

Atmos. Chem. Phys. Discuss., referee comment RC2  
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## Comment on acp-2021-910

Anonymous Referee #2

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Referee comment on "Exploring the inorganic composition of the Asian Tropopause  
Aerosol Layer using medium-duration balloon flights" by Hazel Vernier et al., Atmos.  
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Vernier et al. discuss the results of balloon flights over India in August 2017 and February 2018 analysing the chemical composition of aerosol particles at 15-18 km altitude. To determine the concentration of water-soluble inorganic species they used a particle impactor on the balloon followed by subsequent ion chromatography in the laboratory. They found mainly nitrate and nitrite in terms of mass concentration during summer while sulfate and  $\text{Ca}^{2+}$  were detected during winter as major aerosol constituents. Backward trajectories in combination with chemical transport model simulations have been applied to characterize the history of the sampled air-masses. The main conclusion derived from the observation is, that particles containing nitrogen are a large part of the Asian Tropopause Aerosol Layer (ATAL). As a main result from the model simulations, the general importance of  $\text{NO}_x$  from lightning with respect to the nitrogen budget of the upper troposphere in summer is noted.

The paper presents some of the extremely rare in-situ observations of the composition of particles sampled inside the ATAL. Since this is an active scientific area of general interest and since a novel dataset is described, the work qualifies for publication in 'Atmospheric Chemistry and Physics'. However, in the presentation of the dataset and its interpretation there are some loose ends which are detailed below under 'specific comments'. These should be discussed and presented in more detail before I would recommend publication.

**Specific comments:**

*P2L7-9, 'The sampled air masses in winter 2018 were likely affected by smoke from the Pacific Northwest fire event in Canada, which occurred 7 months prior to our campaign, leading to concentration enhancements of  $SO_4^{2-}$  and  $Ca^{2+}$ .'*

- The influence of fires is not detailed in the main text. Please add a paragraph where this finding is explained in more detail as well as adding references.
- On P8L11,  $Ca^{2+}$  is referred to as an indication for mineral dust and  $K^+$  for biomass burning. How does this fit to the statement here, since  $K^+$  is not measured in the winter 2018 flight (see e.g. Fig. 2)?

*P3L3, 'World Health Organization recommendations':*

Please add a reference.

*P3L19:*

You may add here '(Wagner et al., 2020)'

*P4-5, chapter '1.3 What is known about ATAL's composition?':*

For the sake of completeness, the different views on mineral dust should also be mentioned, which is either predicted to be the major constituent of the ATAL (Fadnavis et al., 2013; Lau et al., 2018; Yuan et al., 2019; Ma et al., 2019; Bossolasco et al., 2020) or of minor importance (Yu et al., 2015; Gu et al., 2016; Yu et al., 2017; Fairlie et al., 2020).

P5L22, 'It translates into a mass concentration of 40 ng/m<sup>3</sup> assuming that the aerosols were liquid sulfate droplets':

- '...of around 40...'
- STP should be mentioned always, if it applies.

P6L28, Fig. 1, Fig. 2:

- Provide exact dates of all flights in the text .
- The flight abbreviations in Fig. 1 and Fig. 3 are mixed up: in Fig. 1 'ZFW' is the winter 2018 flight while in Fig. 3, it is 'ZF-1'. Please present those in a unique way.
- From the four flights listed in Fig. 1, only 3 are mentioned in the text. Please explain why.

P8L10-11, 'with traceable amounts of proxies for mineral dust (Ca<sup>2+</sup>) and biomass burning (K<sup>+</sup>).'

Please provide references for this statement and explain it a bit more.

P8L20, '...but did find the same in the flight samples of winter (Fig. 3).':

Please formulate this sentence clearer. As it is written, one could think that ammonium has been found during the winter flight, which is not the case according to Fig. 3.

*P8L26-P9L6:*

Might this be an explanation that no ammonium has been found, but nitrate? Please elaborate on this.

*P9L9, 'observations from the CALIOP lidar onboard the CALIPSO satellite.':*

How has the CALIOP data been averaged? Which co-incidence criteria have been applied? Has any cloud-clearing been used.

*P9L14, 'likely made of aspherical particles':*

Does this mean that during this part of the balloon flight, air has been sampled within a cirrus cloud? If so, please state this clearly and any implications this might have on the analysis.

*P9L18, 'was done for more than 1 h in a cloud-free region enhanced with aerosols above.':*

Please formulate this clearer – one could interpret it as if the aerosols have been above the balloon.

P9L21-24:

Is this a general comment on cirrus above Gadanki or does it refer to the situation during one of the flights. Please be clearer here.

*P9L24-26, 'Moreover, the increasing fraction of sub-visible cirrus clouds between 1998-2003 probably modified on the temperature and the water vapor budget in the Tropical Tropopause Layer (Pandit et al., 2015).'*

There is something wrong with this sentence.

*P10L1, 'trajectories (black lines) and deep convection influence 1 (red dots).'*

I cannot see this in Figure 5.

*P11L4, 'CO, nitrate, sulfate, and black carbon (BC) aerosol concentrations':*

- To support a discussion on mineral dust and to compare with the observed  $\text{Ca}^{2+}$ , it would be good to show here also the model perspective.
- Please also provide the model maps of  $\text{NH}_4^+$  and discuss differences between model and the lack of ammonium in the observations.

*P11L11, 'nitrate are significantly lower':*

Please provide here numbers (...% lower). From Fig. 7 it seems that these concentrations are not a much lower.

*P11L20, 'The NO<sub>x</sub> lifetime is believed to increase downwind from the outflow':*

Please provide a reference for this statement.

*P11L31-P12L4, 'Fig.5 shows that GEOS-chem could simulate convective activities reaching levels between 14-15 km ...':*

Please describe more clearly where this is the case. Fig. 5 does not reach down to the ground, so one cannot judge if the transported air stems from the boundary layer. Further there are cases with indications of convection from HIMAWARI which are not captured by the model (ZF2, highest altitude at around 105 deg East).

*P12L11, 'We note that sulfate along the trajectories influenced by Chinese pollution during ZF2 increase significantly...':*

- What means 'significantly' here? Please provide numbers (by ...%).
- Please provide possible explanations (including references) why sulfate is modelled too high compared to the measurements.

*P12-13, chapter '7. Summary and Conclusions':*

- Please discuss also the representativeness of these balloon observations lying more at the border of the AMA for the ATAL as a whole.
- Please explain also the relevance of your findings on  $\text{Ca}^{2+}$  wrt the question of mineral dust as a major constituent of the ATAL (see comment and references above).
- Please discuss also your finding of high concentrations of nitrite. What could be a relevant mechanism for its production?

*P13L8-10, 'parcels, the model ability to simulate convective influence at higher altitudes seem to be limited.':*

This is not mentioned in the main text – please explain it, where the influence of convection and the ability of the model to simulate convection is discussed.

*P13L17, 'where smaller nitrate particles were found which could also indicate the influence of new particle formation.':*

Also this is not discussed in the main text. Please provide an explanation where the relevant Figure is explained including possible references.

#### **Technical comments:**

*P1L39, 'STP':*

Explain abbreviation.

*P2L5, 'with particle size radius (0.05-2µm)':*

- 'with particle size radii from 0.05 to 2 µm'
- Here and all over the text: there should be a space between number and unit

*P2L6, 'mass':*

-> 'masses'

*P2L37, 'have':*

-> 'has'

*P3L25, 'of ATAL':*

-> 'of the ATAL'



*P5L6, 'Höpfner et al., 2016':*

-> 'Höpfner et al., 2019'

*P5L21, 'a concentration of 20 particle/cm<sup>3</sup>':*

-> '... of about 20 ...'

*P6L26, ', to':*

-> '. To'

*P8L14, 'Ca<sub>2</sub><sup>+</sup>':*

-> 'Ca<sup>2+</sup>'

*P8L19+22, 'Hopfner':*

-> 'Höpfner'

*P9L16, ', pressure':*

-> ', the pressure'

*P10L8, 'We conduct GEOS-Chem':*

-> 'We have conducted GEOS-Chem'

*P10L30, '2.5° by 2° horizontal':*

-> 'latitude x longitude' or 'longitude x latitude' ?

*P11L12, 'is':*

-> 'are'

*P12L11, 'increase':*

-> 'increases'

*P12L13, 'NO3':*

-> 'NO<sub>3</sub>'

*P12L20, 'onboard':*

-> 'aboard' or 'on board'

*P14-20, refernces:*

doi' s are missing for some the references, please check.

*P25, Fig. 4:*

Why are only positive ascent rates given in the color scale?

P26, Fig. 5:

x-axis title missing in bottom right panel.

### References:

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