Interactive comment on “Heterogeneous uptake of ammonia and dimethylamine into sulfuric and oxalic acid particles” by Sauerwein and Chan

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The authors would like to thank Anonymous Referee #2 for the comments on the manuscript. We respond to the specific comments made by the referee below and identify the changes we have done to the manuscript.

Anonymous Referee #2: p2, line 10: The phrase “highest in marine particles as well as urban and rural aerosols” is confusing. It is not clear whether the authors are highlighting the importance of alkylaminium ions in marine aerosols, or the 140-560 nm size range.

Response: We thank the anonymous referee #2 for the comment. To improve clarity the sentence was changed to

“In fact, mass loadings of alkylaminium ions (R₃NH⁺) are the highest in particles with a diameter of 140-560 nm, both in urban and rural, as well as in marine aerosols (Müller et al., 2009; VandenBoer et al., 2011; Youn et al., 2015).”

Anonymous Referee #2: p2, line 28: Do the particles actually absorb more water than ammonium sulfate particles across different compositions and sizes? Or are there some limits (e.g., only below the deliquescence point of ammonium sulfate)?

Response: Recent studies on the hygroscopicity of methyl- and ethyl-aminium sulfate salts and their mixtures with sulfuric acid (Chu et al. 2015, Sauerwein et al. 2015, Rovelli et al. 2016) have shown that short-chain alkyl-aminium sulfates are significantly more hygroscopic than ammonium sulfate up to 95% RH, hence beyond the deliquescence point of ammonium sulfate. In all three studies, hygroscopicity of alkyl-aminium sulfates was shown to increase from primary to tertiary aminium sulfate. The difference in hygroscopicity decreases as extent of neutralization decreases to bisulfate composition, where aminium and ammonium bisulfate possess similar hygroscopic properties (Sauerwein et al. 2015).

Anonymous Referee #2: p3, line 30: Are the first experimental parameters for the sulfuric acid particles? If so, may want to specify that.
Response: We agree with anonymous referee #2. To no leave any ambiguity, we added the following for clarification

“Combination of the DMA flow (0.15 or 0.9-1.0 ppm) and the NH3 flow (1.8-1.9 ppm) resulted in DMA/NH3 ratios of 0.07±0.01 and 0.46±0.04 at 10% RH, as well as 0.07±0.01 and 0.49±0.02 at 50% RH for experiments with H2SO4 (Table 2), and 0.49±0.02 at 10% and 0.52±0.01 at 70% RH for experiments with H2C2O4.”

Further changes were made as suggested by anonymous referee #2 as follows:

Anonymous Referee #2: p6, line 4: Add ‘2-‘ to SO4 (as appears later in Section 3.2).

Response: Changes were made as follows:

“Figure 2 depicts the temporal profiles of DMAH/SO4, NH4/SO4 and X at the different gas ratios and RH. At 10% RH particles solidified during the experiment (Fig. 2a and b, indicated by crosshatched areas) and needed 2 to >18 hours (for 0.510% and 0.110%, respectively) to completely neutralize sulfate.”

Anonymous Referee #2: p6, section 3.2: Check section numbers here and throughout.

Response: We thank the referee for this comment, the section numbering has been updated throughout the manuscript.

Anonymous Referee #2: Section 3.2: NH3 displaced DMAH...should this be NH4? And then in paragraph below NH4 displaced by DMA...should be DMAH? Recommended to check throughout.

Response: We agree with anonymous referee #2 that the displacement of one alkaline species by another alkaline gas is inseparable connected with the gas absorption and protonation, and hence technically the argumentation, that DMAH+ is displaced by NH4+ is correct. Yet, the authors like to follow the terminology that is commonly used in literature focused on similar kind of displacement reactions such as Qiu et al. 2011.

Anonymous Referee #2: Also recommended to check notation such as NH4 vs. NH4+, which appears to be used inconsistently.

Response: Following the definition in section 2.3, we use DMAH to represent particle phase

\[ n\text{NH}_3(\text{CH}_3)_2^+ + n\text{NH}(\text{CH}_3)_2, \text{ NH}_4 \] to represent \( n\text{NH}_4^+ + n\text{NH}_3, \text{ SO}_4 \) to indicate total amount of sulfate species = \( n\text{H}_2\text{SO}_4 + n\text{HSO}_4^- + n\text{SO}_4^{2-} \), and \( \text{C}_2\text{O}_4 \) to indicated total amount of oxalate species (\( n \) denotes the molar amounts of each compound). The manuscript was reviewed and expressions such as \( \text{NH}_4^+ \) and \( \text{NH}_4 \)-species were consistently named as \( \text{NH}_4 \), the same was done for DMAH.

Anonymous Referee #2: p8, line 40: “During the”
Response: Following the suggestion the sentence was rewritten as:

“During NH₃-DMA co-uptake the absorbed DMAH seems to have suppressed the precipitation of NH₄HSO₄ in particles with a composition of 1.1 < NH₄/SO₄ < 1.5, so that the phase change started only at a NH₄/SO₄ around 1.5 (Fig. 5a and b).”

Anonymous Referee #2: Table 1: The subscript on the N looks like a superscript.

Response: the term XN₂ in Table 1 and the table caption have been updated to XN₂.

Anonymous Referee #2: Fig. 2: x-axis font in panel b appears larger than in other panels. Some other inconsistencies between panels—recommended to check closely

Response: The figure has been updated as shown below

References

