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Low Tropospheric Ozone Over the Indo-Pacific Warm Pool Related to Non-Electrified Convection

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Key Points:

- Deep convective systems and strong lightning activity are observed over Northern Australia in early 2024
- The Indo-Pacific Warm Pool features deep convection in the absence of lightning which coincides with observations of low O_3
- The low observed O_3 is caused by the updraft of O_3-poor air from the marine boundary layer and a dearth of photochemical production

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Lightning is the most important source of nitric oxide (NO) in the tropical upper troposphere and controls the formation of tropospheric ozone (O_3). It is associated with deep convection and occurs mostly over continents. The Chemistry of the Atmosphere Field Experiment in the Pacific (CAFE Pacific) was conducted in early 2024 from Cairns, Australia, taking airborne measurements across the Australian continent and the surrounding maritime regions. Based on cloud top properties, lightning data and in situ observations of NO, O_3 and carbon monoxide, we show that deep convection occurs over Northern Australia and the Indo-Pacific Warm Pool. While we identify strong lightning activity over Australia, we observe deep convection in the Warm Pool that is not electrified. Our observations of low O_3 in the Warm Pool can be attributed to O_3-poor air from the marine boundary layer, which is not replenished by photochemical production from NO at high altitudes.

Plain Language Summary Nitrogen oxides (NO_x) represent the sum of nitric oxide (NO) and nitrogen dioxide (NO_2) and are catalytic in tropospheric ozone formation. At elevated altitudes in the tropics, NO_x is predominantly produced by lightning due to high temperatures in the flash (>30,000°C). Therefore, lightning strongly affects photochemistry in the tropics, particularly the formation of ozone (O_3). This is important because O_3 is a greenhouse gas and hazardous to human health. We performed measurements of trace gases with a research aircraft over Australia and the Indo-Pacific Warm Pool between January and March 2024. The Warm Pool is a region of elevated sea surface temperatures northeast of Australia. The measured trace gases indicate lightning and the upward transport of air from the surface to high altitudes by deep convection. We find that deep convection and strong lightning occur over terrestrial Northern Australia. In contrast, deep convection occurs without lightning over the marine Warm Pool. This explains the low O_3 over the Warm Pool: O_3-poor air from the surface is transported to high altitudes, where it is not chemically replenished due to the absence of NO from lightning.

1. Introduction

Lightning is the dominant source of nitrogen oxides ($NO_x \equiv NO + NO_2$) in the upper tropical troposphere and accounts for around 10% of the global budget (Nussbaumer et al., 2023; Pusede et al., 2015; Schumann & Huntrieser, 2007). In tropical latitudes, lightning NO_x drives photochemical processes and regulates the formation of tropospheric O_3, a greenhouse gas and hazardous pollutant. It also regulates the abundance of OH radicals, which determine the lifetime of many gases, for example, methane (CH_4) and carbon monoxide (CO) (Crutzen, 1988; Lelieveld et al., 2016).

Lightning occurs in cumulonimbus systems through convective up- and downdrafts that carry ice and supercooled water at low temperatures, causing charge separation and the generation of a strong electric field. This process goes hand in hand with the upward transport of air from low altitudes and promotes reaction pathways for trace gases from the boundary layer under upper tropospheric conditions. It is well established that lightning over continents is substantially more frequent than over maritime regions (Avila et al., 2010; Christian et al., 2003; Lal et al., 2014; Rudlosky & Virts, 2021). While the relatively rapid heating of land masses and the presence of aerosol particles are important factors, the reason behind this uneven distribution of lightning has not been fully

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resolved. The North of Australia is known for its intense lightning activity along with deep convective updrafts (Heyes et al., 2009; Huntrieser et al., 2009; Vaughan et al., 2008).

The Indo-Pacific or tropical Western Pacific Warm Pool (in the following referred to as Warm Pool) is an oceanic region to the northeast of Australia. It is characterized by high sea surface temperatures above 28°C throughout the entire year which promote deep convection and a high tropopause (Bhat et al., 1996; Liu & Zipser, 2015; De Deckker, 2016; L. L. Pan et al., 2017). It is therefore not only relevant to local processes but of high importance to the global climate (De Deckker, 2016; Fasullo & Webster, 1999; Hu et al., 2020). The meteorological conditions give rise to unique photochemistry. Multiple studies over a range of several decades have presented in situ observations of O_3 in the Warm Pool or the surrounding areas, including (but not limited to) the PEM Tropics missions (1996), the BIBLE campaign (1998/99), the CAST and CONTRAST campaign (2014), and O_3 sonde measurements at several sites in the remote Pacific (Anderson et al., 2016; Gao et al., 2014; Harris et al., 2017; Hoell et al., 1999; Kondo et al., 2002; Labrador et al., 2005; Müller et al., 2024; Nicely et al., 2016; Oltmans et al., 2001; L. L. Pan et al., 2015, 2017). All agree on exceptionally low mixing ratios of O_3 at the surface (as low as 5 ppbv) and in the upper troposphere (around 20–30 ppbv). In contrast, observations of NO_x are sparse and range between 10 and 50 pptv. Gao et al. (2014) hypothesized that NO- and O_3-free air from the marine boundary layer is transported upwards through non-electrified deep convection, leading to low NO and O_3 at high altitudes. L. L. Pan et al. (2015) identified deep convection as the reason for low upper tropospheric O_3. Rex et al. (2014) further suggested a minimum of OH radicals concurrent with the low NO and O_3 levels and, consequently, enhanced lifetimes of greenhouse gases that are removed by OH in this region.

Trace gases with characteristic boundary layer concentrations can identify deep convective updraft when they are transported upwards and measured in the upper troposphere. Various studies have reported deep convection based on in situ observations including Cohan et al. (1999), Colomb et al. (2006), L. L. Pan et al. (2017). In Nussbaumer, Tadic, et al. (2021), Nussbaumer, Parchatka, et al. (2021) we have shown that deep convection occurs in a developing tropical cyclone with and without lightning dependent on the strength of the system. This conclusion was based on in situ observations during the Chemistry of the Atmosphere Field Experiment in Africa (CAFE Africa) in August and September 2018.

In this study, we present evidence of the occurrence of deep convective processes in the absence of lightning over the Warm Pool. Our analyses are based on in situ observations during the aircraft campaign CAFE Pacific (Chemistry of the Atmosphere Field Experiment in the Pacific), cloud top properties from the geostationary meteorological satellite Himawari and lightning strokes detected by the World Wide Lightning Location Network (WWLLN). In contrast, we report deep convection in the presence of strong lightning over the Australian continent. While previous studies reported indications for low NO and O_3 and the link to convection and lightning, this study is the first to demonstrate that deep convective updraft in the absence of lightning causes persistently low levels of O_3 (and NO) in the upper troposphere over the Warm Pool. With the help of a global atmospheric chemistry-climate model we evaluate the atmospheric implications of our findings and show that these conditions result in low OH levels, which in turn impact the lifetime of greenhouse gases such as methane.

2. Observations and Methods

2.1. Aircraft Campaign CAFE Pacific

CAFE Pacific took place from January to March 2024 over Australia, Papua New Guinea and the Indo-Pacific Warm Pool. Twenty-two local measurement flights were carried out with the HALO (High Altitude LOng range) research aircraft from the campaign airport in Cairns, Australia (16.88°S, 145.75°E). Figure 1 shows an overview of the flight tracks. We focus our analysis on data obtained over the Warm Pool and Australia, as denoted by the dark blue and dark red boxes, respectively. Each box comprises four major research flights (RF), which are RF10 (01/27-01/28), RF18 (separated into RF18a (02/16-02/17) and RF18b (02/17)) and RF21 (02/23-02/24) for the Warm Pool and RF11 (01/31-02/01), RF15 (separated into RF15a (02/09-02/10) and RF15b (02/10)) and RF19 (02/19) for Australia. We chose the boxes in order to compare the maritime influence on the edge of the Warm Pool with the continental conditions over Australia. We discuss the results in these areas in comparison with the continental (RF18 and RF21) and maritime background (mostly RF08: 01/23-01/24 and RF09: 01/26), shown by the light red and light blue boxes, respectively. We refer to the background as regions characterized by the absence of both lightning and convection. The research aircraft carried multiple scientific instruments for measurements of atmospheric trace gases, particles and meteorological data. In this study, we use

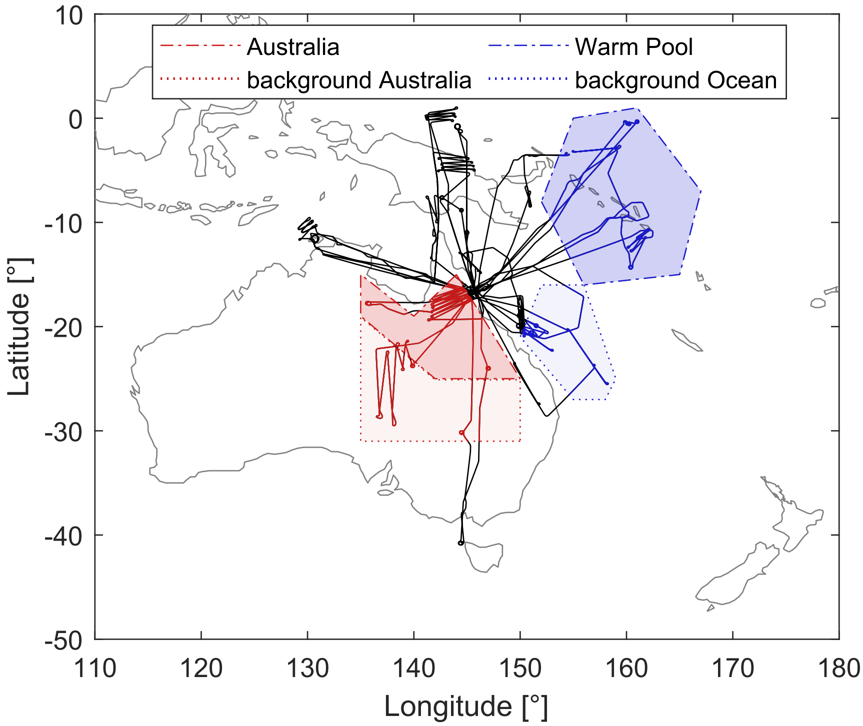

Figure 1. Overview of the CAFE Pacific flight tracks. Red colors show the continental and blue colors the maritime flight tracks. The dark boxes show the areas of interest (Warm Pool and Northern Australia) and the light boxes the continental and maritime background.

measurements of NO, O_3, CO, C_5H_8 and the local meteorology. Details on the instruments and uncertainties can be found in Text S1 of the Supporting Information S1.

2.2. Himawari Geostationary Meteorological Satellite

Data on cloud properties were obtained from the Japanese geostationary meteorological satellite Himawari 8/9 and processed by the Australian Government Bureau of Meteorology (2022). We investigated the cloud top heights, cloud top temperatures and cloud types for each flight day of interest. Details can be found in Text S2 of the Supporting Information S1.

2.3. World Wide Lightning Location Network (WWLLN)

Lightning data were obtained via the WWLLN. Details on the WWLLN and further references can be found at the WWLLN website (Holzworth, 2024). The WWLLN provides the geographic coordinates of lightning strokes, which were detected by at least five stations. This means that remote locations could be less well represented compared to locations in proximity to WWLLN stations. We included lightning data from the flight days over the areas of interest.

2.4. EMAC Model

The model simulations of trace gases and meteorological parameters were performed using the ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) model, which is described in detail in Roeckner et al. (2006) and Jöckel et al. (2016). We carried out two simulations including (2.3 Tg N year^{-1}) and excluding lightning NO_x emissions. Further details can be found in Text S3 of the Supporting Information S1. While the main findings of this study are entirely based on in situ observations, we use the model to investigate the atmospheric implications of our results.

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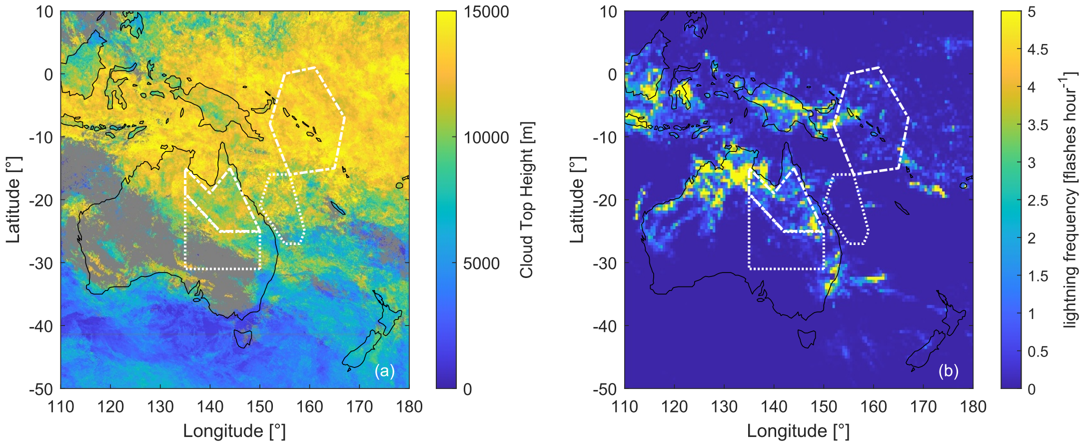

Figure 2. Meteorological conditions in the studied regions on the flight days of interest: (a) Average cloud top heights based on data obtained from the Himawari satellite, (b) Lightning activity over Australia and the Warm Pool. Each grid box represents an 0.5 × 0.5° (in latitude and longitude) area, color-coded according to the lightning flash frequency.

3. Results and Discussion

3.1. Cloud Top Properties

Figure 2a shows the cloud top heights in the studied areas, as an average of the satellite images for each flight day. Gray colors represent areas without cloud measurements on ≥70% of the days. Further cloud top properties, including the cloud top temperatures and detailed information on the cloud types for each flight can be found in Figures S1–S9 of the Supporting Information S1.

High cloud top altitudes mostly ranging between 11 and 16 km in altitude were observed over the Warm Pool during RF10 and RF18. The satellite information on cloud types (as shown in Figures S3c and S7c in Supporting Information S1) demonstrates the dominance of (very) high opaque clouds (cloud type (CT) 8/9) and high semitransparent thick clouds (CT 13) along the flight track. The cloud top heights during RF21 were slightly lower (mostly 7–13 km), corresponding to a higher share of low clouds (CT 6) and high semitransparent moderately thick clouds (CT 12). Taken together, the available cloud top properties are a strong indicator for the presence of deep convective systems over the Warm Pool, which were most pronounced during RF10 and RF18. We observed a higher diversity of cloud top heights and cloud types over Northern Australia throughout the flight days, including both high cloud tops and cloud-free days, resulting in the appearance of mid-level clouds in Figure 2. Similarly, a cloud mix was observed over the Ocean to the East of Australia, which we refer to as the maritime background. We observed mostly cloud-free conditions over Central Australia, referred to as the continental background.

3.2. Lightning

Figure 2 provides an overview of the lightning activity in the studied area as the flash frequency detected by the WWLLN on the days of interest. The continental and maritime backgrounds were mostly lightning-free. This agrees well with the cloud top properties, indicating cloud-free conditions for the continental and a cloud mix for the maritime background. In contrast, lightning activity over Northern Australia was strongly enhanced with values up to 10 flashes per hour. Apart from an area of enhanced lightning between Papua New Guinea and the North of the Solomon islands, lightning over the Warm Pool was scarcer compared to Northern Australia but more abundant compared to the maritime and continental background regions. Of note, the data provided by the WWLLN contain information about the location (latitude and longitude) of the strokes but neither about the altitude nor about the strength of the lightning flash. The temperature and therefore, the amount of NO produced from a lightning flash is highly variable.

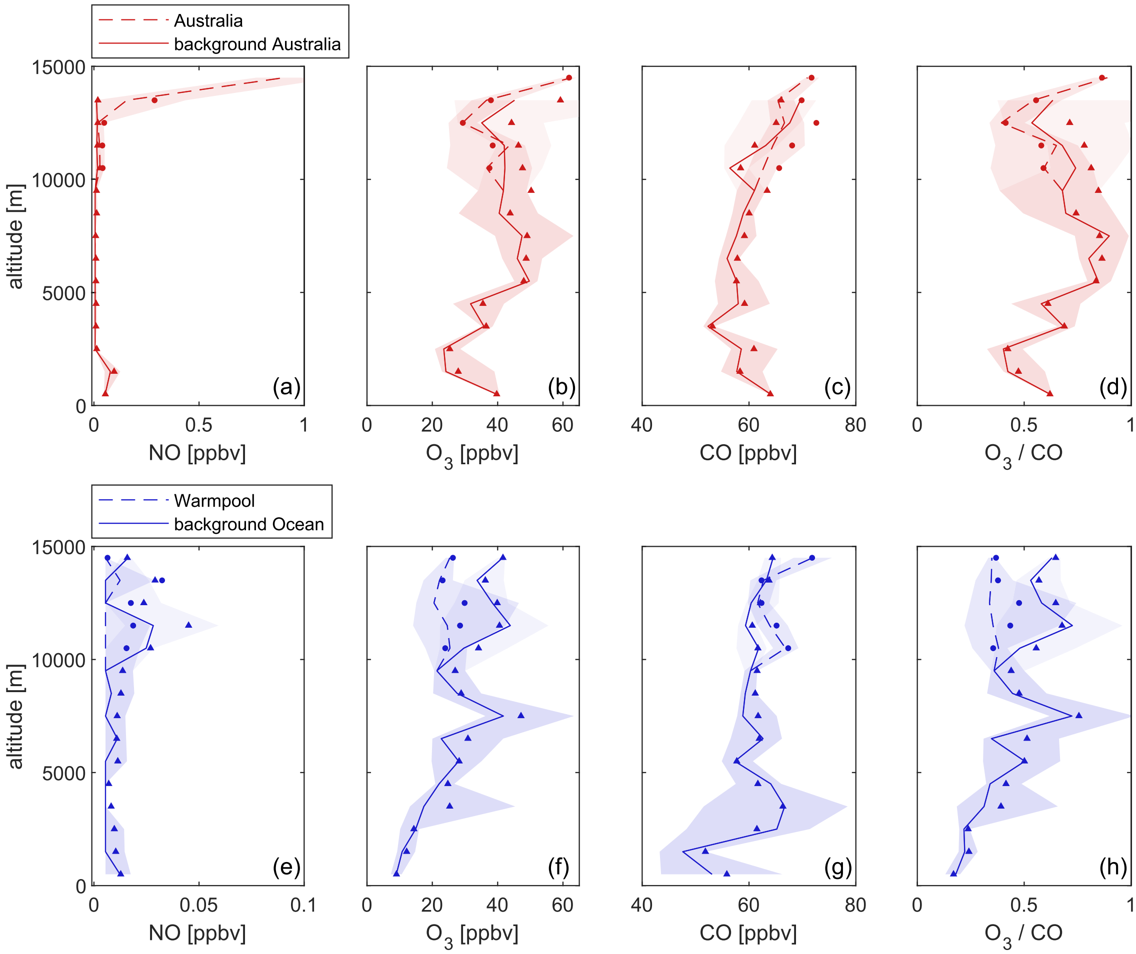

Figure 3. Vertical profiles of trace gases over continental (red) and maritime regions (blue): (a, e) NO, (b, f) O_3, (c, g) CO and (d, h) O_3/CO. Lines and shades represent median values and the 25th/75th percentiles, respectively, at each altitude bin of 1 km. Solid lines show the continental and maritime background and dashed lines present regions with deep convective processes over Australia and the Warm Pool. Circles and triangles show the mean values for the areas of interest and the background, respectively.

3.3. In Situ Observations of Convection

Figure 3 shows the vertical profiles of in situ observations in the areas of interest. We present a joint profile for the continental regions (red, panels (a–d)) and the maritime regions (blue, panels (e–h)) up to an altitude of 10 km, indicated by the solid lines in red and blue, respectively, as we do not expect any differences in the lower and middle troposphere. Above, we show the background with solid lines and the areas of interest over Northern Australia and the Warm Pool with dashed lines. The number of data points in each bin can be found in Tables S1–S3 of the Supporting Information S1.

Figures 3a and 3e show the vertical profiles of NO over the continental and the maritime regions, respectively. For the continental regions, median NO was enhanced at the surface below 2 km. Data points at these altitudes are sparse and the enhancement mostly arises from measurements during RF15. In contrast, NO was close to the detection limit at the surface in the maritime regions, where vessels represent the only source of NO. Particularly in the Warm Pool, a remote maritime area, ship traffic is sparse. In the middle troposphere between 2 and 10 km altitude, median NO was low for both maritime and continental areas, with values up to 15 pptv. At higher altitudes, NO remained low for the maritime and continental background with median values up to 17 and 28 pptv, respectively. For the Warm Pool, NO was mostly below the detection limit between 10 and 15 km altitude. In contrast, NO was strongly enhanced over Northern Australia with median values of 160 pptv between 13 and 14 km and 930 pptv above. These measurements are a clear indicator for the presence of lightning over Northern Australia and the absence of lightning in all other areas, including the background regions and the Warm Pool, despite the presence of deep convective clouds in the latter as shown in Figure 2. Previous studies have shown that lightning is the only relevant source of NO in the upper tropical troposphere (Nussbaumer et al., 2023; Schumann & Huntrieser, 2007). Enhanced NO was mostly arising from flight RF11 with peak values above 2 ppbv. Lightning strokes detected by the WWLLN together with back trajectories, which we show in Figure S10 of the Supporting Information S1 (Nussbaumer et al., 2024b), indicate that NO was not produced locally but transported from a thunderstorm system that occurred several hours before take-off from Cairns.

Figures 3b and 3f show the vertical profiles of O_3. For maritime regions, median values were below 10 ppbv at the surface and increased with rising altitude. For the background, median O_3 increased almost continuously up to around 40 ppbv in the upper troposphere, whereas the profile has an inverse C-shape for the Warm Pool, with maximum values around 40 ppbv between 7 and 8 km and 20–25 ppbv above 10 km. Over Australia, median O_3 was around 40 ppbv at the surface and on average 16 ppbv larger in the lower and middle troposphere compared to the maritime regions - likely due to the higher availability of precursors. In the upper troposphere, median O_3 over Australia was similar to the maritime background around 40 ppbv. Northern Australia showed an enhancement of 60 ppbv median O_3 between 14 and 15 km altitude, indicating O_3 production from lightning NO_x at high altitudes. In contrast, O_3 over the Warm Pool was lower by 40%–50% between 10 and 15 km altitude. O_3 in the upper troposphere can be impacted by various processes. Usually, O_3 increases with altitude due to the decreasing distance to the O_3-rich stratosphere. Additionally, regions with strong lightning activity supply abundant NO as a precursor to photochemical O_3 formation. The lifetime of ozone further increases with altitude, mostly due to the decreasing availability of water vapor. Convective updraft from the boundary layer can provide the opposite effect, transporting O_3-poor air to high altitudes. Long-range transport of air can additionally influence O_3 levels in the upper troposphere in either direction, as O_3 has a lifetime of several weeks. In combination with the cloud top altitudes in Figure 2a indicating convective systems and the vertical profiles of NO providing insights into lightning activity, we suggest that the Warm Pool is characterized by non-electrified, strong convection. Updraft from the O_3-poor maritime boundary layer leads to low mixing ratios of O_3 in the upper troposphere, which is not replenished by photochemical production as NO is absent. In contrast, Northern Australia is characterized by strong lightning activity that provides large amounts of upper tropospheric NO, which in turn affect photochemical O_3 production. The oceanic and continental background regions neither show lightning nor convective activity and upper tropospheric O_3 levels are enhanced due to the proximity to the stratosphere or long-range tropospheric transport.

Figures 3c and 3g show the vertical profiles of CO. The continental profile shows a C-shape with median values of 65 ppbv at the surface, 55–60 ppbv at mid-tropospheric altitudes and 65–70 ppbv in the upper troposphere. No significant differences can be observed between Northern Australia and the continental background at high altitudes. The C-shape indicates convective transport but does not provide insights into the time scale as CO has a lifetime of several weeks to months in the troposphere. At the surface over the ocean, CO was lower with 50 ppbv and peaked between 2 and 5 km with median values around 65 ppbv. For the maritime background, CO was roughly constant with around 60 ppbv at higher altitudes. In contrast, CO was enhanced over the Warm Pool between 10 and 11 km and between 14 and 15 km with 67 and 72 ppbv, respectively. This enhancement indicates upward transport from lower altitudes.

The ratio of O_3 and CO can additionally provide insights into convective processes, which we show in Figures 3d and 3h. Close to the surface, O_3/CO is usually lowest due to low O_3 and high CO and increases with altitude and decreasing distance to the stratosphere, where O_3 peaks and CO reaches a minimum. The shapes are similar to the vertical profiles of O_3, but with more pronounced differences between the Warm Pool/Northern Australia and the respective backgrounds in the upper troposphere. Strong convective updraft over the Warm Pool and Northern Australia is indicated by an inverted C-shape profile with similar O_3/CO values at high altitudes compared to the surface. In contrast, enhanced values over the background regions show the absence of convection. Large values at altitudes of 14 km over Northern Australia (S-shape) additionally indicate the impact of strong lightning on photochemically produced O_3.

An additional indicator for continental convection is the short-lived VOC isoprene, which we show in Figure S11 of the Supporting Information S1. Isoprene mixing ratios were enhanced in the continental boundary layer as well as in the upper troposphere over Northern Australia, indicating recent deep convection. Observations were mostly below the detection limit throughout the maritime tropospheric column due to the absence of strong sources.

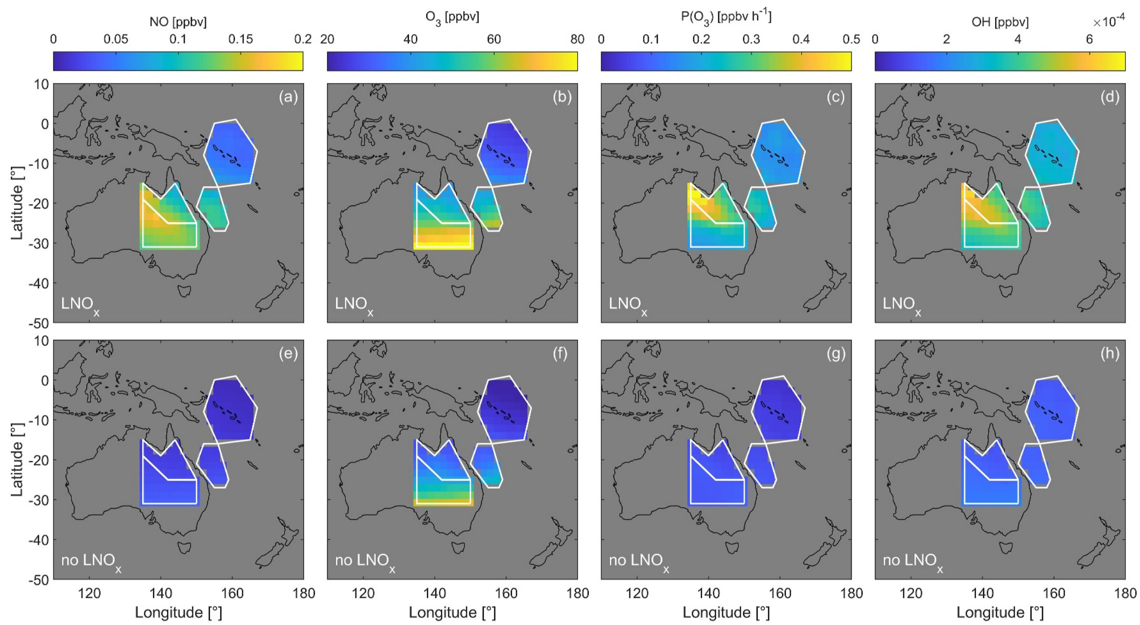

Figure 4. Averaged, modeled NO, O_3, $P(O_3)$ and OH using EMAC in the areas of interest at 180 hPa for daytime values between 01/16 and 02/29 including (a–d) and excluding lightning NO_x (e–h).

3.4. Atmospheric Implications

Figure 4 shows modeled NO, O_3 and OH radicals as well as calculated O_3 production rates ($P(O_3)$) as an average over daytime values ($j(NO_2) > 0$) between 01/16 and 02/29 at 180 hPa, which corresponds to the altitudes at which convection and lightning are observed. Detailed numbers can be found in Tables S4–S8 of the Supporting Information S1. We calculate $P(O_3)$ via the reaction of NO with HO_2 and CH_3O_2. Details are provided in Text S4 of the Supporting Information S1.

Panels (a, e) show NO mixing ratios with (baseline scenario) and without lightning (no LNO_x scenario), respectively. The baseline scenario shows the lowest mixing ratios of 45 pptv (median) over the Warm Pool, followed by 109 pptv over the maritime background and 124 and 137 pptv over Northern and Central Australia, respectively. The modeled values over the Warm Pool and the maritime and continental backgrounds are by around one order of magnitude larger than the in situ observations. In contrast, the measured and modeled mixing ratios over Northern Australia show a much closer agreement, with 266 and 124 pptv, respectively. Comparing the observations with the no-LNO_x scenario, we find a significantly improved agreement over the Warm Pool and the background areas, whereby NO over Northern Australia is underestimated by a factor of 15. These findings indicate that while the model in its actual configuration is able to represent continental lightning over Northern Australia well, it overestimates maritime lightning in this region. As the model predicts much higher NO from lightning over the tropical Pacific, other remote maritime regions could also be overestimated. Central Australia is a desert and cloud top properties, the WWLLN and our in situ observations are strongly suggestive of lightning-free conditions. We hypothesize that the enhanced mixing ratios of NO predicted by the model over the continental background could be the result of transported (overestimated) NO from maritime regions. In contrast, regions like Northern Australia with strong convection and lightning are represented well.

These deviations in NO impact photochemical processes in remote maritime regions and areas to which these air masses are transported. Panels (b, f) show modeled O_3 over the areas of interest. In the baseline scenario, O_3 is overestimated by 25%–50% over the Warm Pool and the background regions compared to the observations, but the values show close agreement in the no-LNO_x scenario. Over Northern Australia, the agreement of O_3 in the baseline scenario is much higher compared to the maritime regions. The model overestimates median O_3 by 18% and underestimates it by 13% in the no-LNO_x scenario. This indicates the impact of both local photochemical production and transport from adjacent areas.

These findings suggest that the baseline scenario represents the actual photochemical state over Northern Australia and the no-LNO$_x$ scenario captures the photochemical state over the Warm Pool and the background regions. Consequently, O_3 production is likely overestimated by the model by a factor of 3–4 and OH radicals by a factor of 2–3 over these remote regions, where our understanding currently still relies on models due to the scarcity of observations. Oxidation by OH radicals represents a major pathway of removing pollutants and greenhouse gases from the troposphere. Significant impacts for estimating the radiative forcing from CH_4 and O_3 could result if our observations are not limited to the studied region, but also apply to other remote maritime areas. The underestimation of OH can further lead to underestimated amounts of halogen species reaching the stratosphere with major impacts on stratospheric O_3 loss (Rex et al., 2014). Further research is required to quantify these impacts.

4. Conclusions

In this study, we presented in situ observations of NO, O_3 and CO from the aircraft campaign CAFE Pacific. In combination with satellite-inferred cloud top properties and lightning observations, we identified deep convection over Northern Australia and the Indo-Pacific Warm Pool. We found that this strong updraft is accompanied by lightning over the continent but is non-electrified over the ocean.

To our knowledge, this is the first study to present evidence of the occurrence of deep convection without lightning over the Warm Pool, an area which impacts chemistry and climate around the entire globe. These observations provide an explanation for the persistently low O_3 values observed in this region up to high altitudes. Deep convection can lift O_3-poor air from the boundary layer to the upper troposphere and the absence of NO prevents the photochemical formation of O_3 locally. By the help of model simulations we show that these conditions lead to very low ozone production rates and OH availability. This OH minimum over the Warm Pool was previously reported by Rex et al. (2014) and we now show that it can be induced by non-electrified, deep convection over maritime regions. Consequently, the OH minimum is not only a result of low primary production from O_3 but also due to minimal secondary production in the absence of NO$_x$. Further research is necessary to investigate if these observations can be extrapolated to other remote maritime regions.

We provide evidence in support of the previously advanced hypothesis on the origin of low O_3 in the Warm Pool. While various studies have investigated the differences between continental and maritime lightning including the convective available potential energy, precipitation, updraft velocities and the impact of aerosols (Lal et al., 2014; Lucas et al., 1994; Z. Pan et al., 2022; Petersen et al., 2005; Romps et al., 2018; Shi et al., 2015; Wang et al., 2018; Wilbourn et al., 2024; Williams & Stanfill, 2002), we still do not fully understand the reason for this imbalance. Future studies should investigate this open question and the implications for chemistry and climate in a changing world.

Data Availability Statement

The supporting data for this study can be found at Nussbaumer et al. (2024a).

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