# Response to reviewers comments for the paper: "Measurement Report: The Palau Atmospheric Observatory and its Ozonesonde Record - Continuous Monitoring of Tropospheric Composition and Dynamics in the Tropical West Pacific"

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 to be easily accessible.

# Review report by reviewer 1:

Measurement Report: The Palau Atmospheric Observatory and its Ozonesonde Record - Continuous Monitoring of Tropospheric Composition and Dynamics in the Tropical West Pacific

By Müller et al.

This paper provides a first overview of the ozonesonde and radiosonde data taken at the Palau Atmospheric Observatory (PAO) in the tropical western Pacific for six years from 2016 to 2021. After describing various ground-based observations at the PAO, the authors discuss variations of ozone, relative humidity (RH), and other meteorological parameters at interannual and annual time scales. They also compare PAO ozonesonde data from those taken at SHADOZ stations and from an aircraft campaign data.

This manuscript, as a Measurement Report, is a very good introduction to the PAO ozonesonde measurements, and is written basically very well. I think that it will be acceptable for publication in Atmospheric Chemistry and Physics after considering my comments below.

## Major comments:

My major comments are regarding Section 3.2.2 Comparison with CONTRAST.

Is the definition of RH the same between Pan et al. (2014) and this paper? I assume that this paper simply uses the radiosonde readings which are RH over liquid water, while it is probably not clearly written in the paper by Pan et al. (2014) whether they also used this, or RH over ice for temperatures colder than e.g. -20 deg.C. For the comparison in this Section 3.2.2, the same definition of RH should be used.

We in deed used the radiosonde readings as processed by Vaisala, which uses the Hyland and Wexler (1983) formulation, and did not clarify the processing of the other data sources - thank you for pointing this out, as we agree, that the comparison is otherwise not physically consistent. We contacted the authors of Pan et al. (2015) who informed us about the hybrid definition of RH in their study: RH w.r.t. water below 5 km and w.r.t. ice above 5 km, as 5 km was the average freezing level in the aircraft data. For Palau this is the case as well, so we decided to also use the altitude threshold for the PAO time series for this particular analysis and point this out in the text (lines 423-424).

The new results are very similar to the previous ones, in particular within the highlighted altitude range between 320 K and 340 K. We updated the plots accordingly.

Related to this, the choice of definition of RH in other subsections of Section 3 might be an issue (e.g. Figure 7). If the authors use the RH over liquid water, RH values tend to be lower at higher altitudes (and never get near 100 %RH above ~5 km in the tropics; see e.g. Fujiwara et al., 2003, their Figure 1), which may give biased results when analyzing data for the whole troposphere simultaneously.

Fujiwara, M., M. Shiotani, F. Hasebe, H. Vömel, S. J. Oltmans, P. W. Ruppert, T. Horinouchi, and T. Tsuda (2003), Performance of the Meteolabor "Snow White" chilled-mirror hygrometer in the tropical troposphere: Comparisons with the Vaisala RS80 A/H-Humicap sensors, Journal of Atmospheric and Oceanic Technology, 20, Issue 11, 1534-1542.

Regarding the SHADOZ data, here for all stations RH over liquid water is shown and thus compared, which we clarified in the text. We use RH as a simple and unique measure of humidity for the intercomparison between different tropical stations, which is not necessarily the physically correct RH of the atmosphere, which we now clarify in the text (lines 406-408). In general, according to Fujiwara et al. 2003 and others (e.g. Miloshevich et al. 2009 for RS92), the sensors can be trusted up to 12 km in the tropics. For a reduced altitude range from 3 to 12 km the analysis results show no significant changes in the overall signature compared to the 3 to 14 km range.

Another comment for Section 3.2.2 is as follows. As the CONTRAST campaign was conducted during January-February, the result for the analysis of PAO ozonesonde data in this season only would also be interesting. Do you obtain similar results when limiting the season? In addition, is there any dependence on the ENSO phase?

We looked into the different seasons and years separately, which had already been further discussed by Müller (2020), and now added more discussion in the manuscript in line 431 and in the Appendix (line 489), as well as new Figures 1, 2 and 3 in the Appendix as A2, A3 and A4.

There are seasons in certain years, when we indeed observe the behaviour postulated by Pan et al., a bimodal distribution in O3. In some but not all cases the high O3 mode disappears when looking only into “wet” data. From February until April (FMA, please see the companion paper, Müller et al, 2023b, for details on the seasonality specific to

Palau) we actually see this more or less clearly in all years except 2018 and 2021, as can be seen in Figure 1. It also shows in the November-December-January (NDJ) season 2019/20, but not as distinct or at all in other years and seasons. The exceptions for FMA in 2018 and 2021 might indeed be related to the ongoing La Nina at the time. Figure 2 shows the respective distributions and histograms on average for all different seasons, Fig. 3 for the different years in total.

We for now did not split the data according to ENSO phases, but the annual averages might already reveal some related characteristics. In 2019, a weak El Nino year, we observed relatively more air masses with O3 VMR > 60 ppb, which could be identified as a secondary mode in the distribution and which correspond to air masses drier than 45 % RH (see Fig. 3). In 2021, a La Nina year, we see little O3 VMR above 60 ppb and no bimodal distribution, even during the FMA season as pointed out above (see Fig. 1). We plan to investigate the impact of ENSO on O3 and RH in more detail including the current El Nino cycle.

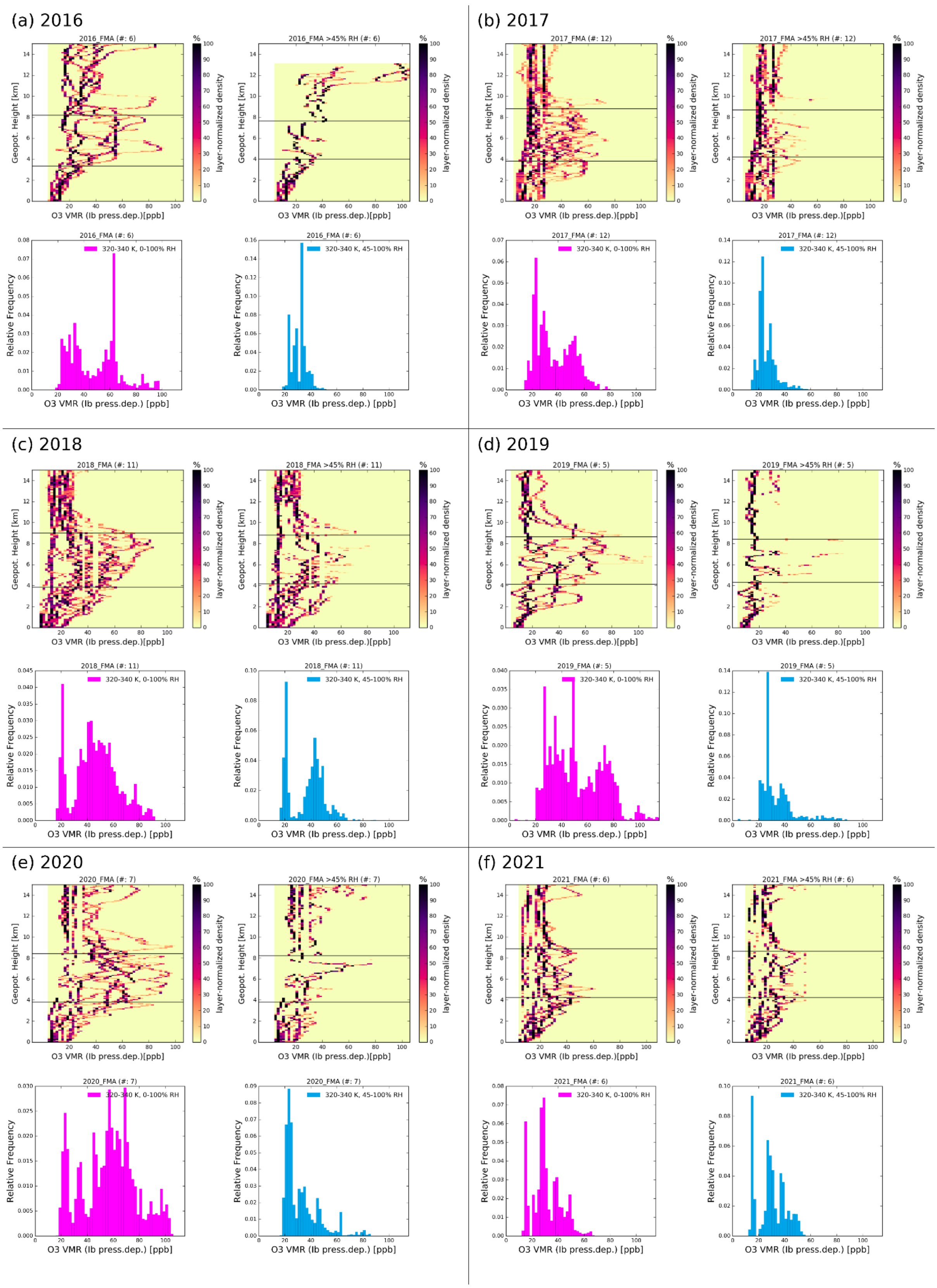


Fig. 1: Relative frequency distributions (normalized to the layer maximum) of tropospheric (0-15 km) O3 VMR with altitude (100 m bins) and histograms for the February-March-April (FMA) season for different years, for further details on the plots, see Fig. 9 in the manuscript.

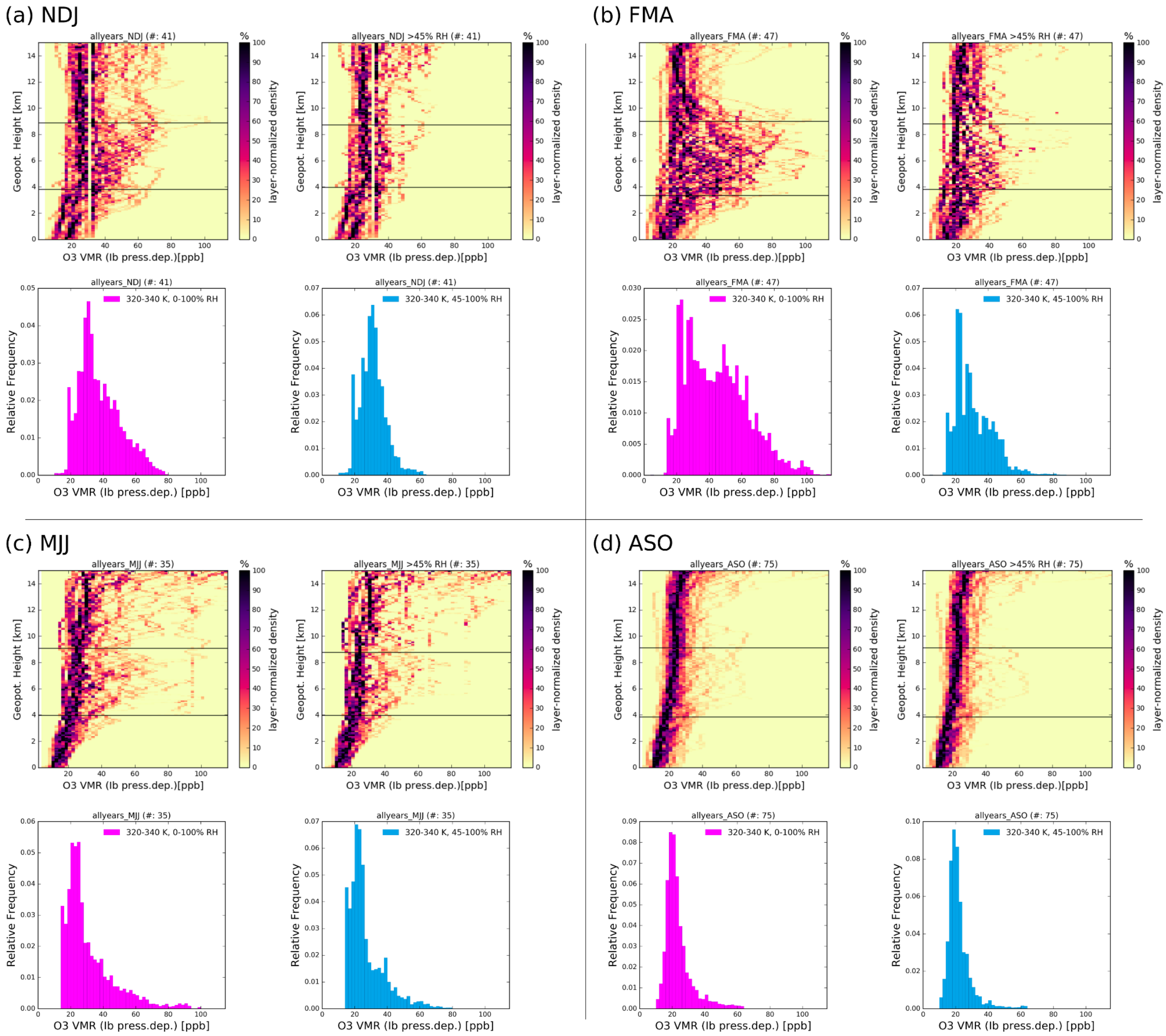


Fig. 2: as Fig. 1, but for 6-year averages of all seasons a) November-December-January (NDJ), b) February-March-April (FMA), c) May-June-July (MJJ), d) August-September-October (ASO).

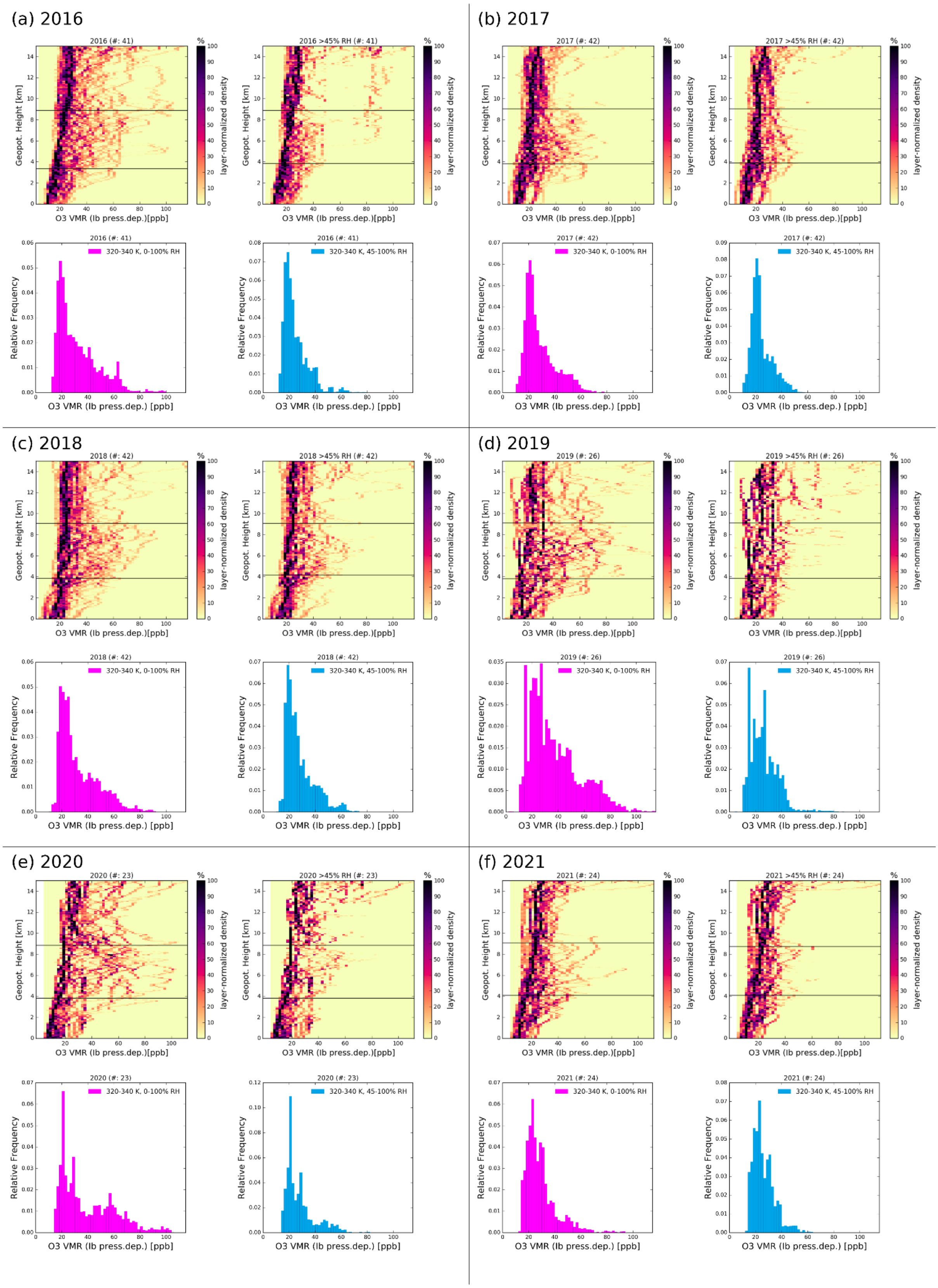


Fig. 3: as Fig. 1 and 2, but for annual averages of all years.

Minor comments:

Below are minor comments and suggestions.

Line 2 and line 42 : The latitude and longitude information of PAO should be shown here.

Done. In line 42 we further included the position information for the altitude level.

Line 128: “The pressure dependent background current correction” should be explained here, by showing the equation. This is because it looks this is the only point where the authors do not follow the published, recent recommendations for ozonesonde data processing.

Done. Please see the comments to reviewer 2 in this regard as well.

Line 174: Vömel et al. (2016) discuss an update on the uncertainties of CFH measurements.

Vömel, H., Naebert, T., Dirksen, R., and Sommer, M.: An update on the uncertainties of water vapor measurements using cryogenic frost point hygrometers, Atmos. Meas. Tech., 9, 3755–3768, doi:10.5194/amt-9-3755-2016, 2016.

Thanks for pointing this out, this reference somehow has slipped out of the manuscript. We included the reference now (again).

Lines 180-: Section 2.3: Reference papers should be shown, at least for Bruker 120 M and SFIT4 code.

We included the following two references here:

J. Notholt, G.C. Toon, C.P. Rinsland, N. Pougatchev, N.B. Jones, B.J., Conner, R. Weller, M. Gautrois, O. Schrems: Latitudinal variations of trace gas concentrations in the free troposphere measured by solar absorption spectroscopy during a ship cruise, J. Geophys. Res., 105, p.1337-1349, 2000.

Hannigan, J. W.; Ortega, I.; Shams, S. B.; Blumenstock, T.; Campbell, J. E.; Conway, S.; Flood, V.; Garcia, O.; Griffith, D.; Grutter, M.; Hase, F.; Jeseck, P.; Jones, N.; Mahieu, E.; Makarova, M.; De Maziere, M.; Morino, I.; Murata, I.; Nagahama, T.; Nakijima, H.; Notholt, J.; Palm, M.; Poberovskii, A.; Rettinger, M.; Robinson, J.; Roehling, A. N.; Schneider, M.; Servais, C.; Smale, D.; Stremme, W.; Strong, K.; Sussmann, R.; Te, Y.; Vigouroux, C. & Wizenberg, T.: Global Atmospheric OCS Trend Analysis From 22 NDACC Stations, J. Geophys. Res.-Atm., 2022, 127.

Lines 186-187: “-1” should be superscript.

Done.

Line 197: / should be ,

Done.

Line 208: Change the locations of (

Done.

Line 241: MEI: Are positive (negative) MEIs indicating El Nino (La Nina) events. Please explain this here.

The sentence introducing the figure and index now reads: “To provide context, the Multivariate ENSO Index (Wolter et al. 2011) is shown in panel c), with positive MEI indicating El Nino events and negative MEI indicating La Nina conditions.”

Line 274 vs. line 284: Use either El Nino/La Nina events or warm/cold ENSO events consistently to avoid confusion for non-experts on the ENSO terminology.

We changed the term “warm ENSO event” to “El Nino event”. Thanks for improving consistency here.

Line 368: Is there any contribution from the dependence on the radiosonde model used? (Kuala Lumpur is the only station where GRAW radiosonde has been used?)

We did not assess the differences between radiosonde models or ECC models for this analysis. A list of the different sonde models can e.g. be found in Thompson et al. 2019, Table 2. Since we cannot fully rule out any influences on the comparison, we included a phrase and reference in the text in Sect. 2.5 (lines 251-252) as follows: “Differences in radiosonde or ECC sensor model are not assessed in this study and might influence the results (see Table 2 in Thompson et al. 2019).”

Line 567-568: Müller et al. (2023) has been cited several times in the manuscript, but currently it is “to be submitted”. While I am very much looking forward to that paper, I point this out here to the editor who might have concerns from the editorial viewpoint.

We apologise that this has caused confusion - it has been mentioned by both reviewers. The two manuscripts are interlinked and were developed at the same time and submitted shortly after one another. We are glad that we can now update the citation as the manuscript is available as a discussion paper and is, as such, citable.
Müller et al. 2023b:
(https://egusphere.copernicus.org/preprints/2023/egusphere-2023-1518/)

# Reviewer report by reviewer 2:

Review of “Measurement Report: The Palau Atmospheric Observatory and its Ozonesonde Record - Continuous Monitoring of Tropospheric Composition and Dynamics in the Tropical West Pacific” by Müller et al.

Summary and General Comments:

This paper presents a detailed introduction to the Palau Atmospheric Observatory (PAO) with specific focus on the ~twice monthly ECC ozonesonde profiles that have been carried out since 2016. The paper is well written and provides excellent background information on PAO and its unique location, the ozonesonde data set, comparisons with nearby SHADOZ sites, the effects of large-scale climate oscillations, and examination of so-called “high ozone/low water vapor”, and vice-versa, features.

## Major comments:

I was surprised and in principle do not agree with the pressure-dependent background for the ECC profiles here and as described in the Müller (2020) PhD thesis. As noted in the paper, this is certainly not the recommendation of the ASOPOS/ASOPOS 2.0 Panels. Nonetheless, the application of this correction avoids the near-zero ozone observations in the UT/LS in this region that previous work has shown to be false. For future studies of UT/LS low ozone amounts in the region, corrections for the slow reaction pathway as outlined in Vömel et al. (2020) and Smit et al. (2023; new reference, see below) and discarding of the pressure-dependent background should be examined. I would like to see more discussion on this data processing choice and the ECC slow time response as justification since it is a critical issue in the UT/LS above PAO. For example, what were typical background currents Ib0, 1, and 2 for this data set? Were background currents sufficiently low so that a pressure-dependent background current correction has little effect on the calculated profiles?

We agree with the reviewer, that the choice of the background current needs to be explained carefully, because the topic remains an active field of research, as the ASOPOS 2.0 panel points out. We therefore added more discussion in the text referring to the latest publications in lines 139, 140-145 and 154-155. More generally, we propose to concentrate future work on careful reprocessing of the time series according to the latest suggested methods and the growing consensus within the community. We believe this suggestion will enable readers to understand the relevance of the background current for ozonesonde measurements at the PAO.

We document all steps of the preparation of our ozonesonde profiles to enable reprocessing in the future and followed the recommendation by the ASOPOS 2.0 panel to not change our procedure for now. That means for example, we still follow the ASOPOS (Smit et al. 2014) procedure to record Ib2 before flight, thus the choice to continue the use of Ib2 for the corrections. Figure 4 gives an overview of the different background currents recorded for the whole time series.

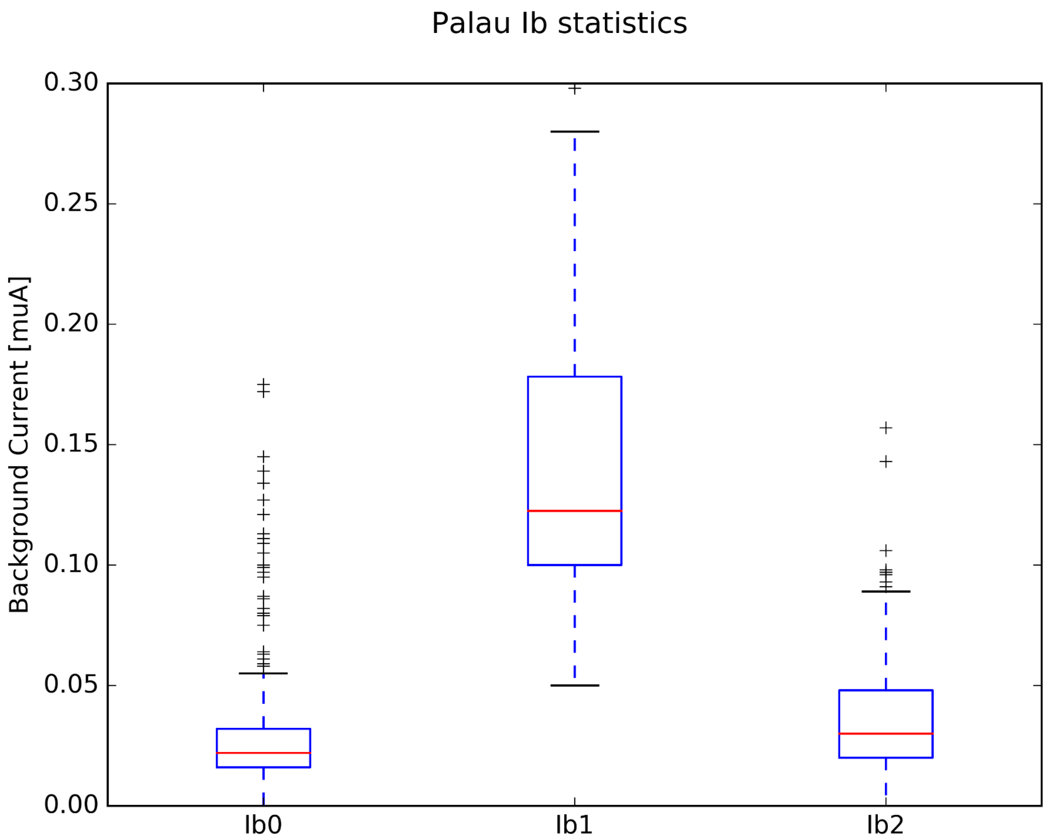


Fig. 4: Boxplots for background currents Ib0, Ib1 and Ib2 recorded during preparation according to ASOPOS, with box edges representing the first (Q1) and third (Q3) quartile and the red line giving the median, whiskers extending from the edges of the box by 1.5 x interquartile range (IQR), i.e. IQR=Q3 - Q1. Values lying outside of 1.5 * IQR are indicated as '+' signs.

We noticed higher background current values when using a new batch of ECC sondes after the first 2 years of the record. Ib2 remained below 0.05 muA with the older serial numbers, which unfortunately could not be achieved for all measurements in later years. For Ib1, the change was even worse (from a median of 0.1 muA to now 0.16 muA). Issues on the manufacturer's side had eventually been found, which the reviewer might be also aware of, but the general trend of higher measurements remains. Hence, a sensible handling of the background current remains crucial. It is exciting to see the detailed work on the time response in the current discussion paper by Smit et al. (2023).

Since a correct background current correction is still discussed (Voemel et al., 2020; Tarasick et al., 2021; Smit et al., 2023) we had to decide between a pressure-dependent or a constant background correction method for the PAO ozonesonde record. We decided against the constant method in particular with regard to the TTL region, as high background currents in the past have caused unrealistic VMR using this method (see e.g. Rex et al. 2014). The pressure-dependent correction, despite having been established under false premises, has been a compromise for us.

Figure 5 shows the differences between VMR derived with pressure dependent correction and either the constant correction (red) or the raw signal without correction (blue). The data set here is a selection of the minimum O3 VMR per sounding for the full time series in the 10 to 13 km altitude range. The O3 VMR with pressure dependent - correction show an overall difference of mostly < 5 ppb from the uncorrected VMR. We are adding this figure in the Appendix of the manuscript (Fig. A1), so that readers can get an idea about the effect of different correction methods at this critical altitude level.

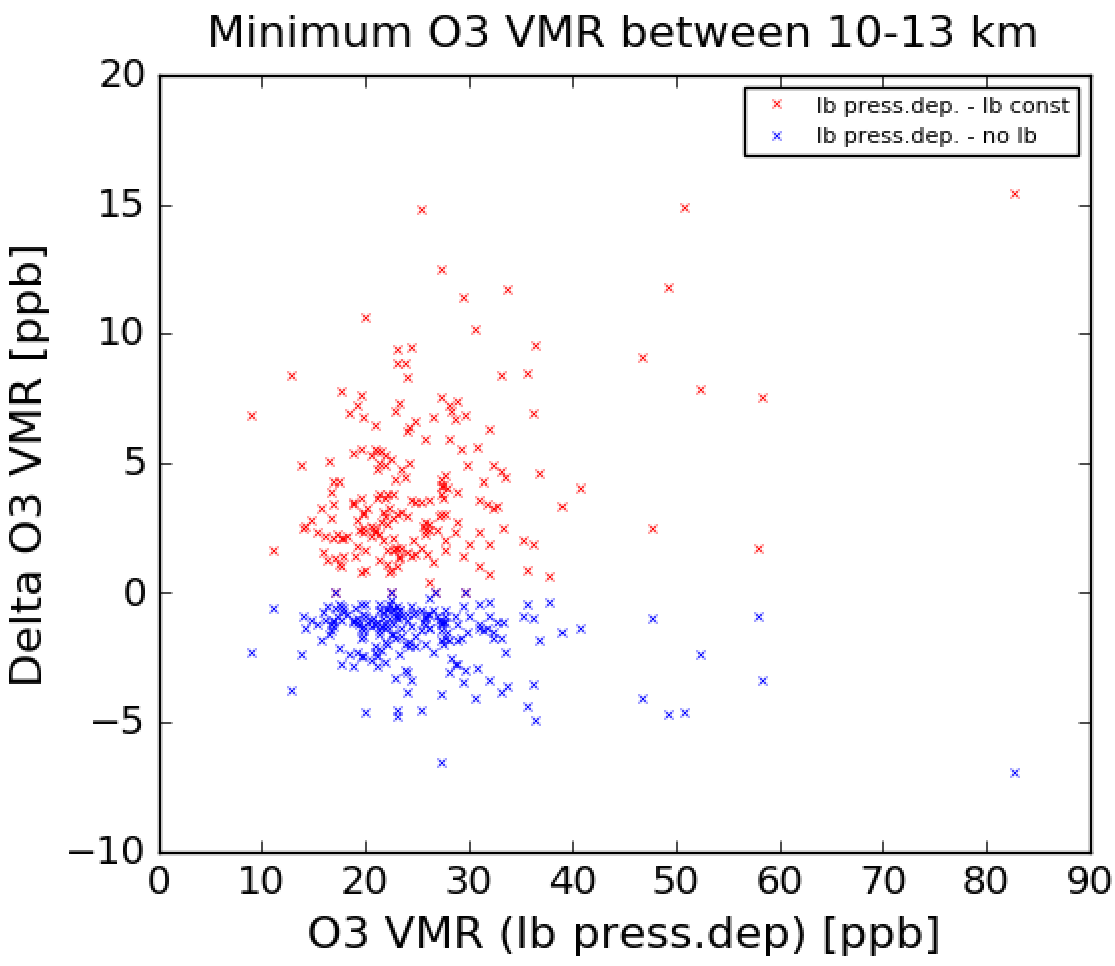


Fig. 5: Difference between O3 VMR calculated using the press.dep. correction and either the constant correction (red) or no correction applied (blue) for daily minimum O3 VMR (press.dep. Ib) between 10 and 13 km altitude.

Smaller note: several times we are asked to consult a yet to be submitted Müller et al. (2023) paper. I suggest removing this reference for now, although by the time a final version of this paper is accepted, that may become available and it will be appropriate to add back in.

We apologise that this has caused confusion - it has been mentioned by both reviewers. The two manuscripts are interlinked and were developed at the same time and submitted shortly after one another. We are glad that we can now update the citation as the manuscript is available as a discussion paper and is, as such, citable.
Müller et al. 2023b:
(https://egusphere.copernicus.org/preprints/2023/egusphere-2023-1518/)

Recommendation:

I recommend this paper for publication after some additional discussion noted above, and very minor technical corrections as follows.

## Minor comments:

Specific and Line-by-Line Comments:

Line 61: Extra space after “/”

Done.

Line 164: I assume you have examined the pump temperatures for potential solution freezing/boiling effects as well, correct?

Thanks for pointing to this issue. We added the sentence: "Monitoring of the pump temperatures yields no explanation for these differences."

Line 193: Fix the quotations here

Done.

Line 212: analyses

Done.

Line 216: What type of radiosondes are launched there? If RS41 with its superior RH measurement, this would be an excellent data set for examining TTL RH.

For the first years of the time series the Palau weather station used Lockheed Martin LMS-06 radiosondes. But when switching the station to the airport, i.e. in 2018, they switched to Vaisala RS41-NG. Thank you for pointing this out, we actually did not know about this switch before as it has not been specifically reported! We included this information in the text in lines 234-235.

Line 231: Thompson et al. (2021; https://doi.org/10.1029/2021JD034691) also used these distinguished characteristics to calculate regional ozone trends from SHADOZ data.

Thanks for finding a good spot to include this citation. We added the sentence in line 248 as: “More recently, Thompson et al. (2021) also used these distinct characteristics to calculate regional ozone trends from SHADOZ data.”

Figure 5 caption: “the a prominent layer” delete “the”

Done.

New paper to consider citing for discussion of the ECC “background” current and correction of the time response delay of the ozonesonde:

Smit, H. G. J., Poyraz, D., Van Malderen, R., Thompson, A. M., Tarasick, D. W., Stauffer, R. M., Johnson, B. J., and Kollonige, D. E.: New Insights From The Jülich Ozone-Sonde Intercomparison Experiments: Calibration Functions Traceable To One Ozone Reference Instrument, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2023-1466, 2023.

We included the citation as discussed above.

# Additional changes to the manuscript made by the authors

Besides updating the bibliography and very minor changes with regard to language, we made some additional small changes to the manuscript, as follows:

Line 106: We added the citation of the dataset in the text here, which had been requested after the change of the article type during the initial submission process.

Fig. 3: Here, we found a small error in the timeline of the ComCAL instrument, pointed out by a friendly reviewer, which we corrected. Whilst at it, we increased font sizes for improved readability.

Lines 193-194: For clarification, we added the sentence “Note, RH measurements used in this study are solely from radio soundings and not from CFH sondes.”

Line 286: We updated the sentence with regard to the current situation of the ENSO cycle. It now reads: “A weak El Nino in 2018/2019 was followed by a longer and stronger La Nina phase that lasted into 2023. Since mid-2023 El Nino conditions occur.