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

Anonymous Referee #3

We would like to thank to the reviewer for her/his detailed and mostly positive comments and suggestions. We discussed each of the points raised by Reviewer 3 among the coauthors and made the changes in the text accordingly. Below each comments, please find our answers and the respective changes made.

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.

Received and published: 14 September 2020

General comments:
This study is very well written and addresses a hot topic in the scope of ACP. It provides interesting hypotheses about the nature of the ATAL, e.g. that there is a double-peak vertical structure, mineral dust dominates aerosol mass, and that the ATAL signature has been increasing from 2000 to 2015. This should be published, considering the following.

The paper would benefit from actually working out if one or more of the above hypotheses have something to do with reality. In the current version, the analyses and discussions are limited almost exclusively to the modelling world, to one simulation.
This simulation is linked to the real world just by a comparison to observed CO. However, the emissions contributing to ATAL have other source distributions than CO, and are affected by other processes.

Furthermore, the uplift of air from the ground to the UTLS - a crucial process for ATAL – might need a closer look in the model: simulated CO compares favorably to the observations in the UTLS, despite being off in the free troposphere (Figure 1e). A much more thorough model evaluation would be appropriate, covering (proxies for) all species, precursors and processes of relevance for the aspects of ATAL that are discussed in this study. Sensibly splitting this between the supplement and the main text would allow the paper to stay concise.

Apart from the mere model evaluation, it would help putting some effort into researching available observations for support of the model-based findings about ATAL.
A more detailed understanding of the strengths and weaknesses of the simulation might also help the discussion of how this study compares to other modelling results.

We thank the reviewer for constructive comments. As discussed in the manuscript and in some previous studies, the chemical

composition of the ATAL remains poorly characterized due to the lack of in in situ measurements in the AMA region. Only over the last recent years some aircraft and balloon campaigns have started to be conducted in the region (e.g. Stratoclim from 2016 to 2018 and BATAL from 2015 to the present), i.e. mainly after the period of our simulation. That is why the present study cannot be exhaustively compared with in situ measurements. In addition, satellite observations of aerosols in terms of their composition are very scarce and mostly limited, for this application, by the interaction of radiation with co-existing clouds (so that the necessary cloud screening likely screened out the lower of the two ATAL peaks, in past works, see discussion about the results of Vernier at al., 2015). However, the double-peak structure of the ATAL was observed before, even if not discussed in past works. So, we have extended our discussion regarding the double-peak vertical profile doing a qualitative comparison with some of these recent measurements (Vernier et al., 2018, Brunamonti et al., 2018, Höpfner et al., 2019) see Sec 4.2, 489:503 in the revised manuscript.
On the other hand, as many studies that have been carried out (Fadnavis et al. 2013,2017; Yu et al., 2015, 2017; Gu et al. 2016; Lau et al. 2018; Ma et al., 2019; Fairlie et al., 2020; etc, cited in the current work), different models simulations provide new insights into the composition, budget, origin and source contribution to the ATAL.

As for the validation using satellite observations of gas specie, please note that these comparisons, for CO, were only meant to illustrate the ability of the model to transport pollutants to the UTLS. CO has been used as a representative pollution tracer in the UTLS. Simulated CO shows a broad maximum over the monsoon anticyclone region, in a reasonable agreement with the spaceborne observations in term of spatial extent.
In this work we do not compare all the gas species with satellite data and do not evaluate all chemical and physical processes computed by CAM5 since this would require a large work and a specific dedication and efforts in itself. As similar past studies (where model validation was generally absent or, in any case, less detailed than ours), here we focus on ATAL aerosols distribution, composition and, more originally, long-term variability of each aerosol type and their integrated optical properties, which has never been reported before. We discuss throughout the text some limitations of the model whenever possible.

Specific comments:
1) L87/ L105 (new line number): have -> has
Changed

2) L105/ L130 (new line number): The bimodality of the AMA has been discussed for longer, see e.g. (Nützel et al. 2016, Pan et al. 2016) and references therein.
The references have been changed and this previous works added.

3) L107/ L130 (new line number): beweekly -> biweekly or bi-weekly
Done

4) L116 /L140 (new line number): larger aerosols composition -> more comprehensive aerosol composition
Changed it

5) L116/ L140 (new line number): Please check the use of aerosols / aerosol / aerosols' / . . . throughout the paper.
Done

6) L149 /L187 (new line number): Isn't anvil associated with convective rather than stratiform clouds?
We have removed the term "anvil" which was, indeed, confusing.

7) L179 /L218 (new line number): Simulated ATAL trends are likely to critically depend these assumptions. Please elaborate on the uncertainties in the emissions' setup, providing the reader with a sense on how this might print through to the results for ATAL.

The paragraph was changed accordingly, and we have added at the end of this paragraph (L225-227) a sentence to explain more in detail the assumptions made by CEDS inventory that introduce uncertainties. This is mentioned in the conclusion as well.

8) L191 /L239 (new line number): Different reanalyses have different peculiarities in representing AMA (see e.g. Nützel et al. 2016). Please shortly note whether there is something specific the reader needs to know about MERRA.

To our knowledge, only the old NCP reanalyses are problematic in representing the AMA (strong bimodality). All modern reanalyses, including MERRA2, agree well on the AMA. There are quite large differences, however between modern reanalysis regarding cloud properties and heating rates. But this is probably not relevant here. MERRA2 reanalyses are compared with other datasets in Long et al. (2017), where they report a very good agreement in temperature seasonally and latitudinally between the surface and 10 hPa, for the more recent reanalyses (CFSR, MERRA, ERA-Interim, JRA-55, and MERRA-2). Zonal winds are in greater agreement than temperatures and this agreement extends to lower pressures than the temperatures. Older reanalyses (NCEP/NCAR, NCEP/DOE,ERA-40, JRA-25) have larger temperature and zonal wind disagreement from the more recent reanalyses.

In Sec 2.1 we have added a sentence to explain that our model is driven by MERRA2 data (and not MERRA like show Nützel et al. 2016) with a constrain of 10%, i.e every time step the offline meteorological fields (horizontal wind components, air temperature, surface temperature, surface pressure, sensible and latent heat flux, and wind stress) are nudged to the online calculated meteorology. The nudging coefficient in our case is 0.01 (10%).

Long, Craig S., Masatomo Fujiwara, Sean Davis, Daniel M. Mitchell, and Corwin J. Wright, Climatology and interannual variability of dynamic variables in multiple reanalyses evaluated by the SPARC Reanalysis Intercomparison Project (S-RIP), Atmos. Chem. Phys., 17, 14593–14629, 2017, https://doi.org/10.5194/acp-17-14593-2017

9) L281: A 30
Corrected

10) L303 /L379 (new line number): Please consider showing the comparison to the corresponding observations.

There aren’t “corresponding observations” for these aerosols during this year. Is not possible to shows such direct comparison. As answered at the beginning, due the lack of information about the ATAL composition derived from observations in the AMA region over the period of 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).

11) L330/ L440 (new line number): Is the following understanding correct? There is no dynamic tracking of the AMA. Rather you choose a static box, which most of the time is part of the AMA. Any averages should thus be dominated by AMA conditions. This is ok, but some rewording might help to make the approach clearer.

We have made some rewording to clarify the approach which is indeed based on a static box and not on dynamic tracking changing with time. See Lines 431-444 in the revised manuscript.

12) L335/ L449 (new line number): Isentropic surfaces might be better to describe horizontal transport and thus the horizontal extent of the ATAL (Santee et al. 2017, Gottschaldt et al. 2018). Please check whether or not your results crucially depend on the choice of the vertical coordinate system.

We have carried out the same analysis doing the plots at different isentropic surfaces (400, 380, 360 K) and the plots look pretty similar. So, we have decided to carry out our analysis in pressure levels since this is the basic coordinate system in our model and this does not require any interpolation at each time step.

13) L363/L476 (new line number): The term “mode” is already in use for aerosol size ranges and for the dynamics of the AMA. Does it refer to different aerosol classes here?

The paragraph was probably confusing, so we have deleted the term bi-modal. The two relative maximum observed (double-peak) refers to two different origin of aerosols that are present at different altitudes one at lower altitudes (~ 250 hPa) associated with “convective” cloud-borne aerosols and one at higher altitudes

(~ 100 hPa) associated with "clear-sky" aerosols. This is discussed later in the paragraph.

14) L363/ L476 (new line number): Is there any observational hint for such a double-peak layering?

Yes (even if not for the time period of our simulations). The works of Vernier et al. 2018, Brunamonti et al. 2018, Höpfner et al 2019 shows evidence of this double peak and a discussion about this was included later in the Sec 4.2, see lines 489-503 in the manuscript.

15) L397 /L525 (new line number): "Double-peak", when used as adjective? Please check throughout the paper.
Done

16) L405/ L531 (new line number): Please mention in the caption that this is modelling only.
Ok, Done.

17) L433 /L552 (new line number): That is rather vague. Several models get an ATAL, so it seems to be a quite stable feature. Interestingly, the exact characteristics vary, probably depending on the various factors listed here. For improving our understanding of ATAL it is therefore important to really understand the model differences, and to find those explanations that are supported by observations.

We agree with the reviewer's comment but as replied to a previous comment (answer 10), there are only few available in situ observations about the chemical composition of aerosols present in the ATAL. We would need a significant number of new in situ observations to make a comprehensive comparison with our model outputs, as well as gathering more information on the "real" ATAL (i.e. from observations). For the moment, we still can have some information using models: we think it useful to provide information on the ATAL's composition and temporal variability by models.
For the other hand we have reorder our discussion in Sec 4.1 regarding the comparison with other models results and regarding specifically to dust an extended discussion and the possible limitations of the model have been added in Sec 4.1 (lines 389-419) and in the conclusions.

18) L457/L558 (new line number): Please use subscripts in chemical compounds throughout the paper.
Done

19) L484 /L606 (new line number): showed -> shown
Done

20) L493 /L612 (new line number): Is there a chance to be more specific: Which aspects of the dust cycle are captured well by your model? Which are not and what are the implications for your conclusions about ATAL?

As mentioned before (answer 17) and in the replies to Reviewer#1 (answer 5 and 9), there are still large uncertainties in dust

modelling across different models. However we have extended our discussion about the possible biases for the modelled dust (Sec 4.1 and Conclusion in the revised Manuscript).

21) L503 /L616, 686 (new line number): Here you state model shows that increased emissions translate into enhanced ATAL, but in L567 alternative explanations are offered. Please check consistency. Furthermore, as already noted by reviewer2, emission trends are more complicated. A more detailed analysis might be needed, e.g. explicitely correlate emissions (by region) with ATAL parameters.

We wish to clarify that we actually do not provide a different explanation since in the first part we analyse the contribution of aerosols in the accumulation mode (a1), which are principally from anthropogenic contribution. For the discussion about AOD, we account for the total modelled extinction which includes all the aerosol types present in CESM-MAM7 and in any case we find an increase of a factor of 2 for the AOD for both ranges of altitude. In the manuscript, we specify that the reasons for the the ATAL AOD increases (increase in Asian emissions, more efficient vertical transport or other reasons) require further investigation. Following the reviewer's suggestion, sensitivity studies could be done by masking emissions from mainly contributing regions (e.g. China for SO_2, Gangetic valley for NH_3) or testing different emission inventories which could be the scope of a dedicated study.
In the conclusion a paragraph regarding the implication of the CEDS emissions used has been added (L733-738).

22) L552 /L663 (new line number): This formulation is kind of suggesting that Vernier et al. might be wrong. Please elaborate.
The paragraph was changed it accordingly.

23) L555 /L669 (new line number): Is CESM1/CARMA from the same model family you are using? Then getting similar results could also indicate a common problem.

CESM1/CARMA is indeed the same family model than our CESM1/CAM5-MAM7. Nevertheless, the aerosol models (CARMA and MAM7) are deeply different. We obtain similar results to Yu et al. 2015 like the features of the maximum in the AOD vs longitude and the AOD values. These simulated values are higher than those reported by Vernier et al. (2015) using the CALIOP space-borne lidar. However, different cloud-screening procedures have been used in Vernier et al. (2015) and in our study. On may argue that aerosols with high extinction, like those we have identified from convective cloud-borne aerosols in our lower altitude peak, might have been removed from the lidar signal during the cloud-screening process, in the paper by Vernier et al (2015).
While the residual differences with respect to Yu et al. 2015 are easily attributable to the different aerosol models, they may also be due to the different emission inventories used. Yu et al., 2015 have used GFED3 for biomass burning emission and GFED2 for SO_2 biomass burning emissions, anthropogenic emissions are taken from EDGAR-FT2000 and biogenic emissions are estimated by

Guetner et al 2006. Different meteorological data used (MERRA, used by Yu et al 2015, MERRA2, used in the present study) may also have played a role.

24) L562/ L672 (new line number): Another interesting hypothesis. Please check whether there are any observations supporting it.

As was mentioned at the beginning and in the answer 14, this hypothesis of different AOD values obtained for the two altitudes ranges can be attributed to the different aerosols present at different altitude ranges, related to the double-peak vertical profile of aerosols found. We have added a discussion regarding the observation of this double-peak structure observed in some recent balloon and aircraft campaigns (Sec. 4.2).

25) L585/ L712 (new line number): Please consider rewording: The results show . . . -> Our modelling results indicate . . .
Done

26) L594: Please make it clear from the beginning that nitrate aerosols might be an important aspect you omit.
Yes, this was added in the abstract

27) L812/ L970 (new line number): space between references missing
Corrected