Response to comments by Referee #2 (Dr. Michelle Santee)

Thank you very much for your review.

Review of "Lower-stratospheric aerosol measurements in eastward shedding vortices over Japan from the Asian summer monsoon anticyclone during the summer of 2018"
by Fujiwara et al.

Ground-based lidar measurements obtained at two stations in Japan during July to September 2018 are analyzed to look for signatures of the transport of aerosols from the Asian summer monsoon (ASM). Particle enhancements were observed in the lower stratosphere in August and September; back trajectories and satellite and reanalysis data are used to show that those air masses originated within the ASM anticyclone and were not influenced by volcanic or biomass burning emissions, and thus they likely reflect extension of the Asian Tropopause Aerosol Layer (ATAL) associated with eastward eddy shedding events. The analysis presented is sound, the manuscript is well written and well organized, and the topic is timely and of interest to the ACP readership. Although I do have some substantive issues that I would like to see considered before the paper is accepted for publication, most of my comments are minor wording suggestions.

Thank you very much for your evaluation.

Specific comments and questions (major substantive issues and minor points of clarification, wording suggestions, and grammar / typo corrections are listed together for each Section in sequential order through the manuscript):

Introduction

- L41-42: It would be better to add "e.g." in front of the list of references for aerosols and water vapor, as is done for trace gases.

Will be added.

- L53: was believed --> is believed

Will be changed.

- L57-59: The way this sentence is constructed – first talking about the behavior observed during a specific week in August 1997 and then stating that the peak in NH_4NO_3 occurs around August – may be slightly confusing for readers, especially those who are not familiar with Höpfner et al.'s paper and the particular satellite data they analyzed. I can understand that the authors do not wish to add extraneous detail to the Introduction, but I think it would be better to break this sentence in two and make it more clear that the findings reported by Höpfner et al. were based on different satellite data sets. As it is now, the week of 8–16 August 1997 appears to hold some special significance, rather than just being when CRISTA data were taken.

We will revise this sentence as: "Their satellite data analysis using Cryogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA) data indicates enhanced NH_4NO_3 signals around the tropopause, both in the ASM region and the western Pacific (including Japan) during 8–16 August 1997 (with the western Pacific signals suggestive of shedding vortices); also, their analysis of satellite Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) data together with CRISTA data show that the mass of NH_4NO_3 in the ASM region at 13–17 km peaks around August."

- L70: data --> the data

Will be corrected.

Section 2

- L86: made --> done

Will be changed.

- L97: The uncertainties of lidar data are discussed here, which are applied to the both systems --> The uncertainties of lidar data discussed here are applicable to both systems

Will be changed.

- L121: Since "PV" is used below, "(PV)" should be added here after "potential vorticity".

Will be added.

- L122 and L126: on to --> onto

Will be corrected.

- L124: Delete the comma after "but".

Will be deleted.

- L126-127: PV … are --> PV … is

Will be corrected.

- L136-137: Young and Vaughan (2009) does not appear in the reference list.

Thank you for pointing this out. We will add it.

Section 3

- L157: mixture --> a mixture

Will be added.

- Figure 1: The color bar, particularly for the PDR panel, could be improved. The pink color denoting the highest values is somewhat difficult to distinguish from the light purple used at the bottom of the PDR range; this would not really be a problem if more were filled in, but it complicates interpretation of such a sparse plot.

We will change the color bar, by making the pink darker, and by removing light purple from the PDR (and TDR) figure. Around the end of this response letter, we attach the revised version of the figures.

- L179: Delete the comma after “2018”.

Will be deleted.

- L191: For clarity, it might be good to repeat verbatim the description in the Figure 1 caption: “the daily (first) lapse-rate tropopause”.

Will be added.

- L192: with --> at

Will be corrected.

- L199: at a 400 K --> at 400 K

Will be removed.

- L200: of boundaries --> of the boundary

Will be added.

- L201: The value for the CO concentration (65 ppbv) selected to identify the ASM anticyclone boundary seems reasonable, but nevertheless it would be appropriate to cite a reference to justify this choice.

We will revise this sentence as: “By comparing the results from Santee et al. (2017) with our own analysis, the 65 ppbv contours . . . are chosen . . . ”

- L210: Although it’s implicit, rather than stating “during July–September 2018”, it would be better to say “on all days during July–September 2018 on which measurements were made”.

Will be corrected as suggested.

- Figures 4 and 5:
  - The vast majority of back trajectories launched from Tsukuba indicate that the air parcels had been transported from altitudes above about 10–12 km over the preceding 10 days (and even slightly higher than that for trajectories run back from Fukuoka). It therefore seemed a bit odd to me that the color bar extends down to Z = 4 km. I had to look closely to spot the single trajectory that appears to originate in the middle of the North Pacific at that altitude (Figure 4, bottom panel). Some explanation for this apparently anomalous parcel should be given.
  - The greens in these color bars are impossible to distinguish, so if the authors feel that the trajectory geopotential height information is important, then they need to use a different color palette. Also, the color bar increments (0.8 km) are awkward.
  - Perhaps both of the above issues could be addressed by reformulating the plots. The existence of the outlier parcel could simply be mentioned in the text and not shown, allowing the geopotential height range to be decreased so fewer colors are needed.

The coloured geopotential height range of Figures 4 and 5 will be changed (narrowed); please see the revised version of these figures at the end of this response letter.

Regarding the trajectory originating in the middle of the North Pacific at that altitude (Figure 4, bottom panel), we will add the following note in the text: “Note that there is a trajectory that originates in the Pacific south of Japan as low as 4 km (Figure 4, bottom, a small-scale spiral in purple); this is associated with upward transport in the typhoon Soulik.”

- L230-232: I don’t find the discussion of PV very illuminating. First, for clarity it would be better to say “with lower values inside than outside the ASM anticyclone at the same latitude (e.g., 30°N)”. More importantly, I’m puzzled by the lack of a clear signature of anticyclonic flow in the PV field. Previous studies have diagnosed eddy shedding through examination of PV maps. In particular, Garny and Randel [JGR, 2013] (a paper that I am surprised to see is not cited in this manuscript) reported an episode of eastward eddy shedding in June 2006 not unlike the one depicted here, and they showed that the contours of CO (from MLS) and PV (from MERRA) follow very similar patterns (although they focused on a lower theta level, 360 K). Perhaps the authors should explore using different PV contours or applying some smoothing to the PV fields to see if a clearer signal emerges. In any case, more discussion of the behavior of PV associated with this event is warranted.

The paper by Garny and Randel (2013) will be cited in the Introduction.

Following the suggestion by Referee #3 (i.e., at and above 400 K, PV is not very useful to see the boundary of the ASM anticyclone), we will change from PV to Montgomery streamfunction (MSF). Please see the response to Referee #3 for detailed revisions. Around the end of this response letter, we show the revised Figure 6 with MSF.

- L236: It seems to me that, in addition to Figure 7, the results of Figure 6 also suggest that the 60-ppbv CO contour is indicative of eastward eddy shedding vortices.

This sentence is specifically for Figure 7 in which we take a latitudinal average. We will revise this sentence as: "In Fig.7, the 60-ppbv CO contour may be a good indicator of eastward shedding vortices."

- L237-238: It is difficult for the reader to judge the timing of these events from Figure 7, especially given that some y-axis tick marks appear to be absent. For example, for the first episode listed, the 60-ppbv CO contour seems to be present at the longitudes of the lidar sites a couple of days before 5 August. Are the 20-25 and 27-31 August events really separable? It might be helpful to guide the eye by overlaying colored horizontal lines to mark each event's start and end dates or perhaps pale (transparent) colored bands spanning the intervals.

We add coloured line segments for the periods when CO concentration was ≥~60 ppbv along the longitude of Tsukuba. Please see the revised Figure 7 at the end of this response letter. (The same line segments will also be added to the MLS figures.) Furthermore, the text describing these periods will be corrected as: 3–15, 20–24, and 28–31 August, and 3–8, 14–17, and 28–29 September.

- L223-240: Since previous studies have used MLS CO to look for evidence of eddy shedding (e.g., Garny and Randel [2013], Honomichl and Pan [2020]), and since MLS water vapor is shown in Figure 8, I am curious why only CO from the CAMS reanalysis – and not from MLS – is used here. Is it because the necessary longitudinal and temporal binning would smear out finer-scale structures too much? Clearly, despite such smoothing, the MLS H_2O data provide valuable information. On the flip side, the authors should probably offer an explanation of why water vapor from MLS was used but not that from CAMS.

Figures R2-1 and R2-2 below show the comparisons of CAMS data and MLS data for CO and for water vapor. As we can see, for both CO and water vapor, CAMS and MLS show qualitatively and broadly similar eastward extension signals over Japan; however, CAMS CO is greater than MLS CO (e.g., the differences are ~10 ppbv around the longitudes of Japan through August–September 2018), and CAMS water vapor mixing ratios are greater than MLS water vapor (e.g., the differences are roughly ~2 ppmv for the wet signals around the longitudes of Japan in August 2018). In this paper, we primarily use CAMS CO data as a high-resolution tracer of the ASM anticyclone. Figure 7 is a companion one for Figure 6. For water vapor, however, we use MLS data because MLS water vapor measurements in the lower stratosphere have been well validated with e.g., balloon measurements (e.g., Hurst et al., 2016; Fujiwara et al., 2010; Vömel et al., 2007), while reanalysis water vapor data in the lower stratosphere are in general less reliable (e.g., Davis et al., 2017).

Thus, we will make the following revisions:

In Section 2.2, in the second paragraph, we will add the following sentences:
"CAMS CO data are originally provided in mass mixing ratio, kg kg^{-1}, which are converted to volume

mixing ratio, ppbv, for this study. It is noted that a quick comparison with MLS Version 4.2 Level 2 CO data (Santee et al., 2017; Livesey et al., 2020) at 400 K isentropic surface (in the form of longitude-time diagram like the one in Section 3.2) shows that CAMS CO data are roughly ~10 ppbv greater than MLS CO over Japan during August–September 2018, but also shows that eastward extension signals coming over Japan agree fairly well qualitatively within the differences in spatio-temporal sampling of the two data sets."

In Section 2.2, we will have the following new (third) paragraph for MLS water vapor data:
"MLS Version 4.2 Level 2 water-vapour data (Santee et al., 2017; Livesey et al., 2020) are analysed because water vapour is also a good tracer of the ASM anticyclone. We use MLS data rather than CAMS data for lower stratospheric water vapour because MLS data have been well validated with e.g., balloon-borne frost-point hygrometers (e.g., Hurst et al., 2016; Fujiwara et al., 2010; Vömel et al., 2007), while reanalysis water vapor data are in general less reliable in the lower stratosphere (e.g., Davis et al., 2017). We found that CAMS water vapour volume mixing ratio data (converted from the original specific humidity data) are greater than MLS data at 400 K isentropic surface over Japan during July–September 2018 (e.g., the differences are roughly ~2 ppmv for the wet signals around the longitudes of Japan in August 2018)."

References (will be added):

Davis, S. M., Hegglin, M. I., Fujiwara, M., Dragani, R., Harada, Y., Kobayashi, C., Long, C., Manney, G. L., Nash, E. R., Potter, G. L., Tegtmeier, S., Wang, T., Wargan, K., and Wright, J. S.: Assessment of upper tropospheric and stratospheric water vapor and ozone in reanalyses as part of S-RIP, Atmos. Chem. Phys., 17, 12743–12778, https://doi.org/10.5194/acp-17-12743-2017, 2017.

Fujiwara, M., Vömel, H., Hasebe, F., Shiotani, M., Ogino, S.-Y., Iwasaki, S., Nishi, N., Shibata, T., Shimizu, K., Nishimoto, E., Valverde-Canossa, J. M., Selkirk, H. B., and Oltmans, S. J.: Seasonal to decadal variations of water vapor in the tropical lower stratosphere observed with balloon-borne cryogenic frostpoint hygrometers, J. Geophys. Res., 115, D18304, https://doi.org/10.1029/2010JD014179, 2010.

Hurst, D. F., Read, W. G., Vömel, H., Selkirk, H. B., Rosenlof, K. H., Davis, S. M., Hall, E. G., Jordan, A. F., and Oltmans, S. J.: Recent divergences in stratospheric water vapor measurements by frost point hygrometers and the Aura Microwave Limb Sounder, Atmos. Meas. Tech., 9, 4447–4457, https://doi.org/10.5194/amt-9-4447-2016, 2016.

Vömel, H., Barnes, J. E., Forno, R. N., Fujiwara, M., Hasebe, F., Iwasaki, S., Kivi, R., Komala, N., Kyrö, E., Leblanc, T., Morel, B., Ogino, S.-Y., Read, W. G., Ryan, S. C., Saraspriya, S., Selkirk, H., Shiotani, M., Valverde Canossa, J., and Whiteman, D. N.: Validation of Aura MLS water vapor by balloonborne Cryogenic Frostpoint Hygrometer measurements, J. Geophys. Res., 112, D24S37, https://doi.org/10.1029/2007JD008698, 2007.

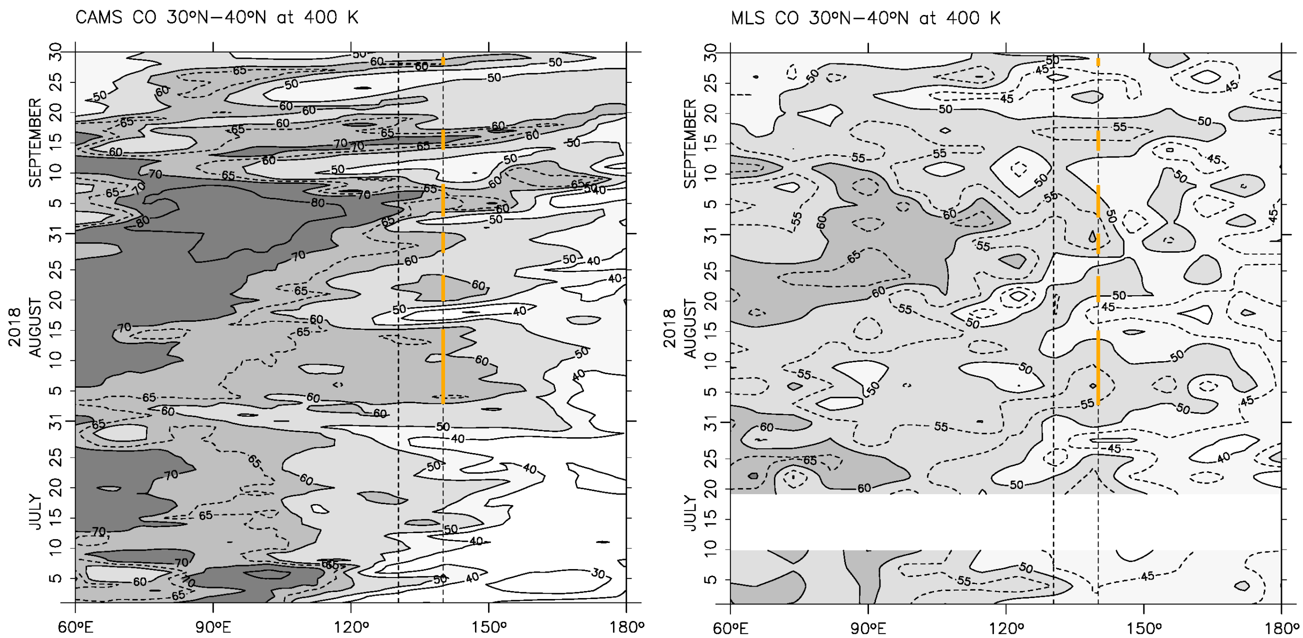


Figure R2-1. (Left) Same as Figure 7 (the revised version). (Right) Same as left but for MLS CO data. Data for the 30°N–40°N region have been aggregated into 3-day and 8°-longitude bins, each constituting about 10 individual data points. The contours for 45 ppbv and 55 ppbv as well as 65 ppbv are added as dotted lines.

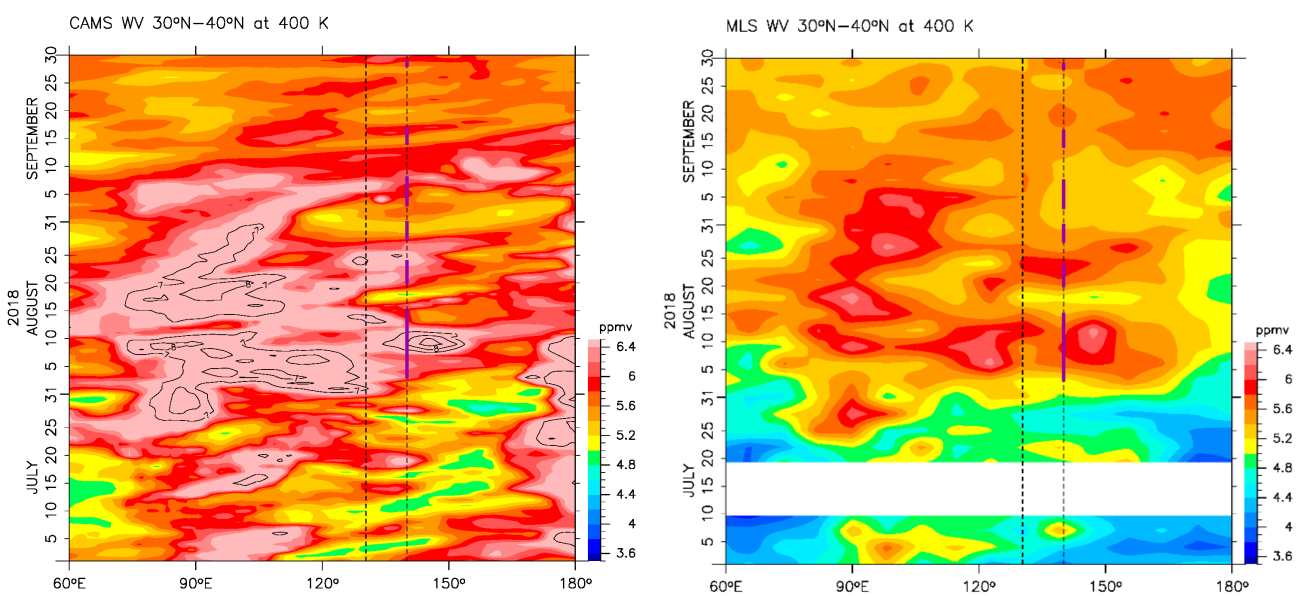


Figure R2-2. (Left) Longitude–time distribution of daily averaged water vapor volume mixing ratio at 400 K potential temperature averaged over 30°N–40°N, using CAMS reanalysis specific humidity data. The contour lines for 7, 8, 9, 10, 11 ppmv have been added. (Right) Same as Figure 8 (the revised version).

- L268-269: types of particle and gas --> types of particles and gases

Will be corrected.

- L286: but not have --> but had not

Will be corrected.

- L287: I think it would be appropriate to add "at least not in a monthly mean view" at the end of this sentence (mirroring the zonal mean caveat in L282).

Will be added.

- L288: unlikely due to --> unlikely to be due to

Will be corrected.

Section 4

- L311: measurements at --> measurements made at; also, delete the comma after "2018"

Will be corrected.

- L314: the BSR --> BSR

Will be deleted.

- L315-316: The authors might want to specifically note that none of the 11 nights on which data were taken at Fukuoka fell during the intervals of strong enhancement observed at Tsukuba.

We will add to this sentence "and due to the fact that the dates of lidar operation at Fukuoka did not overlap those at Tsukuba when strong enhancement was observed"

- L318: are of a few --> are a few

Will be corrected.

- L319: originate in the ASM anticyclone in association with eastward shedding vortices --> originate in the ASM anticyclone and are transported over these sites in association with eastward shedding vortices

Will be added.

- L320: eruptions and extensive --> eruptions or extensive

Will be corrected.

- L325-330: I'm not convinced that this is the best place for the discussion of the OHP measurements. I think it is generally inappropriate to introduce new aspects in the "Summary and Conclusions" section. In fact, these lines might belong in the Introduction.

This sentence will be moved to the Introduction.

- L332: Add a comma after "(3%–10%)".

Will be added.

- L337: Is it necessary to repeat "PDR" 3 times in this line (i.e., is it needed after "8%" and "4%")?

Will be deleted.

- L340-342: It seems to me that here again the Moana Loa and OHP information in these lines is out of place. Since these data add to the evidence that the signals observed at Tsukuba and Fukuoka must have arisen from the ATAL, this discussion could be moved to Section 3.3, which could then be renamed to reflect its exploration of other potential causes of the observed enhancements and not just focus on "Satellite aerosol data".

We will move these sentences to the end of Section 3.3, whose title will be changed to "Investigation of other potential causes".

- L341: any enhancement --> no enhancement

Will be corrected.

- L360: Delete the comma after "extent"; also, is --> are

Will be corrected.

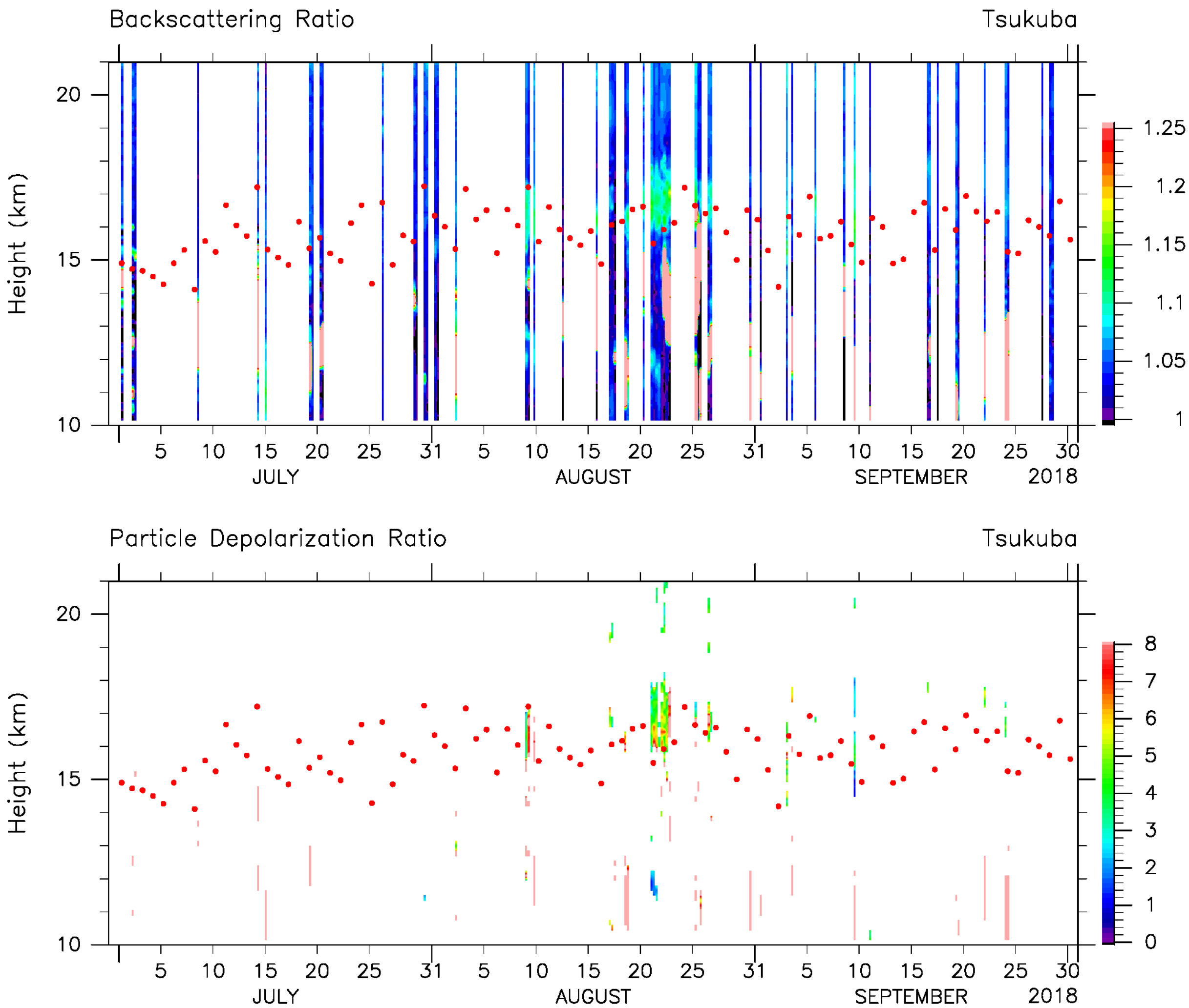
References

- L440: The paper by Hanumanthu et al. has now been accepted for publication in ACP.

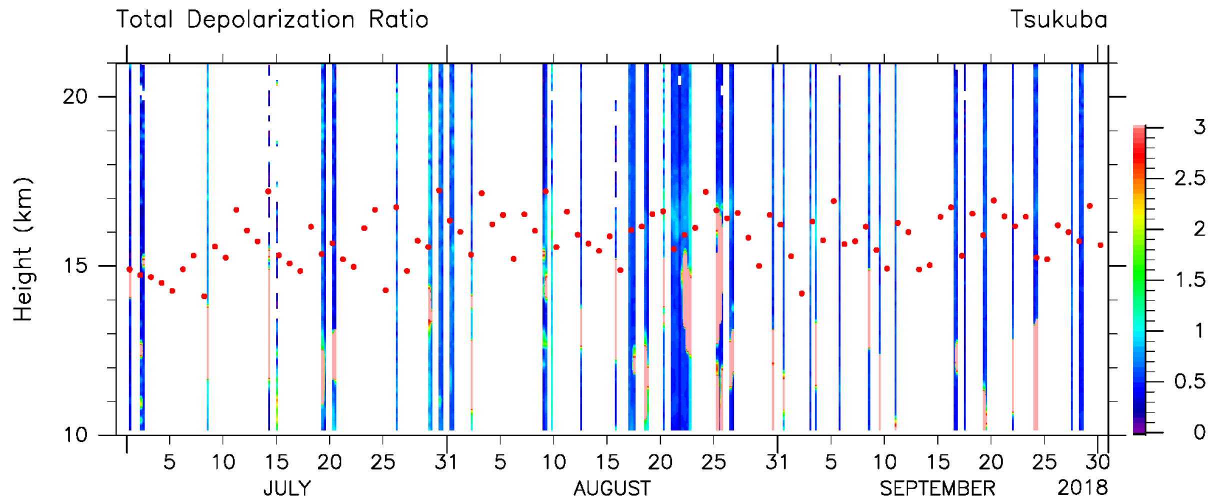
Will be updated.

- L473: The reference for the MLS Data Quality Document (Livesey et al.) is missing the year.

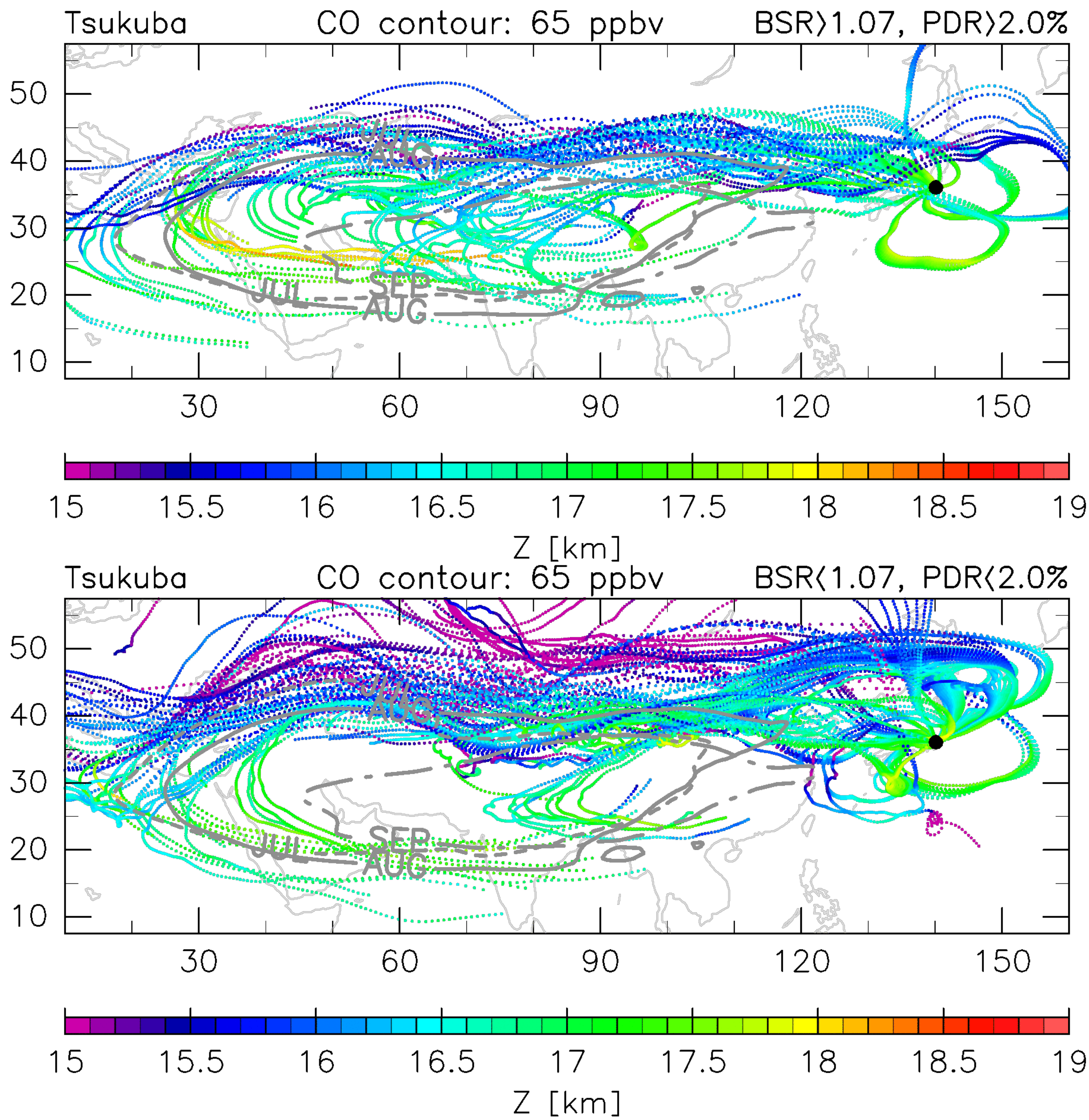
The year 2020 will be added.



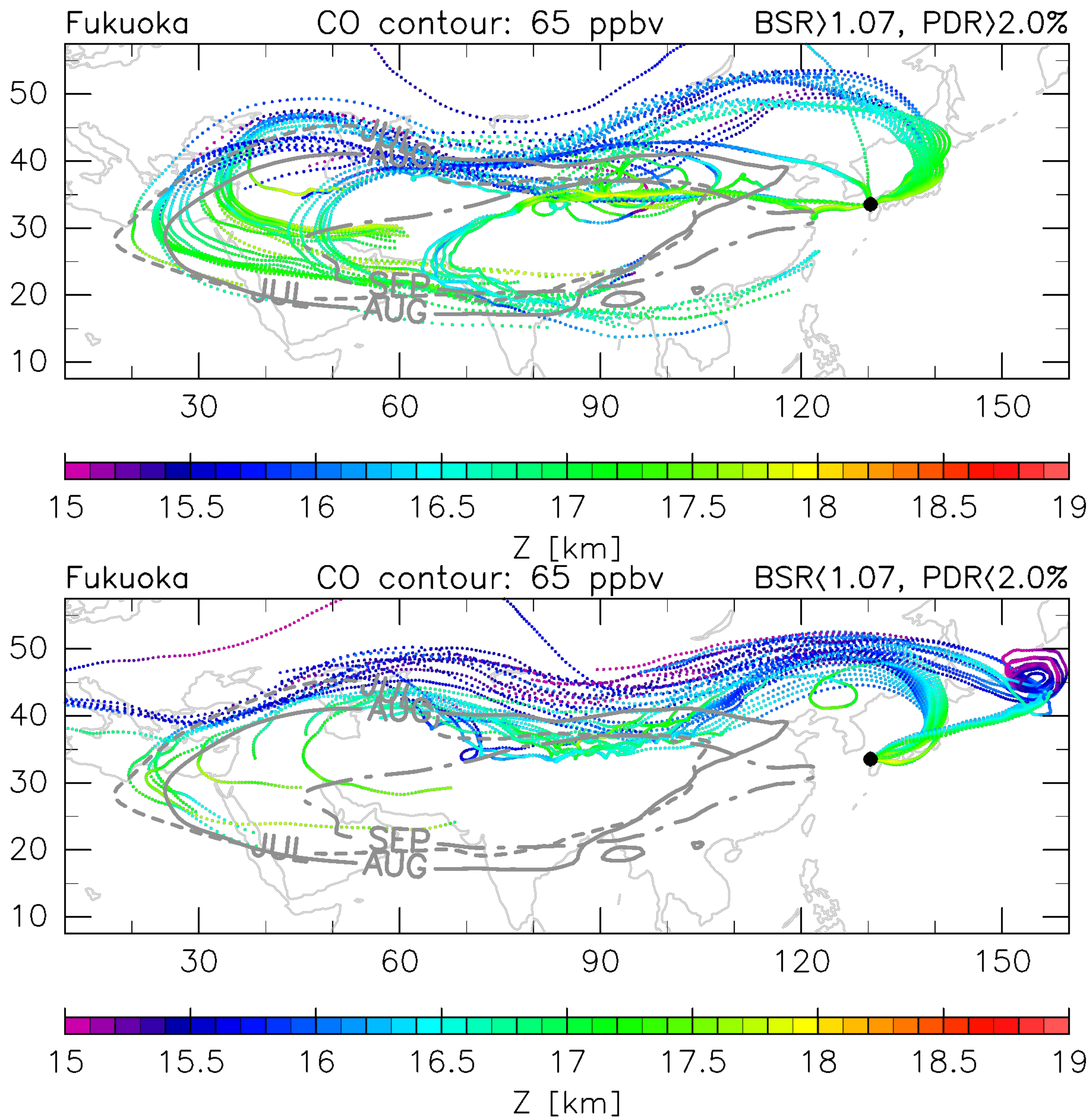
Revised Figure 1: The colour bar has been slightly changed.



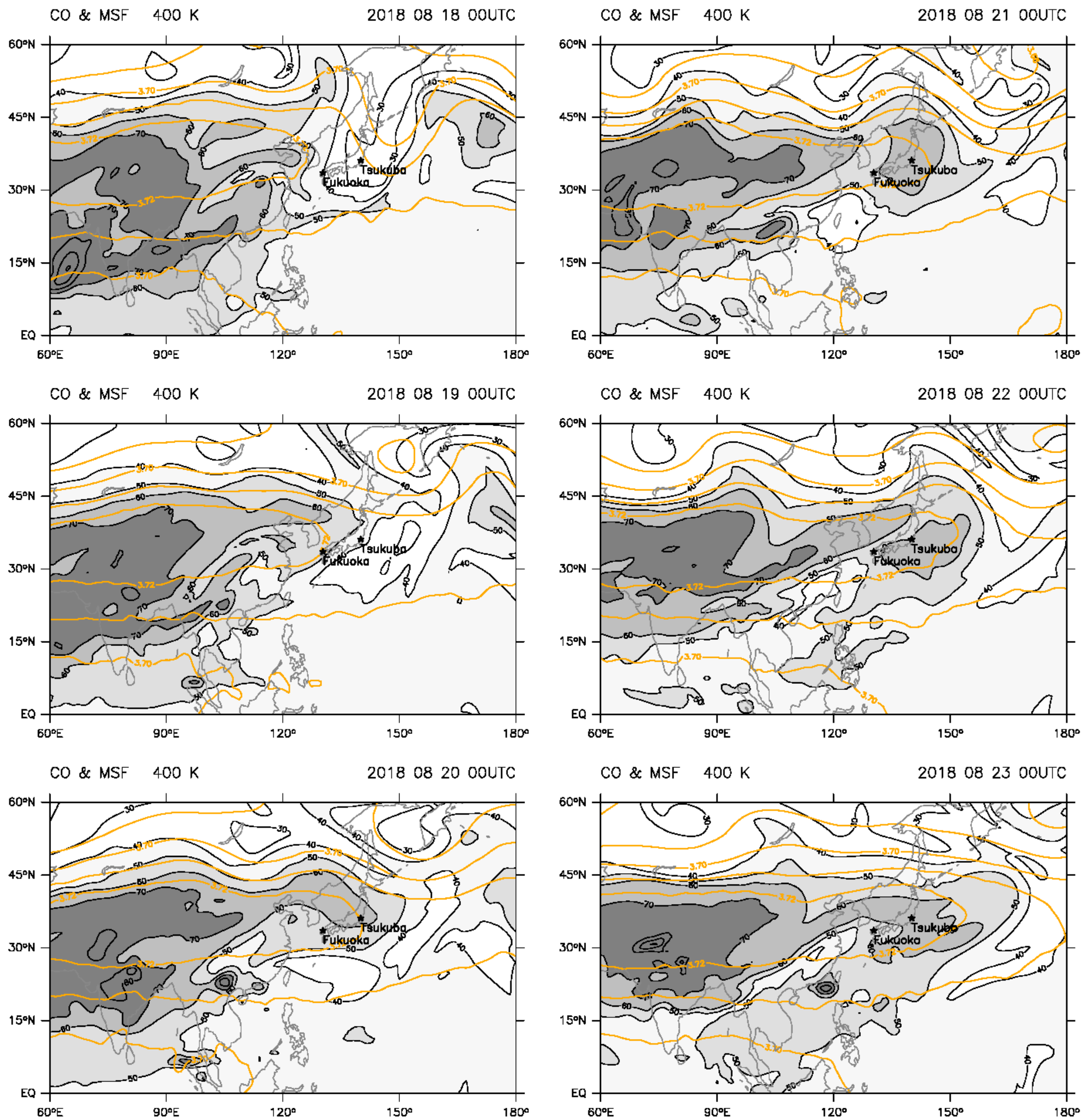
Revised Figure A1: The colour bar has been slightly changed.



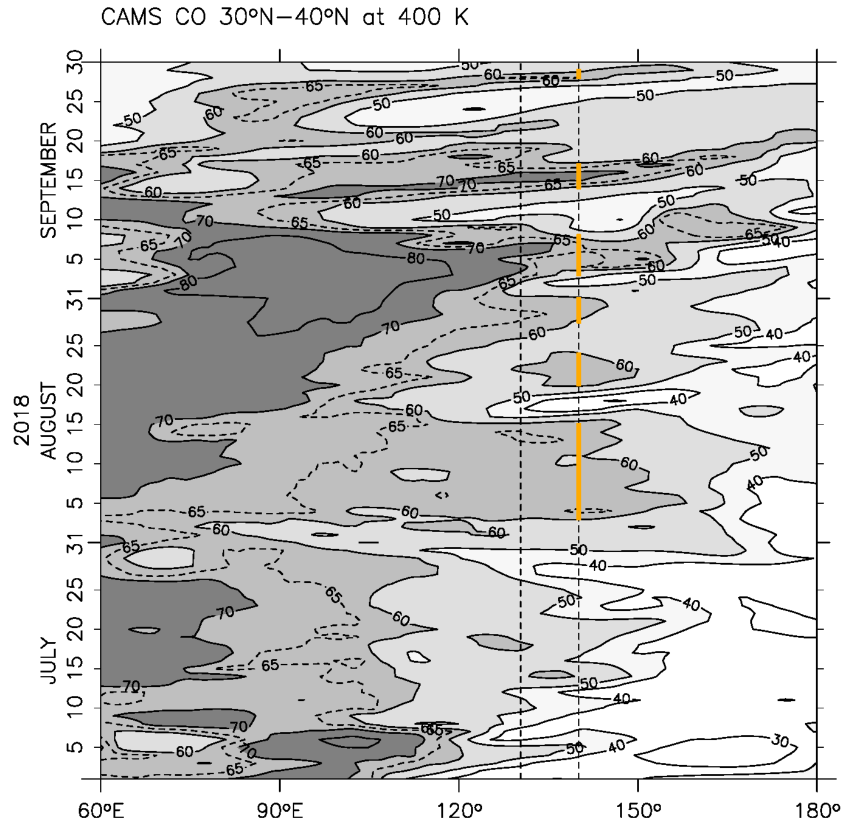
Revised Figure 4: The colour code for geopotential height has been changed (to narrow the range of Z). The CO isolines for different months are expressed with different line styles (i.e., dotted for July, solid for August, and dash-dotted for September).



Revised Figure 5: The colour code for geopotential height has been changed (to narrow the range of Z). The CO isolines for different months are expressed with different line styles (i.e., dotted for July, solid for August, and dash-dotted for September).



Revised Figure 6: PV has been replaced with Montgomery streamfunction (MSF; coloured contours at intervals of 0.01 × 10^5 m^2 s^{-2}). Also, CO and MSF data are now instantaneous at 00 UTC, not daily averages.



Revised Figure 7: Coloured line segments have been added for the periods when CO concentration was ≥~60 ppbv along the longitude of Tsukuba (i.e., 3–15, 20–24, and 28–31 August, and 3–8, 14–17, and 28–29 September).