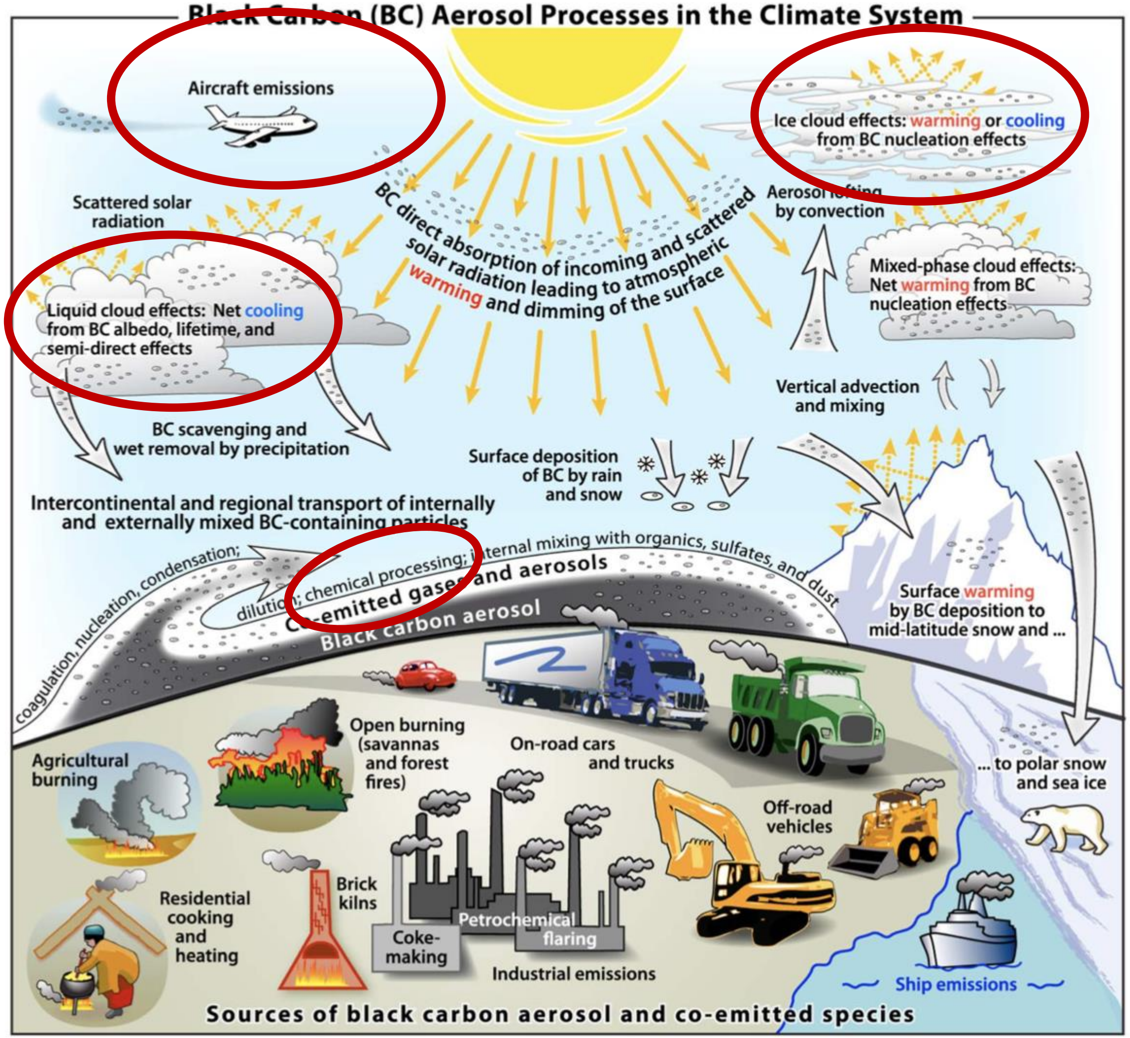
The role of black carbon in cloud formation and climate

<u>Ulrike Lohmann</u>

F. Friebel, Z.A. Kanji, F. Mahrt, A.A. Mensah, D. Neubauer

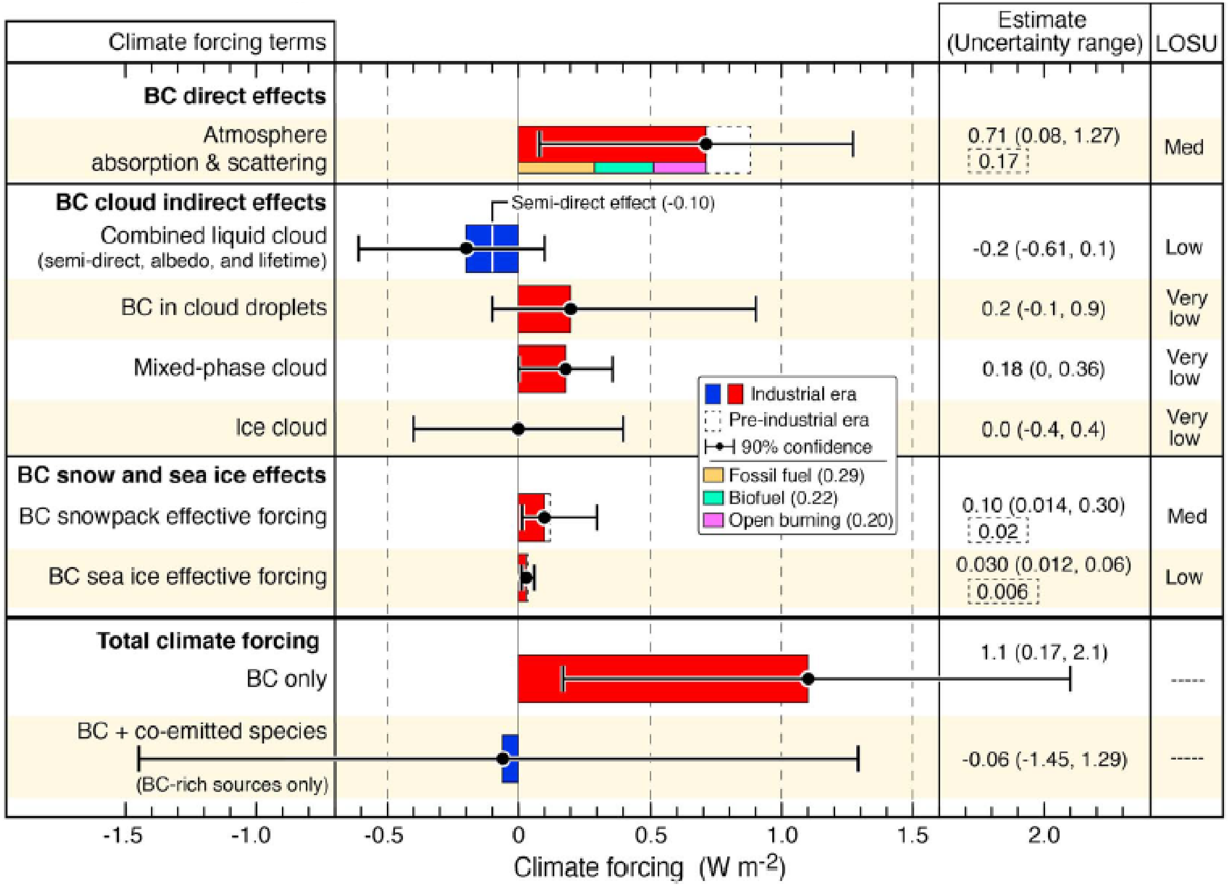
Source: wikipedia

ETH zürich



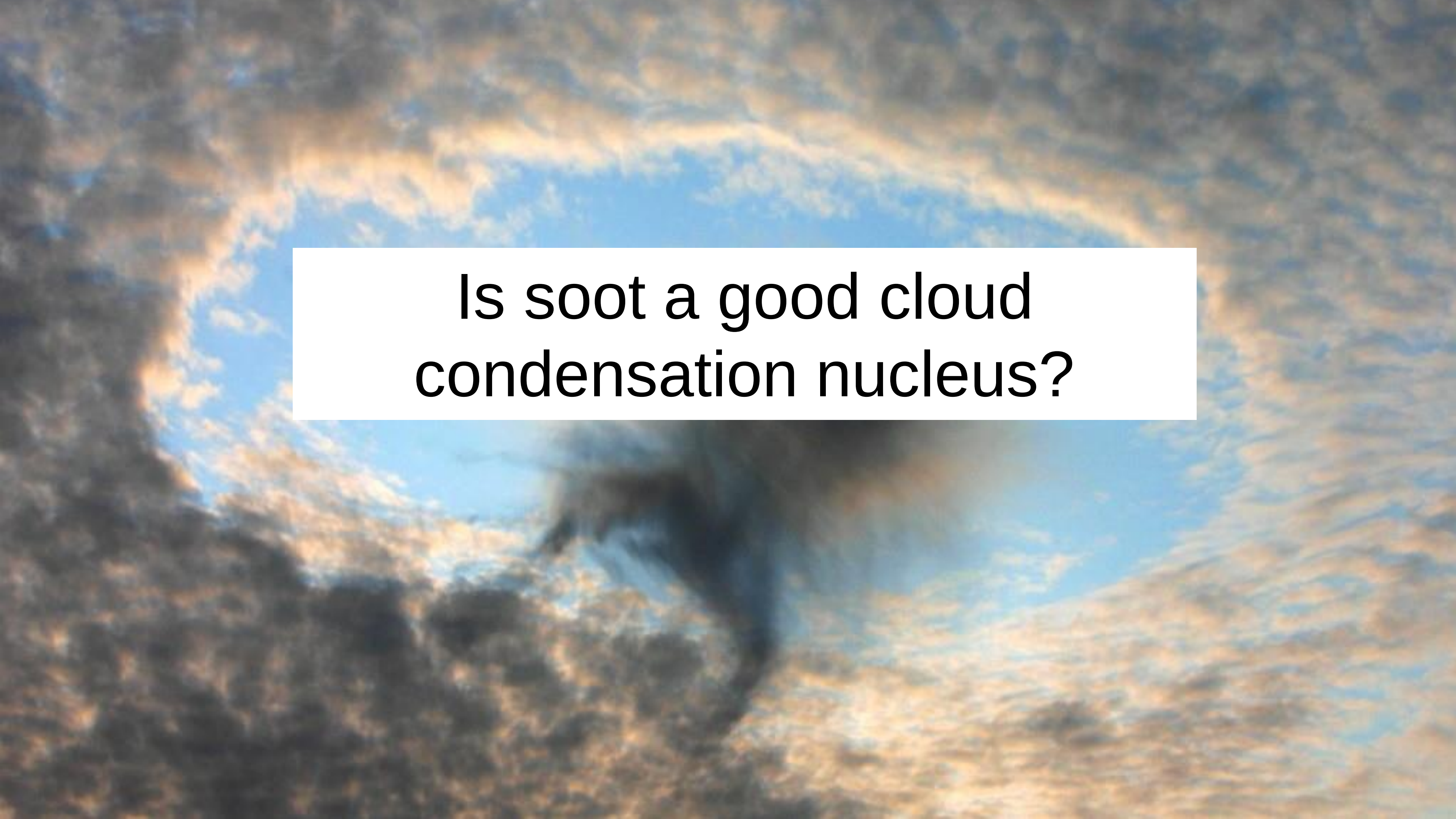
Bond et al., 2013

Climate forcing of soot and co-emitted species (1750-2005)



Bond et al., 2013

- Can soot particles act as cloud condensation nuclei (CCN) at atmospheric conditions?
- Can soot particles act as ice nucleating particles (INPs) at atmospheric conditions?
- Can cloud processing of soot particles improve their INP ability?


Is soot a good cloud condensation nucleus?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Air


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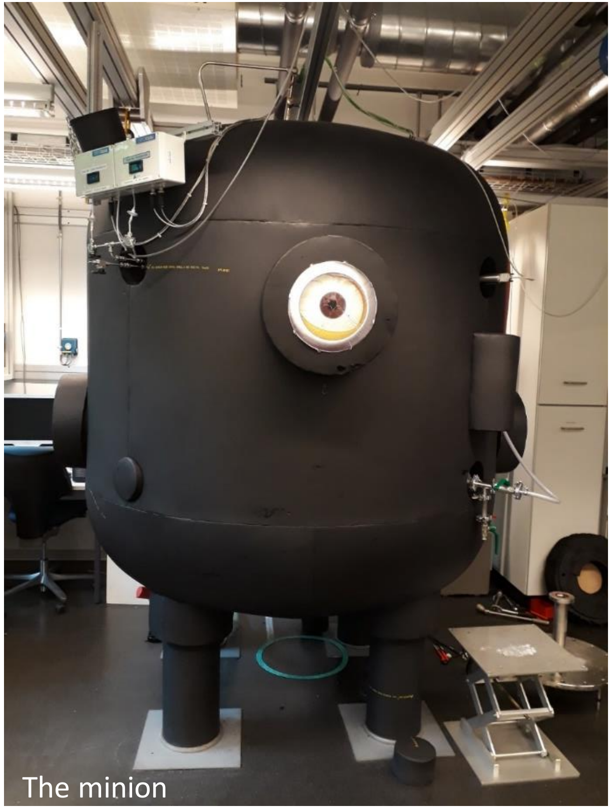
Ozone

Continuous-flow Stirred Tank Reactor (CSTR)

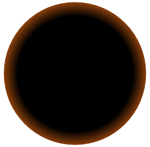
- 100 nm soot particles
- 16 h aging time
- miniCAST brown (organic carbon rich soot)

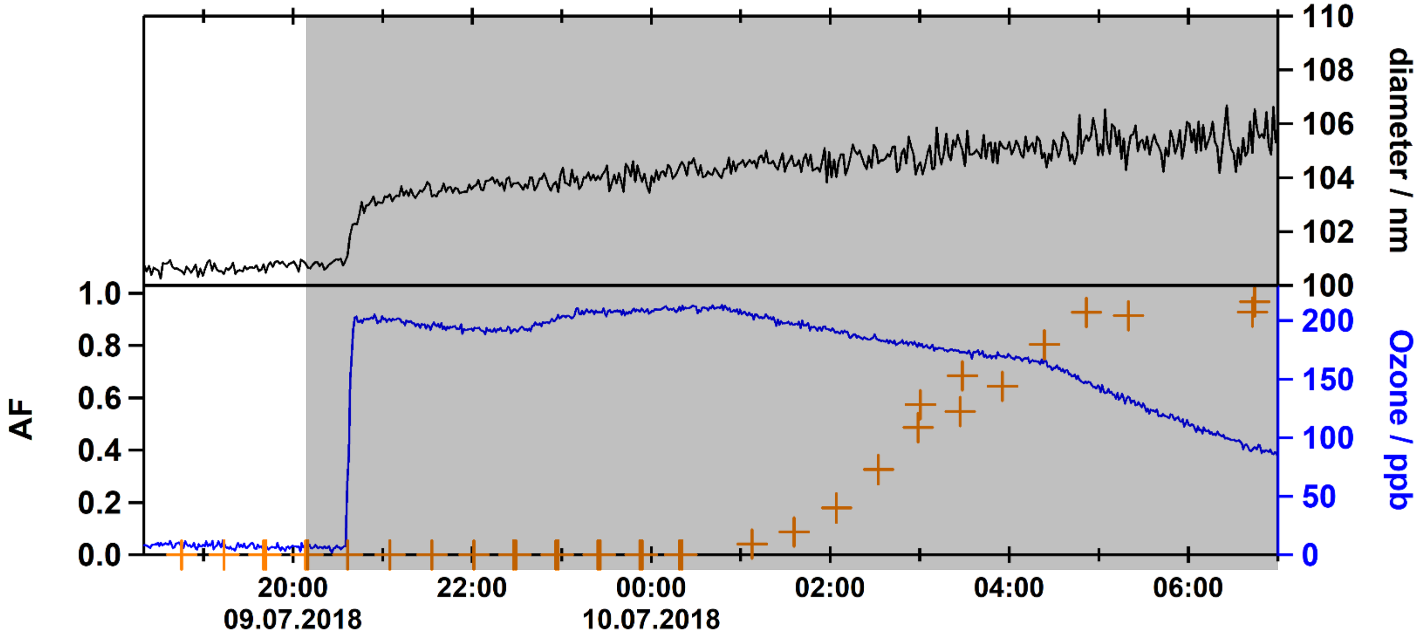
Temperature 5 - 35 °C ⟷ Ozone 0 - 200 ppb



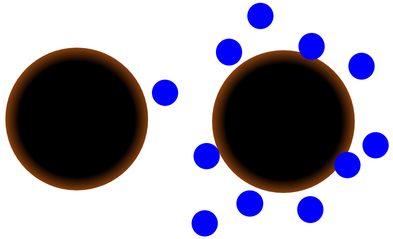
Friebel and Mensah, AMT, 2019

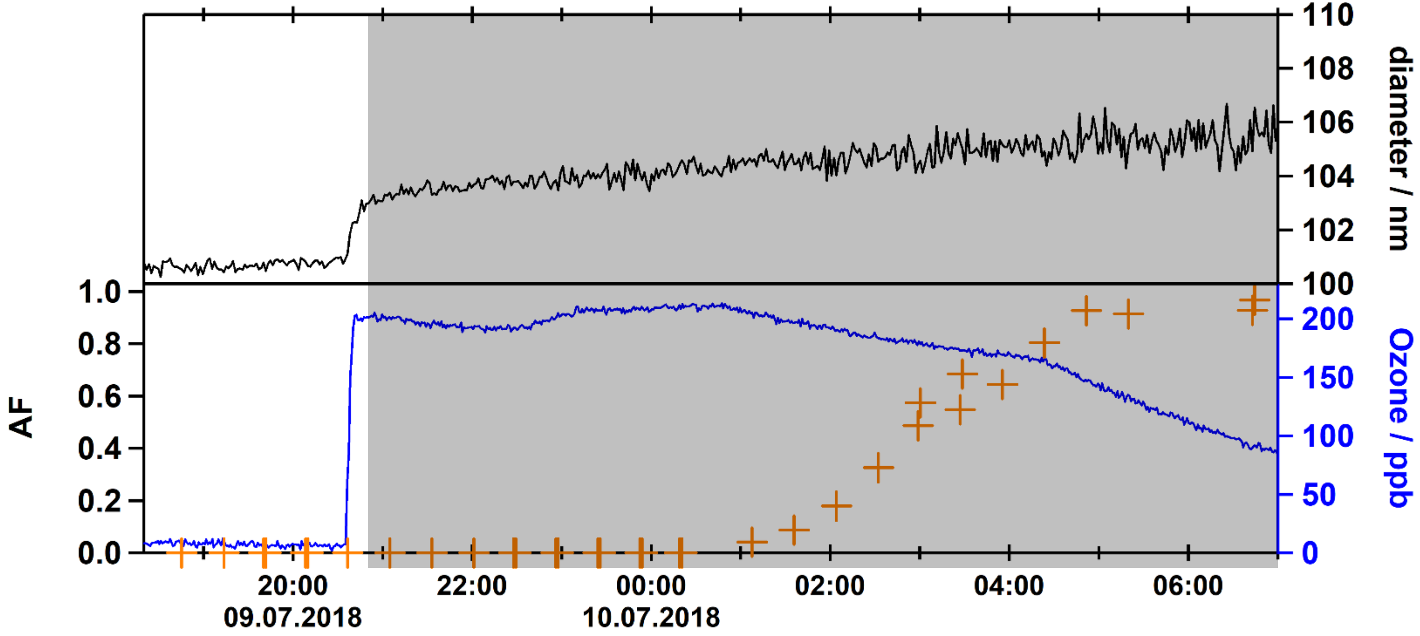
Ozone oxidation of 100 nm organic-rich soot



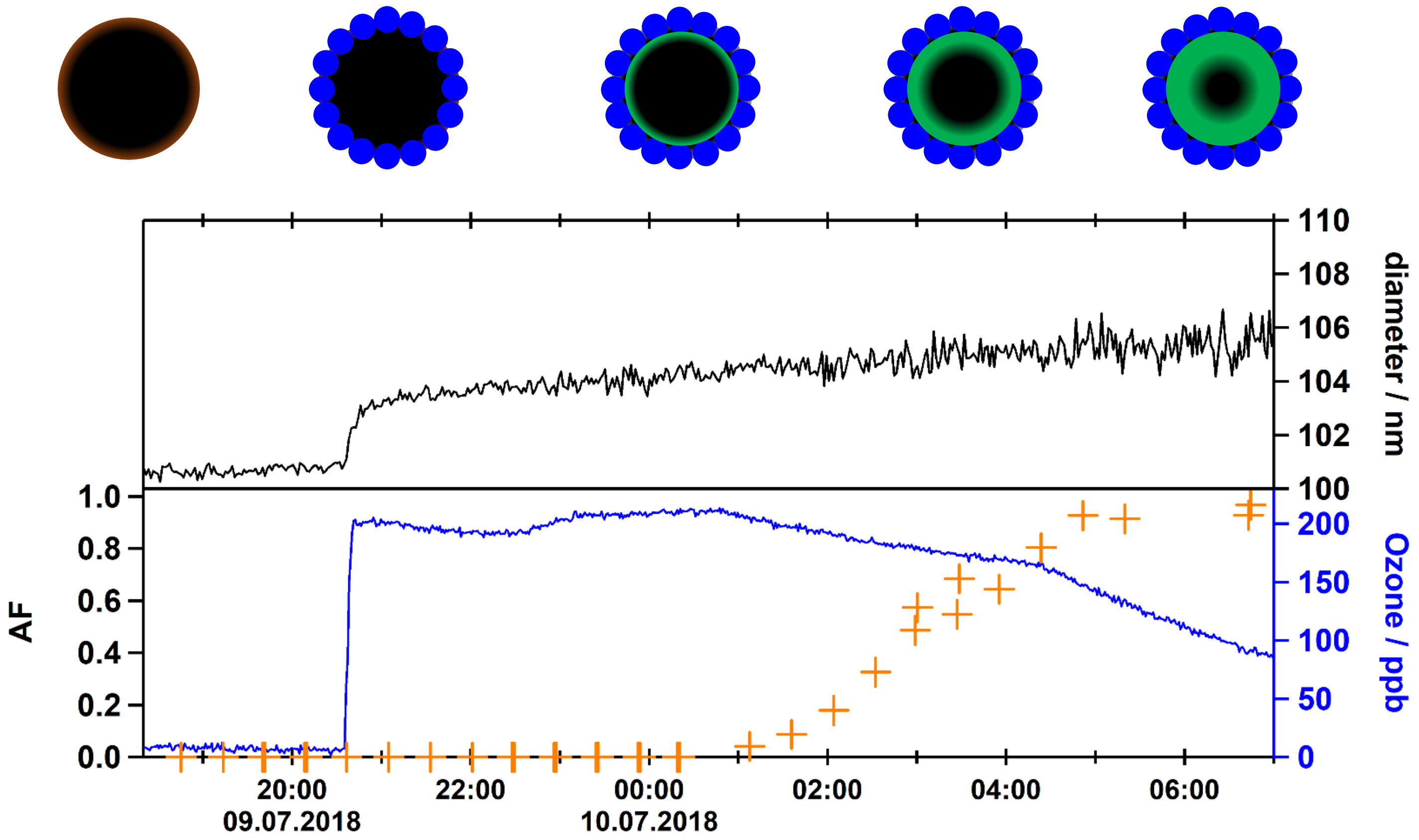


Adding 200 ppb ozone → adsorption

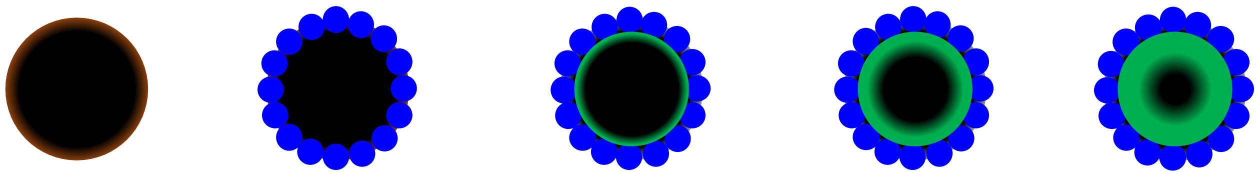




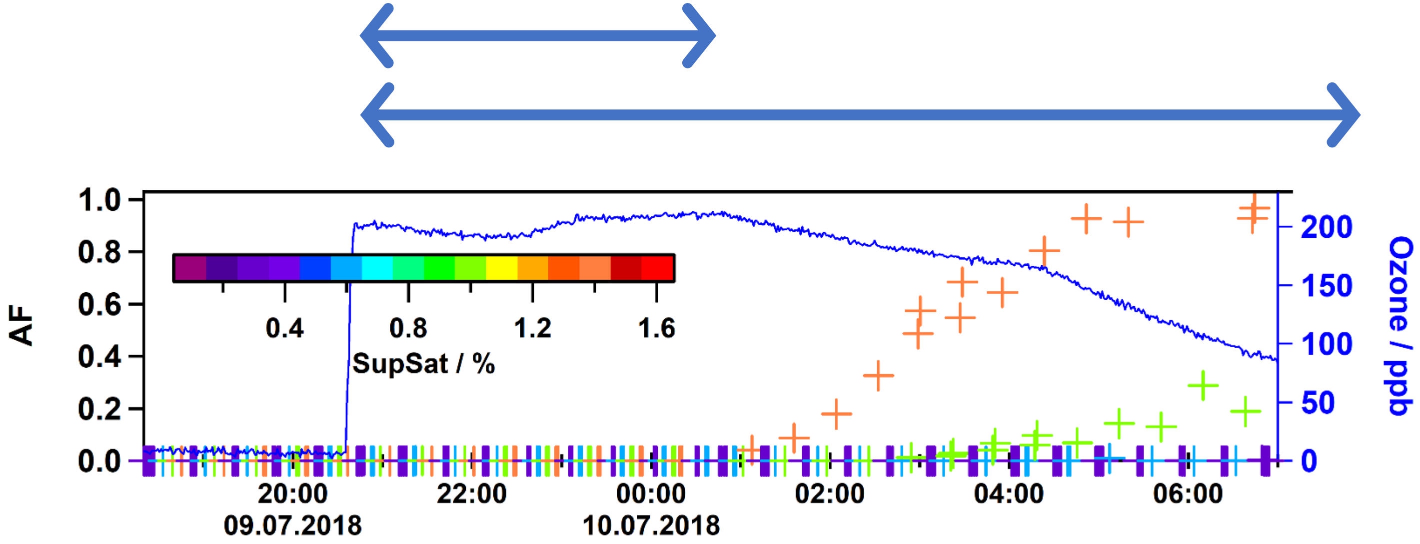
Continuous exposure to 200 ppb ozone



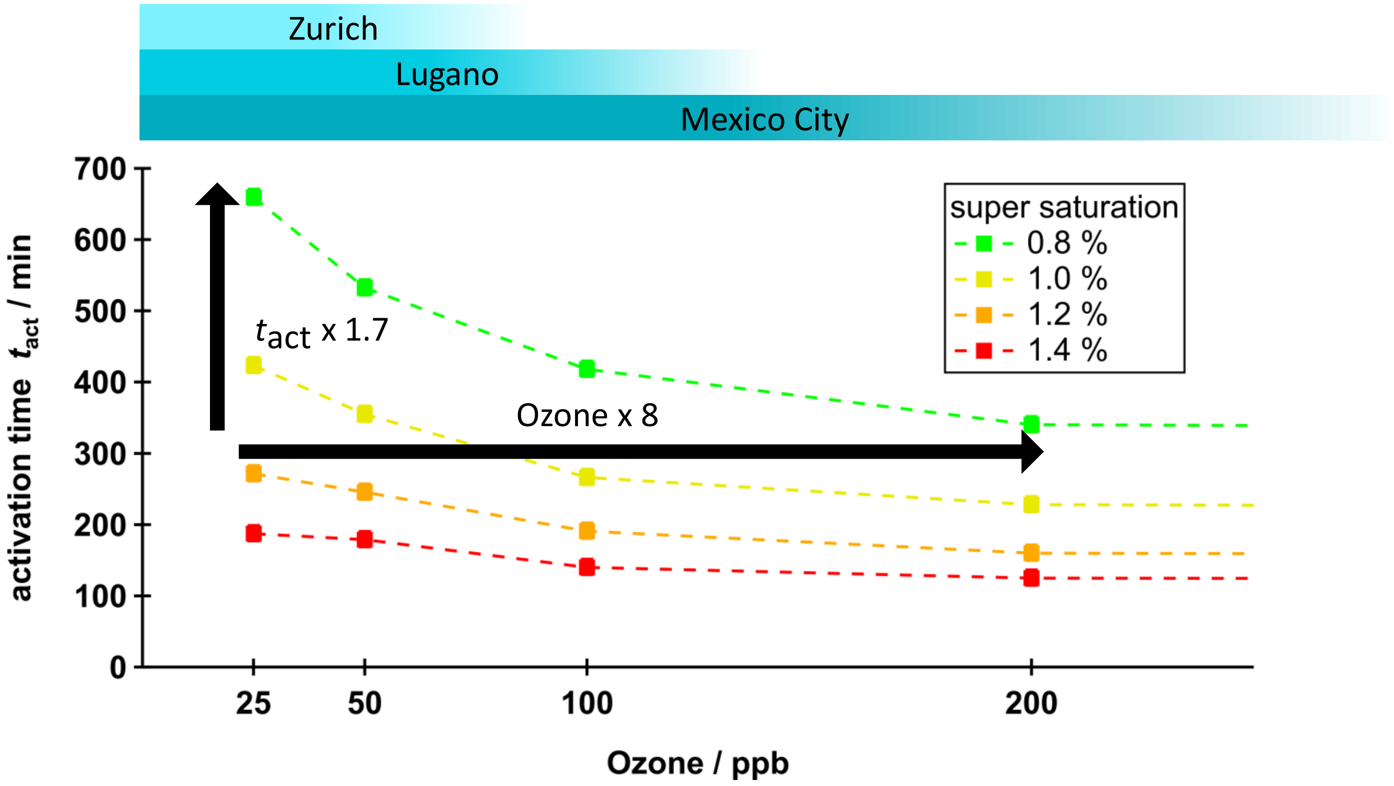
Activation time t_{act}



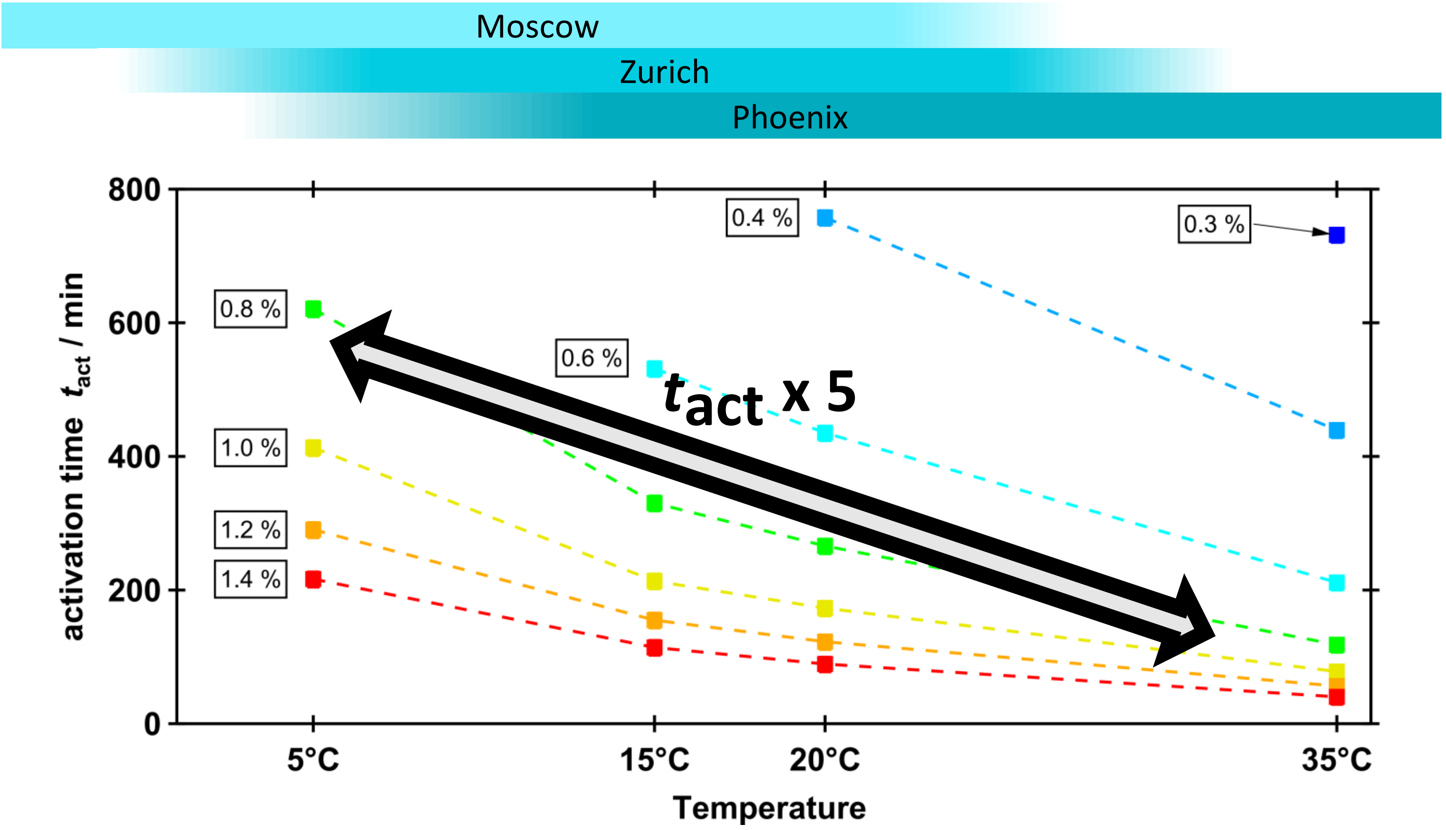
t_{act} = 3 - 12 h



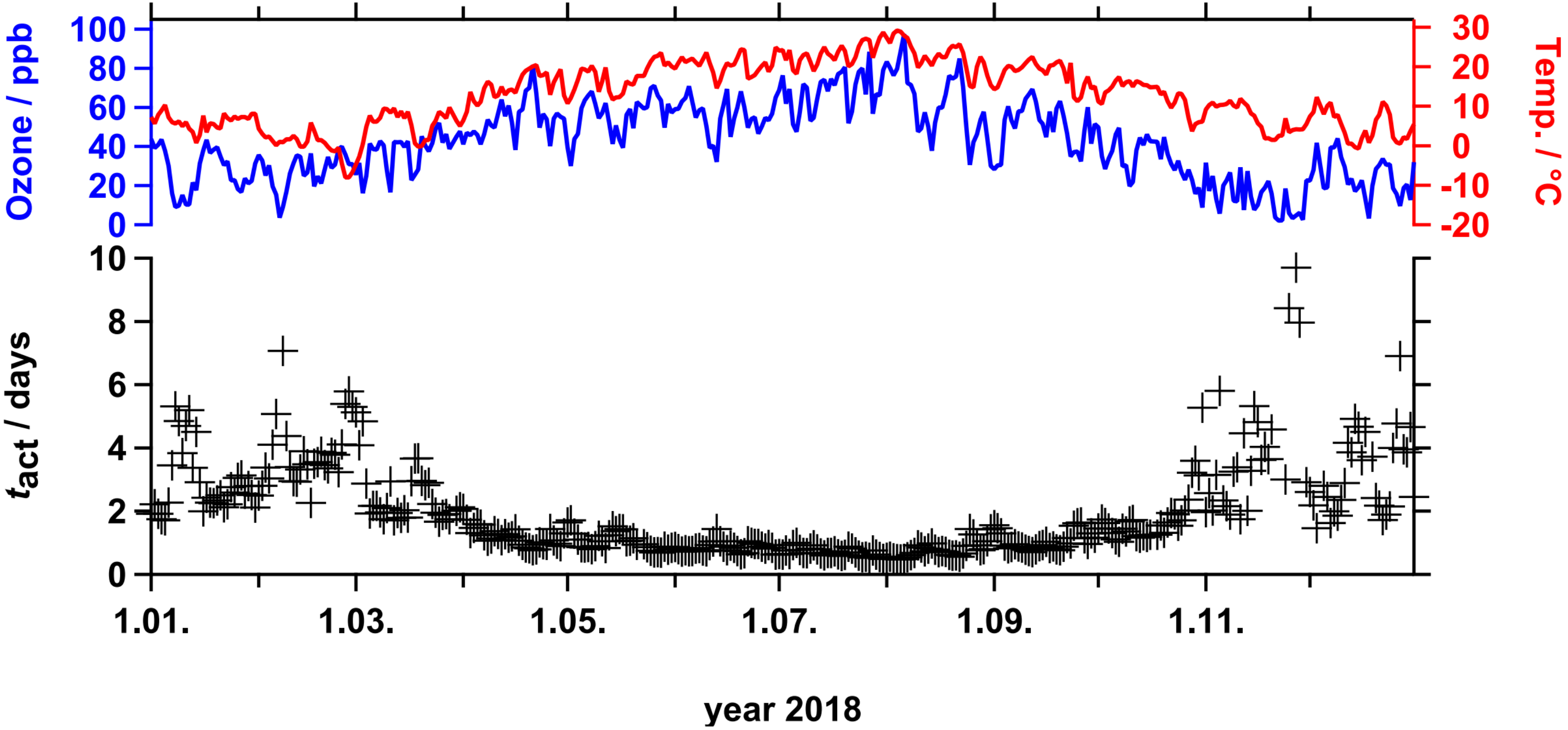
Activation time vs. ozone concentration



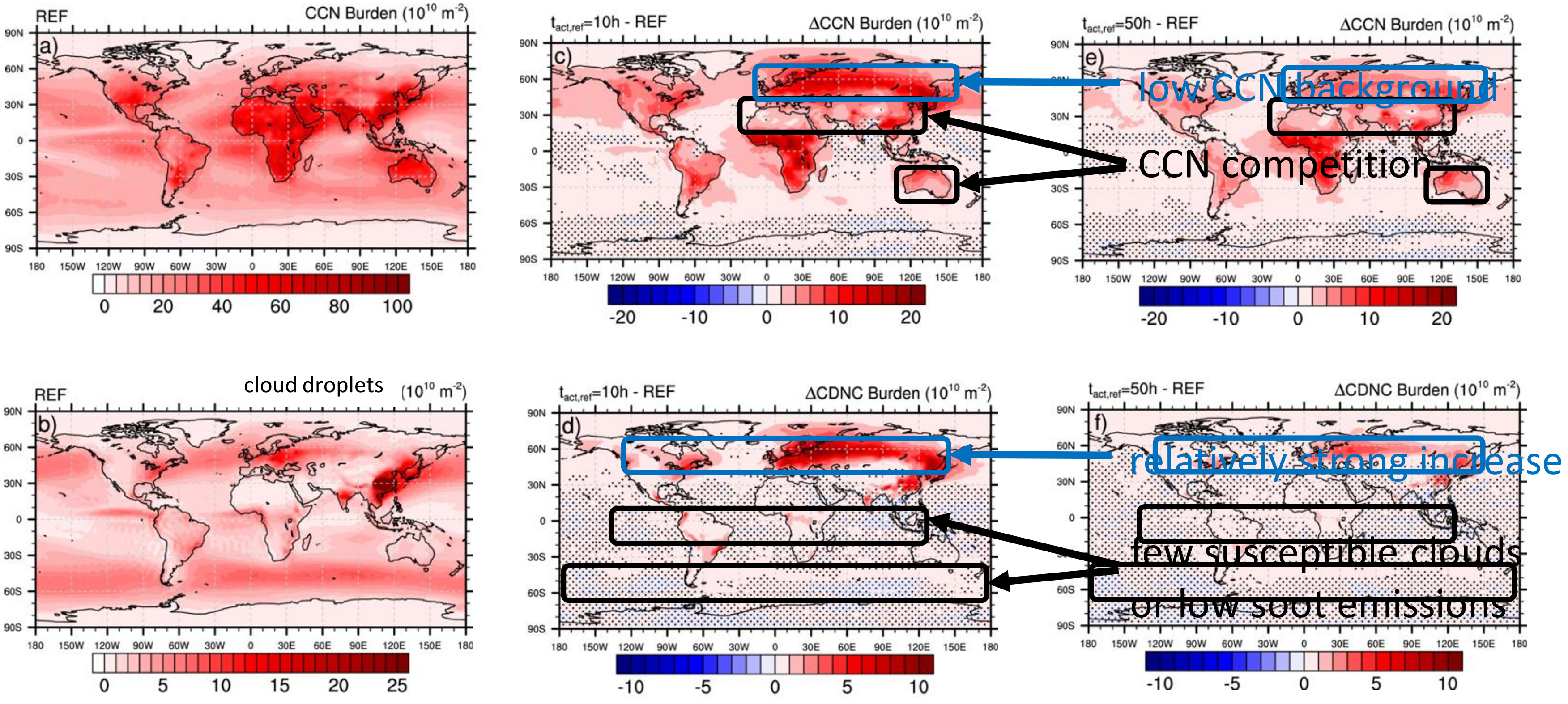
Temperature dependency at 200 ppb O_3



Activation times of soot in Zürich

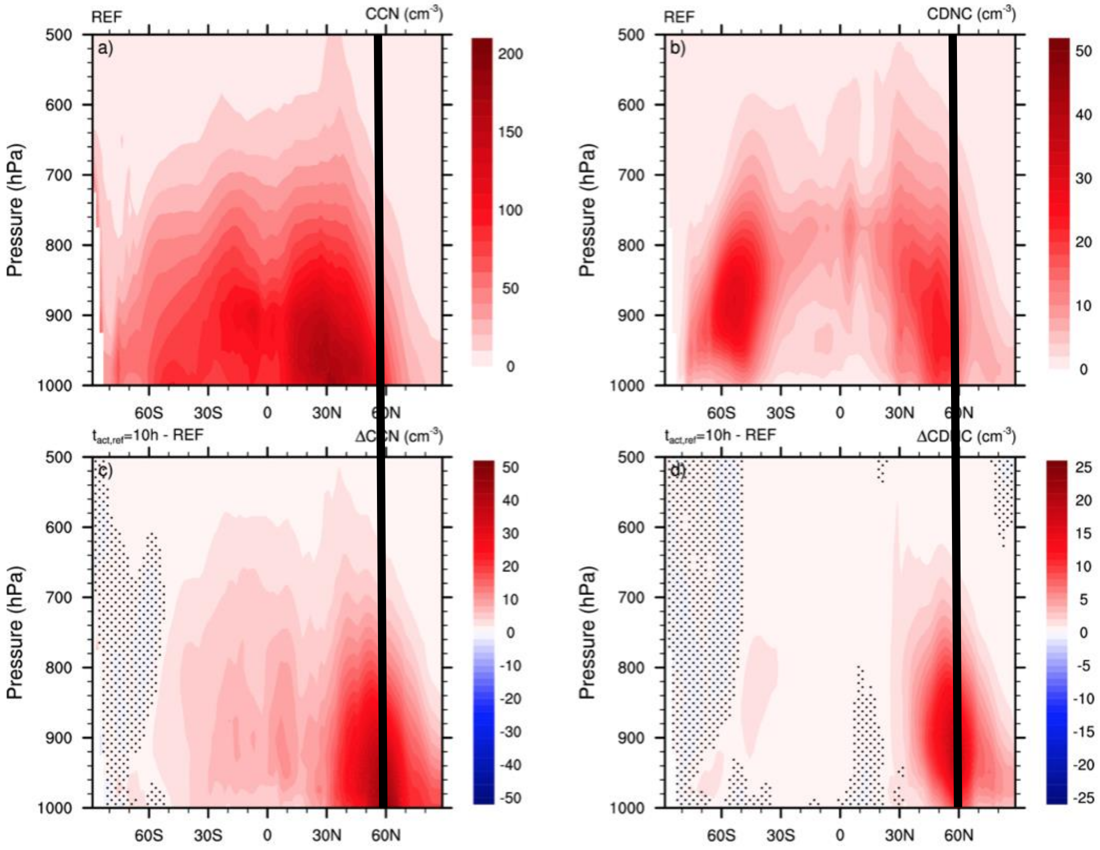


Climate impact of ozone-aged soot



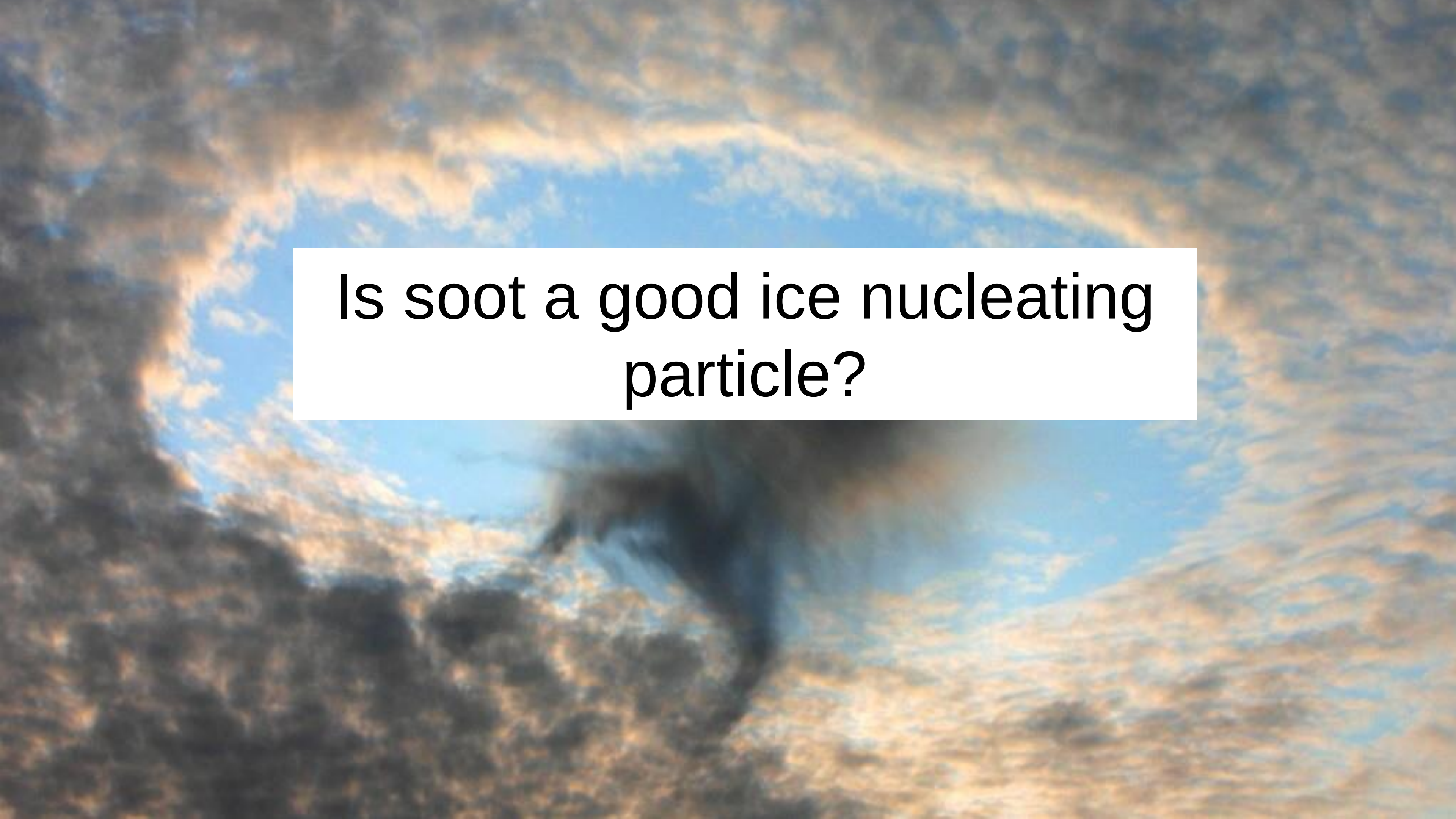
→ 93% increase in cloud droplet burden north of 60 ºN for 10h activation time

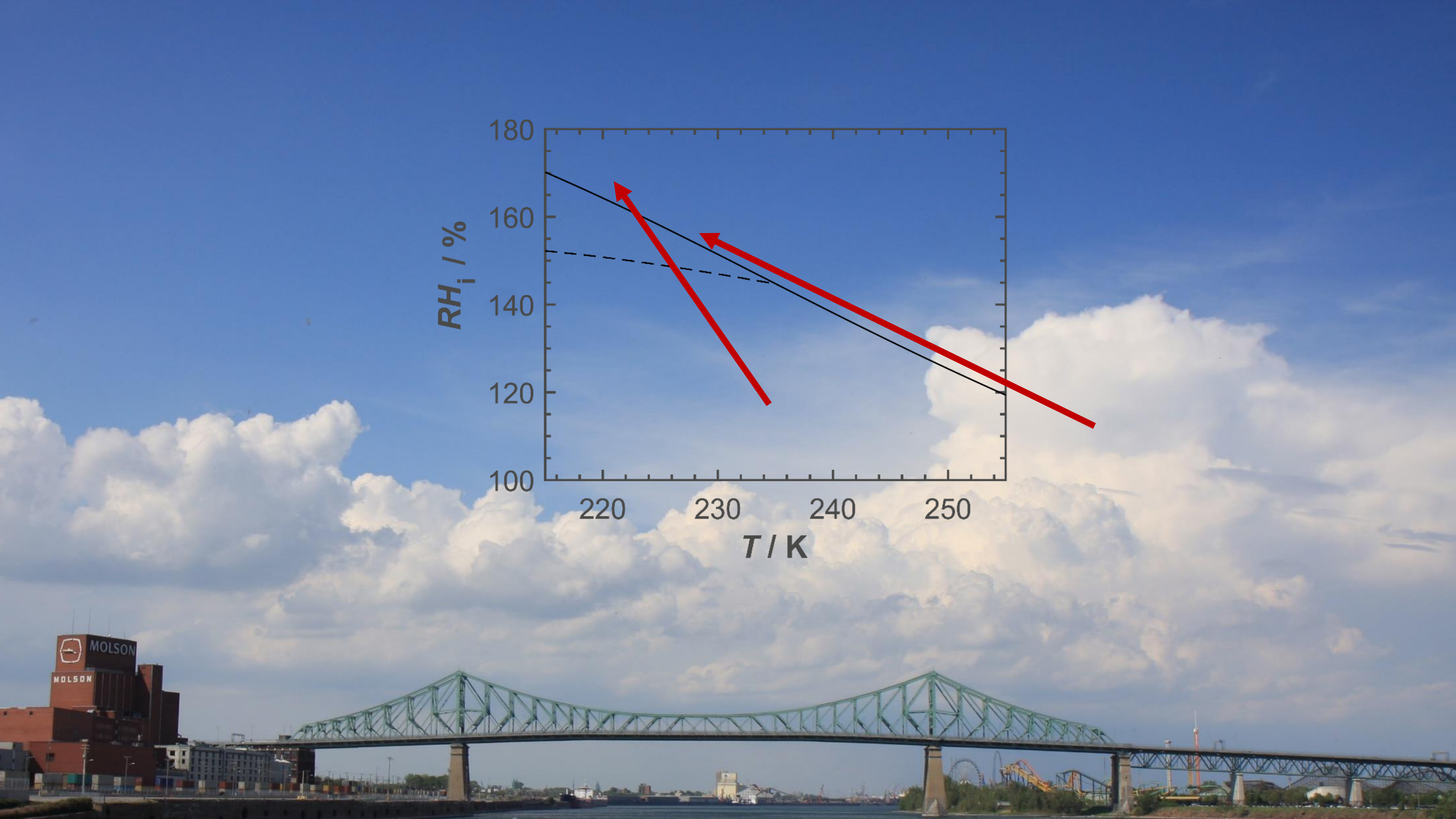
Climate impact of ozone-aged soot



--> Largest impact of ozone as CCN at around 60 ºN

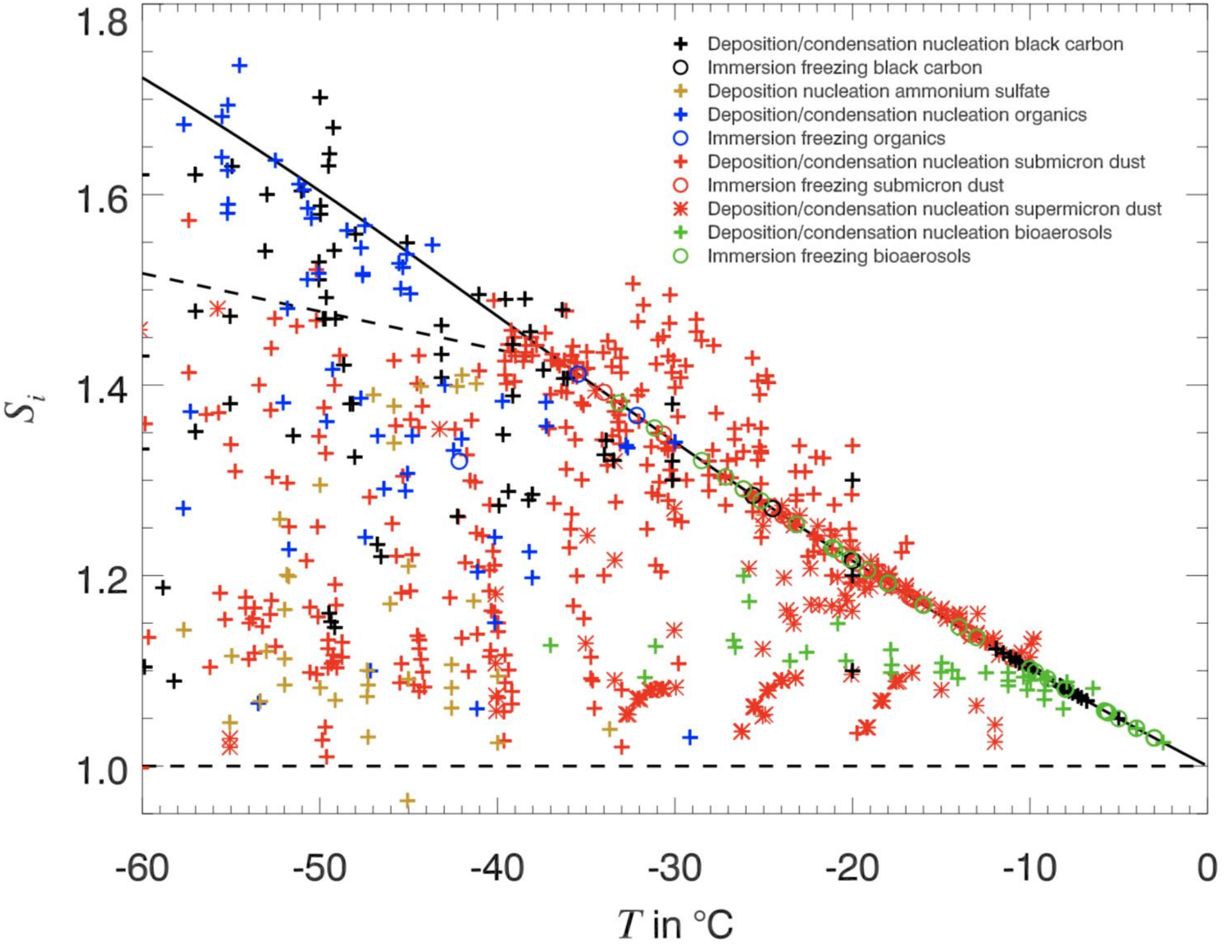
Is soot a good ice nucleating particle?





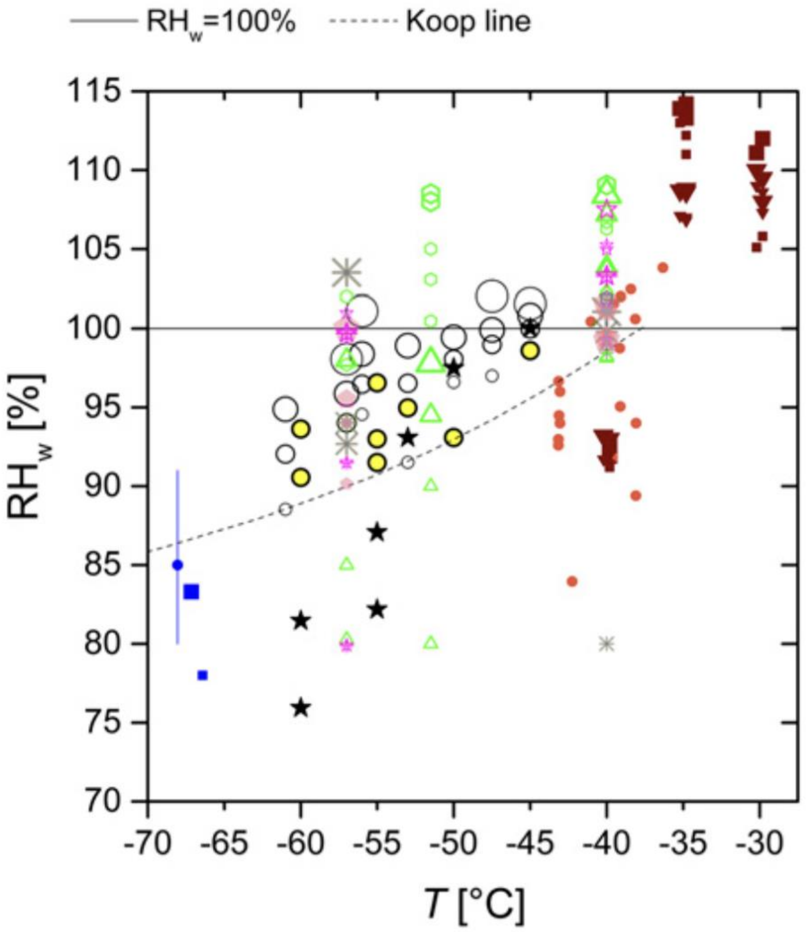
180
160
140
120
100
220
230
240
250
RHi / %
T / K
MOLSON
MOLSON

Ice nucleation onset supersaturations (S_i) and temperatures



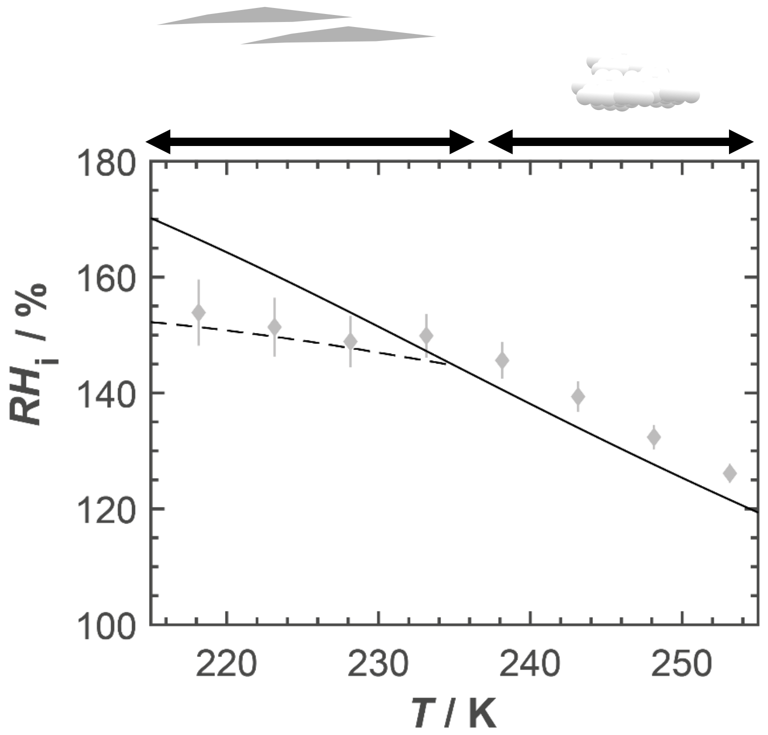
Hoose and Möhler, 2012

Ice nucleation onset for different soot types



Kanji et al., 2017

Ice nucleation activity of soot particles

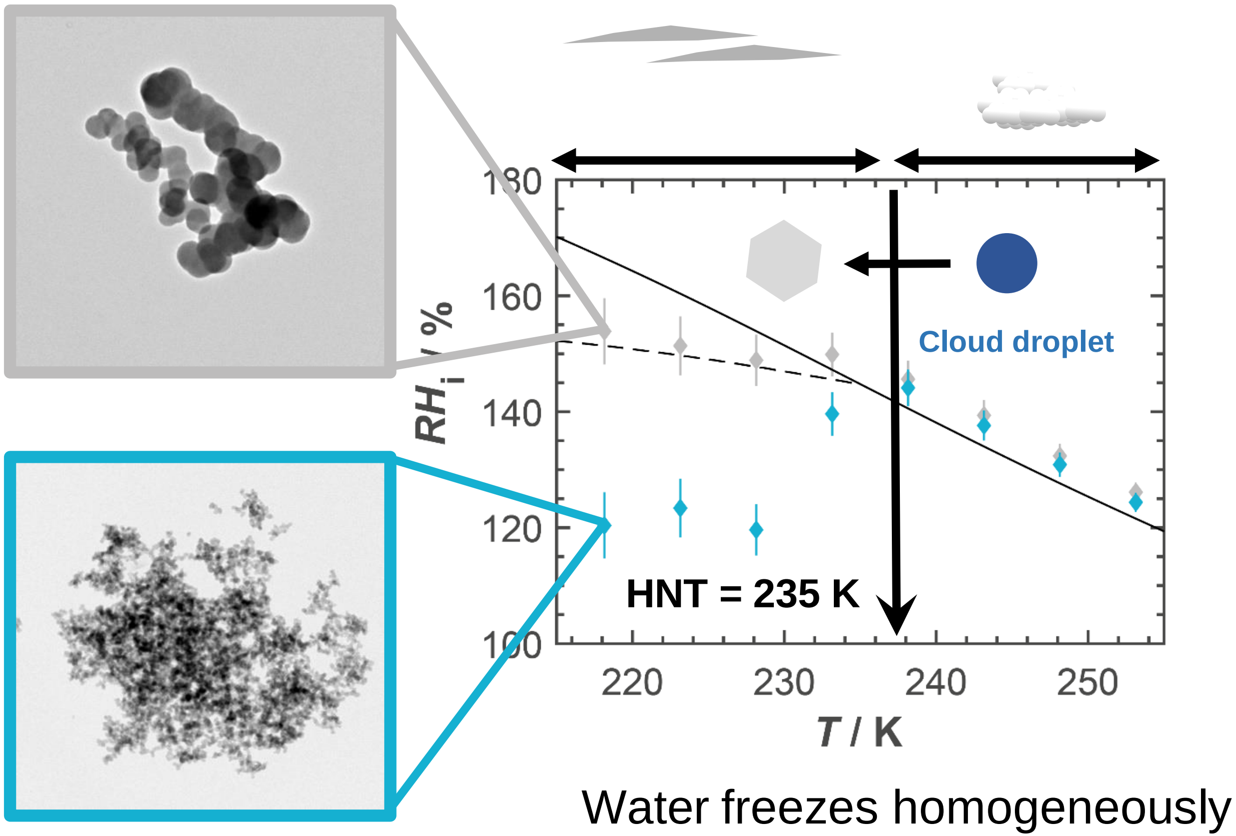


Water saturation (RH_w = 100%)

Homogeneous freezing of solution droplets

Ice nucleation activity of soot particles

- Different soot types have different physicochemical properties.
- Strong temperature dependence of ice formation.
- Implies involvement of liquid water.



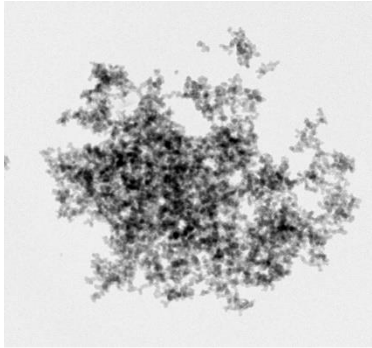
Mahrt et al., ACP, 2018

Pore condensation and freezing

Non-porous particle

- Particle surface
- Condensed water
- Ice

$RH_w \geq 100$ %



Porous particle

$RH_w < 100$ %

T < HNT

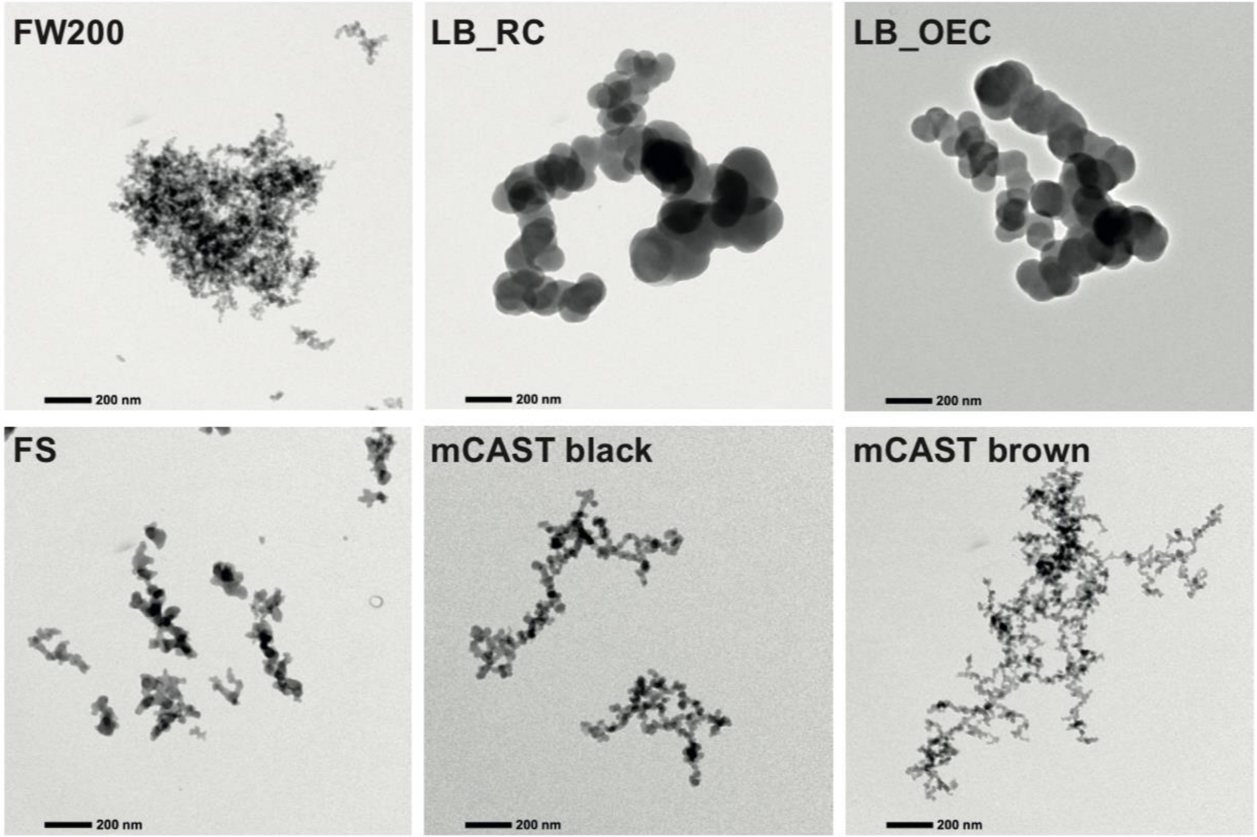
- Water is taken up by capillary condensation at $RH_w < 100\%$.
- Pore water freezes homogeneously at T < 235 K

Marcolli, ACP, 2014; David et al., PNAS, 2019

How often do we find soot with pores?

Atmo-spherically aged soot

Lamb black



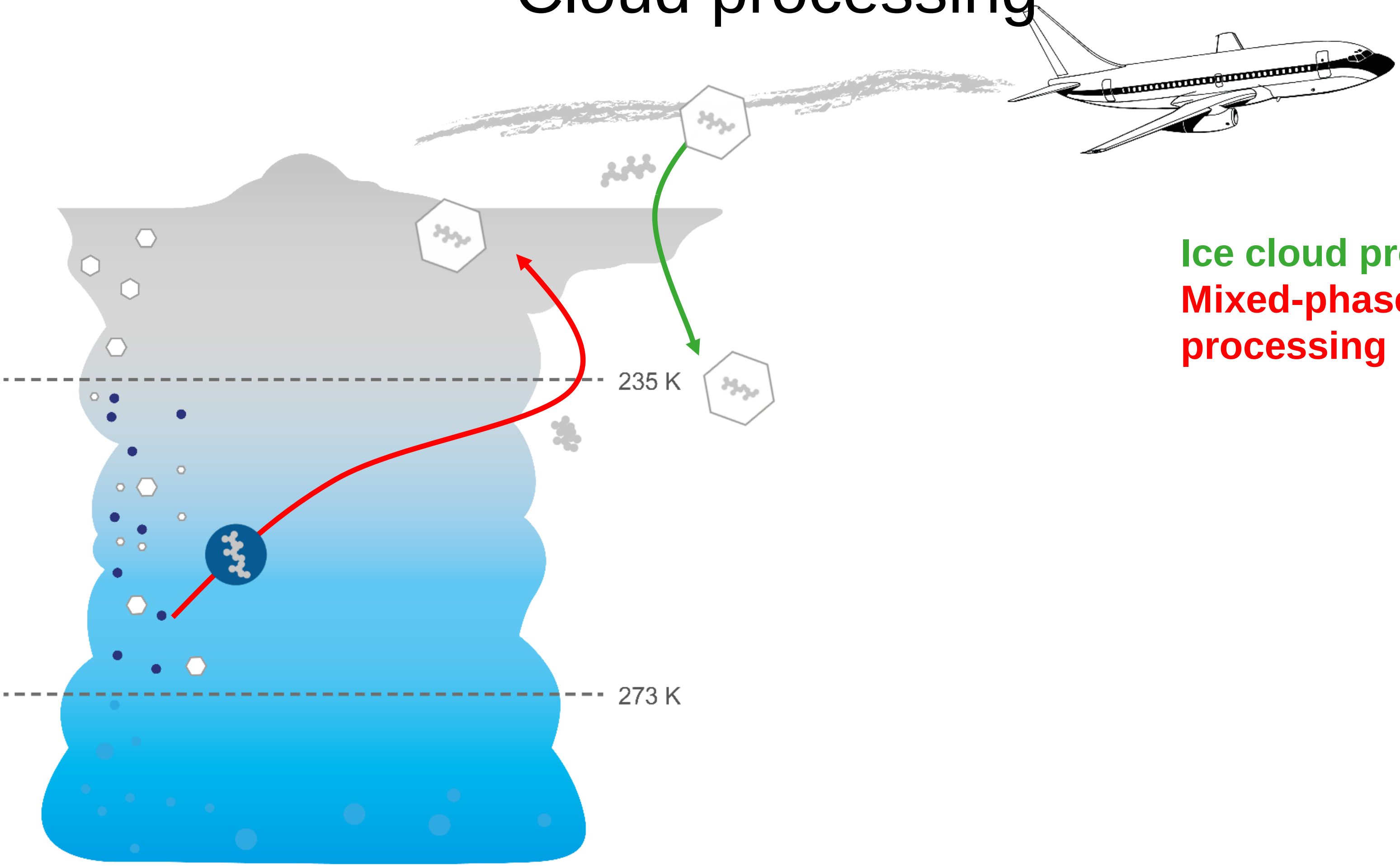
Diesel soot

Aircraft soot

Mahrt et al., ACP, 2018

Cloud processing for cirrus clouds

Cloud processing



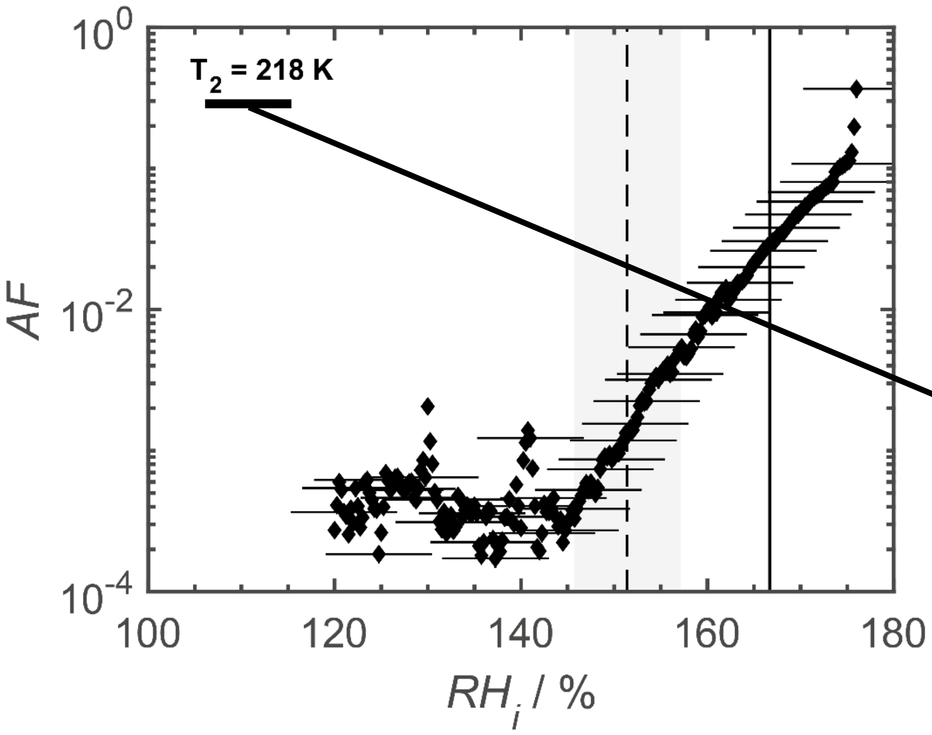
Ice cloud processing

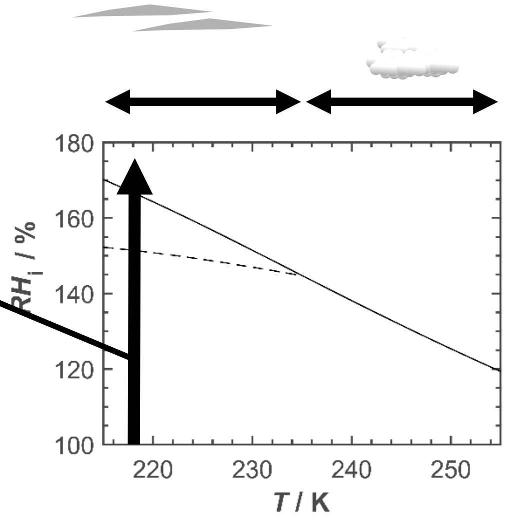
Mixed-phase cloud processing

Mahrt et al., JGR, subm.

Cloud processing

Unprocessed soot





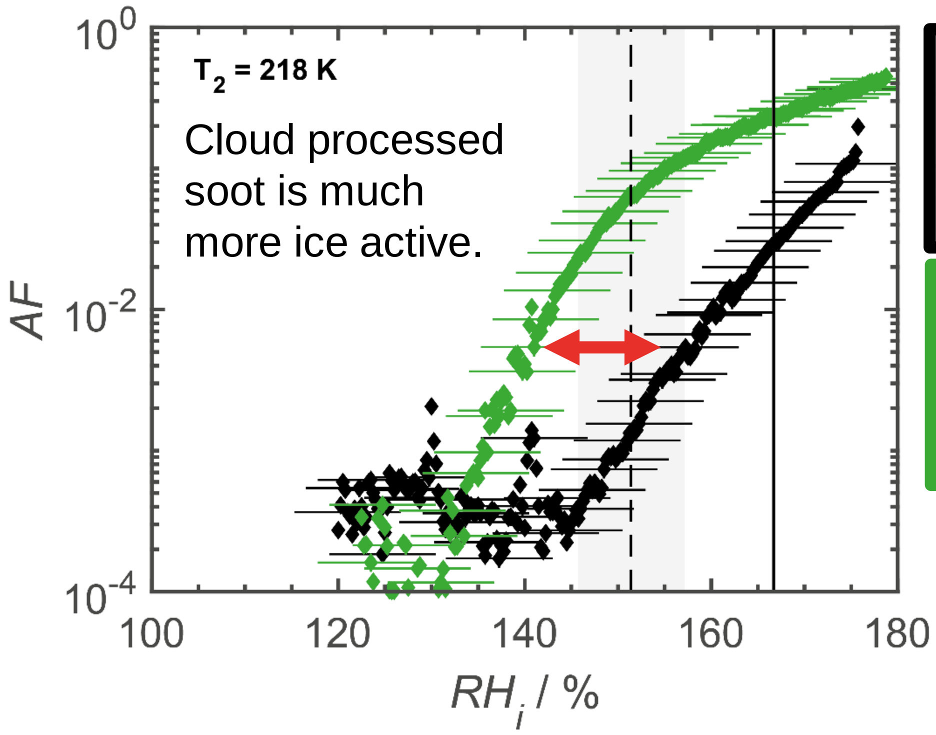
Water saturation (RH_w = 100%)

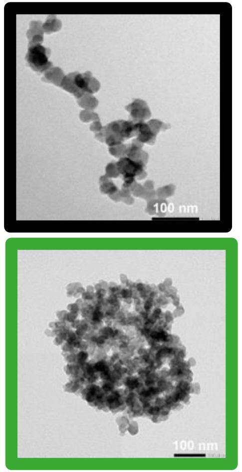
Homogeneous freezing of solution droplets

Cloud processing

Unprocessed soot

Processed (Cirrus)



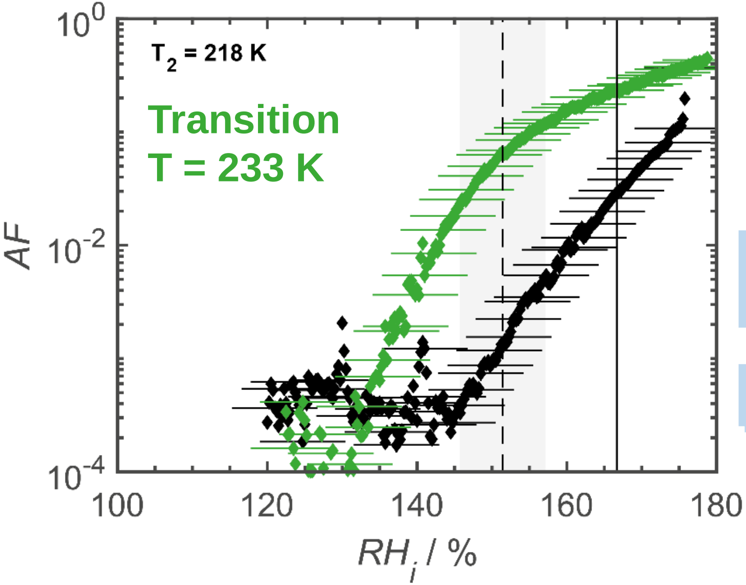


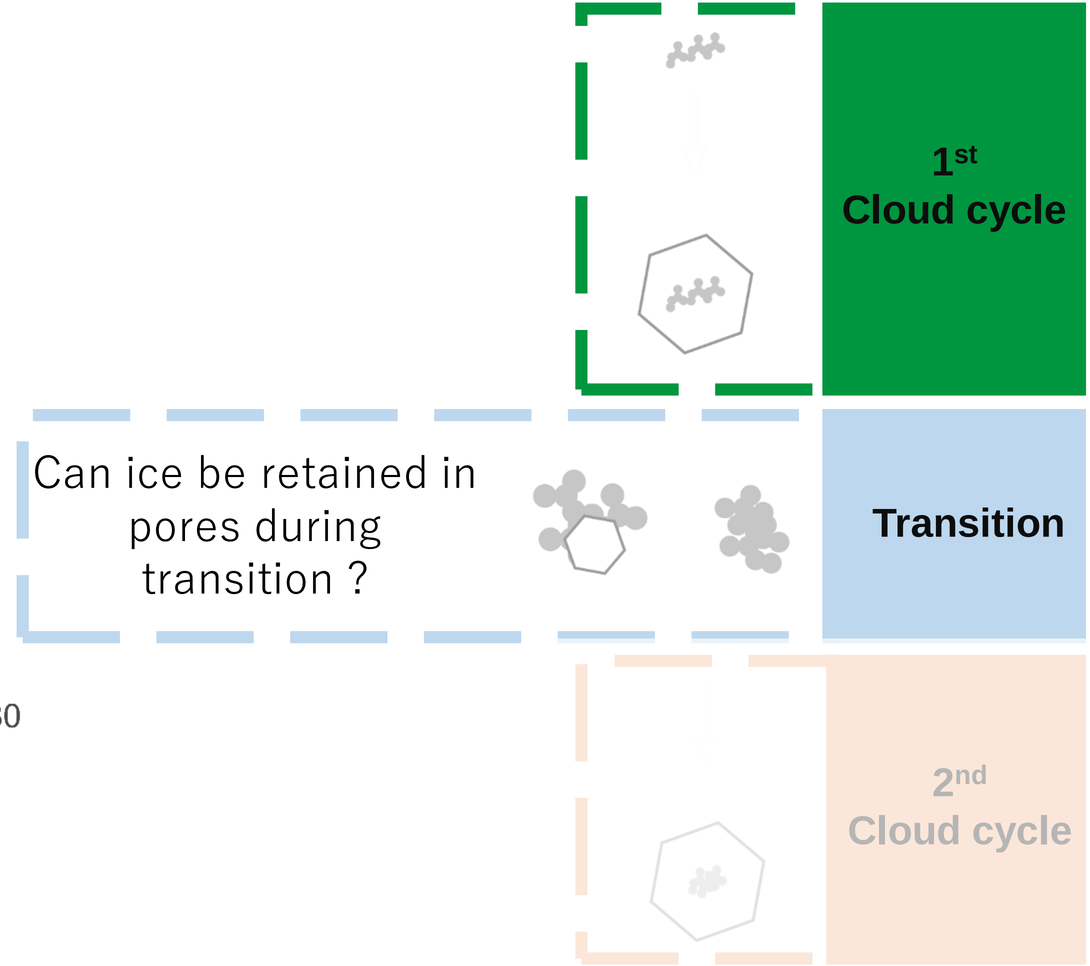
Water saturation (RH_w = 100%)

Homogeneous freezing of solution droplets

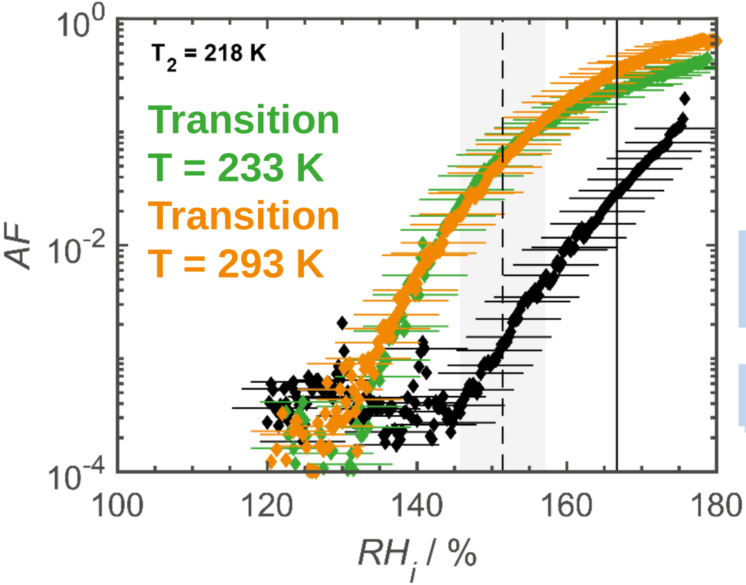
Impact of processing vs. transition

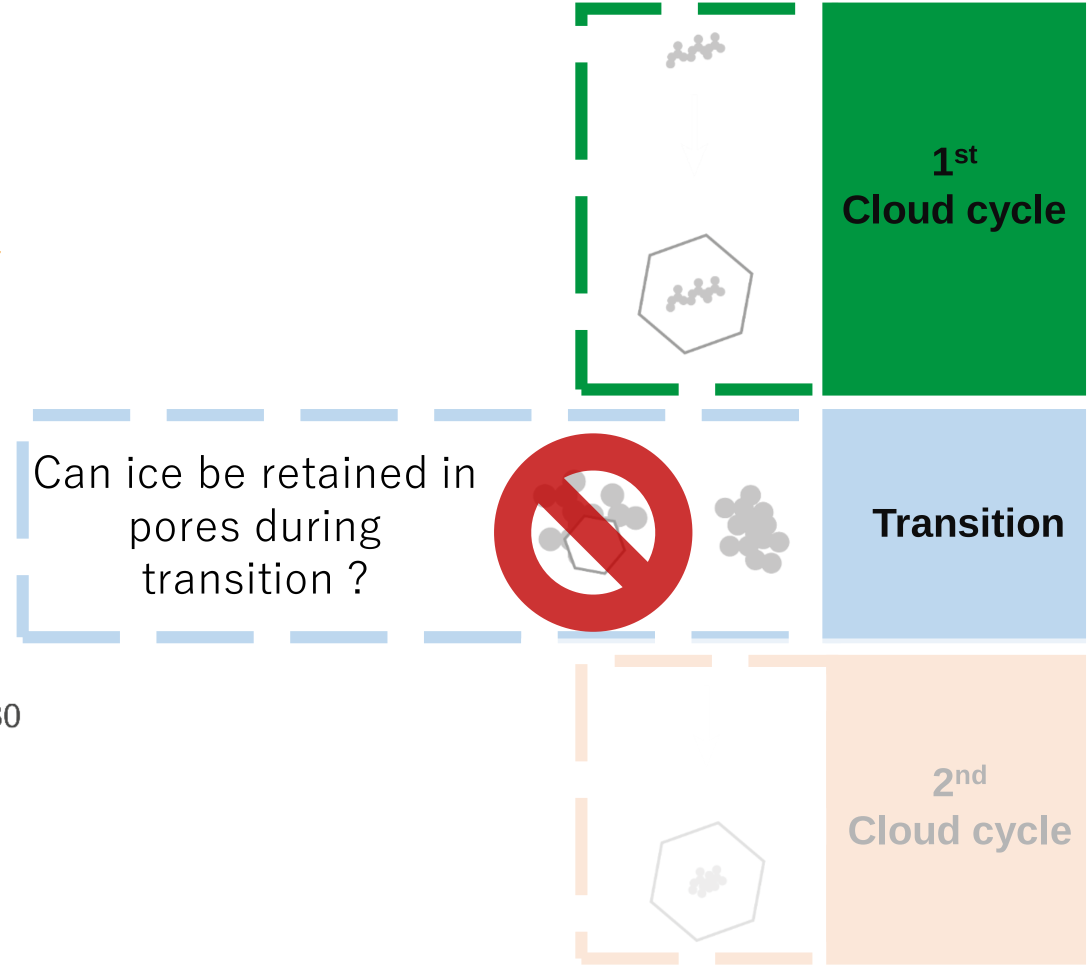
Unprocessed
Processed (Cirrus)





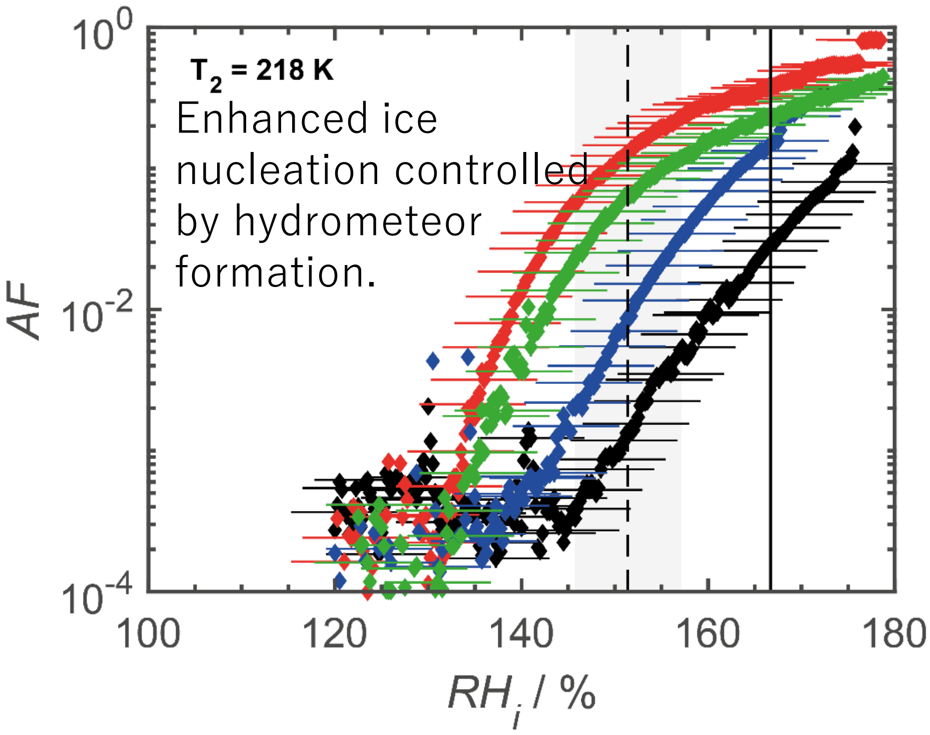
Impact of processing vs. transition





Cloud processing

Unprocessed soot
Processed (Cirrus)
Processed (MPC)
Pre-cooling

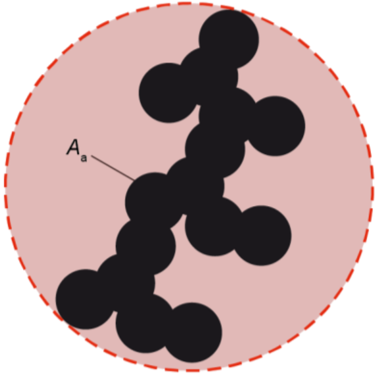


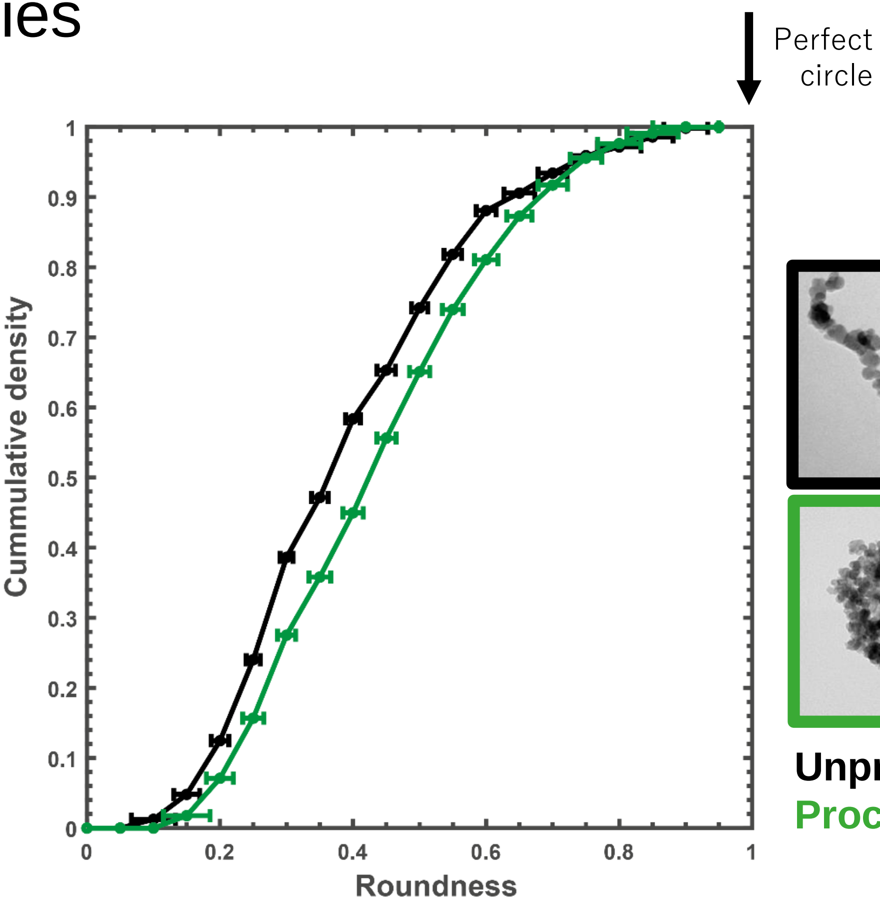
Water saturation (RH_w = 100%)

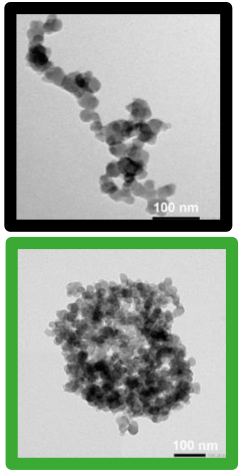
Homogeneous freezing of solution droplets

Hydrometeor formation changes soot morphological properties

- Cloud processed soot aggregates are more compacted.

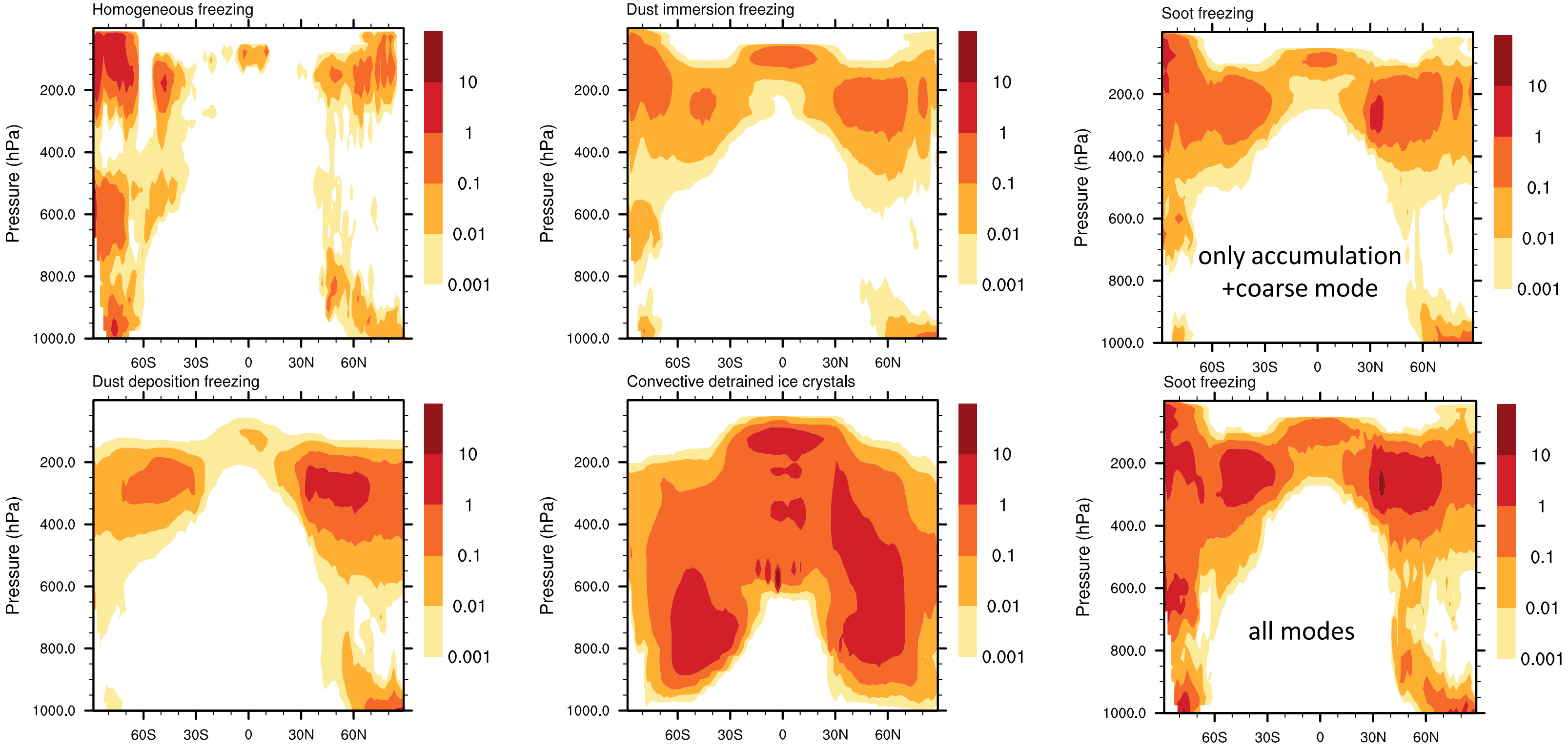






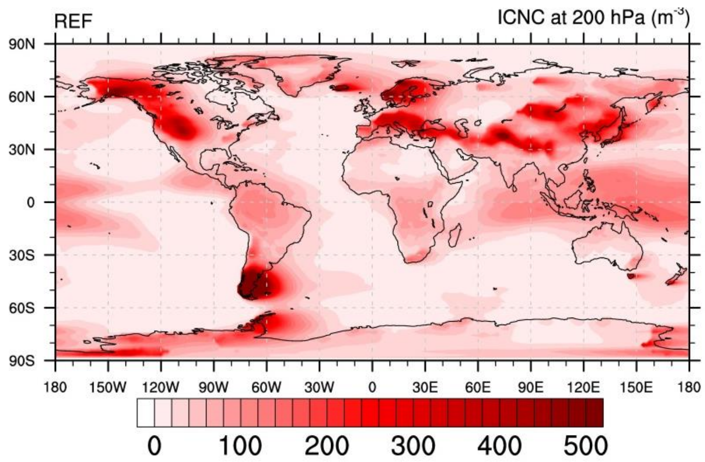
Unprocessed
Processed

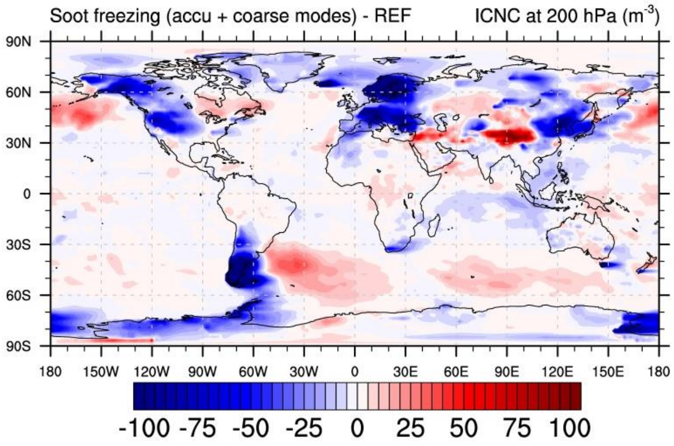
Impact on ice crystal number concentration

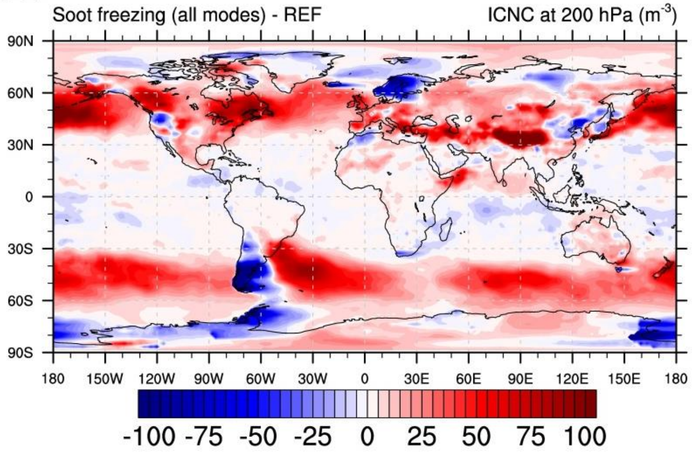


→ Cloud processing of soot makes soot potentially as important as mineral dust

Impact on ice crystal number concentrations (ICNC)





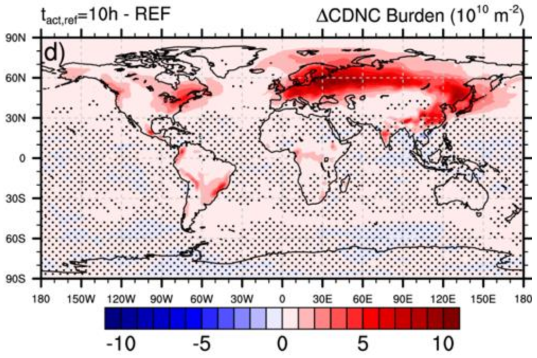


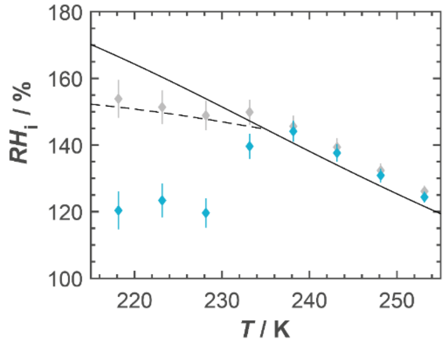
Take-home messages

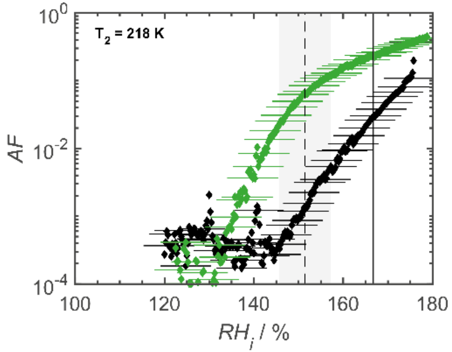
1. How important is soot as a CCN?
   - ➢ It depends on the organic carbon content
   - ➢ It can be important if aged with ozone at atmospheric conditions and can increase the cloud droplet burden significantly on the Northern Hemisphere

1. How important is soot as an INP?
   - ➢ This depends on the soot type; in general soot types with pores are favoured
   - ➢ Soot INP could noticeably increase the ice crystal concentration in mid latitudes

2. How important is cloud processing of soot for cirrus clouds?
   - ➢ It lowers onset RHw by 10% and can then rival with other INPs
   - ➢ Depending on how much soot act as INPs, the global ice crystal burden can be decreased or increased

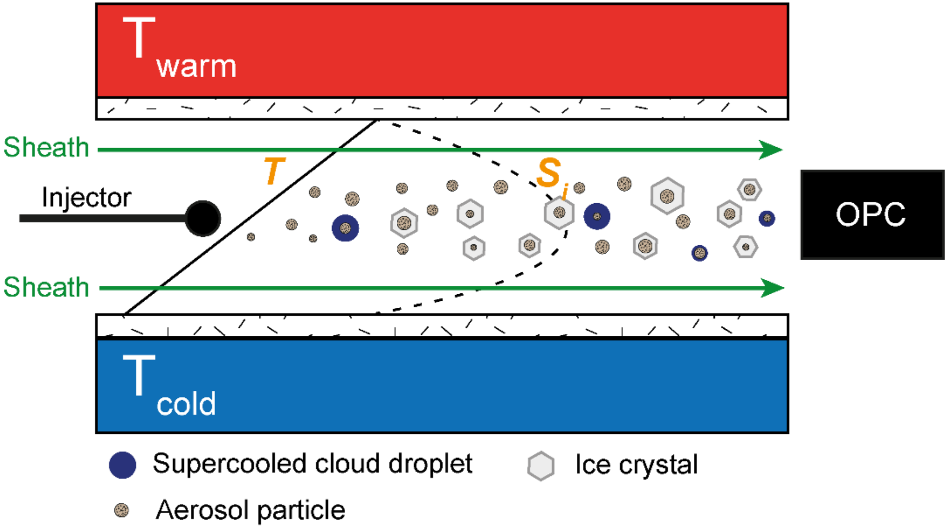


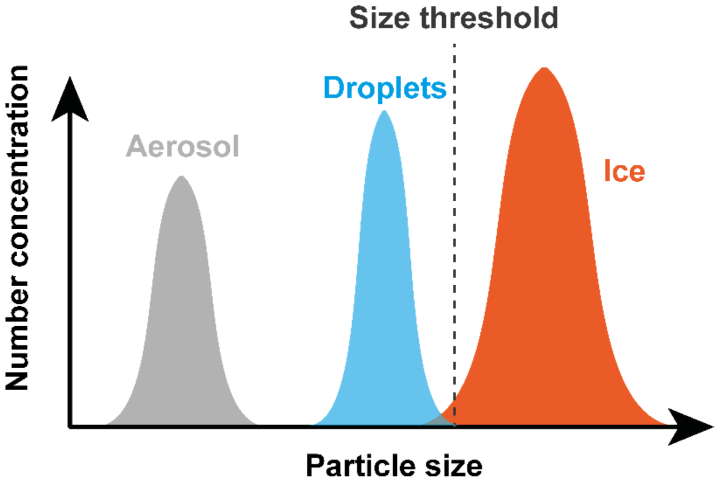





Questions?

Quantifying the ice nucleation activity of soot using the Horizontal Ice Nucleation Chamber (HINC)





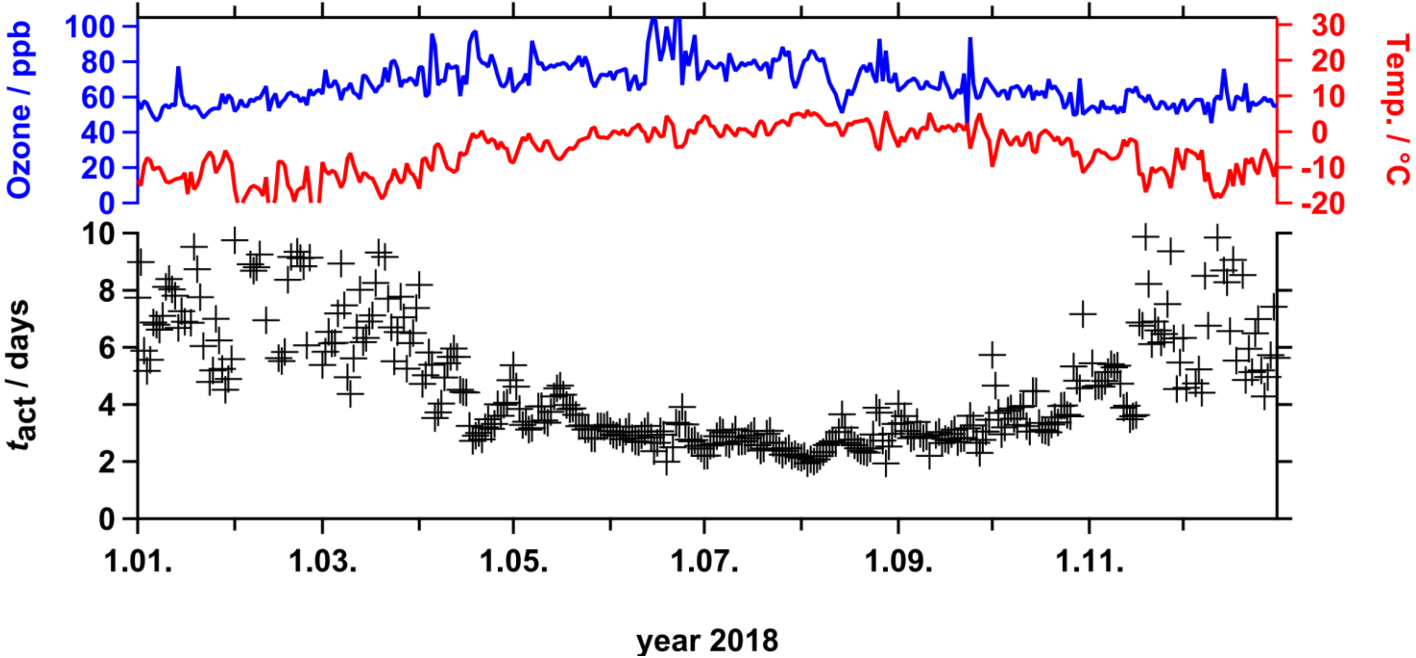
$$\textbf{Active Fraction} = \frac{N(Ice)}{N(Aerosol)}$$

NABEL station: Jungfraujoch

$$t_{act} = \left(A \cdot e^{\frac{-E_A}{R \cdot T}} \cdot \frac{K_{eq} \cdot C_{ozone}}{1 + K_{eq} \cdot C_{ozone}} \right)^{-1}$$

K_{eq} = 0.0229 ppb^{-1}

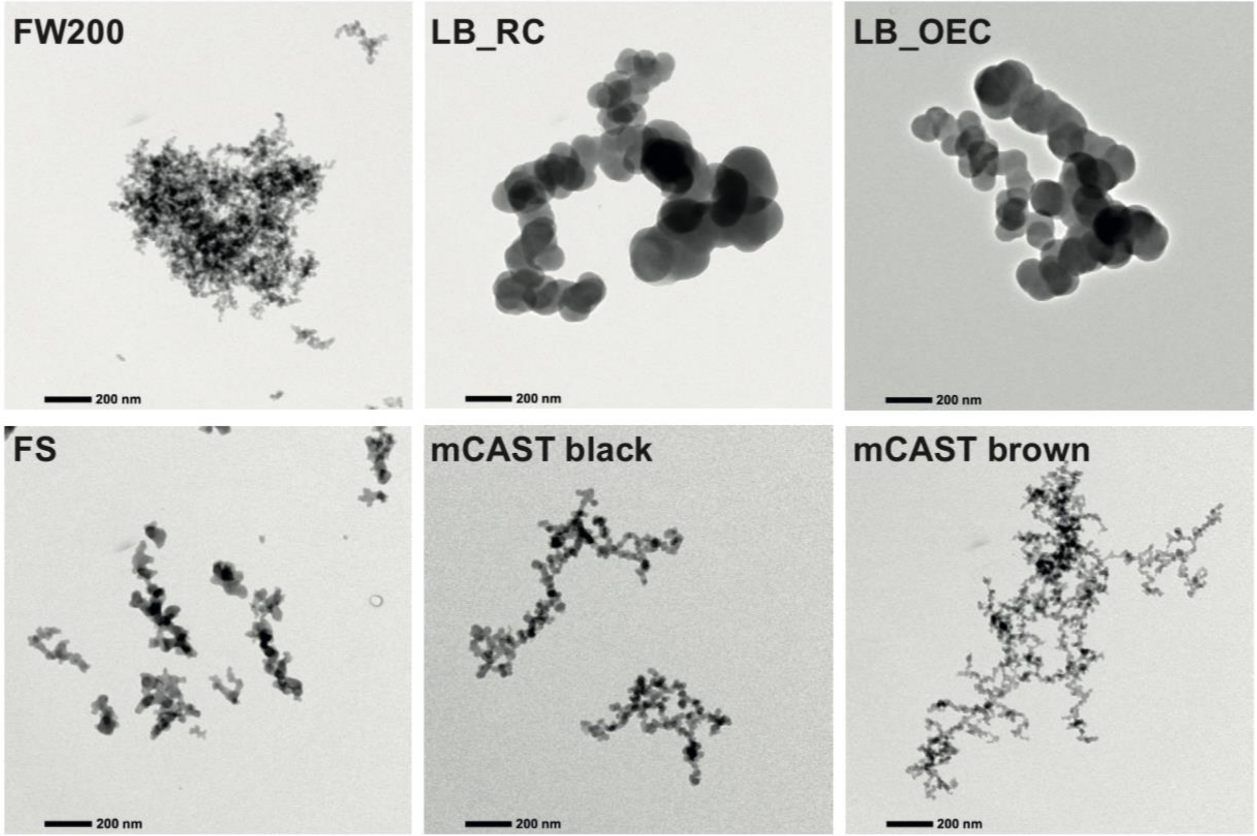
p = 650 mbar



Different soot types

Atmo-
spherically
aged soot

Lamb
black

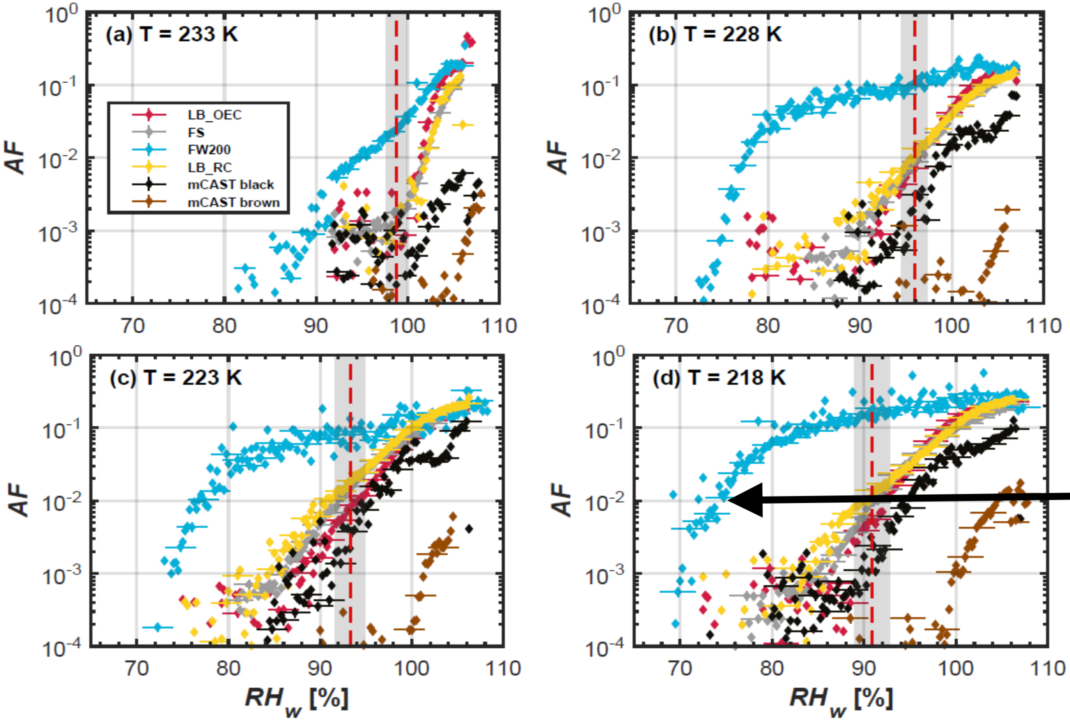


Diesel
soot

Aircraft
soot

Mahrt et al., ACP, 2018

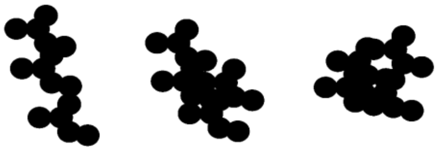
Cirrus cloud regime: 400 nm soot particles



Which mechanism is responsible for ice nucleation?

Low RH (and steep) activation curves of FW200 soot suggest PCF type mechanism

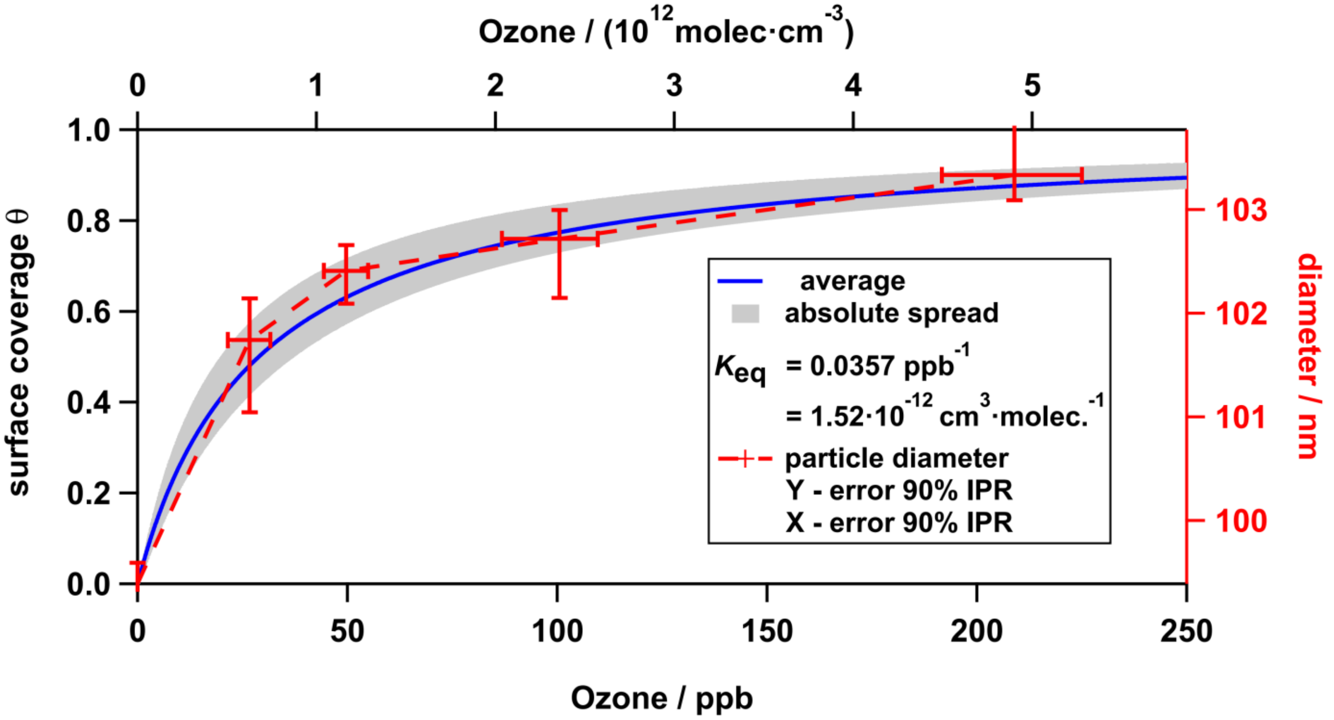
Can particle morphology (pores) give insight?

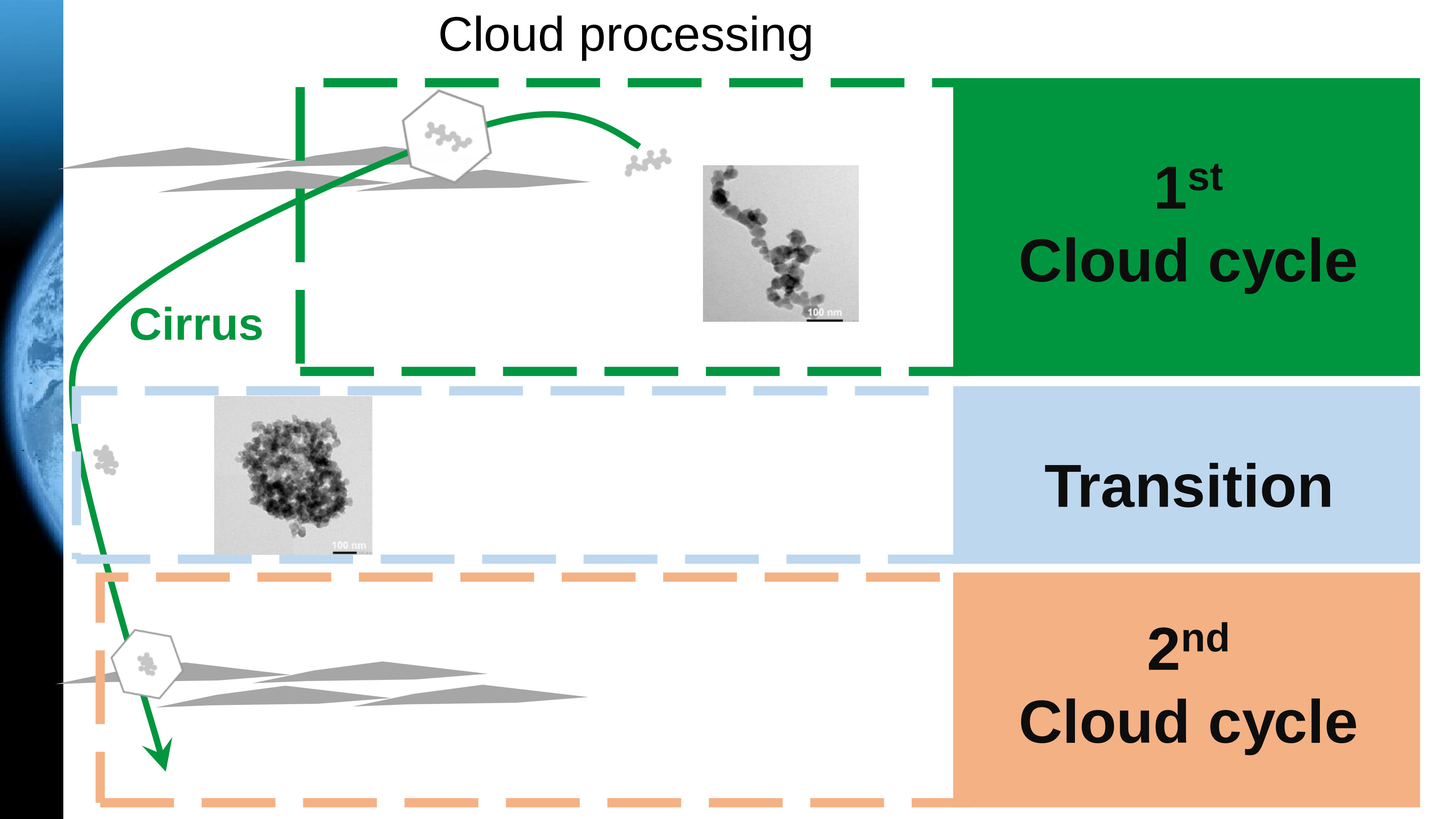


Mahrt et al., ACP, 2018

Langmuir-adsorption: O_3 adsorption is the bottleneck for soot aging

$$\theta = \frac{K_{eq} \cdot C_{ozone}}{1 + K_{eq} \cdot C_{ozone}}$$




Cloud processing
Cirrus
100 nm
1st
Cloud cycle
100 nm
Transition
2nd
Cloud cycle