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Atmospheric Chemistry and Physics Discussions

Interactive comment on "In-cloud sulfate addition to single particles resolved with sulfur isotope analysis during HCCT-2010" *by* E. Harris et al.

Anonymous Referee #2

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This work quantifies the relative magnitude of different sulfate production pathways in orographic clouds during the Hill Cap Cloud Thuringia campaign in Fall 2010. The unique aspect of this work is that the analysis is done on an individual particle level, combining SEM and NanoSIMS analysis, and using the sulfate isotope $\delta^{34}S$ as a marker. The results show that when it comes to in-cloud sulfate production, not all particles are equal, and that the dominating pathway of sulfate production depends not only on particle size but also on the chemical composition of the aerosol particle that grew into a cloud droplet.

This is a very original paper that takes the process analysis of in-cloud chemistry to the next level and reveals a complex picture of sulfate production depending on the per-particle composition of the particles that serve as CCN. Studies like this are important

to guide future model development. The paper fits well into the scope of ACP, and I recommend it after a few minor suggestions for clarification are taken into account.

1. On how many particles in total is this analysis based on? I would assume that given how labor-intensive the procedure is that the analyzed particle collective is rather small. Then, how representative is the particle collective of what is going on in these clouds?

2. Page 2943, line 11: I understand that the question of connected flow is addressed in a separate paper, but it would be helpful to briefly what you mean by the "coefficient of divergence" for particle bins and ozone concentration.

3. Figure 3: What is PBA? Also, based on this figure, I can see how to separate out the dust particles. But how are soot, coated soot, and mixed OA/IA separated without SEM analysis, as stated on page 2950, line 6?

4. Table 1: In the column "Description", you refer to several times to "solid" particles. I believe this does not necessarily have to be the case. For example, the first process ("CON") can occur on a solid or liquid particle, and similar for the processes SCAV and COAG.

5. Caption for Figure 5: Word missing: "Straight lines show the isotopic composition of sulfate that could "be" added to particles in the cloud . . ." Mention that the values of the straight lines are the ones listed in Table 3.

6. Conclusions: It would increase the impact of this paper even further if you could give the modeling community some guidance as to "how wrong" their answer would be if they do not take into account the variation in particle composition for modeling sulfate production. For example, can you compare the estimated sulfate production within the cloud assuming an internal mixture for the particle population to the sulfate production based on the different particle types?

7. Lastly, apart from sulfate there is significant in-cloud production of organic matter.

Is it in principle possible to apply your method to these reactions? A comment on this would be helpful.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 2935, 2014.