

Atmos. Chem. Phys. Discuss., referee comment RC1  
<https://doi.org/10.5194/acp-2021-910-RC1>, 2021  
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## Comment on acp-2021-910

Anonymous Referee #1

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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.  
Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-910-RC1>, 2021

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Vernier et al. present observations of aerosol mass concentrations and other in-situ observations collected with a balloon that sampled the Asian Tropopause Aerosol Layer (ATAL). The authors conclude from the aerosol collected on filters and analyzed offline, that nitrate and nitrite are major constituents of aerosol in the ATAL. Then, they compare with satellite retrieved data to show that the balloon was within the ATAL concurrently as a CALIOP overpass. Finally, they use back trajectories and GEOS-Chem model results to investigate the sources of the aerosol collected onto the filter.

Though it may be of interest to ACP, the authors need to address the comments below for the article to be accepted. Further, as the article is currently written, and depending on how the authors respond to the comments below, the article currently seems more appropriate for Measurement Report instead of a research article for ACP (further discussed below).

### Major Comments

- Here are some examples of how this paper seems more appropriate for Measurement Report.

1.1 The authors mention that both nitrate and nitrite were important constituents of the aerosol composition; however, they only really focus on the potential sources of nitrate. Nitrite is a generally unusual component of aerosol composition and I would expect to have different physicochemical properties than nitrate, including in the ATAL and its impact on cirrus clouds and radiative properties. A discussion on nitrite, including sources and what form it is in the ATAL would improve the paper and make it more appropriate for a research article than a measurement report.

1.2 The authors mention that the GEOS-Chem predictions have much higher sulfate than the filter measurements and provide one short sentence speculation about why without further discussion or ramifications in a model having too high sulfate compared to observations. Ramifications of too high sulfate should be addressed, as that is an important finding. Also, how does the sulfate observations here compare to prior studies?

1.3 The authors use GEOS-Chem as is without any verification if the model predictions match observations (e.g., is CO at the right location / concentration). Satellite retrieved observations of CO could help or even be used instead of GEOS-Chem CO to make this point. Further, other satellite products could be used to try to make the points the authors are trying to make here. On top, it is unclear how well GEOS-Chem is performing for nitrate compared to observations. One obstacle behind this comparison is (a) was ISORROPIA used to predict the thermodynamic partitioning of nitrate with aerosol phase and (b) what nitrate was used within ISORROPIA. If, for example, GEOS-Chem only has ammonium nitrate and not nitric acid trihydrate (NAT) or more refractory aerosol (sodium or calcium nitrate), then the discussion about nitrate with the filter measurements does not work here.

1.4 The nitrate being either NAT or potentially refractory aerosol and not ammonium nitrate is an important finding, especially in regards to the physicochemical properties of aerosol in ATAL. However, this is not explored and expanded upon. Also, could this be a measurement artifact (see next comment)? The fact that prior studies observed ammonium nitrate and not the aerosol reported here needs to be further explored as the findings here are very different.

1.5 The authors make a speculation that Canadian wildfires impacted the ATAL in the abstract and the conclusion. With it being in the abstract, this would lead readers to expect more attention on this detail in the paper instead of just a brief passing sentence in the conclusion. This could be a section in of itself, especially regarding how forest fires could impact ATAL. Currently being in just abstract and conclusion further makes this paper feel more like a measurement report instead of a research article.

2. Page 8, line 26 – 31 and page 9, line 1 – 6, the authors discuss how ammonium nitrate could be lost on filters. However, there was no discussion if experiments were conducted to determine how much loss occurred. Further, the authors mention NAT (page 8, line 15) could explain the lack of charge balance in the observations they show. But, there is no discussion about how stable NAT would be on filters prior to and during the freezing of the filters and during the preparation of the filters for sampling. Further discussion / exploration of this is needed to put the paper into context of prior studies and for use in comparisons against chemical transport models.

3. Page 9, line 17 – 18, the authors quickly mention the pump was on for 16 minutes. This is significantly shorter than the 2 hours the authors said was needed to collect enough aerosol to have measurements above detection limit. This needs to be clarified.

4. The description of GEOS-Chem needs to be moved to methods, and further description of GEOS-Chem needs to be provided—was ISORROPIA used, what forms of nitrate were included, was nitrite in the model, how was SOA modeled, etc.

5. There were four flights of the payload, yet only three flights are shown. Why is one flight not included in Fig. 3 or any discussion?

#### Minor Comments

- Page 10, line 1, the authors have the description of trajectories and deep convection different than in Fig. 5.
- Page 11, line 16, make sure Chem in GEOS-Chem is capitalized.
- Page 11, line 18 – 20, the authors make a statement about upper troposphere (UT) NO<sub>x</sub> lifetime and its sinks. However, the normal thought about UT NO<sub>x</sub> lifetime is that it is about 2 days. Where did this shorter lifetime and sinks come from?
- Page 12, line 4 – 8, the authors use mass concentration for CO instead of volume mixing ratio; however, in the figures, CO is reported in volume mixing ratios. Please clarify which is correct.
- Page 12, line 7, the should not be capitalized.
- Fig. 1, a picture or diagram of the science payload would be good here.
- Fig. 6, the white dot is barely visible and even missing, maybe, in some of the panels. Make it more prevalent to see it.
- Fig. 7, what is the horizontal bar from the left axis to ZF2? What is the nitrate observed for a comparison point? Also, red and green (also for other figures) is generally not a good combination for colored blind readers.
- Fig. 8, it was mentioned dilution of CO is observed. The figure makes CO look relatively flat.