

## ***Interactive comment on “Radiative and chemical implications of the size and composition of aerosol particles in the existing or modified global stratosphere” by Daniel M. Murphy et al.***

**Anonymous Referee #1**

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The manuscript “Radiative and chemical implications of the size and composition of aerosol particles in the existing or modified global stratosphere” by Murphy et al., describes measurements of stratospheric size resolved aerosol composition from optical particle counters and the PALMS aerosol mass spectrometer flown on the ATom aircraft missions in 2016-2018. The authors discuss the compositional signatures that could be indicative of aerosol transport and formation pathways that yield aerosol with two distinctly different size modes. The radiative implications of these two distinct size modes are discussed and finally the implications of these for future geoengineering. Significant anomalies in stratospheric aerosol loading and composition that are associated with volcanic eruptions, stratospheric smoke injections and transport of dust into

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the stratosphere are mentioned but not discussed in detail. Efforts to simulate the observed stratospheric aerosol size distributions using the CESM/CARMA model are briefly described.

The measurements of size resolved composition of lower stratospheric aerosol described in this work are unique, novel and of significant scientific importance. As this manuscript correctly describes, these results have broad implications for stratospheric chemistry and radiation and thus climate. The authors present an overview of the composition measurements in sections 1-4 at a high level that provides a brief description of the inherently complex topic of size dependent aerosol composition in the UTLS. However, the superficial description and analysis of the data set presented in this manuscript is insufficient to support the broad and generalized observations and conclusions presented in Sections 5 and 6. Many of these observations and conclusions are likely to be correct, and may be supported by this ATom data set, but have not been demonstrated to the reader in this work.

As this appears to be the first description of the PALMS composition measurements from the ATom campaign, the brief and qualitative description of the sampling and do not allow for the reader to understand how representative or significant the compositional analysis is, how definitively tropospheric and stratospheric air masses are separated, or even basic information such as how the ozone measurements were acquired. While the authors state that about 10,000 particles were sampled in the Northern Hemisphere (line 238) there is no indication of how these particles were divided between the various flights, how many flights there were, at what locations these particles were sampled, what fraction of the data was from tropopause folds, or how many particles were measured in the Southern Hemisphere. Figure S1 provides some insight into where the measurements may have been made, but as the color bar in S1 does not correspond with the 250 ppbV cut off for stratospheric air, even this provides insufficient information. Due to the low ceiling of the DC-8, the authors indicate that the sampled “stratospheric air was always associated with low tropopauses,

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sometimes in tropopause folds". Given that the upper tropospheric organic aerosol loading can be very substantial, particularly with the wild fire activity likely present for ATom1 and ATom3, it is important to know whether these data are from stratospheric measurements above the broader tropopause (less likely in the summer time) or from tropopause folds (more likely in the summer time) and how edge effects and mixing were considered for the latter.

Without the presentation of a more thorough analysis of these data, it is not clear that some of the major conclusions of the paper are solidly supported. It may be the case that the transport of tropospheric organic-sulfate aerosols is producing a second smaller stratospheric aerosol mode centered around 200 nm in some specific cases. But this is not robustly supported in 5 of the 8 cases show in Figure 1. In ATom4 NH, the peak near 200 nm appears to primarily driven by an increase in pure sulfuric acid aerosol. In only one of the SH cases (ATom 3) is there convincing evidence of a secondary mode, and again in this case it appears to be driven by sulfuric acid aerosol (presumably from Calbuco). Without much more detailed description and analysis it is hard to be sure that the conclusion that tropospheric aerosol significant to the global lower stratosphere (paragraph starting on line 450) are really supported by this data set. This would be a very important result and I encourage the authors to layout a more convincing case based on the ATom data set.

In section 4, the authors state "As expected for their sources, the concentration of meteoric-sulfuric particles increases with altitude and the concentration of organic-sulfate particles decreases with altitude." While it does appear to be clear that concentration of meteoric-sulfuric aerosol increases with ozone/altitude, the data presented here do not appear to provide convincing support for a general decrease in organic-sulfate particles with altitude. This relationship appears to be fundamental to apportioning the source of these particles to transport across the tropopause. Three of the organic-sulfate profiles shown in Figure 4 appear to be constant with respect to ozone/altitude, three may show a decrease and one shows an increase. The signifi-

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cance of these slopes is hard to determine without some metric of the measurement uncertainty. From the narrative, it appears that there was a limited (but unspecified) amount of particle composition data at ozone levels above 500 ppbV, so an indication of how significant the measurements > 500 ppbV is necessary to interpret the importance of this slope. Comparing the ATom 2 data in Fig 3 with Fig 1, it appears that there is in fact no significant difference in organic-sulfate particles in the 200nm mode, and actually a significant increase in the amount of organic-sulfate in the 600nm mode. The tropospheric origin of these organic-sulfate particles could also be indicated by their compositional signature, as the authors state on line 143. This is not demonstrated to the reader, even though the data must surely be available.

Throughout the paper there is no quantitative discussion of the uncertainty or statistics associated with this highly averaged data (in the narrative or in any of the figures). This issue is addressed anecdotally when convenient. For example, in the caption in figure 1 the authors do state that there 'may be artifacts' due to Mie resonance for certain features but other features are robust without any further explanation or justification. If there were other publications describing this data set in detail, some of these considerations could be addressed through references to these publications, however it appears that these publications are planned for the future.

Section 5 of the manuscript primarily describes the radiative and chemical impact that arises from the divergent size modes between aerosol of tropospheric and stratospheric origin. The narrative in section 5 is somewhat disjointed and difficult to follow as it is not always clear how it relates back to the measurements presented in the first sections of the paper. For instance, in section 5.1, the infrared absorption is only discussed in the context of particle volume, yet while the authors acknowledge that particle composition plays a role (line 266) this is not tied back to one of the primary observational findings, that the two size modes have significantly different composition. There are also several results that described in section 5 that are not demonstrated to the reader. For example, on line 329: "The relative contributions to scattering of light at < 240 nm

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are fairly similar to the surface area panel in Figure 7 except that sizes smaller about 80 nm and larger about 600nm contribute less to UV scattering than they do to surface area.” While this statement is quite likely true, it has not been shown and it is hard to quantify what ‘fairly similar’ describes. A similar statement begins on line 313 “On Figure 7, the sedimentation flux as a function of size would be slightly more skewed to large diameters than the light scattering panel.” Such generalized statements need to be shown to be supported by the data.

Section 6 of the manuscript sets out to describe the relevance of this work to volcanic eruptions or to future geoengineering projects. While the discussion is interesting it is highly speculative and largely unrelated to the data presented in Sections 1-4. For example, in the paragraph starting on line 396 it is implied that differences between the modeled sulfuric acid particle sizes and these measurements (Figure 2) is a consequence of one of the main results that is presumably shown by the data – multiple sources of stratospheric aerosol. However, no details on the initialization for the model run or analysis of what is driving these discrepancies is provided, as a result the comparison in Figure 2 is largely anecdotal.

Finally, the title of the paper “Radiative and chemical implications of the size and composition of aerosol particles in the existing or modified global stratosphere” is misleading. While the aerosol composition data is presented in a limited way, the implications (described in sections 5 and 6) are based entirely on the size distribution of the particles not their composition, even when composition would certainly be important to these implications (e.g. 5.2 Scattering and 5.3 chemistry).

Technical Comments:

Line 54: “The moles of oxygen were approximately four times sulfur plus about 0.2 times carbon” - rephrase to make it clear where the brackets are  $O_2 = 4 * (S + 0.2C)$ ?

Paragraph starting line 95: It is unclear why the OPC data is needed if PALMs provides sizing?

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Line 174: Using ozone as a proxy for altitude is understandable, but some indication of what an equivalent tropopause relative altitude or range of altitudes that a given ozone mixing ratio corresponds with would be useful.

Figure 6: What is the unit of measure for ‘Approximate net cooling’ and how was this calculated?

Figure 7: The units of these plots are unclear. Either provide units, or normalize the data to make it clear that it is relative surface/scatter/volume

Figure 8: This figure is confusing. What is meant by the IR-heating trace? Is the peak at 0.5 the most relative cooling per IR heating, and would that mean the least or most IR heating?

Figure S1: It would be useful to start the color bar at 250ppbv so that only the points used in this study are highlighted, with a clear distinction for where the particles shown in Figure 3 were found.

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