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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Surface modification of mineral dust particles by sulphuric acid processing: implications for CCN and IN abilities" *by* P. Reitz et al.

Anonymous Referee #2

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The manuscript by Reitz et al. contains valuable information for a better understanding of the reduced IN ability of mineral dust particles that were coated (and further processed) by sulphuric acid. The manuscript is part of a series of recent publications on the FROST 1 and 2 measurement campaigns with a special focus on the particle characterization by means of aerosol mass spectrometry. Based on the analysis of fragment patterns of differently treated particles, some conclusions (hypotheses) about the potential reaction pathways on the particle surface could be drawn. I consider the presented material as appropriate and important for further publication in ACP, but, in the same way as indicated in the previous referee report, I also had problems to understand all aspects of the discussion. In particular, there is a bad link between the

manuscript text and the related, often very busy figures. On the one hand, some experiments listed in the figures are never referred to in the actual manuscript text. On the other, I sometimes felt like "left alone" to search for a corroboration of a statement made in the manuscript text in one of the busy figures. So the readability of the manuscript must be improved before final publication. In the following, I address my major concerns and try to make some suggestions for the improvement of the discussion.

Major comments:

1) Section 2.2.3, addressing details of the AMS measurements and necessary correction factors, is hard to read for a non-expert in AMS measurements. On page 7243, line 22ff, the authors introduce which information is contained in the following paragraphs, but I would definitely suggest to split the following information into further subsections like "Correction for the collection efficiency", "Correction for the lens transmission" etc. On page 7244, line 1, the first sentence addresses the instrument calibration but the next sentence then already addresses the collection efficiency. The AMS calibration is then again referred to later on page 7246, line 17. These two parts should be grouped together.

I did not really understand how the comparability factors shown in Table 2 were derived. Concerning the statement on page 7246, line 10: Why do you refer to the CCNC measurements in this context? Are they also affected by the diluton stage? The comparability factors imply that there is a 4 – 5 times difference in the uncorrected AMS data for the same experimental conditions between FROST 1 and 2. How does this compare to the assumed uncertainty of only 10% for the dilution stage? What are the other speculative sources mentioned in line 5?

2) Section 3.1, discussion of Figs. 6a-c: The authors have obviously put the results from all individual FROST 1 and 2 experiments into the figures which makes them very difficult to read and to extract the major conclusions. A particularly bad example is Fig. 6c which is only described by a few lines on page 7251 and also contains experiments

(like the one labeled “lr” with the longer residence time) that are not discussed and explained in the manuscript text at all. I understand that the authors want to show all of the experiments – but then they could e.g. include an overview table which shows the experimental conditions of all experiments performed during FROST 1 and 2, and then simplify Figure 6c by just showing a set of examples (e.g. just one conditioning temperature) that underline the major conclusions that are described in the manuscript text. In the manuscript text itself, the authors should add more references to the experiments in the figures to which the current discussion is related to. For example, in the paragraph starting on page 7250, line 28, they presumably address the final experiment shown in Fig. 6a that shows the reduction in the sulphate mass. It would be much easier and a better guide for the reader if this were also announced in the text. Figure 6c e.g. contains too much information. Concerning the statement on page 7251, line23: The reader would more easily see this effect if only an exemplary set of experiments that underlines the subject under discussion would be shown.

3) Section 3.2: Please only show a selection of experiments in Figs. 8a/b that underline the conclusions from the manuscript text (e.g. just show experiments for a single bath temperature in Fig. 8b). If you state in the manuscript text “this difference is clearly visible” (page 7252, line 7), please mention specific experiments in the figures where the reader can actually see this difference. In Fig. 8a, there is an experiment labeled > 300 for the particle mobility diameter which shows a huge signal of the HSO3+ ion although using the water bath. What is the difference to the left-hand counterpart? I really had problems to extract the key aspects from the discussion in this section (e.g. concerning the role of ammonia). There is just one long paragraph, first addressing the results from the fragmentation patterns in Figs. 8a/b, and then e.g. in line 13 on page 7252 - without any break - there is a statement that obviously refers to some experiments from Fig. 6c. As a reader, I also somehow felt like being left alone with the reaction schemes presented at the end of this chapter – I had to read again the previous discussion to reconstruct the proposed reactions. In my opinion, this could be much better arranged. The authors could, e.g., first introduce the reaction scheme and

then explain step by step by addressing the observations from Figs. 6 and 8 why the proposed reactions are the most likely pathways. In a separate paragraph, e.g., the authors could summarise the key differences for the experiments with the water bath compared to those with the unprocessed SA coating, leading to the proposed reactions (R1) and (R2). In the next paragraph, the authors could address the “thermodenuder only” results and finally the water bath + thermodenuder experiments. Why is the the 250°C thermodenuder reaction in line 10 doubly labeled (R6 and R7)?

4) Section 3.4: Some general questions concerning the discussion: Can the authors estimate a coating thickness based on deduced mass per particle results? On page 7260, line 1, they state that only after humidification there is a highly concentrated sulphuric acid solution around the particle – does the humidification not lead to a reduction in the sulphuric acid concentration? What is the RH in the humidification section so to estimate the sulphuric acid concentration? For how long are the particles exposed to the increased RH? (residence time) Are there literature findings that show that less concentrated sulphuric acid solutions more easily react with carbonates etc.? And an important issue that is completely missing in the discussion: How do the applied particle treatments relate to processing that actually occurs in the atmosphere?

Minor comments and technical corrections:

1) Page 7238, line 19: “. . . better understanding . . . for the ability of mineral dust particles . . .”

2) Page 7239, line 4ff: It was not clear to me which median mobility diameters were obtained with the different impaction stages.

3) Page 7239, line 9: mobility

4) Page 7240, line 14: Both CFDC and LACIS data are shown in Fig. 11 and Fig. 12, respectively.

5) Page 7241, line 6: “constant that depends on . . .”

6) Page 7243, line 9: “further restriction of”

7) Page 7243, line 16: Why “nevertheless”? One could state: “Rather, the instrument is sensitive to the more volatile fraction of the particles, thereby enabling the detection . . .”

8) Page 7244, line 22: “in our studies” ?

9) Page 7245, line 2: “. . . the following assumptions were made.”

10) Page 7245, line 10: “to be approximately”

11) Page 7245, line 17: “which were used”

12) Page 7247, line 9: Please include a reference for the “common method”.

13) Page 7247, line 11: “Particles showing a peak at m/z = -97 (HSO4- ?) in the mass spectra were identified as sulphates.”

14) Page 7248, line 22: “is likely related”

15) Page 7248, line 24: “necessarily”

16) Page 7249, line 1: To which particle size do the mass spectra correspond?

17) Page 7249, line 7: “ The higher temperature leads to . . . and to the decomposition . . .”

18) Page 7249, line 10: Could be better phrased, maybe: “From the three organic markers at m/z = 55, 56, and 57 detected during FROST 1, only the signal on m/z 57 is still visible in the FROST 2 experiments with the higher vaporizer temperature.”

19) Page 7249, line 20: “limit of the X-ray photo-electron spectrometer used for this analysis or were not included at all.”

20) Page 7250: line 3: entries

21) Page 7250, line 4: “. . . methyl silicone, whose mass per particle results are sepa-

rately shows on an expanded scale on the right y-axis.”

22) Page 7250, line 13: “to act as an active site”

23) Page 7250, line20: “reduced by the sulphuric acid because it can . . .”

24) Page7251, line18: What means “low amount” in terms of e.g. a coating thickness?

25) Page 7252, line 2: “. . . and of the application of the water bath . . .”

26) Page 7256, line19: “based on our present observations.”

27) Page 7257, line 15: “increased.”

28) Page 7257, line 27: Maybe better: “destroy active sites”.” Niedermeier et al. (2011) address more details . . .”

29) Page 7259, line 3: “possible explanations”

30) Page 7281 and 7282, first lines of the figure captions: “for the FROST2 campaign’s”

31) Page 7287, Fig. 10: Do the measurements refer to a particle size of 300 nm? Then, according to the colour code from Fig. 9, the experimental data should be shown in green.

32) Page 7288, third line of Fig. caption: “different particle classes”

33) Please check the comma placement throughout the text.

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