Dear Dr. Daniel Murphy,

According to the referee comments and also in my point of view, your manuscript still needs minor revision before acceptance for publication in ACP. I am concerned with the content of 'Section 6 Relevance to volcanic or intentional aerosol injection', which seems not to have a strong linkage with (or supported by) measurement results presented in previous sections. I would suggest that Section 6 be skipped over or reduced and then merged into Section 7. Below are specific comments, some of which are technic.

Section 6 has been reduced, with less emphasis on intentionally introduced material. Some text has been deleted and some moved into sections 5 and 7.

L24: The expression 'the surfaces used' is not so clear. Do you mean 'the substances used'?

Done

L31: over 60 years since 1961?

Done.

L59-60: Do you mean most "single" particles larger than about 110 nm? Are these particles internally mixed, e.g., containing an organic core coated by sulfate or vice versa?

The text has been modified.

L89-97: While the UHSA and LAS recorded the data at 1s intervals, how about the sampling frequency of PALMS for particle composition?

PALMS records single particles, so each particle is associated with a particular time. The averaging needed for PALMS data is discussed in the paragraph starting on line 115.

L138-139: What radiative transfer model was used? Better to provide some information here, instead only referring to Murphy (2009).

There is no radiative transfer model required, only Mie scattering. The text has been changed.

L208-209: It is difficult to follow the discussion here. How are the tropospheric mode and stratospheric particles defined? Please describe the method or threshold used to distinguish the modes (aerosols) between the stratosphere and troposphere here and/or in the caption of Figure 2. If applicable, the same legends can be used in Figure 2 as in Figure 1.

Text has been added to the Figure 2 caption to better compare to Figure 1. Text has also been added near line 110.

L244: The phrase 'were both local springtime' can be changed to 'were observed both in local springtime'.

Done.

L255: Since ammonium sulfate aerosol has very low volatility, it might not be so meaningful to discuss the equilibrium of ammonia with it. Did you find ammonium nitrate by the PALMS? Otherwise, I would suggest that ammonia be omitted.

Even very small (~ 1 pptv) amounts of ammonia would be very important for new particle formation, so it is important to constrain the concentration. This is now mentioned in the text/

L268-269: Does the CESA/CARMA model include these species and associated gas-particle processes? If applicable, what specific species used in the model?

The CESA/CARM model does include secondary organic chemistry and gas-particle processes (Yu et al., 2015). This is now mentioned in the text/

L374: Change 'than in the stratosphere than' to 'in the stratosphere than'.

Done.

L486-509: It is difficult to follow the discussions in these two paragraphs. While it is declared at the beginning (L488) that infrared absorption is almost independent of size, the infrared effect (L490), relative infrared effect (L492) and infrared absorption (L496) are said to be size dependent. I could not see the infrared effect on net cooling as a function of size from Figure 7 as stated in L489-490, neither high sedimentation rate (downward triangles) in Figure 9. The 'IR heating' appearing in the legend of Figure 9 is unclear.

The text has been modified here and in the captions of Figures 7 and 9 to clarify the which effects are per unit mass and which are per amount of cooling. The infrared absorption per unit mass is almost independent of size; the infrared absorption per amount of cooling of the Earth is strongly size dependent.

L547-549: This paragraph seems to be repeating the discussion given in previous section.

Yes is it, this is a summary section that repeats some key points. The reorganization of sections 6 and 7 may help this.

Figures 7 and 9: The model and input parameters used to derive these two plots are not well described. What data from the ATom mission are used for the calculation?

These are model calculations. There are no ATom data in Figures 7 or 9. They are, however, crucial to the discussion of ATom data. The middle panel of Figure 8 is essentially the ATom data in Figure 2 (solid lines in Figure 2) multiplied by the calculations in Figure 7. Figure 9 generalizes some implications of Figure 8 to arbitrary sizes.

Sincerely, Jianzhong Ma