Response to reviewers comments for the paper:

“Air Mass Transport to the Tropical West Pacific Troposphere inferred from Ozone and Relative Humidity Balloon Observations above Palau”

First of all, we would like to thank the two reviewers for taking the time to work through our manuscript and make very valuable suggestions. We will respond to these suggestions below and mark the responses in blue. Required changes in the manuscript will be implemented and marked as track changes to be easily accessible. Line references in blue are with regard to the revised manuscript.

Reviewer report by reviewer 1:

Mueller et al examine a four year ozonesonde record from Palau to determine the distribution of air parcels based on their deviation from background values of ozone and relative humidity. They look at the seasonal variability and, using backtrajectories, the origin of these air parcels. They find that background air (O3-RH+) dominates throughout much of the year, but that, particularly in FMA, O3+RH- air parcels make up a substantial fraction of the observed air parcels. They find that the O3 likely originate from biomass burning or other anthropogenic emissions from southeast Asia, while the low RH likely results from large-scale descent in the tropics. Potential vorticity analysis suggested no impact from in-mixing of mid-latitude stratospheric air.

This paper is well-written and presents a convincing conclusions with a thorough analysis of their dataset, and it adds substantively to the debate on the origin of these air masses in the remote tropics. While I recommend publishing the paper as is, I do have two very minor comments.

Line 68: In addition to Pan et al, since you mention CAST, you should also cite Harris et al, 2017.

Harris, N. R. P., and Coauthors, 2017: Coordinated Airborne Studies in the Tropics (CAST) Bull. Amer. Meteor. Soc., **98**, 145–162, https://doi.org/10.1175/BAMS-D-14-00290.1.

Thanks for pointing this out! Done.

Section 3.2: Because convection would "reset" the ozone and water vapor abundance in air parcels to be more reflective of the remote Pacific background, what impacts will this have on your trajectory analysis since you are not explicitly including convection into your analysis? Could this adversely impact your analysis? (Based on your figures this does not seem to be the case but it might be worth a sentence or 2 of discussion explaining why this doesn't matter.)

There are mainly 2 ways that our assumption of conservation of O3 content within an air mass for 5 days fails: Some of the low O3 air masses we observe above Palau could have seen higher O3 in the past (week), but were “reset” during transport by convection. High O3 air masses could have picked up high O3 later during the 5 days of transport. A study by Andersen et al. finds air parcel ages of around 10 days in the TWP in winter 2014, when stopping their trajectories at the point of last precipitating convection based on satellite observations of cloud top height and precipitation. From this point of view, neglecting convection for our 5 days back trajectories seems a reasonable choice.

Generally, we would not observe high O3 levels in the first place, if there was a significant amount of convective events canceling the air parcel history. In addition, potential sources of O3 or its precursors over the Pacific are rather rare compared to those on the continent. So our overall argument, linking O3 rich air masses to an origin on the Asian continent 5 days before arrival in Palau, is not diminished by calculating our trajectories without a convective scheme.

We added some discussion in the text at lines 247ff.

Review report by reviewer 2:

Review of “Air Mass Transport to the Tropical West Pacific Troposphere inferred from Ozone and Relative Humidity Balloon Observations above Palau” by Katrin Müller and coauthors.

General comments

This study examined the transport processes in the Tropical Western Pacific (TWP) using a four-year record of ozonesonde data and trajectory analysis at Palau Atmospheric Observatory (PAO). The research methods are likely well-designed based on a comprehensive literature review, and research findings are clear and reasonably supported by a noble set of observations. Particularly, the tracer-tracer analysis between O3 and H2O (RH) and relevant back trajectory analysis provide meaningful insight on the air mass transport over TWP, a new finding in this field. The only concern is the readability of the manuscript. It is generally well-written, but some sentences are excessively technical and verbose. If the authors could put some effort into clarifying the sentences further, it would greatly benefit the readers of the ACP. I recommend a minor revision before the publication of the manuscript. Detailed comments and suggestions are provided below.

Comments

1. Definition of background is confusing.

In the air mass definition (section 3.1.3), the "background profiles" are defined as 20 and 83.3 percentile for O3 and RH, respectively. However, -5/+15 ppb (O3) and -20/+5 % (RH) ranges are also referred to as background again (in section 4.1.3). Although they are related, it is very confusing. Explicitly defining the latter as "background category" or "background group?" could be helpful, or there could be a better choice of word… Lines 298-303 are related.

Thanks for pointing this out. We carefully checked the manuscript for the consistency of our background terminology. The difficulties arise in the difference between our qualitative, schematic view on the background and the actual quantification, for which we "switch" into the "anomaly space". Now, in Sect. 3.1.3, we introduce the suggestion of the reviewer: "background category". This led to slight rephrasing in Sect. 4.1.3, including a warning not to mix up background profiles with background categories (see also comments below). Please find all the adjustments in the track-changes document.

2. Categorization

Fig. 3 shows four categories, but the actual analysis (Fig. 7) uses nine categories. I understand the authors' intention to provide an easy example for the categorization, but it is actually very confusing…

Line 166: "we propose five qualitative categories…" Is it four?

We understand the reviewer's confusion here and had already tried to help the reader to follow us by speaking of the "qualitative" and "quantitative" scheme. We now improved Fig. 3 to highlight the two different pathways possible for O3+RH-, thus constituting two different air masses, and indeed changed our wording to "four qualitative categories", but "five different pathways" throughout the text. We further tried to improve the text at this particular point (now still lines 166) as follows:

"In conclusion, we propose four qualitative categories of air masses and five different pathways identifiable by our tracers, as illustrated in Fig. 3. Ozone-depleted air masses (O3-, turquoise colors) are of local or Pacific, convective origin and ozone-rich air masses (O3+, brown colors) originate from non-local pollution or the stratosphere. High RH results from a dominant convective uplift of air masses (RH+, darker hues), while low RH indicates a stratospheric origin or dehydration of previously lifted air masses during transport due to clear sky cooling and descent (RH-, lighter hues). O3+RH- air is characteristic for two different pathways (solid and dotted light brown lines in Fig. 3), thus representing two different air masses."

3. Physical interpretation

The “FMA” patterns in Fig. 9 and Fig. 10 are noble findings of this paper. The trajectory pattern seems well related to the seasonal “Gill-type pattern” (Gill 1980; Dima et al. 2005 for reality), and the source region is well matched with agricultural fire in South Asia (Yadav et al. 2017, etc). Some in-depth discussion may be beneficial.

Gill, A. E. (1980). Some simple solutions for heat-induced tropical circulation. *Quarterly Journal of the Royal Meteorological Society*, *106*(449), 447–462. https://doi.org/10.1002/qj.49710644905

Dima, I. M., Wallace, J. M., & Kraucunas, I. (2005). Tropical Zonal Momentum Balance in the NCEP Reanalyses. *J. Atmos. Sci.*, *62*(7), 2499–2513. https://doi.org/10.1175/JAS3486.1

Yadav, I.C., Devi, N.L., Li, J., Syed, J.H., Zhang, G. and Watanabe, H. (2017). Biomass burning in Indo-China peninsula and its impacts on regional air quality and global climate change-a review. Environmental Pollution, 227, pp.414-427.

Thank you for pointing us in the direction of these publications and suggesting more discussion of the FMA pattern. We adapted and extended our text in this regard in Section 5.2, in lines 502ff

As a follow-up on discussion of the source region and the third reference suggestion of the reviewer, we included this reference in the discussion when mentioning biomass burning (in line 523) and also again Anderson et al. (2016) and a paper by Ogino et al. (2022), who target a high-ozone layer over Hanoi and subsequent transport over the Pacific. We did not extend the discussion any further here though.

Ogino, S.-Y., Miyazaki, K., Fujiwara, M., Nodzu, M. I., Shiotani, M., Hasebe, F., et al. (2022). Cause of a lower-tropospheric high-ozone layer in spring over Hanoi. *Journal of Geophysical Research: Atmospheres*, 127, e2021JD035727. https://doi.org/10.1029/2021JD035727

These are suggestions

Line 1-8: This background is somehow redundant with introduction, and authors may want to make it concise.

We believe the readers benefit from the introductory sentences in the abstract. However, we slightly modified the first sentences and rearranged parts of the whole abstract to be more concise.

Line 29: Recommend “Air masses entering the stratosphere largely originate…”

Done.

Line 34: Recommend “tropospheric ozone (O3) concentration sheds light…”

Done.

Line 35: Recommend “both convective (…) and long-range transport processes in this region”

Done.

Line 37-40: The sentence is too long. “The hydroxyl radical…the local troposphere” part can be removed.

We still like to emphasize the importance of the chemical aspect and are happy to divide the idea into two sentences.

Line 43: “humid, marine, and pollutant-free environment”

Done.

Line 49-50: “3.4% per day.” Is it an additional contribution? 3.4% loss rate may not make a lifetime “around 5 days”

Crawford et al. (1997) estimated the net effect of all photochemical processes on O3 for the tropospheric column. Liu et al. 1983 calculate a daytime average rate of the photochemical destruction of ozone corresponding to 5 days for near surface ozone.

We adapted the sentences as follows for clarification:

“Observations in the TWP found low NOx concentrations inhibiting O3 production and thus facilitating an O3 loss rate via the above reactions of 3.4 % per day for the tropospheric column (Crawford et al. 1997). For boundary layer O3 in the equatorial Pacific the efficiency of this loss mechanism results in a lifetime of around 5 days (e.g., Liu1983, Kley1997).”

Line 50-51: “Deep convective outflow and overturning processes lifting the clean boundary layer air to the Tropical Tropopause Layer (TTL)…”

The sentence now reads:

“Deep convective outflow and overturning processes lift the clean boundary layer air to the Tropical Tropopause Layer (TTL). In conjunction with a lack of in situ net O3 production this creates a well-mixed, humid tropospheric profile with a uniform vertical O3 distribution (e.g., Pan et al., 2015).”

Line 55: “enhanced O3 from the lower stratosphere against…”

In this sentence we introduce different studies about the general phenomenon of dry enhanced O3 layer, which not all attribute these layers to a stratospheric origin. Therefore we will leave the sentence as is.

Line 109-121: May need a little more detail. For example, is “diabatic heating rate” computed or provided? Are the spatial and temporal resolution enough for resolving convective transport?

The diabatic heating rates and 3D met fields are parameters from ERA5. We actually think, the sentence is not ambiguous about the data input for the model from the reanalysis data. However, we added a few sentences to address convection within the dataset.

Line 114: two tracers => O3 and RH?

Yes, for clarity, we added these in the sentence.

Line 122, 125, 139: Figure numbers are cited randomly. I am not sure if it is ok... (normally, they are cited in increasing order). Fig. 2 and Fig. 3 can be switched, and “section 4.1.3” can be used instead of “Fig. 7”.

We took the suggestion to use a reference to the section instead of the figure and used it for the “pre-mature” mention of Fig. 3 as well (“section 3.1.1”). This way, the order of the figures can remain as is, which we believe is beneficial for the train of thought in this elaborate Methods section.

Line 181: “layers from balloon and aircraft profiles, different…”

Done.

Line 183: “for various atmospheric constituents”?

Done.

Line 184: “In approach…” => “Using a spike detection approach, they produced extensive statistics for the frequency of anomalous layers in the whole tropical Pacific region for two different seasons”

Done. Thank you for clearing up this complicated sentence!

Line 187: “Besides…” => “Despite the statistical uncertainty, Stoller et al. (1999) recognized the importance of anomalous layers in tropical Pacific profiles and emphasized their role in atmospheric chemistry modeling due to their high occurrence”?

Thank you for making this sentence more concise! We used your suggestion and just ended slightly different with “...due to their frequent occurrence.”

Line 193: “, the study found that approximately 50% of profiles are related to the layer with differing seasonal variations”?

Here, we actually prefer the original version we used, saying “the study found respective layers in approximately 50 % of profiles per year at each station with differing seasonal variations.” and only changed it slightly to the following: “per year and station”.

Line 204-210: Please revise this part. It is important part, but barely understandable…

We rewrote this paragraph (now lines 202ff) and parts of the following paragraphs (please see track changes in the manuscript) to improve readability as follows:

“For a quantitative definition of air masses above Palau, we first determined background profiles in the free troposphere from the PAO O3 and RH time series. These profiles represent humid, ozone-poor, local air masses that are controlled by convective influence and not by long-range transport. This was performed on a statistical and monthly basis and aimed to identify profile shapes similar to the signature profiles in Fig. 2a. For the monthly O3 VMR background profiles we chose the 20th quantile (Q20) profiles. Q20 guarantees sufficiently low O3 values to avoid a “belly” shape of the profiles, typical for tropical average O3 profiles. For the monthly RH background profiles we chose the 83.3th quantile (Q83) profiles. Q83 is the upper boundary of the central 66.6% range, guaranteeing high humidity. Fig.\ \ref{fig:04} shows an example of these background profiles for March. The monthly quantile profiles have been vertically smoothed using first a 1-km-binning and then exponentially weighted moving averages with altitude (see also Figs. A3 and A4). Fig. 4 includes the median and central 66.6% ranges for both tracers separately and is calculated from 16 individual profiles (see Müller, 2020, for more details).

Our method roughly follows the approach of Hayashi et al. 2008. However, Hayashi et al. …”

Line 210: exponentially weighted with height?

Yes, we clarified this further in the text.

Line 211: “While Hayashi et al. (2008) defined the enhanced O3 layer using the 83.3th quantile, we used a less conservative approach in regard to O3 because the Palau background is characterized as a uniform… “

Thanks, this sentence indeed could use some rephrasing. We have done more changes in this whole section and changed the particular sentence to the following: ”However, Hayashi et al. defined air masses above the 83.3th quantile as O3 enhanced layers. With the Q20 limit for O3 we used a less conservative approach, because the Palau background atmosphere is characterized as a uniform, well-mixed low O3 profile, caused by uplift of ozone-poor boundary layer air in active convection.

Line 228: “we did not assess the vertical structure of the anomalous layer…”

Done.

Linę 263: “strong TTL cycle”. The TTL ozone signal in May (column b/w M-J) is too deep to be simply explained by the annual cycle of the Brewer-Dobson circulation. The STE process cannot be excluded in the 12-15 km level. Although it doesn’t affect the main finding, the authors may want to add discussion or adjust the tone of the argument."

At this point, we just stated the results, i.e. our observations, no interpretation. We briefly discussed STE for the MJJ season in lines 439 ff (now lines 444 ff) by referring to the study of Stauffer et al (2018). However, we acknowledge the reviewer’s remark here and extended our discussion with regard to the TTL cycle a little in lines 424 ff and 434f.

Line 268: reoccurring => recurring is a bit better choice.

Done.

Line 269: synoptical => synoptic

Done.

Line 275: winter season => boreal winter (or FMA?)

We clarified this, by adding “i.e.” into the half sentence, now reading: “... winter seasons, i.e. in NDJ and most pronounced in FMA”.

Line 283-287: difficult to understand, please revise…

We rewrote this passage for clarification as follows:

“MJJ and NDJ can be considered as intermediate seasons with respect to the mid-tropospheric cycle, i.e. O3 values here are in between the minimum and maximum. The NDJ profile resembles the annual mean profile up to about 14 km, while the MJJ profile diverges from the annual mean above 10 km towards higher values. The resulting tilted line shape during MJJ, compared to the straight line profile during ASO, implies a more gradual increase of O3 in the UTLS (Müller et al. 2023) and a lack of a pronounced O3 minimum in the UT. In MJJ, the tropopause is at its lowest altitude of the year, thus is the occurrence of high levels of stratospheric O3 VMR.”

Line 301: “as a separate group despite the low population in Fig. 7a?”

We changed the phrasing to “stands out as a low, but separate population in Fig. 7a”

Line 313: “As expected from the previous analysis, ΔO3+ ΔRH- are almost absent in ASO, emphasizing that the season represents a dominant background in the free troposphere.”?

We changed this sentence and noticed, that the following sentence is rather unclear, so we changed that one as follows:

”In contrast to the seasonal mean FMA O3 profile with elevated O3 levels in the mid-troposphere (Fig. 6), the FMA anomalies distribution (Fig. 8b) reveals a dominance of background category air masses over ΔO3+ ΔRH- air masses within the free troposphere.”

Line 310 “A footprint of air mass transport to Palau is analyzed using 10-day backward trajectories for the study period (2016-2019) sorted by…”

Thank you for this phrasing!

Line 322: a day/profile => a profile?

Done.

Additional changes

Apart from the reviewer's comments and clearing a view typos, we made some additional changes as follows:

- One of the co-authors, Ingo Woltmann, withdrew his co-authorship and is now mentioned in the acknowledgements
- We updated the figures 1, 5, 7, 8, 10, 11, A5 and A8 with regard to font sizes as requested by the editor in the first revision after submission. We also made slight modifications in the captions to improve style.
- To improve overall readability of the paper, we rephrased and rearranged parts of the abstract and the discussion. We believe this is still in compliance with the original manuscript, and hope the editor agrees with us.
- Notably, we added some discussion with regard to the RH bimodality in lines 468 ff, which was pointed out to us by a friendly reviewer, also adding new citations on the matter:

  Zhang, C., Mapes, B.E. and Soden, B.J. (2003), Bimodality in tropical water vapour. Q.J.R. Meteorol. Soc., 129: 2847-2866. https://doi.org/10.1256/qj.02.166

  Sherwood, S. C., E. R. Kursinski, and W. G. Read, 2006: A Distribution Law for Free-Tropospheric Relative Humidity. *J. Climate*, **19**, 6267–6277, https://doi.org/10.1175/JCLI3978.1.

  Ruzmaikin, A., H. H. Aumann, and E. M. Manning, 2014: Relative Humidity in the Troposphere with AIRS. *J. Atmos. Sci.*, **71**, 2516–2533, https://doi.org/10.1175/JAS-D-13-0363.1.