025). Another distinction concerns whether an explanation method can be applied to any predictive model, or whether it is tailored to a specific model class. Model-agnostic methods such as Local Interpretable Model Agnostic Explanation (LIME) (Ribeiro et al., 2016), partial dependence plots (Friedman, 2001), and surrogate models can explain model behaviour without information on the internal model structure (Molnar, 2025). On the other hand, model-specific methods exploit the architecture directly, for instance through gradient-based

attribution in neural networks (Simonyan et al., 2013; Sundararajan et al., 2017).

Among these approaches, a particularly impactful class is based on cooperative game theory. In its general form, this class provides local feature attributions and is model-agnostic, but it is possible to derive global information and they can be made model-specific for computational efficiency. It treats the prediction task as a “game” where input features are “players” collaborating to achieve a certain “payout” (the model prediction). It assigns feature contributions according to their marginal effect across all possible subsets of features (Molnar, 2025). This idea can be traced back to the Shapley value (Shapley, 1953). The Shapley value represents the average marginal contribution of feature i over all possible orderings in which the features can be introduced to the prediction, aiming to fairly distribute the total payoff among the players. It is defined as:

$$\phi_i = \sum_{S \subseteq N \setminus \{i\}} \frac{|S|!(M - |S| - 1)!}{M!} [f_x(S \cup \{i\}) - f_x(S)], \quad (2.5)$$

where N is the set of all features, M is the total number of features, S denotes a subset of features excluding feature i , and $f_x(S)$ is the model prediction conditioned on S (i.e. when only the information in subset S is known) (Lundberg and Lee, 2017; Lundberg et al., 2018). This formulation is attractive because it satisfies several desirable properties: *efficiency*, meaning that all feature contributions together exactly recover the difference between the prediction and the baseline; *symmetry*, meaning that two features that contribute equally receive the same attribution; *dummy*, meaning that a feature with no effect on the prediction receives zero attribution; and *additivity*, meaning that attributions remain consistent when combining additive models (Lundberg and Lee, 2017; Molnar, 2025).

Building on this concept, SHAP explains a prediction by assigning each feature an additive contribution based on Shapley values. Each prediction is decomposed into a base value ϕ_0 and a set of individual feature contributions ϕ_i whose sum recovers the model output, which can be expressed as: $\phi_0 + \sum_{i=1}^M \phi_i = f(x)$ (Lundberg and Lee, 2017). The base value corresponds to the model’s expected prediction in the absence of feature-specific information, typically computed as the mean prediction over the training set. Positive (negative) contributions indicate that the specific feature value raises (lowers) the prediction relative to this baseline (base value).

Importantly, SHAP values are therefore local feature contributions of a specific feature instance, not physical sensitivities themselves (e.g. $\frac{dCF}{dN_d}$). These local explanations can be aggregated into global interpretations, such as dependence

relationships, summary plots, and overall feature importance (Lundberg et al., 2020; Molnar, 2025). This distinction between local contributions and global sensitivities is important in the context of this thesis. In the studies presented here, local SHAP values are not interpreted directly as global sensitivities, which are then derived from the distribution of the local contributions. This local-to-global consistency is one of the main reasons why SHAP has become particularly useful in environmental applications (Lundberg et al., 2020; Stirnberg et al., 2021; Zipfel et al., 2022; Li et al., 2022b). A further consideration in the estimation of Shapley values is whether the conditional expectation $f_x(S) = \mathbb{E}[f(x)|x_S]$ is evaluated in a way that is *true to the data*, i.e. respecting observed feature correlations, or *true to the model*, i.e. using an interventional approach that in practice treats features as independent (Chen et al., 2020; Frye et al., 2020). In atmospheric sciences, where meteorological variables are inherently coupled, breaking the feature dependence can force SHAP to explain the model based on physically impossible weather conditions. Thus, the former is preferred to account for the physical covariations among meteorological predictors. This is discussed in detail in Sect. 3.2.3.3. Beyond individual feature attributions, SHAP can also be extended to SHAP interaction values, which quantify how the contribution of one feature to a prediction depends on the value of another feature (Lundberg et al., 2018). This is used in Chap. 3 to examine how meteorological factors influence the $\ln N_d$ –CF sensitivity (Sect. 3.2.3).

A practical limitation of computing exact Shapley values is their exponential computational complexity with respect to the number of features, making the full sum in Eq. 2.5 infeasible for large feature sets. The *TreeSHAP* approach is designed to address this for tree-based models such as random forests and gradient-boosted decision trees (Lundberg et al., 2018, 2020). In this thesis, the tree-based model used is XGBoost (Chen and Guestrin, 2016), a scalable and efficient implementation of gradient-boosted decision trees that builds an ensemble of weak trees sequentially that is widely used because of its strong predictive skill, flexibility for nonlinear tabular data and the ability to capture feature interactions. A more detailed introduction to XGBoost is provided in Sect. 3.2.2. The basic idea is to exploit the hierarchical structure of decision trees to compute feature contributions along each decision path from root to leaf, avoiding the cost of enumerating all possible feature subsets explicitly. This makes the computation of exact Shapley values more efficient in polynomial time, with complexity $\mathcal{O}(TLD^2)$, where T is the number of trees, L the maximum number of leaves per tree, and D the maximum depth. Compared with model-agnostic approximations, TreeSHAP makes Shapley-based explanations computationally feasible for large tree ensembles with many

features, while preserving the key theoretical properties of Shapley values. The resulting SHAP values are exact rather than approximate and satisfy all Shapley axioms. In this thesis, TreeSHAP is applied to the regional XGBoost models using the shap Python package (Lundberg et al., 2020). Details on how SHAP dependence plots are used to derive $\ln N_d$ -CF sensitivities are provided in Sect. 3.2.3.2 and Sect. 4.2.3.

It should be noted that SHAP is not a tool for direct causal inference. Like other post-hoc explanation methods, SHAP describes the behaviour of a trained statistical model rather than the physical system itself. The resulting attributions depend on the training data, model structure, and assumptions made in estimating the Shapley values, particularly when predictors are correlated (Sundararajan and Najmi, 2020; Frye et al., 2020; Chen et al., 2020; Molnar, 2025). In atmospheric sciences, where many variables covary, SHAP-based results should be interpreted as statistically informed and physically guided rather than strictly causal. Nevertheless, SHAP provides a powerful framework for identifying robust feature contributions, exploring interactions, and relating model behaviour to plausible physical mechanisms, making it well suited to the presented work. This is also systematically discussed throughout Chap. 3

Chapter 3

Observation-based cloud fraction response to aerosols

3.1 Specific motivation and aim

As introduced in the Chap. 1, from the perspective of large-scale satellite observations, one of the most challenging aspects in the quantification of the cloud fraction (CF) adjustment is isolating the influence of aerosol on cloud properties from confounding covariations with meteorological parameters (Andersen et al., 2016; Gryspeerd et al., 2019; Bellouin et al., 2020), paired with aerosol retrieval issues related to aerosol swelling and three-dimensional radiative effects in the vicinity of clouds (Loeb and Schuster, 2008; Schwarz et al., 2017).

Recent observational studies have utilized different methods to tackle this issue. One approach is to stratify the data by meteorological factors and thereby account for local meteorology in the relationships (e.g. Su et al., 2010; Chen et al., 2014; Andersen and Cermak, 2015). Another approach is to use cloud droplet number concentration (N_d) as a mediating variable to analyze the causal pathway between aerosol optical depth and CF (Gryspeerd et al., 2016). A further approach is to apply cloud–aerosol pairing strategies (Christensen et al., 2017). However, these methods do not account for aerosol retrieval issues, meteorological influencing factors and confounders at once, which is essential for constraining the CF adjustment.

Recently, several studies have successfully used machine learning (ML) to account for nonlinearities and meteorological factors in the quantification of aerosol–cloud interactions (ACI) (Andersen et al., 2017; Fuchs et al., 2018; Dadashazar et al., 2021; Zipfel et al., 2022). ML regression algorithms allow for the predic-

tion of CF from aerosol and meteorological factors at the same time and treat the aerosol–cloud–meteorology system as a whole. In addition, ML models can represent nonlinear interactive systems, which can be analyzed using explainable ML techniques (Beucler et al., 2023).

In this chapter, region-specific eXtreme Gradient Boosting (XGBoost) models are set up at a near-global scale using MODerate Resolution Imaging Spectroradiometer (MODIS) and fifth generation European Centre for Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis (ERA5) data to predict CF and to analyze N_d -induced changes in marine low clouds (MLCs). The aim is to quantify the global sensitivity patterns of CF to N_d and meteorological factors. In addition, the chapter aims to estimate the magnitude of the dependence of the N_d –CF sensitivity on meteorological factors using SHapley Additive exPlanations (SHAP) interaction values, thereby providing a new pathway to better understand the physical processes relevant to the CF adjustment and to constrain aerosol-induced CF changes globally while accounting for meteorological covariations. The guiding hypotheses of this study are: (i) the response of CF in MLCs to aerosol perturbations is positive but modulated, i.e. reduced or amplified, by ambient meteorology; (ii) CF sensitivities to $\ln N_d$ and meteorological variables, as well as the meteorological modulation of the $\ln N_d$ –CF sensitivity, exhibit distinct regional patterns.

3.2 Data and methods

3.2.1 Data sets

This work combines nine years (2011–2019) of satellite retrievals from MODIS and reanalysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) from 60° N to 60° S. In this study, MLCs are defined as single-layer warm cloud fields with cloud-top temperature (CTT) higher than 268 K. To achieve this, the information on CF (product: Cloud_Retrieval_Fraction_1L_Liquid), cloud droplet effective radius (r_e) (product: Cloud_Effective_Radius_1L_Liquid_Mean), cloud optical depth (τ_c) (product: Cloud_Optical_Thickness_1L_Liquid_Mean), CTT (product: Cloud_Top_Temperature_Mean) and satellite viewing geometry are obtained from MODIS Level-3 collection 6.1 atmosphere daily products on the Terra platform (MOD08_D3), which are gridded into $1^\circ \times 1^\circ$ globally from Level-2 atmospheric products. CF serves as the predictand in this study. The computation of N_d relies on τ_c and r_e , with filtering criteria based on CTT, solar zenith viewing angle and satellite zenith angle, as elaborated in the following.

The MODIS N_d used in this study is calculated following Quaas et al. (2006) (see Eq. 2.1 in Sect. 2.1.3), which depends on the retrievals of r_e and τ_c , as do the uncertainties on the errors propagated from r_e and τ_c . The uncertainties in N_d retrievals are exhaustively evaluated by Grosvenor et al. (2018), which suggests that the uncertainties in averaged N_d over $1^\circ \times 1^\circ$ grid box (spatial resolution of the MODIS products used in this study) decrease by over 50 % compared to pixel-level uncertainties. This derivation approach relies on the assumed adiabaticity in global marine warm clouds where liquid water content (LWC) and r_e increase monotonically and N_d is distributed as constant vertically. Departure from the adiabatic assumption (e.g. due to entrainment) would result in N_d retrieval biases (Merk et al., 2016; Bennartz and Rausch, 2017). The uncertainty related to the estimation of N_d from MODIS also depends on liquid CF. N_d is less biased in regions of larger CF where clouds are more homogeneous, while in regions with lower CF, N_d retrievals are sparser and less reliable (Grosvenor et al., 2018; Zhu et al., 2018). In such heterogeneous cloud fields, subpixel effects in the retrieval of r_e can negatively bias the retrieved N_d values (Zhang and Platnick, 2011; Zhang et al., 2012b; Grosvenor et al., 2018). Such retrieval biases could cause a bias in the N_d –CF relationship as well. Furthermore, the interpretation of the causal effect of N_d on CF can also be obscured by small-scale sampling issues. In particular, apart from the retrieval errors in r_e and τ_c , the natural spatial variability in cloud fields can also propagate to the N_d estimate and distort the N_d –CF relationship (Arola et al., 2022; Liu et al., 2024).

Following the screening criteria for more reliable N_d demarcated by Gryspeerdt et al. (2022), only clouds restricted to single-layer in liquid phase with CTT higher than 268 K are considered. As suggested by Quaas et al. (2006), samples with $r_e < 4\mu\text{m}$ and $\tau_c < 4$ are excluded to cope with the high r_e retrieval uncertainties at low τ_c . In addition, solar and sensor viewing zenith angles respectively greater than 65° and 55° are removed to avoid the large biases in r_e and τ_c retrievals (as in Grosvenor et al., 2018). The pixels selected according to the above sampling strategies generate more reliable N_d estimates.

Atmospheric and oceanic variables are taken from the fifth generation ECMWF atmospheric reanalysis of the global climate (ERA5) at an hourly frequency (Table 3.1) (Hersbach et al., 2020). The ERA5 data sets are harmonized to fit the Level-3 MODIS data by first being resampled to $1^\circ \times 1^\circ$ from their default $0.25^\circ \times 0.25^\circ$ spatial resolution using bilinear interpolation, and they are subsequently collocated to Terra MODIS by extracting hourly data to align with the UTC overpass times of the Terra satellite for each grid cell, yielding a spatiotemporally matched

MODIS-ERA5 combined dataset for training the ML models. For N_d retrievals, only samples within 1st–99th percentiles are retained to exclude potential unrealistic outliers from r_e and τ_c retrievals (Zipfel et al., 2022). Furthermore, the explanation of ML models in this study relies on using linear regressions to capture the distribution of individual prediction instances, and the extreme values may excessively magnify or reduce the sensitivity or interactive effects quantified by SHAP (shown in Fig. 3.1 and discussed in Sect. 3.2.3.2). The threshold of 1–99 percentiles for each predictor is thus adopted to remove the values at the very tails of the specific distribution and to improve the robustness of the estimated sensitivities. To define the sensitivities of CF and the interactive effects of meteorological factors, the natural logarithm of N_d is taken (see Sect. 3.2.3.2 in detail). Estimated inversion strength (EIS) is calculated based on the formulation from Wood and Bretherton (2006) and in this study, it is dependent only on atmospheric temperatures at 700 hPa and at the level of 1000 hPa.

All input predictors for each XGBoost model (i.e. for each $5^\circ \times 5^\circ$ window aggregated from $1^\circ \times 1^\circ$ grid boxes, as detailed in Sect. 3.2.2) are standardized by centering around the mean and scaling to have unit variance as in Scott et al. (2020). Hamby (1994) suggested that the standardization process is a standard practice when aiming for comparability of sensitivity estimates across predictors. This process eliminates the influence of units and aligns data on the same scale instead of the original natural ones, thereby ensuring comparability of the quantified sensitivities and interactive effects with meteorology among different variables. This standardization procedure has been applied in other studies investigating different cloud sensitivities to various cloud-controlling factors (e.g. Ceppi and Nowack, 2021; Andersen et al., 2023). This procedure however may result in reduced spatial comparability due to variations in mean and standard deviation values across different $5^\circ \times 5^\circ$ windows. To assess the trade-off between comparability among different predictors and comparability in space, we provide results without standardization in the supplementary material (Fig. A.2 to Fig. A.7 therein) as done by Grise and Kelleher (2021). In terms of spatial patterns, the results are nearly identical to their corresponding ones presented in the following sections of the main text, suggesting that standardizing the data based on the local mean and standard deviation for each window has only a small impact on comparability across each window. Therefore, we primarily benefit from achieving comparability among different predictors with only a minor compromise in spatial comparability.

Table 3.1: Summary of the predictors from ERA5 reanalysis.

Predictor Name	Abbreviation	Units
Instantaneous pressure level parameters (at 700 hPa, 850 hPa)		
Relative humidity	RH_{700}, RH_{850}	%
Specific humidity	SH_{700}, SH_{850}	kg kg^{-1}
Temperature	t_{700}, t_{850}	K
Vertical velocity	$\omega_{700}, \omega_{850}$	Pa s^{-1}
Eastward wind component	u_{700}, u_{850}	m s^{-1}
Northward wind component	v_{700}, v_{850}	m s^{-1}
Surface and single level parameters (instantaneous or mean rates/fluxes)		
Eastward and northward wind component at 10 m	u_{10}, v_{10}	m s^{-1}
Boundary-layer height	BLH	m
Convective available potential energy	CAPE	J kg^{-1}
Sea surface temperature	SST	K
Total column water vapour	TCWV	kg m^{-2}
Mean large-scale precipitation fraction	PF	–
Mean surface sensible/latent heat flux	SHF/LHF	W m^{-2}
Calculated		
Estimated inversion strength	EIS	K

3.2.2 Machine learning model setup

XGBoost is a distributed tree boosting algorithm aiming to provide a scalable, portable and flexible library under the Gradient Boosting framework (Chen and Guestrin, 2016). The state-of-the-art XGBoost can be implemented efficiently in Python and has been recently used to study clouds and ACI (Andersen et al., 2022; Douglas and L’Ecuyer, 2022). As an extension of previous gradient boosting methods, XGBoost has incorporated regularization techniques which help prevent overfitting and improve model generalization. Besides, the subsampling on training subsets and column (feature) subsampling techniques can shorten the running time and also mitigate overfitting and hence elevate model performance (Chen and Guestrin, 2016). Relevant regularization and subsampling hyperparameters are tuned using Bayesian Optimization to determine the best combination, see Table 3.2 for the search space.

Data from 2011 to 2016 are used for training and data from 2017 to 2019 for testing (independent train/test split about 67 %/33 %). By chronologically splitting the training and test sets without random shuffling, we ensure that the training data will not see future information and the autocorrelation in the data will not

Table 3.2: Overview of the hyperparameters tuned for regional XGBoost models using Bayesian Optimization.

Hyperparameter name	Search Space
learning_rate	0.01–0.5
max_depth	3–10
min_child_weight	1–10
subsample	0.5–1
colsample_bytree	0.5–1
gamma	0–10
alpha	0–10
lambda	0–10

lead to an overoptimistic evaluation of the model’s performance (Beucler et al., 2023; Kapoor et al., 2023). As suggested by Karpatne et al. (2017), a single ML model may not perform well across all regions due to the heterogeneity of relevant processes. Therefore, data at a $1^\circ \times 1^\circ$ spatial resolution are aggregated into $5^\circ \times 5^\circ$ geographical windows, where an individual, independent XGBoost model is trained and tested for each “window”. Hereby a region-specific ML framework is established to potentially capture regional relationships and characteristics and thus the regional patterns of the CF adjustment. The coarser $5^\circ \times 5^\circ$ spatial resolution of the modelling grid increases the sample size by a factor of ≈ 25 which is helpful to establish robust sensitivity estimates. In addition, at the spatial resolution of $1^\circ \times 1^\circ$ summarized in $5^\circ \times 5^\circ$ degree windows, the spatial scale is adequate for ACI sensitivity estimation (Grandey and Stier, 2010). To ensure a sufficient data amount for training and testing the XGBoost models, only the geographical windows with over 6000 available data points are retained. Consequently, 34 out of 1190 oceanic windows have been excluded. These windows located between 47.5° W– 122.5° E and 52.5° S– 57.5° S in the Southern Ocean (Fig. 3.2) contain fewer than 6000 valid samples due to the screening for N_d retrievals. For each model, the hyperparameters are tuned by implementing Bayesian optimization, which uses a Gaussian process prior distribution over hyperparameters to initialize a probabilistic model for the objective function to be optimized. After the initialization, the probabilistic model is updated iteratively and Bayesian optimization suggests the optimal combination of hyperparameters to try for the next iteration according to the previous one and samples gathered from the search space (Table 3.2) (Snoek et al., 2012). Each iteration is evaluated by 5-fold cross-validation using root mean square error (RMSE) as score. The number of boosting rounds (the number of trees) for each XGBoost model is then determined by the early stopping

technique to further avoid overfitting i.e. the training of the model will stop early once it is monitored that the score of cross-validation does not improve within 20 iteration rounds.

3.2.3 Explaining the machine learning models

3.2.3.1 SHapley Additive exPlanation (SHAP) values

SHAP values were proposed by Lundberg and Lee (2017) on the basis of cooperative game theory to explain the outputs of ML models. The SHAP approach has been implemented with XGBoost in Python and it has been reported that outputs from XGBoost models with various number of trees can be well explained by the SHAP framework in different subject areas (e.g. Padarian et al., 2020; Lundberg et al., 2018, 2020; Kim et al., 2021; Li et al., 2022b). The contribution of a predictor value to a specific model prediction is calculated as the difference between the predictions of the model in the presence and absence of this particular predictor for all possible combinations of predictor values. Since this is performed at a “local” level (i.e. for this specific instance’s prediction), it allows for insights into how a certain model outcome is achieved, thereby complementing more traditional “global” (considering all instances) feature importance measures (e.g. partial dependence plot).

The base value in the context of SHAP values is what would be predicted in the absence of any feature information (Lundberg and Lee, 2017) and it is typically computed as the average of all predictions by ML models over the entire training data set. Positive (negative) SHAP values indicate that the specific feature value increases (decreases) the prediction compared to this base value. In other words, the base value serves as the reference point against which the contributions of individual features are measured. SHAP values for all features will always sum up to the difference between the base value and the final model prediction so that SHAP values are additive and internally consistent. The base value could be analogous to the climatological CF for a given geographical window assuming no information about the input parameters is known. In this context, the SHAP values of input features indicate the extent to which knowing information about each feature value would deviate the prediction from the climatological CF (base value).

Furthermore, the quantification of the influence of meteorology on the N_d -CF relationship can be analyzed using SHAP interaction values, which are an extension of SHAP values. They measure the difference between the SHAP values for a feature when another (secondary) feature is included versus when it is not included, offering a potential tool for insights into feature interactions captured by

the tree ensembles. SHAP values have been applied to study atmospheric aerosols in the context of air pollution (Stirnberg et al., 2021), and have been used by Zipfel et al. (2022) to explore satellite-observed N_d –liquid water path (LWP) relationship in MLCs in the Southeast Atlantic, finding that meteorological variables have considerable influences on the N_d –LWP relationship using SHAP interactive values. Moreover, the use of SHAP interaction values in these studies allows for a more profound and in-depth comprehension of the underlying processes with respect to local meteorology. SHAP values provide insights into the behaviour of the XGBoost models, and as all statistical/ML models, they may not necessarily reflect real-world physical causality. Nevertheless, this state-of-the-art technique allows us to account for meteorological covariations when deriving sensitivities and to appraise to what extent the meteorological predictors interact with and influence the N_d –CF relationship beyond traditional global-level feature attributions.

3.2.3.2 Quantification of sensitivities and interactive effects

Figure 3.1 is an exemplary graph for a regional XGBoost model at a specific $5^\circ \times 5^\circ$ window (27.5° S – 32.5° S , 122.5° W – 127.5° W). SHAP values and SHAP interaction values are used to explain this XGBoost model and to quantify and isolate the CF sensitivity to N_d and the interactive effects of meteorological factors (here sea surface temperature (SST)). Each dot in Fig. 3.1 represents an individual data instance (i.e. a single observation at a specific grid cell and time step) and shows how individual N_d or $\ln N_d$ values impact the CF prediction.

Plotting SHAP values of N_d against N_d values without the standardization process (Figure 3.1a) for each data sample illustrates that increased N_d values lead to an increase in the predicted CF, while the rate of the increase ($d\text{SHAP}/dN_d$) drops with N_d as shown by the orange line. For each 20 cm^{-3} wide bin of N_d , $d\text{SHAP}/dN_d$ is calculated as the slope of the linear regression between N_d and N_d SHAP values. The nonlinear positive association between N_d and predicted CF aligns well with findings of prior studies (e.g. Gryspeerd et al., 2016; Rosenfeld et al., 2019) that the aerosol impact on CF saturates at relatively high aerosol loading. This relationship also resembles the one reported by Yuan et al. (2023) which is attributed to the precipitation suppression effect due to relatively high N_d .

Expressing the sensitivity logarithmically in N_d is ideal because cloud processes are prone to respond to a relative change in N_d rather than an absolute one (Carslaw et al., 2013; Bellouin et al., 2020). Furthermore, the log-transformed N_d facilitates the application of simple linear regressions to capture the relationship between

the contribution of N_d to the predicted CF (N_d SHAP values) and its feature values. As depicted in Fig. 3.1b, the contribution of $\ln N_d$ to the predicted CF increases almost linearly with rising $\ln N_d$. Thus, the CF sensitivity to N_d is estimated as the slope of the linear regression between $\ln N_d$ SHAP values and $\ln N_d$ values ($0.098 \text{ CF } \sigma^{-1}$). A similar method to estimate sensitivity has also been used by Li et al. (2022b), where it is also suggested that this method can enhance the robustness of the sensitivity estimation. Because it can leverage the benefits of an XGBoost model, including bagging techniques and no need for distribution assumptions, along with the advantages of SHAP, which provides global interpretations consistent with local explanations (Lundberg et al., 2020; Molnar, 2025). It should be noted that the notably linear relationship in Fig. 3.1b does not hold across all geographical windows. Fig. A.1 displays additional exemplary windows where the relationships exhibit less linearity. Our approach also captures nonlinearity in the system; in these cases, the linear regression helps decrease the convolved relationships as in Gryspeerdt et al. (2016). Note that unlike N_d (cm^{-3}) in panel (a), $\ln N_d$ and SST in (b) and (c) have been standardized and thus sensitivities and interaction indexes (IAIs) are expressed with the unit of cloud fraction change per standard deviation ($\text{CF } \sigma^{-1}$). Standardizing all predictors ensures that the results become comparable across all of them. We also present the SHAP dependence plots for the same example window in Fig. A.2 where non-standardized $\ln N_d$ and SST are used to plot (b) and (c). The patterns are alike and only the magnitudes of the example sensitivity and IAI are different because they are no longer expressed on a physical scale.

The vertical dispersion around the $\ln N_d$ –CF relationship captured by the SHAP dependence plot is due to the dependence of the $\ln N_d$ contribution to the predicted CF on meteorological factors (e.g. SST) in the model, which is captured by SHAP interaction values, as displayed in Fig. 3.1 (c). The colouring of the data points by SST illustrates how interactions with SST split up the $\ln N_d$ –CF relationship, with low SST values amplifying the $\ln N_d$ contribution and vice versa. To quantify this interaction effect, the meteorological data are then divided into a group of above-average feature values and a group of below-average feature values. A linear regression is fit to the $\ln N_d$ values and the SHAP interaction values in each group. An IAI is derived from these regression fits and defined as the slope for the high-value group ($> \text{mean}$) minus the slope for the low-value group ($< \text{mean}$):

$$IAI = \beta_{x,high} - \beta_{x,low} \quad (3.1)$$

where β is the slope of the linear regression between SHAP interaction values

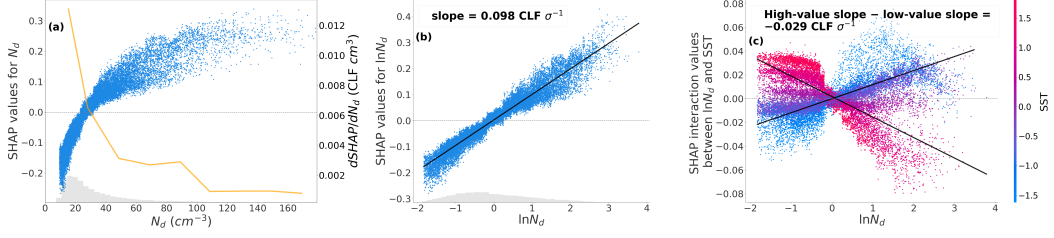


Figure 3.1: SHAP dependence plots for cloud droplet number concentration (N_d) in the region from 27.5° S to 32.5° S and from 122.5° W to 127.5° W. (a) Dots show N_d SHAP values versus N_d values. The orange line shows the change rate of N_d SHAP values with respect to N_d ($d\text{SHAP}/dN_d$) versus N_d values for each N_d bin of 20 cm^{-3} wide. (b) similar to (a) but showing the relationship between $\ln N_d$ SHAP values and $\ln N_d$ with the corresponding sensitivity defined as the slope of the linear regression. (c) SHAP interaction values coloured by sea surface temperature (SST) showing the dependence of $\ln N_d$ –CF relationship on the interactive effects of SST. The interaction values are further divided into two groups by the mean feature value of SST. Linear regressions are performed respectively for the high-value group and low-value group and the interaction index (IAI) is defined as the slope for the high-value group subtracting the slope for the low-value group. The horizontal dashed lines are a demarcation between negative and positive SHAP (interaction) values. Note that N_d in (a) is not standardized while $\ln N_d$ and SST in (b) and (c) are standardized.

and $\ln N_d$ values, and the subscripts denote the high-value group and the low-value group for a specific meteorological variable x (SST in the example), respectively. At the exemplary geographical window, the influence of SST on the $\ln N_d$ –CF sensitivity is quantified by $\text{IAI} = -0.029 \text{ CF } \sigma^{-1}$ (Fig. 3.1c). Similar to sensitivities, the unit of IAIs is also $\text{CF } \sigma^{-1}$. Therefore, for a positive sensitivity such as the $\ln N_d$ –CF sensitivity shown in Fig. 3.1b, a negative IAI value means that the $\ln N_d$ –CF sensitivity is larger with low feature values, as shown in Fig. 3.1c (the positive relationship is weakened by high SST values). On the contrary, a positive IAI value corresponds to a larger positive sensitivity with high feature values.

3.2.3.3 Limitations of observation-based machine-learning of aerosol-cloud processes

In this section, limitations of this study are discussed. A fundamental limitation of our study is that the assertion of causality from the statistical relationships of aerosols/ N_d and cloud fraction/properties is not easily done. While causal inference approaches exist and have been applied in the field of aerosol-cloud interactions (Fons et al., 2023), we employ a more traditional approach of analyzing statistical relationships of instantaneous observations (i.e. correlations). Unless

nonetheless explicitly incorporating such causal inference approaches, studies utilizing statistical or ML models to explore observational aerosol–cloud processes contend with this common limitation. For instance, some studies assessed satellite-based statistical relationships between CF and N_d (Christensen et al., 2016, 2017), between LWP and N_d (Michibata et al., 2016; Rosenfeld et al., 2019), and between N_d and other aerosol proxies (Gryspeerdt et al., 2017; McCoy et al., 2017a), all resting on statistically inferring sensitivities of cloud quantities to aerosol proxies (Forster et al., 2021). While we interpret the derived relationships with respect to the known physical relationships, uncertainties regarding the physical interpretation are mainly driven by two sources: uncertainties in the data and uncertainties from the methods.

1. Data: uncertainties exist for each satellite/reanalysis quantity, but may be particularly large in N_d . For example, the subpixel effect can introduce more bias in the N_d retrieval process within broken-cloud regimes due to increased heterogeneity. The N_d retrieval biases are discussed in Sect. 3.2.1. Also, N_d and CF observations are not fully independent, which may introduce a spurious positive correlation between the two variables. As such, we expect the physical relationship of N_d and CF to be weaker than our estimate, so that the derived sensitivities present an upper bound of the physical relationship.

Another caveat in our data is that N_d values in our study are computed using MODIS Level-3 large-scale mean r_e and τ_c values instead of joint histograms as in Gryspeerdt et al. (2016). This may introduce additional biases considering the nonlinearity of the N_d calculation. In future work, N_d data calculated from underlying joint histograms or pre-filtered data by Gryspeerdt et al. (2022) could be applied to be compared with the results in this study.

2. Methods:

- a. The exact quantification of sensitivities is dependent on the choice of the statistical/machine learning model. While for (more linearly related) monthly data, Andersen et al. (2022) have shown that XGBoost, artificial neural networks and linear models tend to lead to very similar results, this is not expected for more instantaneous data. Here, nonlinear relationships are expected, and a more complex nonlinear model is a more appropriate choice. XGBoost and other tree ensemble methods are a particularly popular choice, because of their interpretability, high accuracy considering computational efficiency (Lundberg et al., 2020)

and their ability to model the interactions between predictors (Elith et al., 2008). They have been used frequently to study aerosols and clouds before (Fuchs et al., 2018; Dadashazar et al., 2021; Andersen et al., 2021; Chen et al., 2022b; Bender et al., 2024). Besides, the TreeSHAP algorithm, specifically tailored for tree-based models to compute exact SHAPley values, can even further enhance their interpretability and has been applied in this field as well (Stirnberg et al., 2021; Zipfel et al., 2022).

- b. The quantification of sensitivities with SHAP values depends on details: the choice of the algorithm to effectively estimate Shapley values is application-specific and comes to the trade-off between being *true to the data* and *true to the model*, which relies on an observational and interventional conditional expectation, respectively (Chen et al., 2020). Here, being true to the data means respecting correlations among input features, whereas being true to the model means explaining the learned model behaviour under independent feature perturbations. The *true to the model* approach is preferable when trying to understand how an ML model makes a prediction, which requires assuming feature independence. In this study, we focus on potential mechanisms behind CF sensitivities and thus we tend to respect the correlations spread among input features (*true to the data*) (Frye et al., 2020; Chen et al., 2022a). Consequently, we suffer from the disadvantage of being *true to the data*: entangled importance attributions of correlated features e.g. a feature not explicitly used by the model for the prediction task might be assigned a non-zero contribution. Yet we refrain from the drawback of being *true to the model*—unrealistic input instances (Sundararajan and Najmi, 2020; Linardatos et al., 2021; Chen et al., 2023). Despite the inherent trade-off, SHAP approach has been employed in the context of being *true to the data* (e.g. Stirnberg et al., 2021; Zipfel et al., 2022; Li et al., 2022b).

The derived estimates of sensitivities and interactive effects in this paper should thus be interpreted with these limitations and uncertainties in mind.

3.3 Results and discussion

3.3.1 Model performance

The skills of the region-specific XGBoost models in predicting CF are evaluated by the coefficient of determination (R^2) on the unseen hold-out test data. The global weighted mean R^2 is 0.45 (about 45 % on weighted average and up to 73.57 % of the variability in CF prediction is explained) and a standard deviation of 0.10. While this means that on average, about half of the variability in CF cannot be explained by the ML models, this is expected as previous studies have shown that the performance of statistical models decreases when going from monthly to daily data (Andersen et al., 2017; Fuchs et al., 2018; Dadashazar et al., 2021), and the performance is on par with that reported by Dadashazar et al. (2021), who used ML models to predict N_d with daily reanalysis data. The models in tropical regions in the Indian Ocean and the Western Pacific explain the variability in CF relatively poorly, while XGBoost models perform well in the stratocumulus regions in the subtropics near the continents, and in the midlatitudes, particularly the southern hemispheric midlatitudes. The high skill of predicting CF in the southern hemispheric midlatitudes is in contrast to a recent study where this region has been found to be particularly difficult to model statistically with monthly data (Andersen et al., 2023). In this region, the day-to-day CF variability is high due to the large influence of synoptic-scale weather systems, and hence data at the daily resolution is more adequate to represent the CF variability in these regions.

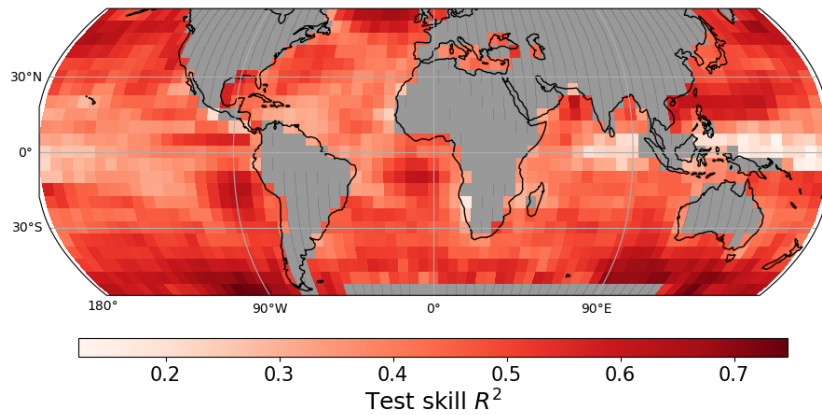


Figure 3.2: R^2 score of regional Extreme Gradient Boosting models predicting cloud fraction of marine boundary layer clouds in the independent test data set (2017–2019).

3.3.2 CF sensitivity: global perspectives and regional characteristics

3.3.2.1 Global overview of CF sensitivities

Figure 3.3 summarizes the means and distributions of the near-global sensitivities of CF to all predictors. The sensitivities are estimated as described in Sect. 3.2.3.2. The sequence is sorted by descending mean values of the absolute sensitivities (i.e. by feature importance) of the predictor variables. A strong and consistently positive N_d -CF sensitivity is found. The fact that CF is most sensitive to N_d is to be expected, as cloud observations from the same sensor are more directly related than a reanalysis product, so that their overall magnitude should not be compared (Zipfel et al., 2022). The entrainment of relatively dry air from the free troposphere into the marine boundary layer (MBL) will be impeded by a stronger inversion (i.e. higher EIS), resulting in a shallower, better-mixed and more humid MBL conducive to stratocumulus clouds (Bretherton and Wyant, 1997; Wood and Hartmann, 2006; Qu et al., 2015a; Myers et al., 2021). The salient positive sensitivity to EIS is in accordance with the links found in previous studies (e.g. Klein and Hartmann, 1993; Qu et al., 2015b; Andersen et al., 2017) suggesting that EIS is a crucial controlling factor for low-marine cloud cover. Note that in some studies the strength of the inversion over the boundary layer is measured by lower tropospheric stability, which can be regarded as a similar metric outperformed by EIS (Wood and Bretherton, 2006). Precipitation fraction is the fraction of the original ERA5 grid box covered by large-scale precipitation. The strong positive CF sensitivity to precipitation fraction is likely caused by the ML model learning that precipitation can be viewed as a proxy for cloudiness, rather than being an indicator of the physical processes via which precipitation exerts controls on the macrophysics of MLCs. Humidity shows positive CF sensitivities greater at 850 hPa, where cloud tops are often located (Gryspeerdt and Stier, 2012), than at 700 hPa which is typically in the free troposphere above the MLCs (Myers and Norris, 2013). Likewise, the atmospheric temperature at 850 hPa (t_{850}) presents stronger CF sensitivity than the temperature at 700 hPa (t_{700}). Nonetheless, in the case of winds the 700 hPa pressure level is more relevant than that at 850 hPa. A relatively pronounced negative sensitivity to the eastward wind component at 700 hPa (u_{700}) seems to indicate that reduced cloudiness is associated with stronger westerlies at this level, which, given the predominantly easterly sub-inversion flow, may increase vertical wind shear across the inversion and enhance dry-air entrainment. CF exhibits negative sensitivities to vertical pressure velocities both at 850 and 700 hPa, showing that large-scale ascending motion is connected to increases in MLCs (Myers and Norris, 2013; Bretherton et al., 2013; Blossey et al., 2013). In

general, the global averages of CF sensitivity in terms of dynamical predictors (i.e. 3-D winds at surface and pressure levels) vary in sign and are less strong. A marked negative sensitivity of CF to SST is found, which is in agreement to many prior studies (e.g. Qu et al., 2015b; Scott et al., 2020), where increases in SST have been found to lead to low cloud breakup and dissipation due to a number of processes as described in e.g. Scott et al. (2020). One of these is that the associated enhancement of mean surface latent heat flux (LHF) deepens MBL and facilitates buoyancy and thus the entrainment of dry free-tropospheric air (Rieck et al., 2012; Andersen et al., 2022). However, CF is much less sensitive to LHF than to SST. This does not necessarily imply that surface moisture fluxes are unimportant, because SST also reflects broader thermodynamic controls such as the inversion strength, and the attribution may be shared between these correlated predictors (Sect. 3.2.3.3). CF exhibits a considerable negative sensitivity to mean surface sensible heat flux (SHF), which quantifies an increase in CF with increasing SHF (upward SHF are negative). While increased SHF can promote the transition from decks of stratus or stratocumulus clouds (high CF) to more convective clouds (low CF) due to the deepening of the boundary layer (Fan et al., 2016), potentially leading to a positive SHF–CF relationship, increased SHF is associated to situations of cold air advection where turbulent surface fluxes are enhanced, which could lead to marked increases in CF (Miyamoto et al., 2018; Zelinka et al., 2018; Grise and Kelleher, 2021)

3.3.2.2 Spatial patterns of the CF sensitivity to N_d

The sensitivity of the MLC CF associated with the aerosol proxy, N_d , is ubiquitously positive, in accordance with the global correlations or sensitivities found in previous studies (e.g. Gryspeerd et al., 2016; Andersen et al., 2017). This is presumably due to the lifetime effect, but could also partially result from N_d retrieval biases discussed in Sect. 3.2.1. The global weighted mean value of the $\ln N_d$ –CF sensitivity is $0.074 \text{ CF } \sigma^{-1}$ with a standard deviation of $0.036 \text{ CF } \sigma^{-1}$. The relationship between CF and N_d is found particularly strong in regions of frequent stratocumulus to cumulus transition off the western continental coasts. These marked positive $\ln N_d$ –CF sensitivities may be caused by high N_d delaying the transition from stratocumulus to cumulus clouds (Gryspeerd et al., 2016; Christensen et al., 2020). However, as this cloud regime transition involves clouds shifting from more overcast to more broken, the strong relationships in these regions may be more affected by N_d retrieval errors. The $\ln N_d$ –CF sensitivity is also pronounced in the southern hemispheric midlatitudes, where stratiform clouds dominate. The $\ln N_d$ –CF sensitivity is weak and close to 0 in the tropics, in particu-

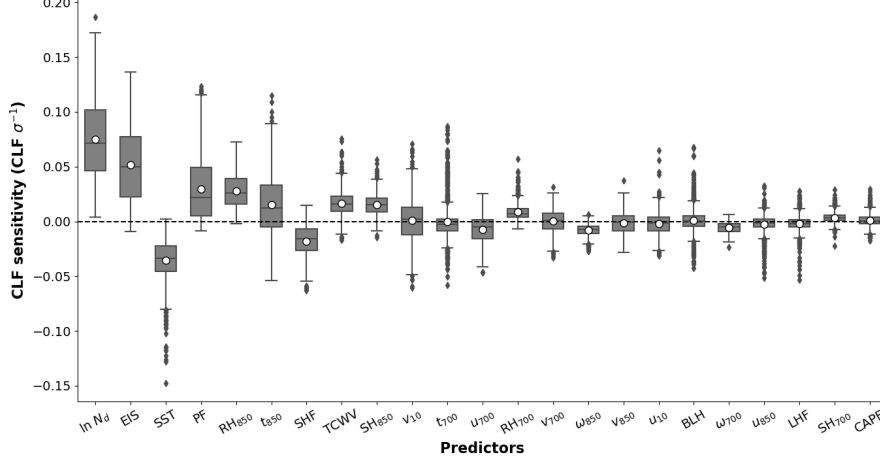


Figure 3.3: The distribution of the sensitivities of cloud fraction to all predictors as listed in Table 3.1. Boxes represent the interquartile range extended by whiskers up to 1.5 interquartile ranges, with outliers shown as points outside this range. The solid line and white dot in each box show the median and mean sensitivity values, respectively. Predictors are sorted by the mean absolute sensitivity value. The dashed line differentiates positive and negative sensitivity values.

lar in the deep convective warm pool region. These spatial patterns of $\ln N_d$ –CF sensitivity resemble those found by Gryspeerd et al. (2016), in particular the ones where they mediated the aerosol optical depth–CF relationship by N_d , but are more pronounced in the southern hemispheric midlatitudes. This difference in estimated sensitivity seems noteworthy and should thus be investigated in future work. As N_d retrievals tend to be negatively biased at lower CF and positively biased at higher CF, the $\ln N_d$ –CF sensitivity may be overestimated, and at the scales considered here, should be interpreted as an upper bound to the physical $\ln N_d$ –CF sensitivity. The global weighted average of the $\ln N_d$ –CF sensitivity without standardization is 0.112 (unitless), and its spatial pattern is shown in Fig. A.4. This value is higher than the upper bound of 0.1 reported by Bellouin et al. (2020), which is based on global climate models and large eddy simulations. This may be partly due to the aforementioned bias. However, it is important to note that our non-standardized $\ln N_d$ –CF sensitivity, shown in Fig. 3.1a, closely mirrors that from Yuan et al. (2023) with a similar range. In addition, the high $\ln N_d$ – $\ln CF$ values estimated in Chen et al. (2022b, 2024) suggest that values exceeding the upper bound of 0.1 might be plausible. These recent observational studies, including quantifying CF adjustment based on ship tracks (Yuan et al., 2023), volcano aerosol perturbations (Chen et al., 2022b, 2024), and our SHAP approach using global satellite observations, indicate

that the 0.1 upper bound may be extended. In future work, estimating a radiative forcing using the SHAP-based sensitivities will make our study more comparable with other research on CF adjustment.

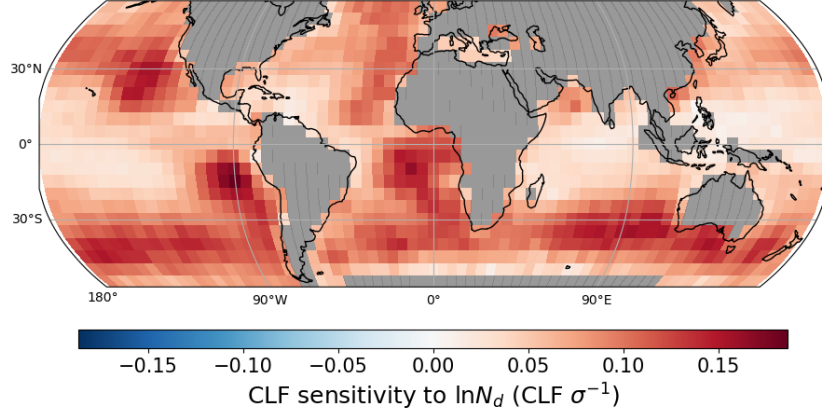


Figure 3.4: Sensitivity of the marine low cloud fraction to $\ln N_d$.

3.3.2.3 Spatial patterns of the CF sensitivity to thermodynamic drivers

There has been a strong consensus that EIS and SST are the two important determinants of CF of MLCs and their corresponding radiative effects across different geographical regions and on varying time scales (e.g. Bretherton, 2015; Myers and Norris, 2015; McCoy et al., 2017b; Wall et al., 2017). Stronger inversions capping MBL (i.e. higher EIS) will hamper the entrainment of aloft dry air from the troposphere and thus lead to a shallower MBL and more moisture trapped within MBL, promoting the development and maintenance of low-level clouds (Andersen et al., 2017). The regional EIS–CF sensitivity patterns (Fig. 3.5a) show that marine-low CF increases ubiquitously in response to stronger EIS, in particular in the tropical and subtropical stratocumulus-capped regions and within the midlatitudes. The sensitivity pattern is in good agreement with that found by Scott et al. (2020) and Andersen et al. (2023), related studies at different time scales (Grise and Medeiros, 2016; Kelleher and Grise, 2019; de Szoeke et al., 2016).

MLC cloud cover reduces globally in response to increased SST, particularly pronounced in the stratocumulus regions over eastern oceanic basins (Fig. 3.5b, consistent with Scott et al. (2020)). SST can favour MLC dissipation through increasing surface LHF and deepening MBL, facilitating dry entrainment and eventually desiccating the MBL and clouds (Rieck et al., 2012; Qu et al., 2015b). Yet as stated in Sect. 3.3.2.1, the weak CF sensitivity to LHF in relation to the strong sensitivity to SST may imply that the other process makes more substantial contributions,

namely that the higher moisture gradient between the troposphere and MBL arising from the increased SST makes the entrained air more efficient in evaporating cloud water (van der Dussen et al., 2015; Qu et al., 2015b). This process has been shown to be the driving mechanism for the observed reduction in marine low cloud cover near the coast of Baja California (Andersen et al., 2022).

Figure 3.5c shows that the CF of MLCs increases with negative (upward) SHF most markedly in the stratocumulus regions. CF can increase in response to increased surface fluxes in situations of cold air advection (Zelinka et al., 2018). Over the South Indian Ocean, a marked SHF–CF sensitivity is also found. Here, enhancements of SHF due to the subtropical anticyclone and midlatitude storm-track activity have been found to increase CF (Miyamoto et al., 2018). The results may be a hint that the increase of CF presumably due to increased SHF (e.g. due to cold air advection) outweighs the influence of SHF on CF by controlling the transition from marine stratocumulus to open-cellular marine clouds (Kazil et al., 2014; Fan et al., 2016) in the core stratocumulus regions. Consequently, the SHF–CF sensitivity is less pronounced in regions of frequent closed- to open-cell and cumulus transitions. Relative humidity at 850 hPa (RH_{850}) is positively related to marine-low CF across the globe. The positive sensitivity is particularly strong in the trade cumulus regions where the 850 hPa level is representative of the boundary layer. In the coastal stratocumulus regions, clouds are frequently below this level (Adebiyi and Zuidema, 2016), so that clouds are not as sensitive to variability in RH at that level.

3.3.2.4 Spatial patterns of the CF sensitivity to dynamical drivers

Large-scale circulations and dynamical conditions play an essential role in controlling CF and the indirect effects of aerosols (Su et al., 2010; Small et al., 2011). The large-scale dynamics are represented by the horizontal and vertical winds at 700 hPa and 850 hPa, which display clear and distinct regional patterns of CF sensitivity (Fig. 3.6). It can also be seen that at the considered scales and pressure levels, horizontal wind vectors have stronger CF sensitivities than large-scale vertical motion. There is a coherent pattern of negative CF sensitivity to the zonal wind at 700 hPa in the stratocumulus-dominated regions (also apparent at 850 hPa), and the southern hemispheric midlatitudes, indicating a decrease in MLCs with westerly anomalies at this pressure level. Recently, a study using monthly data has also found a similar sensitivity pattern of stratocumulus clouds to zonal wind at 700 hPa, finding that the reduced CF is related to increased vertical wind shear (as the boundary layer flow is easterly), leading to increased turbulence and dry-air entrainment (Andersen et al., 2023). Using monthly data, Andersen et al. (2023)

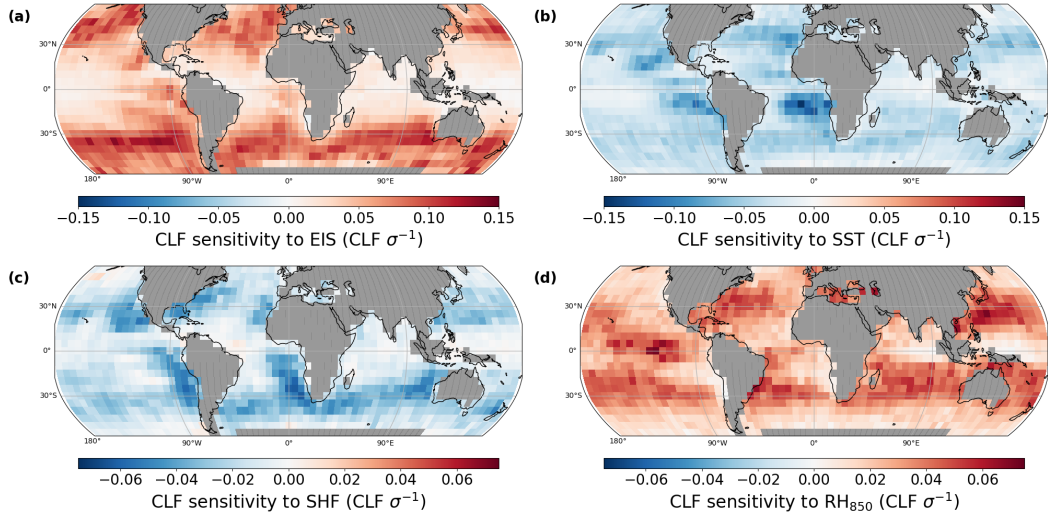


Figure 3.5: Spatial distribution of the sensitivity of marine low-cloud fraction (CF) to estimated inversion strength (EIS), sea surface temperature (SST), sensible heat flux (SHF), and relative humidity at 850 hPa (RH_{850}). Note that the range of colourbars of SHF and RH_{850} (-0.075 – 0.075) is narrower than EIS and SST (-0.15 – 0.15).

did not find a similar CF sensitivity to zonal winds in the southern hemispheric midlatitudes, though. As the CF sensitivity to u_{700} in the southern hemispheric midlatitudes is only apparent using daily data and only at 700 hPa, it seems likely that it is related to synoptic variability that drives day-to-day variability in MLCs in this region (Kelleher and Grise, 2019). Positive CF sensitivities to u_{700} (higher CF with westerly anomalies) and to a lesser degree u_{850} are found off the eastern Asian and North-American continents. CF increases due to cold-air outbreaks in NW Atlantic and NW Pacific may be the reason for these positive sensitivities. Cold-air outbreaks occur during winter as cold continental air moves over warmer SSTs, increasing moisture and heat fluxes into the MBL so that the formation of MLCs is favoured (Young et al., 2002). This leads to wintertime maxima in CF in these regions (Yuan and Oreopoulos, 2013).

The sensitivity of CF to the meridional winds at 700 hPa exhibits two bands straddling the subtropical regions between about 15° and 35° in both hemispheres but opposite in sign (positive in the Northern Hemisphere and negative in the Southern Hemisphere), illustrating that in these regions the poleward winds are associated with an increase in low cloud fraction. The bands are still apparent at 850 hPa, while the negative band in the Southern Hemisphere extends northward to tropical areas. These hemispheric sensitivity bands to the v wind component at 700 hPa closely resemble those found in Andersen et al. (2023), with their analysis

suggesting that the poleward winds on the eastern side of midlatitude cyclones may be related to warm and moist advection, increasing CF. However, they also find a strong correlation of these free-tropospheric poleward winds with large-scale ascending air motion making the assertion of causality difficult. Poleward winds are also found to decrease CF over the southern hemispheric midlatitudes.

CF is negatively connected to the vertical pressure velocity both at 700 hPa and 850 hPa (ω_{700} and ω_{850}) over the entire Earth, indicating that ascending large-scale air motion enhances the cover of MLCs globally. It is shown in the bottom of Fig. 3.6a that the CF sensitivity to ω_{700} is larger in the midlatitude ocean basins, whereas the CF sensitivity to ω_{850} is larger in the subtropical oceans where subsidence is climatologically prevalent (Myers and Norris, 2015, 2016; Scott et al., 2020). This seems indicative of CF being most sensitive to large-scale ascending motion at the typical altitude of the clouds. It is interesting to note that between 30° N and 30° S, no marked CF sensitivity to ω_{700} is found, contrasting the finding of enhanced subsidence at this level reducing MLCs by Myers and Norris (2013). This effect is likely better described in the ω_{850} data which is more related to the altitude of the cloud top.

3.3.3 Dependence of N_d -CF relationship on meteorology

3.3.3.1 Global overview of the interaction indices

In this section, we use the IAI as defined in Sect. 3.2.3.2 to quantitatively show how the CF response in MLCs attributed to the aerosol proxy N_d varies with the meteorological factors. As discussed in Sect. 3.2.3.2, since the sensitivity related to N_d is positive across the globe (Fig. 3.4), a positive IAI can be interpreted as an amplification of the $\ln N_d$ -CF sensitivity with high (above-average) feature values of a meteorological variable, whereas a negative IAI signifies an amplification of the sensitivity at low feature values.

In Fig. 3.7, analogous to Fig. 3.3, the features along the x-axis are arranged in descending order based on their averaged absolute IAIs i.e. by the strength of the impact of each meteorological feature on the $\ln N_d$ -CF sensitivity. Similar to the feature importance summarized by Fig. 3.3, EIS, SST, RH_{850} and SHF have relatively large strength of interaction effect and thus can be regarded as critical controlling factors not only for marine low cloud cover but also for their response to changes in N_d (and in extension aerosols). Compared to the CF sensitivities, the IAIs associated with atmospheric temperatures at 700 and 850 hPa have greater strengths. Furthermore, it can also be seen that the vertical and horizontal winds at the surface and different pressure levels are ranked generally lower. In general,

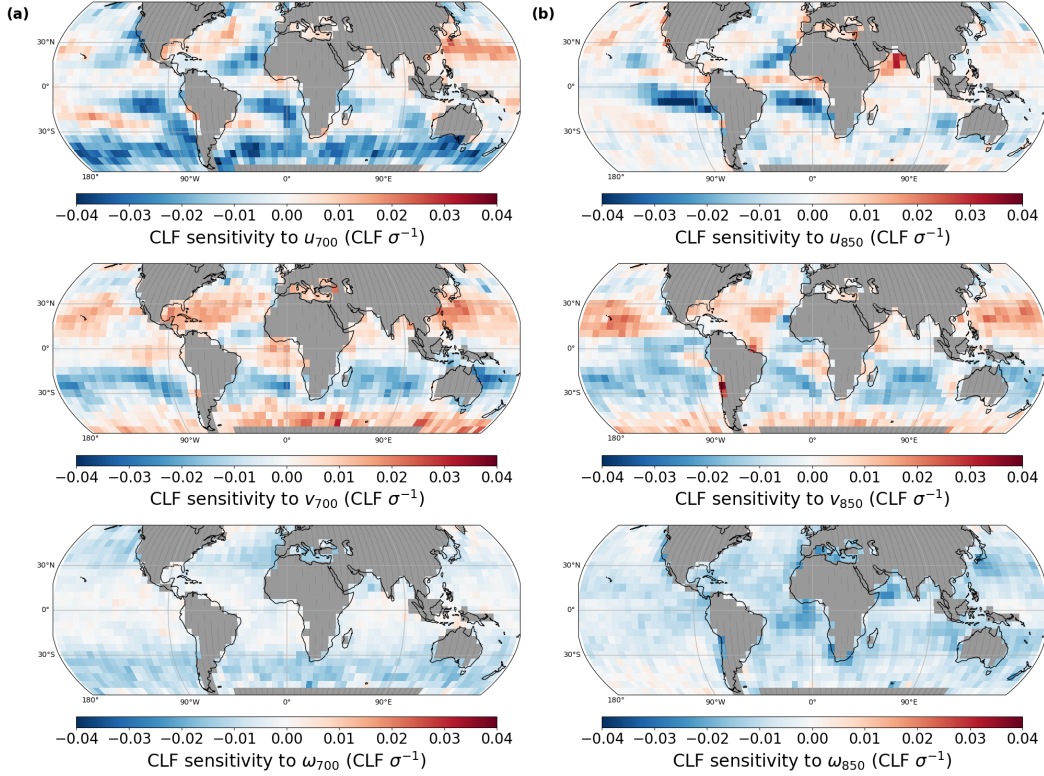


Figure 3.6: Sensitivity of cloud fraction to u , v wind component vectors and vertical velocities at 700 hPa (column (a)) and 850 hPa (column (b)). Note that the range of the colourbars is in general smaller (-0.04 – 0.04) than in Fig. 3.5.

the thermodynamic factors seem to have a stronger influence on the $\ln N_d$ –CF sensitivity than the dynamical factors.

3.3.3.2 Spatial patterns of the interaction indices

Coherent and distinct spatial distributions of the impact of selected meteorological parameters on the N_d –CF relationship can be observed. Hereafter, we show the regional characteristics of the interaction effects of EIS and SST, which are the two most important meteorological factors for CF in MLCs and have the greatest absolute strengths of IAI. EIS exerts the most noticeable positive IAIs over the mid-latitude oceanic areas (Fig. 3.8a), reflecting that stronger temperature inversions capping the MBL over these regions may amplify the positive N_d –CF relationship. The interpretation of possible underlying physical mechanisms of these interaction effects is difficult and remains speculative. The results seem to suggest that in these regions, potentially through hampering the entrainment of drier air from the free troposphere, the stronger inversion and more stable conditions are capable

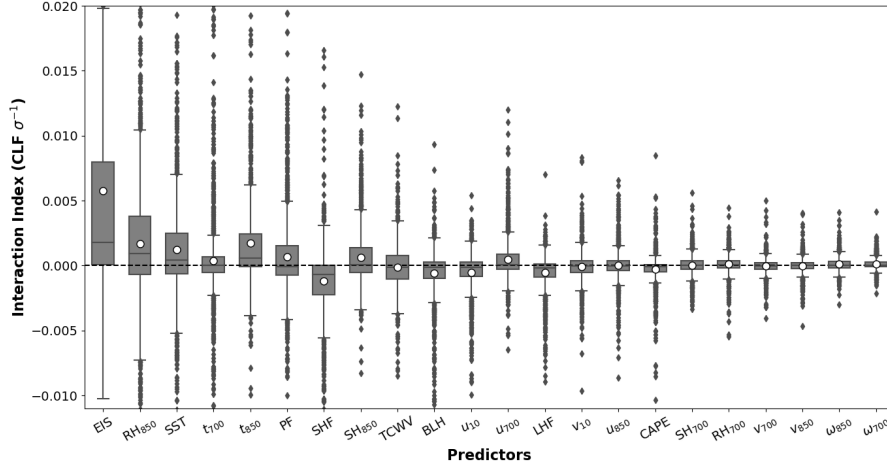


Figure 3.7: Similar to Fig. 3.3 but for the interaction effect of N_d with all meteorological predictors, quantified by the interaction index ($\text{CLF } \sigma^{-1}$).

of trapping more moisture within a shallower MBL and could thus weaken the evaporation-entrainment feedback. As a result, it may ultimately favour a more positive N_d –CF relationship (Chen et al., 2014; Christensen et al., 2020). It is interesting to note that these interactions are not apparent in the stratocumulus regions where EIS is a strong control of CF, and in the stratocumulus-to-cumulus transition regions, where Christensen et al. (2020) found the aerosol effect on this transition to be confined to stable atmospheric conditions. These findings suggest that the proposed entrainment mechanism may operate only over a limited EIS range: sufficient stability may be required, but the interaction does not necessarily strengthen monotonically with EIS and appears most clearly at the intermediate EIS values typically found in the midlatitudes (Scott et al., 2020), rather than at the higher values characteristic of the stratocumulus regions. The observed impact of EIS on the N_d –CF relationship found in the midlatitudes may also have implications within the context of climate change. While in the subtropics, global climate models predict an increase in EIS with a warming climate, in the midlatitudes EIS is predicted to decrease (Myers et al., 2021), the positive IAs found here suggests that decreasing EIS in the midlatitudes could weaken the amplification of the positive N_d –CF relationship and thus reduce the sensitivity of CF to $\ln N_d$ there.

Fig. 3.8b shows that higher SSTs are found to amplify the positive N_d –CF relationship (positive IAI) in the regions of frequent stratocumulus-to-cumulus transition (Cesana and Del Genio, 2021). The physical interpretation could be: Here, higher SSTs tend to lead to the transition from stratocumulus clouds to

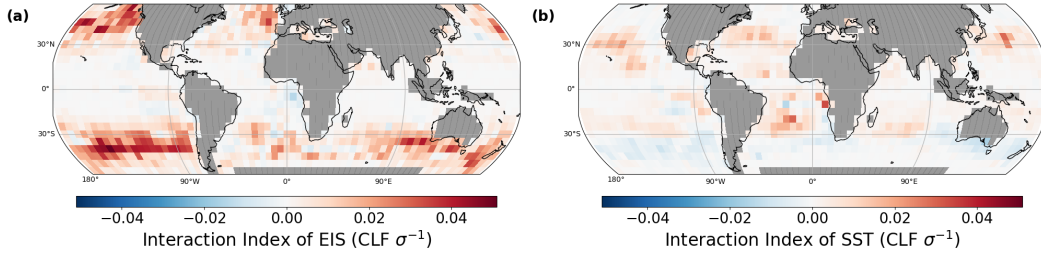


Figure 3.8: Spatial patterns of the interaction index showing the dependence of the $\ln N_d$ –CF relationship on estimated inversion strength (EIS) (a) and sea surface temperature (SST) (b).

shallow convective clouds (Cesana et al., 2019), however, this transition has been found to be delayed when aerosol is increased (Goren et al., 2019; Christensen et al., 2020). Tentatively, the positive IAs in these transition regions may thus point to increased control of N_d on CF at higher SST values, as these are the situations where transitions typically occur and when increased N_d can act to delay this transition. Since SST is robustly projected to increase under warming, higher future SSTs in these regions might increase the sensitivity of MLC CF to N_d , assuming that the interaction inferred here from present-day day-to-day variability also applies to a warmer mean state. It should be noted that the quantification of the dependence of the N_d –CF relationship on meteorological factors (EIS, SST discussed in this section) is also likely subject to the biases in the $\ln N_d$ –CF sensitivity caused by the N_d retrieval biases as a function of CF. This would potentially contribute to the non-causal facets of the relationships and interactive effects quantified by SHAP values.

3.4 Specific conclusions

In this study, nine years (2011–2019) of daily satellite and reanalysis data have been analyzed to better understand the effect of N_d on CF in MLCs, and its dependence on meteorological factors. We have established a near-global ML framework to predict the CF of MLCs using region-specific XGBoost regression models. Including many confounding and influencing factors as a whole, the explainable ML technique of SHAP regression values has been used to explain the regional XGBoost models, to quantify the CF sensitivity to all cloud controlling factors with a specific focus on N_d ; and, moreover, to quantify the meteorological influence on the N_d –CF relationship at a global scale. The statistical sensitivities and interactive effects are interpreted with the guidance of hypothesised causal pathways and the

state-of-the-art physical understanding of the system. The main findings of this study, which should be interpreted in light of the data and methodology limitations discussed in Sect. 3.2.3.3, are summarized as follows:

1. MLC CF shows a notable positive sensitivity to N_d (a surrogate for aerosols) in the regions of stratocumulus to cumulus transition, which may arise from the high N_d delaying this transition. The $\ln N_d$ -CF sensitivity in the southern hemispheric midlatitudes is observed to be higher than in previous studies, which should be investigated in future work. The estimated $\ln N_d$ -CF sensitivity and its magnitude suggest that aerosols likely have a considerable impact on MBL cloudiness, although this may partially result from an overestimation caused by the effect of a positive retrieval bias of N_d at high CF.
2. Consistent with the literature, our statistical method shows that EIS and SST are two important determinants for MLCs by regulating surface fluxes and dry-air entrainment processes. In addition, strong negative CF sensitivity and spatial patterns for SHF are also found, suggesting that the effect of cold air advection might surpass the SHF enhancement of closed- to open-cell and cumulus transitions. Dynamic drivers (meridional and zonal winds) indicate that midlatitude synoptic-scale disturbances and vertical wind shear seemingly make considerable contributions to MLC amounts.
3. In general, thermodynamic parameters exert a more important influence on the N_d -CF relationship than dynamical parameters. EIS, RH_{850} , SST, temperatures at 700 and 850 hPa have the strongest effect on the N_d -CF sensitivity. In the midlatitudes, higher EIS is found to amplify the positive $\ln N_d$ -CF sensitivity, which may be related to a reduced entrainment feedback in these conditions, whereas higher SST is found to amplify the $\ln N_d$ -CF sensitivity in stratocumulus-to-cumulus transition regions, potentially because the transition induced by higher SSTs may be delayed by increased N_d . These findings have potential implications for possible future changes in the sensitivity of CF to aerosols.
4. For the dynamical and thermodynamic factors shown here, both CF sensitivities and the interactive effects (dependence of N_d -CF relationship on meteorology) exhibit distinct regional patterns. These coherent spatial patterns indicate that the proposed explainable ML framework is not only capable of skilfully predicting CF for MLCs but also has the potential to

capture regional characteristics of the relation between CF and N_d , as well as meteorological influences.

In the future, the observation-based sensitivities and interactive effects quantified by the ML framework here will be compared to those in global climate models (GCMs), which have the potential to evaluate GCM parameterizations related to ACI and even help gain insights into how the models could be tuned in this respect. In addition, incorporating causal approaches for SHAP, such as proposed by Heskes et al. (2020); Frye et al. (2020), would help to test the extent to which the observed statistical relationships and interaction effects represent physical processes.

Chapter 4

ICON-HAM based cloud adjustment

4.1 Specific motivation and aim

Over the past decade, systematic differences in the magnitude of cloud adjustments have been found between models and observations (Gryspeerdt et al., 2020; Quaas et al., 2024). In the most recent two eras of the Coupled Model Intercomparison Project (CMIP5 and CMIP6), global climate models (GCMs) consistently show strong cooling driven by the Twomey effect and liquid water path (LWP) adjustments (Smith et al., 2020; Zelinka et al., 2023). By comparison, observational studies generally find a weak or ambiguous LWP response to aerosol perturbations (Malavelle et al., 2017; Bender et al., 2019; Toll et al., 2019).

For the cloud fraction (CF) adjustment, observational studies agree on the sign but disagree on the magnitude of the CF sensitivity to aerosol (proxies) or the resulting negative forcing (Gryspeerdt et al., 2016; Andersen et al., 2017; Fuchs et al., 2018; Rosenfeld et al., 2019; Wall et al., 2022; Jia et al., 2024). GCMs agree with observational studies in terms of the positive relationship between cloud cover and aerosols and therefore a negative radiative forcing from the CF adjustment (Zelinka et al., 2014; Forster et al., 2021), as autoconversion parameterizations in many of these models allow aerosols and thus cloud droplet number concentration (N_d) to suppress precipitation and thereby prolong cloud lifetime (Bellouin et al., 2020). However, while models agree on the sign of the CF adjustment, they diverge on the magnitude and relative importance of the Twomey effect, the LWP adjustment and the CF adjustment.

GCMs simulate a minimal or negligible CF contribution relative to the Twomey

effect and LWP adjustment (Smith et al., 2020; Zelinka et al., 2023). Even when diagnosed using a consistent cloud radiative kernel method, multiple CMIP6 models suggest a CF adjustment being weaker than both the Twomey effect and LWP adjustment (Duran et al., 2025), whereas the same framework applied to satellite observations indicates that the CF adjustment outweighs both (Wall et al., 2023). Consistent with this, a recent model–observation comparison (Wang et al., 2025) showed that multiple GCMs failed to reproduce the strong aerosol-induced increase in cloud cover associated with the Holuhraun volcanic eruption documented by Chen et al. (2022b). Taken together, the discrepancies in the magnitudes and relative importance of LWP and CF adjustments suggest that LWP adjustments are likely overestimated and CF adjustments are underestimated in GCMs. In particular, recent observational evidence increasingly highlights the CF adjustment as potentially the largest contributor to effective radiative forcing due to aerosol–cloud interactions (ACI) (e.g. Chen et al., 2022b; Yuan et al., 2023; Wall et al., 2023; Chen et al., 2024), providing a key motivation for this present work. Moreover, the CF response to aerosols may saturate under already polluted conditions, further complicating its quantification. For example, Wang et al. (2024) found over high-latitude oceans that the CF responses saturate once N_d exceeds around 60 cm^{-3} , suggesting that background cloud conditions influence the CF adjustment.

The challenge comes from the fact that observational constraints on the CF adjustment often rely on opportunistic experiments, such as ship tracks and volcanic events, which can rule out confounding meteorological covariates. However, ship tracks are typically subscale for GCMs, while volcanic eruptions can be episodic and often involve uncertain emissions, making the quantification challenging. Therefore, both approaches may lack global representativeness. An alternative is to use large-scale satellite studies, which are spatiotemporally representative and good for constraining GCMs, but use different ways of dealing with meteorological confounders. For example, Gryspeerdt et al. (2016) has attempted to address this confounding issue through statistical control of confounding variables such as relative humidity (RH), or Andersen et al. (2023) has incorporated them into a machine learning (ML) framework using ridge regression.

In a previous study (Jia et al., 2024, hereafter J24), we quantified the CF adjustment as the sensitivity of marine low cloud (MLC) CF to N_d using an explainable ML framework trained on MODerate Resolution Imaging Spectroradiometer (MODIS) observations and fifth generation European Centre for Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis (ERA5) reanalysis data. This

eXtreme Gradient Boosting (XGBoost) and SHapley Additive exPlanations (SHAP) framework demonstrated global positive N_d –CF relationships.

In this study, we extend the explainable ML framework of J24 to Atmospheric Model Intercomparison Project (AMIP) simulations with the ICosahedral Nonhydrostatic (ICON) atmosphere model coupled with the Hamburg Aerosol Module (HAM) (ICON-HAM). The main goals of this study are twofold: (1) to enable a consistent comparison between observation- and model-based N_d –CF sensitivities; (2) to investigate how structural model choices shape the simulated CF response. To achieve this, we conduct six simulations that combine two cloud-cover schemes (CC-RH, dependent only on RH, and CC-RH-LWC, additionally dependent on cloud water content) with three prescribed lower bounds for cloud droplet number concentration ($N_{d,\min} = 10, 40$ and 100 cm^{-3}), examining their joint impact. As in J24, we quantify the sensitivity of MLC CF to log-transformed N_d ($\ln N_d$) using regional XGBoost models with SHAP, extended via a piecewise linear regression (PLR) to accommodate the stronger nonlinear response in ICON-HAM. Guided by these goals, our hypotheses are: (1) the $\ln N_d$ –CF relationships diagnosed from ICON-HAM differ quantitatively, or maybe even qualitatively, from the MODIS-based relationships in J24; (2) given the stronger coupling between cloud water content and cloud cover in CC-RH-LWC, we expect this scheme to exhibit larger $\ln N_d$ –CF sensitivities than the RH-only scheme with prescribed $N_{d,\min}$ expected to further modulate these sensitivities differently in each scheme.

4.2 Data and methods

4.2.1 ICON-HAM experimental setup

4.2.1.1 ICON-HAM general model description

The host model in this study is the ICON atmospheric model (Giorgetta et al., 2018), coupled with HAM to obtain a global atmosphere-aerosol model. The overall coupled model configuration largely follows that described in Salzmann et al. (2022), although we use a more recently developed ICON version 2.6.4 combined with the same HAM version 2.3 as in their study. Hereafter, without explicit notation, ICON-HAM refers to ICON2.6.4-HAM2.3. The ICON dynamical core provides a nonhydrostatic, compressible atmosphere on an icosahedral grid. HAM simulates aerosol properties such as size distributions and couples them to clouds and radiation, enabling process-level links between aerosol perturbations and cloud responses. In this study, the ICON-HAM model uses an R02B04 horizontal grid, corresponding to a grid spacing of approximately 160 km between neighbouring

grid points, with 47 vertical levels using a hybrid sigma-height coordinate system. A code fix to the total turbulent energy scheme has been implemented to correct an issue introduced when porting from ECHAM to ICON, which caused excessive vertical diffusion in earlier model versions (Giorgetta et al., 2022; Franke and Giorgetta, 2024). This correction ensures a more realistic turbulent contribution to updraft velocity and thus cloud droplet activation.

To explore how CF sensitivity to aerosol perturbations is represented in ICON-HAM, we use N_d as a proxy for cloud-active aerosol, consistent with recent satellite-based studies (e.g. Gryspeerd et al., 2016; Christensen et al., 2020; Chen et al., 2024). All simulations in this study follow the AMIP protocols, with sea surface temperature (SST) and sea ice prescribed from observations (Taylor et al., 2000). We conduct six simulations that pair two different cloud-cover parameterization schemes with three different $N_{d,min}$. The following subsections detail the specific configurations of these simulations.

4.2.1.2 Cloud cover and autoconversion parameterization schemes

A central aspect of this study involves comparing two parameterizations for fractional cloud cover that fundamentally differ in whether cloud water directly controls cloud cover. Throughout this study, we refer to these schemes as CC-RH, which depends only on grid-mean RH, and CC-RH-LWC, which additionally depends on the total cloud water content.

CC-RH follows the implementation in the ECHAM6.3 general circulation model as described by Mauritsen et al. (2019), and is also the default configuration in ICON-HAM. In this scheme, CF is diagnosed as a function of grid-mean RH (Sundqvist et al., 1989):

$$CF = 1 - \sqrt{1 - \frac{RH - RH_0}{1 - RH_0}} \quad (4.1)$$

where RH_0 is a critical threshold RH specified as a function of height, as originally formulated in ECHAM4 (Roeckner et al., 1996). Clouds begin to form within a grid cell once RH_0 is exceeded. Further details on the parameter settings and tuning of CC-RH can be found in Mauritsen et al. (2019).

The CC-RH-LWC scheme follows the parameterization proposed by Xu and Randall (1996), previously referred to as COUPL-CC by Lohmann and Feichter (1997). In this scheme, CF increases with RH and additionally with the cloud water content q_w (the sum of liquid and ice water mixing ratios, which is predominantly liquid for MLCs) through an exponential factor:

$$CF = RH^{0.25} \left[1 - \exp \left(-250 [(1 - RH)q_{vs}]^{-0.45} q_w \right) \right] \quad (4.2)$$

with q_{vs} being the saturation specific humidity. The key distinction is that CC-RH-LWC explicitly depends on cloud water content, whereas CC-RH does not. This fundamental difference allows aerosol-induced changes in autoconversion and precipitation, which affect cloud water and hence LWP, to directly feed back on CF in CC-RH-LWC, a process that is absent in CC-RH.

All experiments use the autoconversion parameterization scheme of Khairoutdinov and Kogan (2000), derived from bin-resolving microphysics in large-eddy simulations (LES) of marine stratocumulus. Although originally developed for LES applications, it has been adopted in many GCMs. In what follows, this scheme is referred to as KK2000. The KK2000 autoconversion rate has a relatively strong inverse dependence on N_d ($N_d^{-1.79}$), and it scales with cloud liquid water content (LWC), allowing for interactions with the cloud-cover schemes described above.

4.2.1.3 Prescribed $N_{d,min}$

It is a common practice in global aerosol-climate models to impose $N_{d,min}$ throughout the modelled atmosphere at all locations and altitudes. This constraint is typically imposed to avoid unrealistically low $N_{d,min}$ values that may arise from the incomplete representation of natural background aerosol species that could contribute to cloud condensation nuclei (CCN) and N_d (e.g. nitrate and biological particles) (Kirkevåg et al., 2008), or from potential underprediction of specific aerosol sources (e.g. fine-mode sea salt) (Wang and Penner, 2009).

However, this practice is unphysical and the specific choice of the lower bound value is somewhat arbitrary. Hoose et al. (2009) systematically investigated the influence of these lower bounds and found that they can suppress the simulated aerosol indirect effect by up to 80 % by artificially creating more uniform N_d fields. By design, these N_d constraints are triggered more frequently where low N_d is more common, even though the observed N_d can drop well below the imposed threshold. This reduces the contrast between clean and polluted regimes. Consequently, under preindustrial (PI) emissions the impact of prescribing $N_{d,min}$ is comparably large over land and ocean, whereas under present-day (PD) emissions, it is most evident over oceans (Hoose et al., 2009).

Typical values of this lower bound have ranged between 5 and 40 cm^{-3} across different models (Hoose et al., 2009). ECHAM4 and ECHAM5 have used a $N_{d,min}$ value of 40 cm^{-3} (Lohmann et al., 1999, 2007), and ECHAM6.3–HAM2.3 added an additional option of 10 cm^{-3} (Neubauer et al., 2019). In our study, in addition

to 10 and 40 cm^{-3} , we also impose a more stringent $N_{\text{d,min}}$ of 100 cm^{-3} to further examine how a prescribed minimum N_{d} interacts with CC-RH and CC-RH-LWC cloud cover schemes and modulates the CF sensitivity, yielding six experiments in total.

4.2.1.4 ICON-HAM output data as machine-learning input

In this study, we apply an explainable ML framework similar to that used in J24 (described below in Sect. 4.2.2), where N_{d} along with large-scale meteorological fields are used to predict CF. The data used as input for the XGBoost models here are ICON-HAM model outputs. Predictors for the XGBoost models are listed in Table 4.1. To facilitate direct comparison with our MODIS-based study J24, we remapped ICON-HAM outputs on the native icosahedral grid to the MODIS $1^\circ \times 1^\circ$ latitude-longitude grid using nearest-neighbour interpolation. The original 47 vertical levels in ICON-HAM’s hybrid sigma-height coordinate system were interpolated to six desired standard pressure levels (1000, 950, 900, 850, and 700 hPa) using conservative remapping. Owing to storage and computational constraints associated with producing global hourly output, the analysis is restricted to five years of data (2011–2015).

The low-level CF used here to approximate MLCs is diagnosed as the fractional cover of liquid clouds with cloud-top temperature (CTT) warmer than 268 K, as viewed from the top of the atmosphere whenever high- and mid-level clouds were absent. This “unobscured” diagnostic is considered more suitable for analyzing drivers of liquid cloud cover, because the coarser resolution of ICON-HAM relative to MODIS increases the likelihood of overlying clouds masking the true extent of the low-level CF. Unlike our observational study (J24), where N_{d} was derived specifically for satellite retrievals from cloud droplet effective radius (r_{e}) and cloud optical depth (τ_{c}), here we use the model’s instantaneous cloud-top, in-cloud N_{d} diagnostic. To improve comparability with J24, we apply analogous filtering criteria here: Low clouds must have a CTT warmer than 268 K; cases with a cloud-top in-cloud r_{e} smaller than 4 μm or a τ_{c} smaller than 4 are excluded.

To characterize the dynamical and thermodynamic state of the atmosphere, the ML framework uses 23 meteorological variables in addition to $\ln N_{\text{d}}$ as predictors in the ML model. This selection largely follows J24 except for several adaptations described below. A full list of predictors is provided in Table 4.1. We diagnose boundary-layer height (BLH) as the lowest model level at which the mean RH first drops below 50 %, following a standard LES-based proxy for the stratocumulus top and inversion height (Bretherton et al., 2013), which effectively captures the

sharp transition from the moist boundary layer to the dry free troposphere above. This threshold has been used in a recent ICON-based analysis of stratocumulus adjustments and was shown to align well with the modelled cloud top (Fons et al., 2024). Convective available potential energy (CAPE) was dropped from the predictor set because it is primarily relevant for deep convective processes rather than for the stratiform and shallow cumulus marine boundary-layer clouds that are the focus of this study. Moreover, in addition to RH at 700 and 850 hPa, we include two more lower pressure levels, 900 and 950 hPa, which are considered more relevant for MLCs.

Unlike J24, we do not standardize the predictor variables in this work. In J24, standardization placed all predictors on a common and unitless scale to allow direct comparisons among all predictors with different units. However, our objective here is to compare the $\ln N_d$ -CF sensitivity across the six ICON-HAM configurations (two different cloud-cover schemes and three $N_{d,\min}$ values), rather than to rank predictors by relative importance. We hence retain predictors on their native physical scales to preserve the interpretability of ICON-HAM’s CF responses across six experiments in this study.

4.2.2 Explainable machine learning framework

4.2.2.1 Regional tree-based machine-learning models

We adopt the explainable machine learning framework from J24 to quantify the sensitivity of MLC CF to $\ln N_d$ (proxy for aerosol) while accounting for meteorological covariability and confounding effects. A tree-based ensemble XGBoost algorithm (Chen and Guestrin, 2016) is employed due to its ability to capture nonlinear relationships and model feature interactions, while achieving high predictive performance with computational efficiency for tabular data. As in J24, an independent XGBoost model is trained within each $5^\circ \times 5^\circ$ geographical window to predict CF, using spatiotemporal samples aggregated from $1^\circ \times 1^\circ$ grid cells (remapped from native icosahedral grids) to capture regional characteristics and relationships.

For better comparability with our MODIS-based study (J24), we configure ICON-HAM to output hourly variables, then match them with MODIS Terra overpass times. The XGBoost models are trained on these outputs from 2011 to 2015 (Sect. 4.2.1.4). Unlike J24, where the train–test split was purely chronological, here we ensure that different climate states are represented in the training data. To sample distinct ENSO phases, we train the models on a La Niña year (2011), a neutral SST year (2013), and an El Niño year (2015), consistent with the documented ENSO

conditions during these periods (Null, 2025). Model testing is performed on the intermediate years 2012 and 2014. The spatial patterns of annual SST anomalies relative to the five-year mean are shown in Fig. B.1.

As in J24, hyperparameters for each regional XGBoost model are tuned via Bayesian optimization with five-fold cross-validation, and the number of boosting rounds is determined by early stopping to prevent overfitting. In this study, we expand the hyperparameter tuning to include XGBoost’s complete family of column-wise subsampling hyperparameters `colsample_bylevel` and `colsample_bynode` in addition to `colsample_bytree` used in J24. These parameters control the fraction of features randomly selected at different stages of tree construction (per depth level, per node and per tree, respectively). With these subsampling parameters, the XGBoost model tends to spread feature importance more evenly across correlated features. In this way, we could prevent unphysical overfitting to any single predictor and obtain more robust SHAP attributions that reflect the collective influence of intercorrelated variables. The XGBoost models from J24 were re-tuned in this study based on this more comprehensive hyperparameter set. Thus, the MODIS-based results presented here are marginally different from those reported in J24.

4.2.2.2 Explaining the machine-learning models

The SHAP approach is used to explain the regional XGBoost models. SHAP provides additive, local attributions of each predictor to the CF prediction (Lundberg et al., 2018, 2020). SHAP decomposes every individual prediction into a base value and contributions from each feature, allowing the specific influence of $\ln N_d$ on CF to be isolated while accounting for the influences of meteorological covariates. The SHAP base value is what the model would predict with no feature information (Lundberg and Lee, 2017), typically calculated as the average prediction over the entire training set. Positive (negative) SHAP values indicate that a predictor raises (lowers) the prediction relative to this baseline. In our framework, the SHAP base value can be interpreted as the climatological mean CF over the training dataset in a given geographical window, representing the expected XGBoost prediction given the mean state of meteorology and N_d . Each feature’s SHAP value then quantifies the magnitude and direction (positive or negative) that the specific feature value contributes to the CF prediction, shifting the final prediction away from this mean baseline. By construction, the sum of all SHAP values plus the base value equals the final prediction of the model, ensuring additivity and consistency.

Following J24, plotting SHAP values of $\ln N_d$ against its feature values yields a

SHAP dependence plot where the slope is taken as the $\ln N_d$ –CF sensitivity for a given geographical window. For the ICON-HAM analysis, however, a methodological adaptation is required: instead of a single linear regression fit, is applied. This choice is motivated by the stronger nonlinear (saturating) behaviour in the ICON-HAM–based $\ln N_d$ –CF relationships compared with those in J24. In ICON-HAM, the CF response to $\ln N_d$ diminishes at higher $\ln N_d$ even after the logarithmic transformation, reflecting a saturation effect that largely disappeared after log transformation in J24. The PLR approach retains comparability to J24’s method while allowing the nonlinearities to be captured. A complete overview of the XGBoost+SHAP global framework is provided in J24. Further details on the ICON-HAM nonlinearities and rationale for PLR are discussed in Sect. 4.3.2, and the breakpoint selection strategies for data segmentation are detailed in Sect. 4.2.3 and Sect. 4.5.2.

4.2.3 Piecewise linear regression with constraint-aware breakpoints

Here we introduce a constraint-aware breakpoint with fixed interval in logarithmic space, which is the primary strategy used in our main analysis. Specifically, it is called “constraint-aware” because it defines the breakpoint relative to the prescribed minimal constraint on cloud droplet number ($N_{d,\min}$). This strategy sets the fixed interval between $N_{d,\min}$ and the breakpoint directly in the predictor space of $\ln N_d$:

$$\ln(N_{d,\text{bp}}) = \ln(N_{d,\min}) + \delta \quad (4.3)$$

where $N_{d,\text{bp}}$ denotes the breakpoint and δ represents the logarithmic offset from the imposed lower bound. In linear space this gives $N_{d,\text{bp}} = N_{d,\min} \times e^\delta$. In this study, we fix the log-space interval $\delta = 0.1$, so $N_{d,\text{bp}} \approx N_{d,\min} \times 1.105$, yielding breakpoints of approximately 11, 44, and 111 cm^{-3} for the $N_{d,\min} = 10, 40$, and 100 cm^{-3} experiments, respectively. Using a constant increment in logarithmic space enforces the same relative (multiplicative) distance from $N_{d,\min}$ across all experiments and is consistent with logarithmic scaling of the $\ln N_d$ predictor. By placing the breakpoints $\sim 10.5\%$ above the imposed lower bounds, this approach is designed to effectively capture the transition from a regime controlled by the lower bound, and a regime free from the lower bound.

This PLR framework is applied to the $\ln N_d$ SHAP dependence plots for each $5^\circ \times 5^\circ$ geographical window. Data are segmented at the identified breakpoint, and separate linear regressions are fitted for the below- and above-breakpoint regimes to yield slopes β_b and β_a , respectively. For numerical stability, we require at least

1000 data points in the below-breakpoint regime and 500 in the above-breakpoint regime for each geographical window. A detailed visual demonstration of this PLR approach applied to an exemplary geographical window is presented later in Sect. 4.3.3.1. To assess robustness, we test two alternative breakpoint strategies: a constraint-aware approach with fixed intervals in linear space, and a statistically optimized approach that minimizes fitting error within a constraint-aware search range. These two alternative strategies and their results are detailed in Sect. 4.5.2.

4.3 Results and discussion

4.3.1 Mean states of marine low clouds in ICON-HAM

Here we present how imposing lower-bound constraints of 10, 40 and 100 cm^{-3} elicits distinct responses in precipitation, liquid water in clouds, and cloud cover across the two cloud-cover parameterization schemes (CC-RH and CC-RH-LWC). Figure 4.1 presents the global distributions of the mean precipitation rate at the surface (PR), LWP and CF for each $5^\circ \times 5^\circ$ geographical window.

As $N_{d,\min}$ increases, autoconversion is suppressed, leading to a reduction in the large-scale precipitation (Fig. 4.1a). This arises because higher $N_{d,\min}$ yields clouds populated by more numerous and smaller droplets, which is less favourable for drizzle formation. The KK2000 autoconversion scheme captures this effect through its relatively strong inverse dependence on N_d (Khairoutdinov and Kogan, 2000). Even the cleanest clouds within each experiment (with N_d at or slightly above $N_{d,\min}$) become inefficient in producing rain, allowing more water to accumulate in the clouds and thus resulting in higher LWP (Fig. 4.1b). We see that PR decreases from $N_{d,\min} = 10$ to 40 cm^{-3} and then levels off, while LWP continues to increase throughout the entire range. This decoupling occurs because the LWP represents a reservoir of cloud water, whereas PR is a flux at the surface. Weakening the internal cloud process of autoconversion directly helps build up the reservoir, but changes in its level do not necessarily scale with changes in the surface flux. Moreover, PR is balanced by surface evaporation, which is mainly governed by the surface energy budget and hence by surface temperature (Held and Soden, 2006). Since SSTs are prescribed in our AMIP simulations, oceanic evaporation is relatively constrained, limiting and converging PR changes.

By construction, the two cloud-cover schemes produce contrasting CF responses. In CC-RH-LWC, the explicit dependence of CF on cloud water content means the rising LWP at high $N_{d,\min}$ translates directly into a steady increase in CF. Meanwhile, CC-RH, relying solely on grid-mean RH, shows a decline followed

by a plateau that mirrors the PR trend. This behaviour likely results from higher $N_{d,min}$ leading to smaller droplets and suppressed precipitation formation. Smaller droplets reduce sedimentation and also evaporate faster at the cloud top, enhancing cloud-top entrainment. Meanwhile, less drizzle evaporates below the cloud and more condensate is retained in the cloud layer, diminishing sub-cloud evaporative moistening. Both act to lower grid-mean RH. While similar drying can occur in CC-RH-LWC, it is offset by the LWP-driven CF increase, yielding an overall rise in cloud cover.

The stronger increase in mean-state CF with the higher artificial N_d floor seen in CC-RH-LWC (relative to CC-RH) is qualitatively consistent with Lohmann and Feichter (1997). In their study, when aerosol loading increased from PI to PD, the cloud-water-dependent scheme (the same as our CC-RH-LWC) showed a roughly 3 % CF increase under elevated aerosol emission levels, compared with only about 1 % in their RH-only scheme (conceptually similar to our CC-RH). Although their mechanisms differ from ours (emission changes versus prescribed lower bounds), both approaches show that CC-RH-LWC’s explicit dependence on cloud water content amplifies the CF response when autoconversion is suppressed by increased N_d and liquid water accumulates. We therefore interpret the divergence between schemes in Fig. 4.1c as consistent in sign with Lohmann and Feichter (1997), even though $N_{d,min}$ experiments are not perfect PI and PD analogues. This supports the view that the pronounced CF response in CC-RH-LWC stems from its direct coupling between cloud cover and liquid water, which becomes particularly evident when autoconversion is weakened due to autoconversion’s dependence on droplet number, which $N_{d,min}$ directly affects.

4.3.2 Observations vs. default ICON-HAM setup using one linear regression

We first compare the CF sensitivity to $\ln N_d$ globally based on the ICON-HAM default model setup (CC-RH cloud-cover parameterization with $N_{d,min} = 40 \text{ cm}^{-3}$ to that of the observational results of J24. The sensitivities here are quantified following the approach in J24, defined as the slope derived from an ordinary least-squares linear regression of the $\ln N_d$ SHAP values against the $\ln N_d$ feature values for each $5^\circ \times 5^\circ$ geographical window.

MODIS-based results (Fig. 4.2a) show more spatially heterogeneous CF sensitivities, strongest in the stratus-to-cumulus transition regions off western coasts and in the Southern Hemispheric midlatitudes. In contrast, ICON-HAM displays more widespread and contiguous areas of high $\ln N_d$ –CF sensitivity, pronounced

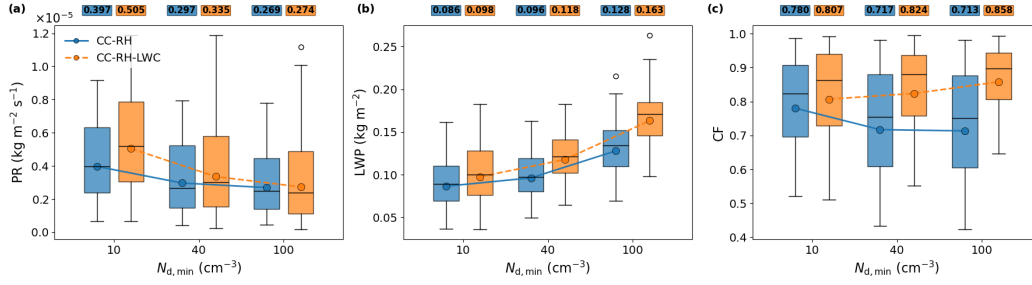


Figure 4.1: Distributions of $5^\circ \times 5^\circ$ geographical-window-mean precipitation rate (PR; panel a), liquid water path (LWP; panel b), and cloud fraction (CF; panel c) over the global oceans for marine liquid clouds during 2011–2015. The simulations compare two cloud-cover parameterization schemes (CC-RH and CC-RH-LWC) and three imposed minima of cloud droplet number concentration ($N_{d,\min} = 10, 40, 100 \text{ cm}^{-3}$). Each window’s value is the spatiotemporal mean of the aggregated $1^\circ \times 1^\circ$ grid cells. The blue boxes and solid lines represent the results from the CC-RH scheme, while the orange boxes and dashed lines represent the results from the CC-RH-LWC scheme. Each box shows the interquartile range. Whiskers extend to 1.5 times the interquartile range, and individual points indicate outliers. Median values are shown by the horizontal lines, while the global area-weighted mean values are represented by circular markers and overlaid lines.

over nearly all subtropical regions, including stratocumulus regions, trade cumulus regions, and tropical ascending regions (Fig. 4.2b). Unlike MODIS results, the regions with the strongest sensitivities in the Pacific and Atlantic Oceans are located more westward, away from the west coasts. Zonal means show that both ICON-HAM and MODIS exhibit weaker sensitivities near the equator compared to subtropical regions, likely reflecting the greater dominance of deep convection and strong meteorological variability in the tropics. These dynamical factors limit the prevalence of stratiform liquid clouds and obscure the signal of aerosol-CF relationships compared to the extensive subtropical stratocumulus decks. Another major difference appears in the Southern Hemispheric midlatitudes, where ICON-HAM shows weak sensitivities. The geographic differences in sensitivity patterns suggest that key controlling processes may differ between observations and ICON-HAM, notably regions where the model frequently computes shallow convection. For instance, differences in droplet size distributions can alter entrainment-evaporation feedbacks and precipitation efficiency, thereby shifting the spatial pattern of the sensitivity.

The distributions of the CF sensitivity values (Fig. 4.2c) show that the global weighted mean sensitivities are of similar magnitude in ICON-HAM (0.137) and MODIS (0.111), with ICON-HAM being somewhat higher and exhibiting greater

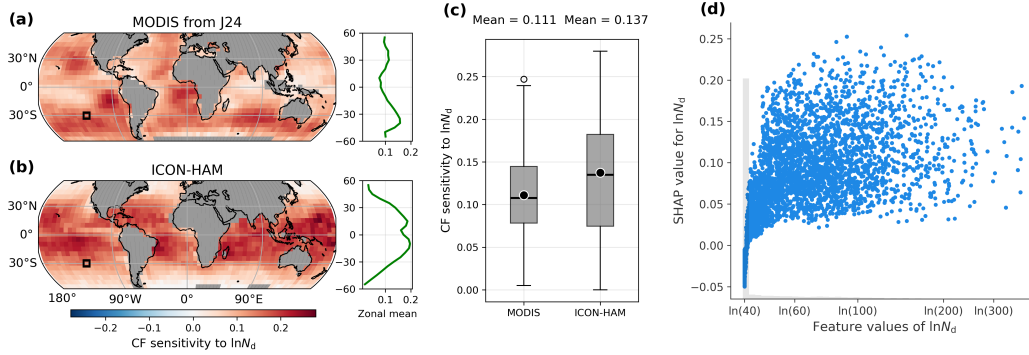


Figure 4.2: Comparison of marine low-cloud fraction (CF) sensitivity to log-transformed cloud droplet number concentration ($\ln N_d$) between MODIS-based results from J24 (recomputed here with retrained XGBoost models using extended column-wise subsampling hyperparameters; see Sect. 4.2.2.1) and ICON-HAM simulations using the default CC-RH scheme with $N_{d,\min} = 40 \text{ cm}^{-3}$. Panels (a) and (b) show spatial patterns and zonal means of the observed and modelled $\ln N_d$ –CF sensitivities, respectively. (c) Distributions of the CF sensitivities; thick black lines denote medians, and black dots show area-weighted global means (values above boxes). (d) ICON-HAM SHAP dependence plot for $\ln N_d$ in the exemplary geographical window (27.5°–32.5° S, 122.5°–127.5° W), whose location is marked by squares in (a) and (b). Grey bars in the background indicate the distribution of $\ln N_d$ feature values.

variability. Note the value of 0.111 differs slightly from that reported in J24 because XGBoost models were re-tuned using the more comprehensive hyperparameters in this study (Table 4.1). The global weighted mean value of ICON-HAM is similar to the MODIS-based estimate but higher than the values reported for other GCMs including ECHAM (Wang et al., 2025). This does not necessarily contradict the broader expectation discussed in the Introduction that models tend to underestimate aerosol–cloud forcing relative to observation-based constraints, as the sensitivity does not translate directly to radiative forcing. However, the larger simulated sensitivities motivate further investigation of their origin (e.g. at individual geographical window) and of potential methodological limitations when applying SHAPs with a single linear regression for ICON-HAMs.

In light of this, we examine the relationship between predicted CF and $\ln N_d$ in the same exemplary geographical window as in J24 (Fig. 4.2d). The ICON-HAM relationship is clearly not as linear as the one based on MODIS. The cloud cover saturation effect, or the dependence of cloud cover sensitivity on background N_d , has already been documented: CF increases quickly with increasing N_d , but the rate of increase approaches zero after reaching an N_d threshold at approximately

60 cm^{-3} (e.g. Hu et al., 2021; Yuan et al., 2023). This effect can be reflected by a sharply decreasing derivative (dCF/dN_d) in Yuan et al. (2023) or ($dSHAP/dN_d$ in J24) before the saturation threshold and a nearly flattened curve after the threshold is exceeded.

As shown in J24, using MODIS data, this effect only manifests evidently as a nonlinear relationship between the N_d feature values and their corresponding SHAP values, which becomes much more linear after applying a logarithmic transformation to N_d . In ICON-HAM data for the same geographical window, the relationship remains strongly nonlinear and the CF saturation effect persists even after the logarithmic transformation (Fig. 4.2d). This more nonlinear CF response in ICON-HAM likely arises because ICON-HAM’s cloud-cover and microphysics schemes are formulated based on process-level understanding of the small-scale nonlinearities observed, for example, in field campaigns (Lohmann and Feichter, 1997) and LES constrained by such field campaigns (Khairoutdinov and Kogan, 2000). At these small scales, cloud systems can shift abruptly once critical thresholds are crossed, resulting in step-like responses. Since these parameterizations include physical constraints such as thresholds and saturation limits, the resulting model output retains its inherent nonlinearity, and this signature of its core physics persists even after aggregation from $1^\circ \times 1^\circ$ grid cells to $5^\circ \times 5^\circ$ geographical windows.

At larger scales, by contrast, the abrupt CF responses that arise from microphysical thresholds are smoothed out. This occurs because higher aerosol concentrations delay the breakup of cloud decks, prolonging their lifetime. As winds advect these systems, the temporal delay in cloud dissipation allows cloud cover to persist over a longer track, which effectively translates into a spatial extension of cloudiness. Consequently, under spatial aggregation across many sampled cloud scenes, a larger fraction of the sampled domain remains cloudy, the spatially aggregated CF response appears gradual and more linear (Goren et al., 2019, 2022; Rosenfeld et al., 2023). This linearity is precisely the characteristic behaviour observed in large-scale satellite analyses, including our previous MODIS-based study (J24).

The simple linear regression between SHAP values and feature values of $\ln N_d$ used in J24 to quantify the $\ln N_d$ –CF sensitivity is not suitable for ICON-HAM output, where the relationships are strongly nonlinear. To account for these while maintaining comparability with J24’s methodology and other linear sensitivity estimates, we apply the PLR approach with a breakpoint strategy defined in Sect. 4.2.3, similar in spirit to the segmented fitting approach applied to the nonlinear N_d –LWP relationships by Gryspeerd et al. (2019). This breakpoint

strategy ensures a consistent proportional offset from the imposed lower bounds across all simulations, which is more appropriate given the logarithmic nature of the $\ln N_d$ predictor. Physically, this approach is intended to effectively capture the transition from a near- $N_{d,\min}$ (lower-bound-constrained) regime with strong CF sensitivity to $\ln N_d$ to a high N_d regime with a weaker CF response. To illustrate how this strategy operates with PLR based on SHAP dependence plots, we zoom in on the SHAP dependence for all six simulations in the same exemplary geographical window as Fig. 4.2d; the detailed visualization and discussions are provided in Sect. 4.3.3.1.

To assess the influence of breakpoint selection on our results, we evaluated two additional strategies beyond the default strategy described above. The first is an alternative constraint-aware approach with a fixed interval on a linear scale of 5 cm^{-3} (Sect. 4.5.2.1). The second is a data-driven approach where the breakpoint is statistically optimized within a constrained search range extending 50 cm^{-3} above each $N_{d,\min}$ value (Sect. 4.5.2.2). This hybrid design combines statistical data-driven flexibility with the $N_{d,\min}$ constraints. The full methodology and global results for these alternative strategies are presented in Sect. 4.5.2.

4.3.3 From ICON-HAM physical states to SHAP-detected sensitivity: the headroom mechanism

4.3.3.1 Piecewise sensitivity and headroom effect at the exemplary geographical window

Figure 4.3 illustrates the application of PLR with constraint-aware breakpoints $\ln(N_{d,bp}) = \ln(N_{d,\min}) + 0.1$ at the exemplary geographical window. The panels confirm the pronounced nonlinearity in the $\ln N_d$ –CF relationship, which justifies our piecewise approach. First, the slopes on either side of the breakpoint (vertical coloured dashed lines) contrast systematically: the below-breakpoint regime has a stronger sensitivity (β_b) than the above-breakpoint regime (β_a) across all experiments. The summary panels (d) and (h) make the two regimes and the associated saturation effect visually explicit. Second, regime occupancy shifts with $N_{d,\min}$ and gets markedly asymmetric at high $N_{d,\min}$: At $N_{d,\min} = 10 \text{ cm}^{-3}$, samples are split roughly evenly between the two regimes. However, at 40 and 100 cm^{-3} , the vast majority fall below the breakpoint, with the below-breakpoint fraction increasing with $N_{d,\min}$, reflecting that more data are truncated by the higher $N_{d,\min}$ constraints and thus concentrated near the imposed bounds.

The below-breakpoint slopes vary systematically with both $N_{d,\min}$ and cloud-cover scheme, whereas the above-breakpoint slopes remain broadly comparable.

At $N_{d,\min} = 10 \text{ cm}^{-3}$, CC-RH-LWC shows a larger β_b than CC-RH. For CC-RH, β_b increases from 10 to 40 cm^{-3} and then decreases from 40 to 100 cm^{-3} with the magnitudes at 40 and 100 cm^{-3} remaining comparable. By contrast, β_b in CC-RH-LWC declines drastically as $N_{d,\min}$ increases, despite its explicit dependence on cloud water content. Consequently, β_b is weaker in CC-RH-LWC than CC-RH at higher $N_{d,\min}$. This apparent paradox reflects a “headroom effect”, more visible when viewing the pattern against the secondary y-axes for predicted CF (Fig. 4.3): At high $N_{d,\min}$, CC-RH-LWC already attains a substantially elevated baseline CF (given by the SHAP base values indicated by the horizontal black solid lines). Therefore, CF predictions approach the physical ceiling of 1.0, and the marginal sensitivity to further $\ln N_d$ increases diminish due to saturation. In contrast, CC-RH maintains lower baseline CF values, preserving a greater dynamic range for $\ln N_d$ to influence CF.

This SHAP-based piecewise approach effectively captures an intrinsic ICON-HAM behaviour in the $\ln N_d$ –CF relationships: a transition from a steeper, $N_{d,\min}$ -constrained regime to a flatter, far-from- $N_{d,\min}$ regime. Although the interval between the breakpoint and $N_{d,\min}$ is fixed in log space across all experiments, the fraction of data points in the first regime increases with $\text{glN}_{d,\min}$, consistent with stronger truncation and tighter constraint near the higher imposed lower bounds. A clear headroom trend emerges: the baseline CF is higher in CC-RH-LWC and increases further with $N_{d,\min}$, leaving less room for additional CF gains and thus dampening the sensitivity.

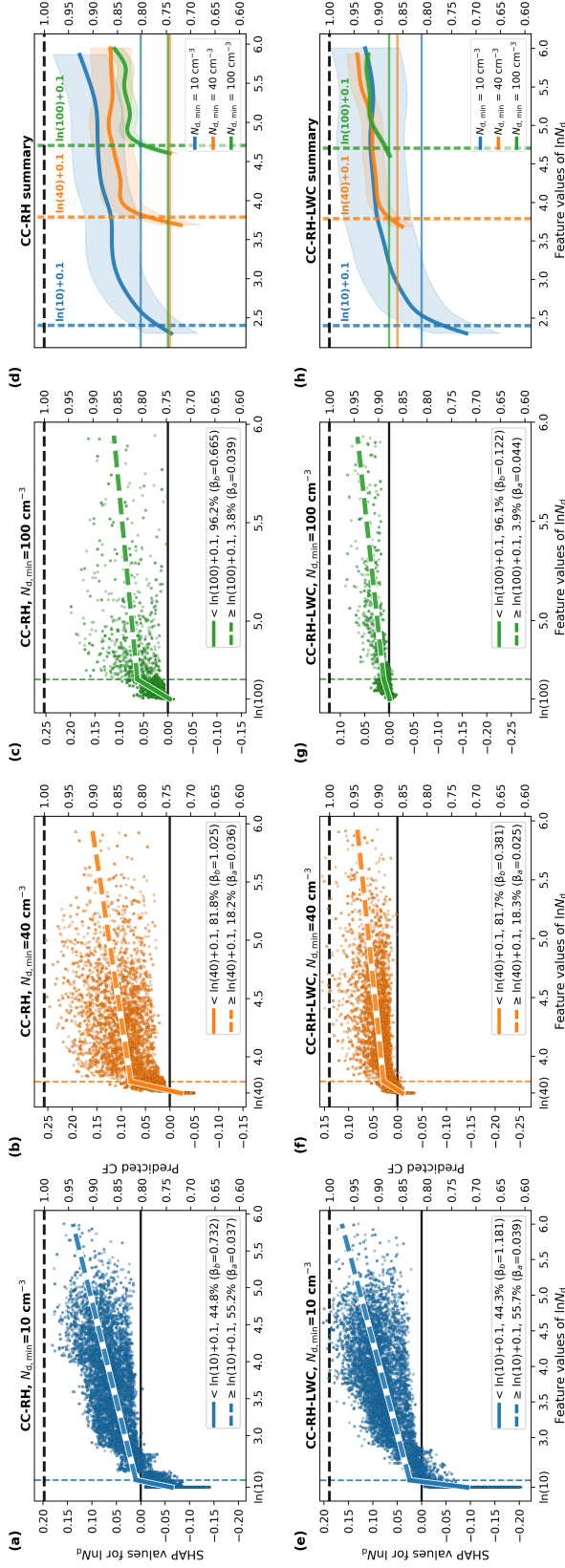


Figure 4.3: SHAP dependence plots for log-transformed cloud droplet number concentration ($\ln N_d$) at the exemplary geographical window (27.5° – 32.5° S, 122.5° – 127.5° W) for six ICON-HAM simulations. Top row (a–d): the CC-RH scheme; bottom row (e–h): the CC-RH-LWC scheme. Columns correspond to $N_{d,\min}$ values of 10, 40 and 100 cm^{-3} (left to right). In panels (a–c) and (e–g), the x-axis shows feature values of $\ln N_d$, and the primary y-axis shows SHAP values for $\ln N_d$. The secondary y-axis (right) shows XGBoost-predicted CF (SHAP base value). Horizontal solid black lines indicate zero SHAP contribution on the primary axis and the baseline CF prediction on the secondary axis; the horizontal dashed line marks $\text{CF} = 1.0$. To quantify $\ln N_d$ –CF sensitivity, piecewise linear regression (PLR) is applied with a single breakpoint at $\ln(N_{d,\text{bp}}) = \ln(N_{d,\min}) + 0.1$. Solid and dashed regression lines show the fitted slopes below (β_a) and above (β_b) the breakpoint, respectively. Vertical dashed lines indicate breakpoint locations. Percentage values in the legend report the fraction of samples in each regime. Panels (d) and (h) summarize the CC-RH and CC-RH-LWC results, respectively, using generalized additive model fits with 95 % prediction intervals. Colours match corresponding scatter plots.

4.3.3.2 Global patterns of SHAP base values and mean absolute SHAP of $\ln N_d$

Section 4.3.1 showed that imposing higher $N_{d,\min}$ values suppresses autoconversion and drizzle, leading to reduced PR and enhanced LWP in both cloud-cover schemes. In CC-RH-LWC, where CF depends explicitly on cloud water, higher LWP directly translates into higher CF, resulting in a mean state characterized by more extensive cloud decks. Conversely, CC-RH, which depends solely on RH, shows little change or even a decline in CF, likely driven by reduced grid-mean RH associated with enhanced cloud-top entrainment and weaker sub-cloud evaporative moistening. As a result, at $N_{d,\min} = 40$ and 100 cm^{-3} , the two schemes produce markedly different baseline CF states, consistent with the behaviour reported by Lohmann and Feichter (1997). In the following, we examine how these mean-state differences are reflected in the SHAP base values and mean absolute SHAP values of $\ln N_d$, which together describe the baseline cloud state and the overall contribution strength of aerosol variability within the SHAP framework.

Figure 4.1 zooms out from the exemplary window to place this behaviour in a global context. Panel (a) presents the global distribution of SHAP base values. Defined as the average of all XGBoost predictions over the training set, the SHAP base value corresponds directly to the local climatological mean-state CF. Indeed, their global distributions resemble the contrasting behaviours of CC-RH and CC-RH-LWC observed in Fig. 4.1c. In other words, variations in SHAP base values across experiments primarily indicate the physical shifts in the simulated CF mean state attributed to the influence of $N_{d,\min}$ on autoconversion and drizzle.

Panel (b) shows the global distribution of the mean absolute SHAP value for $\ln N_d$, which is a measure of its feature importance or average marginal contribution. At 10 cm^{-3} , $\ln N_d$ shows greater feature importance in CC-RH-LWC than in CC-RH, reflecting CC-RH-LWC’s direct CF-LWC coupling, which enables N_d -induced liquid water changes to immediately affect cloud cover. At $N_{d,\min} = 40$ and 100 cm^{-3} , the opposite holds: CC-RH has a larger $\ln N_d$ contribution, since there is limited headroom for $\ln N_d$ variability to drive further CF increases in CC-RH-LWC, constraining the marginal $\ln N_d$ contributions. This global behaviour mirrors the headroom effect identified at the exemplary window in Sect. 4.3.3.1.

In summary, the influence of the differing treatment of liquid-water dependence is twofold both locally and globally: First, it alters the physical mean-state CF (the SHAP base value), and second, this baseline difference makes the system “headroom-mediated” by limiting the room for $\ln N_d$ to contribute to CF increases. This twofold effect is also the key to explaining the resulting sensitivity patterns.

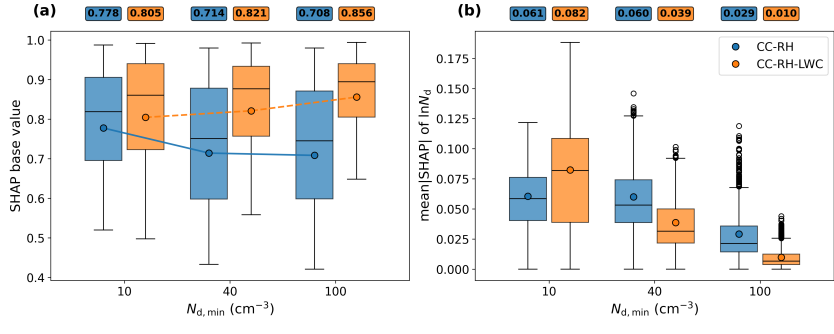


Figure 4.4: Global distributions of per- $5^\circ \times 5^\circ$ -window (a) SHAP base values for the prediction of CF and (b) mean absolute SHAP value of $\ln N_d$ (feature importance) for global marine liquid clouds during 2011–2015, including six experiments (two cloud-cover schemes: CC–RH and CC–RH–LWC; three imposed N_d lower bounds, $N_{d,min} = 10, 40, 100 \text{ cm}^{-3}$). Box plots show the interquartile range (IQR), whiskers extend to $1.5 \times \text{IQR}$, circles denote outliers, and horizontal lines indicate medians. Global area-weighted means are shown as circular markers connected by lines, with the corresponding values annotated above each panel.

4.3.3.3 Global distribution of $\ln N_d$ –CF sensitivity

Having established how elevated SHAP base values (baseline CF) constrain the feature importance of $\ln N_d$ within the SHAP framework, we now examine how this mechanism manifests itself in the global distribution of $\ln N_d$ –CF sensitivities derived from the PLR with the same breakpoint strategy (Sect. 4.2.3). The PLR slopes represent the directional sensitivity of CF to $\ln N_d$. Accordingly, lower mean absolute SHAP values, which indicate weaker marginal contributions, naturally correspond to flatter PLR slopes since the individual SHAP values entering the regression are smaller in magnitude. This leads to flatter SHAP dependence plots when $\ln N_d$ has limited capacity to contribute to CF predictions. Figure 4.5 summarizes the resulting global distributions of PLR-derived $\ln N_d$ –CF sensitivities for the below- and above-breakpoint regimes. Consistently, the global distribution of the mean absolute SHAP values for $\ln N_d$ (Figs. 4.4b) resembles that of the below-breakpoint sensitivities (Fig. 4.5a).

Across all experiments, the below-breakpoint sensitivities are systematically larger than the above-breakpoint ones, typically by about an order of magnitude (note the different y-axis ranges in Fig. 4.5a and b). The below-breakpoint regime reveals the clearest inter-scheme contrasts and directly reflects the headroom mechanism described above. At $N_{d,min} = 10 \text{ cm}^{-3}$, CC–RH–LWC shows stronger sensitivities than CC–RH (global area-weighted means of approximately 1.092 and 0.424, respectively). In line with Fig. 4.4, at this low $N_{d,min}$, both schemes have

similar baseline CF (and hence similar headroom), allowing CC-RH-LWC’s explicit dependence on cloud liquid water to be fully expressed as both stronger feature importance and stronger derived CF sensitivity.

However, as $N_{d,min}$ increases to 40 and 100 cm^{-3} , the pattern reverses. The global weighted means for CC-RH (approximately 1.135 and 1.011, respectively) exceed those of CC-RH-LWC (0.633 and 0.209, respectively). This systematic weakening in CC-RH-LWC aligns precisely with the SHAP diagnostics in Fig. 4.4: the liquid-water-dependent scheme’s high baseline CF (SHAP base values exceeding 0.85 at $N_{d,min} = 100 \text{ cm}^{-3}$) leaves minimal dynamic range for $\ln N_d$ to act, thereby muting the marginal contribution captured by SHAP and leading to weaker PLR-derived sensitivities. Conversely, CC-RH maintains lower baseline CFs, preserving a larger dynamic range for $\ln N_d$ to act, enabling stronger detected sensitivities.

Above the breakpoint, sensitivities are roughly an order of magnitude weaker than below it, and are centred near zero for both schemes (Fig. 4.5b). This attenuation mirrors the flatter upper segments of the SHAP dependence plots after saturation. In this regime, sensitivities remain relatively stable across $N_{d,min}$ values (0.036–0.054 per unit $\ln N_d$), and inter-scheme differences are minimal. This suggests that once $\ln N_d$ values are high and less influenced by the imposed lower bounds, both parameterizations respond similarly weakly to aerosol variability.

Taken together with Sect. 4.3.3.2, the counterintuitive finding of weaker $\ln N_d$ –CF sensitivities in CC-RH-LWC at high $N_{d,min}$ does not indicate that its explicit CF–LWC coupling is ineffective. Instead, it demonstrates that the liquid-water dependence raises baseline CF and accelerates saturation, thereby diminishing the marginal contribution of $\ln N_d$ and the sensitivity detected by SHAP. This reflects a fundamental distinction between physical responsiveness and methodological detectability. In this sense, the global sensitivity patterns are “headroom-mediated”, providing a coherent bridge from ICON-HAM’s physical representation of cloud cover to the explainable-ML diagnostics.

A direct comparison with the MODIS-based results from J24 highlights the structural differences between observation- and model-based sensitivities. The global area-weighted mean sensitivity from J24, derived from a single linear regression, is approximately 0.111 (Fig. 4.2c). This value lies between our two PLR regimes: substantially smaller than the below-breakpoint means (0.209 to 1.135) but larger than the above-breakpoint means (0.036 to 0.054). This intermediate value falls between the two regimes and can be effectively interpreted as an aggregated response. However, the distinct segmentation and spread in ICON-HAM sensitivities indicate that the simulated sensitivity estimates are tricky and also

dictated by model configuration and methodological choices, complicating direct model-observation comparisons and aerosol-cloud forcing estimates.

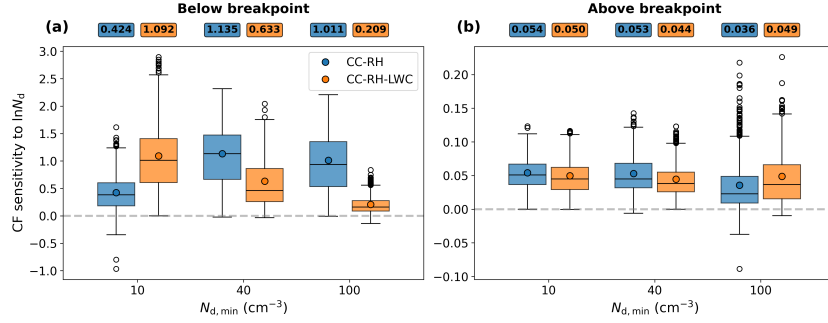


Figure 4.5: Global distributions of the cloud fraction sensitivity to log-transformed cloud droplet number concentration ($\ln N_d$) for global marine liquid clouds during 2011–2015, for six experiments (two cloud-cover parameterization schemes: CC-RH and CC-RH-LWC; three imposed N_d lower bounds $N_{d,min} = 10, 40$ and 100 cm^{-3}). Sensitivities are computed within each $5^\circ \times 5^\circ$ oceanic window using a piecewise linear regression between $\ln N_d$ feature values and $\ln N_d$ SHAP values, with a single breakpoint $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$. (a) below-breakpoint sensitivities; (b) above-breakpoint sensitivities; Box plots display the interquartile range (IQR), whiskers extend to $1.5 \times \text{IQR}$, and circles denote outliers; medians are horizontal lines. Area-weighted means are shown as circular markers with connecting lines, and the floating values above each panel denote the corresponding area-weighted mean sensitivities. Note that the y-axis ranges differ between panels.

4.3.3.4 Robustness of findings and physical plausibility of breakpoints

To ensure our main findings are not an artefact of our primary breakpoint definition $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$, we tested additional strategies: an alternative constraint-aware strategy with a fixed interval in linear space ($N_{d,min} + 5 \text{ cm}^{-3}$, Sect.4.5.2.1), and a statistically optimized constraint-aware approach that determines breakpoints by minimizing the overall fitting error within a search range constrained by $N_{d,min}$ (Sect.4.5.2.2). Despite differences in their underlying rationale and implementation, all three strategies yield qualitatively consistent results. First, the $\ln N_d$ –CF sensitivity in CC-RH-LWC is stronger at $N_{d,min} = 10 \text{ cm}^{-3}$ but becomes weaker than in CC-RH at 40 and 100 cm^{-3} . Second, inter-scheme differences are dominated by the below-breakpoint regime. This confirms the headroom mechanism independently of the specific breakpoint definition.

Beyond methodological consistency, the robustness of our findings is further supported by the physical plausibility of the breakpoint locations themselves. Independent observational studies of MLCs, using opportunistic experiments

(ship tracks and volcanic eruptions) and large-scale satellite data (Hu et al., 2021; Yuan et al., 2023; Wang et al., 2024; Jia et al., 2024), have repeatedly identified a saturation threshold for CF responses to N_d at roughly $50\text{--}60\text{ cm}^{-3}$. In addition, stratocumulus clouds are also known to transition from closed to open cells at a similar N_d threshold (Rosenfeld et al., 2023). In ICON-HAM, the breakpoint locations depend on the imposed $N_{d,\min}$ by design. For the default $N_{d,\min}$ of 40 cm^{-3} , however, they consistently fall close to the observational threshold range across three independently derived breakpoint strategies. The two constraint-aware strategies (Sect. 4.2.3 and 4.5.2.1) yield breakpoints of approximately 44 cm^{-3} and 45 cm^{-3} , respectively. Notably, the statistically optimized strategy (Sect. 4.5.2.2) independently finds a global area-weighted mean breakpoint value of about 48 cm^{-3} for CC-RH and 56 cm^{-3} for CC-RH-LWC (Fig. 4.9), closely matching the observed saturation threshold of $\sim 50\text{--}60\text{ cm}^{-3}$. By contrast, the $N_{d,\min} = 10\text{ cm}^{-3}$ and 100 cm^{-3} experiments produce different breakpoint locations. The agreement for $N_{d,\min} = 40\text{ cm}^{-3}$ therefore suggests that this lower bound choice places the model’s emergent onset of the CF saturation at a more physically plausible N_d level than the more extreme $N_{d,\min}$ settings.

Overall, the consistency of the breakpoint values among the three breakpoint-selection strategies, combined with agreement between the resulting breakpoints and the known range of the physical saturation thresholds in the $N_{d,\min} = 40\text{ cm}^{-3}$ simulations, confirms the robustness of our SHAP-based PLR approach and validates the underlying nonlinearity. This shows that the PLR approach is not merely an ad hoc curve-fitting choice but effectively captures the model’s physical transition from a highly sensitive, aerosol-limited regime to a saturated regime. Moreover, the consistent breakpoint range reinforces that our central findings—the headroom-mediated sensitivity patterns and the counterintuitive $\text{CC-RH} > \text{CC-RH-LWC}$ sensitivity contrast at high $N_{d,\min}$ —are robust and reflect genuine ICON-HAM’s simulated physics.

4.3.4 Implications for the forcing estimate

4.3.4.1 For the bottom-up approach (GCM PI–PD paired simulations)

Our findings reveal a potential challenge in quantifying the radiative forcing from the CF adjustment, as the detected sensitivity is highly state-dependent. Our findings have different implications for different methods to estimate forcing: the bottom-up approach (GCM PI–PD paired simulations) and the top-down approach (observational sensitivity-based).

GCMs typically estimate effective radiative forcing due to aerosol-cloud interac-

tions (ERFaci) using a bottom-up method by running paired PI and PD simulations and taking their difference in the net radiation at the top of atmosphere (TOA) (Ghan, 2013; Boucher et al., 2013; Smith et al., 2020), without relying on an explicit sensitivity value. Instead, the aerosol-cloud forcing emerges from the full nonlinear response across the PI–PD aerosol contrast. Our analysis, in contrast, focuses on the local PD-state $\ln N_d$ –CF sensitivity across six ICON-HAM experiments. These PD-state slopes are therefore not a direct measure of CF-adjustment forcing, but they describe how efficiently additional N_d changes can still modify CF once the system has already reached its PD baseline. Here, we discuss how our findings from these different ICON-HAM configurations potentially affect aerosol forcing estimates.

As systematically investigated by Hoose et al. (2009), imposing $N_{d,\min}$ unphysically alters both the PI and PD states, creating artificially polluted PI conditions over both land and ocean and artificially polluted PD conditions over oceans. This makes the contrast between PI and PD collapse and strongly damps the total aerosol indirect effects. Our experiments add a previously undocumented, complementary dimension to this result by introducing the CC-RH-LWC scheme. When this scheme is combined with high $N_{d,\min}$, suppressed autoconversion raises LWP and thereby elevates baseline CF in both PI and PD equilibrium states. Under naturally pristine PI conditions, this combination can already pre-saturate clouds. In the PD state, increases in anthropogenic aerosols cannot further increase CF because clouds already operate near the $CF \approx 1$ ceiling. In addition to the $N_{d,\min}$ -alone effect (reduced PI–PD N_d contrast) on the total forcing owing to ACI identified by Hoose et al. (2009), this bilateral saturation through the headroom effect compresses the PI–PD ΔCF and physically suppresses the CF-adjustment component of the forcing. For the CC-RH scheme, which lacks the direct LWC–CF link, baseline CF does not saturate to the same extent (Fig. 4.1c), so the forcing is mainly altered by $N_{d,\min}$ alone as in Hoose et al. (2009).

On the other hand, our study also adds a new angle to Lohmann and Feichter (1997). Their work compared an LWC-dependent and an RH-only cloud-cover scheme (referred to as COUPL-CC and COUPL, respectively) and their resulting PI–PD changes in CF and the total aerosol indirect effect, but without imposing $N_{d,\min}$. $N_{d,\min}$ was only introduced in ECHAM4 later by Lohmann et al. (1999). Without $N_{d,\min}$, there is no associated increase in baseline CF and thus PI clouds are not pre-saturated. Consequently, there is sufficient headroom and anthropogenic aerosols can increase CF without being immediately constrained by the CF ceiling in either COUPL or COUPL-CC. Therefore, the stronger dependence on cloud water

in COUPL-CC produces larger CF increases and a more negative total indirect effect than the RH-only COUPL scheme. This behaviour is qualitatively consistent with our ICON-HAM experiments at a low $N_{d,\min}$ of 10 cm^{-3} , where CC-RH-LWC shows a larger marginal contribution of $\ln N_d$ (Fig. 4.4b) and a stronger $\ln N_d$ -CF PD-state sensitivity (Fig. 4.5a) than CC-RH. In both studies, a cloud-water-dependent scheme responds more strongly than an RH-only scheme when $N_{d,\min}$ is effectively low. However, at higher $N_{d,\min}$, the ordering reverses (i.e. CC-RH becomes stronger) due to the headroom effect. Extrapolating this behaviour to the Lohmann and Feichter (1997) framework, one would expect that imposing a high $N_{d,\min}$ in their COUPL versus COUPL-CC comparison would reduce the PI-PD CF increase in both schemes and thereby shrink the difference in total aerosol indirect effect between them. A complete reversal in the forcing order, analogous to the reversal seen in our $\ln N_d$ -CF sensitivities at high $N_{d,\min}$, seems less likely. Lohmann and Feichter (1997) quantified the ACI forcing from PI to PD including non-CF components, whereas the headroom effect identified here acts specifically on the CF-adjustment component and is inferred from PD-state sensitivity variability rather than from an explicit PI-PD forcing calculation.

Overall, if CC-RH-LWC is used, this strong reduction in the CF-adjustment forcing is already operative at $N_{d,\min} = 40\text{ cm}^{-3}$, which is the default value in the ECHAM and ICON model families. Although we do not perform explicit PI-PD runs or compute the radiative forcing from CF adjustments, it is reasonable to expect that the combination of a CC-RH-LWC-type parameterization with high $N_{d,\min}$ would weaken the CF-adjustment contribution to ERF_{aci} in a bottom-up setup, especially over clean oceanic regions. This tendency would further widen the gap between model-based estimates that attribute only a minor role to CF adjustment (e.g. Smith et al., 2020; Zelinka et al., 2023) and observation-based studies that identify CF adjustment as a primary component (e.g. Chen et al., 2022b; Wall et al., 2023; Yuan et al., 2023). These implications remain qualitative, as they are inferred from PD-state sensitivities rather than aerosol forcing diagnostics; a natural next step is to repeat our ICON-HAM experiments for paired PI-PD simulations and explicitly diagnose the CF-adjustment contribution to the aerosol-cloud forcing to test this hypothesis.

4.3.4.2 For the top-down approach (observational sensitivity-based)

In contrast to the bottom-up PI-PD approach, observational studies often follow a top-down strategy, in which CF-adjustment forcing is inferred from an explicit sensitivity value. These strategies vary: some exploit localized perturbations to

isolate aerosol effects by deriving N_d –CF sensitivities from contrasts between relatively clean background conditions and nearby polluted perturbations (e.g. ship tracks or volcanic events) and directly calculate the radiative effect of the associated CF adjustments (e.g. Chen et al., 2022b; Yuan et al., 2023), while others diagnose aerosol–CF sensitivities from statistical relationships across PD variability and apply cloud radiative kernels to the derived cloud-property changes (e.g. Wall et al., 2023). In all of these frameworks, the inferred sensitivity in the sampled cloud regimes is central, as it links observed variability to radiative forcing.

Our results highlight that such sensitivities are strongly state-dependent. In ICON-HAM, the headroom effect appears most clearly when high $N_{d,min}$ is combined with CC-RH-LWC, which pushes baseline CF close to its saturation limit and suppresses the PD-state $\ln N_d$ –CF sensitivity. Part of this pre-saturation is clearly structural to climate models, arising from the imposed $N_{d,min}$ and the chosen cloud-cover formulation. In the real atmosphere, there is neither $N_{d,min}$ nor a prescribed scheme, but there is still physical saturation of CF in regimes with high LWP or RH. Any observationally derived N_d –CF slope will therefore reflect the headroom available in the sampled clouds: sensitivities diagnosed in already cloudy, high-LWP regimes will tend to be weaker than those in thinner, less saturated regimes. This aligns with previous findings that N_d –CF sensitivities are strongest near the edges of the stratocumulus field and in stratocumulus-to-cumulus transition regions, rather than within the overcast decks (Gryspeerdt et al., 2016; Jia et al., 2024).

This state-dependence has implications for interpreting observational constraints on CF-adjustment forcing. Most recent observation-based estimates (e.g. Chen et al., 2022b; Wall et al., 2023; Yuan et al., 2023; Chen et al., 2024) use natural experiments or from PD aerosol–cloud relationships, which do not require assuming a single fixed N_d –CF sensitivity that can be applied to infer changes from PI to PD. However, the underlying N_d –CF relationships in these studies are still diagnosed from specific cloud regimes in the current climate, where baseline CF varies and thus the remaining potential for CF to increase differs. Consequently, the inferred sensitivities are conditional on the mean state and may not be directly transferable to a PI–PD forcing context, since sensitivities in already-cloudy, low-headroom regimes could underestimate the potential response in cleaner, less saturated conditions (representative of PI). A more complete understanding of the CF adjustment to the aerosol forcing would therefore benefit from explicitly conditioning sensitivities on baseline cloud state (e.g. CF, LWP, and precipitation regime) as a complement to regime-aggregated estimates. This is particularly

relevant given the likely reduction in MLC cover in a warming world (Myers et al., 2021; Forster et al., 2021; Bock and Lauer, 2024), which would increase the available headroom and thus potentially enhance the CF sensitivity. This would facilitate more consistent comparisons between model-based and observational estimates by accounting for how the available headroom for CF increases varies systematically across different cloud states.

4.4 Specific conclusions

This study extends the explainable ML framework from our previous observational study (J24) to the ICON-HAM atmosphere-aerosol model. Our primary goals were to compare MODIS- and ICON-HAM-based $\ln N_d$ -CF sensitivities in MLCs, and to understand how the cloud-cover schemes and $N_{d,\min}$ jointly shape the simulated CF response. To achieve this, we conducted six experiments combining two cloud-cover schemes, one dependent solely on RH (CC-RH) and one additionally dependent on cloud water content (CC-RH-LWC), and three different $N_{d,\min} = 10, 40$, and 100 cm^{-3} . The findings of our study are summarized as follows:

1. Mean-state response to $N_{d,\min}$ and cloud-cover schemes: Higher $N_{d,\min}$ suppresses autoconversion and drizzle, and builds up liquid water in both schemes. The CC-RH-LWC scheme translates this LWP increase directly into elevated CF due to its explicit cloud-water dependence, whereas CC-RH, lacking this direct link, shows a weaker or even declining CF response. At $N_{d,\min} = 40$ and 100 cm^{-3} , this creates markedly different mean background cloud conditions between the schemes.
2. ICON-HAM simulates a non-linearity in the $\ln N_d$ -CF relationship: The ICON-HAM $\ln N_d$ -CF relationship is strongly non-linear and shows a clear saturation effect that persists even after log-transformation. This contrasts with the observation-based relationship, which has been shown to be linear in logarithmic space. The difference likely reflects ICON-HAM's process-level parameterizations that retain small-scale saturation effects from field campaigns or LES studies, whereas satellite observations at larger scales show smoother responses due to spatial averaging.
3. Headroom-mediated $\ln N_d$ -CF sensitivity: The mean-state physics in point 1 sets the baseline CF in ICON-HAM. The SHAP approach reveals that high SHAP base values (baseline CF) limit the marginal contribution of $\ln N_d$ (Fig. 4.4), leading to weaker detected sensitivity (Fig. 4.5), despite the direct

physical coupling between CF and cloud water, creating a headroom effect. At $N_{d,\min} = 10 \text{ cm}^{-3}$, CC-RH-LWC is more sensitive to $\ln N_d$ due to its direct dependence on LWC. This pattern reverses at $N_{d,\min} = 40$ and 100 cm^{-3} , as high $N_{d,\min}$ together with CC-RH-LWC leaves little headroom, while CC-RH preserves a lower baseline CF and thus more room for $\ln N_d$ -driven changes.

4. Methodological extension and breakpoint robustness: Three breakpoint strategies consistently separate a steep low- N_d regime and a flatter high- N_d regime, and the contrasting patterns are qualitatively consistent across all experiments. For ICON-HAM's default $N_{d,\min} = 40 \text{ cm}^{-3}$, the statistically optimized breakpoints converge at approximately $50\text{--}60 \text{ cm}^{-3}$, consistent with observed N_d –CF saturation thresholds, supporting the robustness of our framework. More importantly, this suggests that the default $N_{d,\min}$ setup provides a physically consistent representation of the transition into the saturation regime.
5. Implications for CF adjustment forcing and model–observation comparisons: For GCMs using bottom-up PI–PD methods, high $N_{d,\min}$ values combined with LWC-dependent cloud-cover schemes can pre-saturate both PI and PD cloud fields, damping CF adjustment forcing even when the underlying model physics is strong. In top-down, observation-based approaches, any diagnosed N_d –CF sensitivity is inherently state-dependent and reflects the available headroom in the sampled cloud regimes. This implies that robust CF adjustment constraints should explore the option to condition sensitivities on baseline cloud state (e.g. CF, LWP, and precipitation regime), rather than treating the N_d –CF sensitivity as a single, regime-independent quantity.

4.5 Additional methodological details

4.5.1 Predictor variables

Table 4.1: Overview of the predictors from ICON-HAM. Numeric subscripts denote pressure levels or altitudes.

Predictor variable	Short name	Unit
Relative humidity	RH ₇₀₀ , RH ₈₅₀ , RH ₉₀₀ , RH ₉₅₀	%
Specific humidity	SH ₇₀₀ , SH ₈₅₀	kg kg ⁻¹
Air temperature	t_{700} , t_{850}	K
Vertical velocity	ω_{700} , ω_{850}	Pa s ⁻¹
Zonal (eastward) wind	u_{700} , u_{850}	m s ⁻¹
Meridional (northward) wind	v_{700} , v_{850}	m s ⁻¹
Eastward and northward wind component at 10 m	u_{10} , v_{10}	m s ⁻¹
Boundary-layer height (proxy)	BLH	m
Sea surface temperature	SST	K
Vertically integrated water vapour	VIWV	kg m ⁻²
Large-scale precipitation flux (water)	PR	kg m ⁻² s ⁻¹
Sensible heat flux	SHF	W m ⁻²
Latent heat flux	LHF	W m ⁻²
Estimated inversion strength (calculated)	EIS	K
Natural logarithm of cloud droplet number concentration	$\ln N_d$	dimensionless (N_d in cm ⁻³)

4.5.2 Piecewise SHAP sensitivity with two alternative breakpoint strategies

4.5.2.1 Constraint-aware breakpoint with fixed interval in linear space

To assess how the choice of breakpoints influences our results, we introduce two additional breakpoint strategies. The first one is likewise constraint-aware, as the breakpoint is defined relative to $N_{d,\min}$. However, the interval is specified in linear rather than logarithmic space:

$$\ln(N_{d,bp}) = \ln(N_{d,\min} + \Delta N_d) \quad (4.4)$$

where $N_{d,bp}$ denotes the breakpoint and ΔN_d represents the offset in linear space (in cm⁻³) from the imposed lower bound. For this test, we set $\Delta N_d = 5$ cm⁻³, yielding breakpoints of 15, 45 and 105 cm⁻³ for $N_{d,\min} = 10, 40$, and 100 cm⁻³, respectively.

On the logarithmic scale, the interval between $N_{d,\min}$ and the breakpoint equals $\ln(1 + \frac{\Delta N_d}{N_{d,\min}})$, which shrinks as $N_{d,\min}$ increases. The log-space interval width is $\ln(1.5)$, $\ln(1.125)$, $\ln(1.05)$ for $N_{d,\min} = 10, 40$ and 100 cm⁻³, respectively. Thus, as $N_{d,\min}$ increases, the prescribed truncation becomes tighter and causes more data points to pile up near $N_{d,\min}$. This already increases the y-axis (SHAP value)

spread within a narrow x-axis range. The shrinking intervals further compress the near- $N_{d,\min}$ regime on the x-axis while the associated SHAP response variability on the y-axis remains comparatively strong, leading to an inflated β_b at higher $N_{d,\min}$ values.

This inflated β_b is visible both in Figs. 4.6 and 4.7 for the CC-RH parameterization as $N_{d,\min}$ increases. It does not occur for CC-RH-LWC because the compression effect is outweighed by the headroom limitation. Despite these quantitative differences in the slopes, the main qualitative conclusions remain robust: First, below-breakpoint $\ln N_d$ -CF sensitivities are larger than those above the breakpoint regime, and inter-experiment contrasts are only evident in the below-breakpoint regime. Second, at $N_{d,\min} = 10 \text{ cm}^{-3}$, CC-RH-LWC has stronger sensitivities owing to its direct dependence on cloud water content; at $N_{d,\min} = 40$ and 100 cm^{-3} , CC-RH shows stronger sensitivities because of the headroom effect arising from CC-RH-LWC's elevated baseline CF and a CF = 1.0 cap. Choosing this breakpoint strategy primarily affects the trend of sensitivity with increasing $N_{d,\min}$ within each cloud-cover regime. The extent to which this x-range compression influences the results depends on its relative strength versus the headroom effect.

4.5.2.2 Statistically optimized constraint-aware breakpoint

To further evaluate the robustness of our results, we apply a statistically optimized strategy. This data-driven approach determines breakpoints by optimizing their locations that minimize the overall sum of squared errors of the two-segment PLR fit, but constrains the search range to $[\ln(N_{d,\min}), \ln(N_{d,\min} + 50)]$. This hybrid design is therefore constraint-aware while allowing the data to determine the exact split location.

To ensure statistical robustness, this hybrid strategy enforces per-regime sample minima of 1000 data points below the breakpoint and 500 above. These thresholds serve two purposes: first, a pre-fit feasibility check verifies whether any breakpoint within the bounded range can satisfy both minima at the same time, triggering the fallback if no feasible split exists; second, a post-fit validation checks the actual final regime sizes, setting slopes to NaN if either regime is insufficient. Additionally, we set a fit quality screen accepting the piecewise fit only if $R^2 \geq 0.70$ and root mean square error (RMSE) improves by at least 5 % compared to a single linear regression. If either the quality criteria fail, or no feasible split exists within the bounded range, we fall back to $\ln(N_{d,bp}) = \ln(N_{d,\min}) + 0.1$. Per-regime sample minima are enforced at the slope-estimation stage; if only one regime lacks sufficient samples, that slope is set to NaN (no fallback), whereas a complete failure (both slopes invalid) triggers

the fallback. Finally, we apply an interquartile-range-based outlier filter (with a multiplier of 2.0) and a slope-consistency check to remove unphysical or outlying slope estimates.

This hybrid strategy yields Fig. 4.8 and Fig. 4.9. Again, both from the exemplary geographical window perspective and the global perspective, the key pattern persists. Particularly in the below-breakpoint regime, CC-RH-LWC shows stronger sensitivity at $N_{d,\min} = 10 \text{ cm}^{-3}$ through its cloud water dependence, while CC-RH is stronger at 40 and 100 cm^{-3} due to headroom constraints in CC-RH-LWC. Overall, the inter- $N_{d,\min}$ and inter-scheme contrasts obtained with this data-driven strategy (Fig. 4.9) are consistent with those from the constraint-aware approach with a fixed interval in log space (Fig. 4.5), confirming that our key scientific conclusions are robust to the choice of breakpoint strategy.

We additionally show the global distribution of the optimized breakpoints using this strategy in Fig. 4.9c. For the default ICON-HAM configuration ($N_{d,\min} = 40 \text{ cm}^{-3}$), the area-weighted global mean optimized breakpoints are approximately 48 and 56 cm^{-3} on a linear scale for CC-RH and CC-RH-LWC, respectively. These values align closely with the observational saturation thresholds discussed in the main text (Sect. 4.3.3.4), providing physical support for the statistically optimized breakpoint locations (see main text for detailed discussion).

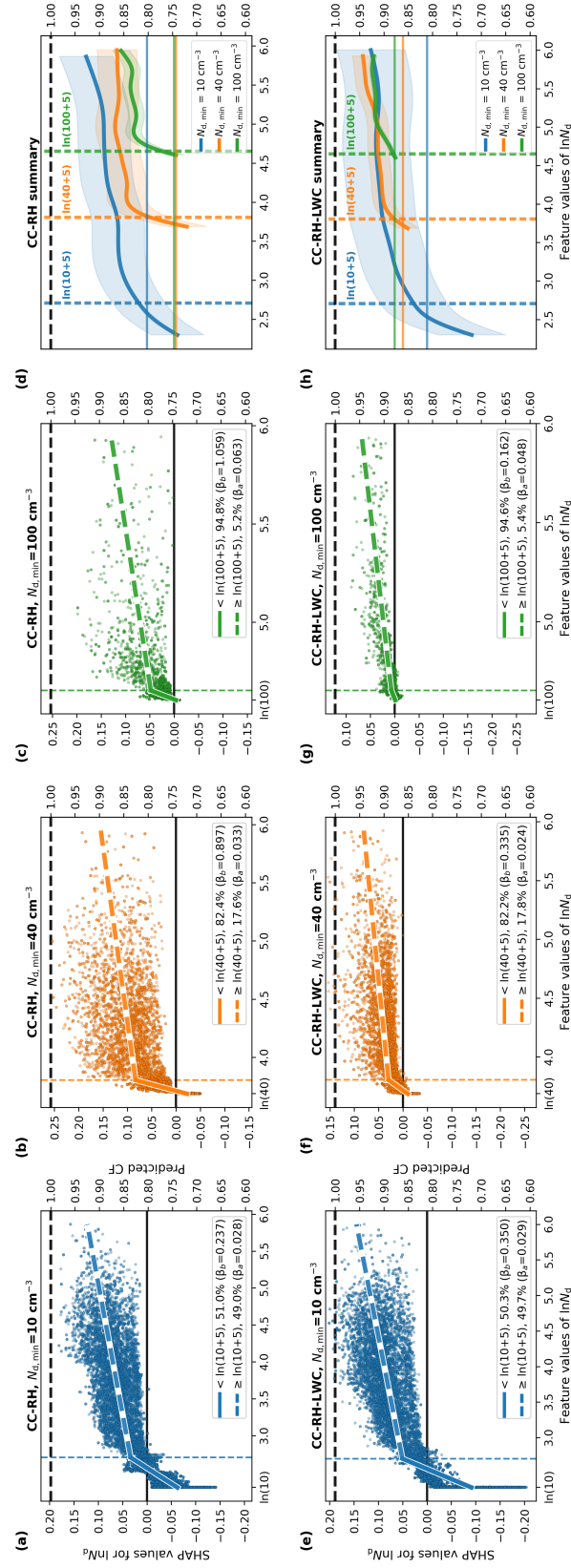


Figure 4.6: As in Fig. 4.3, but the breakpoint for the piecewise linear regression is defined as $\ln(N_{d, \min} + 5)$.

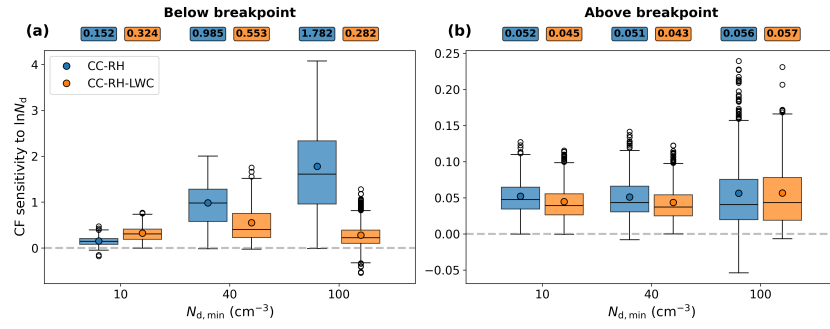


Figure 4.7: Similar to Fig. 4.3, except the breakpoint for the piecewise linear regression is defined as $\ln(N_{d,bp}) = \ln(N_{d,min} + 5)$.

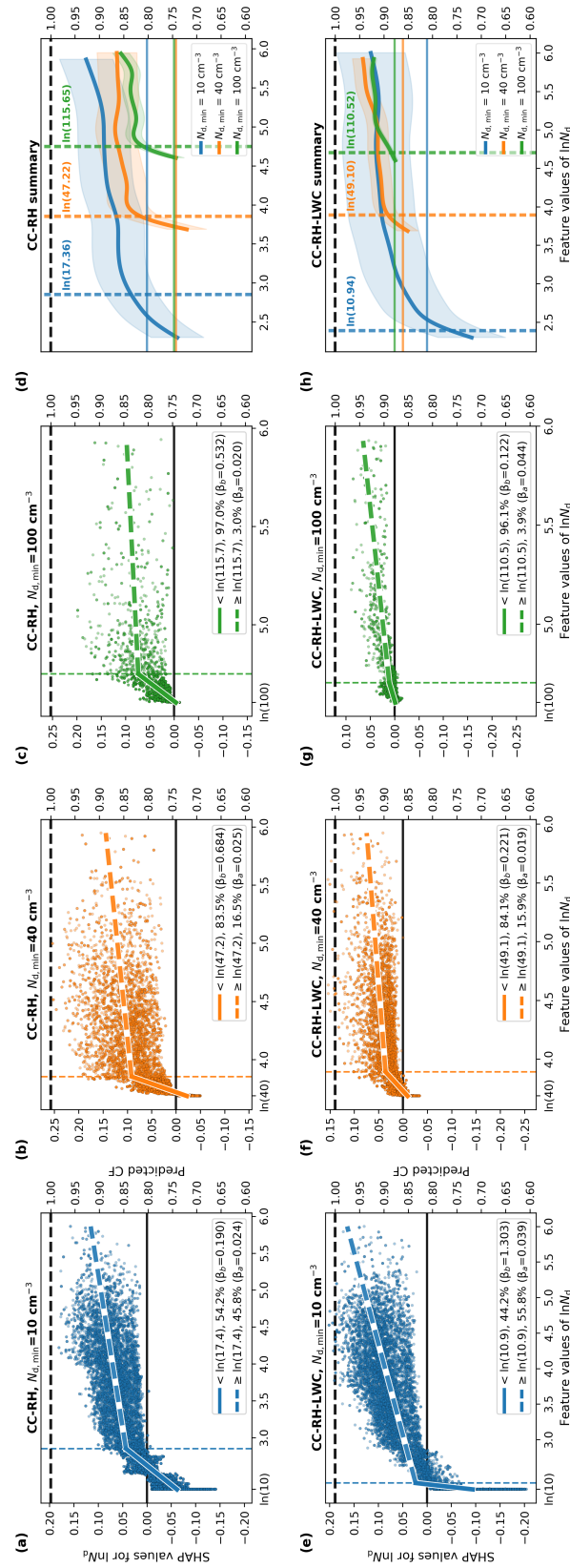


Figure 4.8: Similar to Fig. 4.3, except the breakpoints for the piecewise linear regression are statistically optimized.

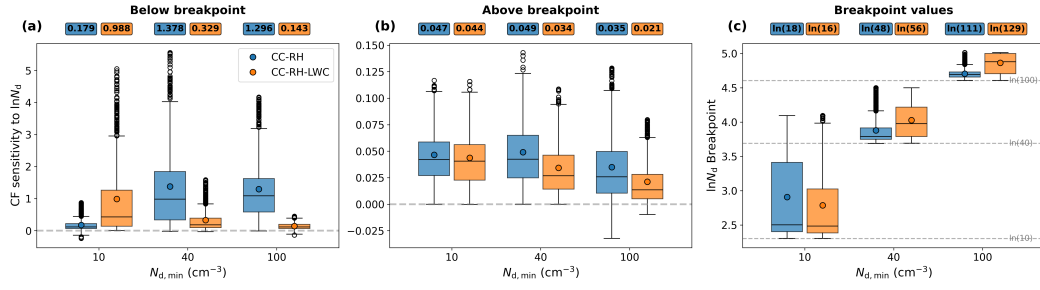


Figure 4.9: As in Fig. 4.5, but the breakpoints for the piecewise linear regression are statistically optimized (their distributions are shown in panel (c)).

Chapter 5

Concluding discussion and outlook

5.1 Concluding Discussion

In Chap. 3 and Chap. 4 of this thesis, two studies were presented that investigated the response of MLC CF to aerosol perturbations using a consistent explainable ML framework. Together, they aim to improve the quantification and physical understanding of this response from an observational and a modelling perspective simultaneously. As outlined in Sect. 1.5, this thesis was guided by two overarching research questions (RQs) which correspond to Chap. 3 and Chap. 4, respectively:

RQ 1: How does MLC CF respond to aerosol perturbations globally, and how is this response influenced by meteorological conditions?

RQ 2: How does the CF response to aerosols emerge in a GCM, and how do model configuration choices affect this response?

These overarching questions were further addressed through more specific questions for the observation-based (**RQ 1.1**, **RQ 1.2**) and model-based (**RQ 2.1**, **RQ 2.2**) parts of the thesis, which guided the design of the individual studies. In the following, the main findings and contributions are discussed and then synthesized at the thesis level.

5.1.1 Observation-based perspective

The observational study presented in Chap. 3 addressed **RQ 1** by establishing a region-specific XGBoost–SHAP framework trained on nine years (2011–2019) of daily MODIS satellite retrievals and ERA5 reanalysis data across $5^\circ \times 5^\circ$ geographical windows. XGBoost models predict CF based on $\ln N_d$ (aerosol proxy) together with a set of thermodynamic and dynamical meteorological predictors at the same

time, and SHAP explain the predictions by providing the feature contributions and feature interactions.

With respect to **RQ 1.1**, this framework provides a useful way to quantify the direction and magnitude of the $\ln N_d$ –CF sensitivity while accounting for meteorological predictors. This observational analysis shows that MLC CF is generally positively associated with N_d , consistent with previous satellite-based evidence for positive aerosol-related CF responses (Gryspeerd et al., 2016; Andersen et al., 2017; Fuchs et al., 2018; Christensen et al., 2020). This indicates that the precipitation suppression pathway (cloud lifetime effect) (Albrecht, 1989; Bellouin et al., 2020) may be the dominant physical response captured by this method in the PI satellite records with $5^\circ \times 5^\circ$ aggregated geographical windows and daily resolution. However, the magnitude of the positive $\ln N_d$ –CF sensitivity varies substantially across different regions. It is strongest in the stratocumulus-to-cumulus transition regions off the western continental coasts and in the Southern Hemispheric midlatitudes, while weaker and even somewhat negligible in the tropics. Therefore, the answer to **RQ 1.1** is that the observed MLC CF response to aerosols, represented by N_d , is positive overall, with a global area-weighted mean $\ln N_d$ –CF of 0.112, while its magnitude varies substantially across cloud regimes and regions.

At the same time, this explainable ML framework addresses **RQ 1.2** by examining the CF sensitivities to all meteorological predictors involved and their influences on the $\ln N_d$ –CF sensitivity. The results show that thermodynamic variables in general exert stronger controls on CF. Among them estimated inversion strength (EIS) and SST are the most important drivers of CF by regulating surface fluxes and entrainment processes. Higher EIS generally favours larger CF, whereas higher SST tends to reduce CF, particularly in stratocumulus regions, generally in agreement with previous studies on MLC controls (Klein and Hartmann, 1993; Wood and Bretherton, 2006; Qu et al., 2015b; Scott et al., 2020; Myers et al., 2021). Interaction indexes (IAIs) based on SHAP interaction values reveal that the $\ln N_d$ –CF sensitivity also depends more strongly on thermodynamic variables than on dynamical ones, similar to CF itself. EIS and SST are still among the most important ones along with RH at 850 hPa and temperatures at 700 and 850 hPa. Thus, the answer to **RQ 1.2** is that thermodynamic drivers are more important for both the direct control of MLC CF and the modulation of the N_d –CF relationship, with EIS and SST as the most important determinants.

The findings support the guiding hypotheses formulated in Sect. 1.5. First, it is confirmed that the CF response of MLCs to aerosol perturbations is positive but modulated—reduced or amplified—by ambient meteorology: the positive near-

global $\ln N_d$ -CF sensitivity supports the expected sign of the response, and the dependence on this response on local thermodynamic and dynamical conditions supports the expected modulation of meteorology. Second, CF sensitivities to $\ln N_d$ and meteorological variables as well as the meteorological modulation of the $\ln N_d$ -CF sensitivity exhibit distinct regional patterns, is also confirmed, and they are physically interpretable. For example, the spatial patterns of horizontal winds in the midlatitudes may suggest the contributions of midlatitude synoptic-scale disturbances and vertical wind shear to CF; the amplification of the $\ln N_d$ -CF sensitivity by higher EIS is a distinct feature of the midlatitudes, while the amplification by higher SST is localized to stratocumulus-to-cumulus transition zones. The coherent spatial distributions of the sensitivities and interaction effects suggest that the framework may capture meaningful regional characteristics.

Taken together, the observational study answers the first overarching research question (**RQ 1**) by showing that MLC CF responds positively to aerosol perturbations globally, but that this response is strongly state-dependent i.e. influenced by the meteorological environment and varies across cloud regimes. However, the observational results should be interpreted with limitations in mind. For example, the derived sensitivities and the meteorological influence on the N_d -CF relationship are subject to satellite retrieval issues. In addition, N_d and CF are not fully independent retrieved quantities. These effects can contribute to a positive bias in the $\ln N_d$ -CF relationship. Therefore, the derived sensitivity should be interpreted as an upper-bound estimate, in light of known uncertainties in satellite-derived N_d , cloud heterogeneity, and sampling choices (Grosvenor et al., 2018; Arola et al., 2022; Gryspeerd et al., 2022). Thus, the quantifications in Chap. 3 should be understood as data-driven estimates of the CF response guided by hypothesized causal pathways and the state-of-the-art physical understanding of the system.

5.1.2 Model-based perspective

The model-based study presented in Chap. 4 addressed **RQ 2** by applying the same XGBoost-SHAP framework from the observation-based study to ICON-HAM output. Six AMIP experiments were conducted, combining two cloud-cover parameterization schemes, CC-RH which depends solely on RH, and CC-RH-LWC where CF is a function of both RH and LWC, and three $N_{d,min}$ values (10, 40, and 100 cm^{-3}). Previous studies have shown that both these prescribed lower bounds and cloud-cover schemes with explicit cloud-water coupling can substantially affect the simulated aerosol indirect effect (Lohmann and Feichter, 1997; Hoose et al., 2009). As in the observational study, regional XGBoost models were trained

in aggregated $5^\circ \times 5^\circ$ geographical windows to predict CF from ICON-HAM $\ln N_d$ and meteorological predictors, and SHAP values were used to estimate the sensitivities. This setup allows the ICON-HAM $\ln N_d$ –CF sensitivities to be compared with the satellite-based estimates and, at the same time, allows the role of model configuration choices to be investigated.

Regarding **RQ 2.1**, the explainable ML framework is transferable to ICON-HAM output in the sense that the regional XGBoost models can predict CF and SHAP can be used to derive the CF response to N_d with meteorological influences taken into account. However, a methodological extension, PLR, is required because of the much more nonlinear $\ln N_d$ –CF relationships in ICON-HAM compared to those found in the MODIS-based study. Whereas the MODIS-based $\ln N_d$ –CF relationship can be reasonably captured by a single linear regression, the ICON-HAM-based relationship shows clear nonlinear saturation behaviour (Hu et al., 2021; Yuan et al., 2023), even after applying the logarithmic transformation to N_d . This nonlinearity is itself a physically meaningful result, as it likely reflects the small-scale threshold-like saturation behaviour in ICON-HAM’s parameterizations, whereas such abrupt local responses are more smoothed in the large-scale satellite-based relationships. The PLR approach divides the relationship into a low- N_d regime with stronger sensitivity and a high- N_d regime with much weaker sensitivity, consistent with previous evidence of saturation behaviour (Yuan et al., 2023; Wang et al., 2024). Hence, the answer to **RQ 2.1** is that the explainable ML framework can be transferred to ICON-HAM, but the stronger nonlinearity of the modelled relationships requires an adapted approach to estimate the $\ln N_d$ –CF sensitivity. The comparison with satellite-based estimates therefore shows not only differences in magnitude, but also a fundamentally different shape in the $\ln N_d$ –CF relationship.

Concerning **RQ 2.2**, the results show that the cloud-cover scheme and $N_{d,\min}$ jointly shape the $\ln N_d$ –CF sensitivity through a *headroom effect*. High $N_{d,\min}$ suppresses autoconversion, resulting in higher LWP in both CC-RH and CC-RH-LWC schemes (Khairoutdinov and Kogan, 2000; Hoose et al., 2009). However, the two schemes translate this increase into CF differently. In CC-RH-LWC, CF depends explicitly on LWC, so the increase in cloud water directly raises the climatological mean-state CF. In CC-RH, CF depends only on RH, so the same microphysical change does not translate into an equally strong increase in cloud cover. Therefore, the elevated climatological mean-state CF in CC-RH-LWC at high $N_{d,\min}$ leaves less room for further aerosol-induced CF increases. This “headroom effect” leads to weaker diagnosed $\ln N_d$ –CF sensitivities in CC-RH-LWC at high $N_{d,\min}$, despite the

explicit coupling between cloud water and cloud cover. In this sense, the answer to **RQ 2.2** is that model configuration choices shape the simulated aerosol-related CF response not only through direct physical pathways, but also through their effect on the mean-state CF and the available headroom.

These results can be discussed against the guiding hypotheses formulated in Sect. 1.5. The results confirm the first guiding hypothesis, as ICON-HAM-based relationships differ quantitatively and qualitatively from MODIS-based estimates by the stronger log-space nonlinearity, i.e. clearer saturation behaviour than the MODIS-based relationships. The second hypothesis, that CC-RH-LWC would exhibit larger $\ln N_d$ -CF sensitivities than CC-RH due to the explicit cloud-water dependence with $N_{d,\min}$ modulating these differently in each scheme, is only partially confirmed by our results and only holds at low $N_{d,\min}$ (10 cm^{-3}) and reverses at higher values (40 and 100 cm^{-3} , where 40 cm^{-3} is the default configuration setting), where the headroom effect outweighs the direct physical coupling in CC-RH-LWC. This reversal demonstrates that the original expectation is not generally valid, and the mean-state of CF can control the diagnosed marginal response.

Overall, the model-based study answers the second overarching research question (**RQ 2**) by showing that the CF response to aerosols in a GCM emerges as a positive and strongly nonlinear and saturating relationship with the consistent explainable ML framework. This response is affected by model configuration choices. In particular, cloud-cover parameterization and N_d values can shift the simulated mean-state CF and thereby alter the diagnosed sensitivity. Thus, background climatological CF state should be considered when interpreting GCM-based CF responses.

5.1.3 Synthesis: state-dependence as a unifying theme

After considering the observation- and model-based results separately, a common thesis-level message is that the CF response should be interpreted together with the physical context in which it is derived. While MLC CF generally increases with N_d globally, the magnitude of this response is state-dependent. Here, “state-dependent” means that the response depends on the background meteorological environment, the prevailing cloud regime, and, in models, the simulated mean-state CF.

In the observational analysis, this state dependence appears through meteorological modulation and regional cloud-regime differences, where the environment modifies CF itself and the local efficiency of the aerosol effect. In the ICON-HAM analysis, it is further shaped by model configuration choices that alter the sim-

ulated climatological mean-state CF and therefore the available headroom for additional aerosol-induced CF increases. Whether in the real atmosphere or a numerical model, treating the derived $\ln N_d$ -CF sensitivity as a single, regime-independent scalar is insufficient. This state-aware interpretation is consistent with the broader ACI literature, which shows that aerosol effects on clouds depend on cloud regime, meteorological environment, and background cloud state (e.g. Stevens and Feingold, 2009; Chen et al., 2014; Rosenfeld et al., 2019; Wang et al., 2024).

In terms of observation–model discrepancies, these findings suggest that the CF response cannot be directly constrained by comparing one globally averaged sensitivity alone, especially when considering that many GCMs simulate relatively weak CF adjustments compared to the Twomey effect and LWP adjustment (e.g. Smith et al., 2020; Zelinka et al., 2023). Instead, it should be clarified under which climatological CF, meteorological conditions, cloud regimes, and model configurations a given sensitivity is derived. From this perspective, differences between observation- and model-based estimates may arise not only from parameterizations of microphysical processes, but also from differences in the climatological background states in which the response occurs.

5.2 Outlook

The results of this thesis, combining global satellite observations and a GCM under a consistent explainable ML framework, advance the understanding of the CF response to aerosol perturbations in MLCs and open several directions for future work.

On the observational side, as discussed in Sects. 2.1.3 and 3.2.3.3, the N_d retrieval can be improved by using retrievals based on joint histograms rather than large-scale mean cloud properties, e.g. the pre-filtered N_d dataset from Gryspeerdt et al. (2022). This issue is expected to be minor because the associated bias is likely small. Another path to improving satellite data quality is to combine active and passive satellite observations, for example from the Earth Cloud, Aerosol and Radiation Explorer (EarthCARE) (Wehr et al., 2023) to resolve cloud vertical structure and identify multilayer scenes (Stephens et al., 2018; Xu et al., 2026), while newer geostationary platforms with improved spatial and temporal resolution such as Meteosat Third Generation (MTG) (Holmlund et al., 2021) could help mitigate biases associated with subpixel heterogeneity in broken-cloud regimes (Werner and Deneke, 2020; Deneke et al., 2021). More broadly, combining complemen-

tary observations from multiple current and upcoming satellite platforms and reanalysis data can be a path to reducing meteorological confounding and retrieval uncertainties in ACI (Jia et al., 2022; Im et al., 2026).

A natural next step is to move from statistical relationships towards more causal interpretations. The XGBoost–SHAP framework used in this thesis accounts for many meteorological covariates by explicitly including them as predictors and provides interpretable estimates of the $\ln N_d$ –CF sensitivity, but it does not by itself establish causality. This direction is meaningful because N_d , CF, and meteorology are strongly coupled, and satellite retrieval uncertainties can introduce non-causal components into the SHAP-based relationships. Future work could hence combine the explainable ML framework with causal-inference approaches, for example causal discovery algorithms (Runge et al., 2019; Nowack et al., 2020), or more directly connected to the SHAP framework in this thesis, causal SHAP methods (Heskes et al., 2020; Frye et al., 2020). The former could help identify and mitigate spurious relationships among N_d , CF, and meteorological variables, while the latter could provide causally grounded feature attributions within the existing SHAP decomposition.

A natural next extension of the present work is to translate the $\ln N_d$ –CF sensitivities derived here into estimates of radiative forcing. The regional sensitivities should be combined with the anthropogenic N_d perturbation and follow a top-down sensitivity-based approach similar to Gryspeerdt et al. (2016), or using a radiative kernel to convert ΔCF into radiative impact (Zelinka et al., 2014; Wall et al., 2023) to derive a global observational constraint on the effective radiative forcing (ERF) due to the CF adjustment. This can be complemented by bottom-up modelling in which PI–PD ICON–HAM simulations can be run to yield this forcing and test whether the headroom mechanism identified in Chap. 4 also mutes the diagnosed CF adjustment forcing under specific model configurations.

Another future direction is to make the sensitivity estimates more explicitly state-aware. As discussed in Sect. 5.1.3, the $\ln N_d$ –CF sensitivity should not be interpreted independently of the background state in which it is derived. Future studies could therefore condition the sensitivity estimates on cloud regime and mean-state cloud properties or specific meteorological regimes, for example CF, LWP, precipitation state, EIS, SST and humidity. Such an approach is consistent with previous studies showing that the MLC response depends on prevailing cloud and meteorological state (Stevens and Feingold, 2009; Chen et al., 2014; Rosenfeld et al., 2019; Chen et al., 2024). In practice, this could mean deriving separate $\ln N_d$ –CF sensitivities for different background CF or LWP ranges, and then testing

whether models reproduce not only the global mean sensitivity but also the distribution of sensitivities across cloud states. Such a state-aware comparison would be particularly useful for evaluating whether the discrepancy is from deficient process-level parameterizations or just muted by the background climatology.

Another direction concerns the generalization of the present explainable ML framework, including its application beyond CF and beyond one GCM. First, the XGBoost–SHAP framework could be extended from the CF adjustment to the LWP adjustment. SHAP values and SHAP interaction values have already been used to explain LWP predictions from tree-based models for the southeast Atlantic (Zipfel et al., 2022) and for five marine stratocumulus regions (Zipfel et al., 2024). This could be extended to the region-specific XGBoost–SHAP framework in this thesis to obtain global spatial characteristics of the N_d –LWP sensitivity. Applying the consistent framework to both CF and LWP for both observations and GCMs would help understand the disagreement between observations and GCMs regarding the relative importance of the two adjustments to ERF_{aci}, specifically, that the LWP adjustment dominates in GCMs while observational evidence suggests the CF adjustment is stronger (e.g. Zelinka et al., 2023; Toll et al., 2019; Chen et al., 2022b). The same idea might also be extended to other cloud properties, such as cloud albedo, precipitation, or cloud radiative effects (CREs). Second, applying the same analysis to other GCMs would test whether the log-space nonlinearity and the headroom effect found in Chap. 4 are specific to ICON–HAM’s cloud-cover parameterizations and $N_{d,min}$, or reflect a more general behaviour. Applying this analysis to a broader range of GCMs, such as those within the CMIP6 ensemble (Eyring et al., 2016), would place these findings in a larger model intercomparison context. Further experiments with alternative cloud-cover parameterizations, e.g. a prognostic scheme (Muench and Lohmann, 2020), or with modified parameters of key microphysical processes such as the autoconversion rate as in (Wang et al., 2025), would help identify which parts of the model cloud representation are most responsible.

Finally, the meteorological interaction effects i.e. the IAI results demonstrated in Chap. 3 could help explore how the CF response to aerosols may change in a warming climate by using projected changes in cloud-controlling factors. Global warming is projected to alter key meteorological drivers of MLCs, including SST, EIS, and humidity (Qu et al., 2015b; Bony et al., 2015; Myers et al., 2021), which are shown in Chap. 3 to amplify or weaken the $\ln N_d$ –CF sensitivity in spatially distinct ways. Their projected changes could therefore in principle be combined with the derived IAIs to estimate how the CF adjustment may shift in the future,

although this should be interpreted cautiously as it assumes that the PD statistical interactions remain valid under warming.

Overall, this thesis shows that the $\ln N_d$ -CF sensitivity can be interpreted more meaningfully when meteorological covariability is accounted for and when the results are considered in the context of the background cloud state. The results provide an observation-based constraint on the $\ln N_d$ -CF sensitivity and show how model configuration choices can shape the diagnosed response. Future progress will require improved observations, more causal and state-aware methods, direct links to radiative forcing, exploration of future climate implications of the IAI patterns, and broader model comparisons to better constrain how aerosols affect marine low-cloud cover.

Appendix A

Supplementary material of Chapter 3

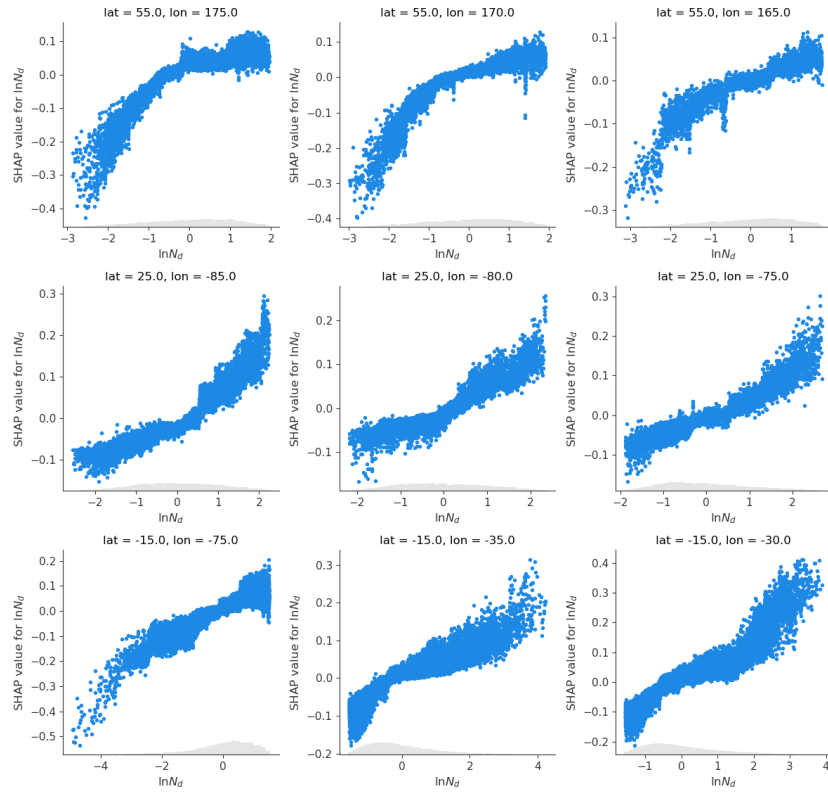


Figure A.1: SHapley Additive exPlanations (SHAP) dependence plots similar to Fig. 3.1b illustrating relatively nonlinear relationships between $\ln N_d$ SHAP values and feature values (standardized). The latitude and longitude values for each subplot represent the midpoint of each $5^\circ \times 5^\circ$ geographical window.

Figures A.2 to A.7 illustrate results from the same analysis but using non-standardized predictor variables. The box plots Figs. 3.3 and 3.7 do not have corresponding non-standardized versions here because the magnitudes of the sensitivities and interaction index (IAI) are not physically comparable without the standardization process.

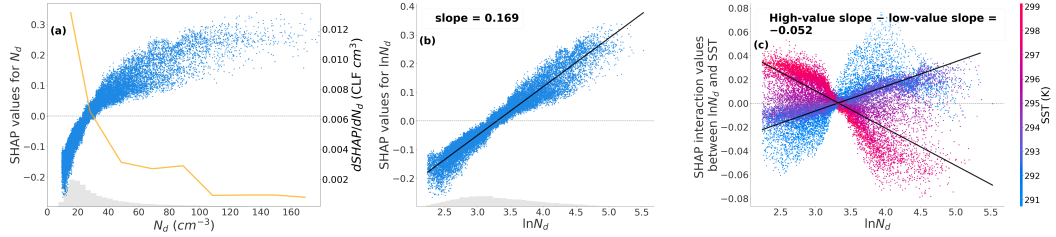


Figure A.2: SHAP dependence plots for cloud droplet number concentration (N_d) in the region from 27.5° S to 32.5° S and from 122.5° W to 127.5° W. Panel (a) is identical to the corresponding panel in Fig. 3.1, while $\ln N_d$ and sea surface temperature (SST) are not standardized in panels (b) and (c).

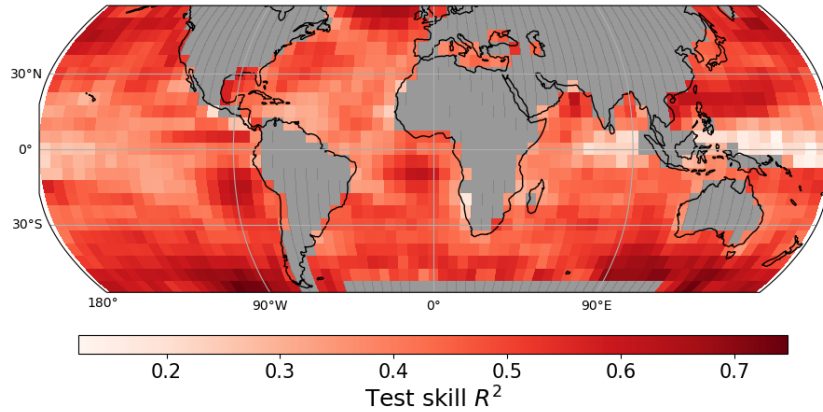


Figure A.3: R^2 score of regional eXtreme Gradient Boosting (XGBoost) models predicting cloud fraction (CF) of marine low clouds (MLCs) in the independent test dataset (2017–2019). Similar to Fig. 3.2 but predictor variables are not standardized.

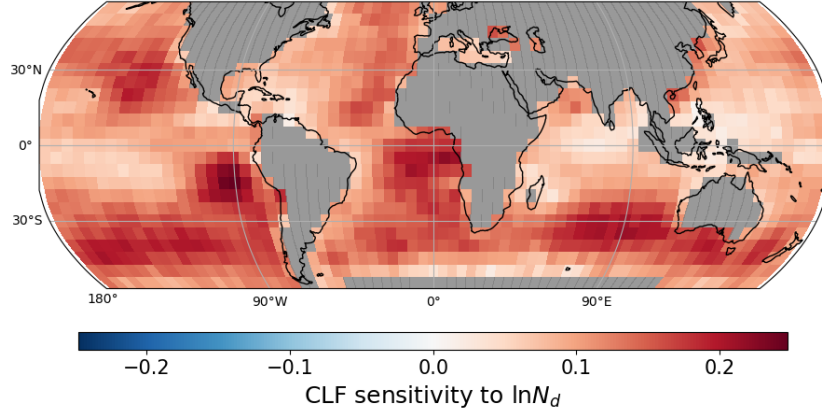


Figure A.4: Sensitivity of MLC CF to $\ln N_d$. Similar to Fig. 3.4 but predictor variables are not standardized.

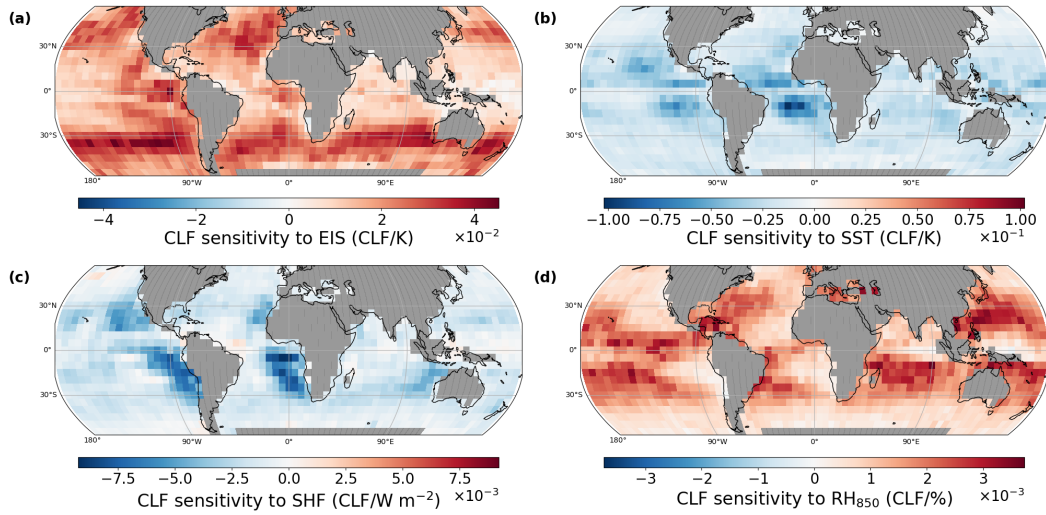


Figure A.5: Sensitivity of MLC CF to estimated inversion strength (EIS), SST, sensible heat flux (SHF), and relative humidity (RH) at 850 hPa. Similar to Fig. 3.5 but predictor variables are not standardized, and thus units and ranges of colourbars vary.

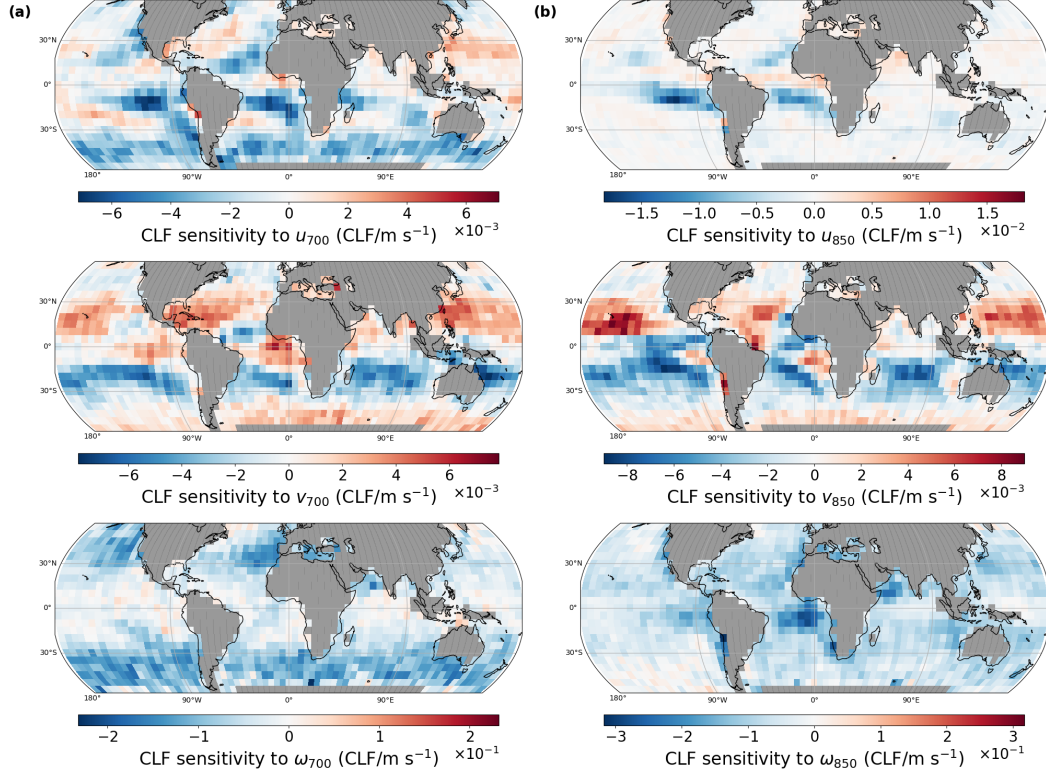


Figure A.6: Sensitivity of CF to zonal (eastward) wind (u), meridional (northward) wind (v) and vertical velocity (ω) at 700 hPa (column a) and 850 hPa (column b). Similar to Fig. 3.6 but predictor variables are not standardized, and thus units and ranges of colourbars vary.

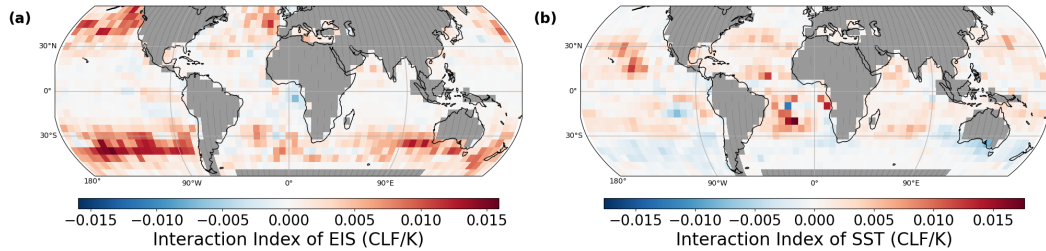


Figure A.7: Patterns of the IAI showing the dependence of the ln-CF relationship on EIS (a) and SST (b). Similar to Fig. 3.8 but predictor variables are not standardized.

Appendix B

Supplementary material of Chapter 4

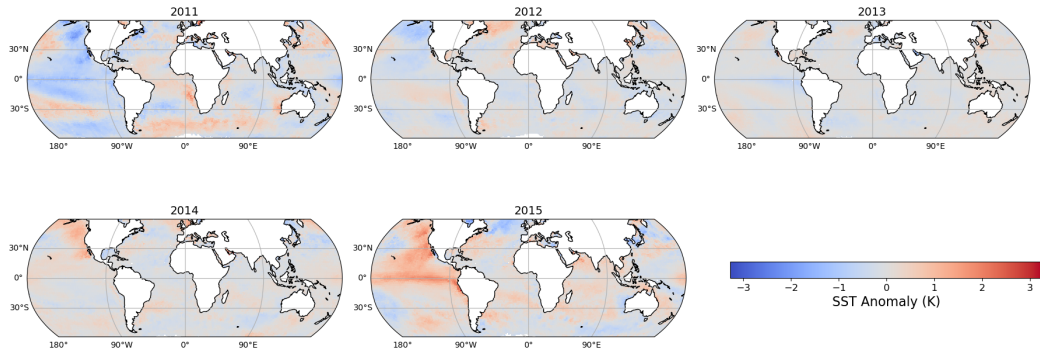


Figure B.1: Spatial distributions of annual mean SST anomalies (K) for 2011–2015 (CC-RH cloud-cover scheme, prescribed lower bound for N_d ($N_{d,\min} = 40 \text{ cm}^{-3}$)). Anomalies are computed as the annual mean minus the 2011–2015 multi-year mean. Panels show individual years; the sixth panel shows the colour scale.

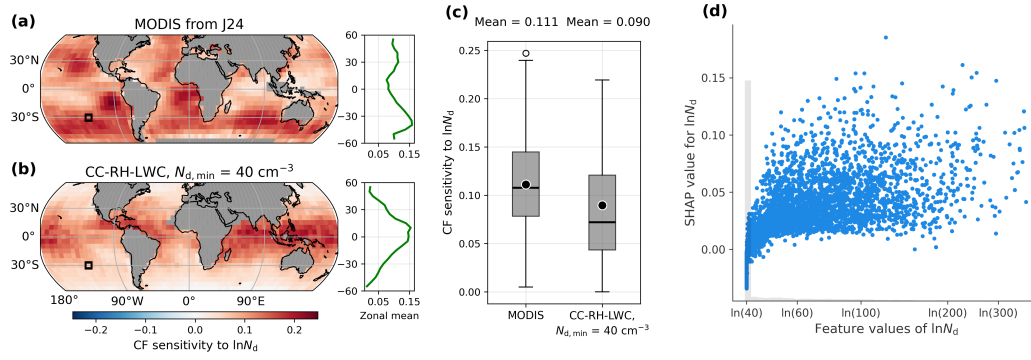


Figure B.2: Similar to Fig. 4.2 in Sect. 4.3.2, but for the ICON-HAM experiment using the CC-RH-LWC cloud-cover scheme with $N_{d,min} = 40 \text{ cm}^{-3}$. This alternative cloud-cover parameterization is evaluated to ensure the robustness of the identified sensitivity patterns across different model structural configurations.

Bibliography

- Abdul-Razzak, H. and Ghan, S. J.: A parameterization of aerosol activation 2. Multiple aerosol types, *Journal of Geophysical Research Atmospheres*, 105, 6837–6844, doi: 10.1029/1999JD901161, 2000.
- Ackerman, A. S., Toon, O. B., Taylor, J. P., Johnson, D. W., Hobbs, P. V., and Ferek, R. J.: Effects of Aerosols on Cloud Albedo: Evaluation of Twomey’s Parameterization of Cloud Susceptibility Using Measurements of Ship Tracks, *Journal of the Atmospheric Sciences*, 57, 2684–2695, doi: [https://doi.org/10.1175/1520-0469\(2000\)057<2684:EOAOCA>2.0.CO;2](https://doi.org/10.1175/1520-0469(2000)057<2684:EOAOCA>2.0.CO;2), 2000.
- Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017, doi: 10.1038/nature03174, 2004.
- Ackerman, S. A., Strabala, K. I., Menzel, W. P., Frey, R. A., Moeller, C. C., and Gumley, L. E.: Discriminating clear sky from clouds with MODIS, *Journal of Geophysical Research: Atmospheres*, 103, 32 141–32 157, doi: <https://doi.org/10.1029/1998JD200032>, 1998.
- Adebisi, A. A. and Zuidema, P.: The role of the southern African easterly jet in modifying the southeast Atlantic aerosol and cloud environments, *Quarterly Journal of the Royal Meteorological Society*, 142, 1574–1589, doi: <https://doi.org/10.1002/qj.2765>, 2016.
- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, doi: 10.1126/science.245.4923.1227, 1989.
- Andersen, H. and Cermak, J.: How thermodynamic environments control stratocumulus microphysics and interactions with aerosols, *Environmental Research Letters*, 10, doi: 10.1088/1748-9326/10/2/024004, 2015.

- Andersen, H., Cermak, J., Fuchs, J., and Schwarz, K.: Global observations of cloud-sensitive aerosol loadings in low-level marine clouds, *Journal of Geophysical Research*, 121, 936–12, doi: 10.1002/2016JD025614, 2016.
- Andersen, H., Cermak, J., Fuchs, J., Knutti, R., and Lohmann, U.: Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks, *Atmospheric Chemistry and Physics*, 17, 9535–9546, doi: 10.5194/acp-17-9535-2017, 2017.
- Andersen, H., Cermak, J., Stirnberg, R., Fuchs, J., Kim, M., and Pauli, E.: Assessment of COVID-19 effects on satellite-observed aerosol loading over China with machine learning, *Tellus B: Chemical and Physical Meteorology*, 73, 1–14, doi: 10.1080/16000889.2021.1971925, 2021.
- Andersen, H., Cermak, J., Zipfel, L., and Myers, T. A.: Attribution of Observed Recent Decrease in Low Clouds Over the Northeastern Pacific to Cloud-Controlling Factors, *Geophysical Research Letters*, 49, 1–10, doi: 10.1029/2021gl096498, 2022.
- Andersen, H., Cermak, J., Douglas, A., Myers, T. A., Nowack, P., Stier, P., Wall, C. J., and Wilson Kemsley, S.: Sensitivities of cloud radiative effects to large-scale meteorology and aerosols from global observations, *Atmospheric Chemistry and Physics*, 23, 10 775–10 794, doi: 10.5194/acp-23-10775-2023, 2023.
- Angal, A., Xiong, X., Twedt, K., Chang, T., Geng, X., Aldoretta, E., and Díaz, C. P.: Status of the Terra and Aqua Modis Collection 7 L1B, in: *IGARSS 2023 - 2023 IEEE International Geoscience and Remote Sensing Symposium*, pp. 4466–4469, doi: 10.1109/IGARSS52108.2023.10283303, 2023.
- Arola, A., Lipponen, A., Kolmonen, P., Virtanen, T. H., Bellouin, N., Grosvenor, D. P., Gryspeerdt, E., Quaas, J., and Kokkola, H.: Aerosol effects on clouds are concealed by natural cloud heterogeneity and satellite retrieval errors, *Nature Communications*, 13, 7357, doi: 10.1038/s41467-022-34948-5, 2022.
- Barredo Arrieta, A., Díaz-Rodríguez, N., Del Ser, J., Bennetot, A., Tabik, S., Barbado, A., Garcia, S., Gil-Lopez, S., Molina, D., Benjamins, R., Chatila, R., and Herrera, F.: Explainable Artificial Intelligence (XAI): Concepts, taxonomies, opportunities and challenges toward responsible AI, *Information Fusion*, 58, 82–115, doi: <https://doi.org/10.1016/j.inffus.2019.12.012>, 2020.
- Baum, B. A., Menzel, W. P., Frey, R. A., Tobin, D. C., Holz, R. E., Ackerman, S. A., Heidinger, A. K., and Yang, P.: MODIS Cloud-Top Property Refinements for

- Collection 6, *Journal of Applied Meteorology and Climatology*, 51, 1145–1163, doi: <https://doi.org/10.1175/JAMC-D-11-0203.1>, 2012.
- Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A. L., Dufresne, J. L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B.: Bounding Global Aerosol Radiative Forcing of Climate Change, *Reviews of Geophysics*, 58, 1–45, doi: 10.1029/2019RG000660, 2020.
- Belmonte Rivas, M. and Stoffelen, A.: Characterizing ERA-Interim and ERA5 surface wind biases using ASCAT, *Ocean Science*, 15, 831–852, doi: 10.5194/os-15-831-2019, 2019.
- Bender, F. A., Frey, L., McCoy, D. T., Grosvenor, D. P., and Mohrmann, J. K.: Assessment of aerosol–cloud–radiation correlations in satellite observations, climate models and reanalysis, *Climate Dynamics*, 52, 4371–4392, doi: 10.1007/s00382-018-4384-z, 2019.
- Bender, F. A., Lord, T., Staffansdotter, A., Jung, V., and Undorf, S.: Machine Learning Approach to Investigating the Relative Importance of Meteorological and Aerosol-Related Parameters in Determining Cloud Microphysical Properties, *Tellus B: Chemical and Physical Meteorology*, 76, 1–18, doi: 10.16993/tellusb.1868, 2024.
- Bennartz, R. and Rausch, J.: Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations, *Atmospheric Chemistry and Physics*, 17, 9815–9836, doi: 10.5194/acp-17-9815-2017, 2017.
- Beucler, T., Ebert-Uphoff, I., Rasp, S., Pritchard, M., and Gentine, P.: Machine Learning for Clouds and Climate, in: *Clouds and Their Climatic Impacts*, edited by Hoose, S. S. and C., *Geophysical Monograph Series*, pp. 325–345, John Wiley & Sons, Inc., doi: <https://doi.org/10.1002/9781119700357.ch16>, 2023.
- Bi, K., Xie, L., Zhang, H., Chen, X., Gu, X., and Tian, Q.: Accurate medium-range global weather forecasting with 3D neural networks, *Nature*, 619, 533–538, doi: 10.1038/s41586-023-06185-3, 2023.

- Blossey, P. N., Bretherton, C. S., Zhang, M., Cheng, A., Endo, S., Heus, T., Liu, Y., Lock, A. P., de Roode, S. R., and Xu, K.-M.: Marine low cloud sensitivity to an idealized climate change: The CGILS LES intercomparison, *Journal of Advances in Modeling Earth Systems*, 5, 234–258, doi: <https://doi.org/10.1002/jame.20025>, 2013.
- Bock, L. and Lauer, A.: Cloud properties and their projected changes in CMIP models with low to high climate sensitivity, *Atmospheric Chemistry and Physics*, 24, 1587–1605, doi: [10.5194/acp-24-1587-2024](https://doi.org/10.5194/acp-24-1587-2024), 2024.
- Bonavita, M. and Laloyaux, P.: Machine Learning for Model Error Inference and Correction, *Journal of Advances in Modeling Earth Systems*, 12, e2020MS002 232, doi: <https://doi.org/10.1029/2020MS002232>, 2020.
- Bony, S. and Dufresne, J.-L.: Marine boundary layer clouds at the heart of tropical cloud feedback uncertainties in climate models, *Geophysical Research Letters*, 32, doi: <https://doi.org/10.1029/2005GL023851>, 2005.
- Bony, S., Colman, R., Kattsov, V. M., Allan, R. P., Bretherton, C. S., Dufresne, J.-L., Hall, A., Hallegatte, S., Holland, M. M., Ingram, W., Randall, D. A., Soden, B. J., Tselioudis, G., and Webb, M. J.: How Well Do We Understand and Evaluate Climate Change Feedback Processes?, *Journal of Climate*, 19, 3445–3482, doi: <https://doi.org/10.1175/JCLI3819.1>, 2006.
- Bony, S., Stevens, B., Frierson, D. M. W., Jakob, C., Kageyama, M., Pincus, R., Shepherd, T. G., Sherwood, S. C., Siebesma, A. P., Sobel, A. H., Watanabe, M., and Webb, M. J.: Clouds, circulation and climate sensitivity, *Nature Geoscience*, 8, 261–268, doi: [10.1038/ngeo2398](https://doi.org/10.1038/ngeo2398), 2015.
- Boucher, O. and Lohmann, U.: The sulfate-CCN-cloud albedo effect, *Tellus B: Chemical and Physical Meteorology*, 47, 281–300, doi: [10.3402/tellusb.v47i3.16048](https://doi.org/10.3402/tellusb.v47i3.16048), 1995.
- Boucher, O., Granier, C., Hoose, C., and Andy, J.: Clouds and aerosols, *Climate Change 2013 the Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, 97811070057, 571–658, doi: [10.1017/CBO9781107415324.016](https://doi.org/10.1017/CBO9781107415324.016), 2013.
- Bracco, A., Brajard, J., Dijkstra, H. A., Hassanzadeh, P., Lessig, C., and Monteleoni, C.: Machine learning for the physics of climate, *Nature Reviews Physics*, 7, 6–20, doi: [10.1038/s42254-024-00776-3](https://doi.org/10.1038/s42254-024-00776-3), 2025.

- Breiman, L.: Bagging predictors, *Machine Learning*, 24, 123–140, doi: 10.1007/BF00058655, 1996.
- Breiman, L.: Random Forests, *Machine Learning*, 45, 5–32, doi: 10.1023/A:1010933404324, 2001.
- Brenguier, J.-L., Pawlowska, H., Schüller, L., Preusker, R., Fischer, J., and Fouquart, Y.: Radiative Properties of Boundary Layer Clouds: Droplet Effective Radius versus Number Concentration, *Journal of the Atmospheric Sciences*, 57, 803–821, doi: [https://doi.org/10.1175/1520-0469\(2000\)057<0803:RPOBLC>2.0.CO;2](https://doi.org/10.1175/1520-0469(2000)057<0803:RPOBLC>2.0.CO;2), 2000.
- Bretherton, C. S.: Insights into low-latitude cloud feedbacks from high-resolution models, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 373, 20140415, doi: 10.1098/rsta.2014.0415, 2015.
- Bretherton, C. S. and Blossey, P. N.: Low cloud reduction in a greenhouse-warmed climate: Results from Lagrangian LES of a subtropical marine cloudiness transition, *Journal of Advances in Modeling Earth Systems*, 6, 91–114, doi: <https://doi.org/10.1002/2013MS000250>, 2014.
- Bretherton, C. S. and Blossey, P. N.: Understanding Mesoscale Aggregation of Shallow Cumulus Convection Using Large-Eddy Simulation, *Journal of Advances in Modeling Earth Systems*, 9, 2798–2821, doi: <https://doi.org/10.1002/2017MS000981>, 2017.
- Bretherton, C. S. and Wyant, M. C.: Moisture Transport, Lower-Tropospheric Stability, and Decoupling of Cloud-Topped Boundary Layers, *Journal of the Atmospheric Sciences*, 54, 148–167, doi: [https://doi.org/10.1175/1520-0469\(1997\)054<0148:MTLTSA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1997)054<0148:MTLTSA>2.0.CO;2), 1997.
- Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, *Geophysical Research Letters*, 34, doi: <https://doi.org/10.1029/2006GL027648>, 2007.
- Bretherton, C. S., Blossey, P. N., and Jones, C. R.: Mechanisms of marine low cloud sensitivity to idealized climate perturbations: A single-LES exploration extending the CGILS cases, *Journal of Advances in Modeling Earth Systems*, 5, 316–337, doi: <https://doi.org/10.1002/jame.20019>, 2013.

- Brueck, M., Nuijens, L., and Stevens, B.: On the Seasonal and Synoptic Time-Scale Variability of the North Atlantic Trade Wind Region and Its Low-Level Clouds, *Journal of the Atmospheric Sciences*, 72, 1428–1446, doi: <https://doi.org/10.1175/JAS-D-14-0054.1>, 2015.
- Brunton, S. L., Noack, B. R., and Koumoutsakos, P.: Machine Learning for Fluid Mechanics, *Annual Review of Fluid Mechanics*, 52, 477–508, doi: <https://doi.org/10.1146/annurev-fluid-010719-060214>, 2020.
- Bühl, J., Alexander, S., Crewell, S., Heymsfield, A., Kalesse, H., Khain, A., Maahn, M., Van Tricht, K., and Wendisch, M.: Remote Sensing, *Meteorological Monographs*, 58, 1–10, doi: <https://doi.org/10.1175/AMSMONOGRAPHIS-D-16-0015.1>, 2017.
- Cairo, F., Di Liberto, L., Dionisi, D., and Snels, M.: Understanding Aerosol–Cloud Interactions through Lidar Techniques: A Review, doi: 10.3390/rs16152788, 2024.
- Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G. W., Spracklen, D. V., Woodhouse, M. T., Regayre, L. A., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing, *Nature*, 503, 67–71, doi: 10.1038/nature12674, 2013.
- Ceppi, P. and Nowack, P.: Observational evidence that cloud feedback amplifies global warming, *Proceedings of the National Academy of Sciences*, 118, e2026290 118, doi: 10.1073/pnas.2026290118, 2021.
- Cesana, G., Del Genio, A. D., Ackerman, A. S., Kelley, M., Elsaesser, G., Fridlind, A. M., Cheng, Y., and Yao, M.-S.: Evaluating models’ response of tropical low clouds to SST forcings using CALIPSO observations, *Atmospheric Chemistry and Physics*, 19, 2813–2832, doi: 10.5194/acp-19-2813-2019, 2019.
- Cesana, G. V. and Del Genio, A. D.: Observational constraint on cloud feedbacks suggests moderate climate sensitivity, *Nature Climate Change*, 11, 213–218, doi: 10.1038/s41558-020-00970-y, 2021.
- Chen, H., Janizek, J. D., Lundberg, S., and Lee, S.-I.: True to the Model or True to the Data?, *ArXiv*, abs/2006.1, URL <https://arxiv.org/abs/2006.16234>, 2020.
- Chen, H., Lundberg, S. M., and Lee, S.-I.: Explaining a series of models by propagating Shapley values, *Nature Communications*, 13, 4512, doi: 10.1038/s41467-022-31384-3, 2022a.

- Chen, H., Covert, I. C., Lundberg, S. M., and Lee, S.-I.: Algorithms to estimate Shapley value feature attributions, *Nature Machine Intelligence*, 5, 590–601, doi: 10.1038/s42256-023-00657-x, 2023.
- Chen, T. and Guestrin, C.: XGBoost: A Scalable Tree Boosting System, in: *Proceedings of the 22nd ACM SIGKDD International Conference on Knowledge Discovery and Data Mining, KDD '16*, pp. 785–794, ACM, doi: 10.1145/2939672.2939785, 2016.
- Chen, T., Rossow, W. B., and Zhang, Y.: Radiative Effects of Cloud-Type Variations, *Journal of Climate*, 13, 264–286, doi: [https://doi.org/10.1175/1520-0442\(2000\)013<0264:REOCTV>2.0.CO;2](https://doi.org/10.1175/1520-0442(2000)013<0264:REOCTV>2.0.CO;2), 2000.
- Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Partridge, D., Fieldsend, J., De Leeuw, J., Schmidt, A., Cho, N., Oreopoulos, L., Platnick, S., Grosvenor, D., Field, P., and Lohmann, U.: Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover, *Nature Geoscience*, 15, 609–614, doi: 10.1038/s41561-022-00991-6, 2022b.
- Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Peace, A., Partridge, D. G., Cho, N., Oreopoulos, L., Grosvenor, D., Field, P., Allan, R. P., and Lohmann, U.: Substantial cooling effect from aerosol-induced increase in tropical marine cloud cover, *Nature Geoscience*, doi: 10.1038/s41561-024-01427-z, 2024.
- Chen, Y. C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds, *Nature Geoscience*, 7, 643–646, doi: 10.1038/ngeo2214, 2014.
- Christensen, M. W., Chen, Y.-C., and Stephens, G. L.: Aerosol indirect effect dictated by liquid clouds, *Journal of Geophysical Research: Atmospheres*, 121, 614–636, doi: <https://doi.org/10.1002/2016JD025245>, 2016.
- Christensen, M. W., Neubauer, D., Poulsen, C. A., Thomas, G. E., McGarragh, G. R., Povey, A. C., Proud, S. R., and Grainger, R. G.: Unveiling aerosol–cloud interactions – Part 1: Cloud contamination in satellite products enhances the aerosol indirect forcing estimate, *Atmospheric Chemistry and Physics*, 17, 13 151–13 164, doi: 10.5194/acp-17-13151-2017, 2017.
- Christensen, M. W., Jones, W. K., and Stier, P.: Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition, *Proceedings of the National Academy of Sciences of the United States of America*, 117, 17 591–17 598, doi: 10.1073/pnas.1921231117, 2020.

- Christensen, M. W., Gettelman, A., Cermak, J., Dagan, G., Diamond, M., Douglas, A., Feingold, G., Glassmeier, F., Goren, T., Grosvenor, D. P., Gryspeerdt, E., Kahn, R., Li, Z., Ma, P.-L., Malavelle, F., McCoy, I. L., McCoy, D. T., McFarquhar, G., Mülmenstädt, J., Pal, S., Possner, A., Povey, A., Quaas, J., Rosenfeld, D., Schmidt, A., Schrödner, R., Sorooshian, A., Stier, P., Toll, V., Watson-Parris, D., Wood, R., Yang, M., and Yuan, T.: Opportunistic experiments to constrain aerosol effective radiative forcing, *Atmospheric Chemistry and Physics*, 22, 641–674, doi: 10.5194/acp-22-641-2022, 2022.
- Coakley, J. A., Bernstein, R. L., and Durkee, P. A.: Effect of Ship-Stack Effluents on Cloud Reflectivity, *Science*, 237, 1020–1022, doi: 10.1126/science.237.4818.1020, 1987.
- Dadashazar, H., Painemal, D., Alipanah, M., Brunke, M., Chellappan, S., Corral, A. F., Crosbie, E., Kirschler, S., Liu, H., Moore, R. H., Robinson, C., Scarino, A. J., Shook, M., Sinclair, K., Thornhill, K. L., Voigt, C., Wang, H., Winstead, E., Zeng, X., Ziemba, L., Zuidema, P., and Sorooshian, A.: Cloud drop number concentrations over the western North Atlantic Ocean: seasonal cycle, aerosol interrelationships, and other influential factors, *Atmospheric Chemistry and Physics*, 21, 10 499–10 526, doi: 10.5194/acp-21-10499-2021, 2021.
- de Szoeke, S. P., Verlinden, K. L., Yuter, S. E., and Mechem, D. B.: The Time Scales of Variability of Marine Low Clouds, *Journal of Climate*, 29, 6463–6481, doi: <https://doi.org/10.1175/JCLI-D-15-0460.1>, 2016.
- Denby, L.: Discovering the Importance of Mesoscale Cloud Organization Through Unsupervised Classification, *Geophysical Research Letters*, 47, e2019GL085 190, doi: <https://doi.org/10.1029/2019GL085190>, 2020.
- Deneke, H., Barrientos-Velasco, C., Bley, S., Hünerbein, A., Lenk, S., Macke, A., Meirink, J. F., Schroedter-Homscheidt, M., Senf, F., Wang, P., Werner, F., and Witthuhn, J.: Increasing the spatial resolution of cloud property retrievals from Meteosat SEVIRI by use of its high-resolution visible channel: implementation and examples, *Atmospheric Measurement Techniques*, 14, 5107–5126, doi: 10.5194/amt-14-5107-2021, 2021.
- Diamond, M. S., Saide, P. E., Zuidema, P., Ackerman, A. S., Doherty, S. J., Fridlind, A. M., Gordon, H., Howes, C., Kazil, J., Yamaguchi, T., Zhang, J., Feingold, G., and Wood, R.: Cloud adjustments from large-scale smoke–circulation interactions strongly modulate the southeastern Atlantic stratocumulus-to-cumulus transi-

- tion, *Atmospheric Chemistry and Physics*, 22, 12 113–12 151, doi: 10.5194/acp-22-12113-2022, 2022.
- Dietterich, T. G.: Ensemble Methods in Machine Learning, in: *Multiple Classifier Systems*, pp. 1–15, Springer Berlin Heidelberg, Berlin, Heidelberg, 2000.
- Douglas, A. R. and L’Ecuyer, T.: Possible evidence of increased global cloudiness due to aerosol-cloud interactions, *Atmospheric Chemistry and Physics Discussions*, 2022, 1–22, URL <https://acp.copernicus.org/preprints/acp-2022-688/>, 2022.
- Duran, B. M., Wall, C. J., Lutsko, N. J., Michibata, T., Ma, P.-L., Qin, Y., Duffy, M. L., Medeiros, B., and Debolskiy, M.: A new method for diagnosing effective radiative forcing from aerosol–cloud interactions in climate models, *Atmospheric Chemistry and Physics*, 25, 2123–2146, doi: 10.5194/acp-25-2123-2025, 2025.
- Elith, J., Leathwick, J. R., and Hastie, T.: A working guide to boosted regression trees, *Journal of Animal Ecology*, 77, 802–813, doi: <https://doi.org/10.1111/j.1365-2656.2008.01390.x>, 2008.
- Eyring, V., Bony, S., Meehl, G. A., Senior, C. A., Stevens, B., Stouffer, R. J., and Taylor, K. E.: Overview of the Coupled Model Intercomparison Project Phase 6 (CMIP6) experimental design and organization, *Geoscientific Model Development*, 9, 1937–1958, doi: 10.5194/gmd-9-1937-2016, 2016.
- Fan, J., Wang, Y., Rosenfeld, D., and Liu, X.: Review of aerosol-cloud interactions: Mechanisms, significance, and challenges, doi: 10.1175/JAS-D-16-0037.1, 2016.
- Field, P. R. and Wood, R.: Precipitation and Cloud Structure in Midlatitude Cyclones, *Journal of Climate*, 20, 233–254, doi: <https://doi.org/10.1175/JCLI3998.1>, 2007.
- Fons, E., Runge, J., Neubauer, D., and Lohmann, U.: Stratocumulus adjustments to aerosol perturbations disentangled with a causal approach, *npj Climate and Atmospheric Science*, 6, 1–10, doi: 10.1038/s41612-023-00452-w, 2023.
- Fons, E., Naumann, A. K., Neubauer, D., Lang, T., and Lohmann, U.: Investigating the sign of stratocumulus adjustments to aerosols in the ICON global storm-resolving model, *Atmospheric Chemistry and Physics*, 24, 8653–8675, doi: 10.5194/acp-24-8653-2024, 2024.
- Forster, P. M., Storelvmo, T., Armour, K., Collins, W., Dufresne, J. L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D., Watanabe, M., Wild, M., and Zhang,

- H.: Chapter 7: The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity, in: *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J., Maycock, T. K., Waterfield, T., Yelekçi, O., Yu, R., and Zhou, B., p. in press, Cambridge University Press, 2021.
- Franke, H. and Giorgetta, M. A.: Toward the Direct Simulation of the Quasi-Biennial Oscillation in a Global Storm-Resolving Model, *Journal of Advances in Modeling Earth Systems*, 16, e2024MS004381, doi: <https://doi.org/10.1029/2024MS004381>, 2024.
- Frey, R. A., Ackerman, S. A., Liu, Y., Strabala, K. I., Zhang, H., Key, J. R., and Wang, X.: Cloud Detection with MODIS. Part I: Improvements in the MODIS Cloud Mask for Collection 5, *Journal of Atmospheric and Oceanic Technology*, 25, 1057–1072, doi: <https://doi.org/10.1175/2008JTECHA1052.1>, 2008.
- Friedman, J. H.: Greedy function approximation: A gradient boosting machine., *The Annals of Statistics*, 29, 1189–1232, doi: 10.1214/aos/1013203451, 2001.
- Frye, C., Rowat, C., and Feige, I.: Asymmetric shapley values: Incorporating causal knowledge into model-agnostic explainability, *Advances in Neural Information Processing Systems*, 2020-Decem, 1229–1239, doi: 10.48550/arXiv.1910.06358, 2020.
- Fuchs, J., Cermak, J., and Andersen, H.: Building a cloud in the southeast Atlantic: Understanding low-cloud controls based on satellite observations with machine learning, *Atmospheric Chemistry and Physics*, 18, 16 537–16 552, doi: 10.5194/acp-18-16537-2018, 2018.
- Gentine, P., Pritchard, M., Rasp, S., Reinaudi, G., and Yacalis, G.: Could Machine Learning Break the Convection Parameterization Deadlock?, *Geophysical Research Letters*, 45, 5742–5751, doi: <https://doi.org/10.1029/2018GL078202>, 2018.
- Gettelman, A.: Putting the clouds back in aerosol–cloud interactions, *Atmospheric Chemistry and Physics*, 15, 12 397–12 411, doi: 10.5194/acp-15-12397-2015, 2015.

- Gettelman, A., Morrison, H., Terai, C. R., and Wood, R.: Microphysical process rates and global aerosol–cloud interactions, *Atmospheric Chemistry and Physics*, 13, 9855–9867, doi: 10.5194/acp-13-9855-2013, 2013.
- Ghan, S., Wang, M., Zhang, S., Ferrachat, S., Gettelman, A., Griesfeller, J., Kipling, Z., Lohmann, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., and Zhang, K.: Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability, *Proceedings of the National Academy of Sciences*, 113, 5804–5811, doi: 10.1073/PNAS.1514036113, 2016.
- Ghan, S. J.: Technical note: Estimating aerosol effects on cloud radiative forcing, *Atmospheric Chemistry and Physics*, 13, 9971–9974, doi: 10.5194/acp-13-9971-2013, 2013.
- Ghan, S. J., Guzman, G., and Abdul-Razzak, H.: Competition between sea salt and sulfate particles as cloud condensation nuclei, *Journal of the Atmospheric Sciences*, 55, 3340–3347, doi: 10.1175/1520-0469(1998)055<3340:CBSSAS>2.0.CO;2, 1998.
- Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Liu, X., Ovchinnikov, M., Shipway, B., Meskhidze, N., Xu, J., and Shi, X.: Droplet nucleation: Physically-based parameterizations and comparative evaluation, *Journal of Advances in Modeling Earth Systems*, 3, 1–34, doi: 10.1029/2011ms000074, 2011.
- Giorgetta, M. A., Brokopf, R., Crueger, T., Esch, M., Fiedler, S., Helmert, J., Hohenegger, C., Kornbluh, L., Köhler, M., Manzini, E., Mauritsen, T., Nam, C., Raddatz, T., Rast, S., Reinert, D., Sakradzija, M., Schmidt, H., Schneck, R., Schnur, R., Silvers, L., Wan, H., Zängl, G., and Stevens, B.: ICON-A, the Atmosphere Component of the ICON Earth System Model: I. Model Description, *Journal of Advances in Modeling Earth Systems*, 10, 1613–1637, doi: <https://doi.org/10.1029/2017MS001242>, 2018.
- Giorgetta, M. A., Sawyer, W., Lapillonne, X., Adamidis, P., Alexeev, D., Clément, V., Dietlicher, R., Engels, J. F., Esch, M., Franke, H., Frauen, C., Hannah, W. M., Hillman, B. R., Kornbluh, L., Marti, P., Norman, M. R., Pincus, R., Rast, S., Reinert, D., Schnur, R., Schulzweida, U., and Stevens, B.: The ICON-A model for direct QBO simulations on GPUs (version icon-cscs:baf28a514), *Geoscientific Model Development*, 15, 6985–7016, doi: 10.5194/gmd-15-6985-2022, 2022.
- Goodfellow, I., Bengio, Y., and Courville, A.: *Deep Learning*, MIT Press, 2016.

- Gordon, H., Glassmeier, F., and T. McCoy, D.: An Overview of Aerosol-Cloud Interactions, in: *Clouds and Their Climatic Impacts*, chap. 2, pp. 13–45, John Wiley & Sons, Ltd, doi: <https://doi.org/10.1002/9781119700357.ch2>, 2023.
- Goren, T., Kazil, J., Hoffmann, F., Yamaguchi, T., and Feingold, G.: Anthropogenic Air Pollution Delays Marine Stratocumulus Breakup to Open Cells, *Geophysical Research Letters*, 46, 14 135–14 144, doi: <https://doi.org/10.1029/2019GL085412>, 2019.
- Goren, T., Feingold, G., Gryspeerdt, E., Kazil, J., Kretzschmar, J., Jia, H., and Quaas, J.: Projecting Stratocumulus Transitions on the Albedo—Cloud Fraction Relationship Reveals Linearity of Albedo to Droplet Concentrations, *Geophysical Research Letters*, 49, e2022GL101 169, doi: <https://doi.org/10.1029/2022GL101169>, 2022.
- Grandey, B. S. and Stier, P.: A critical look at spatial scale choices in satellite-based aerosol indirect effect studies, *Atmospheric Chemistry and Physics*, 10, 11 459–11 470, doi: [10.5194/acp-10-11459-2010](https://doi.org/10.5194/acp-10-11459-2010), 2010.
- Grise, K. M. and Kelleher, M. K.: Midlatitude Cloud Radiative Effect Sensitivity to Cloud Controlling Factors in Observations and Models: Relationship with Southern Hemisphere Jet Shifts and Climate Sensitivity, *Journal of Climate*, 34, 5869–5886, doi: <https://doi.org/10.1175/JCLI-D-20-0986.1>, 2021.
- Grise, K. M. and Medeiros, B.: Understanding the Varied Influence of Midlatitude Jet Position on Clouds and Cloud Radiative Effects in Observations and Global Climate Models, *Journal of Climate*, 29, 9005–9025, doi: <https://doi.org/10.1175/JCLI-D-16-0295.1>, 2016.
- Grosvenor, D. P., Sourdeval, O., Zuidema, P., Ackerman, A., Alexandrov, M. D., Ben-nartz, R., Boers, R., Cairns, B., Chiu, J. C., Christensen, M., Deneke, H., Diamond, M., Feingold, G., Fridlind, A., Hünerbein, A., Knist, C., Kollias, P., Marshak, A., McCoy, D., Merk, D., Painemal, D., Rausch, J., Rosenfeld, D., Russchenberg, H., Seifert, P., Sinclair, K., Stier, P., van Diedenhoven, B., Wendisch, M., Werner, F., Wood, R., Zhang, Z., and Quaas, J.: Remote Sensing of Droplet Number Concentration in Warm Clouds: A Review of the Current State of Knowledge and Perspectives, *Reviews of Geophysics*, 56, 409–453, doi: [10.1029/2017RG000593](https://doi.org/10.1029/2017RG000593), 2018.
- Gryspeerdt, E. and Stier, P.: Regime-based analysis of aerosol-cloud interactions, *Geophysical Research Letters*, 39, 1–5, doi: [10.1029/2012GL053221](https://doi.org/10.1029/2012GL053221), 2012.

- Gryspeerdt, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, *Journal of Geophysical Research*, 121, 3566–3583, doi: 10.1002/2015JD023744, 2016.
- Gryspeerdt, E., Quaas, J., Ferrachat, S., Gettelman, A., Ghan, S., Lohman, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M., and Zhang, K.: Constraining the instantaneous aerosol influence on cloud albedo, *Proceedings of the National Academy of Sciences of the United States of America*, 114, 4899–4904, doi: 10.1073/pnas.1617765114, 2017.
- Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid water path, *Atmospheric Chemistry and Physics*, 19, 5331–5347, doi: 10.5194/acp-19-5331-2019, 2019.
- Gryspeerdt, E., Mülmenstädt, J., Gettelman, A., Malavelle, F. F., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M., and Zhang, K.: Surprising similarities in model and observational aerosol radiative forcing estimates, *Atmospheric Chemistry and Physics*, 20, 613–623, doi: 10.5194/acp-20-613-2020, 2020.
- Gryspeerdt, E., McCoy, D. T., Crosbie, E., Moore, R. H., Nott, G. J., Painemal, D., Small-Griswold, J., Sorooshian, A., and Ziemba, L.: The impact of sampling strategy on the cloud droplet number concentration estimated from satellite data, *Atmospheric Measurement Techniques*, 15, 3875–3892, doi: 10.5194/amt-15-3875-2022, 2022.
- Guidotti, R., Monreale, A., Ruggieri, S., Turini, F., Giannotti, F., and Pedreschi, D.: A Survey of Methods for Explaining Black Box Models, *ACM Comput. Surv.*, 51, doi: 10.1145/3236009, 2018.
- Hamby, D. M.: A review of techniques for parameter sensitivity analysis of environmental models, *Environmental Monitoring and Assessment*, 32, 135–154, doi: 10.1007/BF00547132, 1994.
- Hartmann, D. L., Ockert-Bell, M. E., and Michelsen, M. L.: The Effect of Cloud Type on Earth’s Energy Balance: Global Analysis, *Journal of Climate*, 5, 1281–1304, doi: 10.1175/1520-0442(1992)005<1281:TEOCTO>2.0.CO;2, 1992.
- Hastie, T., Tibshirani, R., and Friedman, J.: *The Elements of Statistical Learning: Data Mining, Inference, and Prediction*, Springer New York, New York, NY, 2 edn., doi: 10.1007/978-0-387-84858-7, 2009.

- Held, I. M. and Soden, B. J.: Robust Responses of the Hydrological Cycle to Global Warming, *Journal of Climate*, 19, 5686–5699, doi: <https://doi.org/10.1175/JCLI3990.1>, 2006.
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 146, 1999–2049, doi: <https://doi.org/10.1002/qj.3803>, 2020.
- Heskes, T., Sijben, E., Bucur, I. G., and Claassen, T.: Causal Shapley Values: Exploiting Causal Knowledge to Explain Individual Predictions of Complex Models, 2020.
- Holmlund, K., Grandell, J., Schmetz, J., Stuhlmann, R., Bojkov, B., Munro, R., Lekouara, M., Coppens, D., Viticchie, B., August, T., Theodore, B., Watts, P., Dobber, M., Fowler, G., Bojinski, S., Schmid, A., Salonen, K., Tjemkes, S., Aminou, D., and Blythe, P.: Meteosat Third Generation (MTG): Continuation and Innovation of Observations from Geostationary Orbit, *Bulletin of the American Meteorological Society*, 102, E990–E1015, doi: <https://doi.org/10.1175/BAMS-D-19-0304.1>, 2021.
- Hoose, C., Kristjánsson, J. E., Iversen, T., Kirkevåg, A., Seland, Ø., and Gettelman, A.: Constraining cloud droplet number concentration in GCMs suppresses the aerosol indirect effect, *Geophysical Research Letters*, 36, doi: <https://doi.org/10.1029/2009GL038568>, 2009.
- Hu, S., Zhu, Y., Rosenfeld, D., Mao, F., Lu, X., Pan, Z., Zang, L., and Gong, W.: The Dependence of Ship-Polluted Marine Cloud Properties and Radiative Forcing on Background Drop Concentrations, *Journal of Geophysical Research: Atmospheres*, 126, e2020JD033852, doi: <https://doi.org/10.1029/2020JD033852>, 2021.
- Im, U., Samset, B. H., Nenes, A., Thomas, J. L., Kokkola, H., Dubovik, O., Amiridis, V., Arola, A., Bellouin, N., Benedetti, A., Bilde, M., Blichner, S., Decesari, S., Ekman, A. M. L., García-Pando, C. P., Gross, S., Gryspeerdt, E., Hasekamp,

- O., Kahn, R. A., Laakso, A., Lohmann, U., Marelle, L., Massling, A. H., Myhre, C. L., Pöhlker, M., Quaas, J., Raatikainen, T., Riipinen, I., Schmale, J., Seifert, P., Skov, H., Smith, C., Sporre, M. K., Stier, P., Storelvmo, T., Tsigaridis, K., van Dierenhoven, B., Virtanen, A., Wandinger, U., Wilcox, L. J., and Zieger, P.: Aerosol-Cloud Interactions: Overcoming a Barrier to Projecting Near-Term Climate Evolution and Risk, *AGU Advances*, 7, e2025AV001 872, doi: <https://doi.org/10.1029/2025AV001872>, 2026.
- Jia, H., Quaas, J., Gryspeerd, E., Böhm, C., and Sourdeval, O.: Addressing the difficulties in quantifying droplet number response to aerosol from satellite observations, *Atmospheric Chemistry and Physics*, 22, 7353–7372, doi: 10.5194/acp-22-7353-2022, 2022.
- Jia, Y., Andersen, H., and Cermak, J.: Analysis of the cloud fraction adjustment to aerosols and its dependence on meteorological controls using explainable machine learning, *Atmospheric Chemistry and Physics*, 24, 13 025–13 045, doi: 10.5194/acp-24-13025-2024, 2024.
- Jia, Y., Andersen, H., Neubauer, D., Lohmann, U., Hoose, C., and Cermak, J.: High climatological cloud cover limits its response to aerosols in ICON-HAM, *EGU sphere*, 2026, 1–32, doi: 10.5194/egusphere-2026-569, 2026.
- Jiang, H., Xue, H., Teller, A., Feingold, G., and Levin, Z.: Aerosol effects on the lifetime of shallow cumulus, *Geophysical Research Letters*, 33, doi: <https://doi.org/10.1029/2006GL026024>, 2006.
- Jiang, S., Sweet, L.-b., Blougouras, G., Brenning, A., Li, W., Reichstein, M., Denzler, J., Shangguan, W., Yu, G., Huang, F., and Zscheischler, J.: How Interpretable Machine Learning Can Benefit Process Understanding in the Geosciences, *Earth's Future*, 12, e2024EF004 540, doi: <https://doi.org/10.1029/2024EF004540>, 2024.
- Jing, X. and Suzuki, K.: The Impact of Process-Based Warm Rain Constraints on the Aerosol Indirect Effect, *Geophysical Research Letters*, 45, 710–729, doi: <https://doi.org/10.1029/2018GL079956>, 2018.
- Jones, A., Roberts, D. L., and Slingo, A.: A climate model study of indirect radiative forcing by anthropogenic sulphate aerosols, *Nature*, 370, 450–453, doi: 10.1038/370450a0, 1994.
- Kapoor, S., Cantrell, E., Peng, K., Pham, T. H., Bail, C. A., Gundersen, O. E., Hoffman, J. M., Hullman, J., Lones, M. A., Malik, M. M., Nanayakkara, P., Poldrack,

- R. A., Raji, I. D., Roberts, M., Salganik, M. J., Serra-Garcia, M., Stewart, B. M., Vandewiele, G., and Narayanan, A.: REFORMS: Reporting Standards for Machine Learning Based Science, URL <https://arxiv.org/abs/2308.07832>, 2023.
- Karniadakis, G. E., Kevrekidis, I. G., Lu, L., Perdikaris, P., Wang, S., and Yang, L.: Physics-informed machine learning, *Nature Reviews Physics*, 3, 422–440, doi: 10.1038/s42254-021-00314-5, 2021.
- Karpatne, A., Ebert-Uphoff, I., Ravela, S., Babaie, H. A., and Kumar, V.: Machine Learning for the Geosciences: Challenges and Opportunities, URL <https://arxiv.org/abs/1711.04708>, 2017.
- Karpatne, A., Ebert-Uphoff, I., Ravela, S., Babaie, H. A., and Kumar, V.: Machine Learning for the Geosciences: Challenges and Opportunities, *IEEE Transactions on Knowledge and Data Engineering*, 31, 1544–1554, doi: 10.1109/TKDE.2018.2861006, 2019.
- Kazil, J., Feingold, G., Wang, H., and Yamaguchi, T.: On the interaction between marine boundary layer cellular cloudiness and surface heat fluxes, *Atmospheric Chemistry and Physics*, 14, 61–79, doi: 10.5194/acp-14-61-2014, 2014.
- Kelleher, M. K. and Grise, K. M.: Examining Southern Ocean Cloud Controlling Factors on Daily Time Scales and Their Connections to Midlatitude Weather Systems, *Journal of Climate*, 32, 5145–5160, doi: <https://doi.org/10.1175/JCLI-D-18-0840.1>, 2019.
- Khain, A. P., Beheng, K. D., Heymsfield, A., Korolev, A., Krichak, S. O., Levin, Z., Pinsky, M., Phillips, V., Prabhakaran, T., Teller, A., van den Heever, S. C., and Yano, J.-I.: Representation of microphysical processes in cloud-resolving models: Spectral (bin) microphysics versus bulk parameterization, *Reviews of Geophysics*, 53, 247–322, doi: <https://doi.org/10.1002/2014RG000468>, 2015.
- Khairoutdinov, M. and Kogan, Y.: A New Cloud Physics Parameterization in a Large-Eddy Simulation Model of Marine Stratocumulus, *Monthly Weather Review*, 128, 229–243, doi: [https://doi.org/10.1175/1520-0493\(2000\)128<0229:ANCPPI>2.0.CO;2](https://doi.org/10.1175/1520-0493(2000)128<0229:ANCPPI>2.0.CO;2), 2000.
- Kidder, S. Q. and Haar, T. H. V.: Meteorological Satellite Instrumentation, in: *Satellite Meteorology*, pp. 87–144, Academic Press, doi: 10.1016/b978-0-08-057200-0.50008-0, 1995.

- Kim, J., Kim, H., Kim, H., Lee, D., and Yoon, S.: A comprehensive survey of deep learning for time series forecasting: architectural diversity and open challenges, *Artificial Intelligence Review*, 58, 216, doi: 10.1007/s10462-025-11223-9, 2025.
- Kim, M., Brunner, D., and Kuhlmann, G.: Importance of satellite observations for high-resolution mapping of near-surface NO₂ by machine learning, *Remote Sensing of Environment*, 264, 112 573, doi: 10.1016/j.rse.2021.112573, 2021.
- King, M. D., Menzel, W. P., Kaufman, Y. J., Tanre, D., Gao, B.-C., Platnick, S., Ackerman, S. A., Remer, L. A., Pincus, R., and Hubanks, P. A.: Cloud and aerosol properties, precipitable water, and profiles of temperature and water vapor from MODIS, *IEEE Transactions on Geoscience and Remote Sensing*, 41, 442–458, doi: 10.1109/TGRS.2002.808226, 2003.
- Kingma, D. P. and Welling, M.: Auto-Encoding Variational Bayes, arXiv preprint arXiv:1312.6114, URL <https://arxiv.org/abs/1312.6114>, 2013.
- Kirkevåg, A., Iversen, T., Seland, Ø., Debernard, J. B., Storelvmo, T., and Kristjánsson, J. E.: Aerosol-cloud-climate interactions in the climate model CAM-Oslo, *Tellus A: Dynamic Meteorology and Oceanography*, 60, 492, doi: 10.1111/j.1600-0870.2007.00313.x, 2008.
- Klein, S. A. and Hartmann, D. L.: The Seasonal Cycle of Low Stratiform Clouds, *Journal of Climate*, 6, 1587–1606, doi: [https://doi.org/10.1175/1520-0442\(1993\)006<1587:TSCOLS>2.0.CO;2](https://doi.org/10.1175/1520-0442(1993)006<1587:TSCOLS>2.0.CO;2), 1993.
- Klein, S. A., Hall, A., Norris, J. R., and Pincus, R.: Low-Cloud Feedbacks from Cloud-Controlling Factors: A Review, *Surveys in Geophysics*, 38, 1307–1329, doi: 10.1007/s10712-017-9433-3, 2017.
- Köhler, H.: The nucleus in and the growth of hygroscopic droplets, *Transactions of the Faraday Society*, 32, 1152–1161, doi: 10.1039/TF9363201152, 1936.
- Kooperman, G. J., Pritchard, M. S., Ghan, S. J., Wang, M., Somerville, R. C. J., and Russell, L. M.: Constraining the influence of natural variability to improve estimates of global aerosol indirect effects in a nudged version of the Community Atmosphere Model 5, *Journal of Geophysical Research: Atmospheres*, 117, doi: <https://doi.org/10.1029/2012JD018588>, 2012.
- Kreidenweis, S. M., Petters, M., and Lohmann, U.: 100 Years of Progress in Cloud Physics, Aerosols, and Aerosol Chemistry Research, *Meteorological*

- Monographs, 59, 1–11, doi: <https://doi.org/10.1175/AMSMONOGRAPHS-D-18-0024.1>, 2019.
- Lai, C.-Y., Hassanzadeh, P., Sheshadri, A., Sonnewald, M., Ferrari, R., and Balaji, V.: Machine Learning for Climate Physics and Simulations, *Annual Review of Condensed Matter Physics*, 16, 343–365, doi: <https://doi.org/10.1146/annurev-conmatphys-043024-114758>, 2025.
- Lam, R., Sanchez-Gonzalez, A., Willson, M., Wirnsberger, P., Fortunato, M., Alet, F., Ravuri, S., Ewalds, T., Eaton-Rosen, Z., Hu, W., Merose, A., Hoyer, S., Holland, G., Vinyals, O., Stott, J., Pritzel, A., Mohamed, S., and Battaglia, P.: Learning skillful medium-range global weather forecasting, *Science*, 382, 1416–1421, doi: [10.1126/science.adi2336](https://doi.org/10.1126/science.adi2336), 2023.
- Leahy, L. V., Wood, R., Charlson, R. J., Hostetler, C. A., Rogers, R. R., Vaughan, M. A., and Winker, D. M.: On the nature and extent of optically thin marine low clouds, *Journal of Geophysical Research: Atmospheres*, 117, doi: <https://doi.org/10.1029/2012JD017929>, 2012.
- LeCun, Y., Bengio, Y., and Hinton, G.: Deep learning, *Nature*, 521, 436–444, doi: [10.1038/nature14539](https://doi.org/10.1038/nature14539), 2015.
- Letu, H., Shi, J., Li, M., Wang, T., Shang, H., Lei, Y., Ji, D., Wen, J., Yang, K., and Chen, L.: A review of the estimation of downward surface shortwave radiation based on satellite data: Methods, progress and problems, *Science China Earth Sciences*, 63, 774–789, doi: [10.1007/s11430-019-9589-0](https://doi.org/10.1007/s11430-019-9589-0), 2020.
- Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The Collection 6 MODIS aerosol products over land and ocean, *Atmospheric Measurement Techniques*, 6, 2989–3034, doi: [10.5194/amt-6-2989-2013](https://doi.org/10.5194/amt-6-2989-2013), 2013.
- Li, J., Carlson, B. E., Yung, Y. L., Lv, D., Hansen, J., Penner, J. E., Liao, H., Ramaswamy, V., Kahn, R. A., Zhang, P., Dubovik, O., Ding, A., Lacis, A. A., Zhang, L., and Dong, Y.: Scattering and absorbing aerosols in the climate system, *Nature Reviews Earth & Environment*, 3, 363–379, doi: [10.1038/s43017-022-00296-7](https://doi.org/10.1038/s43017-022-00296-7), 2022a.
- Li, T., Yu, Y., Wang, X., Liu, X., Mao, Q., and Yuan, Y.: A review of aerosol-cloud interactions: Mechanisms, climate effects, and observation methods, *Atmospheric Research*, 325, 108 267, doi: <https://doi.org/10.1016/j.atmosres.2025.108267>, 2025.

- Li, W., Migliavacca, M., Forkel, M., Denissen, J. M. C., Reichstein, M., Yang, H., Duveiller, G., Weber, U., and Orth, R.: Widespread increasing vegetation sensitivity to soil moisture, *Nature Communications*, 13, 3959, doi: 10.1038/s41467-022-31667-9, 2022b.
- Linardatos, P., Papastefanopoulos, V., and Kotsiantis, S.: Explainable AI: A Review of Machine Learning Interpretability Methods, *Entropy*, 23, doi: 10.3390/e23010018, 2021.
- Liu, Y., Lin, T., Zhang, J., Wang, F., Huang, Y., Wu, X., Ye, H., Zhang, G., Cao, X., and de Leeuw, G.: Opposite effects of aerosols and meteorological parameters on warm clouds in two contrasting regions over eastern China, *Atmospheric Chemistry and Physics*, 24, 4651–4673, doi: 10.5194/acp-24-4651-2024, 2024.
- Loeb, N. G. and Schuster, G. L.: An observational study of the relationship between cloud, aerosol and meteorology in broken low-level cloud conditions, *Journal of Geophysical Research: Atmospheres*, 113, doi: <https://doi.org/10.1029/2007JD009763>, 2008.
- Loeb, N. G., Johnson, G. C., Thorsen, T. J., Lyman, J. M., Rose, F. G., and Kato, S.: Satellite and Ocean Data Reveal Marked Increase in Earth’s Heating Rate, *Geophysical Research Letters*, 48, e2021GL093047, doi: <https://doi.org/10.1029/2021GL093047>, 2021.
- Lohmann, U.: Marine Boundary Layer Clouds, in: *Surface Ocean–Lower Atmosphere Processes*, pp. 57–68, American Geophysical Union (AGU), doi: <https://doi.org/10.1029/2008GM000761>, 2009.
- Lohmann, U. and Feichter, J.: Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity study with the ECHAM4 GCM, *Journal of Geophysical Research: Atmospheres*, 102, 13 685–13 700, doi: <https://doi.org/10.1029/97JD00631>, 1997.
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmospheric Chemistry and Physics*, 5, 715–737, doi: 10.5194/acp-5-715-2005, 2005.
- Lohmann, U. and Hoose, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, *Atmospheric Chemistry and Physics*, 9, 8917–8934, doi: 10.5194/acp-9-8917-2009, 2009.
- Lohmann, U. and Neubauer, D.: The importance of mixed-phase and ice clouds for climate sensitivity in the global aerosol–climate model ECHAM6-HAM2,

- Atmospheric Chemistry and Physics, 18, 8807–8828, doi: 10.5194/acp-18-8807-2018, 2018.
- Lohmann, U. and Roeckner, E.: Design and performance of a new cloud microphysics scheme developed for the ECHAM general circulation model, *Climate Dynamics*, 12, 557–572, doi: 10.1007/BF00207939, 1996.
- Lohmann, U., Feichter, J., Chuang, C. C., and Penner, J. E.: Prediction of the number of cloud droplets in the ECHAM GCM, *Journal of Geophysical Research: Atmospheres*, 104, 9169–9198, doi: <https://doi.org/10.1029/1999JD900046>, 1999.
- Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, *Atmospheric Chemistry and Physics*, 7, 3425–3446, doi: 10.5194/acp-7-3425-2007, 2007.
- Lohmann, U., Lüönd, F., and Mahrt, F.: *An Introduction to Clouds: From the Microscale to Climate*, Cambridge University Press, Cambridge, doi: DOI:10.1017/CBO9781139087513, 2016.
- Lundberg, S. M. and Lee, S. I.: A unified approach to interpreting model predictions, *Advances in Neural Information Processing Systems*, 2017-Decem, 4766–4775, 2017.
- Lundberg, S. M., Erion, G. G., and Lee, S.-I.: Consistent Individualized Feature Attribution for Tree Ensembles, *ArXiv*, abs/1802.0, URL <http://arxiv.org/abs/1802.03888>, 2018.
- Lundberg, S. M., Erion, G., Chen, H., DeGrave, A., Prutkin, J. M., Nair, B., Katz, R., Himmelfarb, J., Bansal, N., and Lee, S.-I.: From local explanations to global understanding with explainable AI for trees, *Nature Machine Intelligence*, 2, 56–67, doi: 10.1038/s42256-019-0138-9, 2020.
- L’Ecuyer, T. S., Hang, Y., Matus, A. V., and Wang, Z.: Reassessing the Effect of Cloud Type on Earth’s Energy Balance in the Age of Active Spaceborne Observations. Part I: Top of Atmosphere and Surface, *Journal of Climate*, 32, 6197–6217, doi: <https://doi.org/10.1175/JCLI-D-18-0753.1>, 2019.
- Maddux, B. C., Ackerman, S. A., and Platnick, S.: Viewing Geometry Dependencies in MODIS Cloud Products, *Journal of Atmospheric and Oceanic Technology*, 27, 1519–1528, doi: <https://doi.org/10.1175/2010JTECHA1432.1>, 2010.

- Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S., Allan, R. P., Karset, I. H. H., Kristjánsson, J. E., Oreopoulos, L., Cho, N., Lee, D., Bellouin, N., Boucher, O., Grosvenor, D. P., Carslaw, K. S., Dhomse, S., Mann, G. W., Schmidt, A., Coe, H., Hartley, M. E., Dalvi, M., Hill, A. A., Johnson, B. T., Johnson, C. E., Knight, J. R., O'Connor, F. M., Partridge, D. G., Stier, P., Myhre, G., Platnick, S., Stephens, G. L., Takahashi, H., and Thordarson, T.: Strong constraints on aerosol–cloud interactions from volcanic eruptions, *Nature*, 546, 485–491, doi: 10.1038/nature22974, 2017.
- Marais, W. J., Holz, R. E., Reid, J. S., and Willett, R. M.: Leveraging spatial textures, through machine learning, to identify aerosols and distinct cloud types from multispectral observations, *Atmospheric Measurement Techniques*, 13, 5459–5480, doi: 10.5194/amt-13-5459-2020, 2020.
- Marchant, B., Platnick, S., Meyer, K., and Wind, G.: Evaluation of the MODIS Collection 6 multilayer cloud detection algorithm through comparisons with CloudSat Cloud Profiling Radar and CALIPSO CALIOP products, *Atmospheric Measurement Techniques*, 13, 3263–3275, doi: 10.5194/amt-13-3263-2020, 2020.
- Mason, B. J.: The Nucleation and Growth of Ice Crystals, in: *Physics of Precipitation: Proceedings of the Cloud Physics Conference*, Woods Hole, Massachusetts, June 3–5, 1959, Geophysical Monograph Series, pp. 226–232, American Geophysical Union (AGU), doi: <https://doi.org/10.1029/GM005p0226>, 1960.
- Mauritsen, T., Bader, J., Becker, T., Behrens, J., Bittner, M., Brokopf, R., Brovkin, V., Claussen, M., Crueger, T., Esch, M., Fast, I., Fiedler, S., Fläschner, D., Gayler, V., Giorgetta, M., Goll, D. S., Haak, H., Hagemann, S., Hedemann, C., Hohenegger, C., Ilyina, T., Jahns, T., Jimenéz-de-la Cuesta, D., Jungclaus, J., Kleinen, T., Kloster, S., Kracher, D., Kinne, S., Kleberg, D., Lasslop, G., Kornbluh, L., Marotzke, J., Matei, D., Meraner, K., Mikolajewicz, U., Modali, K., Möbis, B., Müller, W. A., Nabel, J. E., Nam, C. C., Notz, D., Nyawira, S. S., Paulsen, H., Peters, K., Pincus, R., Pohlmann, H., Pongratz, J., Popp, M., Raddatz, T. J., Rast, S., Redler, R., Reick, C. H., Rohrschneider, T., Schemann, V., Schmidt, H., Schnur, R., Schulzweida, U., Six, K. D., Stein, L., Stemmler, I., Stevens, B., von Storch, J. S., Tian, F., Voigt, A., Vrese, P., Wieners, K. H., Wilkenskjaeld, S., Winkler, A., and Roeckner, E.: Developments in the MPI-M Earth System Model version 1.2 (MPI-ESM1.2) and Its Response to Increasing CO₂, *Journal of Advances in Modeling Earth Systems*, 11, 998–1038, doi: 10.1029/2018MS001400, 2019.
- McCoy, D. T., Bender, F. A.-M., Mohrmann, J. K. C., Hartmann, D. L., Wood, R., and

- Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA, and AeroCom, *Journal of Geophysical Research: Atmospheres*, 122, 1779–1796, doi: <https://doi.org/10.1002/2016JD026141>, 2017a.
- McCoy, D. T., Eastman, R., Hartmann, D. L., and Wood, R.: The Change in Low Cloud Cover in a Warmed Climate Inferred from AIRS, MODIS, and ERA-Interim, *Journal of Climate*, 30, 3609–3620, doi: <https://doi.org/10.1175/JCLI-D-15-0734.1>, 2017b.
- McGovern, A., Lagerquist, R., Gagne, D. J., Jergensen, G. E., Elmore, K. L., Homeyer, C. R., and Smith, T.: Making the black box more transparent: Understanding the physical implications of machine learning, *Bulletin of the American Meteorological Society*, 100, 2175–2199, doi: 10.1175/BAMS-D-18-0195.1, 2019.
- Mellado, J. P.: Cloud-Top Entrainment in Stratocumulus Clouds, *Annual Review of Fluid Mechanics*, 49, 145–169, doi: <https://doi.org/10.1146/annurev-fluid-010816-060231>, 2017.
- Menzel, W. P., Frey, R. A., Zhang, H., Wylie, D. P., Moeller, C. C., Holz, R. E., Maddux, B., Baum, B. A., Strabala, K. I., and Gumley, L. E.: MODIS Global Cloud-Top Pressure and Amount Estimation: Algorithm Description and Results, *Journal of Applied Meteorology and Climatology*, 47, 1175–1198, doi: <https://doi.org/10.1175/2007JAMC1705.1>, 2008.
- Merk, D., Deneke, H., Pospichal, B., and Seifert, P.: Investigation of the adiabatic assumption for estimating cloud micro- and macrophysical properties from satellite and ground observations, *Atmospheric Chemistry and Physics*, 16, 933–952, doi: 10.5194/acp-16-933-2016, 2016.
- Michibata, T. and Takemura, T.: Evaluation of autoconversion schemes in a single model framework with satellite observations, *Journal of Geophysical Research*, 120, 9570–9590, doi: 10.1002/2015JD023818, 2015.
- Michibata, T., Suzuki, K., Sato, Y., and Takemura, T.: The source of discrepancies in aerosol–cloud–precipitation interactions between GCM and A-Train retrievals, *Atmospheric Chemistry and Physics*, 16, 15 413–15 424, doi: 10.5194/acp-16-15413-2016, 2016.
- Michibata, T., Wall, C. J., Hirota, N., Duran, B. M., and Nozawa, T.: Recent Advances in the Observation and Modeling of Aerosol-Cloud Interactions, Cloud Feedbacks, and Earth’s Energy Imbalance: A Review, *Current Pollution Reports*, 11, 50, doi: 10.1007/s40726-025-00382-6, 2025.

- Min, M., Li, J., Wang, F., Liu, Z., and Menzel, W. P.: Retrieval of cloud top properties from advanced geostationary satellite imager measurements based on machine learning algorithms, *Remote Sensing of Environment*, 239, 111 616, doi: <https://doi.org/10.1016/j.rse.2019.111616>, 2020.
- Minnett, P. J., Alvera-Azcárate, A., Chin, T. M., Corlett, G. K., Gentemann, C. L., Karagali, I., Li, X., Marsouin, A., Marullo, S., Maturi, E., Santoleri, R., Saux Picart, S., Steele, M., and Vazquez-Cuervo, J.: Half a century of satellite remote sensing of sea-surface temperature, *Remote Sensing of Environment*, 233, 111 366, doi: <https://doi.org/10.1016/j.rse.2019.111366>, 2019.
- Miyamoto, A., Nakamura, H., and Miyasaka, T.: Influence of the Subtropical High and Storm Track on Low-Cloud Fraction and Its Seasonality over the South Indian Ocean, *Journal of Climate*, 31, 4017–4039, doi: <https://doi.org/10.1175/JCLI-D-17-0229.1>, 2018.
- Molina, M. J., O'Brien, T. A., Anderson, G., Ashfaq, M., Bennett, K. E., Collins, W. D., Dagon, K., Restrepo, J. M., and Ullrich, P. A.: A Review of Recent and Emerging Machine Learning Applications for Climate Variability and Weather Phenomena, *Artificial Intelligence for the Earth Systems*, 2, 220 086, doi: <https://doi.org/10.1175/AIES-D-22-0086.1>, 2023.
- Molnar, C.: *Interpretable Machine Learning*, Christoph Molnar, 3 edn., URL <https://christophm.github.io/interpretable-ml-book>, 2025.
- Morrison, H. and Gettelman, A.: A New Two-Moment Bulk Stratiform Cloud Microphysics Scheme in the Community Atmosphere Model, Version 3 (CAM3). Part I: Description and Numerical Tests, *Journal of Climate*, 21, 3642–3659, doi: <https://doi.org/10.1175/2008JCLI2105.1>, 2008.
- Morrison, H., Curry, J. A., and Khvorostyanov, V. I.: A New Double-Moment Microphysics Parameterization for Application in Cloud and Climate Models. Part I: Description, *Journal of the Atmospheric Sciences*, 62, 1665–1677, doi: <https://doi.org/10.1175/JAS3446.1>, 2005.
- Morrison, H., van Lier-Walqui, M., Fridlind, A. M., Grabowski, W. W., Harrington, J. Y., Hoose, C., Korolev, A., Kumjian, M. R., Milbrandt, J. A., Pawlowska, H., Posselt, D. J., Prat, O. P., Reimel, K. J., Shima, S.-I., van Didenhoven, B., and Xue, L.: Confronting the Challenge of Modeling Cloud and Precipitation Microphysics, *Journal of Advances in Modeling Earth Systems*, 12, e2019MS001 689, doi: <https://doi.org/10.1029/2019MS001689>, 2020.

- Muench, S. and Lohmann, U.: Developing a Cloud Scheme With Prognostic Cloud Fraction and Two Moment Microphysics for ECHAM-HAM, *Journal of Advances in Modeling Earth Systems*, 12, e2019MS001 824, doi: <https://doi.org/10.1029/2019MS001824>, 2020.
- Mülmenstädt, J. and Feingold, G.: The Radiative Forcing of Aerosol–Cloud Interactions in Liquid Clouds: Wrestling and Embracing Uncertainty, *Current Climate Change Reports*, 4, 23–40, doi: 10.1007/s40641-018-0089-y, 2018.
- Myers, T. A. and Norris, J. R.: Observational Evidence That Enhanced Subsidence Reduces Subtropical Marine Boundary Layer Cloudiness, *Journal of Climate*, 26, 7507–7524, doi: <https://doi.org/10.1175/JCLI-D-12-00736.1>, 2013.
- Myers, T. A. and Norris, J. R.: On the Relationships between Subtropical Clouds and Meteorology in Observations and CMIP3 and CMIP5 Models, *Journal of Climate*, 28, 2945–2967, doi: <https://doi.org/10.1175/JCLI-D-14-00475.1>, 2015.
- Myers, T. A. and Norris, J. R.: Reducing the uncertainty in subtropical cloud feedback, *Geophysical Research Letters*, 43, 2144–2148, doi: 10.1002/2015GL067416, 2016.
- Myers, T. A., Scott, R. C., Zelinka, M. D., Klein, S. A., Norris, J. R., and Caldwell, P. M.: Observational constraints on low cloud feedback reduce uncertainty of climate sensitivity, *Nature Climate Change*, 11, 501–507, doi: 10.1038/s41558-021-01039-0, 2021.
- Myers, T. A., Vogel, R., Brient, F., Parishani, H., and Scott, R. C.: Tropical Marine Low Clouds: Feedbacks to Warming and on Climate Variability, in: *Clouds and Their Climatic Impacts*, edited by S.C. Sullivan and C. Hoose, *Geophysical Monograph Series*, chap. 7, pp. 159–177, John Wiley & Sons, Inc., doi: <https://doi.org/10.1002/9781119700357.ch7>, 2023.
- Nakajima, T. and King, M. D.: Determination of the Optical Thickness and Effective Particle Radius of Clouds from Reflected Solar Radiation Measurements. Part I: Theory, *Journal of Atmospheric Sciences*, 47, 1878–1893, doi: [https://doi.org/10.1175/1520-0469\(1990\)047<1878:DOTOTA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1990)047<1878:DOTOTA>2.0.CO;2), 1990.
- Narenpitak, P., Kazil, J., Yamaguchi, T., Quinn, P., and Feingold, G.: From Sugar to Flowers: A Transition of Shallow Cumulus Organization During ATOMIC, *Journal of Advances in Modeling Earth Systems*, 13, e2021MS002 619, doi: <https://doi.org/10.1029/2021MS002619>, 2021.

- Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Stier, P., Partridge, D. G., Tegen, I., Bey, I., Stanelle, T., Kokkola, H., and Lohmann, U.: The global aerosol-climate model ECHAM6.3–HAM2.3 – Part 2: Cloud evaluation, aerosol radiative forcing, and climate sensitivity, *Geoscientific Model Development*, 12, 3609–3639, doi: 10.5194/gmd-12-3609-2019, 2019.
- Norris, J. R. and Iacobellis, S. F.: North Pacific Cloud Feedbacks Inferred from Synoptic-Scale Dynamic and Thermodynamic Relationships, *Journal of Climate*, 18, 4862–4878, doi: <https://doi.org/10.1175/JCLI3558.1>, 2005.
- Norris, J. R. and Klein, S. A.: Low Cloud Type over the Ocean from Surface Observations. Part III: Relationship to Vertical Motion and the Regional Surface Synoptic Environment, *Journal of Climate*, 13, 245–256, doi: [https://doi.org/10.1175/1520-0442\(2000\)013<0245:LCTOTO>2.0.CO;2](https://doi.org/10.1175/1520-0442(2000)013<0245:LCTOTO>2.0.CO;2), 2000.
- Nowack, P., Runge, J., Eyring, V., and Haigh, J. D.: Causal networks for climate model evaluation and constrained projections, *Nature Communications*, 11, 1415, doi: 10.1038/s41467-020-15195-y, 2020.
- Null, J.: El Niño and La Niña Years and Intensities Based on {Oceanic Niño Index (ONI)}, Golden Gate Weather Services, URL <https://ggweather.com/enso/oni.htm>, 2025.
- Padarian, J., McBratney, A. B., and Minasny, B.: Game theory interpretation of digital soil mapping convolutional neural networks, *Soil*, 6, 389–397, doi: 10.5194/soil-6-389-2020, 2020.
- Platnick, S. and Twomey, S.: Determining the Susceptibility of Cloud Albedo to Changes in Droplet Concentration with the Advanced Very High Resolution Radiometer, *Journal of Applied Meteorology and Climatology*, 33, 334–347, doi: [https://doi.org/10.1175/1520-0450\(1994\)033<0334:DTSOCA>2.0.CO;2](https://doi.org/10.1175/1520-0450(1994)033<0334:DTSOCA>2.0.CO;2), 1994.
- Platnick, S., King, M. D., Ackerman, S. A., Menzel, W. P., Baum, B. A., Riédi, J. C., and Frey, R. A.: The MODIS cloud products: Algorithms and examples from terra, *IEEE Transactions on Geoscience and Remote Sensing*, 41, 459–472, doi: 10.1109/TGRS.2002.808301, 2003.
- Platnick, S., Meyer, K. G., King, M. D., Wind, G., Amarasinghe, N., Marchant, B., Arnold, G. T., Zhang, Z., Hubanks, P. A., Holz, R. E., Yang, P., Ridgway, W. L., and Riedi, J.: The MODIS Cloud Optical and Microphysical Products: Collection 6 Updates and Examples From Terra and Aqua, *IEEE Transactions on Geoscience and Remote Sensing*, 55, 502–525, doi: 10.1109/TGRS.2016.2610522, 2017.

- Pugsley, G., Gryspeerdt, E., and Nair, V.: Cloud fraction response to aerosol driven by nighttime processes, *Proceedings of the National Academy of Sciences*, 122, e2509949 122, doi: 10.1073/pnas.2509949122, 2025.
- Qu, X., Hall, A., Klein, S. A., and Caldwell, P. M.: The strength of the tropical inversion and its response to climate change in 18 CMIP5 models, *Climate Dynamics*, 45, 375–396, doi: 10.1007/s00382-014-2441-9, 2015a.
- Qu, X., Hall, A., Klein, S. A., and Deangelis, A. M.: Positive tropical marine low-cloud cover feedback inferred from cloud-controlling factors, *Geophysical Research Letters*, 42, 7767–7775, doi: 10.1002/2015GL065627, 2015b.
- Quaas, J. and Gryspeerdt, E.: Chapter 12 - Aerosol-cloud interactions in liquid clouds, in: *Aerosols and Climate*, edited by Carslaw, K. S., pp. 489–544, Elsevier, doi: <https://doi.org/10.1016/B978-0-12-819766-0.00019-5>, 2022.
- Quaas, J., Boucher, O., and Lohmann, U.: Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data, *Atmospheric Chemistry and Physics*, 6, 947–955, doi: 10.5194/acp-6-947-2006, 2006.
- Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol climate forcing, *Journal of Geophysical Research Atmospheres*, 113, 1–9, doi: 10.1029/2007JD008962, 2008.
- Quaas, J., Arola, A., Cairns, B., Christensen, M., Deneke, H., Ekman, A. M., Feingold, G., Fridlind, A., Gryspeerdt, E., Hasekamp, O., Li, Z., Lipponen, A., Mülmenstädt, J., Nenes, A., Penner, J. E., Rosenfeld, D., Schrödner, R., Sinclair, K., Sourdeval, O., Stier, P., Tesche, M., Van Dierenhoven, B., and Wendisch, M.: Constraining the Twomey effect from satellite observations: Issues and perspectives, *Atmospheric Chemistry and Physics*, 20, 15 079–15 099, doi: 10.5194/acp-20-15079-2020, 2020.
- Quaas, J., Andrews, T., Bellouin, N., Block, K., Boucher, O., Ceppi, P., Dagan, G., Doktorowski, S., Eichholz, H. M., Forster, P., Goren, T., Gryspeerdt, E., Hodnebrog, Ø., Jia, H., Kramer, R., Lange, C., Maycock, A. C., Mülmenstädt, J., Myhre, G., O'Connor, F. M., Pincus, R., Samset, B. H., Senf, F., Shine, K. P., Smith, C., Stjern, C. W., Takemura, T., Toll, V., and Wall, C. J.: Adjustments to Climate Perturbations—Mechanisms, Implications, Observational Constraints, *AGU Advances*, 5, e2023AV001 144, doi: 10.1029/2023AV001144, 2024.
- Quinlan, J. R.: Induction of decision trees, *Machine Learning*, 1, 81–106, doi: 10.1007/BF00116251, 1986.

- Quinlan, J. R.: Simplifying decision trees, *International Journal of Man-Machine Studies*, 27, 221–234, doi: [https://doi.org/10.1016/S0020-7373\(87\)80053-6](https://doi.org/10.1016/S0020-7373(87)80053-6), 1987.
- Raissi, M., Perdikaris, P., and Karniadakis, G. E.: Physics-informed neural networks: A deep learning framework for solving forward and inverse problems involving nonlinear partial differential equations, *Journal of Computational Physics*, 378, 686–707, doi: <https://doi.org/10.1016/j.jcp.2018.10.045>, 2019.
- Ramanathan, V., Cess, R. D., Harrison, E. F., Minnis, P., Barkstrom, B. R., Ahmad, E., and Hartmann, D.: Cloud-Radiative Forcing and Climate: Results from the Earth Radiation Budget Experiment, *Science*, 243, 57–63, doi: [10.1126/science.243.4887.57](https://doi.org/10.1126/science.243.4887.57), 1989.
- Rasp, S., Pritchard, M. S., and Gentine, P.: Deep learning to represent subgrid processes in climate models, *Proceedings of the National Academy of Sciences*, 115, 9684–9689, doi: [10.1073/pnas.1810286115](https://doi.org/10.1073/pnas.1810286115), 2018.
- Rasp, S., Schulz, H., Bony, S., and Stevens, B.: Combining Crowdsourcing and Deep Learning to Explore the Mesoscale Organization of Shallow Convection, *Bulletin of the American Meteorological Society*, 101, E1980–E1995, doi: <https://doi.org/10.1175/BAMS-D-19-0324.1>, 2020.
- Ravuri, S., Lenc, K., Willson, M., Kangin, D., Lam, R., Mirowski, P., Fitzsimons, M., Athanassiadou, M., Kashem, S., Madge, S., Prudden, R., Mandhane, A., Clark, A., Brock, A., Simonyan, K., Hadsell, R., Robinson, N., Clancy, E., Arribas, A., and Mohamed, S.: Skilful precipitation nowcasting using deep generative models of radar, *Nature*, 597, 672–677, doi: [10.1038/s41586-021-03854-z](https://doi.org/10.1038/s41586-021-03854-z), 2021.
- Reichstein, M., Camps-Valls, G., Stevens, B., Jung, M., Denzler, J., Carvalhais, N., and Prabhat: Deep learning and process understanding for data-driven Earth system science, *Nature*, 566, 195–204, doi: [10.1038/s41586-019-0912-1](https://doi.org/10.1038/s41586-019-0912-1), 2019.
- Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., Andreae, M. O., and Pöschl, U.: Aerosol- and updraft-limited regimes of cloud droplet formation: Influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN), *Atmospheric Chemistry and Physics*, 9, 7067–7080, doi: [10.5194/acp-9-7067-2009](https://doi.org/10.5194/acp-9-7067-2009), 2009.
- Ribeiro, M. T., Singh, S., and Guestrin, C.: "Why Should I Trust You?": Explaining the Predictions of Any Classifier, in: *Proceedings of the 22nd ACM SIGKDD*

- International Conference on Knowledge Discovery and Data Mining, KDD '16, p. 1135–1144, Association for Computing Machinery, New York, NY, USA, doi: 10.1145/2939672.2939778, 2016.
- Rieck, M., Nuijens, L., and Stevens, B.: Marine Boundary Layer Cloud Feedbacks in a Constant Relative Humidity Atmosphere, *Journal of the Atmospheric Sciences*, 69, 2538–2550, doi: <https://doi.org/10.1175/JAS-D-11-0203.1>, 2012.
- Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., and Dümenil, L.: The atmospheric general circulation model ECHAM-4: Model description and simulation of present-day climate, Tech. Rep. 218, Max-Planck-Institut für Meteorologie, Hamburg, Germany, 1996.
- Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., de Leeuw, G., Donovan, D. P., Kahn, R., Kinne, S., Kivekäs, N., Kulmala, M., Lau, W., Schmidt, K. S., Suni, T., Wagner, T., Wild, M., and Quaas, J.: Global observations of aerosol-cloud-precipitation-climate interactions, *Reviews of Geophysics*, 52, 750–808, doi: <https://doi.org/10.1002/2013RG000441>, 2014.
- Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., and Yu, S.: Aerosol-driven droplet concentrations dominate coverage and water of oceanic low-level clouds, *Science*, 363, doi: 10.1126/science.aav0566, 2019.
- Rosenfeld, D., Kokhanovsky, A., Goren, T., Gryspeerd, E., Hasekamp, O., Jia, H., Lopatin, A., Quaas, J., Pan, Z., and Sourdeval, O.: Frontiers in Satellite-Based Estimates of Cloud-Mediated Aerosol Forcing, *Reviews of Geophysics*, 61, e2022RG000799, doi: <https://doi.org/10.1029/2022RG000799>, 2023.
- Rozendaal, M. A., Leovy, C. B., and Klein, S. A.: An Observational Study of Diurnal Variations of Marine Stratiform Cloud, *Journal of Climate*, 8, 1795–1809, doi: [https://doi.org/10.1175/1520-0442\(1995\)008<1795:AOSODV>2.0.CO;2](https://doi.org/10.1175/1520-0442(1995)008<1795:AOSODV>2.0.CO;2), 1995.
- Runge, J., Bathiany, S., Bollt, E., Camps-Valls, G., Coumou, D., Deyle, E., Glymour, C., Kretschmer, M., Mahecha, M. D., Muñoz-Marí, J., van Nes, E. H., Peters, J., Quax, R., Reichstein, M., Scheffer, M., Schölkopf, B., Spirtes, P., Sugihara, G., Sun, J., Zhang, K., and Zscheischler, J.: Inferring causation from time series in Earth system sciences, *Nature Communications*, 10, 1–13, doi: 10.1038/s41467-019-10105-3, 2019.
- Saffin, L., Lock, A., Tomassini, L., Blyth, A., Böing, S., Denby, L., and Marsham, J.: Kilometer-Scale Simulations of Trade-Wind Cumulus Capture Processes of

- Mesoscale Organization, *Journal of Advances in Modeling Earth Systems*, 15, e2022MS003 295, doi: <https://doi.org/10.1029/2022MS003295>, 2023.
- Salzmann, M., Ferrachat, S., Tully, C., Münch, S., Watson-Parris, D., Neubauer, D., Siegenthaler-Le Drian, C., Rast, S., Heinold, B., Crueger, T., Brokopf, R., Mülmenstädt, J., Quaas, J., Wan, H., Zhang, K., Lohmann, U., Stier, P., and Tegen, I.: The Global Atmosphere-aerosol Model ICON-A-HAM2.3–Initial Model Evaluation and Effects of Radiation Balance Tuning on Aerosol Optical Thickness, *Journal of Advances in Modeling Earth Systems*, 14, e2021MS002 699, doi: <https://doi.org/10.1029/2021MS002699>, 2022.
- Sandu, I. and Stevens, B.: On the Factors Modulating the Stratocumulus to Cumulus Transitions, *Journal of the Atmospheric Sciences*, 68, 1865–1881, doi: <https://doi.org/10.1175/2011JAS3614.1>, 2011.
- Schiller, J., Stiller, S., and Ryo, M.: Artificial intelligence in environmental and Earth system sciences: explainability and trustworthiness, *Artificial Intelligence Review*, 58, 316, doi: 10.1007/s10462-025-11165-2, 2025.
- Schwarz, K., Cermak, J., Fuchs, J., and Andersen, H.: Mapping the Twilight Zone—What We Are Missing between Clouds and Aerosols, *Remote Sensing*, 9, doi: 10.3390/rs9060577, 2017.
- Scott, R. C., Myers, T. A., Norris, J. R., Zelinka, M. D., Klein, S. A., Sun, M., and Doelling, D. R.: Observed Sensitivity of Low-Cloud Radiative Effects to Meteorological Perturbations over the Global Oceans, *Journal of Climate*, 33, 7717–7734, doi: <https://doi.org/10.1175/JCLI-D-19-1028.1>, 2020.
- Seifert, A. and Beheng, K. D.: A two-moment cloud microphysics parameterization for mixed-phase clouds. Part 1: Model description, *Meteorology and Atmospheric Physics*, 92, 45–66, doi: 10.1007/s00703-005-0112-4, 2006.
- Seifert, A., Heus, T., Pincus, R., and Stevens, B.: Large-eddy simulation of the transient and near-equilibrium behavior of precipitating shallow convection, *Journal of Advances in Modeling Earth Systems*, 7, 1918–1937, doi: 10.1002/2015MS000489, 2015.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric chemistry and physics : from air pollution to climate change*, John Wiley & Sons, Hoboken, N.J, 3rd ed. edn., 2016.

- Seinfeld, J. H., Bretherton, C., Carslaw, K. S., Coe, H., DeMott, P. J., Dunlea, E. J., Feingold, G., Ghan, S., Guenther, A. B., Kahn, R., Kraucunas, I., Kreidenweis, S. M., Molina, M. J., Nenes, A., Penner, J. E., Prather, K. A., Ramanathan, V., Ramaswamy, V., Rasch, P. J., Ravishankara, A. R., Rosenfeld, D., Stephens, G., and Wood, R.: Improving our fundamental understanding of the role of aerosol-cloud interactions in the climate system, *Proceedings of the National Academy of Sciences of the United States of America*, 113, 5781–5790, doi: 10.1073/pnas.1514043113, 2016.
- Shapley, L. S.: A Value for n-Person Games, in: *Contributions to the Theory of Games*, edited by Kuhn, H. W. and Tucker, A. W., vol. 2 of *Annals of Mathematics Studies*, pp. 307–317, Princeton University Press, Princeton, NJ, doi: doi:10.1515/9781400881970-018, 1953.
- Sherwood, S. C., Bony, S., Boucher, O., Bretherton, C., Forster, P. M., Gregory, J. M., and Stevens, B.: Adjustments in the Forcing-Feedback Framework for Understanding Climate Change, *Bulletin of the American Meteorological Society*, 96, 217–228, doi: <https://doi.org/10.1175/BAMS-D-13-00167.1>, 2015.
- Sherwood, S. C., Webb, M. J., Annan, J. D., Armour, K. C., Forster, P. M., Hargreaves, J. C., Hegerl, G., Klein, S. A., Marvel, K. D., Rohling, E. J., Watanabe, M., Andrews, T., Braconnot, P., Bretherton, C. S., Foster, G. L., Hausfather, Z., von der Heydt, A. S., Knutti, R., Mauritsen, T., Norris, J. R., Proistosescu, C., Rugenstein, M., Schmidt, G. A., Tokarska, K. B., and Zelinka, M. D.: An Assessment of Earth’s Climate Sensitivity Using Multiple Lines of Evidence, *Reviews of Geophysics*, 58, e2019RG000 678, doi: <https://doi.org/10.1029/2019RG000678>, 2020.
- Simonyan, K., Vedaldi, A., and Zisserman, A.: Deep Inside Convolutional Networks: Visualising Image Classification Models and Saliency Maps, *arXiv preprint arXiv:1312.6034*, doi: 10.48550/arXiv.1312.6034, 2013.
- Small, J. D., Chuang, P. Y., Feingold, G., and Jiang, H.: Can aerosol decrease cloud lifetime?, *Geophysical Research Letters*, 36, 1–5, doi: 10.1029/2009GL038888, 2009.
- Small, J. D., Jiang, J. H., Su, H., and Zhai, C.: Relationship between aerosol and cloud fraction over Australia, *Geophysical Research Letters*, 38, doi: <https://doi.org/10.1029/2011GL049404>, 2011.
- Smith, C. J., Kramer, R. J., Myhre, G., Alterskjær, K., Collins, W., Sima, A., Boucher, O., Dufresne, J.-L., Nabat, P., Michou, M., Yukimoto, S., Cole, J., Paynter, D.,

- Shiogama, H., O'Connor, F. M., Robertson, E., Wiltshire, A., Andrews, T., Hannay, C., Miller, R., Nazarenko, L., Kirkevåg, A., Olivié, D., Fiedler, S., Lewinschal, A., Mackallah, C., Dix, M., Pincus, R., and Forster, P. M.: Effective radiative forcing and adjustments in CMIP6 models, *Atmospheric Chemistry and Physics*, 20, 9591–9618, doi: 10.5194/acp-20-9591-2020, 2020.
- Snoek, J., Larochelle, H., and Adams, R. P.: Practical Bayesian Optimization of Machine Learning Algorithms, in: *Advances in Neural Information Processing Systems*, edited by Pereira, F., Burges, C. J., Bottou, L., and Weinberger, K. Q., vol. 25, Curran Associates, Inc., URL <https://proceedings.neurips.cc/paper/2012/file/05311655a15b75fab86956663e1819cd-Paper.pdf>, 2012.
- Stengel, K., Glaws, A., Hettinger, D., and King, R. N.: Adversarial super-resolution of climatological wind and solar data, *Proceedings of the National Academy of Sciences*, 117, 16 805–16 815, doi: 10.1073/pnas.1918964117, 2020.
- Stephens, G., Winker, D., Pelon, J., Trepte, C., Vane, D., Yuhas, C., L'Ecuyer, T., and Lebsock, M.: CloudSat and CALIPSO within the A-Train: Ten Years of Actively Observing the Earth System, *Bulletin of the American Meteorological Society*, 99, 569–581, doi: <https://doi.org/10.1175/BAMS-D-16-0324.1>, 2018.
- Stephens, G. L. and Kummerow, C. D.: The Remote Sensing of Clouds and Precipitation from Space: A Review, *Journal of the Atmospheric Sciences*, 64, 3742–3765, doi: <https://doi.org/10.1175/2006JAS2375.1>, 2007.
- Stephens, G. L., Vane, D. G., Boain, R. J., Mace, G. G., Sassen, K., Wang, Z., Illingworth, A. J., O'connor, E. J., Rossow, W. B., Durden, S. L., Miller, S. D., Austin, R. T., Benedetti, A., and Mitrescu, C.: THE CLOUDSAT MISSION AND THE A-TRAIN: A New Dimension of Space-Based Observations of Clouds and Precipitation, *Bulletin of the American Meteorological Society*, 83, 1771–1790, doi: <https://doi.org/10.1175/BAMS-83-12-1771>, 2002.
- Stephens, G. L., Vane, D. G., Tanelli, S., Im, E., Durden, S., Rokey, M., Reinke, D., Partain, P., Mace, G. G., Austin, R., L'Ecuyer, T., Haynes, J., Lebsock, M., Suzuki, K., Waliser, D., Wu, D., Kay, J., Gettelman, A., Wang, Z., and Marchand, R.: CloudSat mission: Performance and early science after the first year of operation, *Journal of Geophysical Research: Atmospheres*, 113, doi: <https://doi.org/10.1029/2008JD009982>, 2008.
- Stephens, G. L., Li, J., Wild, M., Clayson, C. A., Loeb, N., Kato, S., L'Ecuyer, T., Stackhouse, P. W., Lebsock, M., and Andrews, T.: An update on Earth's energy

- balance in light of the latest global observations, *Nature Geoscience*, 5, 691–696, doi: 10.1038/ngeo1580, 2012.
- Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, *Nature*, 461, 607–613, doi: 10.1038/nature08281, 2009.
- Stevens, B., Giorgetta, M., Esch, M., Mauritsen, T., Crueger, T., Rast, S., Salzmann, M., Schmidt, H., Bader, J., Block, K., Brokopf, R., Fast, I., Kinne, S., Kornblueh, L., Lohmann, U., Pincus, R., Reichler, T., and Roeckner, E.: Atmospheric component of the MPI-M Earth System Model: ECHAM6, *Journal of Advances in Modeling Earth Systems*, 5, 146–172, doi: <https://doi.org/10.1002/jame.20015>, 2013.
- Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, doi: 10.5194/acp-5-1125-2005, 2005.
- Stier, P., van den Heever, S. C., Christensen, M. W., Gryspeerdt, E., Dagan, G., Saleeby, S. M., Bollasina, M., Donner, L., Emanuel, K., Ekman, A. M. L., Feingold, G., Field, P., Forster, P., Haywood, J., Kahn, R., Koren, I., Kummerow, C., L’Ecuyer, T., Lohmann, U., Ming, Y., Myhre, G., Quaas, J., Rosenfeld, D., Samset, B., Seifert, A., Stephens, G., and Tao, W.-K.: Multifaceted aerosol effects on precipitation, *Nature Geoscience*, 17, 719–732, doi: 10.1038/s41561-024-01482-6, 2024.
- Stirnberg, R., Cermak, J., Kotthaus, S., Haeffelin, M., Andersen, H., Fuchs, J., Kim, M., Petit, J. E., and Favez, O.: Meteorology-driven variability of air pollution (PM₁) revealed with explainable machine learning, *Atmospheric Chemistry and Physics*, 21, 3919–3948, doi: 10.5194/acp-21-3919-2021, 2021.
- Stubenrauch, C. J., Rossow, W. B., Kinne, S., Ackerman, S., Cesana, G., Chepfer, H., Di Girolamo, L., Getzewich, B., Guignard, A., Heidinger, A., Maddux, B. C., Menzel, W. P., Minnis, P., Pearl, C., Platnick, S., Poulsen, C., Riedi, J., Sun-Mack, S., Walther, A., Winker, D., Zeng, S., and Zhao, G.: Assessment of Global Cloud Datasets from Satellites: Project and Database Initiated by the GEWEX Radiation Panel, *Bulletin of the American Meteorological Society*, 94, 1031–1049, doi: <https://doi.org/10.1175/BAMS-D-12-00117.1>, 2013.
- Su, W., Loeb, N. G., Xu, K.-M., Schuster, G. L., and Eitzen, Z. A.: An estimate of aerosol indirect effect from satellite measurements with concurrent meteorological analysis, *Journal of Geophysical Research: Atmospheres*, 115, doi: <https://doi.org/10.1029/2010JD013948>, 2010.

- Sundararajan, M. and Najmi, A.: The many shapley values for model explanation, in: 37th International Conference on Machine Learning, ICML 2020, vol. PartF16814, pp. 9210–9220, 2020.
- Sundararajan, M., Taly, A., and Yan, Q.: Axiomatic Attribution for Deep Networks, in: Proceedings of the 34th International Conference on Machine Learning, edited by Precup, D. and Teh, Y. W., vol. 70 of *Proceedings of Machine Learning Research*, pp. 3319–3328, PMLR, URL <https://proceedings.mlr.press/v70/sundararajan17a.html>, 2017.
- Sundqvist, H., Berge, E., and Kristjánsson, J. E.: Condensation and Cloud Parameterization Studies with a Mesoscale Numerical Weather Prediction Model, *Monthly Weather Review*, 117, 1641–1657, doi: [https://doi.org/10.1175/1520-0493\(1989\)117<1641:CACPSW>2.0.CO;2](https://doi.org/10.1175/1520-0493(1989)117<1641:CACPSW>2.0.CO;2), 1989.
- Taylor, K. E., Williamson, D., and Zwiers, F.: PCMDI Report - Number 60 - The sea surface temperature and sea-ice concentration boundary conditions for AMIP II simulations, doi: 10.5281/zenodo.13835219, 2000.
- Tegen, I., Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Bey, I., Schutgens, N., Stier, P., Watson-Parris, D., Stanelle, T., Schmidt, H., Rast, S., Kokkola, H., Schultz, M., Schroeder, S., Daskalakis, N., Barthel, S., Heinold, B., and Lohmann, U.: The global aerosol–climate model ECHAM6.3–HAM2.3 – Part 1: Aerosol evaluation, *Geoscientific Model Development*, 12, 1643–1677, doi: 10.5194/gmd-12-1643-2019, 2019.
- Toll, V., Christensen, M., Quaas, J., and Bellouin, N.: Weak average liquid-cloud-water response to anthropogenic aerosols, *Nature*, 572, 51–55, doi: 10.1038/s41586-019-1423-9, 2019.
- Trenberth, K. E., Fasullo, J. T., and Kiehl, J.: Earth’s Global Energy Budget, *Bulletin of the American Meteorological Society*, 90, 311–324, doi: <https://doi.org/10.1175/2008BAMS2634.1>, 2009.
- Turner, D. D., Vogelmann, A. M., Austin, R. T., Barnard, J. C., Cady-Pereira, K., Chiu, J. C., Clough, S. A., Flynn, C., Khaiyer, M. M., Liljegren, J., Johnson, K., Lin, B., Long, C., Marshak, A., Matrosov, S. Y., McFarlane, S. A., Miller, M., Min, Q., Minimis, P., O’Hirok, W., Wang, Z., and Wiscombe, W.: Thin Liquid Water Clouds: Their Importance and Our Challenge, *Bulletin of the American Meteorological Society*, 88, 177–190, doi: <https://doi.org/10.1175/BAMS-88-2-177>, 2007.

- Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, *Journal of the Atmospheric Sciences*, 34, 1149–1152, doi: 10.1175/1520-0469(1977)034<1149:tiopot>2.0.co;2, 1977.
- van der Dussen, J. J., de Roode, S. R., Dal Gesso, S., and Siebesma, A. P.: An LES model study of the influence of the free tropospheric thermodynamic conditions on the stratocumulus response to a climate perturbation, *Journal of Advances in Modeling Earth Systems*, 7, 670–691, doi: <https://doi.org/10.1002/2014MS000380>, 2015.
- van der Dussen, J. J., de Roode, S. R., and Siebesma, A. P.: How large-scale subsidence affects stratocumulus transitions, *Atmospheric Chemistry and Physics*, 16, 691–701, doi: 10.5194/acp-16-691-2016, 2016.
- Van Katwyk, P., Fox-Kemper, B., Hewitt, H. T., and Bergen, K. J.: Rewiring climate modeling with machine learning emulators, *Communications Earth & Environment*, 7, 107, doi: 10.1038/s43247-026-03238-z, 2026.
- Vandal, T., Kodra, E., Ganguly, S., Michaelis, A., Nemani, R., and Ganguly, A. R.: DeepSD: Generating High Resolution Climate Change Projections through Single Image Super-Resolution, in: *Proceedings of the 23rd ACM SIGKDD International Conference on Knowledge Discovery and Data Mining, KDD '17*, p. 1663–1672, Association for Computing Machinery, New York, NY, USA, doi: 10.1145/3097983.3098004, 2017.
- Vaswani, A., Shazeer, N., Parmar, N., Uszkoreit, J., Jones, L., Gomez, A. N., Kaiser, L., and Polosukhin, I.: Attention is All you Need, in: *Advances in Neural Information Processing Systems*, edited by Guyon, I., Luxburg, U. V., Bengio, S., Wallach, H., Fergus, R., Vishwanathan, S., and Garnett, R., vol. 30, Curran Associates, Inc., URL https://proceedings.neurips.cc/paper_files/paper/2017/file/3f5ee243547dee91fbd053c1c4a845aa-Paper.pdf, 2017.
- von Schuckmann, K., Minière, A., Gues, F., Cuesta-Valero, F. J., Kirchengast, G., Adusumilli, S., Straneo, F., Ablain, M., Allan, R. P., Barker, P. M., Beltrami, H., Blazquez, A., Boyer, T., Cheng, L., Church, J., Desbruyeres, D., Dolman, H., Domingues, C. M., Garcia-Garcia, A., Giglio, D., Gilson, J. E., Gorfer, M., Haimberger, L., Hakuba, M. Z., Hendricks, S., Hosoda, S., Johnson, G. C., Killick, R., King, B., Kolodziejczyk, N., Korosov, A., Krinner, G., Kuusela, M., Landerer, F. W., Langer, M., Lavergne, T., Lawrence, I., Li, Y., Lyman, J., Marti, F., Marzeion, B., Mayer, M., MacDougall, A. H., McDougall, T., Monselesan, D. P., Nitzbon,

- J., Otosaka, I., Peng, J., Purkey, S., Roemmich, D., Sato, K., Sato, K., Savita, A., Schweiger, A., Shepherd, A., Seneviratne, S. I., Simons, L., Slater, D. A., Slater, T., Steiner, A. K., Suga, T., Szekely, T., Thiery, W., Timmermans, M.-L., Vanderkelen, I., Wjiffels, S. E., Wu, T., and Zemp, M.: Heat stored in the Earth system 1960–2020: where does the energy go?, *Earth System Science Data*, 15, 1675–1709, doi: 10.5194/essd-15-1675-2023, 2023.
- Wall, C. J., Hartmann, D. L., and Ma, P.-L.: Instantaneous Linkages between Clouds and Large-Scale Meteorology over the Southern Ocean in Observations and a Climate Model, *Journal of Climate*, 30, 9455–9474, doi: <https://doi.org/10.1175/JCLI-D-17-0156.1>, 2017.
- Wall, C. J., Norris, J. R., Possner, A., McCoy, D. T., McCoy, I. L., and Lutsko, N. J.: Assessing effective radiative forcing from aerosol–cloud interactions over the global ocean, *Proceedings of the National Academy of Sciences*, 119, e2210481 119, doi: 10.1073/pnas.2210481119, 2022.
- Wall, C. J., Storelvmo, T., and Possner, A.: Global observations of aerosol indirect effects from marine liquid clouds, *Atmospheric Chemistry and Physics*, 23, 13 125–13 141, doi: 10.5194/acp-23-13125-2023, 2023.
- Wallace, J. M. and Hobbs, P. V.: 6 - Cloud Microphysics, in: *Atmospheric Science (Second Edition)*, edited by Wallace, J. M. and Hobbs, P. V. B. T. A. S. S. E., pp. 209–269, Academic Press, San Diego, doi: <https://doi.org/10.1016/B978-0-12-732951-2.50011-9>, 2006.
- Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, *Atmospheric Chemistry and Physics*, 9, 239–260, doi: 10.5194/acp-9-239-2009, 2009.
- Wang, X., Mao, F., Zhu, Y., Rosenfeld, D., Pan, Z., Zang, L., Lu, X., Liu, F., and Gong, W.: Hidden Large Aerosol-Driven Cloud Cover Effect Over High-Latitude Ocean, *Journal of Geophysical Research: Atmospheres*, 129, e2023JD039 312, doi: <https://doi.org/10.1029/2023JD039312>, 2024.
- Wang, Y., Neubauer, D., Chen, Y., Jordan, G., Malavelle, F., Yuan, T., Partridge, D., Field, P., Wang, H., Wang, M., Michou, M., Nabat, P., Laakso, A., Myhre, G., and Lohmann, U.: Challenges in global climate models to represent cloud response to aerosols: insights from volcanic eruptions, *Nature Communications*, 17, 627, doi: 10.1038/s41467-025-67359-3, 2025.

- Warren, S. G., Hahn, C. J., London, J., Chervin, R. M., and Jenne, R. L.: Global Distribution of Total Cloud Cover and Cloud Type Amounts Over the Ocean, Tech. rep., University Corporation for Atmospheric Research, doi: 10.5065/D6QC01D1, 1988.
- Watson, P. A. G.: Applying Machine Learning to Improve Simulations of a Chaotic Dynamical System Using Empirical Error Correction, *Journal of Advances in Modeling Earth Systems*, 11, 1402–1417, doi: <https://doi.org/10.1029/2018MS001597>, 2019.
- Watson-Parris, D.: Machine learning for weather and climate are worlds apart, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 379, 20200098, doi: 10.1098/rsta.2020.0098, 2021.
- Watson-Parris, D., Rao, Y., Olivié, D., Seland, Ø., Nowack, P., Camps-Valls, G., Stier, P., Bouabid, S., Dewey, M., Fons, E., Gonzalez, J., Harder, P., Jeggle, K., Lenhardt, J., Manshausen, P., Novitasari, M., Ricard, L., and Roesch, C.: ClimateBench v1.0: A Benchmark for Data-Driven Climate Projections, *Journal of Advances in Modeling Earth Systems*, 14, e2021MS002954, doi: <https://doi.org/10.1029/2021MS002954>, 2022.
- Watt-Meyer, O., Brenowitz, N. D., Clark, S. K., Henn, B., Kwa, A., McGibbon, J., Perkins, W. A., and Bretherton, C. S.: Correcting Weather and Climate Models by Machine Learning Nudged Historical Simulations, *Geophysical Research Letters*, 48, e2021GL092555, doi: <https://doi.org/10.1029/2021GL092555>, 2021.
- Wehr, T., Kubota, T., Tzeremes, G., Wallace, K., Nakatsuka, H., Ohno, Y., Koopman, R., Rusli, S., Kikuchi, M., Eisinger, M., Tanaka, T., Taga, M., Deghaye, P., Tomita, E., and Bernaerts, D.: The EarthCARE mission – science and system overview, *Atmospheric Measurement Techniques*, 16, 3581–3608, doi: 10.5194/amt-16-3581-2023, 2023.
- Werner, F. and Deneke, H.: Increasing the spatial resolution of cloud property retrievals from Meteosat SEVIRI by use of its high-resolution visible channel: evaluation of candidate approaches with MODIS observations, *Atmospheric Measurement Techniques*, 13, 1089–1111, doi: 10.5194/amt-13-1089-2020, 2020.
- Wild, M., Hakuba, M. Z., Folini, D., Dörig-Ott, P., Schär, C., Kato, S., and Long, C. N.: The cloud-free global energy balance and inferred cloud radiative effects: an assessment based on direct observations and climate models, *Climate Dynamics*, 52, 4787–4812, doi: 10.1007/s00382-018-4413-y, 2019.

- Wind, G., Platnick, S., King, M. D., Hubanks, P. A., Pavolonis, M. J., Heidinger, A. K., Yang, P., and Baum, B. A.: Multilayer Cloud Detection with the MODIS Near-Infrared Water Vapor Absorption Band, *Journal of Applied Meteorology and Climatology*, 49, 2315–2333, doi: <https://doi.org/10.1175/2010JAMC2364.1>, 2010.
- Winker, D. M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., Hunt, W. H., and Young, S. A.: Overview of the CALIPSO Mission and CALIOP Data Processing Algorithms, *Journal of Atmospheric and Oceanic Technology*, 26, 2310–2323, doi: <https://doi.org/10.1175/2009JTECHA1281.1>, 2009.
- Wood, R.: Stratocumulus Clouds, *Monthly Weather Review*, 140, 2373–2423, doi: [10.1175/MWR-D-11-00121.1](https://doi.org/10.1175/MWR-D-11-00121.1), 2012.
- Wood, R. and Bretherton, C. S.: Boundary Layer Depth, Entrainment, and Decoupling in the Cloud-Capped Subtropical and Tropical Marine Boundary Layer, *Journal of Climate*, 17, 3576–3588, doi: [https://doi.org/10.1175/1520-0442\(2004\)017<3576:BLDEAD>2.0.CO;2](https://doi.org/10.1175/1520-0442(2004)017<3576:BLDEAD>2.0.CO;2), 2004.
- Wood, R. and Bretherton, C. S.: On the relationship between stratiform low cloud cover and lower-tropospheric stability, *Journal of Climate*, 19, 6425–6432, doi: [10.1175/JCLI3988.1](https://doi.org/10.1175/JCLI3988.1), 2006.
- Wood, R. and Hartmann, D. L.: Spatial Variability of Liquid Water Path in Marine Low Cloud: The Importance of Mesoscale Cellular Convection, *Journal of Climate*, 19, 1748–1764, doi: <https://doi.org/10.1175/JCLI3702.1>, 2006.
- Wood, R., Bretherton, C. S., and Hartmann, D. L.: Diurnal cycle of liquid water path over the subtropical and tropical oceans, *Geophysical Research Letters*, 29, 4–7, doi: <https://doi.org/10.1029/2002GL015371>, 2002.
- Wood, R., Kubar, T. L., and Hartmann, D. L.: Understanding the Importance of Microphysics and Macrophysics for Warm Rain in Marine Low Clouds. Part II: Heuristic Models of Rain Formation, *Journal of the Atmospheric Sciences*, 66, 2973–2990, doi: <https://doi.org/10.1175/2009JAS3072.1>, 2009.
- Wood, R., Wyant, M., Bretherton, C. S., Rémillard, J., Kollias, P., Fletcher, J., Stemmler, J., de Szoeko, S., Yuter, S., Miller, M., Mechem, D., Tselioudis, G., Chiu, J. C., Mann, J. A. L., O'Connor, E. J., Hogan, R. J., Dong, X., Miller, M., Ghate, V., Jefferson, A., Min, Q., Minnis, P., Palikonda, R., Albrecht, B., Luke, E., Hannay, C., and Lin, Y.: Clouds, Aerosols, and Precipitation in the Marine Boundary Layer:

- An Arm Mobile Facility Deployment, *Bulletin of the American Meteorological Society*, 96, 419–440, doi: 10.1175/BAMS-D-13-00180.1, 2015.
- Xu, K.-M. and Randall, D. A.: A Semiempirical Cloudiness Parameterization for Use in Climate Models, *Journal of Atmospheric Sciences*, 53, 3084–3102, doi: [https://doi.org/10.1175/1520-0469\(1996\)053<3084:ASCPFU>2.0.CO;2](https://doi.org/10.1175/1520-0469(1996)053<3084:ASCPFU>2.0.CO;2), 1996.
- Xu, Z., Kollias, P., Sasikumar, S., Battaglia, A., Puigdomènech Treserras, B., and McLinden, M. L. W.: EarthCARE Cloud Profiling Radar observations of the vertical structure of marine stratocumulus clouds, *Atmospheric Chemistry and Physics*, 26, 4619–4632, doi: 10.5194/acp-26-4619-2026, 2026.
- Xue, H. and Feingold, G.: Large-Eddy Simulations of Trade Wind Cumuli: Investigation of Aerosol Indirect Effects, *Journal of the Atmospheric Sciences*, 63, 1605–1622, doi: 10.1175/JAS3706.1, 2006.
- Ying, X.: An Overview of Overfitting and its Solutions, *Journal of Physics: Conference Series*, 1168, 22 022, doi: 10.1088/1742-6596/1168/2/022022, 2019.
- Young, G. S., Kristovich, D. A. R., Hjelmfelt, M. R., and Foster, R. C.: ROLLS, STREETS, WAVES, AND MORE: A Review of Quasi-Two-Dimensional Structures in the Atmospheric Boundary Layer, *Bulletin of the American Meteorological Society*, 83, 997–1002, doi: [https://doi.org/10.1175/1520-0477\(2002\)083<0997:RSWAMA>2.3.CO;2](https://doi.org/10.1175/1520-0477(2002)083<0997:RSWAMA>2.3.CO;2), 2002.
- Yuan, T. and Oreopoulos, L.: On the global character of overlap between low and high clouds, *Geophysical Research Letters*, 40, 5320–5326, doi: <https://doi.org/10.1002/grl.50871>, 2013.
- Yuan, T., Song, H., Wood, R., Oreopoulos, L., Platnick, S., Wang, C., Yu, H., Meyer, K., and Wilcox, E.: Observational evidence of strong forcing from aerosol effect on low cloud coverage, doi: 10.1126/sciadv.adh7716, 2023.
- Zelinka, M. D., Andrews, T., Forster, P. M., and Taylor, K. E.: Quantifying components of aerosol-cloud-radiation interactions in climate models, *Journal of Geophysical Research: Atmospheres*, 119, 7599–7615, doi: 10.1002/2014JD021710, 2014.
- Zelinka, M. D., Grise, K. M., Klein, S. A., Zhou, C., DeAngelis, A. M., and Christensen, M. W.: Drivers of the Low-Cloud Response to Poleward Jet Shifts in the North Pacific in Observations and Models, *Journal of Climate*, 31, 7925–7947, doi: <https://doi.org/10.1175/JCLI-D-18-0114.1>, 2018.

- Zelinka, M. D., Myers, T. A., McCoy, D. T., Po-Chedley, S., Caldwell, P. M., Ceppi, P., Klein, S. A., and Taylor, K. E.: Causes of Higher Climate Sensitivity in CMIP6 Models, *Geophysical Research Letters*, 47, e2019GL085782, doi: <https://doi.org/10.1029/2019GL085782>, 2020.
- Zelinka, M. D., Smith, C. J., Qin, Y., and Taylor, K. E.: Comparison of methods to estimate aerosol effective radiative forcings in climate models, *Atmospheric Chemistry and Physics*, 23, 8879–8898, doi: 10.5194/acp-23-8879-2023, 2023.
- Zhang, K., O'Donnell, D., Kazil, J., Stier, P., Kinne, S., Lohmann, U., Ferrachat, S., Croft, B., Quaas, J., Wan, H., Rast, S., and Feichter, J.: The global aerosol-climate model ECHAM-HAM, version 2: Sensitivity to improvements in process representations, *Atmospheric Chemistry and Physics*, 12, 8911–8949, doi: 10.5194/acp-12-8911-2012, 2012a.
- Zhang, K., Wan, H., Liu, X., Ghan, S. J., Kooperman, G. J., Ma, P.-L., Rasch, P. J., Neubauer, D., and Lohmann, U.: Technical Note: On the use of nudging for aerosol–climate model intercomparison studies, *Atmospheric Chemistry and Physics*, 14, 8631–8645, doi: 10.5194/acp-14-8631-2014, 2014.
- Zhang, Z. and Platnick, S.: An assessment of differences between cloud effective particle radius retrievals for marine water clouds from three MODIS spectral bands, *Journal of Geophysical Research: Atmospheres*, 116, doi: <https://doi.org/10.1029/2011JD016216>, 2011.
- Zhang, Z., Ackerman, A. S., Feingold, G., Platnick, S., Pincus, R., and Xue, H.: Effects of cloud horizontal inhomogeneity and drizzle on remote sensing of cloud droplet effective radius: Case studies based on large-eddy simulations, *Journal of Geophysical Research: Atmospheres*, 117, doi: <https://doi.org/10.1029/2012JD017655>, 2012b.
- Zhu, Y., Rosenfeld, D., and Li, Z.: Under What Conditions Can We Trust Retrieved Cloud Drop Concentrations in Broken Marine Stratocumulus?, *Journal of Geophysical Research: Atmospheres*, 123, 8754–8767, doi: 10.1029/2017JD028083, 2018.
- Zipfel, L., Andersen, H., and Cermak, J.: Machine-Learning Based Analysis of Liquid Water Path Adjustments to Aerosol Perturbations in Marine Boundary Layer Clouds Using Satellite Observations, *Atmosphere*, 13, doi: 10.3390/atmos13040586, 2022.

BIBLIOGRAPHY

Zipfel, L., Andersen, H., Grosvenor, D. P., and Cermak, J.: How Cloud Droplet Number Concentration Impacts Liquid Water Path and Precipitation in Marine Stratocumulus Clouds—A Satellite-Based Analysis Using Explainable Machine Learning, doi: 10.3390/atmos15050596, 2024.

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Yichen Jia contributed to the development of the study concept and methodology. Yichen Jia obtained and analyzed the datasets, implemented the explainable machine-learning framework, performed the visualization, interpreted the results and wrote the original draft of the manuscript. Yichen Jia also revised the manuscript during the peer-review process.

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Analysis of the cloud fraction adjustment to aerosols and its dependence on meteorological controls using explainable machine learning

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Abstract. Aerosol–cloud interactions (ACI) have a pronounced influence on the Earth’s radiation budget but continue to pose one of the most substantial uncertainties in the climate system. Marine boundary-layer clouds (MBLCs) are particularly important since they cover a large portion of the Earth’s surface. One of the biggest challenges in quantifying ACI from observations lies in isolating adjustments of cloud fraction (CLF) to aerosol perturbations from the covariability and influence of the local meteorological conditions. In this study, this isolation is attempted using 9 years (2011–2019) of near-global daily satellite cloud products in combination with reanalysis data of meteorological parameters. With cloud-droplet number concentration (N_d) as a proxy for aerosol, MBLC CLF is predicted by region-specific gradient boosting machine learning (ML) models. By means of SHapley Additive exPlanation (SHAP) regression values, CLF sensitivity to N_d and meteorological factors as well as meteorological influences on the N_d –CLF sensitivity are quantified. The regional ML models are able to capture, on average, 45 % of the CLF variability. Based on our statistical approach, global patterns of CLF sensitivity suggest that CLF is positively associated with N_d , particularly in the stratocumulus-to-cumulus transition regions and the Southern Hemispheric midlatitudes. However, N_d retrieval bias may contribute to non-causality in these positive sensitivities, and hence they should be considered upper-bound estimates. CLF sensitivity to estimated inversion strength (EIS) is ubiquitously positive and strongest in tropical and subtropical regions topped by stratocumulus and within the midlatitudes. Globally, increased sea-surface temperature (SST) reduces CLF, particularly in stratocumulus regions. The spatial patterns of CLF sensitivity to horizontal wind components in the free troposphere may point to the impact of synoptic-scale weather systems and vertical wind shear on MBLCs. The N_d –CLF relationship is found to depend more on the selected thermodynamical variables than dynamical variables and in particular on EIS and SST. In the midlatitudes, a stronger inversion is found to amplify the N_d –CLF relationship, while this is not observed in the stratocumulus regions. In the stratocumulus-to-cumulus transition regions, the N_d –CLF sensitivity is found to be amplified by higher SSTs, potentially pointing to N_d more frequently delaying this transition in these conditions. The expected climatic changes in EIS and SST may thus influence future forcings from the CLF adjustment. The novel data-driven framework, whose limitations are also discussed, produces a quantification of the response of MBLC CLF to aerosols, taking into account the covariations with meteorology.

1 Introduction

The emission of aerosols into the atmosphere affects the Earth's climate in particular by masking part of the warming effect from greenhouse gases by reflecting solar radiation and changing cloud properties. Aerosol–cloud interactions (ACIs) can strongly influence the Earth's energy distribution and thus also contribute a substantial uncertainty to past and future climate projections. The effective radiative forcing due to ACI (ERF_{aci}) is assessed to be -1.0 W m^{-2} , with an uncertainty range of -1.7 to -0.3 W m^{-2} (Forster et al., 2021) albeit decades of effort and headway have been made in understanding the complex system of aerosols, clouds, and their environmental controls. The correct representation of ACI in Earth system models (ESMs) remains a tremendous challenge because of the lack of accurate global quantification of the cloud-related fine-scale processes and the lack of larger-scale constraints from the existing measurement systems at the ESM spatiotemporal resolution (Fan et al., 2016; Seinfeld et al., 2016; Sato et al., 2018).

Marine boundary-layer clouds (MBLCs) cover over 23 % of the global ocean surface (Wood, 2012). Due to relatively small temperature differences between MBLC top and the sea surface, they only weakly impact outgoing longwave radiation but greatly reflect incoming shortwave radiation, leading to a strong net cooling effect (Hartmann et al., 1992). MBLCs play a critical role in the Earth's radiative balance (Zheng et al., 2021) and, in this regard, are the most important cloud type (Chen et al., 2014). Furthermore, MBLCs are especially susceptible to aerosol perturbations due to their relatively low optical depths (Turner, 2007; Leahy et al., 2012) and their formation in environments typically characterized by lower anthropogenic aerosol loading than continental clouds (Platnick and Twomey, 1994). Therefore, a deeper understanding of the aerosol–MBLC interactions is crucial to reduce the uncertainties in climate predictions. Atmospheric aerosols are critical for the formation of clouds as cloud condensation nuclei (CCN). Increases in aerosols are associated with increases in cloud-droplet number concentration (N_d). As the cloud water is distributed among more droplets, cloud-droplet effective radius (r_e) shrinks at constant liquid water content, resulting in an enhancement of cloud brightness and a negative instantaneous radiative forcing (Twomey, 1977). The likelihood of collision and coalescence subsequently decreases due to smaller drop sizes, hampering rainfall formation, which can prolong cloud lifetime and thus increase cloud fraction (CLF) (Albrecht, 1989). However, the aerosol–CLF relationship is complex, and the sign of the CLF adjustment can also be the opposite. This has been found in particular for non-precipitating clouds, stemming from enhanced entrainment mixing with ambient air over the clouds owing to shorter evaporation timescales (Wang et al., 2003; Jiang et al., 2006; Small et al., 2009) or reduced sedimentation (Ackerman et al., 2004; Bretherton et al., 2007) because of smaller droplet sizes.

From the perspective of observations at satellite scales, though there are studies suggesting a negative relationship between aerosols and CLF (Dey et al., 2011; Small et al., 2011), it has been documented by multiple studies that the overall CLF increases in response to increasing aerosols (e.g. Kaufman and Koren, 2006; Yuan et al., 2011; Gryspeerdt et al., 2016; Christensen et al., 2017; Andersen et al., 2017; Fuchs et al., 2018; Rosenfeld et al., 2019; Christensen et al., 2020). Likewise, studies based on ESMs reported substantial negative ERF_{aci} due to liquid water path (LWP) and CLF adjustments (e.g. Zelinka et al., 2014). In spite of the attribution of such adjustments in ESMs primarily to LWP adjustments (Ghan et al., 2016), a global satellite-based study by Bender et al. (2019) suggested that LWP adjustments are overestimated in ESMs and that aerosol impact on CLF dominates the negative aerosol forcing. This is supported by observational evidence presented by Toll et al. (2019), who also reported an overestimation of LWP adjustment in climate models, and by Y. Chen et al. (2022), who recently highlighted the role of CLF increases due to aerosols from a large volcano eruption as the main cause of the associated forcing. Some large-eddy simulations have, however, suggested a negative response of CLF of trade wind cumulus to aerosol perturbations (Xue and Feingold, 2006; Seifert et al., 2015). While most studies, from both observational and model points of view, are in agreement that generally CLF increases with increasing aerosols due to a prolonged lifetime (Douglas and L'Ecuyer, 2022), the magnitude of the response of CLF to aerosols and its corresponding adjustments are still highly uncertain. For satellite-based analyses, one of the most challenging aspects in the quantification of CLF adjustment is isolating the influence of the aerosol loading on cloud properties from confounding covariations with meteorological parameters (Andersen et al., 2016; Gryspeerdt et al., 2019; Bellouin et al., 2020) paired with aerosol retrieval issues related to aerosol swelling and 3D radiative effects in the vicinity of clouds (Loeb and Schuster, 2008; Schwarz et al., 2017). Recent observational studies have utilized different methods to tackle this issue. A first approach is to stratify the data by meteorological factors, therefore accounting for local meteorology in the relationships (e.g. Su et al., 2010; Chen et al., 2014; Andersen and Cermak, 2015). Secondly, using N_d as a mediating variable was proposed by Gryspeerdt et al. (2016) to analyse the causal pathway between aerosol optical depth and CLF. Another approach is to use a sampling strategy that applies a cloud–aerosol pairing algorithm (Christensen et al., 2017). However, these methods do not account for aerosol retrieval issues, meteorological influencing factors, and confounders at once, which is essential to constrain the CLF adjustment. Recently, several studies have successfully used machine learning (ML) to account for non-linearities and meteorological factors to quantify ACI (Andersen et al., 2017; Fuchs et al., 2018; Dadashazar et al., 2021; Zipfel et al., 2022). ML regression algorithms allow for the prediction of CLF (predictand) on the basis of aerosol and me-

teological factors at the same time and treat the aerosol–cloud–meteorology system as a whole. In addition, ML models can represent non-linear interactive systems, which can be analysed in sensitivity analyses with explainable ML techniques. Explainable ML refers to the techniques explaining the predictions of a trained ML model by explicitly quantifying the relationships, which helps improve the understandability, transparency, and trustworthiness of the ML models (Beucler et al., 2023).

In this study, we set up region-specific ML models at a global scale using satellite and reanalysis data sets to predict CLF to analyse N_d -induced changes in MBLCs. The goal of the explainable ML framework is to quantify the global sensitivity patterns of CLF to N_d and meteorological factors. In addition, we aim to estimate the magnitude of the dependence of N_d –CLF sensitivity on the meteorological factors using SHapley Additive exPlanation (SHAP) interaction values, providing a new and insightful pathway to more profound knowledge of the physical processes relevant to the CLF adjustment and, hence, to a global constraint on aerosol-induced CLF changes accounting for meteorological covariations. The hypothesis of this study is that the response of cloud fraction of MBLCs to aerosol perturbations is positive but buffered, i.e. reduced or amplified, by ambient meteorology and that both the sensitivities and the interactions with meteorological factors have distinct regional patterns.

2 Data and methods

2.1 Data sets

This work combines 9 years (2011–2019) of satellite retrievals from Moderate Resolution Imaging Spectroradiometer (MODIS) and reanalysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) from 60° N to 60° S. In this study, MBLCs are defined as single-layer warm cloud fields with cloud top temperatures higher than 268 K. To achieve this, the information on CLF (Cloud_Retrieval_Fraction_1L_Liquid product), r_e (Cloud_Effective_Radius_1L_Liquid_Mean product), cloud optical depth (τ_c ; Cloud_Optical_Thickness_1L_Liquid_Mean product), cloud top temperature (CTT; Cloud_Top_Temperature_Mean product), and satellite viewing geometry are obtained from MODIS level-3 collection-6.1 atmosphere daily products on the Terra platform (MOD08_D3), which are gridded into $1^\circ \times 1^\circ$ globally from level-2 atmospheric products. CLF serves as the predictand in this study. The computation of N_d relies on τ_c and r_e , with filtering criteria based on CTT, solar zenith viewing angle, and satellite zenith angle, as elaborated in the following.

The equation used to calculate the MODIS N_d is from Quaas et al. (2006) depends on the retrievals of r_e and τ_c and so do the uncertainties in the errors propagated from r_e

and τ_c :

$$N_d = \alpha \tau_c^{0.5} r_e^{-2.5}, \quad (1)$$

where $\alpha = 1.37 \times 10^{-5} \text{ m}^{-0.5}$ is a constant related to adiabatic growth rate. The uncertainties in N_d retrievals are exhaustively evaluated by Grosvenor et al. (2018), which suggests that the uncertainties in averaged N_d over a $1^\circ \times 1^\circ$ grid box (spatial resolution of the MODIS products used in this study) decrease by over 50 % compared to pixel-level uncertainties. This derivation approach relies on the assumed adiabaticity in global marine warm clouds where liquid water content and r_e increase monotonically and N_d is distributed as constant vertically. Departure from the adiabatic assumption (e.g. due to entrainment) would result in N_d retrieval biases (Merk et al., 2016; Bennartz and Rausch, 2017). The uncertainty related to the estimation of N_d from MODIS also depends on liquid CLF. N_d is less biased in the regions of larger CLF, where clouds are more homogeneous, while in the regions with lower CLF N_d retrievals are sparser and less reliable (Grosvenor et al., 2018; Zhu et al., 2018). In such heterogeneous cloud fields, subpixel effects in the retrieval of r_e can negatively bias the retrieved N_d values (Zhang and Platnick, 2011; Zhang et al., 2012; Grosvenor et al., 2018). Such retrieval biases could cause a bias in the N_d –CLF relationship as well. Furthermore, the interpretation of the causal effect of N_d on CLF can also be obscured by small-scale sampling issues. In particular, apart from the retrieval errors in r_e and τ_c , the natural spatial variability in cloud fields can also propagate to the N_d estimate and distort the N_d –CLF relationship (Arola et al., 2022; Liu et al., 2024).

Following the screening criteria for a more reliable N_d demarcated by Gryspeerd et al. (2022), only clouds restricted to a single layer in the liquid phase with a CTT higher than 268 K are considered. As suggested by Quaas et al. (2006), samples with $r_e < 4 \mu\text{m}$ and $\tau_c < 4$ are excluded to cope with the high r_e retrieval uncertainties at low τ_c . In addition, solar and sensor viewing zenith angles respectively greater than 65° and 55° are removed to avoid the large biases in r_e and τ_c retrievals (as in Grosvenor et al., 2018). The pixels selected according to the above sampling strategies generate more reliable N_d estimates.

Atmospheric and oceanic variables are taken from the fifth-generation ECMWF atmospheric reanalysis of the global climate (ERA5) at an hourly frequency (Table 1) (Hersbach et al., 2020). The ERA5 data sets are harmonized to fit the level-3 MODIS data by first being resampled to $1^\circ \times 1^\circ$ from their default $0.25^\circ \times 0.25^\circ$ spatial resolution using bilinear interpolation; they are subsequently collocated with Terra MODIS by extracting hourly data to align with the UTC overpass times of the Terra satellite for each grid cell, yielding a spatiotemporally matched MODIS–ERA5 combined data set for training the ML models. For N_d retrievals, only samples within 1st–99th percentiles are retained to exclude potential unrealistic outliers from r_e and

τ_c retrievals (Zipfel et al., 2022). Furthermore, the explanation of ML models in this study relies on using linear regressions to capture the distribution of individual prediction instances, and the extreme values may excessively magnify or reduce the sensitivity or interactive effects quantified by SHAP (shown in Fig. 1 and discussed in Sect. 2.3.2). The threshold of 1st–99th percentiles for each predictor is thus adopted to remove the values at the very tails of the specific distribution and to improve the robustness of the estimated sensitivities. To define the sensitivities of CLF and the interactive effects of meteorological factors, the natural logarithm of N_d is taken (see Sect. 2.3.2 in detail). Estimated inversion strength (EIS) is calculated based on the formulation from Wood and Bretherton (2006), and in this study, it is dependent only on atmospheric temperatures at 700 hPa and at the level of 1000 hPa.

All input predictors for each Extreme Gradient Boosting (XGB) model (i.e. for each $5^\circ \times 5^\circ$ window aggregated from $1^\circ \times 1^\circ$ grid boxes, as detailed in Sect. 2.2) are standardized by centring around the mean and scaling to have unit variance as in Scott et al. (2020). Hamby (1994) suggested that the standardization process is a standard practice when aiming for comparability of sensitivity estimates across predictors. This process eliminates the influence of units and aligns data on the same scale instead of the original natural ones, thereby ensuring the comparability of the quantified sensitivities and interactive effects with meteorology among different variables. This standardization procedure has been applied in other studies investigating different cloud sensitivities to various cloud-controlling factors (e.g. Ceppi and Nowack, 2021; Andersen et al., 2023). This procedure, however, may result in reduced spatial comparability due to variations in mean and standard deviation values across different $5^\circ \times 5^\circ$ windows. To assess the trade-off between comparability among different predictors and comparability in space, we provide results without standardization in the Supplement (Figs. S2 to S7 therein) as done by Grise and Kelleher (2021). In terms of spatial patterns, the results are nearly identical to their corresponding ones presented in the following sections of the main text, suggesting that standardizing the data based on the local mean and standard deviation for each window has only a small impact on comparability across each window. Therefore, we primarily benefit from achieving comparability among different predictors while making only a minor compromise in spatial comparability.

2.2 Machine learning model setup

Extreme Gradient Boosting (XGB) is a distributed tree boosting algorithm aiming to provide a scalable, portable, and flexible library under the gradient boosting framework (Chen and Guestrin, 2016). The state-of-the-art XGB algorithm can be implemented efficiently in Python and has been recently used to study clouds and ACI (Andersen et al., 2022; Douglas and L'Ecuyer, 2022). As an extension of previous gradi-

ent boosting methods, XGB has incorporated regularization techniques which help prevent overfitting and improve model generalization. Besides, the subsampling on training subsets and column (feature) subsampling techniques can shorten the running time and also avert overfitting and hence elevate model performance (Chen and Guestrin, 2016). Relevant regularization and subsampling hyperparameters are tuned using Bayesian optimization to determine the best combination; see Table 2 for the search space.

Data from 2011 to 2016 are used for training and data from 2017 to 2019 for testing (ratio of independent train to test split of about 67 % / 33 %). By chronologically splitting the training and test sets without random shuffling, we ensure that the training data does not see future information and the autocorrelation in data does not lead to overoptimistic evaluation of the model's performance (Beucler et al., 2023; Kapoor et al., 2023). As suggested by Karpatne et al. (2017), a single ML model may not perform well across all regions due to the heterogeneity of relevant processes. Therefore, data at a $1^\circ \times 1^\circ$ spatial resolution are aggregated into $5^\circ \times 5^\circ$ geographical windows, where an individual independent XGB model is trained and tested for each “window”. Hereby, a region-specific ML framework is established to potentially capture regional relationships and characteristics and thus the regional patterns of CLF adjustment. The coarser $5^\circ \times 5^\circ$ spatial resolution of the modelling grid increases the sample size by a factor of ≈ 25 , which is helpful to establish robust sensitivity estimates. In addition, at the spatial resolution of $1^\circ \times 1^\circ$ summarized in $5^\circ \times 5^\circ$ windows, the spatial scale is adequate for ACI sensitivity estimation (Grandey and Stier, 2010). To ensure a sufficient data amount for training and testing the XGB models, only the geographical windows with over 6000 available data points are retained. Consequently, 34 out of 1190 oceanic windows have been excluded. These windows located between 47.5° W and 122.5° E and 52.5° and 57.5° S in the Southern Ocean (Fig. 2) contain fewer than 6000 valid samples due to the screening for N_d retrievals. For each model, the hyperparameters are tuned by implementing Bayesian optimization, which uses a Gaussian process prior distribution over hyperparameters to initialize a probabilistic model for the objective function to be optimized. After the initialization, the probabilistic model is updated iteratively, and Bayesian optimization suggests the optimal combination of hyperparameters to try for the next iteration according to the previous one and samples gathered from the search space (Table 2) (Snoek et al., 2012). Each iteration is evaluated by five-fold cross-validation using the root mean square error (RMSE) as score. The number of boosting rounds (the number of trees) for each XGB model is then determined by the early stopping technique to further avoid overfitting; i.e. the training of the model stops early once it is monitored, so the score of cross-validation does not improve within 20 iteration rounds.

Table 1. Summary of the predictors from ERA5 reanalysis.

Predictor name	Abbreviation	Unit
Instantaneous pressure-level parameters (at 700 hPa, 850 hPa)		
Relative humidity	RH ₇₀₀ , RH ₈₅₀	%
Specific humidity	SH ₇₀₀ , SH ₈₅₀	kg kg ⁻¹
Temperature	<i>t</i> ₇₀₀ , <i>t</i> ₈₅₀	K
Vertical velocity	ω_{700} , ω_{850}	Pa s ⁻¹
Eastward wind component	<i>u</i> ₇₀₀ , <i>u</i> ₈₅₀	m s ⁻¹
Northward wind component	<i>v</i> ₇₀₀ , <i>v</i> ₈₅₀	m s ⁻¹
Surface and single-level parameters (instantaneous or mean rates/fluxes)		
Eastward and northward wind component at 10 m	<i>u</i> ₁₀ , <i>v</i> ₁₀	m s ⁻¹
Boundary-layer height	BLH	m
Convective available potential energy	CAPE	J kg ⁻¹
Sea surface temperature	SST	K
Total column water vapour	TCWV	kg m ⁻²
Mean large-scale precipitation fraction	PF	proportion
Mean surface sensible/latent heat flux	SHF/LHF	W m ⁻²
Calculated		
Estimated inversion strength	EIS	K

Table 2. Overview of the hyperparameters tuned for regional Extreme Gradient Boosting models using Bayesian optimization.

Hyperparameter name	Search space
learning_rate	0.01–0.5
max_depth	3–10
min_child_weight	1–10
subsample	0.5–1
colsample_bytree	0.5–1
gamma	0–10
alpha	0–10
lambda	0–10

2.3 Explaining the machine learning models

2.3.1 SHapley Additive exPlanation (SHAP) values

SHAP values were proposed by Lundberg and Lee (2017) on the basis of cooperative game theory to explain the outputs of ML models. The SHAP approach has been implemented with XGB in Python, and it has been reported that outputs from XGB models with various number of trees can be well explained by the SHAP framework in different subject areas (e.g. Padarian et al., 2020; Lundberg et al., 2018, 2020; Kim et al., 2021; Li et al., 2022). The contribution of a predictor value to a specific model prediction is calculated as the difference between the predictions of the model in the presence and absence of this particular predictor for all possible combinations of predictor values. Since this is performed at a “local” level (i.e. for this specific instance’s prediction),

it allows for insights into how a certain model outcome is achieved, thereby complementing more traditional “global” (considering all instances) feature importance measures (e.g. partial dependence plot).

The base value in the context of SHAP values is what would be predicted in the absence of any feature information (Lundberg and Lee, 2017), and it is typically computed as the average of all predictions by ML models over the entire training data set. Positive (negative) SHAP values indicate that the specific feature value increases (decreases) the prediction compared to this base value. In other words, the base value serves as the reference point against which the contributions of individual features are measured. SHAP values for all features always sum up to the difference between the base value and the final model prediction so that SHAP values are additive and internally consistent. The base value could be analogous to the climatological CLF for a given geographical window, assuming no information about the input parameters is known. In this context, the SHAP values of input features indicate the extent to which knowing information about each feature value would deviate the prediction from the climatological CLF (base value).

Furthermore, the quantification of the influence of meteorology on the N_d –CLF relationship can be analysed using SHAP interaction values, which are an extension of SHAP values. They measure the difference between the SHAP values for a feature when another (secondary) feature is included versus when it is not included, offering a potential tool for insights into feature interactions captured by the tree ensembles. SHAP values have been applied to study at-

mospheric aerosols in the context of air pollution Stirnberg et al. (2021) and have been used by Zipfel et al. (2022) to explore satellite-observed N_d –LWP relationship in MBLCs in the southeast Atlantic, finding that meteorological variables have considerable influences on the N_d –LWP relationship using SHAP interactive values. Moreover, the use of SHAP interaction values in these studies allows for a more profound and in-depth comprehension of the underlying processes with respect to local meteorology. SHAP values provide insights into the behaviour of the XGB models, and as all statistical/ML models, they may not necessarily reflect real-world physical causality. Nevertheless, this state-of-the-art technique allows us to account for meteorological covariations when deriving sensitivities and to appraise to what extent the meteorological predictors interact with and influence the N_d –CLF relationship beyond traditional global-level feature attributions.

2.3.2 Quantification of sensitivities and interactive effects

Figure 1 is an exemplary graph for a regional XGB model at a specific $5^\circ \times 5^\circ$ window (27.5 – 32.5° S, 122.5 – 127.5° W). SHAP values and SHAP interaction values are used to explain this XGB model and to quantify and isolate the CLF sensitivity to N_d and the interactive effects of meteorological factors (here sea-surface temperature, SST). Each dot in Fig. 1 represents an individual data instance (i.e. a single observation at a specific grid cell and time step) and shows how individual N_d or $\ln N_d$ values impact the CLF prediction.

Plotting SHAP values of N_d against N_d values without the standardization process (Fig. 1a) for each data sample illustrates that increased N_d values lead to an increase in the predicted CLF, while the rate of the increase ($d\text{SHAP}/dN_d$) drops with N_d as shown by the orange line. For each 20 cm^{-3} wide bin of N_d , $d\text{SHAP}/dN_d$ is calculated as the slope of the linear regression between N_d and N_d SHAP values. The non-linear positive association between N_d and predicted CLF aligns well with findings of prior studies (e.g. Gryspeerd et al., 2016; Rosenfeld et al., 2019) that the aerosol impact on CLF saturates at relatively high aerosol loading. This relationship also resembles the one reported by Yuan et al. (2023), which is attributed to the precipitation suppression effect due to a relatively high N_d .

Expressing the sensitivity logarithmically in N_d is ideal because cloud processes are prone to respond to a relative change in N_d rather than an absolute one (Carslaw et al., 2013; Bellouin et al., 2020). Furthermore, the log-transformed N_d facilitates the application of simple linear regressions to capture the relationship between the contribution of N_d and the predicted CLF (N_d SHAP values) and its feature values. As depicted in Fig. 1b, the contribution of $\ln N_d$ to the predicted CLF increases almost linearly with a rising $\ln N_d$. Thus, the CLF sensitivity to N_d is estimated as the slope of the linear regression between $\ln N_d$ SHAP values

and $\ln N_d$ values ($0.098\text{ CLF } \sigma^{-1}$). A similar method to estimate sensitivity has also been used by Li et al. (2022), where it is also suggested that this method can enhance the robustness of the sensitivity estimation. Because it can leverage the benefits of an XGB model, including bagging techniques and no need for distribution assumptions, along with the advantages of SHAP, which provides global interpretations consistent with local explanations (Lundberg et al., 2020; Molnar, 2022). It should be noted that the notably linear relationship in Fig. 1b does not hold across all geographical windows. Figure S1 displays additional exemplary windows where the relationships exhibit less linearity. Our approach also captures non-linearity in the system; in these cases, the linear regression helps decrease the convolved relationships as in Gryspeerd et al. (2016). Note that unlike N_d (cm^{-3}) in panel (a), $\ln N_d$ and SST in (b) and (c) have been standardized, and thus sensitivities and interaction indices (IAIs) are expressed with the unit of cloud fraction change per standard deviation ($\text{CLF } \sigma^{-1}$). Standardizing all predictors ensures that the results become comparable across all of them. We also present the SHAP dependence plots for the same example window in Fig. S2 where non-standardized $\ln N_d$ and SST are used to plot panels (b) and (c). The patterns are alike and only the magnitudes of the example sensitivity and IAI are different because they are no longer expressed on a physical scale.

The vertical dispersion around the $\ln N_d$ –CLF relationship captured by the SHAP dependence plot is due to the dependence of the $\ln N_d$ contribution to the predicted CLF on meteorological factors (e.g. SST) in the model, which is captured by SHAP interaction values, as displayed in Fig. 1c. The colouring of the data points by SST illustrates how interactions with SST split up the $\ln N_d$ –CLF relationship, with low SST values amplifying the $\ln N_d$ contribution and vice versa. To quantify this interaction effect, the meteorological data are then divided into a group of above-average feature values and a group of below-average feature values. A linear regression is fit to the $\ln N_d$ values and the SHAP interaction values in each group. An interaction index (IAI) is derived from these regression fits and defined as the slope for the high-value group ($> \text{mean}$) with the slope for the low-value group ($< \text{mean}$) subtracted:

$$\text{IAI} = \beta_{x,\text{high}} - \beta_{x,\text{low}}, \quad (2)$$

where β is the slope of the linear regression between SHAP interaction values and $\ln N_d$ values and the subscripts denote the high-value group and the low-value group for a specific meteorological variable x (SST in the example) respectively. At the exemplary geographical window, the influence of SST on the N_d –CLF sensitivity is quantified by $\text{IAI} = -0.029\text{ CLF } \sigma^{-1}$ (Fig. 1c). Similar to sensitivities, the unit of IAIs is also $\text{CLF } \sigma^{-1}$. Therefore, for a positive sensitivity such as the N_d –CLF sensitivity shown in Fig. 1b, a negative IAI value means that the N_d –CLF sensitivity is

larger with low feature values, as shown in Fig. 1c (the positive relationship is weakened by high SST values). On the contrary, a positive IAI value corresponds to a larger positive sensitivity with high feature values.

2.3.3 Limitations of observation-based machine learning of aerosol-cloud processes

In this section, limitations of this study are discussed. A fundamental limitation of our study is that the assertion of causality from the statistical relationships of aerosols/ N_d and cloud fraction/properties is not easily done. While causal inference approaches exist and have been applied in the field of aerosol–cloud interactions (Fons et al., 2023), we employ a more traditional approach of analysing statistical relationships of instantaneous observations (i.e. correlations). Unless nonetheless explicitly incorporating such causal inference approaches, studies utilizing statistical or ML models to explore observational aerosol–cloud processes contend with this common limitation. For instance, some studies assessed satellite-based statistical relationships between CLF and N_d (Christensen et al., 2016, 2017), between LWP and N_d (Michibata et al., 2016; Rosenfeld et al., 2019), and between N_d and other aerosol proxies (Gryspeerdt et al., 2017; McCoy et al., 2017a), all resting on statistically inferring sensitivities of cloud quantities to aerosol proxies (Forster et al., 2021). While we interpret the derived relationships with respect to the known physical relationships, uncertainties regarding the physical interpretation are mainly driven by two sources: uncertainties in the data and uncertainties from the methods.

1. *Data.* Uncertainties exist for each satellite/reanalysis quantity, but may be particularly large in N_d . For example, the subpixel effect can introduce more bias in the N_d retrieval process within broken-cloud regimes due to increased heterogeneity. The N_d retrieval biases are discussed in Sect. 2.1. Also, N_d and CLF observations are not fully independent, which may introduce a spurious positive correlation between the two variables. As such, we expect the physical relationship of N_d and CLF to be weaker than our estimate so that the derived sensitivities present an upper bound of the physical relationship.

Another caveat in our data is that N_d values in our study are computed using MODIS level-3 large-scale mean r_e and τ_c values instead of joint histograms as in Gryspeerdt et al. (2016). This may introduce additional biases considering the non-linearity of the N_d calculation. In future work, N_d data calculated from underlying joint histograms or pre-filtered data by Gryspeerdt et al. (2022) could be applied to be compared with the results in this study.

2. *Methods.*

- a. The exact quantification of sensitivities is dependent on the choice of the statistical/machine learning model. While for (more linearly related) monthly data, Andersen et al. (2022) have shown that XGB, artificial neural networks, and linear models tend to lead to very similar results, this is not expected for more instantaneous data. Here, non-linear relationships are expected, and a more complex non-linear model is a more appropriate choice. XGB and other tree ensemble methods are a particularly popular choice because of their interpretability, high accuracy considering computational efficiency (Lundberg et al., 2020), and ability to model the interactions between predictors (Elith et al., 2008). They have been frequently used to study aerosols and clouds in the past (Fuchs et al., 2018; Dadashazar et al., 2021; Andersen et al., 2021; Y. Chen et al., 2022; Bender et al., 2024). Besides, the Tree SHAP algorithm, specifically tailored for tree-based models to compute exact Shapley values, can even further enhance their interpretability and has been applied in this field as well (Stirnberg et al., 2021; Zipfel et al., 2022).
- b. The quantification of sensitivities with SHAP values depends on details: the choice of the algorithm to effectively estimate Shapley values is application-specific and comes to the trade-off between being true to the data and true to the model, which relies on an observational and interventional conditional expectation respectively (Chen et al., 2020). The true to the model approach is preferable when trying to understand how an ML model makes a prediction, which requires assuming feature independence. In this study, we focus on potential mechanisms behind CLF sensitivities, and thus we tend to respect the correlations spread among input features (true to the data) (Frye et al., 2021; Chen et al., 2022). Consequently, we suffer from the disadvantage of being true to the data: entangled importance attributions of correlated features, e.g. a feature not explicitly used by the model for the prediction task, might be assigned a non-zero contribution. Yet we refrain from the drawback of being true to the model – unrealistic input instances (Sundararajan and Najmi, 2020; Linardatos et al., 2021; H. Chen et al., 2023). Despite the inherent trade-off, SHAP approach has been employed in the context of being true to the data (e.g. Stirnberg et al., 2021; Zipfel et al., 2022; Li et al., 2022).

The derived estimates of sensitivities and interactive effects in this paper should thus be interpreted with these limitations and uncertainties in mind.

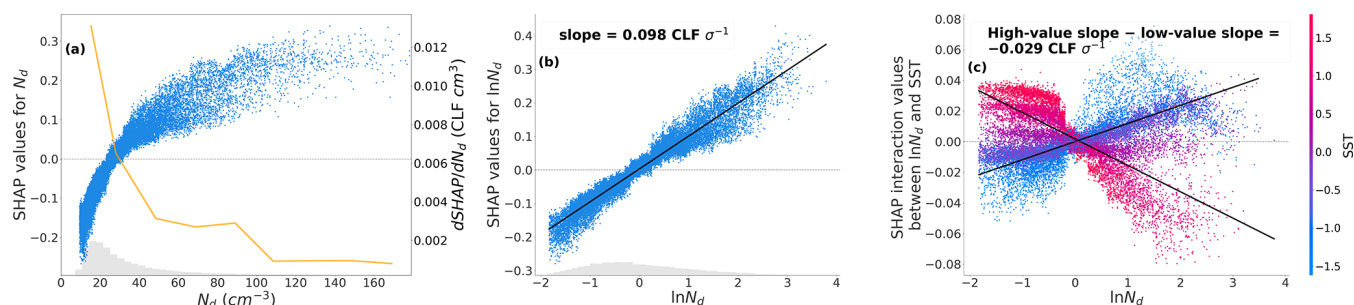


Figure 1. SHAP dependence plots for the cloud-droplet number concentration (N_d) in the region from 27.5 to 32.5° S and from 122.5 to 127.5° W. **(a)** Dots show N_d SHAP values versus N_d values. The orange line shows the change rate of N_d SHAP values with respect to N_d ($d\text{SHAP}/dN_d$) versus N_d values for each N_d bin of 20 cm^{-3} wide. Panel **(b)** is similar to panel **(a)** but shows the relationship between $\ln N_d$ SHAP values and $\ln N_d$ with the corresponding sensitivity defined as the slope of the linear regression. Panel **(c)** shows SHAP interaction values coloured by sea-surface temperature (SST) showing the dependence of $\ln N_d$ –CLF relationship on the interactive effects of SST. The interaction values are further divided into two groups by the mean feature value of SST. Linear regressions are performed respectively for the high-value group and low-value group and the interaction index (IAI) is defined as the slope for the high-value group by subtracting the slope for the low-value group. The horizontal dashed lines are a demarcation between negative and positive SHAP (interaction) values. Note that N_d in **(a)** is not standardized, while $\ln N_d$ and SST in **(b)** and **(c)** are standardized.

3 Results and discussion

3.1 Model performance

The skills of the region-specific XGB models in predicting CLF are evaluated by the coefficient of determination (R^2) on the unseen hold-out test data. The global weighted mean R^2 is 0.45 (about 45 % on weighted average and up to 73.57 % of the variability in CLF prediction is explained) and the standard deviation 0.10. While this means that, on average, about half of the variability in CLF cannot be explained by the machine learning models, this is expected as previous studies have shown that the performance of statistical models decreases when going from monthly to daily data (Andersen et al., 2017; Fuchs et al., 2018; Dadashazar et al., 2021), and the performance is on par with that reported by Dadashazar et al. (2021), who used machine learning models to predict N_d with daily reanalysis data. The models in tropical regions in the Indian Ocean and the western Pacific relatively poorly explain the variability in CLF, while XGB models perform well in the stratocumulus regions in the subtropics near the continents and in the midlatitudes, particularly the Southern Hemispheric midlatitudes. The high skill of predicting CLF in the Southern Hemispheric midlatitudes is in contrast to a recent study where this region has been found to be particularly difficult to model statistically with monthly data (Andersen et al., 2023). In this region, the day-to-day CLF variability is high due to the large influence of synoptic-scale weather systems, and hence data at the daily resolution are more adequate to represent the CLF variability in these regions.

3.2 CLF sensitivity: global perspectives and regional characteristics

3.2.1 Global overview of CLF sensitivities

Figure 3 summarizes the means and distributions of the near-global sensitivities of CLF to all predictors. The sensitivities are estimated as described in Sect. 2.3.2. The sequence is sorted by descending mean values of the absolute sensitivities (i.e. by feature importance) of the predictor variables. A strong and consistently positive N_d –CLF sensitivity is found. The fact that CLF is the most sensitive to N_d is to be expected, as cloud observations from the same sensor are more directly related than a reanalysis product, so their overall magnitude should not be compared (Zipfel et al., 2022). The entrainment of relatively dry air from the free troposphere into the MBL is impeded by a stronger inversion (i.e. higher EIS), resulting in a shallower, better-mixed, and more humid MBL conducive to stratocumulus clouds (Bretherton and Wyant, 1997; Wood and Hartmann, 2006; Qu et al., 2015a; Myers et al., 2021). The salient positive sensitivity to EIS is in accordance with the links found in previous studies (e.g. Klein and Hartmann, 1993; Qu et al., 2015b; Andersen et al., 2017), suggesting that EIS is a crucial controlling factor for low marine cloud cover. Note that in some studies, the strength of the inversion over the boundary layer is measured by lower tropospheric stability, which can be regarded as a similar metric outperformed by EIS (Wood and Bretherton, 2006). Precipitation fraction is the fraction of the original ERA5 grid box covered by large-scale precipitation. The strong positive CLF sensitivity to precipitation fraction is likely caused by the ML model learning that precipitation can be viewed as a proxy for cloudiness rather than being an indicator of the physical processes via which precipitation exerts controls on the macrophysics of MBLs. Humidity shows

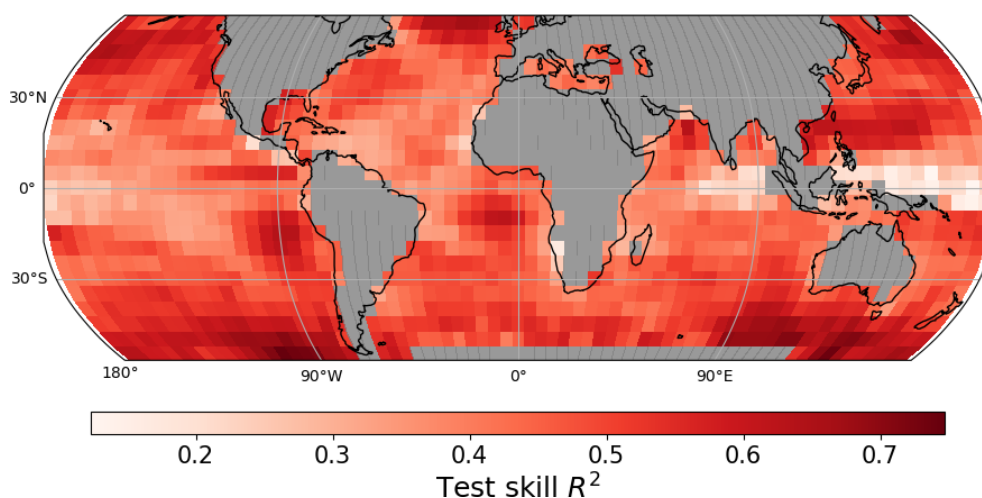


Figure 2. R^2 score of regional Extreme Gradient Boosting models predicting the cloud fraction of marine boundary-layer clouds in the independent test data set (2017–2019).

positive CLF sensitivities greater at 850 hPa, where cloud tops are often located (Gryspeerdt and Stier, 2012), than at 700 hPa, which is typically in the free troposphere above the MBLs (Myers and Norris, 2013). Likewise, the atmospheric temperature at 850 hPa (t_{850}) presents stronger CLF sensitivity than the temperature at 700 hPa (t_{700}). Nonetheless, in the case of winds the 700 hPa pressure level is more relevant than that at 850 hPa. A relatively pronounced negative sensitivity to the eastward wind component at 700 hPa (u_{700}) seems to indicate that clouds are depleted due to more westerlies at this level. CLF exhibits negative sensitivities to vertical pressure velocities at both 850 and 700 hPa, showing that large-scale ascending motion is connected to increases in MBLs (Myers and Norris, 2013; Bretherton et al., 2013; Blosssey et al., 2013). In general, the global averages of CLF sensitivity in terms of dynamical predictors (i.e. 3D winds at surface and pressure levels) vary in sign and are less strong. A marked negative sensitivity of CLF to SST is found, which is in agreement with many prior studies (e.g. Qu et al., 2015b; Scott et al., 2020), where increases in SST have been found to lead to low cloud breakup and dissipation due to a number of processes as described in, for example, Scott et al. (2020). One of these is that the associated enhancement of mean surface latent heat flux (LHF) deepens MBL and facilitates buoyancy and thus the entrainment of dry free-tropospheric air (Rieck et al., 2012; Andersen et al., 2022). However, CLF is much less sensitive to LHF than to SST, which may indicate that this mechanism is less important at the spatial scale and timescale considered in this study. CLF exhibits a considerable negative sensitivity to mean surface sensible heat flux (SHF), which quantifies an increase in CLF with increasing SHF (upward SHF is negative). While increased SHF can promote the transition from decks of stratus or stratocumulus clouds (high CLF) to more convective clouds (low CLF) due to the deepening of the boundary layer (Fan et al.,

2016), potentially leading to a positive SHF–CLF relationship, increased SHF is associated with situations of cold air advection where turbulent surface fluxes are enhanced, which could lead to marked increases in CLF (Miyamoto et al., 2018; Zelinka et al., 2018; Grise and Kelleher, 2021).

3.2.2 Spatial patterns of the CLF sensitivity to N_d

The sensitivity of the MBLC fraction associated with the aerosol proxy, N_d , is ubiquitously positive in accordance with the global correlations or sensitivities found in, for example, Gryspeerdt et al. (2016) and Andersen et al. (2017). This is presumably due to the lifetime effect but could also partially result from N_d retrieval biases discussed in Sect. 2.1. The global weighted mean value of the N_d –CLF sensitivity is $0.074 \text{ CLF } \sigma^{-1}$, with a standard deviation of $0.036 \text{ CLF } \sigma^{-1}$. The relationship between CLF and N_d is found to be particularly strong in the regions of frequent stratocumulus-to-cumulus transition off the western continental coasts. These marked positive N_d –CLF sensitivities may be caused by high N_d , delaying the transition from stratocumulus to cumulus clouds (Gryspeerdt et al., 2016; Christensen et al., 2020). However, as this cloud regime transition involves clouds shifting from more overcast to more broken, the strong relationships in these regions may be more affected by N_d retrieval errors. The N_d –CLF sensitivity is also pronounced in the Southern Hemispheric midlatitudes, where stratiform clouds dominate. The N_d –CLF sensitivity is weak and close to zero in the tropics, in particular in the deep convective warm-pool region. These spatial patterns of N_d –CLF sensitivity resemble those found by Gryspeerdt et al. (2016), in particular the ones where they mediated the aerosol optical depth–CLF relationship by N_d but are more pronounced in the Southern Hemispheric midlatitudes. This difference in estimated sensitivity seems note-

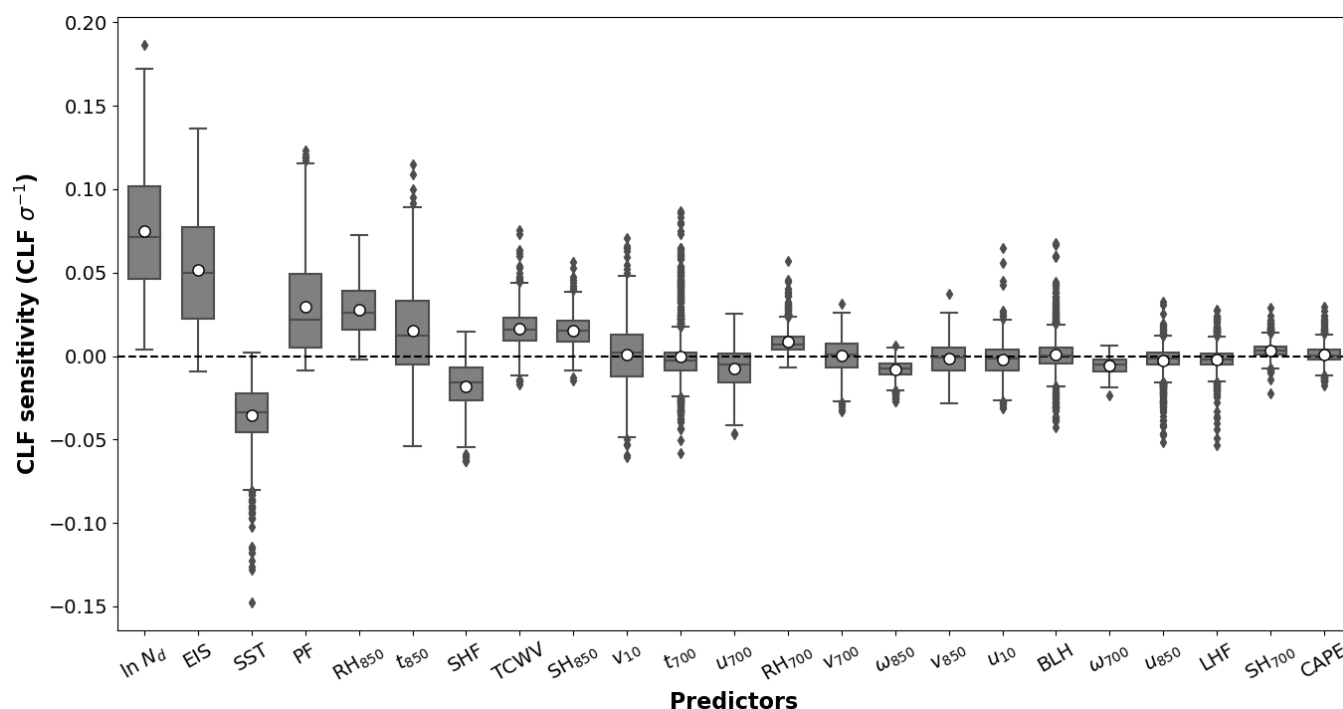


Figure 3. The distribution of the sensitivities of the cloud fraction to all predictors as depicted in Table 1. Boxes represent the interquartile range, which is extended by whiskers to up to 1.5 interquartile ranges, with outliers shown as points outside the range. The solid line and white dot in each box show the median and mean values of the sensitivities respectively. Predictors are sorted by the mean values of absolute sensitivity values. The dashed line across the figure separates positive and negative sensitivity values.

worthy and should thus be investigated in future work. As N_d retrievals tend to negatively bias at lower CLF and positively bias at higher CLF, the N_d –CLF sensitivity may be overestimated and, at the scales considered here, should be interpreted as an upper bound to the physical N_d –CLF sensitivity. The global weighted average of the CLF– $\ln N_d$ sensitivity without standardization is 0.112 (unitless), and its spatial pattern is shown in Fig. S4. This value is higher than the upper bound of 0.1 reported by Bellouin et al. (2020), which is based on global climate models and large-eddy simulations. This may be partly due to the aforementioned bias. However, it is important to note that our non-standardized CLF– N_d sensitivity, shown in Fig. 1a, closely mirrors that from Yuan et al. (2023), with a similar range. In addition, the high $\ln \text{CLF}$ – $\ln N_d$ values estimated in Y. Chen et al. (2022) and Chen et al. (2024) suggest that values exceeding the upper bound of 0.1 might be plausible. These recent observational studies, including quantifying cloud fraction adjustment based on ship tracks (Yuan et al., 2023), volcano aerosol perturbations (Y. Chen et al., 2022; Chen et al., 2024), and our SHAP approach using global satellite observations, indicate that the 0.1 upper bound may be extended. In future work, estimating a radiative forcing using the SHAP-based sensitivities will make our study more comparable with other research on cloud fraction adjustment.

3.2.3 Spatial patterns of the CLF sensitivity to thermodynamical drivers

There has been a strong consensus that EIS and SST are the two important determinants of cloud fraction of marine boundary clouds and their corresponding radiative effects across different geographical regions and on varying timescales (e.g. Bretherton, 2015; Myers and Norris, 2015; McCoy et al., 2017b; Wall et al., 2017). Stronger inversions capping MBL (i.e. higher EIS) will hamper the entrainment of aloft dry air from the troposphere and thus lead to a shallower MBL and more moisture trapped within MBL, promoting the development and maintenance of low-level clouds (Andersen et al., 2017). The regional EIS–CLF sensitivity patterns (Fig. 5a) show that low marine cloud fraction increases ubiquitously in response to stronger EIS, in particular in the tropical and subtropical stratocumulus-capped regions and within the midlatitudes. The sensitivity pattern is in good agreement with that found by Scott et al. (2020) and Andersen et al. (2023), related studies at different timescales (Grise and Medeiros, 2016; Kelleher and Grise, 2019; de Szoeke et al., 2016).

MBLC cover reduces globally in response to increased SST, particularly pronounced in the stratocumulus regions over eastern oceanic basins (Fig. 5b), consistent well with (Scott et al., 2020). SST can favour MBLC dissipation through increasing surface latent heat fluxes and deepening

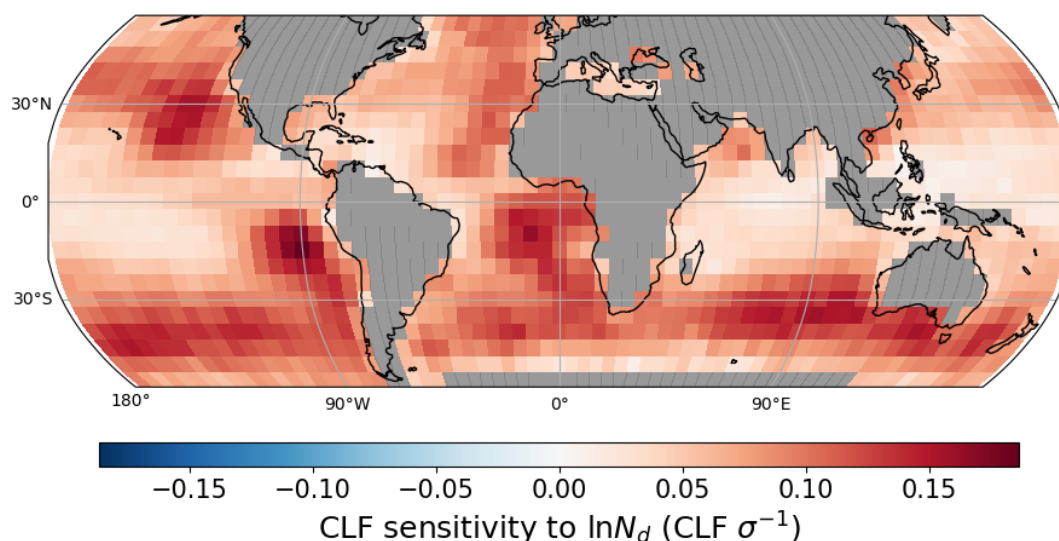


Figure 4. Sensitivity of the marine boundary-layer cloud fraction to $\ln N_d$.

ing MBL, facilitating dry entrainment and eventually desiccating the MBL and clouds (Rieck et al., 2012; Qu et al., 2015b). Yet as stated in Sect. 3.2.1, the weak CLF sensitivity to LHF in relation to the strong sensitivity to SST may imply that the other process makes more substantial contributions – namely, that the higher moisture gradient between the troposphere and MBL arising from the increased SST makes the entrained air more efficient in evaporating cloud water (van der Dussen et al., 2015; Qu et al., 2015b). This process has been shown to be the driving mechanism for the observed reduction in marine low cloud cover near the coast of Baja California (Andersen et al., 2022).

Figure 5c shows that low marine cloud fraction increases with negative (upward) SHF most markedly in the stratocumulus regions. CLF can increase in response to increased surface fluxes in situations of cold advection (Zelinka et al., 2018). Over the south Indian Ocean, a marked SHF–CLF sensitivity is also found. Here, enhancements of SHF due to the subtropical anticyclone and midlatitude storm-track activity have been found to increase CLF (Miyamoto et al., 2018). The results may be a hint that the increase in CLF presumably due to increased SHF (e.g. due to cold advection) outweighs the influence of SHF on CLF by controlling the transition from marine stratocumulus to open-cellular marine clouds (Kazil et al., 2014; Fan et al., 2016) in the core stratocumulus regions. Consequently, the SHF–CLF sensitivity is less pronounced in regions of frequent closed- to open-cell and cumulus transitions. Relative humidity at 850 hPa (RH_{850}) is positively related to marine low liquid cloud fraction across the globe. The positive sensitivity is particularly strong in the trade cumulus regions, where the 850 hPa level is representative of the boundary layer. In the coastal stratocumulus regions, clouds are frequently below this level

(Adebiyi and Zuidema, 2016), so that clouds are not as sensitive to variability in RH at that level.

3.2.4 Spatial patterns of the CLF sensitivity to dynamical drivers

Large-scale circulations and dynamical conditions play an essential role in controlling cloud fraction and the indirect effects of aerosols (Su et al., 2010; Small et al., 2011). The large-scale dynamics are represented by the horizontal and vertical winds at 700 and 850 hPa, which display clear and distinct regional patterns of CLF sensitivity (Fig. 6). It can also be seen that at the considered scales and pressure levels, horizontal wind vectors have stronger CLF sensitivities than large-scale vertical motion. There is a coherent pattern of negative CLF sensitivity to the zonal wind at 700 hPa in the stratocumulus-dominated regions (also apparent at 850 hPa), and the Southern Hemispheric midlatitudes, indicating a decrease in MBLCs with westerly anomalies at this pressure level. Recently, a study using monthly data has also found a similar sensitivity pattern of stratocumulus clouds to zonal wind at 700 hPa, finding that the reduced CLF is related to increased vertical wind shear (as the boundary-layer flow is easterly), leading to increased turbulence and dry-air entrainment (Andersen et al., 2023). However, using monthly data, Andersen et al. (2023) did not find a similar CLF sensitivity to zonal winds in the Southern Hemispheric midlatitudes. As the CLF sensitivity to u_{700} in the Southern Hemispheric midlatitudes is only apparent using daily data and only at 700 hPa, it seems likely that it is related to synoptic variability that drives day-to-day variability in MBLCs in this region (Kelleher and Grise, 2019). Positive CLF sensitivities to u_{700} (higher CLF with westerly anomalies) and, to a lesser degree, u_{850} are found off the eastern Asian and North Amer-

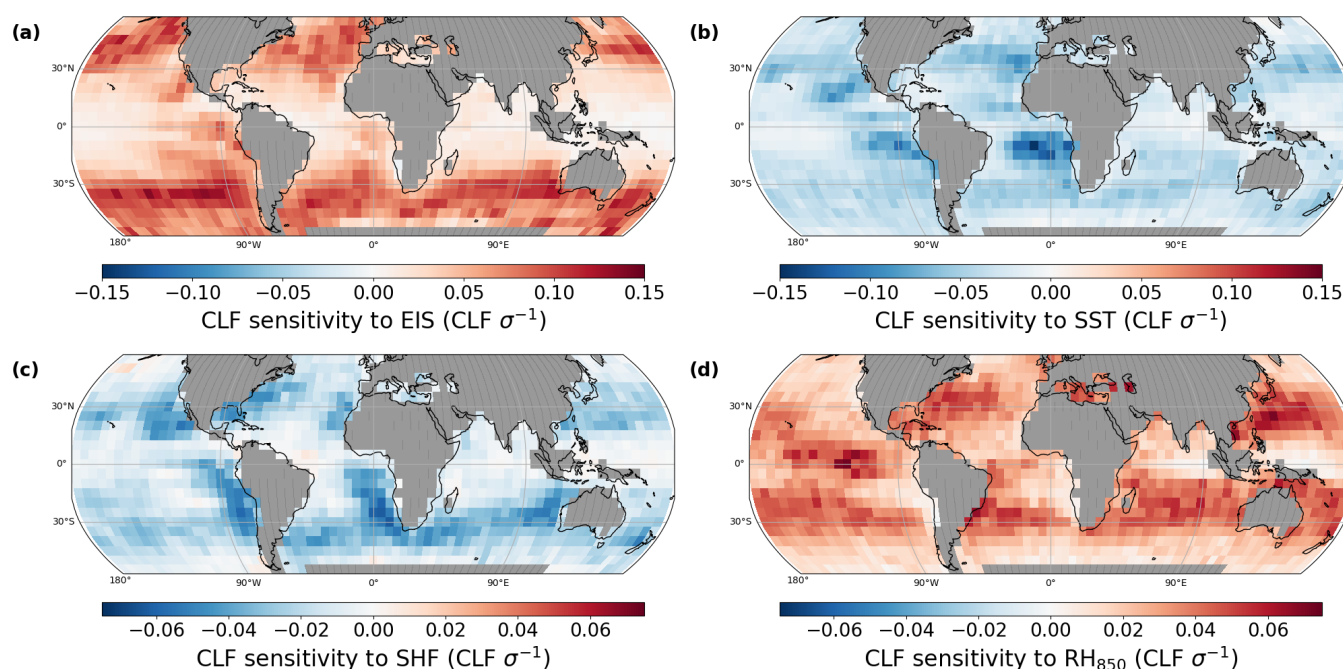


Figure 5. Sensitivity of the marine boundary-layer cloud fraction to the estimated inversion strength (EIS), sea-surface temperature (SST), sensible heat flux (SHF), and relative humidity at 850 hPa (RH_{850}). Note that the range of colour bars of SHF and RH_{850} (-0.075 to 0.075) is narrower than EIS and SST (-0.15 to 0.15).

ican continents. CLF increases due to cold-air outbreaks in NW Atlantic and NW Pacific may be the reason for these positive sensitivities. Cold-air outbreaks occur during winter as cold continental air moves over warmer SSTs, increasing moisture and heat fluxes into the MBL so that the formation of MBLCs is favoured (Young et al., 2002). This leads to wintertime maxima in CLF in these regions (Yuan and Oreopoulos, 2013).

The sensitivity of CLF to the meridional winds at 700 hPa exhibits two bands straddling the subtropical regions between about 15° and 35° in both hemispheres but opposite in sign (positive in the Northern Hemisphere and negative in the Southern Hemisphere), illustrating that in these regions, the poleward winds are associated with an increase in low cloud fraction. The bands are still apparent at 850 hPa, while the negative band in the Southern Hemisphere extends northward to tropical areas. These hemispheric sensitivity bands to the v wind component at 700 hPa closely resemble those found in Andersen et al. (2023), with their analysis suggesting that the poleward winds on the eastern side of midlatitude cyclones may be related to warm and moist advection, increasing CLF. However, they also find a strong correlation of these free-tropospheric poleward winds with large-scale ascending air motion making the assertion of causality difficult. Poleward winds are also found to decrease CLF over the Southern Hemispheric midlatitudes.

CLF is negatively connected to the vertical pressure velocity at both 700 and 850 hPa (ω_{700} and ω_{850}) over the en-

tire Earth, indicating that ascending large-scale air motion enhances the cover of MBLCs globally. It is shown in the bottom of Fig. 6 column (a) that the CLF sensitivity to ω_{700} is larger in the midlatitude ocean basins, whereas the CLF sensitivity to ω_{850} is larger in the subtropical oceans, where subsidence is climatologically prevalent (Myers and Norris, 2015, 2016; Scott et al., 2020). This seems indicative of CLF being the most sensitive to large-scale ascending motion at the typical altitude of the clouds. It is interesting to note that between 30°N and 30°S , no marked CLF sensitivity to ω_{700} is found, contrasting the finding of enhanced subsidence at this level reducing MBLCs by Myers and Norris (2013). This effect is likely better described in the ω_{850} data, which is more related to the altitude of the cloud top.

3.3 Dependence of N_d –CLF relationship on meteorology

3.3.1 Global overview of the interaction indices

In this section, we use the IAI as defined in Sect. 2.3.2 to quantitatively show how the response of MBLC fraction attributed to the aerosol proxy N_d varies with the meteorological factors. As discussed in Sect. 2.3.2, since the sensitivity related to N_d is positive across the globe (Fig. 5d), a positive IAI can be interpreted as an amplification of the N_d –CLF sensitivity with high (above-average) feature values of a meteorological variable, whereas a negative IAI signifies an amplification of the sensitivity at low feature values.

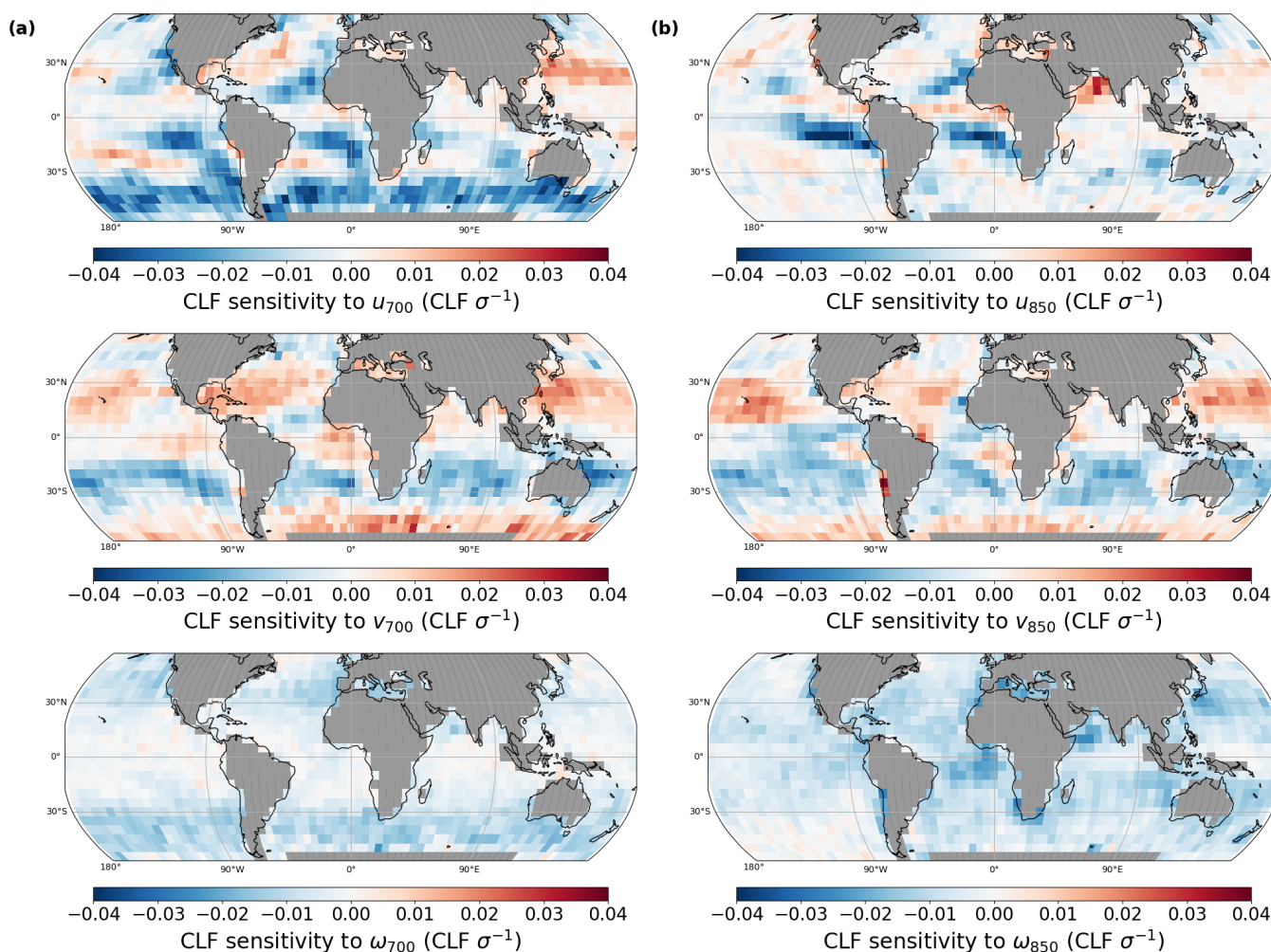


Figure 6. Sensitivity of cloud fraction to wind component vectors u and v and vertical velocities at 700 hPa (column **a**) and 850 hPa (column **b**). Note that the range of the colour bars is in general smaller (-0.04 – 0.04) than in Fig. 5.

In Fig. 7, analogous to Fig. 3, the features along the x axis are arranged in descending order based on their averaged absolute IAI, that is, by the strength of the impact of each meteorological feature on the N_d –CLF sensitivity. Similar to the feature importance summarized by Fig. 3, EIS, SST, RH₈₅₀, and SHF have relatively large strength of interaction effect and can thus be regarded as critical controlling factors for not only marine low cloud cover but also their response to changes in N_d (and in extension aerosols). Compared to the CLF sensitivities, the IAIs associated with atmospheric temperatures at 700 and 850 hPa have greater strengths. Furthermore, it can also be seen that the vertical and horizontal winds at the surface and different pressure levels are generally ranked lower. In general, the thermodynamical factors seem to have a stronger influence on the N_d –CLF sensitivity than the dynamical factors.

3.3.2 Spatial patterns of the interaction indices

Coherent and distinct spatial distributions of the impact of selected meteorological parameters on the N_d –CLF relationship can be observed. Hereafter, we show the regional characteristics of the interaction effects of EIS and SST, which are the two most important meteorological factors for CLF in MBLCs and have the greatest absolute strengths of IAI. EIS exerts the most noticeable positive IAIs over the midlatitude oceanic areas (Fig. 8a), reflecting that stronger temperature inversions capping the MBL over these regions may amplify the positive N_d –CLF relationship. The interpretation of possible underlying physical mechanisms of these interaction effects is difficult and remains speculative. The results seem to suggest that in these regions, potentially through hampering the entrainment of drier air from the free troposphere, the stronger inversion and more stable conditions are capable of trapping more moisture within a shallower MBL and could thus weaken the evaporation–entrainment feedback. As a re-

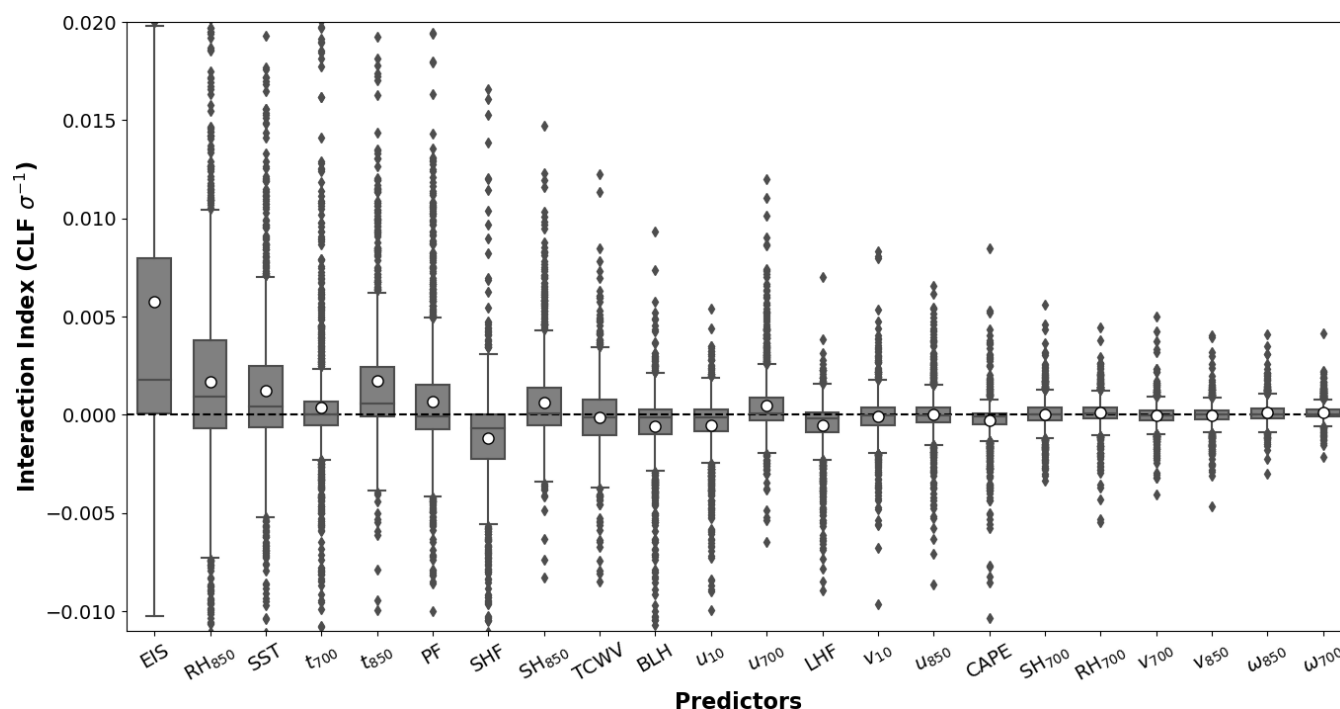


Figure 7. Similar to Fig. 3 but for the interaction effect of N_d with all environmental parameters, quantified by the interaction index ($\text{CLF } \sigma^{-1}$).

sult, it may ultimately favour a more positive N_d –CLF relationship (Chen et al., 2014; Christensen et al., 2020). It is interesting to note that these interactions are not apparent in the stratocumulus regions, where EIS is a strong control of CLF, and in the stratocumulus-to-cumulus transition regions, where Christensen et al. (2020) found the aerosol effect on this transition to be confined to stable atmospheric conditions. This may imply that the suggested entrainment effect is dependent on the EIS and stronger at slightly lower EIS values typically found in the midlatitudes (Scott et al., 2020). The observed impact of EIS on the N_d –CLF relationship found in the midlatitudes may also have implications within the context of climate change. While in the subtropics global climate models predict an increase in EIS with a warming climate, in the midlatitudes EIS is predicted to decrease (Myers et al., 2021), potentially decreasing the sensitivity of CLF to N_d there.

Figure 8b shows that higher SSTs are found to amplify the positive N_d –CLF relationship (positive IAI) in the regions of frequent stratocumulus-to-cumulus transition (Cesana and Del Genio, 2021). The physical interpretation could be the following: here, higher SSTs tend to lead to the transition from stratocumulus clouds to shallow convective clouds (Cesana et al., 2019); however, this transition has been found to be delayed when aerosol is increased (Goren et al., 2019; Christensen et al., 2020). Tentatively, the positive IAIs in these transition regions may thus point to increased control of N_d on CLF at higher SST values as these are the situations

where transitions typically occur and when increased N_d can act to delay this transition. In these regions, higher SSTs in the future might thus increase the sensitivity of MBLC CLF to aerosols. It should be noted that the quantification of the dependence of the N_d –CLF relationship on meteorological factors (EIS, SST discussed in this section) is also likely subject to the biases in the N_d –CLF sensitivity caused by the N_d retrieval biases as a function of CLF. This would potentially contribute to the non-causal facets of the relationships and interactive effects quantified by SHAP values.

4 Conclusions

In this study, 9 years (2011–2019) of daily satellite and re-analysis data have been analysed to better understand the effect of N_d on CLF in MBLC and its dependence on meteorological factors. We have established a near-global machine learning framework to predict the cloud fraction of marine boundary clouds using regionally specific XGB regression models. Including many confounding and influencing factors as a whole, the explainable machine learning technique of SHAP regression values has been used to explain the regional XGB models; to quantify the CLF sensitivity to all cloud controlling factors with a specific focus on N_d ; and, moreover, to quantify the meteorological influence on the N_d –CLF relationship at a global scale. The statistical sensitivities and interactive effects are interpreted with the guidance of hypothesized causal pathways and the state-of-the-art

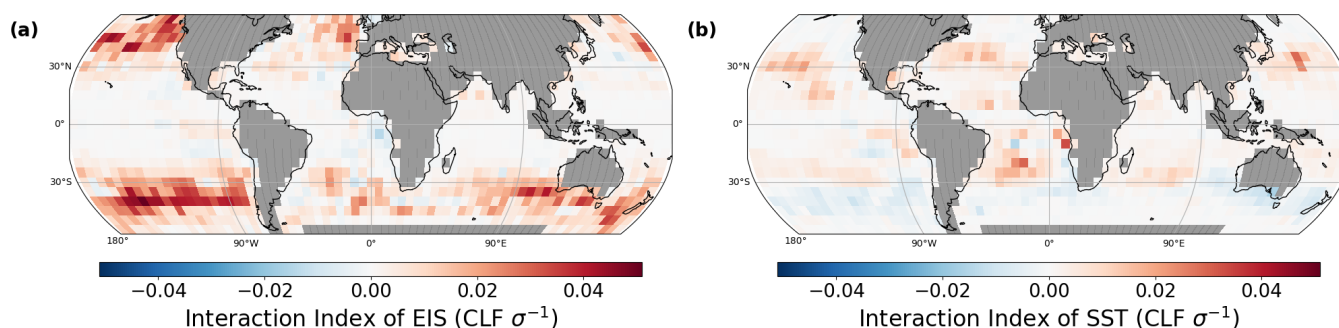


Figure 8. Patterns of the interaction index showing the dependence of the N_d –CLF relationship on estimated inversion strength (EIS) (a) and sea-surface temperature (SST) (b).

physical understanding of the system. The main findings of this study, which should be interpreted in light of the data and methodology limitations discussed in Sect. 2.3.3), are summarized as follows:

1. The marine boundary-layer cloud fraction shows a notable positive sensitivity to N_d (a surrogate for aerosols) in the regions of stratocumulus-to-cumulus transition, which may arise from the high N_d delaying this transition. The N_d –CLF sensitivity in the Southern Hemispheric midlatitudes is observed to be higher than in previous studies, which should be investigated in future work. The estimated N_d –CLF sensitivity and its magnitude suggest that aerosols likely have a considerable impact on MBL cloudiness although this may partially result from an overestimation caused by the effect of a positive retrieval bias of N_d at high CLF.
2. Consistent with the literature, our statistical method shows that EIS and SST are two important determinants for low marine clouds by regulating surface fluxes and dry-air entrainment processes. In addition, strong negative CLF sensitivity and spatial patterns for SHF are also found, suggesting that the effect of cold air advection might surpass the SHF enhancement of closed-to-open-cell and cumulus transitions. Dynamic drivers (meridional and zonal winds) indicate that midlatitude synoptic-scale disturbances and vertical wind shear seemingly make considerable contributions to marine low cloud amounts.
3. In general, thermodynamical parameters exert a more important influence on the N_d –CLF relationship than dynamical parameters. EIS, RH_{850} , SST, and temperatures at 700 and 850 hPa have the strongest effect on the N_d –CLF sensitivity. In the midlatitudes, higher EIS is found to amplify the positive N_d –CLF sensitivity, which may be related to a reduced entrainment feedback in these conditions, whereas higher SST is found to amplify the N_d –CLF sensitivity in stratocumulus-to-cumulus transition regions, which is potentially because

the transition induced by higher SSTs may be delayed by increased N_d . These findings have potential implications for possible future changes in the sensitivity of CLF to aerosols.

4. For the dynamical and thermodynamical factors shown here, both CLF sensitivities and interactive effects (dependence of N_d –CLF relationship on meteorology) exhibit distinct regional patterns. These coherent spatial patterns indicate that the proposed explainable machine learning framework not only is capable of skilfully predicting CLF for marine low clouds but also has the potential to capture regional characteristics of the relation between CLF and N_d as well as meteorological influences.

In the future, the observation-based sensitivities and interactive effects quantified by the ML framework here will be compared to those in ESMs, which have the potential to evaluate ESM parameterizations related to ACI and even help gain insights into how the models could be tuned in this respect. In addition, incorporating causal approaches for SHAP, such as those proposed by Heskes et al. (2020) and Frye et al. (2021), would help to test to which extent the observed statistical relationships and interaction effects represent physical processes.

Code availability. Code is available from the corresponding author upon reasonable request.

Data availability. All data sets used in this study are publicly available. The MODIS data set (https://doi.org/10.5067/MODIS/MOD08_D3.061, Platnick et al., 2015) was acquired from the Level-1 and Atmosphere Archive and Distribution System (LAADS) Distributed Active Archive Center (DAAC) (NASA: MODIS Data Collection, <https://ladsweb.modaps.eosdis.nasa.gov/search/>, last access: 17 November 2024); the hourly reanalysis data at single levels (<https://doi.org/10.24381/cds.adbb2d47>, Hersbach et al., 2023a) and pressure levels (<https://doi.org/10.24381/cds.bd0915c6>, Hers-

bach et al., 2023b) are obtained from the Copernicus Climate Change Service (C3S) Climate Data Store.

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Author contributions. HA and JC designed the initial research idea. YJ, HA, and JC developed the study concept and methodology. YJ and HA obtained and analysed the data sets. YJ implemented the explainable machine learning framework, performed the visualization, and wrote the original draft. All authors contributed to interpreting the results and reviewing and improving the paper.

Competing interests. The contact author has declared that none of the authors has any competing interests.

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References

Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017, <https://doi.org/10.1038/nature03174>, 2004.

- Adebisi, A. A. and Zuidema, P.: The role of the southern African easterly jet in modifying the southeast Atlantic aerosol and cloud environments, *Q. J. Roy. Meteorol. Soc.*, 142, 1574–1589, <https://doi.org/10.1002/qj.2765>, 2016.
- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, <https://doi.org/10.1126/science.245.4923.1227>, 1989.
- Andersen, H. and Cermak, J.: How thermodynamic environments control stratocumulus microphysics and interactions with aerosols, *Environ. Res. Lett.*, 10, 024004, <https://doi.org/10.1088/1748-9326/10/2/024004>, 2015.
- Andersen, H., Cermak, J., Fuchs, J., and Schwarz, K.: Global observations of cloud-sensitive aerosol loadings in low-levelmarine clouds, *J. Geophys. Res.*, 121, 936–12, <https://doi.org/10.1002/2016JD025614>, 2016.
- Andersen, H., Cermak, J., Fuchs, J., Knutti, R., and Lohmann, U.: Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks, *Atmos. Chem. Phys.*, 17, 9535–9546, <https://doi.org/10.5194/acp-17-9535-2017>, 2017.
- Andersen, H., Cermak, J., Störner, R., Fuchs, J., Kim, M., and Pauli, E.: Assessment of COVID-19 effects on satellite-observed aerosol loading over China with machine learning, *Tellus B*, 73, 1–14, <https://doi.org/10.1080/16000889.2021.1971925>, 2021.
- Andersen, H., Cermak, J., Zipfel, L., and Myers, T. A.: Attribution of Observed Recent Decrease in Low Clouds Over the Northeastern Pacific to Cloud-Controlling Factors, *Geophys. Res. Lett.*, 49, 1–10, <https://doi.org/10.1029/2021gl096498>, 2022.
- Andersen, H., Cermak, J., Douglas, A., Myers, T. A., Nowack, P., Stier, P., Wall, C. J., and Wilson Kemsley, S.: Sensitivities of cloud radiative effects to large-scale meteorology and aerosols from global observations, *Atmos. Chem. Phys.*, 23, 10775–10794, <https://doi.org/10.5194/acp-23-10775-2023>, 2023.
- Arola, A., Lipponen, A., Kolmonen, P., Virtanen, T. H., Bellouin, N., Grosvenor, D. P., Gryspeerdt, E., Quaas, J., and Kokkola, H.: Aerosol effects on clouds are concealed by natural cloud heterogeneity and satellite retrieval errors, *Nat. Commun.*, 13, 7357, <https://doi.org/10.1038/s41467-022-34948-5>, 2022.
- Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A. L., Dufresne, J. L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B.: Bounding Global Aerosol Radiative Forcing of Climate Change, *Rev. Geophys.*, 58, 1–45, <https://doi.org/10.1029/2019RG000660>, 2020.
- Bender, F. A., Frey, L., McCoy, D. T., Grosvenor, D. P., and Mohrmann, J. K.: Assessment of aerosol–cloud–radiation correlations in satellite observations, climate models and reanalysis, *Clim. Dynam.*, 52, 4371–4392, <https://doi.org/10.1007/s00382-018-4384-z>, 2019.
- Bender, F. A., Lord, T., Staffansdotter, A., Jung, V., and Undorf, S.: Machine Learning Approach to Investigating the Relative Importance of Meteorological and Aerosol-Related Parameters in Determining Cloud Microphysical Properties, *Tellus B*, 76, 1–18, <https://doi.org/10.16993/tellusb.1868>, 2024.

- Bennartz, R. and Rausch, J.: Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations, *Atmos. Chem. Phys.*, 17, 9815–9836, <https://doi.org/10.5194/acp-17-9815-2017>, 2017.
- Beucler, T., Ebert-Uphoff, I., Rasp, S., Pritchard, M., and Gentine, P.: Machine Learning for Clouds and Climate, in: *Clouds and Their Climatic Impacts*, Geophysical Monograph Series, 325–345, ISBN 9781119700357, <https://doi.org/10.1002/9781119700357.ch16>, 2023.
- Blossey, P. N., Bretherton, C. S., Zhang, M., Cheng, A., Endo, S., Heus, T., Liu, Y., Lock, A. P., de Roode, S. R., and Xu, K.-M.: Marine low cloud sensitivity to an idealized climate change: The CGILS LES intercomparison, *J. Adv. Model. Earth Sy.*, 5, 234–258, <https://doi.org/10.1002/jame.20025>, 2013.
- Bretherton, C. S.: Insights into low-latitude cloud feedbacks from high-resolution models, *Philos. T. R. Soc. A*, 373, 20140415, <https://doi.org/10.1098/rsta.2014.0415>, 2015.
- Bretherton, C. S. and Wyant, M. C.: Moisture Transport, Lower-Tropospheric Stability, and Decoupling of Cloud-Topped Boundary Layers, *J. Atmos. Sci.*, 54, 148–167, [https://doi.org/10.1175/1520-0469\(1997\)054<0148:MTLSTA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1997)054<0148:MTLSTA>2.0.CO;2), 1997.
- Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, *Geophys. Res. Lett.*, 34, L03813, <https://doi.org/10.1029/2006GL027648>, 2007.
- Bretherton, C. S., Blossey, P. N., and Jones, C. R.: Mechanisms of marine low cloud sensitivity to idealized climate perturbations: A single-LES exploration extending the CGILS cases, *J. Adv. Model. Earth Sy.*, 5, 316–337, <https://doi.org/10.1002/jame.20019>, 2013.
- Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G. W., Spracklen, D. V., Woodhouse, M. T., Regayre, L. A., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing, *Nature*, 503, 67–71, <https://doi.org/10.1038/nature12674>, 2013.
- Ceppi, P. and Nowack, P.: Observational evidence that cloud feedback amplifies global warming, *P. Natl. Acad. Sci. USA*, 118, e2026290118, <https://doi.org/10.1073/pnas.2026290118>, 2021.
- Cesana, G., Del Genio, A. D., Ackerman, A. S., Kelley, M., Elsaesser, G., Fridlind, A. M., Cheng, Y., and Yao, M.-S.: Evaluating models' response of tropical low clouds to SST forcings using CALIPSO observations, *Atmos. Chem. Phys.*, 19, 2813–2832, <https://doi.org/10.5194/acp-19-2813-2019>, 2019.
- Cesana, G. V. and Del Genio, A. D.: Observational constraint on cloud feedbacks suggests moderate climate sensitivity, *Nat. Clim. Change*, 11, 213–218, <https://doi.org/10.1038/s41558-020-00970-y>, 2021.
- Chen, H., Janizek, J. D., Lundberg, S., and Lee, S.-I.: True to the Model or True to the Data?, *ArXiv*, abs/2006.16234, <https://arxiv.org/abs/2006.16234> (last access: 17 November 2024), 2020.
- Chen, H., Lundberg, S. M., and Lee, S.-I.: Explaining a series of models by propagating Shapley values, *Nat. Commun.*, 13, 4512, <https://doi.org/10.1038/s41467-022-31384-3>, 2022.
- Chen, H., Covert, I. C., Lundberg, S. M., and Lee, S.-I.: Algorithms to estimate Shapley value feature attributions, *Nat. Mach. Intell.*, 5, 590–601, <https://doi.org/10.1038/s42256-023-00657-x>, 2023.
- Chen, T. and Guestrin, C.: XGBoost: A scalable tree boosting system, *Proceedings of the ACM SIGKDD International Conference on Knowledge Discovery and Data Mining*, KDD'16: The 22nd ACM SIGKDD International Conference on Knowledge Discovery and Data Mining San Francisco California USA, 13–17 August, Association for Computing Machinery, New York, NY, United States, 785–794, <https://doi.org/10.1145/2939672.2939785>, 2016.
- Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Partridge, D., Fieldsend, J., De Leeuw, J., Schmidt, A., Cho, N., Oreopoulos, L., Platnick, S., Grosvenor, D., Field, P., and Lohmann, U.: Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover, *Nat. Geosci.*, 15, 609–614, <https://doi.org/10.1038/s41561-022-00991-6>, 2022.
- Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Peace, A., Partridge, D. G., Cho, N., Oreopoulos, L., Grosvenor, D., Field, P., Allan, R. P., and Lohmann, U.: Substantial cooling effect from aerosol-induced increase in tropical marine cloud cover, *Nat. Geosci.*, 17, 404–410, <https://doi.org/10.1038/s41561-024-01427-z>, 2024.
- Chen, Y. C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds, *Nat. Geosci.*, 7, 643–646, <https://doi.org/10.1038/ngeo2214>, 2014.
- Christensen, M. W., Chen, Y.-C., and Stephens, G. L.: Aerosol indirect effect dictated by liquid clouds, *J. Geophys. Res.-Atmos.*, 121, 614–636, <https://doi.org/10.1002/2016JD025245>, 2016.
- Christensen, M. W., Neubauer, D., Poulsen, C. A., Thomas, G. E., McGarragh, G. R., Povey, A. C., Proud, S. R., and Grainger, R. G.: Unveiling aerosol–cloud interactions – Part 1: Cloud contamination in satellite products enhances the aerosol indirect forcing estimate, *Atmos. Chem. Phys.*, 17, 13151–13164, <https://doi.org/10.5194/acp-17-13151-2017>, 2017.
- Christensen, M. W., Jones, W. K., and Stier, P.: Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition, *P. Natl. Acad. Sci. USA*, 117, 17591–17598, <https://doi.org/10.1073/pnas.1921231117>, 2020.
- Dadashazar, H., Painemal, D., Alipanah, M., Brunke, M., Chellappan, S., Corral, A. F., Crosbie, E., Kirschler, S., Liu, H., Moore, R. H., Robinson, C., Scarino, A. J., Shook, M., Sinclair, K., Thornhill, K. L., Voigt, C., Wang, H., Winstead, E., Zeng, X., Ziemba, L., Zuidema, P., and Sorooshian, A.: Cloud drop number concentrations over the western North Atlantic Ocean: seasonal cycle, aerosol interrelationships, and other influential factors, *Atmos. Chem. Phys.*, 21, 10499–10526, <https://doi.org/10.5194/acp-21-10499-2021>, 2021.
- de Szoek, S. P., Verlinden, K. L., Yuter, S. E., and Mechum, D. B.: The Time Scales of Variability of Marine Low Clouds, *J. Clim.*, 29, 6463–6481, <https://doi.org/10.1175/JCLI-D-15-0460.1>, 2016.
- Dey, S., Di Girolamo, L., Zhao, G., Jones, A. L., and McFarquhar, G. M.: Satellite-observed relationships between aerosol and trade-wind cumulus cloud properties over the Indian Ocean, *Geophys. Res. Lett.*, 38, L01804, <https://doi.org/10.1029/2010GL045588>, 2011.
- Douglas, A. R. and L'Ecuyer, T.: Possible evidence of increased global cloudiness due to aerosol-cloud interactions, *Atmos. Chem. Phys. Discuss.* [preprint], <https://doi.org/10.5194/acp-2022-688>, 2022.

- Elith, J., Leathwick, J. R., and Hastie, T.: A working guide to boosted regression trees, *J. Anim. Ecol.*, 77, 802–813, <https://doi.org/10.1111/j.1365-2656.2008.01390.x>, 2008.
- Fan, J., Wang, Y., Rosenfeld, D., and Liu, X.: Review of aerosol-cloud interactions: Mechanisms, significance, and challenges, *J. Atmos. Sci.*, 73, 4221–4252, <https://doi.org/10.1175/JAS-D-16-0037.1>, 2016.
- Fons, E., Runge, J., Neubauer, D., and Lohmann, U.: Stratocumulus adjustments to aerosol perturbations disentangled with a causal approach, *npj Clim. Atmos. Sci.*, 6, 1–10, <https://doi.org/10.1038/s41612-023-00452-w>, 2023.
- Forster, P. M., Storelvmo, T., Armour, K., Collins, W., Dufresne, J. L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D., Watanabe, M., Wild, M., and Zhang, H.: Chapter 7: The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity, in: *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J., Maycock, T. K., Waterfield, T., Yelekçi, O., Yu, R., and Zhou, B., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 923–1054, <https://doi.org/10.1017/9781009157896.009>, 2021.
- Frye, C., Rowat, C., and Feige, I.: Asymmetric Shapley values: incorporating causal knowledge into model-agnostic explainability, *Adv. Neural Informat. Process. Syst.*, 33, 1229–1239, <https://doi.org/10.48550/arXiv.1910.06358>, 2021.
- Fuchs, J., Cermak, J., and Andersen, H.: Building a cloud in the southeast Atlantic: Understanding low-cloud controls based on satellite observations with machine learning, *Atmos. Chem. Phys.*, 18, 16537–16552, <https://doi.org/10.5194/acp-18-16537-2018>, 2018.
- Ghan, S., Wang, M., Zhang, S., Ferrachat, S., Gettelman, A., Griesfeller, J., Kipling, Z., Lohmann, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., and Zhang, K.: Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability, *P. Natl. Acad. Sci. USA*, 113, 5804–5811, <https://doi.org/10.1073/PNAS.1514036113>, 2016.
- Goren, T., Kazil, J., Hoffmann, F., Yamaguchi, T., and Feingold, G.: Anthropogenic Air Pollution Delays Marine Stratocumulus Breakup to Open Cells, *Geophys. Res. Lett.*, 46, 14135–14144, <https://doi.org/10.1029/2019GL085412>, 2019.
- Grandey, B. S. and Stier, P.: A critical look at spatial scale choices in satellite-based aerosol indirect effect studies, *Atmos. Chem. Phys.*, 10, 11459–11470, <https://doi.org/10.5194/acp-10-11459-2010>, 2010.
- Grise, K. M. and Kelleher, M. K.: Midlatitude Cloud Radiative Effect Sensitivity to Cloud Controlling Factors in Observations and Models: Relationship with Southern Hemisphere Jet Shifts and Climate Sensitivity, *J. Clim.*, 34, 5869–5886, <https://doi.org/10.1175/JCLI-D-20-0986.1>, 2021.
- Grise, K. M. and Medeiros, B.: Understanding the Varied Influence of Midlatitude Jet Position on Clouds and Cloud Radiative Effects in Observations and Global Climate Models, *J. Clim.*, 29, 9005–9025, <https://doi.org/10.1175/JCLI-D-16-0295.1>, 2016.
- Grosvenor, D. P., Sourdeval, O., Zuidema, P., Ackerman, A., Alexandrov, M. D., Bennartz, R., Boers, R., Cairns, B., Chiu, J. C., Christensen, M., Deneke, H., Diamond, M., Feingold, G., Fridlind, A., Hünerbein, A., Knist, C., Kollias, P., Marshak, A., McCoy, D., Merk, D., Painemal, D., Rausch, J., Rosenfeld, D., Russchenberg, H., Seifert, P., Sinclair, K., Stier, P., van Diedenoven, B., Wendisch, M., Werner, F., Wood, R., Zhang, Z., and Quaas, J.: Remote Sensing of Droplet Number Concentration in Warm Clouds: A Review of the Current State of Knowledge and Perspectives, *Rev. Geophys.*, 56, 409–453, <https://doi.org/10.1029/2017RG000593>, 2018.
- Gryspeerd, E. and Stier, P.: Regime-based analysis of aerosol-cloud interactions, *Geophys. Res. Lett.*, 39, 1–5, <https://doi.org/10.1029/2012GL053221>, 2012.
- Gryspeerd, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, *J. Geophys. Res.*, 121, 3566–3583, <https://doi.org/10.1002/2015JD023744>, 2016.
- Gryspeerd, E., Quaas, J., Ferrachat, S., Gettelman, A., Ghan, S., Lohman, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M., and Zhang, K.: Constraining the instantaneous aerosol influence on cloud albedo, *P. Natl. Acad. Sci. USA*, 114, 4899–4904, <https://doi.org/10.1073/pnas.1617765114>, 2017.
- Gryspeerd, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid water path, *Atmos. Chem. Phys.*, 19, 5331–5347, <https://doi.org/10.5194/acp-19-5331-2019>, 2019.
- Gryspeerd, E., McCoy, D. T., Crosbie, E., Moore, R. H., Nott, G. J., Painemal, D., Small-Griswold, J., Sorooshian, A., and Ziemba, L.: The impact of sampling strategy on the cloud droplet number concentration estimated from satellite data, *Atmos. Meas. Tech.*, 15, 3875–3892, <https://doi.org/10.5194/amt-15-3875-2022>, 2022.
- Hamby, D. M.: A review of techniques for parameter sensitivity analysis of environmental models, *Environ. Monit. Assess.*, 32, 135–154, <https://doi.org/10.1007/BF00547132>, 1994.
- Hartmann, D. L., Ockert-Bell, M. E., and Michelsen, M. L.: The Effect of Cloud Type on Earth's Energy Balance: Global Analysis, *J. Clim.*, 5, 1281–1304, [https://doi.org/10.1175/1520-0442\(1992\)005<1281:TEOCTO>2.0.CO;2](https://doi.org/10.1175/1520-0442(1992)005<1281:TEOCTO>2.0.CO;2), 1992.
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Q. J. Roy. Meteorol. Soc.*, 146, 1999–2049, <https://doi.org/10.1002/qj.3803>, 2020.
- Hersbach, H., Bell, B., Berrisford, P., Biavati, G., Horányi, A., Muñoz Sabater, J., Nicolas, J., Peubey, C., Radu, R., 25 Rozum, I., Schepers, D., Simmons, A., Soci, C., Dee, D., and Thépaut, J.-N.: ERA5 hourly data on single levels from 1940 to present, Copernicus Climate Change Service (C3S) Climate Data Store (CDS) [data set], <https://doi.org/10.24381/cds.adbb2d47>, 2023a.

- Hersbach, H., Bell, B., Berrisford, P., Biavati, G., Horányi, A., Muñoz Sabater, J., Nicolas, J., Peubey, C., Radu, R., Rozum, I., Schepers, D., Simmons, A., Soci, C., Dee, D., and Thépaut, J.-N.: ERA5 hourly data on pressure levels from 1940 to present, Copernicus Climate Change Service (C3S) Climate Data Store (CDS) [data set], <https://doi.org/10.24381/cds.bd0915c6>, 2023b.
- Heskes, T., Sijben, E., Bucur, I. G., and Claassen, T.: Causal Shapley Values: Exploiting Causal Knowledge to Explain Individual Predictions of Complex Models, *Adv. Neural Informat. Process. Syst.*, 33, 4778–4789, <https://doi.org/10.48550/arXiv.2011.01625>, 2020.
- Jiang, H., Xue, H., Teller, A., Feingold, G., and Levin, Z.: Aerosol effects on the lifetime of shallow cumulus, *Geophys. Res. Lett.*, 33, L14806, <https://doi.org/10.1029/2006GL026024>, 2006.
- Kapoor, S., Cantrell, E., Peng, K., Pham, T. H., Bail, C. A., Gundersen, O. E., Hofman, J. M., Hullman, J., Lones, M. A., Malik, M. M., Nanayakkara, P., Poldrack, R. A., Raji, I. D., Roberts, M., Salganik, M. J., Serra-Garcia, M., Stewart, B. M., Vandewiele, G., and Narayanan, A.: REFORMS: Reporting Standards for Machine Learning Based Science, *Arxiv*, <https://arxiv.org/abs/2308.07832>, 2023.
- Karpatne, A., Ebert-Uphoff, I., Ravela, S., Babaie, H. A., and Kumar, V.: Machine Learning for the Geosciences: Challenges and Opportunities, *Arxiv*, <https://arxiv.org/abs/1711.04708>, 2017.
- Kaufman, Y. J. and Koren, I.: Smoke and Pollution Aerosol Effect on Cloud Cover, *Science*, 313, 655–658, <https://doi.org/10.1126/science.1126232>, 2006.
- Kazil, J., Feingold, G., Wang, H., and Yamaguchi, T.: On the interaction between marine boundary layer cloudiness and surface heat fluxes, *Atmos. Chem. Phys.*, 14, 61–79, <https://doi.org/10.5194/acp-14-61-2014>, 2014.
- Kelleher, M. K. and Grise, K. M.: Examining Southern Ocean Cloud Controlling Factors on Daily Time Scales and Their Connections to Midlatitude Weather Systems, *J. Clim.*, 32, 5145–5160, <https://doi.org/10.1175/JCLI-D-18-0840.1>, 2019.
- Kim, M., Brunner, D., and Kuhlmann, G.: Importance of satellite observations for high-resolution mapping of near-surface NO₂ by machine learning, *Remote Sens. Environ.*, 264, 112573, <https://doi.org/10.1016/j.rse.2021.112573>, 2021.
- Klein, S. A. and Hartmann, D. L.: The Seasonal Cycle of Low Stratiform Clouds, *J. Clim.*, 6, 1587–1606, [https://doi.org/10.1175/1520-0442\(1993\)006<1587:TSCOLS>2.0.CO;2](https://doi.org/10.1175/1520-0442(1993)006<1587:TSCOLS>2.0.CO;2), 1993.
- Leahy, L. V., Wood, R., Charlson, R. J., Hostetler, C. A., Rogers, R. R., Vaughan, M. A., and Winker, D. M.: On the nature and extent of optically thin marine low clouds, *J. Geophys. Res.-Atmos.*, 117, D22201, <https://doi.org/10.1029/2012JD017929>, 2012.
- Li, W., Migliavacca, M., Forkel, M., Denissen, J. M. C., Reichstein, M., Yang, H., Duveiller, G., Weber, U., and Orth, R.: Widespread increasing vegetation sensitivity to soil moisture, *Nat. Commun.*, 13, 3959, <https://doi.org/10.1038/s41467-022-31667-9>, 2022.
- Linardatos, P., Papastefanopoulos, V., and Kotsiantis, S.: Explainable AI: A Review of Machine Learning Interpretability Methods, *Entropy*, 23, 18, <https://doi.org/10.3390/e23010018>, 2021.
- Liu, Y., Lin, T., Zhang, J., Wang, F., Huang, Y., Wu, X., Ye, H., Zhang, G., Cao, X., and de Leeuw, G.: Opposite effects of aerosols and meteorological parameters on warm clouds in two contrasting regions over eastern China, *Atmos. Chem. Phys.*, 24, 4651–4673, <https://doi.org/10.5194/acp-24-4651-2024>, 2024.
- Loeb, N. G. and Schuster, G. L.: An observational study of the relationship between cloud, aerosol and meteorology in broken low-level cloud conditions, *J. Geophys. Res.-Atmos.*, 113, D14214, <https://doi.org/10.1029/2007JD009763>, 2008.
- Lundberg, S. M. and Lee, S. I.: A unified approach to interpreting model predictions, *Adv. Neur. In.*, abs/1705.07874, <https://doi.org/10.48550/arXiv.1705.07874>, 2017.
- Lundberg, S. M., Erion, G. G., and Lee, S.-I.: Consistent Individualized Feature Attribution for Tree Ensembles, *ArXiv*, <http://arxiv.org/abs/1802.03888>, 2018.
- Lundberg, S. M., Erion, G., Chen, H., DeGrave, A., Prutkin, J. M., Nair, B., Katz, R., Himmelfarb, J., Bansal, N., and Lee, S.-I.: From local explanations to global understanding with explainable AI for trees, *Nat. Mach. In.*, 2, 56–67, <https://doi.org/10.1038/s42256-019-0138-9>, 2020.
- McCoy, D. T., Bender, F. A.-M., Mohrmann, J. K. C., Hartmann, D. L., Wood, R., and Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA, and AeroCom, *J. Geophys. Res.-Atmos.*, 122, 1779–1796, <https://doi.org/10.1002/2016JD026141>, 2017a.
- McCoy, D. T., Eastman, R., Hartmann, D. L., and Wood, R.: The Change in Low Cloud Cover in a Warmed Climate Inferred from AIRS, MODIS, and ERA-Interim, *J. Clim.*, 30, 3609–3620, <https://doi.org/10.1175/JCLI-D-15-0734.1>, 2017b.
- Merk, D., Deneke, H., Pospichal, B., and Seifert, P.: Investigation of the adiabatic assumption for estimating cloud micro- and macro-physical properties from satellite and ground observations, *Atmos. Chem. Phys.*, 16, 933–952, <https://doi.org/10.5194/acp-16-933-2016>, 2016.
- Michibata, T., Suzuki, K., Sato, Y., and Takemura, T.: The source of discrepancies in aerosol–cloud–precipitation interactions between GCM and A-Train retrievals, *Atmos. Chem. Phys.*, 16, 15413–15424, <https://doi.org/10.5194/acp-16-15413-2016>, 2016.
- Miyamoto, A., Nakamura, H., and Miyasaka, T.: Influence of the Subtropical High and Storm Track on Low-Cloud Fraction and Its Seasonality over the South Indian Ocean, *J. Clim.*, 31, 4017–4039, <https://doi.org/10.1175/JCLI-D-17-0229.1>, 2018.
- Molnar, C.: Interpretable Machine Learning, 2nd Edn., <https://christophm.github.io/interpretable-ml-book> (last access: 17 November 2024), 2022.
- Myers, T. A. and Norris, J. R.: Observational Evidence That Enhanced Subsidence Reduces Subtropical Marine Boundary Layer Cloudiness, *J. Clim.*, 26, 7507–7524, <https://doi.org/10.1175/JCLI-D-12-00736.1>, 2013.
- Myers, T. A. and Norris, J. R.: On the Relationships between Subtropical Clouds and Meteorology in Observations and CMIP3 and CMIP5 Models, *J. Clim.*, 28, 2945–2967, <https://doi.org/10.1175/JCLI-D-14-00475.1>, 2015.
- Myers, T. A. and Norris, J. R.: Reducing the uncertainty in subtropical cloud feedback, *Geophys. Res. Lett.*, 43, 2144–2148, <https://doi.org/10.1002/2015GL067416>, 2016.
- Myers, T. A., Scott, R. C., Zelinka, M. D., Klein, S. A., Norris, J. R., and Caldwell, P. M.: Observational constraints on low cloud feedback reduce uncertainty of climate sensitivity, *Nat. Clim. Change*, 11, 501–507, <https://doi.org/10.1038/s41558-021-01039-0>, 2021.

- Padarian, J., McBratney, A. B., and Minasny, B.: Game theory interpretation of digital soil mapping convolutional neural networks, *Soil*, 6, 389–397, <https://doi.org/10.5194/soil-6-389-2020>, 2020.
- Platnick, S. and Twomey, S.: Determining the Susceptibility of Cloud Albedo to Changes in Droplet Concentration with the Advanced Very High Resolution Radiometer, *J. Appl. Meteorol. Climatol.*, 33, 334–347, [https://doi.org/10.1175/1520-0450\(1994\)033<0334:DTSOCA>2.0.CO;2](https://doi.org/10.1175/1520-0450(1994)033<0334:DTSOCA>2.0.CO;2), 1994.
- Platnick, S., King, M., and Hubanks, P.: MODIS Atmosphere L3 Daily Product, NASA MODIS Adaptive Processing System, Goddard Space Flight Center [data set], https://doi.org/10.5067/MODIS/MOD08_D3.061, 2015.
- Qu, X., Hall, A., Klein, S. A., and Caldwell, P. M.: The strength of the tropical inversion and its response to climate change in 18 CMIP5 models, *Clim. Dynam.*, 45, 375–396, <https://doi.org/10.1007/s00382-014-2441-9>, 2015a.
- Qu, X., Hall, A., Klein, S. A., and Deangelis, A. M.: Positive tropical marine low-cloud cover feedback inferred from cloud-controlling factors, *Geophys. Res. Lett.*, 42, 7767–7775, <https://doi.org/10.1002/2015GL065627>, 2015b.
- Quaas, J., Boucher, O., and Lohmann, U.: Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data, *Atmos. Chem. Phys.*, 6, 947–955, <https://doi.org/10.5194/acp-6-947-2006>, 2006.
- Rieck, M., Nuijens, L., and Stevens, B.: Marine Boundary Layer Cloud Feedbacks in a Constant Relative Humidity Atmosphere, *J. Atmos. Sci.*, 69, 2538–2550, <https://doi.org/10.1175/JAS-D-11-0203.1>, 2012.
- Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., and Yu, S.: Aerosol-driven droplet concentrations dominate coverage and water of oceanic low-level clouds, *Science*, 363, 6427, <https://doi.org/10.1126/science.aav0566>, 2019.
- Sato, Y., Goto, D., Michibata, T., Suzuki, K., Takemura, T., Tomita, H., and Nakajima, T.: Aerosol effects on cloud water amounts were successfully simulated by a global cloud-system resolving model, *Nat. Commun.*, 9, 1–7, <https://doi.org/10.1038/s41467-018-03379-6>, 2018.
- Schwarz, K., Cermak, J., Fuchs, J., and Andersen, H.: Mapping the Twilight Zone – What We Are Missing between Clouds and Aerosols, *Remote Sens.*, 9, 577, <https://doi.org/10.3390/rs9060577>, 2017.
- Scott, R. C., Myers, T. A., Norris, J. R., Zelinka, M. D., Klein, S. A., Sun, M., and Doelling, D. R.: Observed Sensitivity of Low-Cloud Radiative Effects to Meteorological Perturbations over the Global Oceans, *J. Clim.*, 33, 7717–7734, <https://doi.org/10.1175/JCLI-D-19-1028.1>, 2020.
- Seifert, A., Heus, T., Pincus, R., and Stevens, B.: Large-eddy simulation of the transient and near-equilibrium behavior of precipitating shallow convection, *J. Adv. Model. Earth Sy.*, 7, 1918–1937, <https://doi.org/10.1002/2015MS000489>, 2015.
- Seinfeld, J. H., Bretherton, C., Carslaw, K. S., Coe, H., DeMott, P. J., Dunlea, E. J., Feingold, G., Ghan, S., Guenther, A. B., Kahn, R., Kraucunas, I., Kreidenweis, S. M., Molina, M. J., Nenes, A., Penner, J. E., Prather, K. A., Ramanathan, V., Ramaswamy, V., Rasch, P. J., Ravishankara, A. R., Rosenfeld, D., Stephens, G., and Wood, R.: Improving our fundamental understanding of the role of aerosol-cloud interactions in the climate system, *P. Natl. Acad. Sci. USA*, 113, 5781–5790, <https://doi.org/10.1073/pnas.1514043113>, 2016.
- Small, J. D., Chuang, P. Y., Feingold, G., and Jiang, H.: Can aerosol decrease cloud lifetime?, *Geophys. Res. Lett.*, 36, 1–5, <https://doi.org/10.1029/2009GL038888>, 2009.
- Small, J. D., Jiang, J. H., Su, H., and Zhai, C.: Relationship between aerosol and cloud fraction over Australia, *Geophys. Res. Lett.*, 38, L23802, <https://doi.org/10.1029/2011GL049404>, 2011.
- Snoek, J., Larochelle, H., and Adams, R. P.: Practical Bayesian Optimization of Machine Learning Algorithms, in: *Advances in Neural Information Processing Systems*, edited by Pereira, F., Burges, C. J., Bottou, L., and Weinberger, K. Q., vol. 25, Curran Associates, Inc., 9 pp., <https://arxiv.org/abs/1206.2944> (last access: 17 November 2024), 2012.
- Stirnberg, R., Cermak, J., Kotthaus, S., Haeffelin, M., Andersen, H., Fuchs, J., Kim, M., Petit, J. E., and Favez, O.: Meteorology-driven variability of air pollution (PM₁) revealed with explainable machine learning, *Atmos. Chem. Phys.*, 21, 3919–3948, <https://doi.org/10.5194/acp-21-3919-2021>, 2021.
- Su, W., Loeb, N. G., Xu, K.-M., Schuster, G. L., and Eitzen, Z. A.: An estimate of aerosol indirect effect from satellite measurements with concurrent meteorological analysis, *J. Geophys. Res.-Atmos.*, 115, D18219, <https://doi.org/10.1029/2010JD013948>, 2010.
- Sundararajan, M. and Najmi, A.: The many shapley values for model explanation, in: *37th International Conference on Machine Learning, ICML 2020, Part F16814*, 9210–9220, ISBN 9781713821120, 2020.
- Toll, V., Christensen, M., Quaas, J., and Bellouin, N.: Weak average liquid-cloud-water response to anthropogenic aerosols, *Nature*, 572, 51–55, <https://doi.org/10.1038/s41586-019-1423-9>, 2019.
- Turner, D. D.: Improved ground-based liquid water path retrievals using a combined infrared and microwave approach, *J. Geophys. Res.-Atmos.*, 112, D15204, <https://doi.org/10.1029/2007JD008530>, 2007.
- Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, *J. Atmos. Sci.*, 34, 1149–1152, [https://doi.org/10.1175/1520-0469\(1977\)034<1149:tiopot>2.0.co;2](https://doi.org/10.1175/1520-0469(1977)034<1149:tiopot>2.0.co;2), 1977.
- van der Dussen, J. J., de Roode, S. R., Dal Gesso, S., and Siebesma, A. P.: An LES model study of the influence of the free tropospheric thermodynamic conditions on the stratocumulus response to a climate perturbation, *J. Adv. Model. Earth Sy.*, 7, 670–691, <https://doi.org/10.1002/2014MS000380>, 2015.
- Wall, C. J., Hartmann, D. L., and Ma, P.-L.: Instantaneous Linkages between Clouds and Large-Scale Meteorology over the Southern Ocean in Observations and a Climate Model, *J. Clim.*, 30, 9455–9474, <https://doi.org/10.1175/JCLI-D-17-0156.1>, 2017.
- Wang, S., Wang, Q., and Feingold, G.: Turbulence, Condensation, and Liquid Water Transport in Numerically Simulated Nonprecipitating Stratocumulus Clouds, *J. Atmos. Sci.*, 60, 262–278, [https://doi.org/10.1175/1520-0469\(2003\)060<0262:TCALWT>2.0.CO;2](https://doi.org/10.1175/1520-0469(2003)060<0262:TCALWT>2.0.CO;2), 2003.
- Wood, R.: Stratocumulus Clouds, *Mon. Weather Rev.*, 140, 2373–2423, <https://doi.org/10.1175/MWR-D-11-00121.1>, 2012.
- Wood, R. and Bretherton, C. S.: On the relationship between stratiform low cloud cover and lower-tropospheric stability, *J. Clim.*, 19, 6425–6432, <https://doi.org/10.1175/JCLI3988.1>, 2006.
- Wood, R. and Hartmann, D. L.: Spatial Variability of Liquid Water Path in Marine Low Cloud: The Importance of

- Mesoscale Cellular Convection, *J. Clim.*, 19, 1748–1764, <https://doi.org/10.1175/JCLI3702.1>, 2006.
- Xue, H. and Feingold, G.: Large-Eddy Simulations of Trade Wind Cumuli: Investigation of Aerosol Indirect Effects, *J. Atmos. Sci.*, 63, 1605–1622, <https://doi.org/10.1175/JAS3706.1>, 2006.
- Young, G. S., Kristovich, D. A. R., Hjelmfelt, M. R., and Foster, R. C.: Rolls, Streets, Waves, And More: A Review of Quasi-Two-Dimensional Structures in the Atmospheric Boundary Layer, *Bull. Am. Meteorol. Soc.*, 83, 997–1002, [https://doi.org/10.1175/1520-0477\(2002\)083<0997:RSWAMA>2.3.CO;2](https://doi.org/10.1175/1520-0477(2002)083<0997:RSWAMA>2.3.CO;2), 2002.
- Yuan, T. and Oreopoulos, L.: On the global character of overlap between low and high clouds, *Geophys. Res. Lett.*, 40, 5320–5326, <https://doi.org/10.1002/grl.50871>, 2013.
- Yuan, T., Remer, L. A., and Yu, H.: Microphysical, macrophysical and radiative signatures of volcanic aerosols in trade wind cumulus observed by the A-Train, *Atmos. Chem. Phys.*, 11, 7119–7132, <https://doi.org/10.5194/acp-11-7119-2011>, 2011.
- Yuan, T., Song, H., Wood, R., Oreopoulos, L., Platnick, S., Wang, C., Yu, H., Meyer, K., and Wilcox, E.: Observational evidence of strong forcing from aerosol effect on low cloud coverage, *Sci. Adv.*, 9, eadh7716, <https://doi.org/10.1126/sciadv.adh7716>, 2023.
- Zelinka, M. D., Andrews, T., Forster, P. M., and Taylor, K. E.: Quantifying components of aerosol-cloud-radiation interactions in climate models, *J. Geophys. Res.-Atmos.*, 119, 7599–7615, <https://doi.org/10.1002/2014JD021710>, 2014.
- Zelinka, M. D., Grise, K. M., Klein, S. A., Zhou, C., DeAngelis, A. M., and Christensen, M. W.: Drivers of the Low-Cloud Response to Poleward Jet Shifts in the North Pacific in Observations and Models, *J. Clim.*, 31, 7925–7947, <https://doi.org/10.1175/JCLI-D-18-0114.1>, 2018.
- Zhang, Z. and Platnick, S.: An assessment of differences between cloud effective particle radius retrievals for marine water clouds from three MODIS spectral bands, *J. Geophys. Res.-Atmos.*, 116, D20215, <https://doi.org/10.1029/2011JD016216>, 2011.
- Zhang, Z., Ackerman, A. S., Feingold, G., Platnick, S., Pincus, R., and Xue, H.: Effects of cloud horizontal inhomogeneity and drizzle on remote sensing of cloud droplet effective radius: Case studies based on large-eddy simulations, *J. Geophys. Res.-Atmos.*, 117, D19208, <https://doi.org/10.1029/2012JD017655>, 2012.
- Zheng, G., Wang, Y., Wood, R., Jensen, M. P., Kuang, C., McCoy, I. L., Matthews, A., Mei, F., Tomlinson, J. M., Shilling, J. E., Zawadowicz, M. A., Crosbie, E., Moore, R., Ziemba, L., Andreae, M. O., and Wang, J.: New particle formation in the remote marine boundary layer, *Nat. Commun.*, 12, 527, <https://doi.org/10.1038/s41467-020-20773-1>, 2021.
- Zhu, Y., Rosenfeld, D., and Li, Z.: Under What Conditions Can We Trust Retrieved Cloud Drop Concentrations in Broken Marine Stratocumulus?, *J. Geophys. Res.-Atmos.*, 123, 8754–8767, <https://doi.org/10.1029/2017JD028083>, 2018.
- Zipfel, L., Andersen, H., and Cermak, J.: Machine-Learning Based Analysis of Liquid Water Path Adjustments to Aerosol Perturbations in Marine Boundary Layer Clouds Using Satellite Observations, *Atmosphere*, 13, 586, <https://doi.org/10.3390/atmos13040586>, 2022.

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High climatological cloud cover limits its response to aerosols in ICON-HAM

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Abstract.

Marine low-level clouds substantially cool the climate and are sensitive to aerosols. Observations suggest strong cooling from cloud fraction (CF) adjustments, yet global climate models (GCMs) may underestimate this process. We apply explainable machine-learning with SHapley Additive exPlanations (SHAP) to the ICOSahedral Nonhydrostatic atmosphere model coupled with the Hamburg Aerosol Module (ICON-HAM) to quantify CF sensitivity to cloud droplet number concentration (N_d). We analyze six experiments combining two cloud cover parameterizations, an RH-only scheme (CC–RH) and a cloud-water-dependent scheme (CC–RH–LWC), with three prescribed lower bounds for N_d ($N_{d,min} = 10, 40, 100 \text{ cm}^{-3}$). The $\ln N_d$ –CF relationships in ICON–HAM are more nonlinear than in satellite observations and exhibit saturation. We quantify sensitivities using piecewise linear regression (PLR) separating low- and high- N_d regimes. Sensitivities are stronger in the low- N_d regime, while inter-scheme and inter- $N_{d,min}$ differences are negligible at high N_d . Unexpectedly, CC–RH–LWC shows weaker sensitivity than CC–RH at high $N_{d,min}$, despite explicit CF–cloud water coupling. We attribute this to a “headroom effect”: higher $N_{d,min}$ suppresses autoconversion and enhances liquid water path in both schemes, but CC–RH–LWC converts this into higher mean CF, limiting headroom for aerosol-induced CF increases. These findings are robust across three PLR breakpoint strategies. Our results demonstrate that GCM CF responses are state-dependent and strongly influenced by model configuration choices, which can pre-saturate cloud cover responses and true CF adjustment magnitudes. We suggest that model–observation comparisons should account for baseline mean CF to avoid misinterpretation.



1 Introduction

More than 23 % of the Earth's ocean surface is covered by marine low-level clouds (MLCs) (Wood, 2012). Due to the small temperature difference between MLC tops and the ocean surface, their impact on outgoing longwave radiation is limited. However, MLCs greatly reflect incoming shortwave radiation and thus exert a strong net cooling effect on the climate system (Hartmann et al., 1992). Because they are so widespread over the dark ocean surface, MLCs are extremely important for the Earth's radiative balance (Wood, 2012; Chen et al., 2014). Anthropogenic aerosols modify the radiative effect of MLCs by acting as cloud condensation nuclei (CCN), thus influencing the coverage and properties of clouds. Compared to continental clouds and other marine cloud types, MLCs tend to be particularly responsive to aerosol perturbations (Wood et al., 2015) owing to their large areal coverage, the prevalence of relatively optically thin clouds (optical depth <3) (Leahy et al., 2012), and their development in relatively clean regions with less anthropogenic aerosol influence (Platnick and Twomey, 1994).

Cloud droplet number concentration (N_d) increases with more atmospheric aerosols, leading to smaller cloud droplet effective radii, assuming a constant liquid water path (LWP), thus leads to brighter clouds and an instantaneous negative top-of-atmosphere (TOA) radiation change (Twomey effect, (Twomey, 1977)). Subsequently, the reduced collision and coalescence efficiency of the smaller-sized droplets delays precipitation and prolongs the lifetime of clouds, ultimately resulting in a greater cloud fraction (CF) and LWP (Albrecht, 1989). However, particularly in non-precipitating clouds, reducing droplet sizes can also reduce CF and LWP by enhancing cloud-top entrainment, either via shorter evaporation timescales (Wang et al., 2003; Small et al., 2009) or via decreased sedimentation, which can retain liquid water in the entrainment zone (Ackerman et al., 2004; Bretherton et al., 2007). These cloud processes that exert influence independent of surface air temperature changes are referred to as cloud adjustments (Boucher et al., 2013; Sherwood et al., 2015), including CF and LWP adjustments. The instantaneous effect of aerosols on the TOA radiation balance due to changes in N_d , and the subsequent CF and LWP adjustments together are known as effective radiative forcing from aerosol-cloud interactions (ACI), contributing to a large source of uncertainty in the quantification of climate change (Forster et al., 2021; Watson-Parris and Smith, 2022).

Over the past decade, systematic differences in the magnitude of cloud adjustments have been found between models and observations (Gryspeerdt et al., 2020; Quaas et al., 2024). In the most recent two eras of the Coupled Model Intercomparison Project (CMIP5 and CMIP6) in particular, GCMs consistently show strong cooling driven by the Twomey effect and LWP adjustments (Smith et al., 2020; Zelinka et al., 2023). By comparison, observational studies generally find a weak or ambiguous LWP response to aerosol perturbations (Malavelle et al., 2017; Bender et al., 2019; Toll et al., 2019).

For the CF adjustment, observational studies agree on the sign while disagree on the magnitude of the CF sensitivity to aerosol (proxies) or resulting negative forcing (Gryspeerdt et al., 2016; Andersen et al., 2017; Fuchs et al., 2018; Rosenfeld et al., 2019; Wall et al., 2022; Jia et al., 2024). GCMs agree with observational studies in terms of the positive relationship between cloud cover and aerosols and therefore a negative radiative forcing from the CF adjustment (Zelinka et al., 2014; Forster et al., 2021), as autoconversion parameterizations in many of these models allow aerosols and thus N_d to suppress precipitation and thereby prolong cloud lifetime (Bellouin et al., 2020). However, while models agree on the sign of the CF



adjustment, they diverge on the magnitude and relative importance of the Twomey effect, the LWP adjustment and the CF adjustment.

GCMs simulate a minimal or negligible CF contribution relative to the Twomey effect and LWP adjustment (Smith et al., 2020; Zelinka et al., 2023). Even when diagnosed using a consistent cloud radiative kernel method, multiple CMIP6 models suggest a CF adjustment being weaker than both the Twomey effect and LWP adjustment (Duran et al., 2025), whereas the same framework applied to satellite observations indicates that the CF adjustment outweighs both (Wall et al., 2023). Consistent with this, a recent model–observation comparison (Wang et al., 2025) showed that multiple GCMs failed to reproduce the strong aerosol-induced increase in cloud cover associated with the Holuhraun volcanic eruption documented by Chen et al. (2022). Taken together, the discrepancies in the magnitudes and relative importance of LWP and CF adjustments suggest that LWP adjustments are likely overestimated and CF adjustments are underestimated in GCMs. In particular, recent observational evidence increasingly highlights the CF adjustment as potentially the largest contributor to effective radiative forcing due to ACI (e.g. Chen et al., 2022; Yuan et al., 2023; Wall et al., 2023; Chen et al., 2024), providing a key motivation for this present work. Moreover, the CF response to aerosols may saturate under already polluted conditions, further complicating its quantification. For example, Wang et al. (2024) found over high-latitude oceans that the CF responses saturate once N_d exceeds around 60 cm^{-3} , suggesting that background cloud conditions influence the CF adjustment.

The challenge comes from the fact that observational constraints on the CF adjustment often rely on opportunistic experiments, such as ship tracks and volcanic events, which can rule out confounding meteorological covariates. However, ship tracks are typically subscale for GCMs, while volcanic eruptions can be episodic and often involve uncertain emissions, making the quantification challenging. Therefore, both approaches may lack global representativeness. The alternative is to use large-scale satellite studies, which are spatiotemporally representative and good for constraining GCMs, but use different ways of dealing with meteorological confounders. For example, Gryspeerdt et al. (2016) has attempted to address this confounding issue through statistical control of confounding variables such as relative humidity (RH), or Andersen et al. (2023) has incorporated them into a machine-learning (ML) framework using ridge regression.

In a previous study (Jia et al., 2024, hereafter J24), we quantified the CF adjustment as the sensitivity of MLC CF to N_d using an explainable ML framework trained on Moderate Resolution Imaging Spectroradiometer (MODIS) observations and reanalysis data from the European Centre for Medium-Range Weather Forecasts. This eXtreme Gradient Boosting (XGBoost) and SHapley Additive exPlanation (SHAP) framework demonstrated global positive N_d –CF relationships.

In this study, we extend the explainable ML framework of J24 to Atmospheric Model Intercomparison Project (AMIP) simulations with the ICOSahedral Nonhydrostatic (ICON) atmosphere model coupled with the Hamburg Aerosol Module (HAM) (ICON-HAM). The main goals of this study are twofold: (1) to enable a consistent comparison between observation- and model-based N_d –CF sensitivities; (2) to investigate how structural model choices shape the simulated CF response. To achieve this, we conduct six simulations that combine two cloud-cover schemes (CC-RH, dependent only on RH, and CC-RH-LWC, additionally dependent on cloud water content) with three prescribed lower bounds for cloud droplet number concentration ($N_{d,\min} = 10, 40$ and 100 cm^{-3}), examining their joint impact. As in J24, we quantify the sensitivity of MLC CF to log-transformed N_d ($\ln N_d$) using regional XGBoost models with SHAP, extended via a piecewise linear regression (PLR) to



accommodate the stronger nonlinear response in ICON-HAM. Guided by these goals, our hypotheses are: (1) the $\ln N_d$ –CF relationships diagnosed from ICON-HAM differ quantitatively, or maybe even qualitatively, from the MODIS-based relationships in J24; (2) given the stronger coupling between cloud water content and cloud cover in CC-RH-LWC, we expect this scheme to exhibit larger $\ln N_d$ –CF sensitivities than the RH-only scheme with prescribed $N_{d,min}$ expected to further modulate these sensitivities differently in each scheme.

2 Data and Methods

2.1 ICON-HAM experimental setup

2.1.1 ICON-HAM general model description

The host model in this study is the ICON atmospheric model (Giorgetta et al., 2018), coupled with HAM to obtain a global atmosphere-aerosol model. The overall coupled model configuration largely follows that described in Salzmann et al. (2022), although we use a more recently developed ICON version 2.6.4 combined with the same HAM version 2.3 as in their study. Hereafter, without explicit notation, ICON-HAM refers to ICON2.6.4–HAM2.3. The ICON dynamical core provides a nonhydrostatic, compressible atmosphere on an icosahedral grid. HAM simulates aerosol properties such as size distributions and couples them to clouds and radiation, enabling process-level links between aerosol perturbations and cloud responses. In this study, the ICON-HAM model uses an R02B04 horizontal grid, corresponding to a grid spacing of approximately 160 km between neighbouring grid points, with 47 vertical levels using a hybrid sigma-height coordinate system. A code fix to the total turbulent energy scheme has been implemented to correct an issue introduced when porting from ECHAM to ICON, which caused excessive vertical diffusion in earlier model versions (Giorgetta et al., 2022; Franke and Giorgetta, 2024). This correction ensures a more realistic turbulent contribution to updraft velocity and thus cloud droplet activation.

To explore how CF sensitivity to aerosol perturbations is represented in ICON-HAM, we use N_d as a proxy for cloud-active aerosol, consistent with recent satellite-based studies (e.g. Gryspeerd et al., 2016; Christensen et al., 2020; Chen et al., 2024). All simulations in this study follow the AMIP protocols, with sea-surface temperatures (SST) and sea ice prescribed from observations (Taylor et al., 2000). We conduct six simulations that pair two different cloud cover parameterization schemes with three different prescribed lower bounds for cloud droplet number concentration ($N_{d,min}$). The following subsections detail the specific configurations of these simulations.

2.1.2 Cloud cover and autoconversion parameterization schemes

A central aspect of this study involves comparing two parameterizations for fractional cloud cover that fundamentally differ in whether cloud water directly controls cloud cover. Throughout this study, we refer to these schemes as CC-RH, which depends only on grid-mean RH, and CC-RH-LWC, which additionally depends on the total cloud water content.



115 CC-RH follows the implementation in the ECHAM6.3 general circulation model as described by Mauritsen et al. (2019), and is also the default configuration in ICON-HAM. In this scheme, CF is diagnosed as a function of grid-mean RH (Sundqvist et al., 1989):

$$CF = 1 - \sqrt{1 - \frac{RH - RH_0}{1 - RH_0}} \quad (1)$$

where RH_0 is a critical threshold RH specified as a function of height, as originally formulated in ECHAM4 (Roeckner et al., 1996). Clouds begin to form within a grid cell once RH_0 is exceeded. Further details on the parameter settings and tuning of CC-RH can be found in Mauritsen et al. (2019).

The CC-RH-LWC scheme follows the parameterization proposed by Xu and Randall (1996), previously referred to as COUPL-CC by Lohmann and Feichter (1997). In this scheme, CF increases with RH and additionally with the cloud water content q_w (the sum of liquid and ice water mixing ratios, which is predominantly liquid for MLCs) through an exponential factor:

$$CF = RH^{0.25} \left[1 - \exp \left(-250 [(1 - RH)q_{vs}]^{-0.45} q_w \right) \right] \quad (2)$$

with q_{vs} being the saturation specific humidity. The key distinction is that CC-RH-LWC explicitly depends on cloud water content, whereas CC-RH does not. This fundamental difference allows aerosol-induced changes in autoconversion and precipitation, which affect cloud water and hence LWP, to directly feed back on CF in CC-RH-LWC, a process that is absent in CC-RH.

All experiments use the autoconversion parameterization scheme of Khairoutdinov and Kogan (2000), derived from bin-resolving microphysics in large-eddy simulations (LES) of marine stratocumulus. Although originally developed for LES applications, it has been adopted in many GCMs. In what follows, this scheme is referred to as KK2000. The KK2000 autoconversion rate has a relatively strong inverse dependence on N_d ($N_d^{-1.79}$), and it scales with cloud liquid water content, allowing for interactions with the cloud-cover schemes described above.

2.1.3 Prescribed $N_{d,min}$

It is a common practice in global aerosol-climate models to impose $N_{d,min}$ throughout the modelled atmosphere at all locations and altitudes. This constraint is typically imposed to avoid unrealistically low N_d values that may arise from the incomplete representation of natural background aerosol species that could contribute to CCN and N_d (e.g. nitrate and biological particles) (Kirkevåg et al., 2008), or from potential underprediction of specific aerosol sources (e.g. fine-mode sea salt) (Wang and Penner, 2009).

However, this practice is unphysical and the specific choice of the lower bound value is somewhat arbitrary. Hoose et al. (2009) systematically investigated the influence of these lower bounds and found that they can suppress the simulated aerosol indirect effect by up to 80 % by artificially creating more uniform N_d fields. By design, these N_d constraints are triggered



145 more frequently where low N_d is more common, even though the observed N_d can drop well below the imposed threshold. This reduces the contrast between clean and polluted regimes. Consequently, under pre-industrial emissions the impact of prescribing $N_{d,min}$ is comparably large over land and ocean, whereas under present-day emissions, it is most evident over oceans (Hoose et al., 2009).

Typical values of this lower bound have ranged between 5 and 40 cm^{-3} across different models (Hoose et al., 2009).
 150 ECHAM4 and ECHAM5 have used a $N_{d,min}$ value of 40 cm^{-3} (Lohmann et al., 1999, 2007), and ECHAM6.3–HAM2.3 added an additional option of 10 cm^{-3} (Neubauer et al., 2019). In our study, in addition to 10 and 40 cm^{-3} , we also impose a more stringent $N_{d,min}$ of 100 cm^{-3} to further examine how a prescribed minimum N_d interacts with CC-RH and CC-RH-LWC cloud cover schemes and modulates the CF sensitivity, yielding six experiments in total.

2.1.4 ICON-HAM output data as ML input

155 In this study, we apply an explainable ML framework similar to that used in J24 (described below in Sect. 2.2), where N_d along with large-scale meteorological fields are used to predict CF. The data used as input for the XGBoost models here are ICON-HAM model outputs. Predictors for the XGBoost models are listed in Table A1. To facilitate direct comparison with our MODIS-based study J24, we remapped ICON-HAM outputs on the native icosahedral grid to the MODIS $1^\circ \times 1^\circ$ latitude-longitude grid using nearest-neighbour interpolation. The original 47 vertical levels in ICON-HAM’s hybrid sigma-
 160 height coordinate system were interpolated to six desired standard pressure levels (1000, 950, 900, 850, and 700 hPa) using conservative remapping. Owing to storage and computational constraints associated with producing global hourly output, the analysis is restricted to five years of data (2011–2015).

The low-level CF used here to approximate MLCs is diagnosed as the fractional cover of liquid clouds with cloud-top temperature warmer than 268 K, as viewed from the top of the atmosphere whenever high- and mid-level clouds were absent.
 165 This “unobscured” diagnostic is considered more suitable for analyzing drivers of liquid cloud cover, because the coarser resolution of ICON-HAM relative to MODIS increases the likelihood of overlying clouds masking the true extent of the low-level CF. Unlike our observational study (J24), where N_d was derived specifically for satellite retrievals from cloud droplet effective radius and cloud optical depth, here we use the model’s instantaneous cloud-top, in-cloud N_d diagnostic. To improve comparability with J24, we apply analogous filtering criteria here: Low clouds must have a cloud-top temperature warmer than
 170 268 K; cases with a cloud-top in-cloud effective radius smaller than 4 μm or a cloud optical depth smaller than 4 are excluded.

To characterize the dynamic and thermodynamic state of the atmosphere, the ML framework uses 23 meteorological variables in addition to $\ln N_d$ as predictors in the ML model. This selection largely follows J24 except for several adaptations described below. A full list of predictors is provided in Table A1. We diagnose boundary-layer height (BLH) as the lowest model level at which the mean RH first drops below 50 %, following a standard LES-based proxy for the stratocumulus top
 175 and inversion height (Bretherton et al., 2013), which effectively captures the sharp transition from the moist boundary layer to the dry free troposphere above. This threshold has been used in a recent ICON-based analysis of stratocumulus adjustments and was shown to align well with the modelled cloud top (Fons et al., 2024). Convective available potential energy (CAPE) was dropped from the predictor set because it is primarily relevant for deep convective processes rather than for the stratiform



and shallow cumulus marine boundary-layer clouds that are the focus of this study. Moreover, in addition to RH at 700 and
 180 850 hPa, we include two more lower pressure levels, 900 and 950 hPa, which are considered more relevant for MLCs.

Unlike J24, we do not standardize the predictor variables in this work. In J24, standardisation placed all predictors on a
 common and unitless scale to allow direct comparisons among all predictors with different units. However, our objective here
 is to compare the $\ln N_d$ -CF sensitivity across the six ICON-HAM configurations (two different cloud cover schemes and three
 $N_{d,min}$ values), rather than to rank predictors by relative importance. We hence retain predictors on their native physical scales
 185 to preserve the interpretability of ICON-HAM's CF responses across six experiments in this study.

2.2 Explainable Machine Learning framework

2.2.1 Regional tree-based machine-learning models

We adopt the explainable machine learning framework from J24 to quantify the sensitivity of MLC CF to $\ln N_d$ (proxy for
 aerosol) while accounting for meteorological covariability and confounding effects. A tree-based ensemble XGBoost algorithm
 190 (Chen and Guestrin, 2016) is employed due to its ability to capture nonlinear relationships and model feature interactions, while
 achieving high predictive performance with computational efficiency for tabular data. As in J24, an independent XGBoost
 model is trained within each $5^\circ \times 5^\circ$ geographical window to predict CF, using spatiotemporal samples aggregated from $1^\circ \times 1^\circ$
 grid cells (remapped from native icosahedral grids) to capture regional characteristics and relationships.

For better comparability with our MODIS-based study (J24), we configure ICON-HAM to output hourly variables, then
 195 match them with MODIS Terra overpass times. The XGBoost models are trained on these outputs from 2011 to 2015 (Sect. 2.1.4.
 Unlike J24, where the train–test split was purely chronological, here we ensure that different climate states are represented in
 the training data. To sample distinct ENSO phases, we train the models on a La Niña year (2011), a neutral SST year (2013),
 and an El Niño year (2015), consistent with the documented ENSO conditions during these periods (Null, 2025). Model testing
 is performed on the intermediate years 2012 and 2014. The spatial patterns of annual SST anomalies relative to the five-year
 200 mean are shown in Fig. S1.

As in J24, hyperparameters for each regional XGBoost model are tuned via Bayesian optimization with five-fold cross-
 validation, and the number of boosting rounds is determined by early stopping to prevent overfitting. In this study, we ex-
 pand the hyperparameter tuning to include XGBoost's complete family of column-wise subsampling hyperparameters *col-*
sample_bylevel and *colsample_bynode* in addition to *colsample_bytree* used in J24. These parameters control the fraction of
 205 features randomly selected at different stages of tree construction (per depth level, per node and per tree, respectively). With
 these subsampling parameters, the XGBoost model tends to spread feature importance more evenly across correlated features.
 In this way, we could prevent unphysical overfitting to any single predictor and obtain more robust SHAP attributions that
 reflect the collective influence of intercorrelated variables. The XGBoost models from J24 were re-tuned in this study based
 on this more comprehensive hyperparameter set. Thus, the MODIS-based results presented here are marginally different from
 210 those reported in J24.



2.2.2 Explaining the machine-learning models

The SHAP approach is used to explain the regional XGBoost models. SHAP provides additive, local attributions of each predictor to the CF prediction (Lundberg et al., 2018, 2020). SHAP decomposes every individual prediction into a base value and contributions from each feature, allowing the specific influence of $\ln N_d$ on CF to be isolated while accounting for the influences of meteorological covariates. The SHAP base value is what the model would predict with no feature information (Lundberg and Lee, 2017), typically calculated as the average prediction over the entire training set. Positive (negative) SHAP values indicate that a predictor raises (lowers) the prediction relative to this baseline. In our framework, the SHAP base value can be interpreted as the climatological mean CF over the training dataset in a given geographical window, representing the expected XGBoost prediction given the mean state of meteorology and N_d . Each feature's SHAP value then quantifies the magnitude and direction (positive or negative) that the specific feature value contributes to the CF prediction, shifting the final prediction away from this mean baseline. By construction, the sum of all SHAP values plus the base value equals the final prediction of the model, ensuring additivity and consistency.

Following J24, plotting SHAP values of $\ln N_d$ against its feature values yields a SHAP dependence plot where the slope is taken as the $\ln N_d$ –CF sensitivity for a given geographical window. For the ICON-HAM analysis, however, a methodological adaptation is required: instead of a single linear regression fit, piecewise linear regression (PLR) is applied. This choice is motivated by the stronger nonlinear (saturating) behaviour in the ICON-HAM–based $\ln N_d$ –CF relationships compared with those in J24. In ICON-HAM, the CF response to $\ln N_d$ diminishes at higher $\ln N_d$ even after the logarithmic transformation, reflecting a saturation effect that largely disappeared after log transformation in J24. The PLR approach retains comparability to J24's method while allowing the nonlinearities to be captured. A complete overview of the XGBoost+SHAP global framework is provided in J24. Further details on the ICON-HAM nonlinearities and rationale for PLR are discussed in Sect. 3.2, and the breakpoint selection strategies for data segmentation are detailed in Sect. 2.3 and Appendix B.

2.3 Piecewise linear regression with constraint-aware breakpoints

Here we introduce a constraint-aware breakpoint with fixed interval in logarithmic space, which is the primary strategy used in our main analysis. Specifically, it is called “constraint-aware” because it defines the breakpoint relative to the prescribed minimal constraint on cloud droplet number ($N_{d,\min}$). This strategy sets the fixed interval between $N_{d,\min}$ and the breakpoint directly in the predictor space of $\ln N_d$:

$$\ln(N_{d,bp}) = \ln(N_{d,\min}) + \delta \quad (3)$$

where $N_{d,bp}$ denotes the breakpoint and δ represents the logarithmic offset from the imposed lower bound. In linear space this gives $N_{d,bp} = N_{d,\min} \times e^\delta$. In this study, we fix the log-space interval $\delta = 0.1$, so $N_{d,bp} \approx N_{d,\min} \times 1.105$, yielding breakpoints of approximately 11, 44, and 111 cm^{-3} for the $N_{d,\min} = 10, 40$, and 100 cm^{-3} experiments, respectively. Using a constant increment in logarithmic space enforces the same relative (multiplicative) distance from $N_{d,\min}$ across all experiments and



is consistent with logarithmic scaling of the $\ln N_d$ predictor. By placing the breakpoints $\sim 10.5\%$ above the imposed lower bounds, this approach is designed to effectively capture the transition from a regime controlled by the lower bound, and a regime free from the lower bound.

245 This PLR framework is applied to the $\ln N_d$ SHAP dependence plots for each $5^\circ \times 5^\circ$ geographical window. Data are segmented at the identified breakpoint, and separate linear regressions are fitted for the below- and above-breakpoint regimes to yield slopes β_b and β_a , respectively. For numerical stability, we require at least 1000 data points in the below-breakpoint regime and 500 in the above-breakpoint regime for each geographical window. A detailed visual demonstration of this PLR approach applied to an exemplary geographical window is presented later in Sect. 3.3.1. To assess robustness, we test two
 250 alternative breakpoint strategies: a constraint-aware approach with fixed intervals in linear space, and a statistically optimized approach that minimizes fitting error within a constraint-aware search range. These two alternative strategies and their results are detailed in Appendix B.

3 Results and discussion

3.1 Mean states of marine low clouds in ICON-HAM

255 Here we present how imposing lower-bound constraints of 10, 40 and 100 cm^{-3} elicits distinct responses in precipitation, liquid water in clouds, and cloud cover across the two cloud cover parameterization schemes (CC-RH and CC-RH-LWC). Figure 1 presents the global distributions of the mean precipitation rate at the surface (PR), LWP and CF for each $5^\circ \times 5^\circ$ geographical window.

As $N_{d,\min}$ increases, autoconversion is suppressed, leading to a reduction in the large-scale precipitation (Fig. 1a). This
 260 arises because higher $N_{d,\min}$ yields clouds populated by more numerous and smaller droplets, which is less favourable for drizzle formation. The KK2000 autoconversion scheme captures this effect through its relatively strong inverse dependence on N_d (Khairoutdinov and Kogan, 2000). Even the cleanest clouds within each experiment (with N_d at or slightly above $N_{d,\min}$) become inefficient in producing rain, allowing more water to accumulate in the clouds and thus resulting in higher LWP (Fig. 1b). We see that PR decreases from $N_{d,\min} = 10$ to 40 cm^{-3} and then levels off, while LWP continues to increase
 265 throughout the entire range. This decoupling occurs because the LWP represents a reservoir of cloud water, whereas PR is a flux at the surface. Weakening the internal cloud process of autoconversion directly helps build up the reservoir, but changes in its level do not necessarily scale with changes in the surface flux. Moreover, PR is balanced by surface evaporation, which is mainly governed by surface energy budget and hence by surface temperature (Held and Soden, 2006). Since SSTs are prescribed in our AMIP simulations, oceanic evaporation is relatively constrained, limiting and converging PR changes.

270 By construction, the two cloud-cover schemes produce contrasting CF responses. In CC-RH-LWC, the explicit dependence of CF on cloud water content means the rising LWP at high $N_{d,\min}$ translates directly into a steady increase in CF. Meanwhile, CC-RH, relying solely on grid-mean RH, shows a decline followed by a plateau that mirrors the PR trend. This behaviour likely results from higher $N_{d,\min}$ leading to smaller droplets and suppressed precipitation formation. Smaller droplets reduce sedimentation and also evaporate faster at the cloud top, enhancing cloud-top entrainment. Meanwhile, less drizzle evaporates

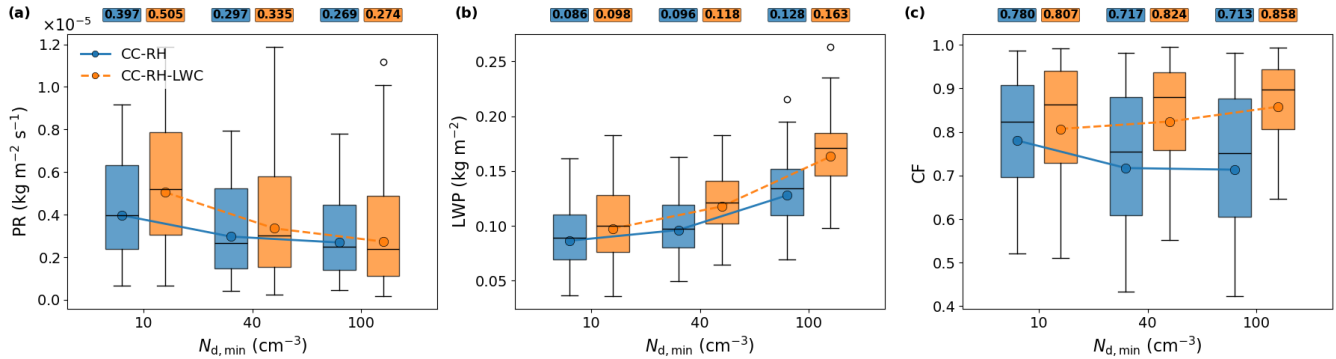


Figure 1. Distributions of $5^\circ \times 5^\circ$ geographical-window-mean precipitation rate (PR; panel a), liquid water path (LWP; panel b) and cloud fraction (CF; panel c) over the global oceans for marine liquid clouds during 2011–2015. The simulations compare two cloud cover parameterization schemes (CC-RH and CC-RH-LWC) and three imposed minima of cloud droplet number concentration ($N_{d,min} = 10, 40, 100 \text{ cm}^{-3}$). Each window’s value is the spatiotemporal mean of the aggregated $1^\circ \times 1^\circ$ grid cells. The blue boxes and solid lines represent the results from the CC-RH scheme, while the orange boxes and dashed lines represent the CC-RH-LWC scheme. Each box shows the interquartile range. Whiskers expand the boxes by up to 1.5 interquartile ranges, and individual points are outliers. Median values are represented by the horizontal lines, while the global area-weighted mean values are represented by circular markers and overlaid lines.

below the cloud and more condensate is retained in the cloud layer, diminishing sub-cloud evaporative moistening. Both act to lower grid-mean RH. While similar drying can occur in CC-RH-LWC, it is offset by the LWP-driven CF increase, yielding an overall rise in cloud cover.

The stronger increase in mean-state CF with the higher artificial N_d floor seen in CC-RH-LWC (relative to CC-RH) is qualitatively consistent with Lohmann and Feichter (1997). In their study, when aerosol loading increased from preindustrial (PI) to present-day (PD), the cloud-water-dependent scheme (the same as our CC-RH-LWC) showed a roughly 3 % CF increase under elevated aerosol emission levels, compared with only about 1 % in their RH-only scheme (conceptually similar to our CC-RH). Although their mechanisms differ from ours (emission changes versus prescribed lower bounds), both approaches show that CC-RH-LWC’s explicit dependence on cloud water content amplifies the CF response when autoconversion is suppressed by increased N_d and liquid water accumulates. We therefore interpret the divergence between schemes in Fig. 1c as consistent in sign with Lohmann and Feichter (1997), even though $N_{d,min}$ experiments are not perfect PI and PD analogues. This supports the view that the pronounced CF response in CC-RH-LWC stems from its direct coupling between cloud cover and liquid water, which becomes particularly evident when autoconversion is weakened due to autoconversion’s dependence on droplet number, which $N_{d,min}$ directly affects.

3.2 Observations vs. default ICON-HAM setup using one linear regression

We first compare the CF sensitivity to $\ln N_d$ globally based on the ICON-HAM default model setup (CC-RH cloud cover parameterization with $N_{d,min} = 40 \text{ cm}^{-3}$) to that of the observational results of J24. The sensitivities here are quantified

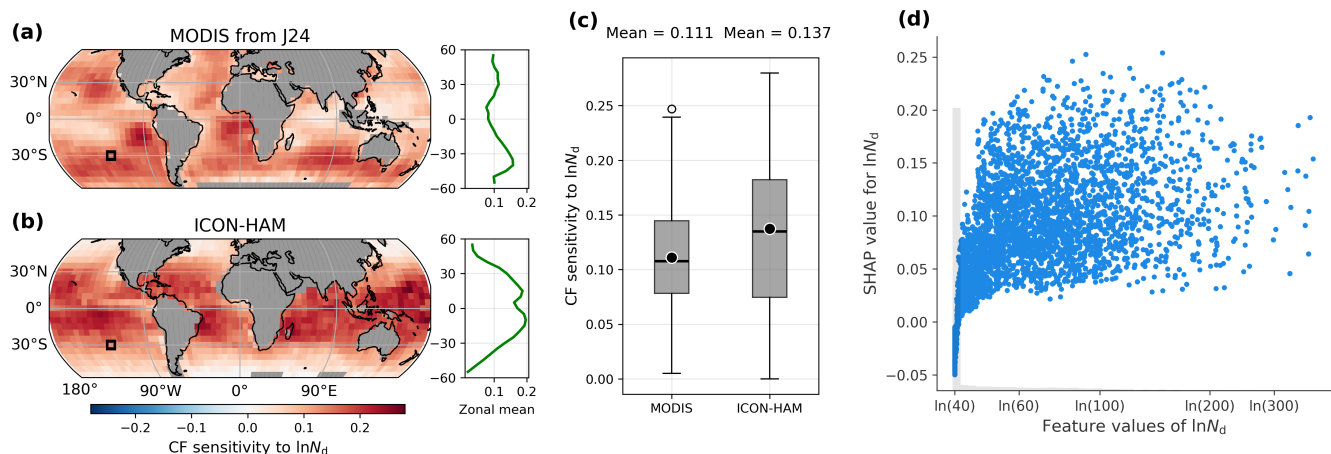


Figure 2. Comparison of marine low-cloud fraction (CF) sensitivity to log-transformed cloud-droplet number concentration ($\ln N_d$) between MODIS-based results from J24 (recomputed here with retrained XGBoost models using extended column-wise subsampling hyperparameters; see Sect. 2.2.1) and ICON-HAM simulations using the default CC-RH cloud-cover scheme with $N_{d,\min} = 40 \text{ cm}^{-3}$. Panels (a) and (b) show spatial patterns and zonal means of the observed and modelled $\ln N_d$ –CF sensitivities, respectively. (c) Distributions of the CF sensitivities; thick black lines denote medians, and black dots show area-weighted global means (values above boxes). (d) ICON-HAM SHAP dependence plot for $\ln N_d$ in the exemplary geographical window at 27.5°–32.5°S, 122.5°–127.5°W, whose location is marked by squares in panels (a) and (b). Grey bars in the background indicate the distribution of $\ln N_d$ feature values.

following the approach in J24, defined as the slope derived from an ordinary least-squares linear regression of the $\ln N_d$ SHAP values against the $\ln N_d$ feature values for each $5^\circ \times 5^\circ$ geographical window.

MODIS-based results (Fig. 2a) show more spatially heterogeneous CF sensitivities, strongest in the stratus-to-cumulus transition regions off western coasts and in the Southern Hemispheric midlatitudes. In contrast, ICON-HAM displays more widespread and contiguous areas of high $\ln N_d$ –CF sensitivity, pronounced over nearly all subtropical regions, including stratocumulus regions, trade cumulus regions, and tropical ascending regions (Fig. 2b). Unlike MODIS results, the regions with the strongest sensitivities in the Pacific and Atlantic Oceans are located more westward, away from the west coasts. Zonal means show that both ICON-HAM and MODIS exhibit weaker sensitivities near the equator compared to subtropical regions, likely reflecting the greater dominance of deep convection and strong meteorological variability in the tropics. These dynamical factors limit the prevalence of stratiform liquid clouds and obscure the signal of aerosol–CF relationships compared to the extensive subtropical stratocumulus decks. Another major difference appears in the Southern Hemispheric midlatitudes, where ICON-HAM shows weak sensitivities. The geographic differences in sensitivity patterns suggest that key controlling processes may differ between observations and ICON-HAM, notably regions where the model frequently computes shallow convection. For instance, differences in droplet size distributions can alter entrainment–evaporation feedbacks and precipitation efficiency, thereby shifting the spatial pattern of the sensitivity.



The distributions of the CF sensitivity values (Fig. 2c) show that the global weighted mean sensitivities are of similar magnitude in ICON-HAM (0.137) and MODIS (0.111), with ICON-HAM being somewhat higher and exhibiting greater variability. Note the value of 0.111 differs slightly from that reported in J24 because XGBoost models were re-tuned using the more comprehensive hyperparameters in this study (Table A1). The global weighted mean value of ICON-HAM is similar to the MODIS-based estimate but higher than the values reported for other GCMs including ECHAM (Wang et al., 2025). This does not necessarily contradict the broader expectation discussed in the Introduction that models tend to underestimate aerosol–cloud forcing relative to observation-based constraints, as the sensitivity does not translate directly to radiative forcing. However, the larger simulated sensitivities motivate further investigation of their origin (e.g. at individual geographical window) and of potential methodological limitations when applying SHAP with a single linear regression for ICON-HAM.

In light of this, we examine the relationship between predicted CF and $\ln N_d$ in the same exemplary geographical window as in J24 (Fig. 2d). The ICON-HAM relationship is clearly not as linear as the one based on MODIS. The cloud cover saturation effect, or the dependence of cloud cover sensitivity on background N_d , has already been documented: CF increases quickly with increasing N_d , but the rate of increase approaches zero after reaching an N_d threshold at approximately 60 cm^{-3} (e.g. Hu et al., 2021; Yuan et al., 2023). This effect can be reflected by a sharply decreasing derivative (dCF/dN_d in Yuan et al. (2023) or $dSHAP/dN_d$ in J24) before the saturation threshold and a nearly flattened curve after the threshold is exceeded.

As shown in J24, using MODIS data, this effect only manifests evidently as a nonlinear relationship between the N_d feature values and their corresponding SHAP values, which becomes much more linear after applying a logarithmic transformation to N_d . In ICON-HAM data for the same geographical window, the relationship remains strongly nonlinear and the CF saturation effect persists even after the logarithmic transformation (Fig. 2d). This more nonlinear CF response in ICON-HAM likely arises because ICON-HAM’s cloud-cover and microphysics schemes are formulated based on process-level understanding of the small-scale nonlinearities observed, for example, in field campaigns (Lohmann and Feichter, 1997) and LES constrained by such field campaigns (Khairoutdinov and Kogan, 2000). At these small scales, cloud systems can shift abruptly once critical thresholds are crossed, resulting in step-like responses. Since these parameterizations include physical constraints such as thresholds and saturation limits, the resulting model output retains its inherent nonlinearity, and this signature of its core physics persists even after aggregation from $1^\circ \times 1^\circ$ grid cells to $5^\circ \times 5^\circ$ geographical windows.

At larger scales, by contrast, the abrupt CF responses that arise from microphysical thresholds are smoothed out. This occurs because higher aerosol concentrations delay the breakup of cloud decks, prolonging their lifetime. As winds advect these systems, the temporal delay in cloud dissipation allows cloud cover to persist over a longer track, which effectively translates into a spatial extension of cloudiness. Consequently, under spatial aggregation across many sampled cloud scenes, a larger fraction of the sampled domain remains cloudy, the spatially aggregated CF response appears gradual and more linear (Goren et al., 2019, 2022; Rosenfeld et al., 2023). This linearity is precisely the characteristic behaviour observed in large-scale satellite analyses, including our previous MODIS-based study (J24).

The simple linear regression between SHAP values and feature values of $\ln N_d$ used in J24 to quantify the $\ln N_d$ –CF sensitivity is not suitable for ICON-HAM output, where the relationships are strongly nonlinear. To account for these while maintaining comparability with J24’s methodology and other linear sensitivity estimates, we apply the PLR approach with a



breakpoint strategy defined in Sect. 2.3, similar in spirit to the segmented fitting approach applied to the nonlinear N_d –LWP relationships by Gryspeerdt et al. (2019). This breakpoint strategy ensures a consistent proportional offset from the imposed lower bounds across all simulations, which is more appropriate given the logarithmic nature of the $\ln N_d$ predictor. Physically, this approach is intended to effectively capture the transition from a near- $N_{d,min}$ (lower-bound-constrained) regime with strong CF sensitivity to $\ln N_d$ to a high N_d regime with a weaker CF response. To illustrate how this strategy operates with PLR based on SHAP dependence plots, we zoom in on the SHAP dependence for all six simulations in the same exemplary geographical window as Fig. 2d; the detailed visualization and discussions are provided in Sect. 3.3.1.

To assess the influence of breakpoint selection on our results, we evaluated two additional strategies beyond the default strategy described above. The first is an alternative constraint-aware approach with a fixed interval on a linear scale of 5 cm^{-3} (Appendix B1). The second is a data-driven approach where the breakpoint is statistically optimized within a constrained search range extending 50 cm^{-3} above each $N_{d,min}$ value (Appendix B2). This hybrid design combines statistical data-driven flexibility with the $N_{d,min}$ constraints. The full methodology and global results for these alternative strategies are presented in Appendix B.

3.3 From ICON-HAM physical states to SHAP-detected sensitivity: the headroom mechanism

3.3.1 Piecewise sensitivity and headroom effect at the exemplary geographical window

Figure 3 illustrates the application of PLR with constraint-aware breakpoints $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$ at the exemplary geographical window. The panels confirm the pronounced nonlinearity in the $\ln N_d$ –CF relationship, which justifies our piecewise approach. First, the slopes on either side of the breakpoint (vertical coloured dashed lines) contrast systematically: the below-breakpoint regime has a stronger sensitivity (β_b) than the above-breakpoint regime (β_a) across all experiments. The summary panels (d) and (h) make the two regimes and the associated saturation effect visually explicit. Second, regime occupancy shifts with $N_{d,min}$ and gets markedly asymmetric at high $N_{d,min}$: At $N_{d,min} = 10 \text{ cm}^{-3}$, samples are split roughly evenly between the two regimes. However, at 40 and 100 cm^{-3} , the vast majority fall below the breakpoint, with the below-breakpoint fraction increasing with $N_{d,min}$, reflecting that more data are truncated by the higher $N_{d,min}$ constraints and thus concentrated near the imposed bounds.

The below-breakpoint slopes vary systematically with both $N_{d,min}$ and cloud-cover scheme, whereas the above-breakpoint slopes remain broadly comparable. At $N_{d,min} = 10 \text{ cm}^{-3}$, CC-RH-LWC shows a larger β_b than CC-RH. For CC-RH, β_b increases from 10 to 40 cm^{-3} and then decreases from 40 to 100 cm^{-3} with the magnitudes at 40 and 100 cm^{-3} remaining comparable. By contrast, β_b in CC-RH-LWC declines drastically as $N_{d,min}$ increases, despite its explicit dependence on cloud water content. Consequently, β_b is weaker in CC-RH-LWC than CC-RH at higher $N_{d,min}$. This apparent paradox reflects a “headroom effect”, more visible when viewing the pattern against the secondary y-axes for predicted CF (Fig. 3): At high $N_{d,min}$, CC-RH-LWC already attains a substantially elevated baseline CF (given by the SHAP base values indicated by the horizontal black solid lines). Therefore, CF predictions approach the physical ceiling of 1.0, and the marginal sensitivity to

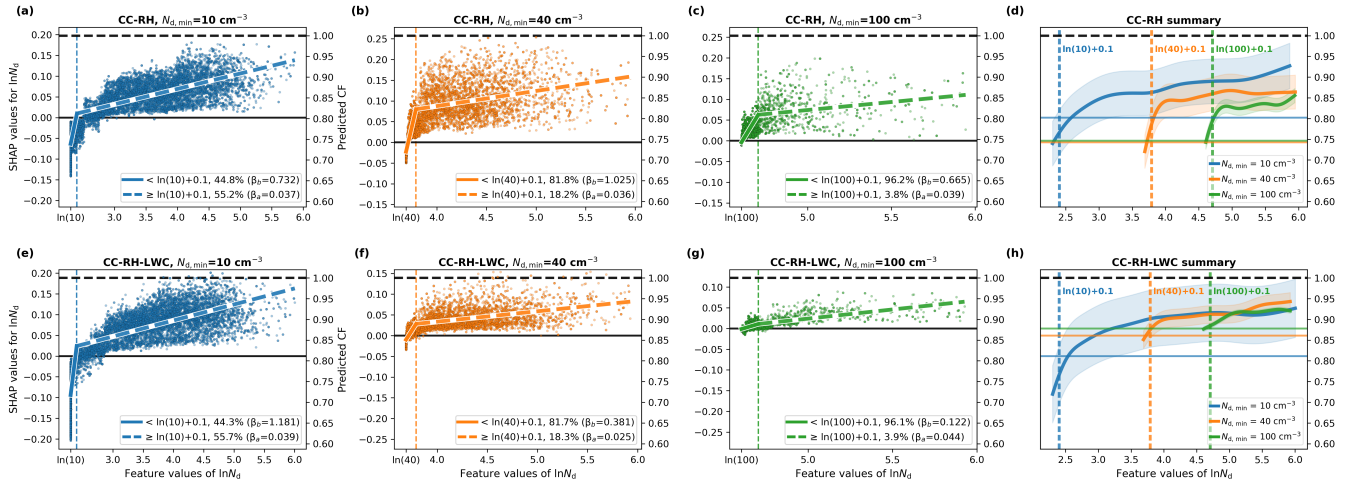


Figure 3. SHAP dependence plots for log-transformed cloud droplet number concentration ($\ln N_d$) at the exemplary geographical window (27.5° - 32.5° S, 122.5° - 127.5° W) for six ICON-HAM simulations. Top row (a–d): the CC-RH scheme; bottom row (e–h): the CC-RH-LWC scheme. Columns correspond to $N_{d,min}$ values of 10, 40 and 100 cm^{-3} (left to right). In panels (a–c) and (e–g), the x-axis shows feature values of $\ln N_d$, and the primary y-axis shows SHAP values for $\ln N_d$. The secondary y-axis (right) shows XGBoost-predicted CF (SHAP values + SHAP base value). Horizontal solid black lines indicate zero SHAP contribution on the primary axis and the baseline CF prediction on the secondary axis; the horizontal dashed line marks CF = 1.0. To quantify $\ln N_d$ –CF sensitivity, piecewise linear regression (PLR) is applied with a single breakpoint at $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$. Solid and dashed regression lines show the fitted slopes below (β_b) and above (β_a) the breakpoint, respectively. Vertical dashed lines indicate breakpoint locations. Percentage values in the legend report the fraction of samples in each regime. Panels (d) and (h) summarize the CC-RH and CC-RH-LWC results, respectively, using generalized additive model fits with 95 % prediction intervals. Colours match corresponding scatter plots.

further $\ln N_d$ increases diminish due to saturation. In contrast, CC-RH maintains lower baseline CF values, preserving a greater
 375 dynamic range for $\ln N_d$ to influence CF.

This SHAP-based piecewise approach effectively captures an intrinsic ICON-HAM behaviour in the $\ln N_d$ –CF relationships: a transition from a steeper, $N_{d,min}$ -constrained regime to a flatter, far-from- $N_{d,min}$ regime. Although the interval between the breakpoint and $N_{d,min}$ is fixed in log space across all experiments, the fraction of data points in the first regime increases with $N_{d,min}$, consistent with stronger truncation and tighter constraint near the higher imposed lower bounds. A clear headroom
 380 trend emerges: the baseline CF is higher in CC-RH-LWC and increases further with $N_{d,min}$, leaving less room for additional CF gains and thus dampening the sensitivity.

3.3.2 Global patterns of SHAP base values and mean absolute SHAP of $\ln N_d$

Section 3.1 showed that imposing higher $N_{d,min}$ values suppresses autoconversion and drizzle, leading to reduced PR and enhanced LWP in both cloud-cover schemes. In CC-RH-LWC, where CF depends explicitly on cloud water, higher LWP
 385 directly translates into higher CF, resulting in a mean state characterized by more extensive cloud decks. Conversely, CC-RH,



which depends solely on RH, shows little change or even a decline in CF, likely driven by reduced grid-mean RH associated with enhanced cloud-top entrainment and weaker sub-cloud evaporative moistening. As a result, at $N_{d,min} = 40$ and 100 cm^{-3} , the two schemes produce markedly different baseline CF states, consistent with the behaviour reported by Lohmann and Feichter (1997). In the following, we examine how these mean-state differences are reflected in the SHAP base values and mean absolute SHAP values of $\ln N_d$, which together describe the baseline cloud state and the overall contribution strength of aerosol variability within the SHAP framework.

Figure 4 zooms out from the exemplary window to place this behaviour in a global context. Figure 4a presents the global distribution of SHAP base values. Defined as the average of all XGBoost predictions over the training set, the SHAP base value corresponds directly to the local climatological mean-state CF. Indeed, their global distributions resemble the contrasting behaviours of CC-RH and CC-RH-LWC observed in Fig. 1c. In other words, variations in SHAP base values across experiments primarily indicate the physical shifts in the simulated CF mean state attributed to the influence of $N_{d,min}$ on autoconversion and drizzle.

Panel (b) shows the global distribution of the mean absolute SHAP value for $\ln N_d$, which is a measure of its feature importance or average marginal contribution. At 10 cm^{-3} , $\ln N_d$ shows greater feature importance in CC-RH-LWC than in CC-RH, reflecting CC-RH-LWC's direct CF-LWC coupling, which enables N_d -induced liquid water changes to immediately affect cloud cover. At $N_{d,min} = 40$ and 100 cm^{-3} , the opposite holds: CC-RH has a larger $\ln N_d$ contribution, since there is limited headroom for $\ln N_d$ variability to drive further CF increases in CC-RH-LWC, constraining the marginal $\ln N_d$ contributions. This global behaviour mirrors the headroom effect identified at the exemplary window in Sect. 3.3.1.

In summary, the influence of the differing treatment of liquid-water dependence is twofold both locally and globally: First, it alters the physical mean-state CF (the SHAP base value), and second, this baseline difference makes the system “headroom-mediated” by limiting the room for $\ln N_d$ to contribute to CF increases. This twofold effect is also the key to explaining the resulting sensitivity patterns.

3.3.3 Global distribution of $\ln N_d$ –CF sensitivity

Having established how elevated SHAP base values (baseline CF) constrain the feature importance of $\ln N_d$ within the SHAP framework, we now examine how this mechanism manifests itself in the global distribution of $\ln N_d$ –CF sensitivities derived from the PLR with the same breakpoint strategy (Sect. 2.3). The PLR slopes represent the directional sensitivity of CF to $\ln N_d$. Accordingly, lower mean absolute SHAP values, which indicate weaker marginal contributions, naturally correspond to flatter PLR slopes since the individual SHAP values entering the regression are smaller in magnitude. This leads to flatter SHAP dependence plots when $\ln N_d$ has limited capacity to contribute to CF predictions. Figure 5 summarizes the resulting global distributions of PLR-derived $\ln N_d$ –CF sensitivities for the below- and above-breakpoint regimes. Consistently, the global distribution of the mean absolute SHAP values for $\ln N_d$ (Figs. 4b) resembles that of the below-breakpoint sensitivities (Fig. 5a).

Across all experiments, the below-breakpoint sensitivities are systematically larger than the above-breakpoint ones, typically by about an order of magnitude (note the different y-axis ranges in Fig. 5a and b). The below-breakpoint regime reveals the

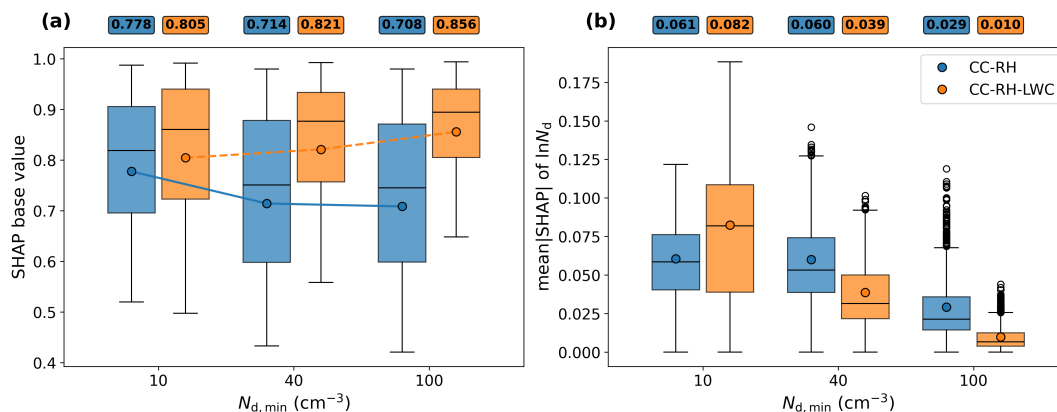


Figure 4. Global distributions of per-5° × 5°-window (a) SHAP base values for the prediction of CF and (b) mean absolute SHAP value of $\ln N_d$ (feature importance) for global marine liquid clouds during 2011–2015, including six experiments (two cloud-cover schemes: CC–RH and CC–RH–LWC; three imposed N_d lower bounds, $N_{d,min} = 10, 40, 100 \text{ cm}^{-3}$). Box plots show the interquartile range (IQR), whiskers extend to 1.5 × IQR, circles denote outliers, and horizontal lines indicate medians. Global area-weighted means are shown as circular markers connected by lines, with the corresponding values annotated above each panel.

420 clearest inter-scheme contrasts and directly reflects the headroom mechanism described above. At $N_{d,min} = 10 \text{ cm}^{-3}$, CC-RH-LWC shows stronger sensitivities than CC-RH (global area-weighted means of approximately 1.092 and 0.424, respectively). In line with Fig. 4, at this low $N_{d,min}$, both schemes have similar baseline CF (and hence similar headroom), allowing CC-RH-LWC’s explicit dependence on cloud liquid water to be fully expressed as both stronger feature importance and stronger derived CF sensitivity.

425 However, as $N_{d,min}$ increases to 40 and 100 cm^{-3} , the pattern reverses. The global weighted means for CC-RH (approximately 1.135 and 1.011, respectively) exceed those of CC-RH-LWC (0.633 and 0.209, respectively). This systematic weakening in CC-RH-LWC aligns precisely with the SHAP diagnostics in Fig.4: the liquid-water-dependent scheme’s high baseline CF (SHAP base values exceeding 0.85 at $N_{d,min} = 100 \text{ cm}^{-3}$) leaves minimal dynamic range for $\ln N_d$ to act, thereby muting the marginal contribution captured by SHAP and leading to weaker PLR-derived sensitivities. Conversely, CC-RH maintains
 430 lower baseline CFs, preserving a larger dynamic range for $\ln N_d$ to act, enabling stronger detected sensitivities.

Above the breakpoint, sensitivities are roughly an order of magnitude weaker than below it, and are centred near zero for both schemes (Fig. 5b). This attenuation mirrors the flatter upper segments of the SHAP dependence plots after saturation. In this regime, sensitivities remain relatively stable across $N_{d,min}$ values (0.036–0.054 per unit $\ln N_d$), and inter-scheme differences are minimal. This suggests that once $\ln N_d$ values are high and less influenced by the imposed lower bounds, both
 435 parameterizations respond similarly weak to aerosol variability.

Taken together with Sect. 3.3.2, the counterintuitive finding of weaker $\ln N_d$ –CF sensitivities in CC-RH-LWC at high $N_{d,min}$ does not indicate that its explicit CF–LWC coupling is ineffective. Instead, it demonstrates that the liquid-water dependence raises baseline CF and accelerates saturation, thereby diminishing the marginal contribution of $\ln N_d$ and the sensitivity de-



ected by SHAP. This reflects a fundamental distinction between physical responsiveness and methodological detectability. In this sense, the global sensitivity patterns are “headroom-mediated”, providing a coherent bridge from ICON-HAM’s physical representation of cloud cover to the explainable-ML diagnostics.

A direct comparison with the MODIS-based results from J24 highlights the structural differences between observation- and model-based sensitivities. The global area-weighted mean sensitivity from J24, derived from a single linear regression, is approximately 0.111 (Fig. 2c). This value lies between our two PLR regimes: substantially smaller than the below-breakpoint means (0.209 to 1.135) but larger than the above-breakpoint means (0.036 to 0.054). This intermediate value falls between the two regimes and can be effectively interpreted as an aggregated response. However, the distinct segmentation and spread in ICON-HAM sensitivities indicate that the simulated sensitivity estimates are tricky and also dictated by model configuration and methodological choices, complicating direct model-observation comparisons and aerosol-cloud forcing estimates.

3.3.4 Robustness of findings and physical plausibility of breakpoints

To ensure our main findings are not an artefact of our primary breakpoint definition $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$, we tested additional strategies: an alternative constraint-aware strategy with a fixed interval in linear space ($N_{d,min} + 5 \text{ cm}^{-3}$, Sect.B1), and a statistically optimized constraint-aware approach that determines breakpoints by minimizing the overall fitting error within a search range constrained by $N_{d,min}$ (Sect.B2). Despite differences in their underlying rationale and implementation, all three strategies yield qualitatively consistent results. First, the $\ln N_d$ -CF sensitivity in CC-RH-LWC is stronger at $N_{d,min} = 10 \text{ cm}^{-3}$ but becomes weaker than in CC-RH at 40 and 100 cm^{-3} . Second, inter-scheme differences are dominated by the below-breakpoint regime. This confirms the headroom mechanism independently of the specific breakpoint definition.

Beyond methodological consistency, the robustness of our findings is further supported by the physical plausibility of the breakpoint locations themselves. Independent observational studies of MLCs, using opportunistic experiments (ship tracks and volcanic eruptions) and large-scale satellite data (Hu et al., 2021; Yuan et al., 2023; Wang et al., 2024; Jia et al., 2024), have repeatedly identified a saturation threshold for CF responses to N_d at roughly $50\text{--}60 \text{ cm}^{-3}$. In addition, stratocumulus clouds are also known to transition from closed to open cells at a similar N_d threshold (Rosenfeld et al., 2023). In ICON-HAM, the breakpoint locations depend on the imposed $N_{d,min}$ by design. For the default $N_{d,min}$ of 40 cm^{-3} , however, they consistently fall close to the observational threshold range across three independently derived breakpoint strategies. The two constraint-aware strategies (Sect. 2.3 and B1) yield breakpoints of approximately 44 cm^{-3} and 45 cm^{-3} , respectively. Notably, the statistically optimized strategy (Sect. B2) independently finds a global area-weighted mean breakpoint value of about 48 cm^{-3} for CC-RH and 56 cm^{-3} for CC-RH-LWC (Fig. B4), closely matching the observed saturation threshold of $\sim 50\text{--}60 \text{ cm}^{-3}$. By contrast, the $N_{d,min} = 10 \text{ cm}^{-3}$ and 100 cm^{-3} experiments produce different breakpoint locations. The agreement for $N_{d,min} = 40 \text{ cm}^{-3}$ therefore suggests that this lower bound choice places the model’s emergent onset of the CF saturation at a more physically plausible N_d level than the more extreme $N_{d,min}$ settings.

Overall, the consistency of the breakpoint values among the three breakpoint-selection strategies, combined with agreement between the resulting breakpoints and the known range of the physical saturation thresholds in the $N_{d,min} = 40 \text{ cm}^{-3}$ simulations, confirms the robustness of our SHAP-based PLR approach and validates the underlying nonlinearity. This shows that the

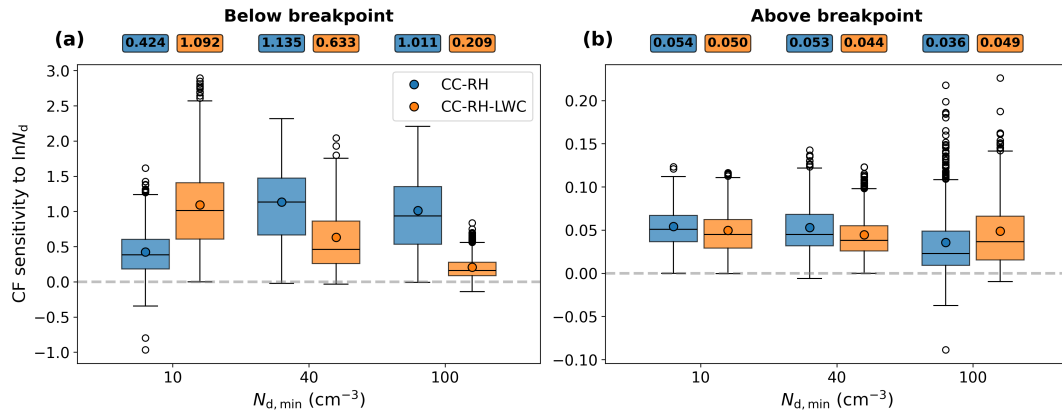


Figure 5. Global distributions of the cloud fraction sensitivity to log-transformed cloud droplet number concentration ($\ln N_d$) for global marine liquid clouds during 2011–2015, for six experiments (two cloud cover parameterization schemes: CC-RH and CC-RH-LWC; three imposed N_d lower bounds $N_{d,min} = 10, 40$ and 100 cm^{-3}). Sensitivities are computed within each $5^\circ \times 5^\circ$ oceanic window using a piecewise linear regression between $\ln N_d$ feature values and $\ln N_d$ SHAP values, with a single breakpoint $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$. (a) below-breakpoint sensitivities; (b) above-breakpoint sensitivities; Box plots display the interquartile range (IQR), whiskers extend to $1.5 \times \text{IQR}$, and circles denote outliers; medians are horizontal lines. Area-weighted means are shown as circular markers with connecting lines, and the floating values above each panel denote the corresponding area-weighted mean sensitivities. Note that the y-axis ranges differ between panels.

PLR approach is not merely an ad hoc curve-fitting choice but effectively captures the model’s physical transition from a highly sensitive, aerosol-limited regime to a saturated regime. Moreover, the consistent breakpoint range reinforces that our central findings—the headroom-mediated sensitivity patterns and the counterintuitive CC-RH > CC-RH-LWC sensitivity contrast at high $N_{d,min}$ —are robust and reflect genuine ICON-HAM’s simulated physics.

3.4 Implications for the forcing estimate

3.4.1 For the bottom-up approach (GCM PI–PD paired simulations)

Our findings reveal a potential challenge in quantifying the radiative forcing from the CF adjustment, as the detected sensitivity is highly state-dependent. Our findings have different implications for different methods to estimate forcing: the bottom-up approach (GCM PI–PD paired simulations) and the top-down approach (observational sensitivity-based).

GCMs typically estimate effective radiative forcing due to ACI using a bottom-up method by running paired PI and PD simulations and taking their difference in the net radiation at the TOA (Ghan, 2013; Boucher et al., 2013; Smith et al., 2020), without relying on an explicit sensitivity value. Instead, the aerosol-cloud forcing emerges from the full nonlinear response across the PI–PD aerosol contrast. Our analysis, in contrast, focuses on the local PD-state $\ln N_d$ –CF sensitivity across six ICON-HAM experiments. These PD-state slopes are therefore not a direct measure of CF-adjustment forcing, but they describe how



efficiently additional N_d changes can still modify CF once the system has already reached its PD baseline. Here, we discuss how our findings from these different ICON-HAM configurations potentially affect aerosol forcing estimates.

As systematically investigated by Hoose et al. (2009), imposing $N_{d,min}$ unphysically alters both the PI and PD states, creating artificially polluted PI conditions over both land and ocean and artificially polluted PD conditions over oceans. This makes the contrast between PI and PD collapse and strongly damps the total aerosol indirect effects. Our experiments add a previously undocumented, complementary dimension to this result by introducing the CC-RH-LWC scheme. When this scheme is combined with high $N_{d,min}$, suppressed autoconversion raises LWP and thereby elevates baseline CF in both PI and PD equilibrium states. Under naturally pristine PI conditions, this combination can already pre-saturates clouds. In the PD state, anthropogenic aerosols increases cannot further increase CF because clouds already operate near the $CF \approx 1$ ceiling. In addition to the $N_{d,min}$ -alone effect (reduced PI–PD N_d contrast) on the total forcing owing to ACI identified by Hoose et al. (2009), this bilateral saturation through the headroom effect compresses the PI–PD ΔCF and physically suppresses the CF-adjustment component of the forcing. For the CC-RH scheme, which lacks the direct LWC–CF link, baseline CF does not saturate to the same extent (Fig. 1c), so the forcing is mainly altered by $N_{d,min}$ alone as in Hoose et al. (2009).

On the other hand, our study also adds a new angle to Lohmann and Feichter (1997). Their work compared an LWC-dependent and an RH-only cloud-cover scheme (referred to as COUPL-CC and COUPL, respectively) and their resulting PI–PD changes in CF and the total aerosol indirect effect, but without imposing $N_{d,min}$. $N_{d,min}$ was only introduced in ECHAM4 later by Lohmann et al. (1999). Without $N_{d,min}$, there is no associated increase in baseline CF and thus PI clouds are not pre-saturated. Consequently, there is sufficient headroom and anthropogenic aerosols can increase CF without being immediately constrained by the CF ceiling in either COUPL or COUPL-CC. Therefore, the stronger dependence on cloud water in COUPL-CC produces larger CF increases and a more negative total indirect effect than the RH-only COUPL scheme. This behaviour is qualitatively consistent with our ICON-HAM experiments at a low $N_{d,min}$ of 10 cm^{-3} , where CC-RH-LWC shows a larger marginal contribution of $\ln N_d$ (Fig. 4b) and a stronger $\ln N_d$ –CF PD-state sensitivity (Fig. 5a) than CC-RH. In both studies, a cloud-water-dependent scheme responds more strongly than an RH-only scheme when $N_{d,min}$ is effectively low. However, at higher $N_{d,min}$, the ordering reverses (i.e. CC-RH becomes stronger) due to the headroom effect. Extrapolating this behaviour to the Lohmann and Feichter (1997) framework, one would expect that imposing a high $N_{d,min}$ in their COUPL versus COUPL-CC comparison would reduce the PI–PD CF increase in both schemes and thereby shrink the difference in total aerosol indirect effect between them. A complete reversal in the forcing order, analogous to the reversal seen in our $\ln N_d$ –CF sensitivities at high $N_{d,min}$, seems less likely. Lohmann and Feichter (1997) quantified the ACI forcing from PI to PD including non-CF components, whereas the headroom effect identified here acts specifically on the CF-adjustment component and is inferred from PD-state sensitivity variability rather than from an explicit PI–PD forcing calculation.

Overall, if CC-RH-LWC is used, this strong reduction in the CF-adjustment forcing is already operative at $N_{d,min} = 40 \text{ cm}^{-3}$, which is the default value in the ECHAM and ICON model families. Although we do not perform explicit PI–PD runs or compute the radiative forcing from CF adjustments, it is reasonable to expect that the combination of a CC-RH-LWC-type parameterization with high $N_{d,min}$ would weaken the CF-adjustment contribution to effective radiative forcing from ACI in a bottom-up setup, especially over clean oceanic regions. This tendency would further widen the gap between model-based



estimates that attribute only a minor role to CF adjustment (e.g. Smith et al., 2020; Zelinka et al., 2023) and observation-based studies that identify CF adjustment as a primary component (e.g. Chen et al., 2022; Wall et al., 2023; Yuan et al., 2023). These implications remain qualitative, as they are inferred from PD-state sensitivities rather than aerosol forcing diagnostics; a natural next step is to repeat our ICON-HAM experiments for paired PI–PD simulations and explicitly diagnose the CF-adjustment contribution to the aerosol-cloud forcing to test this hypothesis.

3.4.2 For the top-down approach (observational sensitivity-based)

In contrast to the bottom-up PI–PD approach, observational studies often follow a top-down strategy, in which CF-adjustment forcing is inferred from an explicit sensitivity value. These strategies vary: some exploit localized perturbations to isolate aerosol effects by deriving N_d –CF sensitivities from contrasts between relatively clean background conditions and nearby polluted perturbations (e.g. ship tracks or volcanic events) and directly calculate the radiative effect of the associated CF adjustments (e.g. Chen et al., 2022; Yuan et al., 2023), while others diagnose aerosol–CF sensitivities from statistical relationships across PD variability and apply cloud radiative kernels to the derived cloud-property changes (e.g. Wall et al., 2023). In all of these frameworks, the inferred sensitivity in the sampled cloud regimes is central, as it links observed variability to radiative forcing.

Our results highlight that such sensitivities are strongly state-dependent. In ICON-HAM, the headroom effect appears most clearly when high $N_{d,min}$ is combined with CC-RH-LWC, which pushes baseline CF close to its saturation limit and suppresses the PD-state $\ln N_d$ –CF sensitivity. Part of this pre-saturation is clearly structural to climate models, arising from the imposed $N_{d,min}$ and the chosen cloud-cover formulation. In the real atmosphere, there is neither $N_{d,min}$ nor a prescribed scheme, but there is still physical saturation of CF in regimes with high LWP or RH. Any observationally derived N_d –CF slope will therefore reflect the headroom available in the sampled clouds: sensitivities diagnosed in already cloudy, high-LWP regimes will tend to be weaker than those in thinner, less saturated regimes. This aligns with previous findings that N_d –CF sensitivities are strongest near the edges of the stratocumulus field and in stratocumulus-to-cumulus transition regions, rather than within the overcast decks (Gryspeerdt et al., 2016; Jia et al., 2024).

This state-dependence has implications for interpreting observational constraints on CF-adjustment forcing. Most recent observation-based estimates (e.g. Chen et al., 2022; Wall et al., 2023; Yuan et al., 2023; Chen et al., 2024) use natural experiments or from PD aerosol–cloud relationships, which do not require assuming a single fixed N_d –CF sensitivity that can be applied to infer changes from PI to PD. However, the underlying N_d –CF relationships in these studies are still diagnosed from specific cloud regimes in the current climate, where baseline CF varies and thus the remaining potential for CF to increase differs. Consequently, the inferred sensitivities are conditional on the mean state and may not be directly transferable to a PI–PD forcing context, since sensitivities in already-cloudy, low-headroom regimes could underestimate the potential response in cleaner, less saturated conditions (representative of PI). A more complete understanding of the CF adjustment to the aerosol forcing would therefore benefit from explicitly conditioning sensitivities on baseline cloud state (e.g. CF, LWP, and precipitation regime) as a complement to regime-aggregated estimates. This is particularly relevant given the likely reduction in MLC cover in a warming world (Myers et al., 2021; Forster et al., 2021; Bock and Lauer, 2024), which would increase the



available headroom and thus potentially enhance the CF sensitivity. This would facilitate more consistent comparisons between model-based and observational estimates by accounting for how the available headroom for CF increases varies systematically across different cloud states.

4 Conclusions

560 This study extends the explainable ML framework from our previous observational study (J24) to the ICON-HAM atmosphere-aerosol model. Our primary goals were to compare MODIS- and ICON-HAM-based $\ln N_d$ -CF sensitivities in MLCs, and to understand how the cloud-cover schemes and $N_{d,min}$ jointly shape the simulated CF response. To achieve this, we conducted six experiments, which are combined with two cloud-cover schemes, one dependent solely on relative humidity (CC-RH) and one additionally dependent on cloud water content (CC-RH-LWC), and three different $N_{d,min} = 10, 40, \text{ and } 100 \text{ cm}^{-3}$. The findings of our study are summarized as follows:

1. Mean-state response to $N_{d,min}$ and cloud-cover schemes: Higher $N_{d,min}$ suppresses autoconversion and drizzle, and builds up liquid water in both schemes. The CC-RH-LWC scheme translates this LWP increase directly into elevated CF due to its explicit cloud-water dependence, whereas CC-RH, lacking this direct link, shows a weaker or even declining CF response. At $N_{d,min} = 40 \text{ and } 100 \text{ cm}^{-3}$, this creates markedly different mean background cloud conditions between the schemes.
2. ICON-HAM simulates a non-linearity in the $\ln N_d$ -CF relationship: The ICON-HAM $\ln N_d$ -CF relationship is strongly non-linear and shows a clear saturation effect that persists even after log-transformation. This contrasts with the observation-based relationship, which has been shown to be linear in logarithmic space. The difference likely reflects ICON-HAM's process-level parameterizations that retain small-scale saturation effects from field campaigns or LES studies, whereas satellite observations at larger scales show smoother responses due to spatial averaging.
3. Headroom-mediated $\ln N_d$ -CF sensitivity: The mean-state physics in point 1 sets the baseline CF in ICON-HAM. The SHAP approach reveals that high SHAP base values (baseline CF) limit the marginal contribution of $\ln N_d$ (Fig. 4), leading to weaker detected sensitivity (Fig. 5), despite the direct physical coupling between CF and cloud water, creating a headroom effect. At $N_{d,min} = 10 \text{ cm}^{-3}$, CC-RH-LWC is more sensitive to $\ln N_d$ due to its direct dependence on LWC. This pattern reverses at $N_{d,min} = 40 \text{ and } 100 \text{ cm}^{-3}$, as high $N_{d,min}$ together with CC-RH-LWC leaves little headroom, while CC-RH preserves a lower baseline CF and thus more room for $\ln N_d$ -driven changes.
4. Methodological extension and breakpoint robustness: Three breakpoint strategies consistently separate a steep low- N_d regime and a flatter high- N_d regime, and the contrasting patterns are qualitatively consistent across all experiments. For ICON-HAM's default $N_{d,min} = 40 \text{ cm}^{-3}$, the statistically optimized breakpoints converge at approximately 50–60 cm^{-3} , consistent with observed N_d -CF saturation thresholds, supporting the robustness of our framework. More importantly, this suggests that the default $N_{d,min}$ setup provides a physically consistent representation of the transition into the saturation regime.



5. Implications for CF adjustment forcing and model–observation comparisons: For GCMs using bottom-up PI–PD methods, high $N_{d,min}$ values combined with LWC-dependent cloud-cover schemes can pre-saturate both PI and PD cloud fields, damping CF adjustment forcing even when the underlying model physics is strong. In top-down, observation-based approaches, any diagnosed N_d –CF sensitivity is inherently state-dependent and reflects the available headroom in the sampled cloud regimes. This implies that robust CF adjustment constraints should explore the option to condition sensitivities on baseline cloud state (e.g. CF, LWP, and precipitation regime), rather than treating the N_d –CF sensitivity as a single, regime-independent quantity.

595 *Code availability.* Code is available from the corresponding author upon reasonable request.

Data availability. All model output analyzed in this study was generated using ICON-HAM version 2.6.4. Due to the large data volume, the model output is not publicly archived but is available from the corresponding author upon reasonable request. The MODIS satellite observations and reanalysis data used for comparison in Fig. 2a and Fig. S2a were obtained from the sources described in Jia et al. (2024).

Appendix A: Predictor variables

600 Appendix B: Piecewise SHAP sensitivity with two alternative breakpoint strategies

B1 Constraint-aware breakpoint with fixed interval in linear space

To assess how the choice of breakpoints influences our results, we introduce two additional breakpoint strategies. The first one is likewise constraint-aware, as the breakpoint is defined relative to $N_{d,min}$. However, the interval is specified in linear rather than logarithmic space:

$$605 \quad \ln(N_{d,bp}) = \ln(N_{d,min} + \Delta N_d) \quad (B1)$$

where $N_{d,bp}$ denotes the breakpoint and ΔN_d represents the offset in linear space (in cm^{-3}) from the imposed lower bound. For this test, we set $\Delta N_d = 5 \text{ cm}^{-3}$, yielding breakpoints of 15, 45 and 105 cm^{-3} for $N_{d,min} = 10, 40$, and 100 cm^{-3} , respectively.

On the logarithmic scale, the interval between $N_{d,min}$ and the breakpoint equals $\ln(1 + \frac{\Delta N_d}{N_{d,min}})$, which shrinks as $N_{d,min}$ increases. The log-space interval width is $\ln(1.5)$, $\ln(1.125)$, $\ln(1.05)$ for $N_{d,min} = 10, 40$ and 100 cm^{-3} , respectively. Thus, as $N_{d,min}$ increases, the prescribed truncation becomes tighter and causes more data points to pile up near $N_{d,min}$. This already increases the y-axis (SHAP value) spread within a narrow x-axis range. The shrinking intervals further compress the near- $N_{d,min}$ regime on the x-axis while the associated SHAP response variability on the y-axis remains comparatively strong, leading to an inflated β_b at higher $N_{d,min}$ values.



Table A1. Overview of the predictors from ICON-HAM. Numeric subscripts denote pressure levels or altitudes.

Predictor variable	Short name	Unit
Relative humidity	$RH_{700}, RH_{850}, RH_{900}, RH_{950}$	%
Specific humidity	SH_{700}, SH_{850}	kg kg^{-1}
Air temperature	t_{700}, t_{850}	K
Vertical velocity	$\omega_{700}, \omega_{850}$	Pa s^{-1}
Zonal (eastward) wind	u_{700}, u_{850}	m s^{-1}
Meridional (northward) wind	v_{700}, v_{850}	m s^{-1}
Eastward and northward wind component at 10 m	u_{10}, v_{10}	m s^{-1}
Boundary-layer height (proxy)	BLH	m
Sea surface temperature	SST	K
Vertically integrated water vapour	VIWV	kg m^{-2}
Large-scale precipitation flux (water)	PR	$\text{kg m}^{-2} \text{s}^{-1}$
Sensible heat flux	SHF	W m^{-2}
Latent heat flux	LHF	W m^{-2}
Estimated inversion strength (calculated)	EIS	K
Natural logarithm of cloud droplet number concentration	$\ln N_d$	dimensionless (N_d in cm^{-3})

615 This inflated β_b is visible both in Figs. B1 and B2 for the CC-RH parameterization as $N_{d,\min}$ increases. It does not occur for
 CC-RH-LWC because the compression effect is outweighed by the headroom limitation. Despite these quantitative differences
 in the slopes, the main qualitative conclusions remain robust: First, below-breakpoint $\ln N_d$ -CF sensitivities are larger than
 those above the breakpoint regime, and inter-experiment contrasts are only evident in the below-breakpoint regime. Second, at
 $N_{d,\min} = 10 \text{ cm}^{-3}$, CC-RH-LWC has stronger sensitivities owing to its direct dependence on cloud water content; at $N_{d,\min}$
 620 $= 40$ and 100 cm^{-3} , CC-RH shows stronger sensitivities because of the headroom effect arising from CC-RH-LWC's elevated
 baseline CF and a CF = 1.0 cap. Choosing this breakpoint strategy primarily affects the trend of sensitivity with increasing
 $N_{d,\min}$ within each cloud-cover regime. The extent to which this x-range compression influences the results depends on its
 relative strength versus the headroom effect.

B2 Statistically optimized constraint-aware breakpoint

625 To further evaluate the robustness of our results, we apply a statistically optimized strategy. This data-driven approach deter-
 mines breakpoints by optimizing their locations that minimize the overall sum of squared errors of the two-segment PLR fit, but
 constrains the search range to $[\ln(N_{d,\min}), \ln(N_{d,\min} + 50)]$. This hybrid design is therefore constraint-aware while allowing
 the data to determine the exact split location.

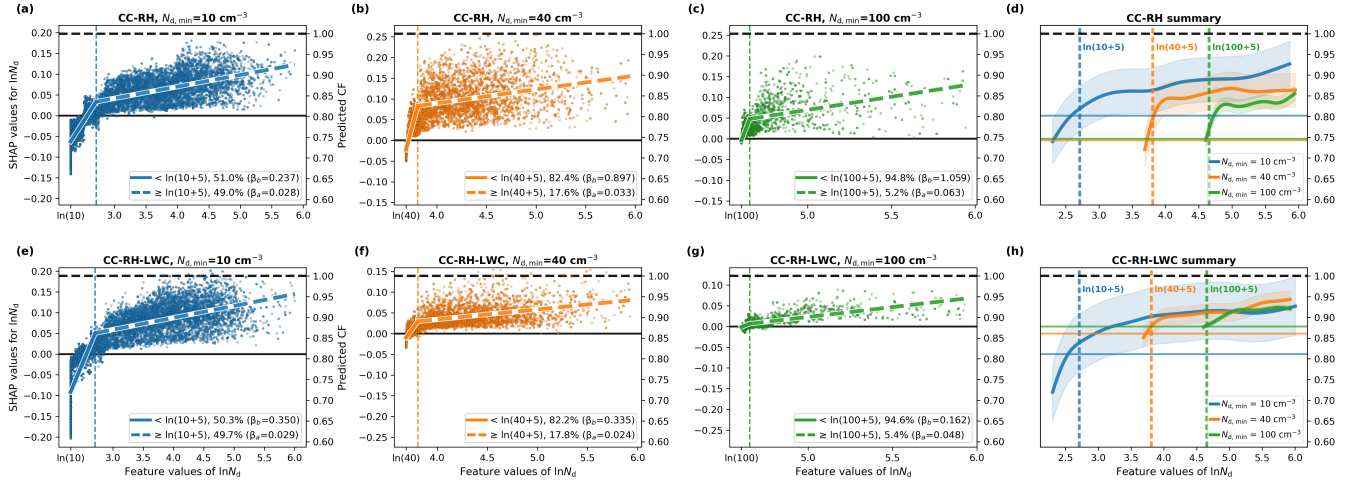


Figure B1. As in Fig. 3, but the breakpoint for the piecewise linear regression is defined as $\ln(N_{d,bp}) = \ln(N_{d,min} + 5)$.

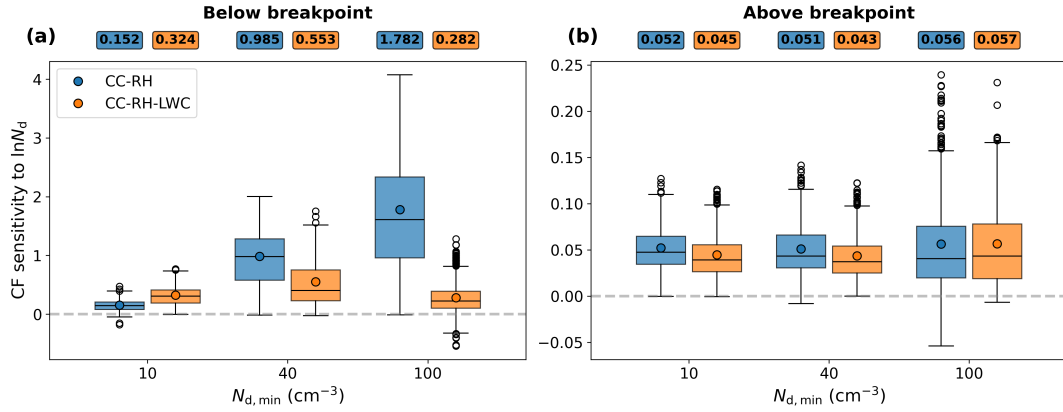


Figure B2. Similar to Fig. 5, except the breakpoint for the piecewise linear regression is defined as $\ln(N_{d,bp}) = \ln(N_{d,min} + 5)$.

To ensure statistical robustness, this hybrid strategy enforces per-regime sample minima of 1000 data points below the
 630 breakpoint and 500 above. These thresholds serve two purposes: first, a pre-fit feasibility check verifies whether any breakpoint
 within the bounded range can satisfy both minima at the same time, triggering the fallback if no feasible split exists; second, a
 post-fit validation checks the actual final regime sizes, setting slopes to NaN if either regime is insufficient. Additionally, we set
 a fit quality screen accepting the piecewise fit only if $R^2 \geq 0.70$ and root mean square error (RMSE) improves by at least 5 %
 compared to a single linear regression. If either the quality criteria fail, or no feasible split exists within the bounded range, we
 635 fall back to $\ln(N_{d,bp}) = \ln(N_{d,min}) + 0.1$. Per-regime sample minima are enforced at the slope-estimation stage; if only one
 regime lacks sufficient samples, that slope is set to NaN (no fallback), whereas a complete failure (both slopes invalid) triggers

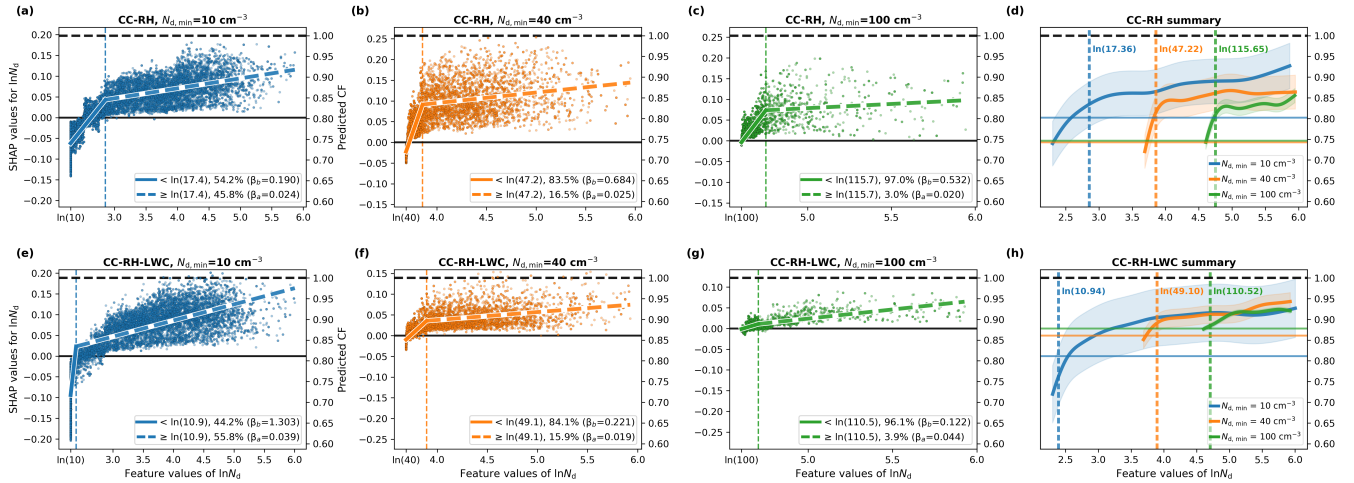


Figure B3. Similar to Fig. 3, except the breakpoints for the piecewise linear regression are statistically optimized.

the fallback. Finally, we apply an interquartile-range-based outlier filter (with a multiplier of 2.0) and a slope-consistency check to remove unphysical or outlying slope estimates.

This hybrid strategy yields Fig. B3 and Fig. B4. Again, both from the exemplary geographical window perspective and the global perspective, the key pattern persists. Particularly in the below-breakpoint regime, CC-RH-LWC shows stronger sensitivity at $N_{d,min} = 10 \text{ cm}^{-3}$ through its cloud water dependence, while CC-RH is stronger at 40 and 100 cm^{-3} due to headroom constraints in CC-RH-LWC. Overall, the inter- $N_{d,min}$ and inter-scheme contrasts obtained with this data-driven strategy (Fig. B4) are consistent with those from the constraint-aware approach with a fixed interval in log space (Fig. 5), confirming that our key scientific conclusions are robust to the choice of breakpoint strategy.

We additionally show the global distribution of the optimized breakpoints using this strategy in Fig. B4c. For the default ICON-HAM configuration ($N_{d,min} = 40 \text{ cm}^{-3}$), the area-weighted global mean optimized breakpoints are approximately 48 and 56 cm^{-3} on a linear scale for CC-RH and CC-RH-LWC, respectively. These values align closely with the observational saturation thresholds discussed in the main text (Sect. 3.3.4), providing physical support for the statistically optimized breakpoint locations (see main text for detailed discussion).

Author contributions. YJ, HA, JC, DN and UL designed the study. YJ, HA, DN, and UL developed the methodology. DN developed the ICON-HAM model configuration. YJ performed the model simulations, implemented the machine learning framework, analyzed the data, and created the visualizations. HA contributed extensively to the data analysis and interpretation throughout the study. UL, DN, and CH provided specific scientific insights regarding model configuration and the interpretation of findings. YJ wrote the original draft, and all authors reviewed and improved the manuscript.

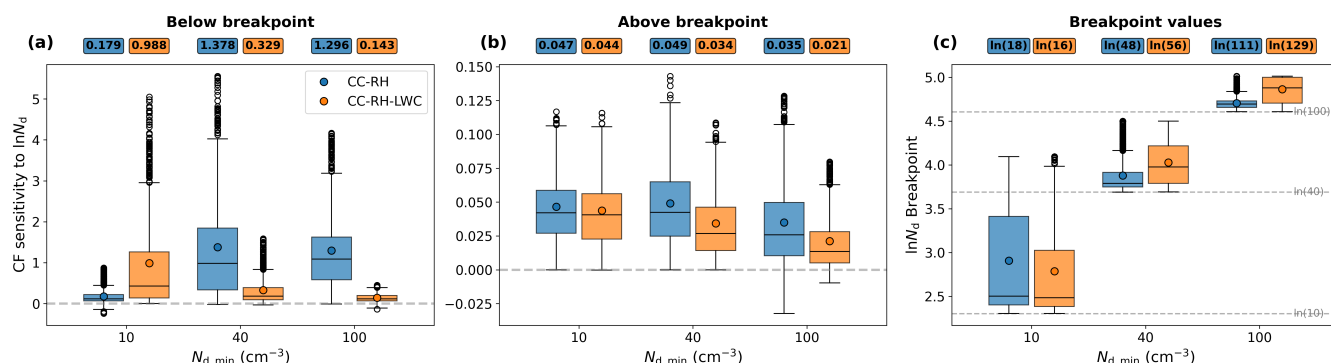


Figure B4. As in Fig. 5, but the breakpoints for the piecewise linear regression are statistically optimized (their distributions are shown in panel (c)).

655 *Competing interests.* The contact author has declared that none of the authors has any competing interests.

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References

- Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017, <https://doi.org/10.1038/nature03174>, 2004.
- 665 Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, <https://doi.org/10.1126/science.245.4923.1227>, 1989.
- Andersen, H., Cermak, J., Fuchs, J., Knutti, R., and Lohmann, U.: Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks, *Atmospheric Chemistry and Physics*, 17, 9535–9546, <https://doi.org/10.5194/acp-17-9535-2017>, 2017.
- 670 Andersen, H., Cermak, J., Douglas, A., Myers, T. A., Nowack, P., Stier, P., Wall, C. J., and Wilson Kemsley, S.: Sensitivities of cloud radiative effects to large-scale meteorology and aerosols from global observations, *Atmospheric Chemistry and Physics*, 23, 10775–10794, <https://doi.org/10.5194/acp-23-10775-2023>, 2023.
- Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A. L., Dufresne, J. L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B.: Bounding Global Aerosol Radiative Forcing of Climate Change, *Reviews of Geophysics*, 58, 1–45, <https://doi.org/10.1029/2019RG000660>, 2020.
- Bender, F. A., Frey, L., McCoy, D. T., Grosvenor, D. P., and Mohrmann, J. K.: Assessment of aerosol–cloud–radiation correlations in satellite observations, climate models and reanalysis, *Climate Dynamics*, 52, 4371–4392, <https://doi.org/10.1007/s00382-018-4384-z>, 2019.
- 680 Bock, L. and Lauer, A.: Cloud properties and their projected changes in CMIP models with low to high climate sensitivity, *Atmospheric Chemistry and Physics*, 24, 1587–1605, <https://doi.org/10.5194/acp-24-1587-2024>, 2024.
- Boucher, O., Granier, C., Hoose, C., and Andy, J.: Clouds and aerosols, *Climate Change 2013 the Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, 9781107057, 571–658, <https://doi.org/10.1017/CBO9781107415324.016>, 2013.
- 685 Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, *Geophysical Research Letters*, 34, <https://doi.org/https://doi.org/10.1029/2006GL027648>, 2007.
- Bretherton, C. S., Blossey, P. N., and Jones, C. R.: Mechanisms of marine low cloud sensitivity to idealized climate perturbations: A single-LES exploration extending the CGILS cases, *Journal of Advances in Modeling Earth Systems*, 5, 316–337, <https://doi.org/https://doi.org/10.1002/jame.20019>, 2013.
- 690 Chen, T. and Guestrin, C.: XGBoost: A Scalable Tree Boosting System, in: *Proceedings of the 22nd ACM SIGKDD International Conference on Knowledge Discovery and Data Mining, KDD '16*, pp. 785–794, ACM, <https://doi.org/10.1145/2939672.2939785>, 2016.
- Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Partridge, D., Fieldsend, J., De Leeuw, J., Schmidt, A., Cho, N., Oreopoulos, L., Platnick, S., Grosvenor, D., Field, P., and Lohmann, U.: Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover, *Nature Geoscience*, 15, 609–614, <https://doi.org/10.1038/s41561-022-00991-6>, 2022.
- 695 Chen, Y., Haywood, J., Wang, Y., Malavelle, F., Jordan, G., Peace, A., Partridge, D. G., Cho, N., Oreopoulos, L., Grosvenor, D., Field, P., Allan, R. P., and Lohmann, U.: Substantial cooling effect from aerosol-induced increase in tropical marine cloud cover, *Nature Geoscience*, <https://doi.org/10.1038/s41561-024-01427-z>, 2024.



- Chen, Y. C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds, *Nature Geoscience*, 7, 643–646, <https://doi.org/10.1038/ngeo2214>, 2014.
- 700 Christensen, M. W., Jones, W. K., and Stier, P.: Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition, *Proceedings of the National Academy of Sciences of the United States of America*, 117, 17591–17598, <https://doi.org/10.1073/pnas.1921231117>, 2020.
- Duran, B. M., Wall, C. J., Lutsko, N. J., Michibata, T., Ma, P.-L., Qin, Y., Duffy, M. L., Medeiros, B., and Debolskiy, M.: A new method for diagnosing effective radiative forcing from aerosol–cloud interactions in climate models, *Atmospheric Chemistry and Physics*, 25, 2123–2146, <https://doi.org/10.5194/acp-25-2123-2025>, 2025.
- 705 Fons, E., Naumann, A. K., Neubauer, D., Lang, T., and Lohmann, U.: Investigating the sign of stratocumulus adjustments to aerosols in the ICON global storm-resolving model, *Atmospheric Chemistry and Physics*, 24, 8653–8675, <https://doi.org/10.5194/acp-24-8653-2024>, 2024.
- Forster, P. M., Storelvmo, T., Armour, K., Collins, W., Dufresne, J. L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D., Watanabe, M., Wild, M., and Zhang, H.: Chapter 7: The Earth’s Energy Budget, Climate Feedbacks, and Climate Sensitivity, in: *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J., Maycock, T. K., Waterfield, T., Yelekçi, O., Yu, R., and Zhou, B., p. in press, Cambridge University Press, 2021.
- 710 Franke, H. and Giorgetta, M. A.: Toward the Direct Simulation of the Quasi-Biennial Oscillation in a Global Storm-Resolving Model, *Journal of Advances in Modeling Earth Systems*, 16, e2024MS004381, <https://doi.org/https://doi.org/10.1029/2024MS004381>, 2024.
- Fuchs, J., Cermak, J., and Andersen, H.: Building a cloud in the southeast Atlantic: Understanding low-cloud controls based on satellite observations with machine learning, *Atmospheric Chemistry and Physics*, 18, 16537–16552, <https://doi.org/10.5194/acp-18-16537-2018>, 2018.
- 720 Ghan, S. J.: Technical note: Estimating aerosol effects on cloud radiative forcing, *Atmospheric Chemistry and Physics*, 13, 9971–9974, <https://doi.org/10.5194/acp-13-9971-2013>, 2013.
- Giorgetta, M. A., Brokopf, R., Crueger, T., Esch, M., Fiedler, S., Helmert, J., Hohenegger, C., Kornblueh, L., Köhler, M., Manzini, E., Mauritsen, T., Nam, C., Raddatz, T., Rast, S., Reinert, D., Sakradzija, M., Schmidt, H., Schneck, R., Schnur, R., Silvers, L., Wan, H., Zängl, G., and Stevens, B.: ICON-A, the Atmosphere Component of the ICON Earth System Model: I. Model Description, *Journal of Advances in Modeling Earth Systems*, 10, 1613–1637, <https://doi.org/https://doi.org/10.1029/2017MS001242>, 2018.
- 725 Giorgetta, M. A., Sawyer, W., Lapillonne, X., Adamidis, P., Alexeev, D., Clément, V., Dietlicher, R., Engels, J. F., Esch, M., Franke, H., Frauen, C., Hannah, W. M., Hillman, B. R., Kornblueh, L., Marti, P., Norman, M. R., Pincus, R., Rast, S., Reinert, D., Schnur, R., Schulzweida, U., and Stevens, B.: The ICON-A model for direct QBO simulations on GPUs (version icon-cscs:baf28a514), *Geoscientific Model Development*, 15, 6985–7016, <https://doi.org/10.5194/gmd-15-6985-2022>, 2022.
- 730 Goren, T., Kazil, J., Hoffmann, F., Yamaguchi, T., and Feingold, G.: Anthropogenic Air Pollution Delays Marine Stratocumulus Breakup to Open Cells, *Geophysical Research Letters*, 46, 14135–14144, <https://doi.org/https://doi.org/10.1029/2019GL085412>, 2019.
- Goren, T., Feingold, G., Gryspeerdt, E., Kazil, J., Kretzschmar, J., Jia, H., and Quaas, J.: Projecting Stratocumulus Transitions on the Albedo—Cloud Fraction Relationship Reveals Linearity of Albedo to Droplet Concentrations, *Geophysical Research Letters*, 49, e2022GL101169, <https://doi.org/https://doi.org/10.1029/2022GL101169>, 2022.



- 735 Gryspeerdt, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, *Journal of Geophysical Research*, 121, 3566–3583, <https://doi.org/10.1002/2015JD023744>, 2016.
- Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M.: Constraining the aerosol influence on cloud liquid water path, *Atmospheric Chemistry and Physics*, 19, 5331–5347, <https://doi.org/10.5194/acp-19-5331-2019>, 2019.
- 740 Gryspeerdt, E., Mülmenstädt, J., Gettelman, A., Malavelle, F. F., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M., and Zhang, K.: Surprising similarities in model and observational aerosol radiative forcing estimates, *Atmospheric Chemistry and Physics*, 20, 613–623, <https://doi.org/10.5194/acp-20-613-2020>, 2020.
- Hartmann, D. L., Ockert-Bell, M. E., and Michelsen, M. L.: The Effect of Cloud Type on Earth’s Energy Balance: Global Analysis, *Journal of Climate*, 5, 1281–1304, [https://doi.org/10.1175/1520-0442\(1992\)005<1281:TEOCTO>2.0.CO;2](https://doi.org/10.1175/1520-0442(1992)005<1281:TEOCTO>2.0.CO;2), 1992.
- 745 Held, I. M. and Soden, B. J.: Robust Responses of the Hydrological Cycle to Global Warming, *Journal of Climate*, 19, 5686–5699, <https://doi.org/https://doi.org/10.1175/JCLI3990.1>, 2006.
- Hoose, C., Kristjánsson, J. E., Iversen, T., Kirkevåg, A., Seland, Ø., and Gettelman, A.: Constraining cloud droplet number concentration in GCMs suppresses the aerosol indirect effect, *Geophysical Research Letters*, 36, <https://doi.org/https://doi.org/10.1029/2009GL038568>, 2009.
- 750 Hu, S., Zhu, Y., Rosenfeld, D., Mao, F., Lu, X., Pan, Z., Zang, L., and Gong, W.: The Dependence of Ship-Polluted Marine Cloud Properties and Radiative Forcing on Background Drop Concentrations, *Journal of Geophysical Research: Atmospheres*, 126, e2020JD033 852, <https://doi.org/https://doi.org/10.1029/2020JD033852>, 2021.
- Jia, Y., Andersen, H., and Cermak, J.: Analysis of the cloud fraction adjustment to aerosols and its dependence on meteorological controls using explainable machine learning, *Atmospheric Chemistry and Physics*, 24, 13 025–13 045, <https://doi.org/10.5194/acp-24-13025-2024>, 755 2024.
- Khairoutdinov, M. and Kogan, Y.: A New Cloud Physics Parameterization in a Large-Eddy Simulation Model of Marine Stratocumulus, *Monthly Weather Review*, 128, 229–243, [https://doi.org/https://doi.org/10.1175/1520-0493\(2000\)128<0229:ANCPPI>2.0.CO;2](https://doi.org/https://doi.org/10.1175/1520-0493(2000)128<0229:ANCPPI>2.0.CO;2), 2000.
- Kirkevåg, A., Iversen, T., Seland, Ø., Debernard, J. B., Storelvmo, T., and Kristjánsson, J. E.: Aerosol-cloud-climate interactions in the climate model CAM-Oslo, *Tellus A: Dynamic Meteorology and Oceanography*, 60, 492, <https://doi.org/10.1111/j.1600-0870.2007.00313.x>, 760 2008.
- Leahy, L. V., Wood, R., Charlson, R. J., Hostetler, C. A., Rogers, R. R., Vaughan, M. A., and Winker, D. M.: On the nature and extent of optically thin marine low clouds, *Journal of Geophysical Research: Atmospheres*, 117, <https://doi.org/https://doi.org/10.1029/2012JD017929>, 2012.
- Lohmann, U. and Feichter, J.: Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity study with the ECHAM4 GCM, 765 *Journal of Geophysical Research: Atmospheres*, 102, 13 685–13 700, <https://doi.org/https://doi.org/10.1029/97JD00631>, 1997.
- Lohmann, U., Feichter, J., Chuang, C. C., and Penner, J. E.: Prediction of the number of cloud droplets in the ECHAM GCM, *Journal of Geophysical Research: Atmospheres*, 104, 9169–9198, <https://doi.org/https://doi.org/10.1029/1999JD900046>, 1999.
- Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, *Atmospheric Chemistry and Physics*, 7, 3425–3446, <https://doi.org/10.5194/acp-7-3425-2007>, 770 2007.
- Lundberg, S. M. and Lee, S. I.: A unified approach to interpreting model predictions, *Advances in Neural Information Processing Systems*, 2017-Decem, 4766–4775, 2017.



- Lundberg, S. M., Erion, G. G., and Lee, S.-I.: Consistent Individualized Feature Attribution for Tree Ensembles, ArXiv, abs/1802.0, <http://arxiv.org/abs/1802.03888>, 2018.
- 775 Lundberg, S. M., Erion, G., Chen, H., DeGrave, A., Prutkin, J. M., Nair, B., Katz, R., Himmelfarb, J., Bansal, N., and Lee, S.-I.: From local explanations to global understanding with explainable AI for trees, *Nature Machine Intelligence*, 2, 56–67, <https://doi.org/10.1038/s42256-019-0138-9>, 2020.
- Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S., Allan, R. P., Karset, I. H. H., Kristjánsson, J. E., Oreopoulos, L., Cho, N., Lee, D., Bellouin, N., Boucher, O., Grosvenor, D. P., Carslaw, K. S., Dhomse, S., Mann, G. W., Schmidt, A.,
 780 Coe, H., Hartley, M. E., Dalvi, M., Hill, A. A., Johnson, B. T., Johnson, C. E., Knight, J. R., O'Connor, F. M., Partridge, D. G., Stier, P., Myhre, G., Platnick, S., Stephens, G. L., Takahashi, H., and Thordarson, T.: Strong constraints on aerosol–cloud interactions from volcanic eruptions, *Nature*, 546, 485–491, <https://doi.org/10.1038/nature22974>, 2017.
- Mauritsen, T., Bader, J., Becker, T., Behrens, J., Bittner, M., Brokopf, R., Brovkin, V., Claussen, M., Crueger, T., Esch, M., Fast, I., Fiedler, S., Fläschner, D., Gayler, V., Giorgetta, M., Goll, D. S., Haak, H., Hagemann, S., Hedemann, C., Hohenegger, C., Ilyina, T., Jahns, T.,
 785 Jimenéz-de-la Cuesta, D., Jungclaus, J., Kleinen, T., Kloster, S., Kracher, D., Kinne, S., Kleberg, D., Lasslop, G., Kornblueh, L., Marotzke, J., Matei, D., Meraner, K., Mikolajewicz, U., Modali, K., Möbis, B., Müller, W. A., Nabel, J. E., Nam, C. C., Notz, D., Nyawira, S. S., Paulsen, H., Peters, K., Pincus, R., Pohlmann, H., Pongratz, J., Popp, M., Raddatz, T. J., Rast, S., Redler, R., Reick, C. H., Rohrschneider, T., Schemann, V., Schmidt, H., Schnur, R., Schulzweida, U., Six, K. D., Stein, L., Stemmler, I., Stevens, B., von Storch, J. S., Tian, F., Voigt, A., Vrese, P., Wieners, K. H., Wilkenskjeld, S., Winkler, A., and Roeckner, E.: Developments in the MPI-M Earth System
 790 Model version 1.2 (MPI-ESM1.2) and Its Response to Increasing CO₂, *Journal of Advances in Modeling Earth Systems*, 11, 998–1038, <https://doi.org/10.1029/2018MS001400>, 2019.
- Myers, T. A., Scott, R. C., Zelinka, M. D., Klein, S. A., Norris, J. R., and Caldwell, P. M.: Observational constraints on low cloud feedback reduce uncertainty of climate sensitivity, *Nature Climate Change*, 11, 501–507, <https://doi.org/10.1038/s41558-021-01039-0>, 2021.
- Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Stier, P., Partridge, D. G., Tegen, I., Bey, I., Stanelle, T., Kokkola, H., and Lohmann,
 795 U.: The global aerosol–climate model ECHAM6.3–HAM2.3 – Part 2: Cloud evaluation, aerosol radiative forcing, and climate sensitivity, *Geoscientific Model Development*, 12, 3609–3639, <https://doi.org/10.5194/gmd-12-3609-2019>, 2019.
- Null, J.: El Niño and La Niña Years and Intensities Based on {Oceanic Niño Index (ONI)}, Golden Gate Weather Services, <https://ggweather.com/enso/oni.htm>, 2025.
- Platnick, S. and Twomey, S.: Determining the Susceptibility of Cloud Albedo to Changes in Droplet Concentration with the Advanced Very
 800 High Resolution Radiometer, *Journal of Applied Meteorology and Climatology*, 33, 334–347, [https://doi.org/10.1175/1520-0450\(1994\)033<0334:DTSOCA>2.0.CO;2](https://doi.org/10.1175/1520-0450(1994)033<0334:DTSOCA>2.0.CO;2), 1994.
- Quaas, J., Andrews, T., Bellouin, N., Block, K., Boucher, O., Ceppi, P., Dagan, G., Doktorowski, S., Eichholz, H. M., Forster, P., Goren, T., Gryspeerdt, E., Hodnebrog, Ø., Jia, H., Kramer, R., Lange, C., Maycock, A. C., Mülmenstädt, J., Myhre, G., O'Connor, F. M., Pincus, R., Samset, B. H., Senf, F., Shine, K. P., Smith, C., Stjern, C. W., Takemura, T., Toll, V., and Wall, C. J.: Ad-
 805 justments to Climate Perturbations—Mechanisms, Implications, Observational Constraints, *AGU Advances*, 5, e2023AV001144, <https://doi.org/10.1029/2023AV001144>, 2024.
- Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., and Dümenil, L.: The atmospheric general circulation model ECHAM-4: Model description and simulation of present-day climate, Tech. Rep. 218, Max-Planck-Institut für Meteorologie, Hamburg, Germany, 1996.



- 810 Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., and Yu, S.: Aerosol-driven droplet concentrations dominate coverage and water of oceanic low-level clouds, *Science*, 363, <https://doi.org/10.1126/science.aav0566>, 2019.
- Rosenfeld, D., Kokhanovsky, A., Goren, T., Gryspeerd, E., Hasekamp, O., Jia, H., Lopatin, A., Quaas, J., Pan, Z., and Sourdeval, O.: Frontiers in Satellite-Based Estimates of Cloud-Mediated Aerosol Forcing, *Reviews of Geophysics*, 61, e2022RG000799, <https://doi.org/https://doi.org/10.1029/2022RG000799>, 2023.
- 815 Salzmann, M., Ferrachat, S., Tully, C., Münch, S., Watson-Parris, D., Neubauer, D., Siegenthaler-Le Drian, C., Rast, S., Heinold, B., Crueger, T., Brokopf, R., Mülmenstädt, J., Quaas, J., Wan, H., Zhang, K., Lohmann, U., Stier, P., and Tegen, I.: The Global Atmosphere-aerosol Model ICON-A-HAM2.3–Initial Model Evaluation and Effects of Radiation Balance Tuning on Aerosol Optical Thickness, *Journal of Advances in Modeling Earth Systems*, 14, e2021MS002699, <https://doi.org/https://doi.org/10.1029/2021MS002699>, 2022.
- Sherwood, S. C., Bony, S., Boucher, O., Bretherton, C., Forster, P. M., Gregory, J. M., and Stevens, B.: Adjustments in the forcing-feedback framework for understanding climate change, *Bulletin of the American Meteorological Society*, 96, 217–228, <https://doi.org/10.1175/BAMS-D-13-00167.1>, 2015.
- 820 Small, J. D., Chuang, P. Y., Feingold, G., and Jiang, H.: Can aerosol decrease cloud lifetime?, *Geophysical Research Letters*, 36, 1–5, <https://doi.org/10.1029/2009GL038888>, 2009.
- Smith, C. J., Kramer, R. J., Myhre, G., Alterskjær, K., Collins, W., Sima, A., Boucher, O., Dufresne, J.-L., Nabat, P., Michou, M., Yukimoto, S., Cole, J., Paynter, D., Shiogama, H., O'Connor, F. M., Robertson, E., Wiltshire, A., Andrews, T., Hannay, C., Miller, R., Nazarenko, L., Kirkevåg, A., Olivé, D., Fiedler, S., Lewinschal, A., Mackallah, C., Dix, M., Pincus, R., and Forster, P. M.: Effective radiative forcing and adjustments in CMIP6 models, *Atmospheric Chemistry and Physics*, 20, 9591–9618, <https://doi.org/10.5194/acp-20-9591-2020>, 2020.
- Sundqvist, H., Berge, E., and Kristjánsson, J. E.: Condensation and Cloud Parameterization Studies with a Mesoscale Numerical Weather Prediction Model, *Monthly Weather Review*, 117, 1641–1657, [https://doi.org/https://doi.org/10.1175/1520-0493\(1989\)117<1641:CACPSW>2.0.CO;2](https://doi.org/https://doi.org/10.1175/1520-0493(1989)117<1641:CACPSW>2.0.CO;2), 1989.
- 830 Taylor, K. E., Williamson, D., and Zwiers, F.: PCMDI Report - Number 60 - The sea surface temperature and sea-ice concentration boundary conditions for AMIP II simulations, <https://doi.org/10.5281/zenodo.13835219>, 2000.
- Toll, V., Christensen, M., Quaas, J., and Bellouin, N.: Weak average liquid-cloud-water response to anthropogenic aerosols, *Nature*, 572, 51–55, <https://doi.org/10.1038/s41586-019-1423-9>, 2019.
- 835 Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, *Journal of the Atmospheric Sciences*, 34, 1149–1152, [https://doi.org/10.1175/1520-0469\(1977\)034<1149:tiopot>2.0.co;2](https://doi.org/10.1175/1520-0469(1977)034<1149:tiopot>2.0.co;2), 1977.
- Wall, C. J., Norris, J. R., Possner, A., McCoy, D. T., McCoy, I. L., and Lutsko, N. J.: Assessing effective radiative forcing from aerosol–cloud interactions over the global ocean, *Proceedings of the National Academy of Sciences*, 119, e2210481119, <https://doi.org/10.1073/pnas.2210481119>, 2022.
- 840 Wall, C. J., Storelvmo, T., and Possner, A.: Global observations of aerosol indirect effects from marine liquid clouds, *Atmospheric Chemistry and Physics*, 23, 13 125–13 141, <https://doi.org/10.5194/acp-23-13125-2023>, 2023.
- Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, *Atmospheric Chemistry and Physics*, 9, 239–260, <https://doi.org/10.5194/acp-9-239-2009>, 2009.
- Wang, S., Wang, Q., and Feingold, G.: Turbulence, Condensation, and Liquid Water Transport in Numerically Simulated Non-precipitating Stratocumulus Clouds, *Journal of the Atmospheric Sciences*, 60, 262–278, [https://doi.org/https://doi.org/10.1175/1520-0469\(2003\)060<0262:TCALWT>2.0.CO;2](https://doi.org/https://doi.org/10.1175/1520-0469(2003)060<0262:TCALWT>2.0.CO;2), 2003.



- Wang, X., Mao, F., Zhu, Y., Rosenfeld, D., Pan, Z., Zang, L., Lu, X., Liu, F., and Gong, W.: Hidden Large Aerosol-Driven Cloud Cover Effect Over High-Latitude Ocean, *Journal of Geophysical Research: Atmospheres*, 129, e2023JD039312, <https://doi.org/https://doi.org/10.1029/2023JD039312>, 2024.
- 850 Wang, Y., Neubauer, D., Chen, Y., Jordan, G., Malavelle, F., Yuan, T., Partridge, D., Field, P., Wang, H., Wang, M., Michou, M., Nabat, P., Laakso, A., Myhre, G., and Lohmann, U.: Challenges in global climate models to represent cloud response to aerosols: insights from volcanic eruptions, *Nature Communications*, 17, 627, <https://doi.org/10.1038/s41467-025-67359-3>, 2025.
- Watson-Parris, D. and Smith, C. J.: Large uncertainty in future warming due to aerosol forcing, *Nature Climate Change*, 12, 1111–1113, <https://doi.org/10.1038/s41558-022-01516-0>, 2022.
- 855 Wood, R.: Stratocumulus Clouds, *Monthly Weather Review*, 140, 2373–2423, <https://doi.org/10.1175/MWR-D-11-00121.1>, 2012.
- Wood, R., Wyant, M., Bretherton, C. S., Rémillard, J., Kollias, P., Fletcher, J., Stemmler, J., de Szoek, S., Yuter, S., Miller, M., Mechem, D., Tselioudis, G., Chiu, J. C., Mann, J. A. L., O'Connor, E. J., Hogan, R. J., Dong, X., Miller, M., Ghate, V., Jefferson, A., Min, Q., Minnis, P., Palikonda, R., Albrecht, B., Luke, E., Hannay, C., and Lin, Y.: Clouds, Aerosols, and Precipitation in the Marine Boundary Layer: An Arm Mobile Facility Deployment, *Bulletin of the American Meteorological Society*, 96, 419–440, [https://doi.org/10.1175/BAMS-D-13-](https://doi.org/10.1175/BAMS-D-13-00180.1)
- 860 00180.1, 2015.
- Xu, K.-M. and Randall, D. A.: A Semiempirical Cloudiness Parameterization for Use in Climate Models, *Journal of Atmospheric Sciences*, 53, 3084–3102, [https://doi.org/https://doi.org/10.1175/1520-0469\(1996\)053<3084:ASCPFU>2.0.CO;2](https://doi.org/https://doi.org/10.1175/1520-0469(1996)053<3084:ASCPFU>2.0.CO;2), 1996.
- Yuan, T., Song, H., Wood, R., Oreopoulos, L., Platnick, S., Wang, C., Yu, H., Meyer, K., and Wilcox, E.: Observational evidence of strong forcing from aerosol effect on low cloud coverage, <https://doi.org/10.1126/sciadv.adh7716>, 2023.
- 865 Zelinka, M. D., Andrews, T., Forster, P. M., and Taylor, K. E.: Quantifying components of aerosol-cloud-radiation interactions in climate models, *Journal of Geophysical Research: Atmospheres*, 119, 7599–7615, <https://doi.org/10.1002/2014JD021710>, 2014.
- Zelinka, M. D., Smith, C. J., Qin, Y., and Taylor, K. E.: Comparison of methods to estimate aerosol effective radiative forcings in climate models, *Atmospheric Chemistry and Physics*, 23, 8879–8898, <https://doi.org/10.5194/acp-23-8879-2023>, 2023.