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Discussions

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Interactive comment on "Composition and oxidation state of sulfur in atmospheric particulate matter" *by* Amelia F. Longo et al.

Anonymous Referee #1

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This paper discusses the compositon and the oxidation state of sulfur in atmospheric aerosols collected in the general Atlanta area. The overall work is of interest to the commiunity and is well written and has adequate discussion regarding the basic results. Some better presentation of the data and results and clarification of data processing methods however, should be included in at least the supplemental materials.

Specific Comments:

page 5, line 15: I very much like the addition of the sulfate standard database in the supplemental material. However, can the authors address the potential of self-absorption effects of sulfate standards? Particularly as this may possibly contribute to broadness and amplitude reduction of some of the standard peaks? Can the authors elaborate more on wether these data were collected in the bulk or microscale mode?

Any more info on the standards particle size, more than "homogenized"?

Suplemental material should at least include some plots of NEXFS spectra and their fit results of some typical sample to be able to evaluate the data quality and method of fitting. Both types of fitting with the linear combination of sulfate standards and the use of gaussian functions should be shown.

page 5, line 34: The authors follow a rigourous methods of following the monochromator energy drift. However, they do not reference what was mono energy was calibrated with initially? This information is critical to allow comparison to any other published dataset of sulfur spectroscopy. Was there any calibration of the sulfur concentration in the samples? If so, was this a theoretical calculation or an empirical calibration. It would be useful to discuss not only the relative changes in oxidation states, but the overall concentrations observed as well. This information would help in examining the various sources.

Fig S1. I would like to see more information regarding the multiple energy maps. What are the units of the map for each sulfur oxidation state? How were the units determined. Is the intensity here the intensity of the sulfur at each of the white line energies, or was a fit done to calculate the various proportions of each oxidation state? The latter would be a much more rigourous method, as there could be intensity of S0 and SVI is not completely unique to each of the white lines – that is there are contributions of each species to each of the measured energies. A proper method would be to measure the intensities at least N+1 energies (given S0 and SVI, N=2) and do a linear combination fit to determine the N species present at each map pixel. The choice of white line energies comared to the standard library seems off – there white line in the library is at ~2483 eV for sulfate typically, whereas the maps were stated to have been measured at 2480 eV. One would expected, based on the apparent energy calibration, that S0 would appear at 2474 eV. Given the 2480 energy, one may observe some signifcant intensity of sulfates such as the ammonium sulfates around the energies of S0. Could this mislead the interpretation of S0 in the multiple energy maps if a careful fitting was

not performed?

Technical Corrections : page 5, line33: Did the experiment actually use Vortex SDD with 5mm2 area? This seems very small for standard XRF analysis. The more standard value for analysis is 50mm2. page 9, line 5: Text refers to Figure S4. This does not exist. Do the authors mean Figure S1?

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