

Interactive comment on “Estimating the NH₃:H₂SO₄ ratio of nucleating clusters in atmospheric conditions using quantum chemical methods” by T. Kurtén et al.

T. Kurtén et al.

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Thank you for your constructive comments. We do not know the definite answer to the question "what favors the increase of ammonia concentration in the larger particles"; the topic is certainly worthy of further study. However, we will add some discussion of the possible reasons in the revised version of the manuscript.

One possibility is that crystallization processes (responsible for producing the solid-state sulfate-ammonia mixtures in larger particles) favor pure ammonium bisulfate or ammonium sulfate over more ammonium-poor mixtures, even though the latter are more stable in the small gas-phase clusters. It is not uncommon for small clusters or surfaces to have notably different chemical compositions from bulk liquids or solids;

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one of the most common examples is surface enrichment by surfactants. From an "atmospheric physics" point of view this would correspond to the fact that the strength of the Kelvin effect differs from one molecular species to another. Though we do not know exactly which mechanism is at work in this specific case, it should be noted that ammonia has a significantly higher vapor pressure than sulfuric acid. This could perhaps make it more difficult for ammonia to accumulate in very small clusters.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2937, 2007.

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