

Interactive comment on “Global modelling studies of composition and decadal trends of the Asian Tropopause Aerosol Layer” by Adriana Bossolasco et al.

Anonymous Referee #1

Received and published: 1 September 2020

In this manuscript, Bossolasco et al. present global model investigations on the composition and evolution of the aerosol layer present in the upper troposphere in the region of the Asian summer monsoon, the so-called Asian Tropopause Aerosol Layer (ATAL). The identification of two separate layers with different origin of aerosols has to my knowledge not been described before. Further, the investigation of long-term trends provides new insights into the possible variability and anthropogenic influence. It would, however, be helpful to discuss and evaluate the finding of mineral dust and sulfate aerosol particles as the major constituents of the ATAL, more thoroughly in light of recent modelling studies and observational results. Provided that the detailed comments below are considered properly, I strongly suggest the paper for publication in

C1

ACP.

Specific comments:

L32:

You may add a short note that nitrate aerosols have not been considered here. In my opinion this would help the reader from the beginning and does not at all diminish the value of the investigation.

L58, ‘while it was not observed prior to that year’:

In this context it should be mentioned that an ammonium nitrate aerosol layer has been observed already in 1997 (Fig., 1 in Höpfner et al., 2019).

L96, ‘dust is one of the predominant aerosol over the Tibetan Plateau’:

Please add the information that it has been detected up to 10 km altitude, otherwise one could be misled to think that it has been observed at altitudes of the ATAL.

L124, chapter ‘2.1-The CESM-MAM7 model’:

Could you add a paragraph how wet scavenging of gases, e.g. SO₂ and NH₃, and aerosols is handled in the model? As e.g. shown in Fairlie et al. (2020), this might be important for the modelling of sulfate in the ATAL.

L310, ‘These results agree with some previous modelling studies (e.g. Fadnavis et al., 2013, Ma et al., 2019)’

I miss a bit more quantitative discussion about the degree of agreement between the actual study and the most recent ones. E.g. add also in the discussion the results by Fairlie et al. (2020).

Figure 2:

Do the units ‘ng/m³’ refer to STP (as e.g. in Fairlie et al., 2020, Fig. 3) or are these absolute values at the given pressure levels?

C2

L543, chapter '4.4 - Aerosol Optical Depth (AOD) of the ATAL':

To be able to compare not only the absolute values but also the year-to-year variability (a strength of the actual study), I would strongly suggest to present a plot vs. time, like in Vernier et al. (2015), Fig. 6. This would allow a discussion model vs. measurements being more independent from the absolute values of AOD.

L555, 'It is important to mention that Vernier et al. (2015) have used hypotheses based on LiDAR observations and hypotheses on the LiDAR ratio value to derive the extinction coefficient to estimate the AOD.'

Vernier et al. (2015) have also used a depolarization filter ('cloudy pixels in the upper troposphere are removed using a volume depolarization ratio threshold of 5%') – could you discuss the possibility that due to this filter, also signals from dust may have been dismissed from the observations and what this would mean for your comparison?

L588, 'The results show that dust is the dominating aerosol type in terms of mass in the ATAL in agreement with other studies (e.g. Ma et al., 2019).'

This conclusion is too absolute in this context. I miss here a bit more balanced discussion with respect to other model results (1) and with respect to observations (2).

(1) Other models, like Fairlie et al. (2020) or Yu et al. (2015), do not predict dust as the dominating type of ATAL aerosols. E.g. Ma et al. (2019) refer to Brühl et al. (2018) who 'showed high sensitivity of mineral dust reaching the UTLS to model resolution, owing mostly to the differences in convection top height and overshooting convection in the parameterizations'. Could you discuss your results with respect to possible reasons why these differences between models occur? Can you detect a single cause why your model results indicate a stronger contribution of dust than other models?

(2) As long as measurements do not confirm the model results, one cannot conclude as firmly as done here about the composition of ATAL aerosols. E.g. in-situ airborne observations during the StratoClim campaign have neither identified dust nor sulfate

C3

as a major constituents of the ATAL layer (e.g. Höpfner et al., 2019).

Technical comments:

L23, 'We identify a "double-peak" aerosols vertical profile':

e.g. 'vertical profile of aerosols' 'aerosols' is in this way often used incorrectly. Please check and correct throughout the manuscript.

L75, 'ammont':

'amount'

L76, 'niitrate':

'nitrate'

L83, 'principal aerosols typology':

'principal typology of aerosols'

L85, 'enhancement':

'enhanced'

L90, 'This region have been'

'...has been'

L112, 'aerosols':

'aerosol'

L390, 'aerosols':

'aerosol'

L384,387, '1.0 10-3 km-1'

Please use correct notation for ACP.

C4

L391, 'seen.'

'seen.'

L450, 'details'

'detail'

References:

Brühl, C., Schallock, J., Klingmüller, K., Robert, C., Bingen, C., Clarisse, L., Heckel, A., North, P., and Rieger, L.: Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using Aerosol CCI satellite data, *Atmos. Chem. Phys.*, 18, 12845–12857, <https://doi.org/10.5194/acp-18-12845-2018>, 2018.

Fairlie, T. D., Liu, H., Vernier, J. P., Campuzano-Jost, P., Jimenez, J. L., Jo, D. S., Zhang, B., Natarajan, M., Avery, M. A., and Huey, G.: Estimates of Regional Source Contributions to the Asian Tropopause Aerosol Layer Using a Chemical Transport Model, *J. Geophys. Res.*, 125, <https://doi.org/10.1029/2019JD031506>, 2020.

Höpfner, M., UngermaNN, J., Borrmann, S., Wagner, R., Spang, R., Riese, M., Stiller, G., Appel, O., Batenburg, A. M., Bucci, S., Cairo, F., Dragoneas, A., Friedl-Vallon, F., Hünig, A., Johansson, S., Krasauskas, L., Legras, B., Leisner, T., Mahnke, C., Möhler, O., Molleker, S., Müller, R., Neubert, T., Orphal, J., Preusse, P., Rex, M., Saathoff, H., Stroh, F., Weigel, R., and Wohltmann, I.: Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons, *Nature Geosci.*, 12, 608–612, <https://doi.org/10.1038/s41561-019-0385-8>, 2019.

Ma, J., Brühl, C., He, Q., Steil, B., Karydis, V. A., Klingmüller, K., Tost, H., Chen, B., Jin, Y., Liu, N., Xu, X., Yan, P., Zhou, X., Abdelrahman, K., Pozzer, A., and Lelieveld, J.: Modeling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon, *Atmos. Chem. Phys.*, 19, 11587–11612, <https://doi.org/10.5194/acp-19-11587-2019>, 2019.

C5

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.

Yu, P., Toon, O. B., Neely, R. R., Martinsson, B. G., and Brenninkmeijer, C. A. M.: Composition and physical properties of the Asian Tropopause Aerosol Layer and the North American Tropospheric Aerosol Layer, *Geophys. Res. Lett.*, 42, 2540–2546, <https://doi.org/10.1002/2015GL063181>, 2015.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2020-677>, 2020.

C6