ACPD – Referee #1 Comments

RC1: 'Comment on egusphere-2024-3832', Anonymous Referee #1, 13 Jan 2025 reply

The manuscript "Aircraft Observations of Continental Pollution In the Equatorial Lower Stratosphere over the Tropical Western Pacific During Boreal Winter" by Pittmann et al. presents observations from the ATTREX campaign 2014 over the tropical western Pacific (TWP) region obtained with the Global Hawk.

The paper describes the dataset and comes to the conclusion that the UTLS composition in the TWP is dominated during the campaign time by continental pollution.

In my point of view the paper is in general well written, but some major and minor revisions should be conducted for a final publication in ACP.

1) The pollution is termed "continental pollution". Even though the analysed pollution is of continental origin, the authors state that this is mostly from biomass burning pollution. Consequently, a re-naming of the manuscript should be considered, as the influence of anthropogenic pollution in the TWP UTLS is minor.
We appreciate the suggestion. We replaced the term "Continental Pollution" with "Biomass Burning Pollutants" in the title.

2) The authors make both in the abstract as well as in the conclusions statements about aerosol particles. However, the measurement data does not obtain any data point on aerosol particles. Even though aerosol particles are potentially included in the emission sources, their fate and transformation in the convective ascent as well as microphysical processing and / or sedimentation along the relatively long transit times is uncertain. I would expect substantial scavenging of primary emitted particles in the ascent, either by precipitation scavenging or by ice nucleation such that only a minor fraction of primary aerosol particles will be transported into the UTLS. Whether new particle formation also takes place in the TWP or in the transit to the measurement region is not yet certain. Therefore, the TWP cannot be named an ascent region of aerosols into the LS. And as this is not shown in this manuscript (and it is not its intention), these statements should be either weakened and discussed or should be removed from the manuscript.

Similarly, the role of VSLS are prominently mentioned in the beginning, but the analysis showed that they do not play a significant role in this context, i.e., as lofted species with a high ozone destruction potential.

Aerosols and VSLS are important players in the recovery of the stratospheric O_3 layer, so they provide motivation for the analysis we conducted.
In terms of aerosols, it is indeed impossible to determine if the pollution plumes we sampled carried the expected emitted aerosols along the way, especially over multiple

days and long distances, given the absence of measurements. The only data we have are of Secondary Organic Aerosol (SOA) precursors, such as Benzene. Elevated values of this trace gas suggest an increased potential for SOA formation, which is all we can deduce. Our study confirms delivery of SOA precursors to the TWP and can only suggest that the presence of Benzene could impact stratospheric O_3 via future SOA formation, which could favor novel conditions for stratospheric O_3 destruction according to recent hypothesis (e.g., Solomon et al, 2023).
In terms of VSLS, our study shows that these compounds were not transported in these pollution plumes despite their continental origin. We find that this is an important statement to keep. Other aircraft studies (different time of year, and similar area of sampling over the tropical western Pacific) showed the opposite, that VSLS (Chlorine, in particular) were indeed transported from urban regions in SE Asia to the TTL over the TWP (e.g., Treadaway et al, 2022).

3) The back-trajectories are calculated for 40 days. This is a very long time period for air mass transport, without the consideration of diabatic processes (cloud processing), turbulence and small-scale mixing, such that the longest trajectories only represent a very crude approximation for the air mass history. Especially trajectories who travel over the Indian Ocean to Africa and from there back to the TWP region must be treated carefully. These limitations should be discussed in the manuscript.

The occurrence of convection associated with the trajectories also has to be discussed in a little more detail. As the trajectories are calculated from ERA5 dynamics, there is no guarantee that individual convective cells and elements on the order of <30 km are represented properly in the meteorological re-analysis data. Of course, the satellite data represent reality, but it should be analysed / discussed whether the convection in the re-analysis is co-located with the observed convective activity or whether this activity is sufficiently wide-spread such that air parcels in the convective region have a high probability to be lofted.

The analysis of air parcels in convection also reveals that it is not guaranteed that an air parcel actually has seen the convection, only because it has been in its vicinity: it could also happen that air parcels simply stream around the anvil of the convective towers and are not substantially modified by lofted near surface air.

These are all very good points to consider when using trajectory analysis. In Sections 2.4 and 3.4, we point out the uncertainties associated with winds and provide references that caution against the use of single trajectories. A more robust approach to interpreting trajectory calculations is to consider trajectory ensembles, instead, which provide a more robust statistical perspective. That is the approach we felt was most suitable in our analysis. With regards to convection, our calculations are anchored to satellite observations of precipitating clouds, which require multiple considerations (see Pfister et al., 2022)

4) The selected / presented compounds often have quite enhanced life times, especially CH4 and CO2, such that convective signals of events more than a few days ago can hardly be distinguished from background conditions. Consequently, the profiles from Fig.2 for methane and carbon dioxide provide a measure for the substantial background variability. For species with shorter lifetime, like CO, the variability is smaller, even though the regimes of convectively and non-convectively influenced air masses cannot be easily distinguished (in Fig.2). In Fig.3 some of the flights show a distinct enhanced CO in the UTLS, which could indicate convective outflow; however, after travel times of more than a week, it is unlikely, that these structures remain that clearly visible, indicating more local or regional convection and further lofting, e.g., by convectively induced gravity waves and their mixing processes.

On the other hand, the shorter lived compounds (C3H8, C2H2) hardly show enhancements in the profiles. Given their shorter chemical lifetime, the air masses must have been lofted several days prior to the observations, if only in the Northern observations any signal can be identified. This rules out transport from Africa, but indicates more regional continental pollution. Even the CO signal in the Southern observations is quite low, and hardly shows UT enhancements. This does not fit well with the analysis from Fig. 14 that the corresponding trajectories could have seen convection only two weeks ago. A separation of continental convection or biomass burning in the vicinity of convection or anthropogenic activity near the convection might elucidate the origin or missing signals a bit better. This is most obvious for the Southern observations, but would also hold true for the rest of the data.

Our analysis focuses on data in the equatorial TTL. As mentioned in the Introduction, air masses at these locations are convective in origin, some more recent than others. Where that convection occurs is reflected in the variability observed in Fig. 2. The lifetimes of the trace gases shown in Fig. 2 (CO2, CH_4, CO, with O_3 added now) are indeed much longer than convective transport timescales. The magnitude of the variability observed in all tracers is primarily driven by hemispheric gradients (varying strengths of sources and sinks) as indicated by latitude (variable used for color-coding) and discussed in Section 3.1. Based on trace gas measurements of elevated pollutants (e.g., CO and various hydrocarbons), we know that those air masses originated over continents. Local convection (in our case, marine convection) would be secondary after those air masses traveled from the continental source regions and entrained into the marine convective systems. This sequence of transport processes would be difficult to pin point, however.

In terms of strength of signal from pollution over time, it is true that the structures would evolve – likely decrease in magnitude as a result of mixing. Previous studies based on aircraft observations provided evidence that pollution plumes can indeed remain fairly distinct against the background after nearly 2 weeks of transport (e.g., Jost et al., 2004).

This prior evidence makes air masses with elevated CO originating from Africa quite feasible, given the similar time scales.

5) The MLS data does not really sheds light onto the observations, as the signal cannot be properly distinguished from the background. Therefore, the manuscript could be shortened here, and the figures 12 and 13 could be moved to an appendix or supplement.
We think it is important to highlight the capabilities of each platform and what can and cannot be observed. Our analysis illustrates this point. For instance, if you cannot see the signal of pollution due to lower spatial resolution in your measurement (e.g., satellite measurement), that does not mean that the pollution is not present. In order to be more succinct in this section of the manuscript, we consolidated the presentation of the results to a single figure and moved additional figures to the Supplement section.

6) The pollution is mostly determined in the cloud-free air. As the authors state, that the pollution is mostly transported for several days, and that the trajectories are slowly lofted by radiative cooling, this is not very surprising. On the other hand this is a bit contradictionary to the finding of Fig. 11, which states that the pollution is usually associated with warmer air masses. As the typical temperature profile does not increase quickly in the low latitudes above the thermal tropopause, this needs an explanation.

Encounters of the pollution plumes were indeed mostly in cloud-free air. The statement regarding warmer temperatures relates only to air masses with 3.5 – 5 ppmv H_2O, which is a subset of the data, as described in Section 3.2. We revised the text to clarify this point. We also added a new panel (now Fig. 10) that illustrates warming in the wetter range of H_2O and no warming in the drier range of H_2O.

7) I am missing an explanation, why the pollution is not found in cloudy or moist regions. Is this because the polluted air is at higher altitude than the clouds, i.e., because of the above mentioned lofting?

We added a new figure to the Supplement material illustrating how pollution plumes were located above the clouds, the maximum level of convective detrainment. We also included a statement in Section 3.2 highlighting a potential link between pollution and cloud formation and the need for a more comprehensive data set (e.g., physical properties and chemical composition of aerosols, which are not available in our data set) to examine the radiative characteristics of these air masses.

8) What information can be deduced from Fig. 9, which is not also included in Fig. 10? Can you (re)move Fig. 9 to the supplement, in order to slightly shorten the manuscript and reformulate the statements from the corresponding paragraph? Basically, the same

conclusions, i.e., that the highest pollution levels are found in non-saturated regions, can be drawn from Fig. 10, which has better representation via the color coded visibility.
We appreciate this suggestion and made the proposed changes.

However, the color scale of Fig.10 (using rainbow colours) could be modified to be more perceptually uniform and color-blind / black-white friendly.
The color scale was modified to use the "turbo" instead of the "jet" colormap.

9) Please discuss the variability in the vertical motion of the individual trajectories: especially trajectories from Africa travel more than 50 hPa upwards and downwards on timescales of a day or two (Fig. 14c, around day 13 to 21). What processes would you associate these relatively rapid air motions including downwelling, before the radiatively driven ascent prior to the measurements?

The rapid changes in short time scales observed in trajectories linked to Africa as a source region is a good question. That was the case for a few trajectories, but not for the bulk. As mentioned earlier, encouraged by earlier comments and suggestions, we decided to take a more statistical approach to the analysis of these calculations and revised the figure, now Fig. 12.

References:

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Reply

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