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

We would like to thank Reviewer 1 for her/his comments and his/her fast response during the discussion phase, which helped us improve the quality of the manuscript. We have discussed the suggestions/corrections raised by Reviewer 1 with the coauthors and made the changes in the text accordingly. Below each comment you can find our answers and the respective changes made in the manuscript.

Note:
Due to changes made in the manuscript, some of the line numbers referred by the reviewers have changed. These changes are shown in green when applicable.

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 ACP

Specific comments:
1) L32:
You may add a short not 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.

Done

2) 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).

Done. A small sentence about this was added at the end of the paragraph.

3) 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 mislead to think that it has been observed at altitudes of the ATAL.

The paragraph was changed accordingly, as follow:

“In several studies, dust has been shown as a major contributor to the aerosol burden in the Asian upper troposphere during summer. Xu et al. (2015), using CALIOP and MISR (Multi-angle Imaging SpectroRadiometer) satellite data, **have found that dust is one of the predominant aerosol over the Tibetan Plateau most probably originating from the Taklamakan desert and lofted from the surface to an altitude of about 10 km**.”

4) L124/L145 (new line number), 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.

We have added two paragraphs to explain shortly the wet removal of soluble gases and aerosols (L168-176 and 200-207, in the revised manuscript). The wet scavenging used in CESM-MAM7 is the standard scheme in CAM5, although as has been noted by Fairlie et al. (2020) a more physically based treatment of wet scavenging of SO_2 in convective updrafts increases the amount of sulfate. A more detailed study to evaluate this will be done in a future.

5) L310/L388 (new line number), ‘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).

We thank the Reviewer for this correction. We have extended the discussion and added a more detailed discussion about the degree of agreementbetween our study and the recent ones and the possible biases of dust modelled. See the lines 388-419 in the revised manuscript.

6) 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?

They are absolute values. These units have been used to quantify aerosol burdens in several previous studies (e.g. Fadnavis et al., 2019; Fadnavis et al., 2017; Ma et al., 2019). We use them for the sake of comparison with these previous studies; we acknowledge that other authors use other units, as volume mixing ratios.

Fadnavis, S., Müller, R., Kalita, G., Rowlinson, M., Rap, A., Li, J.-L. F., Gasparini, B. and Laakso, A.: The impact of recent changes in Asian anthropogenic emissions of SO2 on sulfate loading in the upper troposphere and lower stratosphere and the associated radiative changes, Atmos. Chem. Phys., 19(15), 9989–10008, doi:10.5194/acp-19-9989-2019, 2019.

7) L543/L653 (new line number), 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.

In the revised manuscript, we have now provided such plots and a corresponding discussion. Here we summarize that discussion. On the plot in Fig. 6 in the revised manuscript: the associated summer-averaged AOD values are larger than those of Vernier et al. (2015) (their Fig. 6). It is difficult to directly and quantitatively compare our simulated AODs with the measurements of Vernier et al. 2015 because of the different considered periods and, more important, the cloud filtering in the AOD observations (see next answer). First, periods impacted by volcanic aerosol perturbations are somewhat different between our model analysis and Vernier et al. (2015) (e.g. we have excluded years 2006-2008). We are confident that our selection of volcanic-free periods is state-of-the-art (see manuscript for details). Second, we may expect that the cloud screening result in different average AODs and trends for simulations and observations. This is an obvious reason why we have found a lower altitude/in-cloud ATAL component (lower-altitude peak in the double-peak structure), which is not observed in Vernier et al. 2015. Our new AOD time-series shows that accounting for the double-peak ATAL structure leads to different trends and reflects the importance of the altitude range used to estimate the year-to-year variability. We then conclude that both studies reveal an increasing ATAL AOD trend but without directly reconciling the two datasets, as a result of different methods applied.

8) L555/L663 (new line number), '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?

The paragraph was changed accordingly, and we discuss irregularly-shaped particles might have been removed due to the depolarization filter applied by Vernier et al. 2015, with a possible impact on dust.

9) L588/L712 (new line number), '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?

The paragraph has been changed and a more detailed discussion has been included in Sect. 4.1 (see answer 5) and in the conclusion. A brief summary of the new section is in the following. We are aware of the larger amounts of dust in the UTLS region, in our simulation, with respect to some previous works. Nevertheless, dust modelling is still uncertain, at the present time. As discussed in previous works and especially in Wu et al. 2019, who compare different dust schemes with satellite observations, the GCMs have large uncertainties in the simulated dust cycle in terms of spatial distribution and temporal variation. Possible reasons, which would require thorough analyses, could be linked to uncertainties in the physical process leading to dust erosion, the representation of convection in the models and/or effects of vertical resolution on the transport. The simulations of Brühl et al. (2018) have shown that the amounts of dust reaching the UTLS region in the EMAC model are sensitive to model resolution. In our work with CESM-MAM7, we use a 1.9 x 2.5° horizontal resolution and 56 vertical levels which is standard for CESM1 and has been used in previous studies of aerosol properties (Yu et al. 2015, 2017). These resolutions are lower than the values in Brühl et al. (2018) and this could indeed impact the dust reaching the UTLS in CAM5 as result of differences in convection top height and overshooting convection.
We use also use the standard configuration of CAM5 for the vertical transport (Zhang and McFarlane, 1995). This information has been added in Sec. 2 (L243-249).
We feel that detailed analyses on resolution and convective parameterization would be largely out from the scope of the paper. It is worth noticing that large uncertainties exist also in satellite retrievals of dust, which makes difficult the validation of the models.

Zhang, G. J., and N. A. McFarlane (1995). Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian Climate Centre general circulation model, Atmos. Ocean, 33, 407–446.

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

We agree and we have toned down some statements and discussion throughout the manuscript, including the "validation" of the model. However due the lack of information about the ATAL composition derived from observations in the AMA region over the period of the simulation we can only qualitatively compare our model results with the ion chromatography analysis from aerosol samples collected in summer 2015 in India during the BATAL balloon campaign (Vernier et al., 2018). As discussed in Vernier et al 2018, the undetectable concentration of sulfate ions (<10 ng.m^{-3}) seems to be contradictory with the expectation of a major contribution of sulfur (and influence emissions in Asia over the past few decades) in the aerosol layer in the UTLS. This result strongly differs from the observations of the StratoClim campaign which has identified a very high proportion of nitrates and sulfates in the summer 2017 ATAL, reflecting the complexity of the processes controlling the ATAL composition and variability. The ATAL might be so variable that a single campaign is not sufficient to characterise it. This is reflected by the revised text in the new manuscript version.

Technical comments:

11) 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.
Changed

12) L75/L88 (new line number), 'ammont':
'amount'
Corrected

13) L76/L89 (new line number), 'niitrate':
'nitrate'
Corrected

14) L83/L85 (new line number), 'principal aerosols typology':
'principal typology of aerosols'
Corrected

15) L85/L91 (new line number), 'enhancement':
'enhanced'
Corrected

16) L90/L108 (new line number), 'This region have been'
'. . .has been'
Changed. We have deleted the paragraph because it was redundant and it was extended in accordance with comments of reviewer #2.

17) L112/L133 (new line number), 'aerosols':
'aerosol'
Corrected

18) L390/L514 (new line number), 'aerosols':

‘aerosol’
Corrected

19) L384,387/ L508,511 (new line number), ‘1.0 10-3 km-1’
Please use correct notation for ACP.
Corrected

20) L391/L515 (new line number), ‘seen .’
‘seen.’
Corrected

21) L450/L568 (new line number), ‘details’
‘detail’
Corrected