Response to comments by Anonymous Referee #3

Thank you very much for your review.

Comments on Manuscript No. ACP-2020-980

In the last decade the Asian Tropopause Aerosol Layer (ATAL) becomes in the focus of attention. This study shows that the transport of aerosol out of the ATAL by eastward shedding vortices were measured over Japan by two lidar systems during summer 2018. Several eddy shedding events were observed and backward trajectory calculations indicate that eddies including air masses with enhanced aerosol particles originate in the Asian monsoon anticyclone. The analysis of satellite observations and meteorological reanalysis confirm the eddy events and further show that the considered time period was free of the impact of volcanic eruptions and high forest fires.

This is a very interesting study, which merits its publication in ACP. The scientific content, the quality of the study and its presentation is good. Therefore, I suggest only some minor revisions before publication by ACP

Thank you very much for your evaluation.

1) P2/L51: 'The enhanced aerosol particle signature in the ASM anticyclone at 14–18 km altitude is known as the Asian Tropopause Aerosol Layer (ATAL), which was believed to consist of carbonaceous and sulphate materials, mineral dust, and nitrate particles (Vernier et al., 2015, 2018; Brunamonti et al., 2018; Bossolasco et al., 2020; Hanumanthu et al., 2020).'

From this statement it is not clear if the knowledge about the chemical composition of the the ATAL particles is based on in situ measurements, remotes sensing observations or model simulations. The discussion about the chemical composition of the ATAL should be much more improved and clarified. I recommend to add a short summery about the current knowledge of ATAL particle characteristics (e.g. chemical composition, particle size distribution, particle form, possible sources etc.). This would help to better bring the results of the lidar measurements presented in this study into the context of previous publications.

We would like to point out that one of the latest ATAL studies by Bossolasco et al. (2020) (a modeling study under review) discuss in the Introduction that "The sources, chemical composition and spatial and temporal variability of the ATAL are not yet well understood." Also, Hanumanth et al. (2020) (a study using balloon borne backscatter sondes) discuss in the Introduction that "The source regions of ATAL aerosols and their chemical precursors on the Earth's surface (origin) as well as the transport pathways from the surface to ATAL altitudes are poorly understood." Note also that Referee #2 suggested us to change from "was believed" to "is believed" in this sentence. Furthermore, in the text, in the following sentences, we introduce the recent study by Höpfner et al. (2019) rather extensively. We believe that this is the current status of our knowledge regarding the chemical composition of ATAL. Thus, we believe that the current text is not inappropriate for the introduction to a study using ground-based lidar systems.

2) P4/L114: ERA5 is a very new product from ECMWF, therefore I think it is worth to add a few references demonstrating the quality of ERA5 compared to the former ERA-Interim reanalysis.

We will add the following sentences: “ERA5 temperature data in the tropical tropopause layer have been evaluated by Tegtmeier et al. (2020). Lagrangian transport calculations using ERA5 and its predecessor ERA-Interim have been compared by Hoffmann et al. (2019) and Li et al. (2020).”

Tegtmeier, S., Anstey, J., Davis, S., Dragani, R., Harada, Y., Ivanciu, I., Pilch Kedzierski, R., Krüger, K., Legras, B., Long, C., Wang, J. S., Wargan, K., and Wright, J. S.: Temperature and tropopause characteristics from reanalyses data in the tropical tropopause layer, Atmos. Chem. Phys., 20, 753–770, https://doi.org/10.5194/acp-20-753-2020, 2020.

Hoffmann, L., Günther, G., Li, D., Stein, O., Wu, X., Griessbach, S., Heng, Y., Konopka, P., Müller, R., Vogel, B., and Wright, J. S.: From ERA-Interim to ERA5: the considerable impact of ECMWFs next -generation reanalysis on Lagrangian transport simulations, Atmos. Chem. Phys., 19, 3097–3124, https://doi.org/10.5194/acp-19-3097-2019, 2019.

Li, D., Vogel, B., Müller, R., Bian, J., Günther, G., Ploeger, F., Li, Q., Zhang, J., Bai, Z., Vömel, H., and Riese, M.: Dehydration and low ozone in the tropopause layer over the Asian monsoon caused by tropical cyclones: Lagrangian transport calculations using ERA-Interim and ERA5 reanalysis data, Atmos. Chem. Phys., 20, 4133–4152, https://doi.org/10.5194/acp-20-4133-2020, 2020.

3) P4/L122: Please add a statement like this: ’CO and the ATAL have not necessarily the same emission sources, however CO is a good chemical tracer to indicated the location of the Asian monsoon anticyclone.’

We will add the following sentence:
Although CO and ATAL aerosol particles do not necessarily have the same emission sources, CO is a good chemical tracer to indicate the location of the ASM anticyclone.

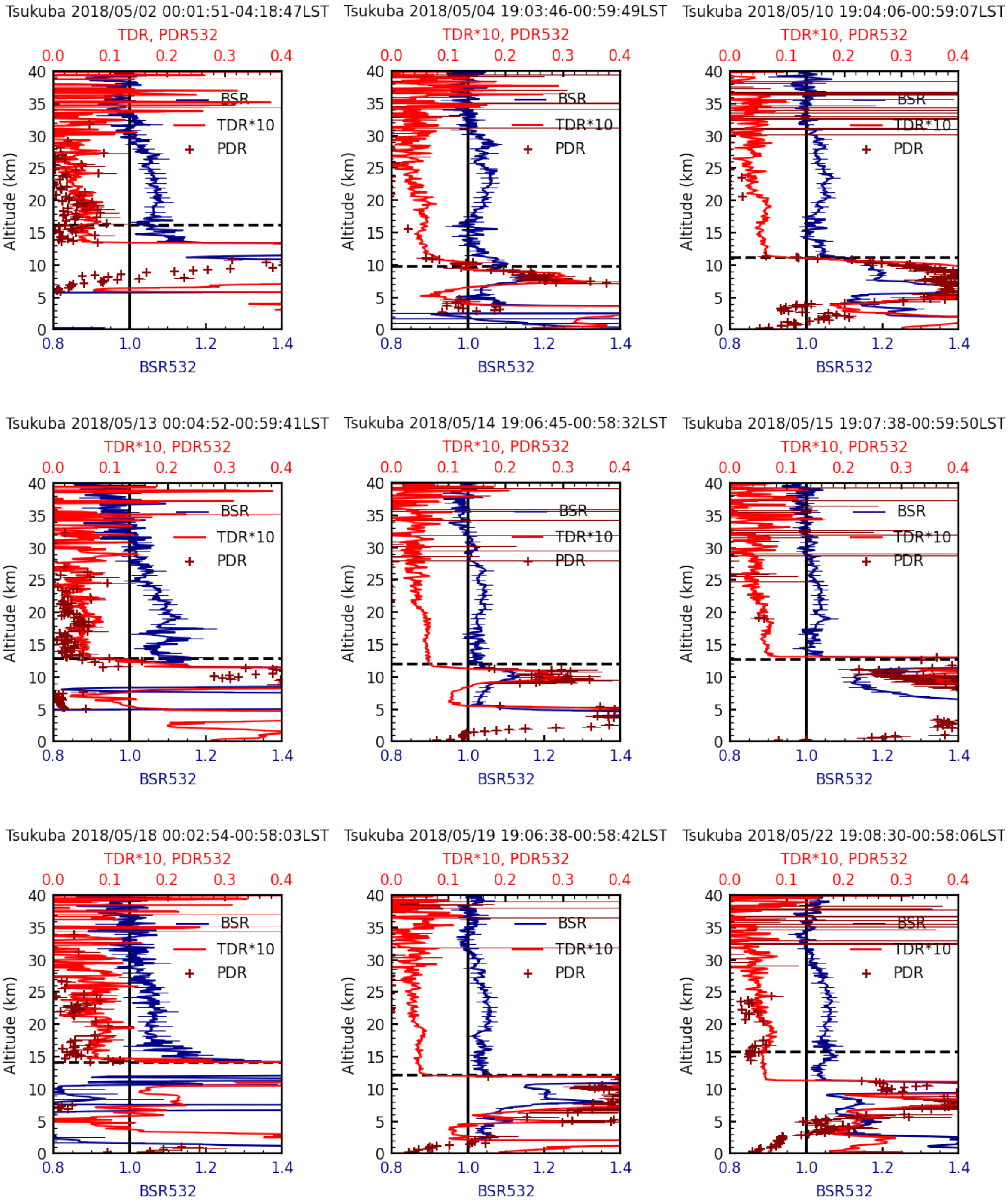
4) P5/L152: Is it possible that cirrus signal overlays the aerosol signal, so that cirrus and aerosol can coexist simultaneously? Or can you exclude this with your method?

Yes, it is always possible that cirrus signal overlays the aerosol signal if they coexist because of the larger backscattering cross section of cirrus particles. In that case, the BSR and PDR values would become those for cirrus particles.

5) P9/Fig.3: You could add a BSR profile from pre- or post-monsoon to show the difference. The difference can be use to better highlight the signal in BSR from the ATAL.

Figures R3-1 and R3-2 below show the lidar profiles at Tsukuba on some days in May 2018 and in October 2018, respectively. These figures show that BSR in these months did not show enhancements of >1.1 like those found in August and September (Figs. 2c, e). However, we note that TDR slightly increased below 20 km, suggesting a possibility of presence of minute amount of non-spherical particles. The origins of the particles are unknown and a subject of our future study.

Figure R3-3 below shows the lidar profiles at Fukuoka on some days in May-June 2018 and in October-November 2018. Again, we did not detect depolarization enhanced layers (with depolarization ratio higher than 2%) in these months.



Tsukuba 2018/05/02 00:01:51-04:18:47LST
TDR, PDR532
Tsukuba 2018/05/04 19:03:46-00:59:49LST
TDR*10, PDR532
Tsukuba 2018/05/10 19:04:06-00:59:07LST
TDR*10, PDR532
Tsukuba 2018/05/13 00:04:52-00:59:41LST
TDR*10, PDR532
Tsukuba 2018/05/14 19:06:45-00:58:32LST
TDR*10, PDR532
Tsukuba 2018/05/15 19:07:38-00:59:50LST
TDR*10, PDR532
Tsukuba 2018/05/18 00:02:54-00:58:03LST
TDR*10, PDR532
Tsukuba 2018/05/19 19:06:38-00:58:42LST
TDR*10, PDR532
Tsukuba 2018/05/22 19:08:30-00:58:06LST
TDR*10, PDR532
BSR
TDR*10
PDR
Altitude (km)
BSR532

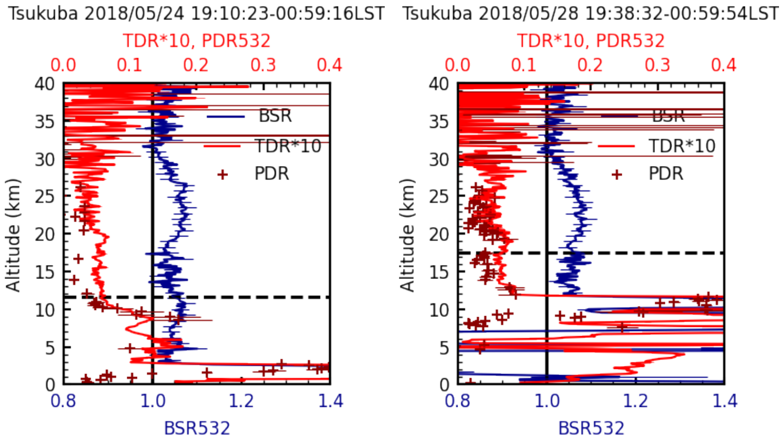
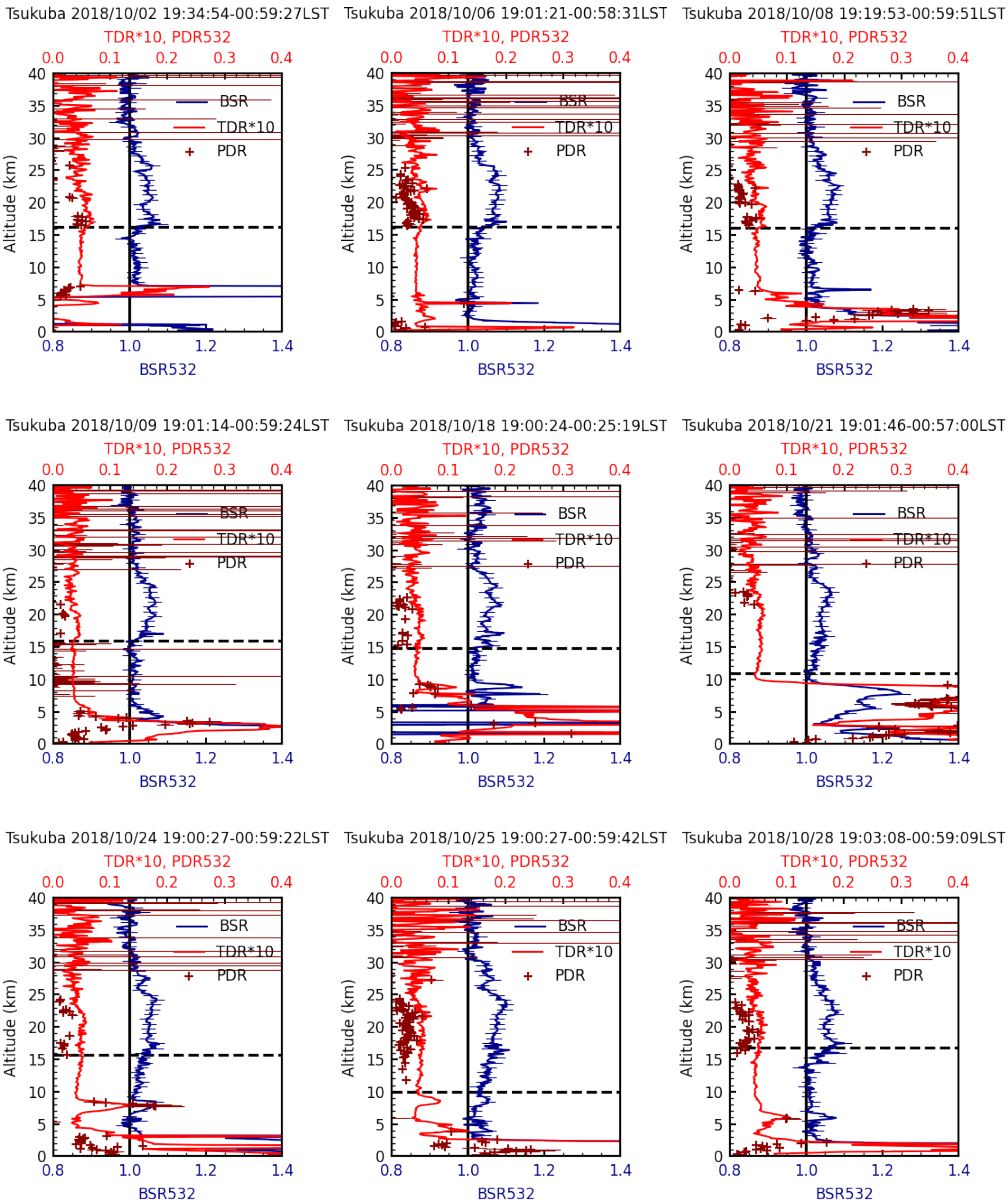


Figure R3-1. Lidar profiles taken at Tsukuba on May 2, 4, 10, 13, 14, 15, 18, 19, 22, 24, and 28, 2018. The horizontal dashed line in each panel indicates the location of the first lapse rate tropopause. It is noted that for "TDR*10" = 0.05 means TDR = 0.05/10 = 0.005, i.e., 0.5%TDR. This is close to the depolarization ratio value, 0.366% for air molecules.



Tsukuba 2018/10/02 19:34:54-00:59:27LST
Tsukuba 2018/10/06 19:01:21-00:58:31LST
Tsukuba 2018/10/08 19:19:53-00:59:51LST
Tsukuba 2018/10/09 19:01:14-00:59:24LST
Tsukuba 2018/10/18 19:00:24-00:25:19LST
Tsukuba 2018/10/21 19:01:46-00:57:00LST
Tsukuba 2018/10/24 19:00:27-00:59:22LST
Tsukuba 2018/10/25 19:00:27-00:59:42LST
Tsukuba 2018/10/28 19:03:08-00:59:09LST
TDR*10, PDR532
Altitude (km)
BSR532
BSR
TDR*10
PDR

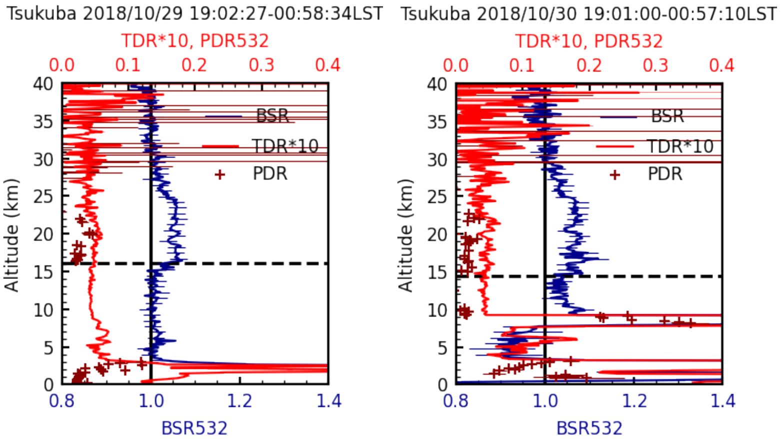


Figure R3-2. As for Figure R1-1, but for October 2, 6, 8, 9, 18, 21, 24, 25, 28, 29, and 30, 2018.

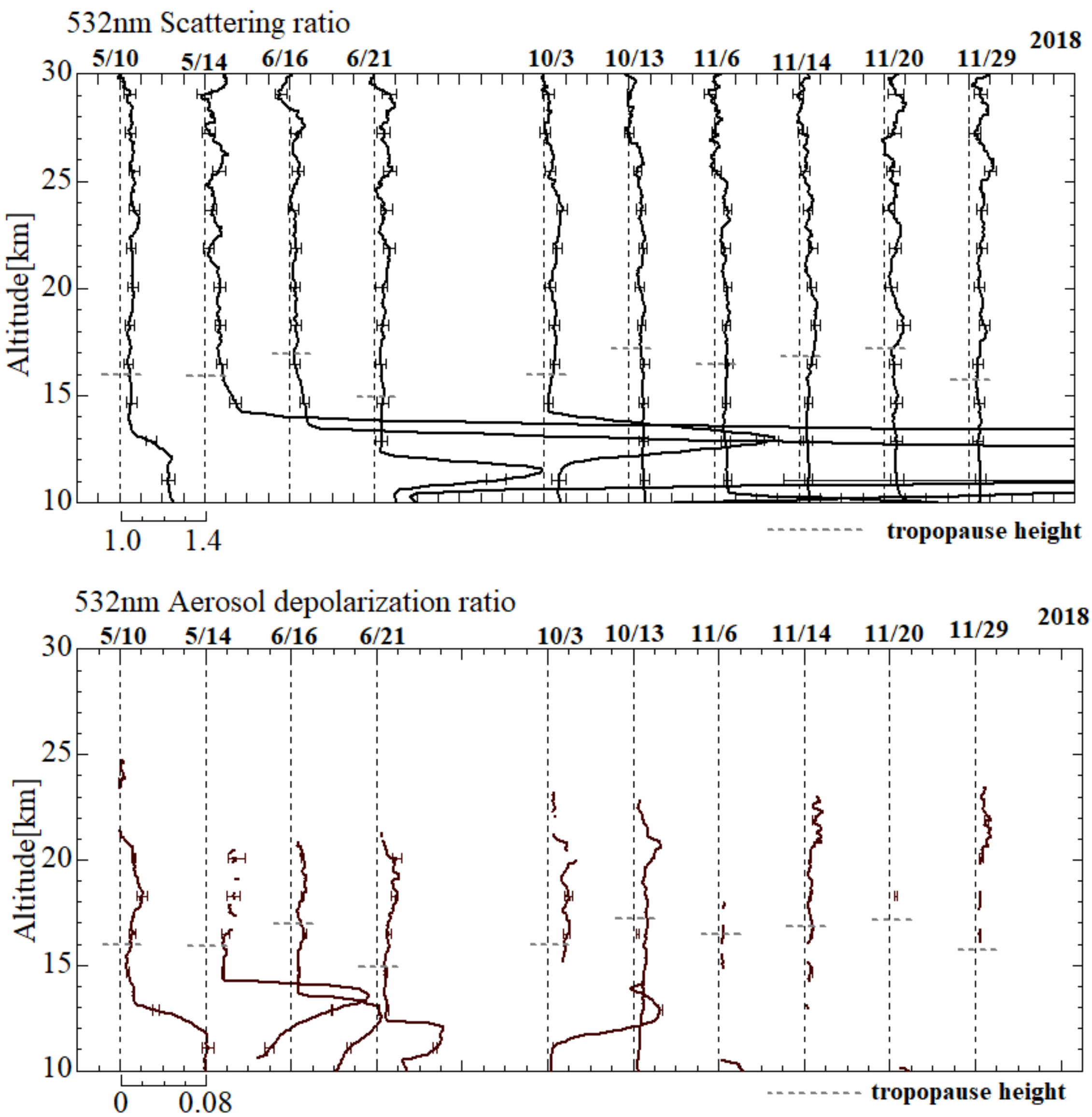


Figure R3-3. Lidar profiles taken at Fukuoka on the days in May-June 2018 and in October-November 2018 when the lidar was operated.

6) P10/L204: '... whereas those without enhanced aerosol particles tend to originate from edge regions surrounding the anticyclone.' and from the extratropical lower stratosphere. Right?

At the northern edge regions, yes. But, at the southern edge regions, they are from tropical lower stratosphere.

Why do you use only ten-days backward trajectories? What about 15- or 20-day backward calculations? In somewhat longer trajectories, the difference between air masses from the core the anticyclone or from the edge (or outside from the anticyclone) should be more pronounced.

Figure R3-4 below shows the 15-day backward trajectories. We had found that if we plot longer trajectories, we have more trajectories within the ASM anticyclone, although the density is still lower for the cases without enhanced aerosols measured at Tsukuba/Fukuoka. Also, we had received a comment before the paper submission that longer trajectories are less reliable. Thus, we decided to show the results from 10-day backward trajectories which better shows the differences.

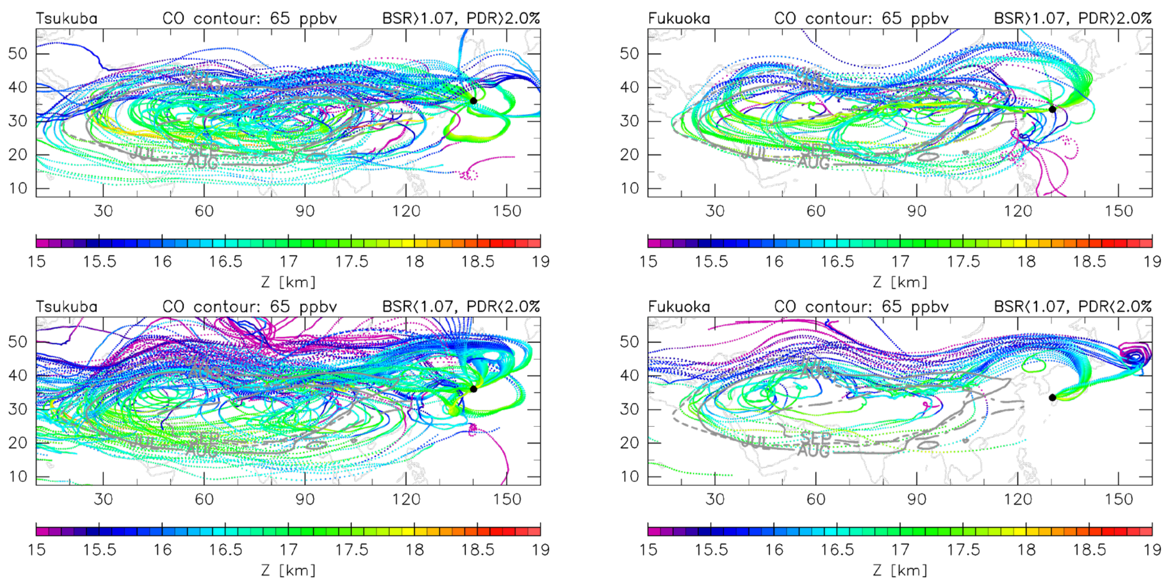


Figure R3-4. As for Figures 4 (for Tsukuba, for the left two panels) and 5 (for Fukuoka, for the right two panels), but for 15-day backward trajectories. (Note that the colour code for the geopotential height has been revised (to narrow the range of Z); please see the next QA.)

7) P11/Fig.4: Is it possible to adjust the color bar more to the Z range of the trajectories to better highlight the gradients along the trajectories. The bluish colors are only used for one trajectory over the Pacific in Fig. 4b. It looks like that this trajectory is influenced by a tropical cyclone. If that is true that could be mentioned as a side remark.

The colored 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 reply 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.”

8) P13/L230 : ’PV can be regarded as a dynamical tracer, with lower values in the ASM anticyclone along the same latitudes (e.g., 30°N), although background positive gradients in latitude and its noisier nature give more complicated features.’

PV can be very useful to see the edge of the Asian monsoon anticyclone at around 380K (e.g. Ploeger et al., 2015), above around 400K as shown in Fig. 6, the PV is not so useful. Instead you could try to use the (Montgomery) stream function or the geopotential height.

Ploeger, F., Gottschling, C., Grießbach, S., Grooß, J.-U., Günther, G., Konopka, P., Müller, R., Riese, M., Stroh, F., Tao, M., Ungermann, J., Vogel, B., and von Hobe, M.: A potential vorticity-based determination of the transport barrier in the Asian summer monsoon anticyclone, Atmos. Chem. Phys., 15, 13 145–13 159, https://doi.org/doi:10.5194/acp-15-13145-2015, 2015.

Thank you very much for your comment and suggestion. We will change from PV to Montgomery streamfunction. Please see the response letter to Referee #2 for the revised Figure 6. We will add the following sentences in the second paragraph of Section 2.2:

"Carbon monoxide (CO), temperature (T), and geopotential (Φ) data are analysed in this paper."

"Montgomery streamfunction (MSF), defined as MSF = c_p T + Φ, where c_p is specific heat of dry air at constant pressure, in isentropic coordinates corresponds to geopotential (height) in pressure coordinates (e.g., Amemiya and Sato, 2018) , and thus is a good dynamical indicator of the ASM anticyclone. Potential vorticity (PV) on isentropic surfaces (e.g., at 360–380 K) is often used as a dynamical tracer in studies of the ASM anticyclone (e.g., Popovic and Plumb, 2001; Garny and Randel, 2013; Ploeger et al., 2015; Amemiya and Sato, 2018); however, PV at and above 400 K (this isentropic surface will be focused in Section 3.2) is not very useful to analyse the ASM anticyclone boundary. Thus, we will analyse MSF at 400 K surface calculated from CAMS data."

Also, we will revise the second paragraph of Section 3.2 accordingly.

9) P16/Fig.8: Why do you show H2O from MLS and not CO from MLS? CO would be a better chemical tracer for transport as H2O which is in addition affected by microphysics. You could also use MLS O3 which should be anticorrelated to CO (low O3 in the anticyclone and high O3 in the lower stratosphere).

Figures R3-5 and R3-6 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.

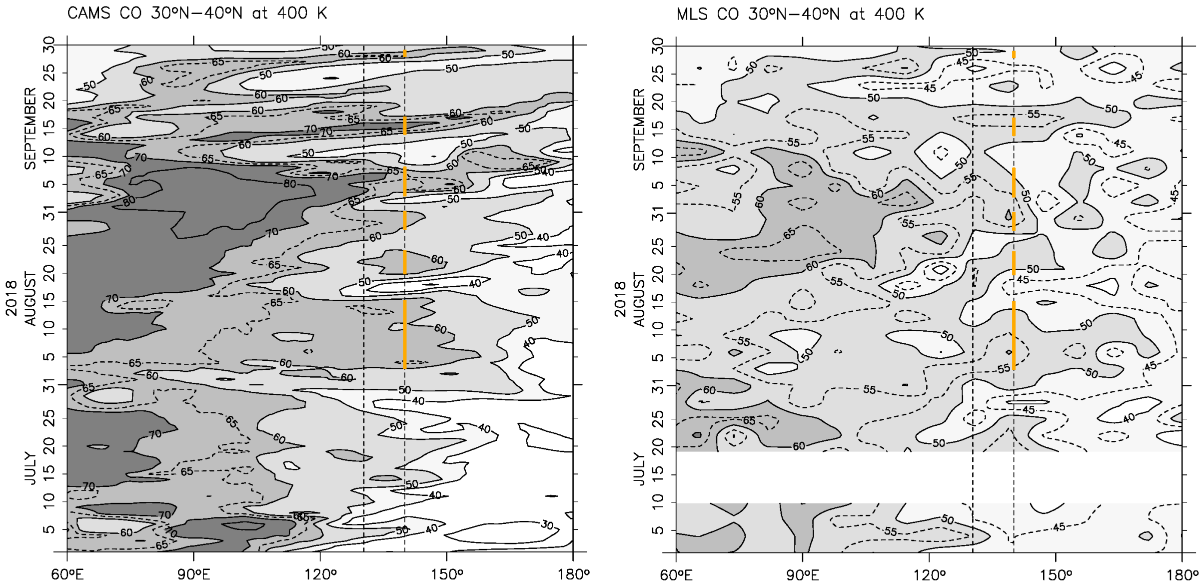


Figure R3-5. (Left) Same as Figure 7 (the revised version). (Right) Same as left but for MLS CO data. Data for the 30°N–40°N region have been aggregated into 3-day and 8°-longitude bins, each constituting about 10 individual data points. The contours for 45 ppbv and 55 ppbv as well as 65 ppbv are added as dotted lines.

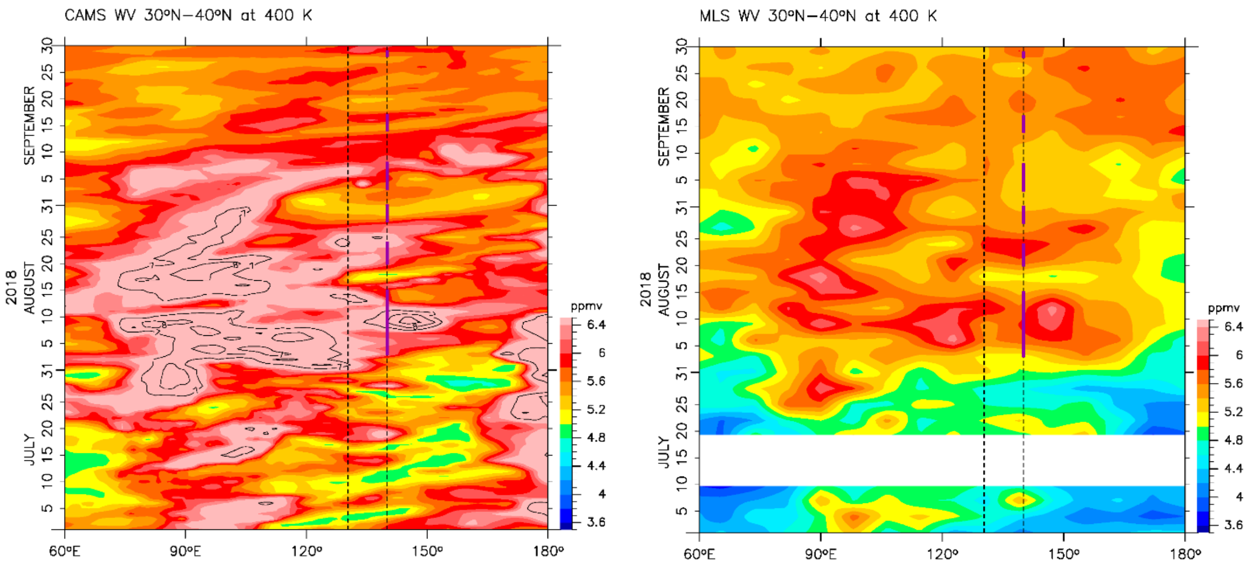


Figure R3-6. (Left) Longitude–time distribution of daily averaged water vapor volume mixing ratio at 400 K potential temperature averaged over 30°N–40°N, using CAMS reanalysis specific humidity data. The contour lines for 7, 8, 9, 10, 11 ppmv have been added. (Right) Same as Figure 8 (the revised version).

Two figures for ozone from CAMS and MLS are shown below. Figure R3-7 shows horizontal distributions of daily ozone at 400 K during 18–23 August 2018 using CAMS reanalysis data, similar to Figure 6. Figure R3-8 shows longitude–time distribution (Hovmöller diagram) of ozone at 400 K averaged over 30°N–40°N using CAMS reanalysis and MLS data. We see that ozone and CO are anticorrelated around the northern edge of the ASM anticyclone (in other words, along the westerly jet stream). In particular, around 16–19 August, a strong stratosphere-to-troposphere intrusion occurred over Japan, which are clearly observed as a strong ozone enhancement event. Around the southeastern and southern part of the ASM anticyclone, however, the relationship between ozone and CO becomes less clear (i.e., not always clearly anticorrelated); ozone concentration is often still relatively high in CO enhanced regions there. This is probably in part due to the transport from the north (i.e., ozone of stratospheric origin) and in part due to upward transport from Asian countries where ozone is photochemically produced (i.e., ozone of tropospheric origin). Thus, in this paper, we decided not to show ozone results as the interpretation is more complicated than CO and water vapour.

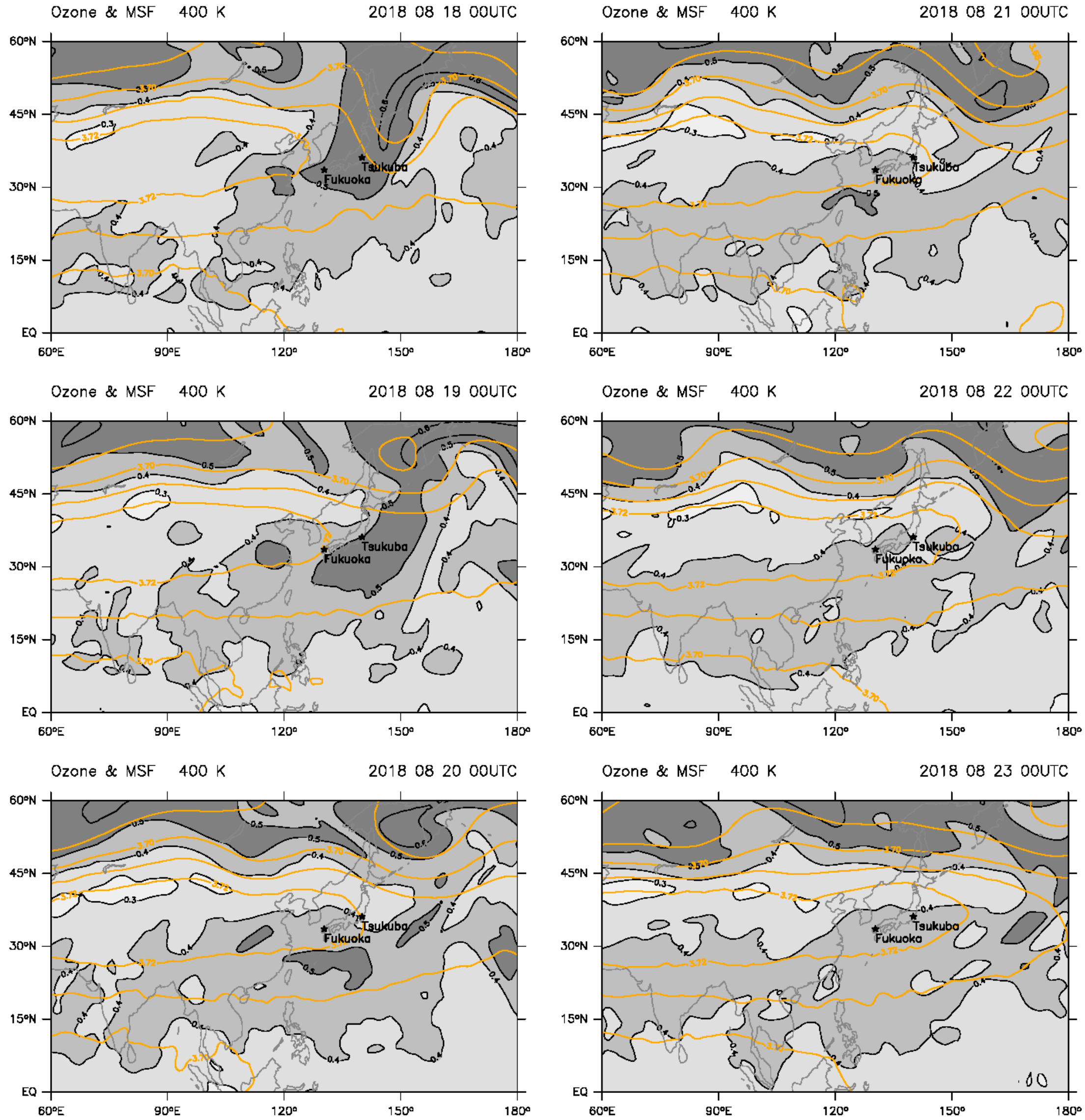


Figure R3-7. As for the revised Figure 6, but for ozone volume mixing ratio (black contours with grey tone, with intervals of 0.1 ppmv) from CAMS reanalysis data. CAMS ozone data are originally provided in mass mixing ratio, kg kg^{-1}, which are converted to volume mixing ratio, ppmv, for this response letter.

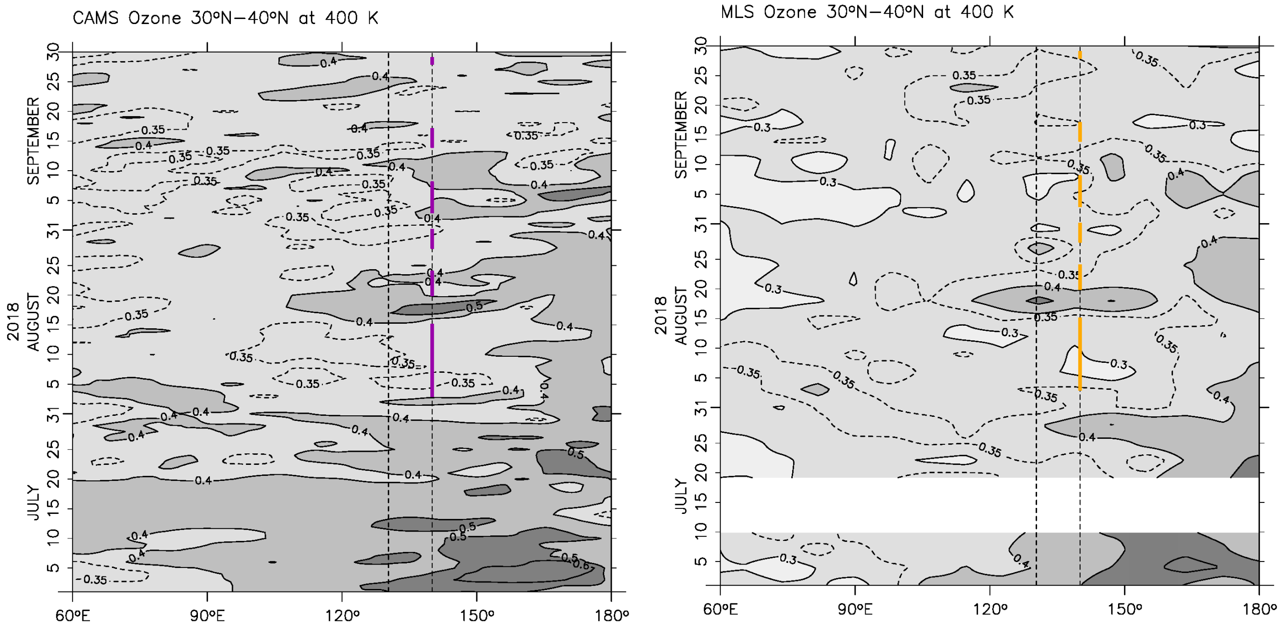


Figure R3-8. (Left) Same as Figure 7 (the revised version), but for CAMS ozone in ppmv. The contour interval is 0.1 ppmv, with 0.35 ppmv contours added (dotted). (Right) Same as left but for MLS ozone data. Data for the 30°N–40°N region have been aggregated into 3-day and 8°-longitude bins, each constituting about 10 individual data points.

10) P20/L331: 'The PDR values obtained at Tsukuba, i.e., ~5% (3%–10%) suggest that these enhanced particles are solid particles, rather than spherical, liquid H2SO4 particles (PDR ~0%) or cirrus ice particles (PDR > 25%–30%). The observed values may be consistent with those of solid NH4NO3 particles recently suggested by Höpfner et al. (2019).'

Using the particle depolarization ratio, the study shows that the aerosol particles are most likely solid and it is concluded that the aerosol particles possibly contain NH4NO3. In the literature it is discussed that also carbonaceous aerosols, dust, nitrate-containing aerosol, black carbon and organic carbon could contribute to the chemical composition of the ATAL. Can you exclude with your measurements such types of aerosol particles? Please clarify this point.

The lidar measurements cannot exclude the co-existence/existence of other types of aerosol particles. We will add the following sentence: "However, it should be noted that the lidar BSR and PDR measurements cannot exclude the possibility of co-existence of other types of solid aerosol particles such as mineral dust, black carbon, and some types of carbonaceous aerosols which are solid."

Minor comments:

1) P3/L84: (senkrecht in German) –> (= "senkrecht" in German) ?

Will be added.

2) P5/L37: remove large white spaces

This will be handled at the type-setting phase (if this paper is accepted).

3) P13/L223: ’Horizontal distributions of CO and PV’ add ’from CAMS’

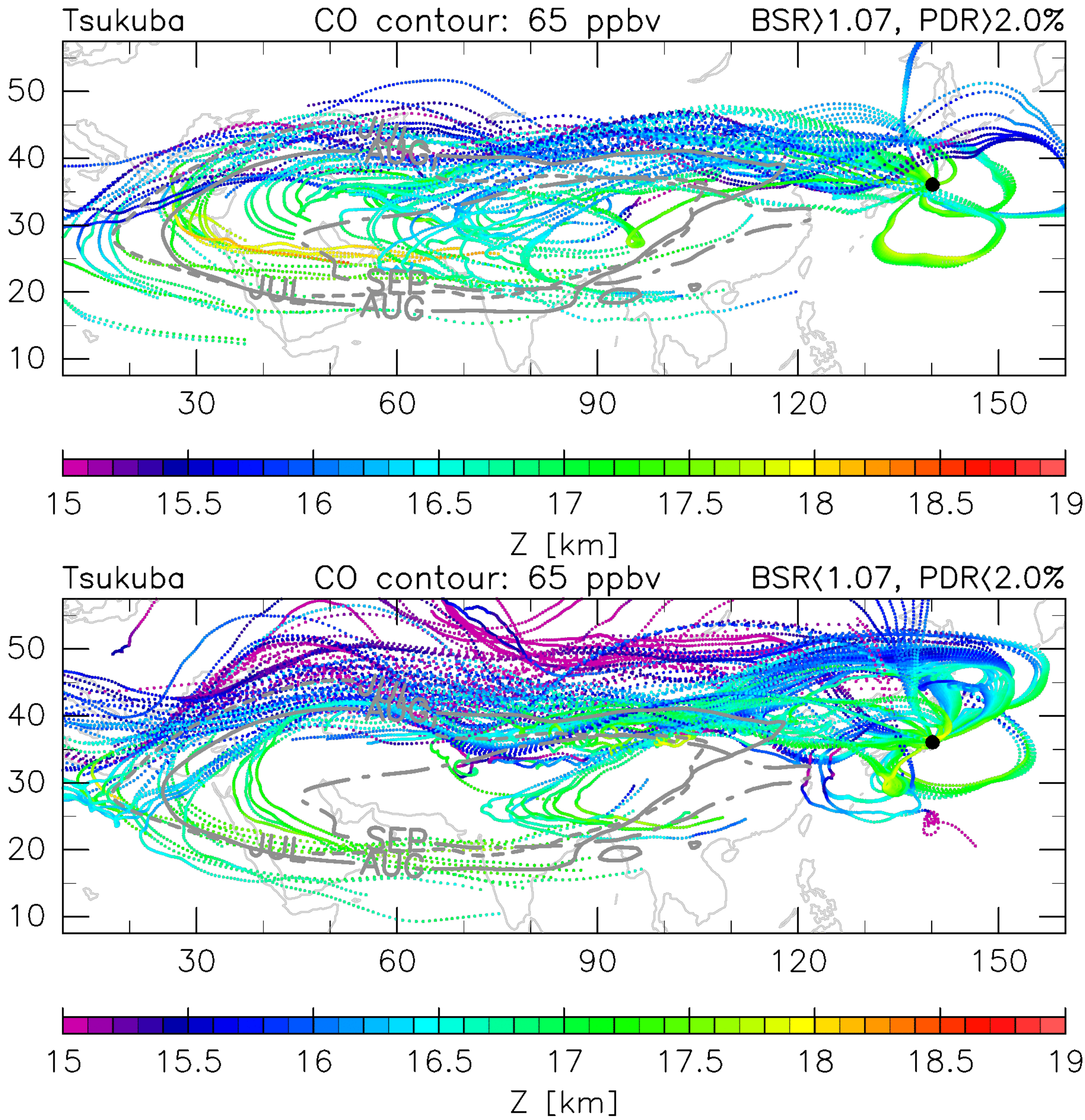
Will be added.

4) P13/L233: ’are shown in Figure 7’ -> ’are shown as Hovmöller diagrams in Fig. 7’

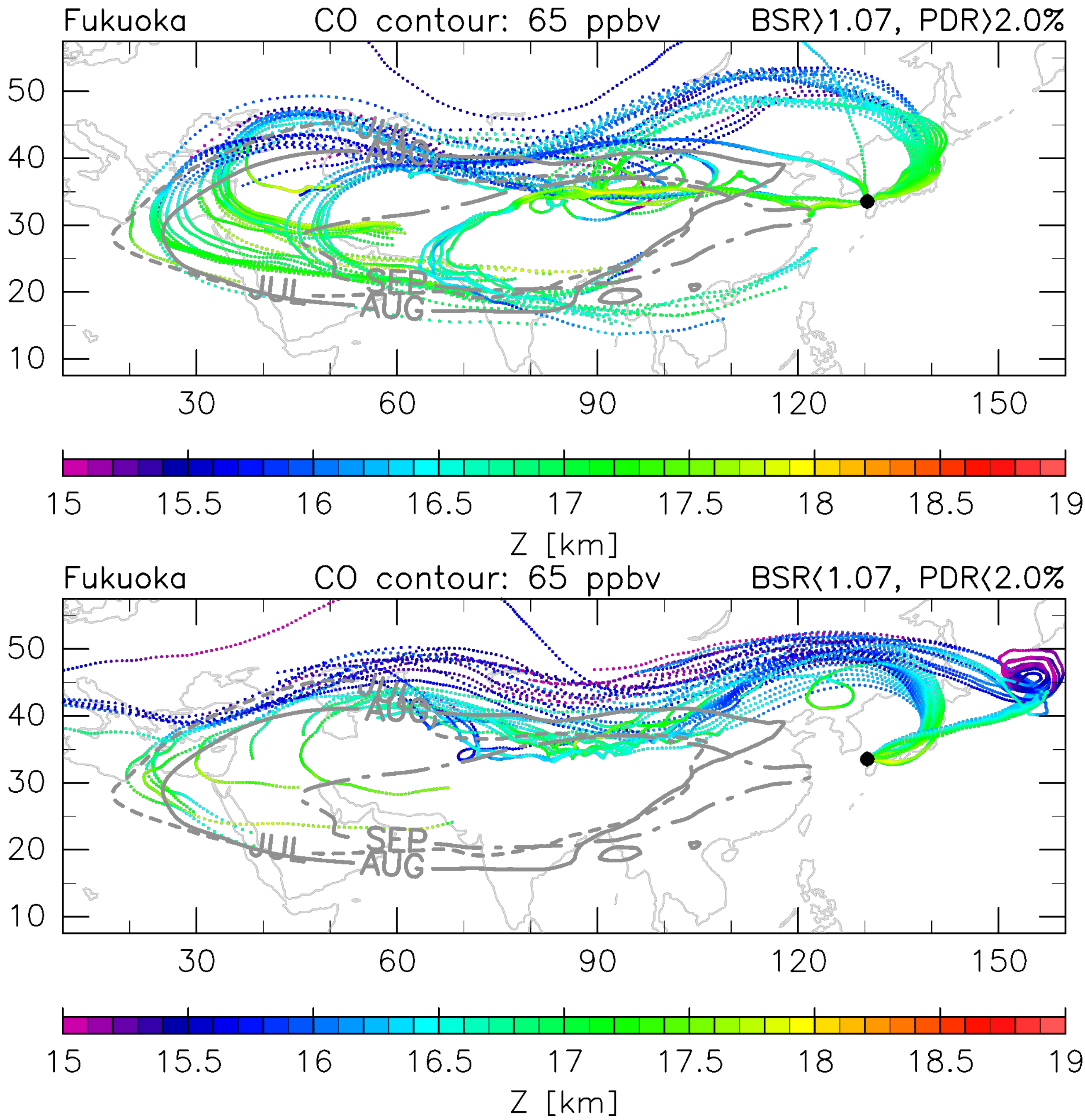
We chose not to use the term “Hovmöller diagram” because this may not be understandable for some of the readers of the journal Atmospheric Chemistry and Physics. But, we are happy to make the change if the editor recommends to do so.

5) Fig.7/8: You could say that the Figures are ’Hovmöller diagrams’

Please see above.



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).