Response to comments by Anonymous Referee #1

Thank you very much for your review.

In this manuscript, Fujiwara and co-authors present observational data from two lidars in Japan indicating the presence of aerosol particles from the Asian Tropopause Aerosol Layer (ATAL) over the stations just above the tropopause in August and September 2018. The origin of these particles from inside the Asian monsoon anticyclone is indicated by trajectory analyses as well as CAMS reanalysis data of CO and MLS satellite observations of water vapor. Further, the authors exclude the influence of volcanic eruptions and forest fires by inspecting the global condition of aerosols in the UTLS through space borne limb-scatter (OMPS LP) and lidar (CALIOP) observations.

A central point of this work is the depolarization of the observed particles of around 5%. This indicates that at least a part of the aerosols was non-spherical (i.e. not liquid) but solid, however, not as strongly depolarizing as cirrus particles.

In general, the manuscript is well written and logically organised with clear figures. It presents a novel dataset on aerosols from the ATAL embedded in the interpretation of the general atmospheric situation. To my knowledge it is also the first one of an eastward shedding event over Japan and explicitly including the particle depolarization. Therefore, I recommend publication in ACP after taking into account the comments below.

Thank you very much for your evaluation.

Specific comments

L97-L110, Lidar error estimation:
In L98 the BSR uncertainties are stated as 2-3%, but in L106, additional BSR errors are discussed. I would suggest to clearly state first all error terms for the BSR (random and possible systematic ones) and then include those in the discussion on PDR.

This sentence will be revised as follows:
"The BSR uncertainties were estimated as follows. The random component was estimated from the photon counts of the backscatter signals at 532 nm after temporal and vertical averaging by assuming Poisson statistics. Other sources of BSR uncertainties (biases) were estimated by assuming the uncertainty of the normalization value of BSR with 8.5×10^{-3} (Russell et al., 1979, 1982) and that of the extinction-to-backscatter ratio with 30 sr (Jäger and Hofmann, 1991; Jäger et al., 1995). The total uncertainty of BSR were then estimated to be 2–3 % typically around the tropopause."

References:

Jäger, H., and Hofmann, D.: Midlatitude lidar backscatter to mass, area, and extinction conversion model based on in situ aerosol measurements from 1980 to 1987, Appl. Opt., 30(1), 127, https://doi.org/10.1364/ao.30.000127, 1991.

Jäger, H., Deshler, T., and Hofmann, D. J.: Midlatitude lidar backscatter conversions based on balloonborne aerosol measurements, Geophys. Res. Lett., 22(13), 1729–1732, https://doi.org/10.1029/95GL01521, 1995.

Russell, P. B., Swissler, T. J., and McCormick, M. P.: Methodology for error analysis and simulation of lidar aerosol measurements, Appl. Opt., 18(22), 3783, https://doi.org/10.1364/ao.18.003783, 1979.

Russell, P. B., Morley, B. M., Livingston, J. M., Grams, G. W., and Patterson, E. M.: Orbiting lidar simulations 1: Aerosol and cloud measurements by an independent-wavelength technique, Appl. Opt. 21(9), 1541, https://doi.org/10.1364/ao.21.001541, 1982.

L135, ‘in this data product, clouds and Polar Stratospheric Clouds (PSCs) have been removed’:
Please add the information, how cirrus clouds have been removed.

The algorithms to classify the CALIOP backscatter signals into various types of clouds and aerosols, presented by Young and Vaughan (2009), are quite complicated, but in short, they are based on the particulate extinction-to-backscatter (lidar) ratio and the multiple-scattering factor profile. At the end of the sentence, we will add “based on the information of particulate extinction-to-backscatter (lidar) ratio and the multiple-scattering factor profile”. Also, we will add more recent publication by Kim et al. (2018).

Reference:
Kim, M.-H., Omar, A. H., Tackett, J. L., Vaughan, M. A., Winker, D. M., Trepte, C. R., Hu, Y., Liu, Z., Poole, L. R., Pitts, M. C., Kar, J., and Magill, B. E.: The CALIPSO version 4 automated aerosol classification and lidar ratio selection algorithm, Atmos. Meas. Tech., 11, 6107–6135, https://doi.org/10.5194/amt-11-6107-2018, 2018.

L145:
Please discuss also the event around Aug, 9th, over Tsukuba since there has been a clear eastward shedding as can be seen in Figs. 7 and 8. It is not clear to me if the particles were above or below the tropopause since there is quite a strong change in tropopause height visible by the red dots in Fig. 1.

We will add the following sentence just after describing the 18–26 August case:
“We also observe another strong event around 9 August at 15–17 km, although missing observations before and after this date prevent from characterizing the temporal scale of the event; furthermore, the tropopause height was highly variable around this date and was located at 17 km on that date, situating the aerosol enhanced layer temporarily in the troposphere.”
We will also add this information to Section 3.2 in the discussion for Figure 7 as “. . . in the fact that only the 20–25

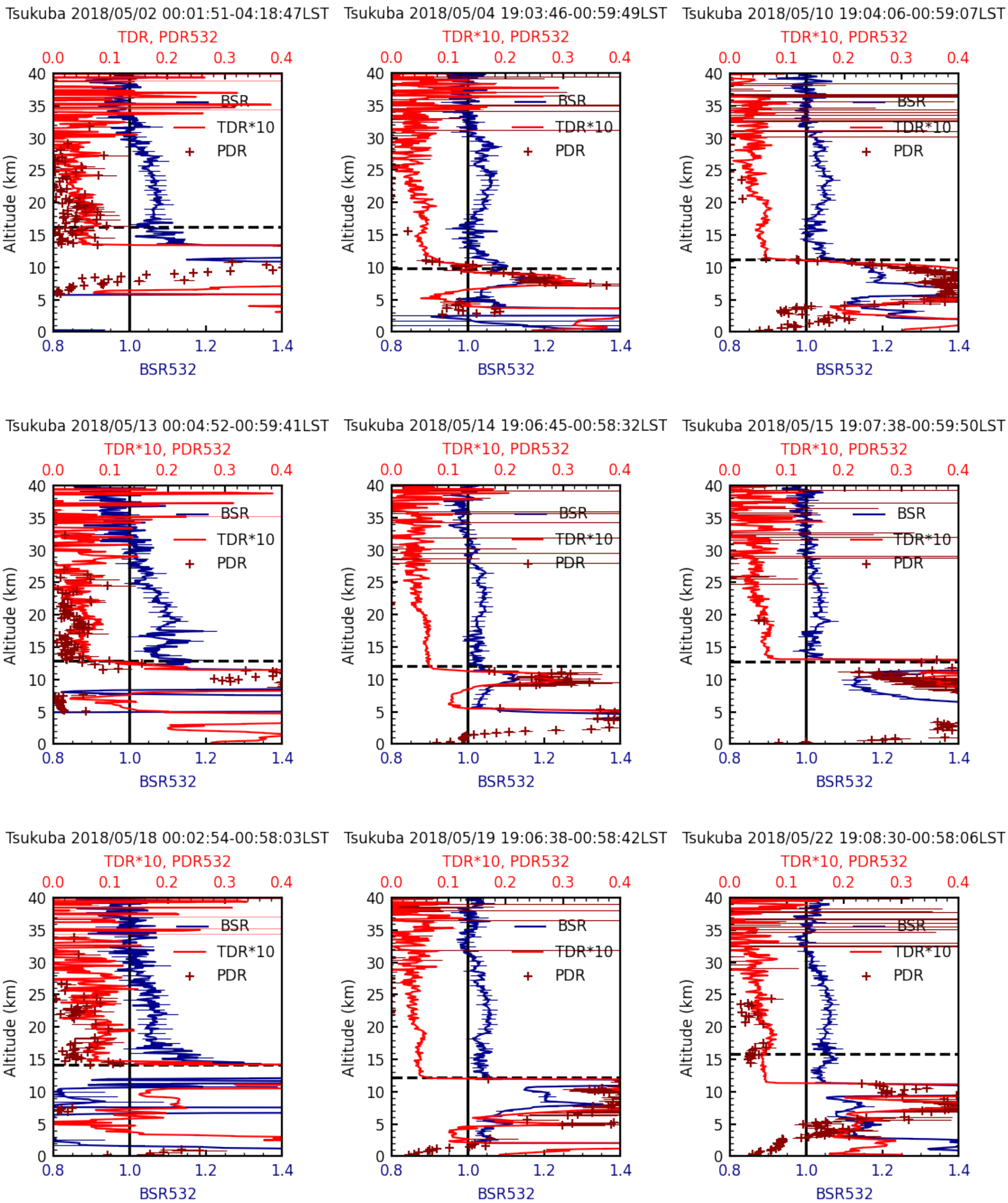
August event was relatively well observed, with the 5–15 August event being captured only on 9 August."

L181, 'PDR values of 1%–3%':

The difference between the PDR values between Tsukuba, showing clearly enhanced signals, and Fukuoka is tentatively explained by the different measurement periods. In case of Tsukuba, quasi no event has been shown with values of PDR less than 2 (see Fig. 2), while there are many above Fukuoka. Please discuss whether this might hint to some unidentified bias in one of the instruments. It would e.g. be informative to present some observations before June or after September where both instruments show consistently low/high values of PDR.

The reason why PDR values less than 2% were rare in the plot for Tsukuba (Fig. 2) is that we do not plot the data with "relative" uncertainty of PDR larger than 30%; this treatment resulted in removing data points with BSR values lower than ~1.05 where background spherical sulphate particles (with PDR values of <2%) were presumably predominant. We will note this treatment for Tsukuba data in the revised manuscript. Figures R1-1 and R1-2 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 R1-3 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
0.0 0.1 0.2 0.3 0.4
Altitude (km)
0 5 10 15 20 25 30 35 40
BSR
TDR*10
PDR
0.8 1.0 1.2 1.4
BSR532

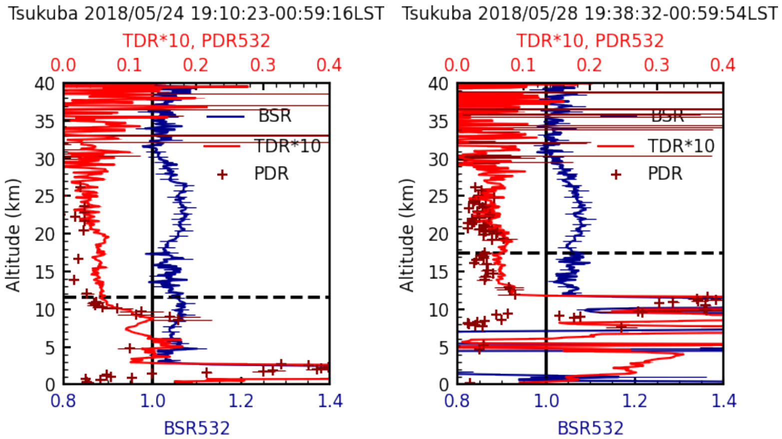
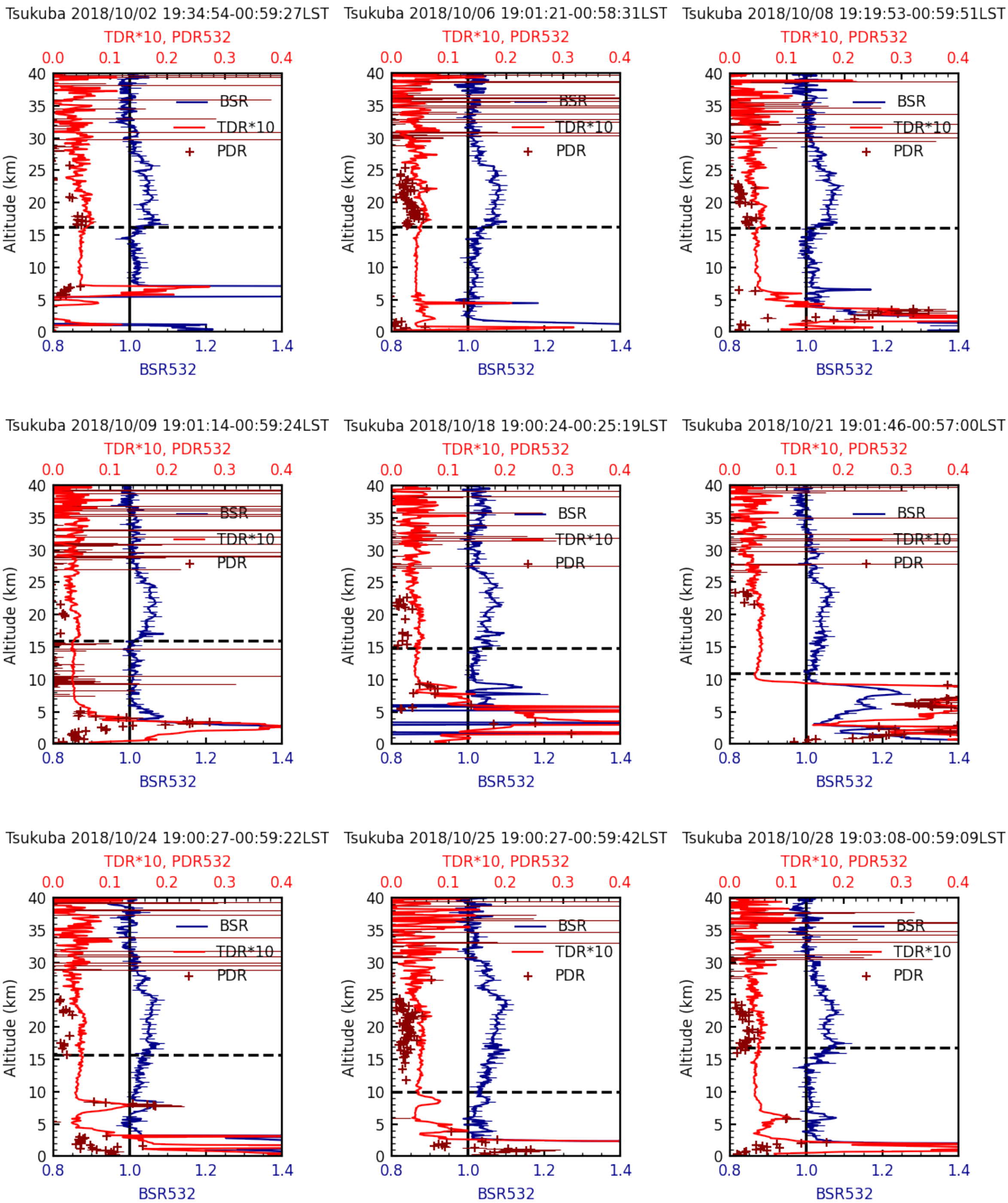


Figure R1-1. Lidar profiles taken at Tsukuba on May 2, 4, 10, 13, 14, 15, 18, 19, 22, 24, and 28, 2018. The horizontal dashed line in each panel indicates the location of the first lapse rate tropopause. It is noted that for "TDR*10" = 0.05 means TDR = 0.05/10 = 0.005, i.e., 0.5%TDR. This is close to 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
0.0
0.1
0.2
0.3
0.4
Altitude (km)
0
5
10
15
20
25
30
35
40
BSR
TDR*10
PDR
0.8
1.0
1.2
1.4
BSR532

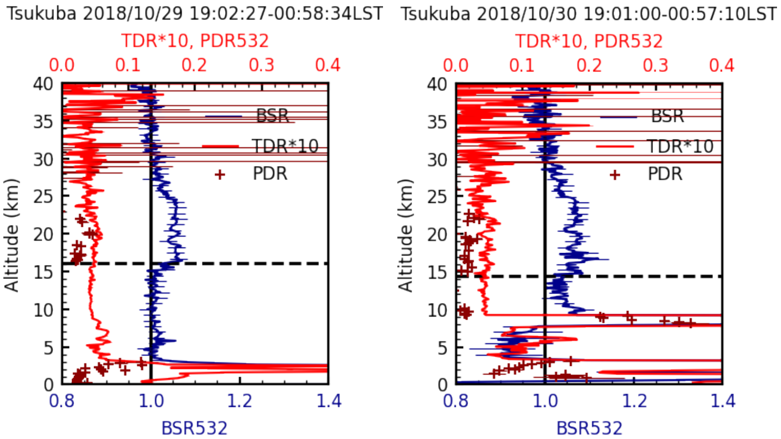


Figure R1-2. As for Figure R1-1, but for October 2, 6, 8, 9, 18, 21, 24, 25, 28, 29, and 30, 2018.

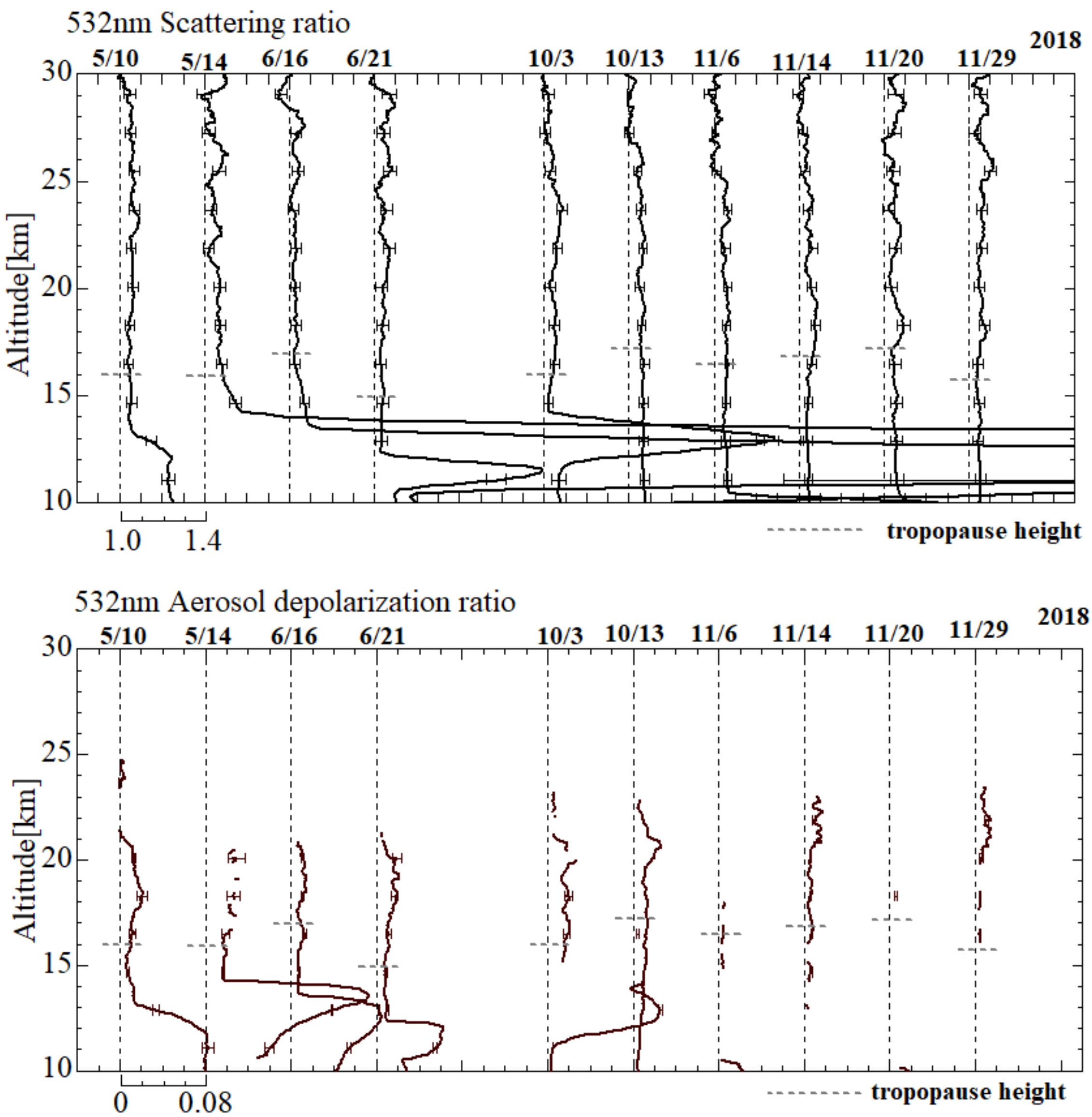


Figure R1-3. Lidar profiles taken at Fukuoka on the days in May-June 2018 and in October-November 2018 when the lidar was operated.

L196, '3.2 Trajectories and airmasses':

From the trajectories shown, it is not clear if they reach altitudes below the tropopause. Could you provide any discussion on this point?

Following the comments by other reviewers, 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 response letter.

We will revise the sentence,

"They also indicate that airmasses with enhanced aerosol particles at this height tend to originate in regions within the ASM anticyclone, whereas those without enhanced aerosol particles tend to originate from edge regions surrounding the anticyclone."

as:

"They also indicate that airmasses with enhanced aerosol particles at this height tend to originate in regions within the ASM anticyclone at the altitudes 16.5–18 km, i.e., around or just below the tropopause, whereas those without enhanced aerosol particles tend to originate from edge regions surrounding the anticyclone." (i.e., the underlined part will be added.)

L240:

I would be interested if the CAMS CO data could be supported by MLS measurements of CO. This should be easy by providing a figure similar to Fig. 8 but for MLS CO.

Thank you for this suggestion. Please see Figure R1-4 below. We see that CAMS CO data are roughly ~10 ppbv greater than MLS CO over Japan during August–September 2018, but also that eastward extension signals coming over Japan agree fairly well qualitatively within the differences in spatio-temporal sampling of the two data sets. We will add this note to Section 2.2.

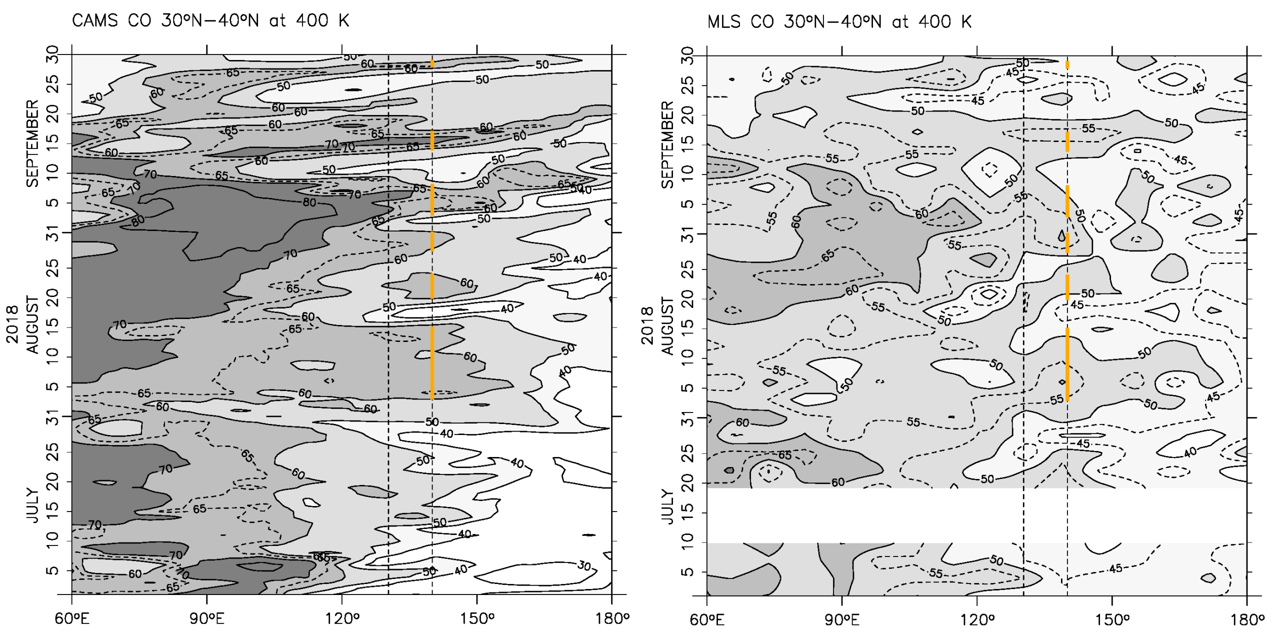


Figure R1-4. (Left) Same as Figure 7 (the revised version following Referee #2's suggestion: Coloured line segments have been added for the periods when CO concentration was ≥~60 ppbv along the longitude of Tsukuba (i.e., 3–15, 20–24, and 28–31 August, and 3–8, 14–17, and 28–29 September)). (Right) Same 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.

L283:

Is any direct comparison/match of the ground based lidars with the CALIOP lidar possible during the relevant time period?

We have looked at the CALIOP lidar data on 21 August 2018 when the lidar at Tsukuba (36.1°N, 140.1°E) observed the strong signal and when the CALIPSO flew over the region relatively close to Tsukuba: https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_v4_detail.php?s=production&v=V4-10&browse_date=2018-08-21&orbit_time=03-36-39&page=3&granule_name=CAL_LID_L1-Standard-V4-10.2018-08-21T03-36-39ZD.hdf

As you see, we do not observe the corresponding signals around the orbital track (31.71°N, 139.38°E) to (37.78°N, 137.66°E) around 15.5–18 km in CALIOP data. Young and Vaughan (2009) noted for the CALIOP measurements, "The usual method of increasing the SNR (signal-to-noise ratio) is to average many profiles. However, along-track inhomogeneities in the atmospheric features, combined with the high speed (typically 7.5 km s^{-1}) of the satellite across these features and the relatively low firing rate of the laser (~20 s^{-1}), lead to situations where it is simply not possible to acquire a sufficient number of profiles before the subsatellite atmosphere changes significantly." Note also that the ground-based lidar data from Tsukuba (Fukuoka) have been averaged for 3 (4) hours in this study. We think that appropriate quality control (e.g., taking only high-quality nighttime measurements) and spatio-temporal averaging for CALIOP data might give us ATAL eastward extension signals over Japan in a Hovmöller diagram like Figures 7 and 8, but this would be a potential future work.

L317:

For ATAL studies, the applied CALIOP filter on cirrus clouds has been a depolarization ratio threshold of 5% (e.g. Vernier et al., 2015): (1) why has a different limit been applied for the present ground based observations? (2) could you discuss which effect the finding of this work indicating 5% depolarization and more for ATAL particles would have on the CALIOP data analysis?

Regarding (1), Vernier et al. (2009) masked ice clouds by using their total depolarization ratio (TDR'), defined by the perpendicular to total (particular plus molecular) backscatter signal, and not by using PDR. In that case, TDR' values would be small if BSR values of cirrus are small. Theoretically, for example, TDR' would be less than 5% if BSR values are smaller than 1.2 for cirrus for the case that the PDR value is 35% (see Figure R1-5 below). In addition, averaging the data for a large grid volume (2° longitude × 1° latitude × 200 m height) could reduce BSR and TDR' values if cirrus clouds are partially present in the volume. Thus, we presume that they removed the data even if it contained some cirrus clouds.

Reference:

Vernier, J. P., Pommereau, J. P., Garnier, A., Pelon, J., Larsen, N., Nielsen, J., Christensen, T., Cairo, F., Thomason, L. W., Leblanc, T., and McDermid, I. S.: Tropical stratospheric aerosol layer from CALIPSO lidar observations, J.

Geophys. Res., 114, https://doi.org/10.1029/2009jd011946, 2009.

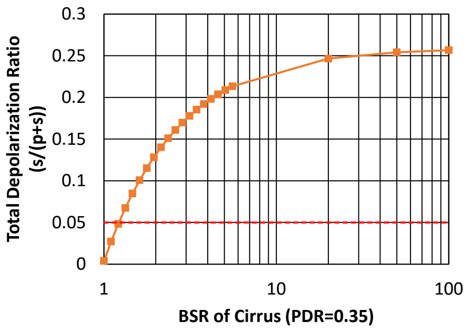


Figure R1-5. A theoretical total depolarization ratio (TDR') curve as a function of BSR for cirrus cloud particles with the PDR value of 35%. The horizontal dotted red line shows the threshold value of TDR' (5%) used in the papers by Vernier et al. (2009, 2015).

Regarding (2), we do not argue that future ATAL studies using CALIOP data should always use smaller threshold values. The choice of the threshold would depend on the purpose of each study. It is also possible that appropriate threshold numbers differ for different regions, e.g., either over the Tibetan Plateau or over Japan, in part because the chemical composition and crystal shape (if they are solid particles) may differ for different regions due to different life stages of the ATAL particles.

L327, 'with an average BSR value of 1.05 being a systematic feature':
Is there any information on the depolarization available from the OHP-lidar?

Unfortunately, the lidar at OHP has no capability of measuring the depolarization.

L337:
One may add the information from Fig. 2 in Wagner et al., 2020a, that the depolarization ratio obtained in the laboratory for solid ammonium nitrate particles was around 9%. Further, in Wagner et al., 2020b, from electron microscope images of ammonium nitrate particles Fig. 2c reveals 'that the crystalline AN particles are of rather compact shape with aspect ratios predominantly in the range from 0.80 to 1.25.'

Thank you very much for the information of these very important and relevant papers. We will modify the relevant part of the text. The revised, whole paragraph will be as follows:
"The PDR values obtained at Tsukuba, i.e., ~5% (3%–10%) suggest that these enhanced particles are solid particles, rather than spherical, liquid H_2SO_4 particles (PDR ~0%) or cirrus ice particles (PDR > 25%–30%). A recent laboratory experiment by Wagner et al. (2020a) showed the PDR values of ~9.5% for solid NH_4NO_3 particles at 488 nm. (Also, Wagner et al. (2020b) showed electron microscope images of solid NH_4NO_3 particles, which are "of rather compact shape with aspect ratios predominantly in the range from 0.80 to 1.25.") Thus, the values obtained with our lidars in Japan might be consistent

with those of solid NH_4NO_3 particles suggested by Höpfner et al. (2019). (Note that Sakai et al. (2010) investigated PDR values of other particle types at 532 nm in laboratory experiments; among these particles, sub-micrometre sea-salt and ammonium sulphate crystals (e.g., Plate 9 (pages 237–239) of Pruppacher and Klett, 1997) were found to have PDR values of ~8% PDR and ~4% PDR, respectively.) Small non-zero PDR values can occur if enhanced liquid H_2SO_4 particles and fresh ash particles from volcanic eruptions are mixed, although satellite data indicate this is less plausible (Sect. 3.3). Lidar measurements at Mauna Loa, Hawaii, indicated no signals from volcanic eruptions during the summer of 2018 (Chouza et al., 2020). Also, at the OHP lidar site in France, no enhancement in the lower stratospheric aerosol abundance was observed during the summer of 2018."

Technical corrections

Figure 4:
The CO isolines for different months cannot be distinguished easily. Perhaps use different line styles.

We will revise this figure as suggested. (Please see the revised version of these figures at the end of this response letter.)

L223:
Please add in this sentence the information 'from the CAMS reanalysis data'.

We will add this information.

L224, 'A potential temperature of 400 K corresponds to altitudes of 17.1 km at Tsukuba and 17.3 km at Fukuoka, on average, during July–September 2018':
This information should be provided before, e.g. where the trajectories are discussed.

This information will be moved to the first paragraph of Section 3.2.

L286, 'not have reached':
'have not reached'

Will be corrected.

L298, '. Rectangular':
'. The rectangular'

Will be corrected.

L341, ‘France, any enhancement’:
‘France, no enhancement’

Will be corrected.

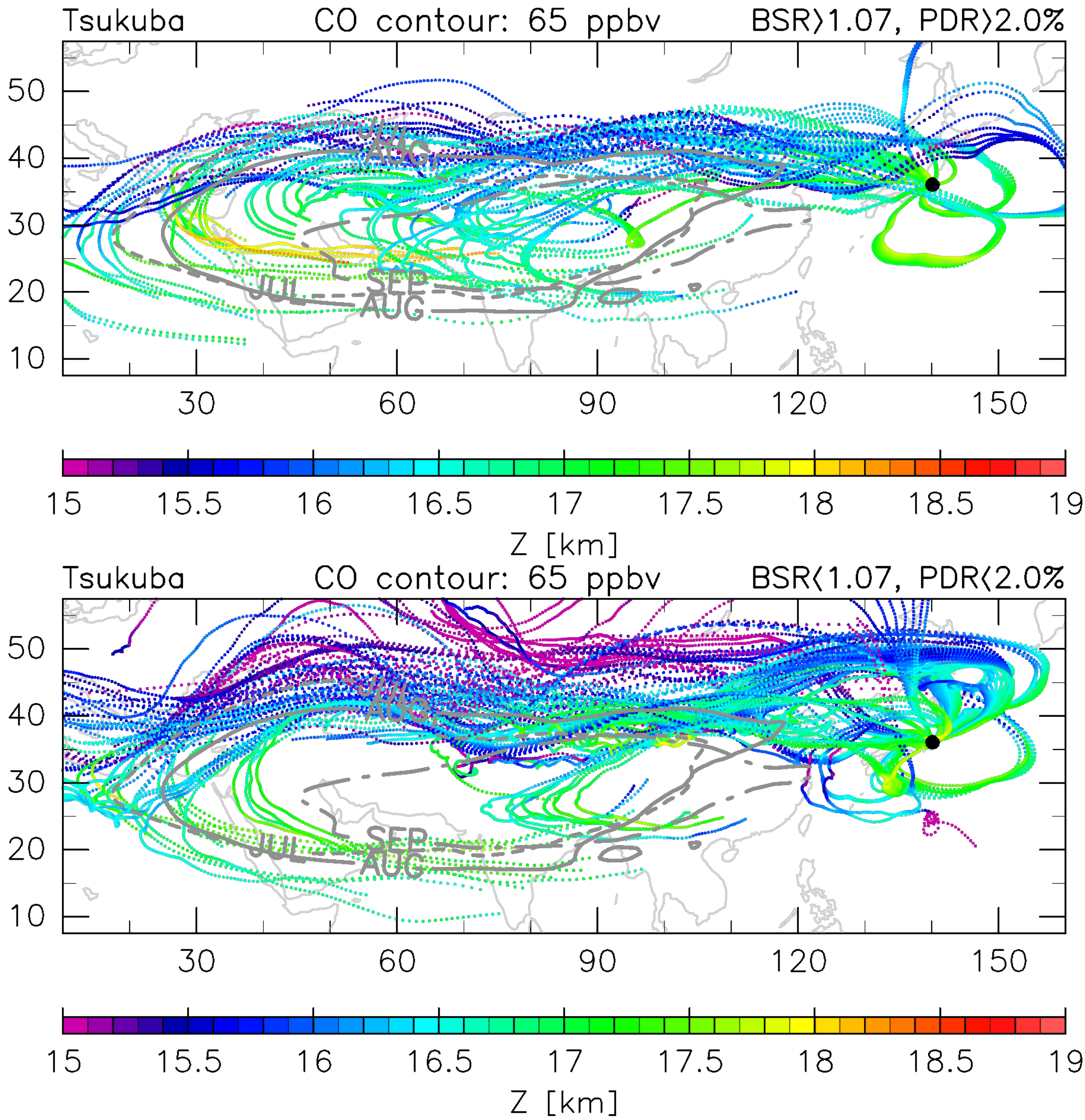
References

Vernier, J.-P., Fairlie, T. D., Natarajan, M., Wienhold, F. G., Bian, J., Martinsson, B. G., Crumeyrolle, S., Thomason, L. W., and Bedka, K. M.: Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution, Journal of geophysical research. Atmospheres JGR, 120, 1608–1619, https://doi.org/10.1002/2014JD022372, 2015.

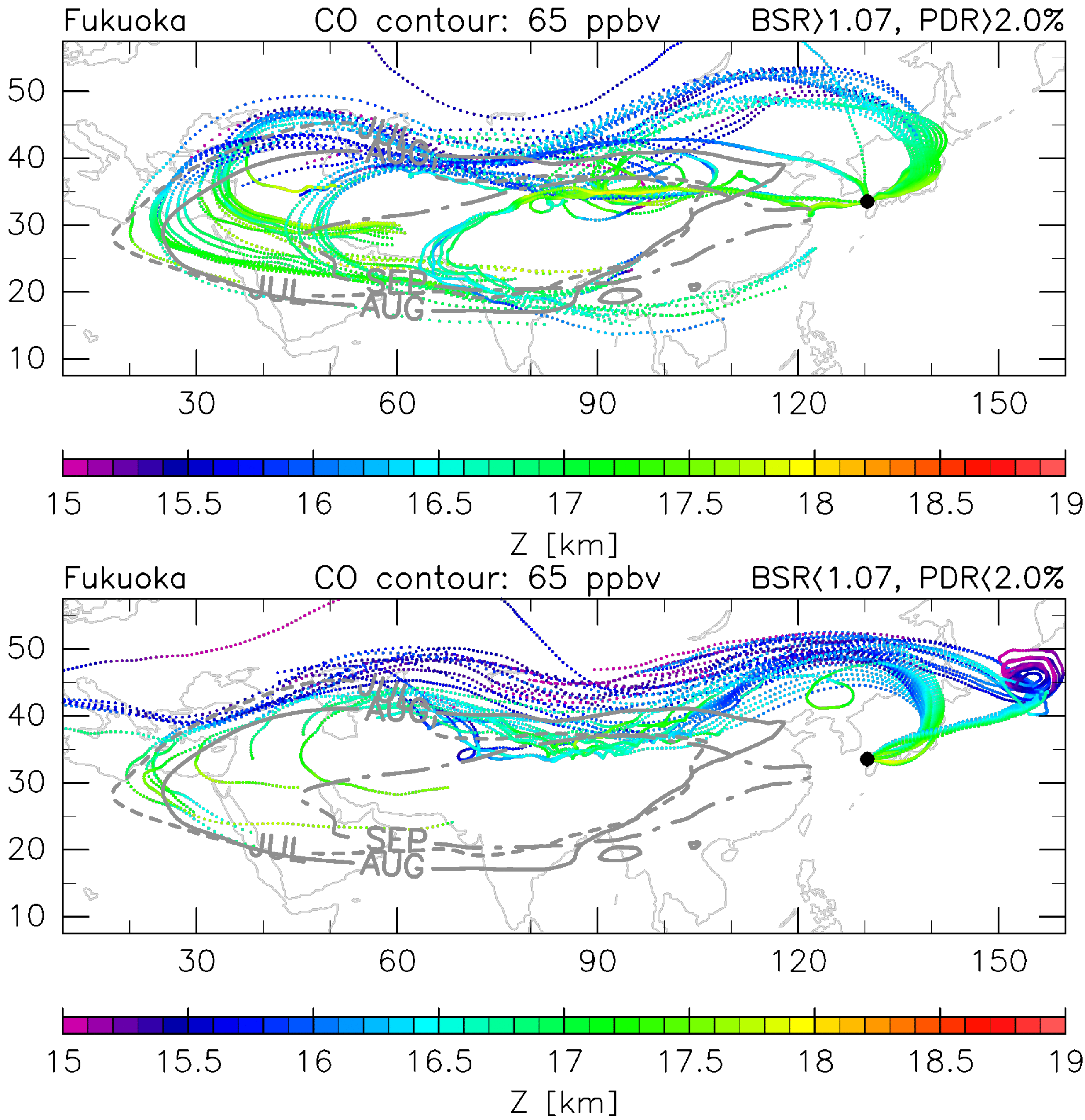
Wagner, R., Bertozzi, B., Höpfner, M., Höhler, K., Möhler, O., Saathoff, H., and Leisner, T.: Solid Ammonium Nitrate Aerosols as Efficient Ice Nucleating Particles at Cirrus Temperatures, J. Geophys. Res., 125, e2019JD032248, https://doi.org/10.1029/2019JD032248, 2020a.

Wagner, R., Testa, B., Höpfner, M., Kiselev, A., Möhler, O., Saathoff, H., Ungermann, J., and Leisner, T.: High-resolution optical constants of crystalline ammonium nitrate for infrared remote sensing of the Asian Tropopause Aerosol Layer, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-262, 2020b.

The two papers by Wagner et al. will be cited in the revised manuscript. The paper by Vernier et al. has already been cited.



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).