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 2 for the time spent and the detailed comments and suggestions (including additional literature). This helped us improving our manuscript. In the following, we address each comment individually, including the changes we made to the manuscript accordingly.

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 #2
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Comments on Manuscript No. ACP-2020-677
In the last decade the Asian Tropopause Aerosol Layer (ATAL) becomes in the focus of attention. However, the current knowledge of the ATAL is limited. This study presents model results based on the Community Earth System Model (CESM 1.2) with the focus to simulate the chemical composition of the ATAL and its decadal trends. A vertical 'double-peak' structure is found for the ATAL. Mineral dust is the dominant aerosol by mass in the ATAL. Further, the ATAL is composed of around 40 % of sulfate, 30% of secondary and 15% of primary organic aerosols, 14% of ammonium aerosols and less than 3% of black carbon. A positive trend for all aerosols was simulated using the Modal Aerosol Model MAM7.

Despite of the somewhat weak discussion of the scientific results compared to the current scientific knowledge, that could be improved, this is an interesting study, which merits its publication in ACP. However, I suggest some revisions to make this possible.

1) The vertical 'double-peak' structure in the ATAL presented in this paper is very interesting. It would be an added value for the paper to discuss whether also measured vertical ATAL profiles from in situ balloon or aircraft measurements show such a 'double-peak' structure (e.g. published by Vernier et al, 2018; Brunamonti et al, 2018; Höpfner et al, 2019;...). Such a discussion could be presented in a separate 'discussion section'.
All these references are already in the publication list.

A discussion regarding the "double-peak" structure observations has been added in the Sec 4.2. See lines 489-503 in the revised manuscript.

2) At several places within the paper, I am missing the discussion about what is new or different to previous publications (see specific comments below).

We have added more discussion about how our results compare with previous studies (Fadnavis et al., 2013; Yu et al., 2015; Ma et al., 2019, Fairlie et al., 2020).
The new contribution of our work is the ATAL aerosol modeled trends over 16 years (not shown before) and the new ATAL double-peak structure, which, although it has been observed in some recent measurements has not been extensively discussed. We modified the text in different places so to make it clear these two major novel results of our work.

3) The comparison of CO between model and satellite measurements is somewhat weak in particular the comparison of vertical CO profiles (see specific comment below).
We address the answer in the specific comments below.

Specific comments:
4) P1/L19: ’pollutants’ -> ’ATAL aerosols and their chemical precursors’
The paragraph was changed

5) P1/L19: Please clarify: ’its atmospheric chemical processes’
The paragraph was changed

6) P1/L19: What about the variabiliy of the sources/emissions contributing to ATAL?
The reviewer is right, we have added a small sentence to mention it.

7) P1/L14-19: For better understanding, I recommend to separate this long sentence into two or more sentences.
The paragraph was changed.

8) P2/L27: ’We find that mineral dust is the dominant aerosol by mass in the ATAL showing a large interannual variability, but no long-term trend, due to its natural variation.’ Here it is unclear if mineral dust is dominant in both ATAL peaks. Please clarify.
This clarification was added in parentheses.

9) P2/L40/ L43 (new line number): ’The upper atmospheric circulation is dominated by the related Asian Monsoon Anticyclone (AMA), which is known to contain enhanced concentration of tropospheric trace gases and aerosols, ...’ Please add some references.
Done

10) P2/L59/ L60 (new line number): Höpfner et al, 2019 should be also mentioned. They reported that enhanced concentrations of solid ammonium nitrate particles were found in the Asian monsoon anticyclone in 1997.
This has been added

11) P2/L74/ L86 (new line number): model, Fairlie ... -> model. Fairlie ...

“ Using GEOS-Chem (Goddard Earth Observing System with Chemistry) chemical transport model, Fairlie et al. (2020) have found significant amont of sulfate....”
We think using a comma is correct here.

12) P2/L76 / L89 (new line number): niitrate -> nitrate
Changed it

13) P2/L80/ L93 (new line number): One result of the paper is that dust is the dominant aerosol by mass in the ATAL. However, only Ma et al (2019) is discussed here in the introduction as a model study that also found that mineral dust contribute to the ATAL. In the literature there are more studies analyzing the contribution of mineral dust to ATAL (e.g. Lau et al, 2018; Fadnavis et al, 2013, ...). Please discuss here also the results of more previous publications to give them credit.

To clarify, we first mention the previous studies that are based on modelling and their results for all aerosols types in the ATAL. Then we emphasize the previous studies that showed an important contribution of dust in the ATAL region (Lau et., al 2018, Ma et., al 2019). To further clarify the text, we have moved the paragraph regarding the discussion of dust to some lines above (Line:93-104 in the revised manuscript) and we rephrased it.

14) P3/L90/ L115 (new line number): What about the contribution from the Sichuan Basin (China)?
We have added a paragraph to mentioned it (including the citation to Lau et al 2018).

15) P3/L93/ L108 (new line number): ’Continental convective regions have also been shown to be the main contributors to the air trapped within the AMA with North India and South of the Tibetan Plateau as specific source areas (Tissier and Legras, 2016; Legras et al., 2019).’ -> (e.g. Tissier and Legras, 2016; Legras et al., 2019).’ There are several other studies related to the possible source regions of AMA. Please discuss a few more of these studies. Moreover, Fairlie et al. (2020) indicated the dominance of the contribution of regional anthropogenic emissions from China and the Indian subcontinent to the ATAL. Therefore, please add here also studies discussing possible source regions of the ATAL (e.g. Fairlie et al., 2020;...)

This discussion has been extended and the references added. We have modified the paragraph as follow:
“ Continental convective regions have also been shown to be the main contributors to the air trapped within the AMA with North India and South of the Tibetan Plateau as specific source areas (e.g. Tissier and Legras, 2016; Legras et al., 2019). Bergman et al. (2013), using Lagrangian backward trajectories, have shown that the anticyclone is connected to the boundary layer through a vertical conduit centred over Northeast India, Nepal, and southern Tibet. In the recent BATAL campaign, Vernier et al. (2018) have used back-trajectory calculations to point at North of India as a principal region source for ATAL. Lau et al. (2018), based on MERRA-2 reanalysis have reported that the Himalayas Gangetic Plain (HGP) region and the Sichuan Basin (SB) of southwestern China, are two important regions with

strong vertical transport of CO, carbonaceous aerosols and dust from the surface to the UTLS. On the other hand, the simulations of Fairlie et al. (2020) have suggested that the anthropogenic sources from India contribute to up to 40% of sulfate and up to 65% of organic and ammonium aerosols in the western ATAL region, whereas China contributes up to 60% (both sulfate and organic aerosols) in the eastern ATAL region."

16) P3/L94: Its confusing that the contribution of dust to the ATAL was discussed in several places within the introduction (see comment above).

We agree. The paragraph has been changed accordingly and we have moved the discussion of dust (see answer 13 above,L93).

17) P3/L108/ L128 (new line number): 'Wei et al., (2019) have also found that the AMA exhibits intraseasonal variability between the Iranian Plateau and the Tibetan Plateau with a quasy-biweekly oscillation.' The bimodal distribution of the AMA is already discussed in previous publications (e.g. Zhang et al., 2002; Yan et al., 2011; Vogel et al., 2015; Nützel et al., 2016). Please add some of these references.
Zhang, Q., Wu, G., and Qian, Y.: The Bimodality of the 100 hPa South Asia High and its Relationship to the Climate Anomaly over East Asia in Summer, J. Meteorol. Soc.Jpn., 80, 733–744, 2002.

Yan, R.-C., Bian, J.-C., and Fan, Q.-J.: The impact of the South Asia High Bimodality on the chemical composition of the upper troposphere and lower stratosphere, Atmos. Oceanic Sci. Lett., 4, 229–234, 2011.

Nützel, M., Dameris, M., and Garny, H.: Movement, drivers and bimodality of the South Asian High, Atmos. Chem. Phys., 16, 14 755–14 774, https://doi.org/10.5194/acp-16-14755-2016, 2016.

The reviewer is right, thanks, the paragraph has been changed as follow:

" Several studies have shown that the AMA exhibits intraseasonal variability between the Iranian Plateau and the Tibetan Plateau with a quasi-biweekly oscillation(e.g. Zhang et al., 2002; Yan et al., 2011; Nützel et al., 2016; Pan et al. 2016; Wei et al., 2019).**"**

18) P7/L256 /L307 (new line number): 'This could explain the low bias in CO mixing ratios for our comparisons with satellite measurements.' What about the impact of vertical transport from surface sources to the UTLS in the model. In Fig. 1e the CO values at around 500hPa are underestimated in the model, however above ~150hPa model and measurement agree. However the CO increase between 500 and 150hPa in the measurements is much lower compared to the model. Could that be a hint on vertical transport issues in the model?

The reasons for the differences between observed and modelled CO with respect to altitude are still unclear. We have added a

paragraph to discuss about the possibility of this discrepencies are linked to the treatment of convection by CESM1/CAM5 (L335-347) together with discrepancies in emission inventories.
See more details below:
The model is in general able to simulate much of the large-scale behavior for CO found in space-borne observations, although the degree of consistency of simulated and observed CO amounts depends on the season and latitude of the comparison as reported with CAM4 by Park et al. (2013) (note that convection is parameterized in the same way in CAM4 and CAM5).
He et al. (2015) using CESM1/CAM5 have reported an under predictions of CO at the surface over Asia, but the global tropospheric column of CO seems to be over predicted in their study. These authors suggest uncertainties in terms of spatial allocations of CO emissions as well as convective transport treatments. The convection in CESM-MAM7 is parameterized using the Zhang-McFarlane scheme (Zhang and McFarlane, 1995) for deep convection and the Hack scheme (Hack, 1994) for shallow convection (See L243:249 in the revised manuscript). This is a typical parameterization used in numerous studies involving the CAM5 (or previous versions) model. For more details about the convection schemes used in CAM5 please see Liu et al 2012 (Supplement) http://www.geosci-model-dev.net/5/709/2012/gmd-5-709-2012-supplement.pdf.
Brühl et al. (2018) have reported that model resolution affects transport (of aerosols in their study). The model resolution used is likely to impact the calculated transport of gases by convection. 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; Yu et al. 2017).
The model is “nudged” using external meteorological fields (here MERRA2) and although this nudging does not directly change the convection parameterization in the model, it is expected to influence the representation of convection, which is still to be properly assessed. The impacts of nudging in CCMs (including CESM1) on the vertical transport have been studied by Chrysanthou et al. (2019), who have shown some limitations in simulating the mean vertical transport in the stratosphere for these models (but interestingly with realistic representations of fast horizontal transport in their work).
Our goal is to investigate the ability of the model to simulate ATAL’s properties in its typical set-up and tests about resolution and standard diagnostics of atmospheric convection in CAM 5.1 would deviate from the scope of the paper.

Finally, another possibilities could be: the uncertainties in the extrapolation emissions using CEDS inventory (this discussion has been added in the revised manuscript L308:313) and the reactivity of CO with OH which is different in the gas and liquid phase; in this case, a thorough analysis of the pertinence of simulated OH amounts and of the reaction rate of oxidation of CO by OH in presence of clouds (more predominant below the 150 hPa level

where the difference is larger) could be conducted in a next study. In the UTLS, possible underpredictions of temperature could lead to smaller loss of CO with OH (He et al., 2015).
Following the reviewer's comment, we have toned down our statement that the model-observation comparisons shown in figure 1 tend to "validate" the model calculation of transport.

Park, M., W. J. Randel, D. E. Kinnison, L. K. Emmons, P. F. Bernath, K. A. Walker, C. D. Boone, and N. J. Livesey (2013), Hydrocarbons in the upper troposphere and lower stratosphere observed from ACE-FTS and comparisons with WACCM, J. Geophys. Res. Atmos., 118, 1964–1980, doi:10.1029/2012JD018327.

He, J., Y. Zhang, T. Glotfelty, R. He, R. Bennartz, J. Rausch, and K. Sartelet (2015), Decadal simulation and comprehensive evaluation of CESM/CAM5.1 with advanced chemistry, aerosol microphysics, and aerosol cloud interactions, J. Adv. Model. Earth Syst., 7, 110–141, doi:10.1002/2014MS000360.

Chrysanthou, Andreas, Amanda C. Maycock, Martyn P. Chipperfield, Sandip Dhomse, Hella Garny, Douglas Kinnison, Hideharu Akiyoshi, Makoto Deushi, Rolando R. Garcia, Patrick Jöckel, Oliver Kirner, Giovanni Pitari, David A. Plummer, Laura Revell, Eugene Rozanov, Andrea Stenke, Taichu Y. Tanaka, Daniele Visioni, and Yousuke Yamashita, The effect of atmospheric nudging on the stratospheric residual circulation in chemistry–climate models, Atmos. Chem. Phys., 19, 11559–11586, 2019, https://doi.org/10.5194/acp-19-11559-2019.

19) P7/L262/ L317(new line number): In the literature 'eddy shedding' is not the same as the bimodality of the AMA. Please clarify.
We agree, "eddy shedding" word was not used correctly in our manuscript, so we have changed in the paragraph.

20) P7/L265/ L320 (new line muber): 'They show a distributed pattern with maxima above eastern Asia, but also above western Asia (Fig. 1d), ...'. What about the maxima over Africa near the Equator?
The CO maximum at 150 hPa over north Africa is expected to result from the Asian pollution uplifted to the upper troposphere and recirculated by the ASM as described in Barret et al. (2008).

B. Barret, P. Ricaud, C. Mari, J.-L. Attié, N. Bousserez, B. Josse, E. Le Flochmoën, N. J. Livesey, S. Massart, V.-H. Peuch, A. Piacentini, B. Sauvage, V. Thouret, and J.-P. Cammas, Transport pathways of CO in the African upper troposphere during the monsoon season: a study based upon the assimilation of spaceborne observations, Atmos. Chem. Phys., 8, 3231–3246, 2008.

21) P8/L274/ L326 (new line number): 'We have also tested the vertical structures of CESM-MAM7 simulations, using an ACE-FTS CO mixing ratio profile in the UTLS (Fig. 1e).' One single vertical CO profile is not very

representative for a simulation over 16 years. Please could you provide more vertical CO profiles and maybe present their mean value and its variability over an larger time frame perhaps for June, July and August (similar as Fig. 3). Is the vertical 'double-peak' found in aerosol also present in simulated CO?

Only a few ACE-FTS profiles are available each year in the AMA (and even less within our 20-35°N/60-105°E box) due to sparse sampling and presence of clouds. This sampling is too limited to derive a robust averaged CO profile and do subsequent statistically significant analysis. However, following the reviewer's comment, we have added in the supplementary material a figure showing a comparison between MLS profiles and the model, and an inherent discussion is added in the manuscript (see 348:352 in the revised manuscrpt).

The double peak is not detected in modelled CO conversely to aerosols, because the lower peak is only linked to aqueous phase aerosol microphysics and not expected for gaseous precursors. This is very reasonable: we attribute the higher-altitude aerosol peak to gas phase chemistry (homogeneous nucleation) and this is reflected by the increased gaseous precursors concentration due to AMA-related convection.

22) P10/L322/ L430 (new line number): '(Randel and Park, 2006; Garny...)'-> '(e.g. Randel and Park, 2006;Garny...)'
Done

23) P12/L359/ L468 (new line number): 'The vertical structure of the AMA-related dynamics has been investigated by several authors (Bergman, J. et al., 2013; Garny and Randel, 2013; Brunamonti et al., 2018)..' Remove 'J' after Bergman and add 'e.g.' . There are more previous publications studying the vertical structure of the AMA (e.g. Park et al. 2009; Vogel et al, 2019; Bian et al, 2020,..)
Done and references added.

Park M, Randel WJ and Emmons LK et al. Transport pathways of carbon monoxide in the Asian summer monsoon diagnosed from Model of Ozone and Related Tracers (MOZART). J Geophys Res 2009; 114: D08303.
Vogel, B., Müller, R., Günther, G., Spang, R., Hanumanthu, S., Li, D., Riese, M., and Stiller, G. P.: Lagrangian simulations of the transport of young air masses to the top of the Asian monsoon anticyclone and into the tropical pipe, Atmos. Chem. Phys.,19, 6007–6034, https://doi.org/10.5194/acp-19-6007-2019, https://www.atmos-chem-phys.net/19/6007/2019/, 2019.
Bian, J., Li, D., Bai, Z., Li, Q., Lyu, D., and Zhou, X.: Transport of Asian surface pollutants to the global stratosphere from the Tibetan Plateau region during the Asian summer monsoon, Natl. Sci. Rev., 7, 516–533, https://doi.org/10.1093/nsr/nwaa005, https://doi.org/10. 1093/nsr/nwaa005, 2020.
Some of these references were added.

24) P14/Fig.3: Please explain briefly in the Figure caption why an application of an extinction filter is shown.

Done

25) P18/L485/ L603 (new line number): Please explain the meaning of the p-value in words.

The meaning was added in parentheses.

26) P18/L506/ L616 (new line number): This mirrors the increase of the emissions in Asia.' Zheng et al. (2018) shows that after 2013 China's anthropogenic emission of some pollutants decreased substantially (e.g., SO2) because of the implementation of new emission control measures. How does that fit to your results about increasing emissions in Asia? Are the new Chinese emission control measures considered in the Regional Emission inventory that is used in this study?
Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18, 14095–14 111, https://doi.org/10.5194/acp-18-14095-2018, https://www.atmos-chem-phys.net/18/14095/2018/, 2018.

Unfortunately, the CEDS emissions inventory does not include this recent regional emission inventory. The data for CMIP6 were published before our study and it usually takes time to introduce this kind of changes in regional inventories in the global emissions inventories. As has been detailed in the paper of Hoesly et al. (2018) and in the description of the Emissions (see Section 2 of our manuscript) REAS is the regional emission inventory used for the Asian region (covering the period 2000-2008) and MEIC (MEIC-Multi-resolution Emission Inventory for China)(Li et al. 2017) for China (having years 2008, 2010 and 2012).

Li, M., Zhang, Q., Kurokawa, J.-I., Woo, J.-H., He, K., Lu, Z.,Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng,Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H.,and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework ofthe MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 935–963,https://doi.org/10.5194/acp-17-935-2017, 2017

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R. and Zhang, Q.: Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11(1), 369–408, doi:10.5194/gmd-11-369-2018, 2018.

27) p17/Fig.4: Figure 4 shows very nicely the impact of volcanic eruptions. Is there also a modulation by El Niño?

This is an interesting and complex question raised by the reviewer. ENSO affects remote regions of the globe with regional responses in atmospheric dynamics, precipitation, temperature, etc. The way it might impact transport and atmospheric burdens of aerosols and their precursors is an open question.
Several studies have shown that ENSO clearly impacts tropopause temperatures which control the amounts of water vapour in the UTLS. On a basic way of thinking, this could affect the oxidation capacity (through OH radical production) and the microphysics of UTLS aerosols. During El Niño positive anomalies of up to 10% in lower stratospheric H2O can be induced (Diallo et al., 2018). Such an investigation would require to analyse in details the alignment of ENSO with the phase of the QBO because the two mechanisms give rise to different patterns of variability in the tropical cold point tropopause temperatures with as a consequence different degrees of moistening or drying of the lower stratosphere depending on the QBO phase (Diallo et al., 2018). The QBO alone produces more H2O (and ozone) anomalies than the ENSO alone so the question could be raised for QBO also.

Perhaps one first step to address the questions of modulation of AOD by ENSO, QBO, volcanoes over the period covered in our work would be to use multilinear regression through a dedicated study as done in the Diallo et al.'s paper for Age of Air. However, the fact that ENSO exerts its impacts on remote regions of the globe through nonlinear atmospheric teleconnections and that patterns of these teleconnection have changed throughout time (possibly due to anthropogenic forcing) may complicate a robust statistical analysis with this kind of method.

This is definitely matter of a different dedicated paper.

Diallo, Mohamadou, Martin Riese, Thomas Birner, Paul Konopka, Rolf Müller, Michaela I. Hegglin, Michelle L. Santee, Mark Baldwin, Bernard Legras, and Felix Ploeger Response of stratospheric water vapor and ozone to the unusual timing of El Niño and the QBO disruption in 2015–2016, Atmos. Chem. Phys., 18, 13055–13073, 2018, https://doi.org/10.5194/acp-18-13055-2018.

28) P18/L511/ L621 (new line number): Please clarify the meaning of 'increment' and 'correlation'.

The sentence was changed accordingly.

29) P21/L559/ L670 (new line number): Please clarify 'Our double-peak ATAL features highlighted in Fig. 6a'. I assume the meaning is that two maxima of AOD at different longitudes are found (corresponding to the bimodality of the AMA). It is confusing here because the expression 'double-peak structure' was already used for the two maxima

found in the vertical structure of the ATAL. Or is there an misunderstanding? Please clarify.

The reviewer is right, the paragraph was confusing. We refer to the shape and maximum found in Fig 6a which are comparable with those found by Yu et al 2015. The paragraph was changed accordingly.

30) P21/L564/ L673 (new line number): ’The difference between the AOD values obtained for the two altitude ranges in Fig 6a and 6b points at the importance of what we have identified as convective incloud aerosols.’ Please explain this in more detail.

We refer to the fact that the AOD difference found between the two range of altitude: 200-80 hPa (Fig 6a) and 120-80 hPa (Fig 6b) highlights the contribution of convective in-cloud aerosols, which makes that the AOD values for 200-80 hPa are larger than for 120-80 hPa.
The paragraph was extended accordingly.

31) P21/L567: ’full double-peak ATAL’ (see above L559)
Changed, see answer 29.

32) P22/L585/ L706 (new line number): ’The model evaluation with MLS and ACE-FTS satellite data reflects that transport and convection features are well represented in our simulations, despite a possible underestimation of the biomass burning emissions.’ In the paper, a rough comparison between simulated CO and measured CO is shown. I would not call this ’model evaluation’. Further, I am not sure if the transport and convection features are overall well represented in the model (see comment to L256). Please rephrase this sentence and use a somewhat more cautious formulation.

This paragraph and the title of section 3 have been changed accordingly, see answer n°18 and 21. We have also rewriting the conslusion as follow:
“The model results show overall good agreement with the space-time behaviour of CO in the UTLS region observed by the MLS and ACE-FTS space-borne instruments, despite a possible underestimation in the CO burden due to the underestimation of surface emissions. In particular, the horizontal distribution of modelled CO is in good agreement with MLS data and the vertical structure in the AMA shows a maximum near 150 hPa in agreement with the available ACE-FTS observations.”

33) P22/L590 /L716 (new line number): ’..what has been reported in the past’. Please add some references.
Done

34) P22/L595: ’Apart from dust, the average partitioning for other aerosol types contained in the ATAL (from anthropogenic and from biomass burning emissions) is the following: 40% Sulfate, 30% Secondary Organic Aerosols, 15% Primary Organic Matter, 14% Ammonium and less than 3% Black

Carbon.’ What is new or different compared to previous results regarding the chemical composition of ATAL?

As for a previous reply (see answer n° 2) new contribution of our work is the ATAL aerosol modeled trends over 16 years (not studied before) and the new ATAL double-peak structure, which, although it has been observed in some recent measurements has not been further discussed.
Regarding specifically to the chemical composition, we have compared our results with some previous works (e.g. Yu et al 2015). With respect to our work, they have also reported approximately the same % of sulfate, but a larger % of organic aerosols (45% in our model, 60% in theirs). Yu et al. 2015 have also simulated large amounts of dust but they don’t explicitly report the percentage. Unlikely Fadnavis et al. 2013,2017, our model doesn't show a maximum of black carbon in the ATAL. Some others works, like Fadnavis et al 2013 and Fairlie et al 2020, include nitrate in his models, while CESM-MAM7 doesn't treat nitrates.

35) P22/L602/ L731 (new line number): ’... a marked positive trend of anthropogenic and biomass burning aerosol concentrations is found, with up to a factor two increase of mass concentrations between 2000 and 2015. ’ What are the consequences if the ATAL over Asia is increasing further in future?

The consequences of the continuous anthropogenic emissions increase in Asia (principally of SO_2 and volatile organic compounds), and likely therefore aerosols in the ATAL, could be an impact in the radiative balance, stratospheric ozone chemistry, and properties/occurrence of cirrus clouds. However, as discussed before (see answer 27), the new emission control measures for SO_2 emissions in China is not considered in our CEDS emissions inventories and therefore could have different implications in the trends showed.