

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

Anonymous Referee #1

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The manuscript describes time-dependent measurements of the competitive partitioning of ammonia and dimethyl amine to droplets/particles containing sulfuric acid or oxalic acid deposited on a substrate. Although many previous measurements have studied the partitioning of one of these gas phase species, this paper provides insight into the relative abundances of the species in the gas and condensed phase at different gas phase concentrations and relative humidities when acting in competition. The manuscript is helpful in providing a qualitative exploration of the problem although I believe some of the quantitative aspects need to be interpreted with some caution due to experimental uncertainties, particles sizes, etc. The inherently qualitative interpretation of the measurements does limit the more general application of the work

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(and is sometimes quite hard to follow). However, the insights provided are sufficiently new that the manuscript merits publication - I do suggest the authors respond to the following suggestions before the paper can be accepted.

Page 5, line 40: The authors suggest that re-volatilization occurs at long times with the neutralization ratio decreasing from 1.7 to 1.5. Looking at the data in Figure 1, this seems to me to be over-interpreting the data. Firstly the “noise” in the calculated ratio seems to be larger than the error bars included with the points (e.g. see the fluctuations in the points around 17 hours for the DMA 0.15 ppm measurement at 50 % RH) – I suggest the authors consider again the uncertainties in their measurements. Secondly, how do the authors know that any such small change is not just due to a drift in environmental conditions (e.g. RH or temperature)? In fact, there is very little discussion of experimental errors in the analyses and the magnitude of uncertainties is quite key in verifying the veracity of trends identified in the data. I recommend a fuller discussion of experimental reproducibility and uncertainties.

Pages 6-7: I find the discussion of the change in neutralization and DMAH/NH₄ ratios along with the surface acidity quite confusing. Given the large size of the droplets and the long timescales for the measurements, I can't see that different surface kinetic parameters (uptake coefficients) for the amine and ammonia can govern the change in these ratios. Indeed, I don't think this is what the authors are saying – if they are, then they need to consider competing rates of gas diffusion limited uptake, diffusional mixing within these liquid droplets, sensitivity of such large droplets to surface kinetics etc. Whichever is correct, the discussions is very confusing. This need for clarity is even more true when the authors then state that Figure 4 confirms that the uptake of DMA and NH₃ proceed independently.

Page 12, lines 30-36: The authors consider the relevance of the high gas phase concentrations studied here when compared to atmospheric concentrations. It seems equally important to consider the importance of the particle size range studied. How do the authors expect their results to impact on our understanding of the much smaller

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particles that must be considered in the atmosphere? There is a brief comment on this at the very end of the summary section.

Minor changes/corrections:

Page 3, line 20: The method for checking the gas phase ratios is not clearly described. The sentence describing this procedure needs to be expanded on: "To ensure accuracy of the gas ratio, measures were taken including conditioning the setup for a prolonged period, separating RH conditioning cells and reaction cells."

Page 6, line 6: Presumably the authors are referring to Sections 3.3 and 3.4 here?

Page 9, line 21: The error bar on this line is 0.00 – this is presumably not correct.

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