Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-677-RC2, 2020

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

Received and published: 8 September 2020

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.

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).

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).

Specific comments:

P1/L19: 'pollutants' -> 'ATAL aerosols and their chemical precursors'

P1/L19: Please clarify: 'its atmospheric chemical processes'

P1/L19: What about the variabiliy of the sources/emissions contributing to ATAL?

P1/L14-19: For better understanding, I recommend to separate this long sentence into two or more sentences.

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.

P2/L40: '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.

P2/L59: 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.

P2/L74: model, Fairlie ... -> model. Fairlie ...

P2/L76: niitrate -> nitrate

P2/L80: 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.

P3/L90: What about the contribution from the Sichuan Basin (China)?

P3/L93: '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;...)

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

P3/L108: '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.

P7/L256: '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 $\sim$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?

P7/L262: In the literature 'eddy shedding' is not the same as the bimodality of the AMA. Please clarify.

P7/L265: '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?

P8/L274: '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?

P10/L322: '(Randel and Park, 2006; Garny...)'-> '(e.g. Randel and Park, 2006; Garny...)'

P12/L359: '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,..)

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.

P14/Fig.3: Please explain briefly in the Figure caption why an application of an extinc-

tion filter is shown.

P18/L485: Please explain the meaning of the p-value in words.

P18/L506: 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.

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

P18/L511: Please clarify the meaning of 'increment' and 'correlation'.

P21/L559: 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.

P21/L564: '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.

P21/L567: 'full double-peak ATAL' (see above L559)

P22/L585: '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.

P22/L590: '..what has been reported in the past'. Please add some references.

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?

P22/L602: '... 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?

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