

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 #2

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The LMS is an important part of the stratosphere and difficult to observe. Its composition differs from the rest of the stratosphere in many ways due to the mixing of tropospheric and stratospheric air. The study by Murphy et al. is based on in-situ data from aircraft of aerosol particle size distributions and composition of individual particles. Particles were classified depending on their composition to study the history of the particles. Radiative impact was discussed in relation to volcanic eruptions and climate engineering.

I find large issues with the authors' interpretation of the data. One reason is the generalization in sections 5-7 based on few data (a single season in a single year). Another

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is the data on organics, and the claim that the organic aerosol comes from the troposphere only, which contradicts previous work. No uncertainties are presented for the observations as far as I can tell, and there are no statistical analysis to support the claims of trends.

I find several shortcomings in the manuscript and cannot recommend publication without changes in the data analysis / interpretation of the data.

Major comments

1. Uncertainties and quantification a. How large are the uncertainties in the observations? I don't find any numbers on that. b. How well can one quantify different aerosol constituents from the PALMS data? c. Is it possible to tell whether there is a trend, as in Figure 4, if there are no statistical analysis to support it?

2. There are too little organics in the data compared to previous work, and this is not discussed in relation to those studies. a. There is no discussion on organics after Calbuco. Several other groups have found large amounts of organics in volcanic aerosol. Two examples are Schmale 2010 and Andersson 2013 reported much higher organics (or carbon) to sulfate ratios in volcanic particles. Those data were from AMS and Ion beam analysis. b. Vertical gradient in organics is different from Martinsson 2019, who reported higher carbon abundance deeper into the LMS. I cite from their abstract: "...the carbonaceous and sulfurous components of the aerosol in the lowermost stratosphere (LMS) show strong increases in concentration connected with springtime subsidence from overlying stratospheric layers. The LMS concentrations significantly exceed those in the troposphere, thus clearly indicating a stratospheric production of not only the well-established sulfurous aerosol, but also a considerable but less understood carbonaceous component. . ." c. There is no real discussion on wildfire smoke. The smoke from the Aug 2017 fires in western North America is evident in Figure 4. The relatively small impact on the organics after the wildfire in that figure is strange given that the event was almost volcanic sized (Peterson 2018). In Figure 1, it is appar-

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ently less organics 1-2 months after the fire than in spring the next year. Why is that? The discrepancy between the data in the manuscript with data from other studies is not discussed as far as I can tell. Identifying particles containing organics is not the same as measuring the mass (of organics or carbon), which other techniques do.

3. Large part of the manuscript is focused on a single season in the Northern Hemisphere (Atom2). It is unclear to me why this is the case. Sulfate concentrations in the LMS varies with season due to both seasonal variation in subsidence from the stratospheric overworld and varying cross TP transport. Stratospheric influence is large in winter and spring, and low in summer and fall. Thus: a. Data presented for a specific season is representative for that season only, and not for the entire year. b. General climatic conclusions cannot be drawn from a single season in a single year.

4. Data after the Calbuco eruption are almost not discussed at all. The authors mention the eruption and that it had some impact on the sulfuric acid but no more details. I understand that the authors focused on the Northern Hemisphere, but this omission is strange to me. I expect some discussion on data that are included in a manuscript.

Other comments

1. I would like to see a more clear discussion on the history of sulfuric particles. Meteoric ones from the upper stratosphere, and pure sulfuric from the lowest stratosphere (directly from the tropical lower stratosphere in the BDC).

2. The ExTL has very different composition than the rest of the LMS and (stratosphere). The author never mention the ExTL. Why is that?

3. I think that the phrase biomass burning shall be changed to wildfire smoke since it comes from PyroCb intrusion(s) to the stratosphere. Biomass burning leads the reader to believe that it is a general upwelling from diffuse fires instead of PyroCb formation in enormous fires.

4. I think that the manuscript should have a concise conclusions section after the

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discussions section.

L45. "...The local tropopause and slightly above the altitude of the tropical tropopause...". This is not true. The tropical TP is located at ~17 km, and the LMS extends to ~14-15 km in the extratropics. L135. "...The primary source of sulfuric acid in the stratosphere, oxidation of carbonyl sulfide, is similar in the two hemispheres...". Do you mean the background stratospheric aerosol? It is still an open question whether SO₂ or COS is the most important for the background stratospheric aerosol. L183. Change to: ...Northern hemisphere than in the Southern Hemisphere... L191. I do not understand the statement: "...the organic-sulfate particles contain on average about 40 to 80 organic material by mass..." L260. "...Alumina with the size distribution from rocket emissions was calculated to cause net warming (Ross and Sheaffer, 2014)...". This is a strange comment in the end of a section on volcanic particles and IR. Are there a sentence missing? L361. Change "will it" to "it will" L469. "...the modeled particles have about 65% of the climate impact per unit mass as the observations, 160% of the surface area, and sediment about 60% as fast..." Is this referenced to findings in the manuscript? Are there actual observations of these quantities or do you mean estimated from the PALMS observations?

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