

Interactive comment on “Hygroscopic Properties of Aminium Sulphate Aerosols” by Grazia Rovelli et al.

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Response to Anonymous Referee #2

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

Anonymous Referee #2: *This paper describes a new method of CK-EDB which can determine the hygroscopic properties of aerosol particles. In order to validate the method, the authors provided reproducible data for hygroscopic growth factor over the wide range of water activity. The results shown here agreed well with the results of previous studies that were performed with different methods. The manuscript presents in a clear, concise, and well-structured way, but I am afraid whether this paper is suitable for the scope of Atmos. Chem. Phys publication or not. Since the main focus of this work is to validate the new method, it would be better to be in a technical journal. If the authors would still like to publish the manuscript to ACP, the authors should address and implement my comments as below.*

Major comments: The measurements of hygroscopic properties of six aminium sulfate aerosols over the water activity range of 0.5 – 1.0 are in remarkably good agreement with the calculations and the previous studies. However, I do wonder to where/for what we could apply these results? To understand why this work is important in the area of atmospheric chemistry and physics, please describe atmospheric implications in more detail with a separate section.

Response: In order to address the concerns of Referee #2 about the atmospheric relevance of this study, we have added some considerable detail in the Introduction section about the different processes aminium sulphates are involved in, together with a number of references to the literature. In particular, we added more detail about the role of gaseous amines molecules in the formation of new particles. In addition, we now clearly stress the fact that nanoparticles deriving from such new particles formation events and containing aminium sulphates have the potential ability to act as cloud condensation nuclei (CCN). For this reason, investigating and quantifying precisely the hygroscopic properties of aminium sulphates is particularly atmospherically relevant, since this information is valuable in understanding of the role of such compounds in cloud activation and therefore in the indirect effects of atmospheric aerosols on climate. Please refer to the Introduction (specifically the modifications already requested by Referee #1) and to the Summary and Conclusions (section 4), which is now titled “Atmospheric Importance and Conclusions”. To highlight the atmospheric relevance, we now write at the beginning of Section 4:

"Quantifying the hygroscopic properties of aminium sulphates is important for understanding and modelling of the atmospheric processes in which they are involved. In particular, the role of short-chained alkylamines in the formation of new particles has been investigated in recent literature studies and found to be significant (Section 1). Aminium sulphate-rich nanoparticles that derive from new particles formation events can potentially act as CCN, and their hygroscopic properties must be well-characterised with the aim of reducing the overall uncertainties that currently affect our understanding of the indirect effects of atmospheric aerosols on climate. Robust and accurate data are essential for improving microphysical models of aerosol hygroscopicity; this study presents an extensive data set for an homologous series of six compounds, compared to ammonium sulphate, extending over a wide range in RH. In addition, it represents the most comprehensive characterisation of the hygroscopic response of aminium sulphate aerosol so far, complementing previous bulk phase measurements (comparable in accuracy but limited to higher water activity) and aerosol measurements at lower RH (with lower accuracy than achieved here). Previously, the bulk and aerosol measurements reported in the literature were in disagreement. Here, we report aerosol measurements that are in good agreement with the previously most accurate bulk phase data, resolving this discrepancy."

In addition, we already state later in this section the significance of these new data when compared with the earlier bulk phase data, stating:

"The main differences in approaches are that: we perform aerosol measurements that cover a wider range in water activity as compared with the bulk measurements of Sauerwein et al. (2015); and we provide direct measurement at amine-to-sulphate ratios of exactly 2:1, whereas Sauerwein et al. (2015) performed a ZSR fitting on data from solutions with variable amine-to-sulphates ratios and extrapolated water content for the exact 2:1 ratio. These new CK-EDB measurements suggest a higher level of hygroscopic growth for the aminium sulphates than previously reported by Sauerwein and co-workers when inferred from measurements over a range of amine-to-sulphates ratios; we have provided a refined parameterisation for all compositions."

Minor comments

Anonymous Referee #2: 1. Page 4, line 23, Please add the RH and temperature values.

Response: This information has now been added at line 16, page 4.

"Temperature and gas phase RH ranges that are accessible with this experimental setup are -25 to 50 °C and 0 to 99%, respectively. All the comparative evaporation kinetics measurements presented here were performed at 20 °C and at gas phase RH values between ~50-90%."

Anonymous Referee #2: 2. Page 6, line 13, Remove the comma after "mixed,"

Response: This comma has now been removed.

Anonymous Referee #2: 3. Page 6, line 18, Please state at what temperature the amine solution was kept in an ice bath.

Response: We have now explicitly said that the temperature was 0 °C.

"During both the titration of the amine stock solution with HCl and the preparation of the aminium sulphates solutions with H₂SO₄, the amine solution was kept in an ice bath (0 °C) and the addition of the acid was performed slowly and dropwise, in order to dissipate the heat generated by the neutralisation reaction and to avoid any possible amine volatilization."

Anonymous Referee #2: 4. Page 8, line 12, Cite only once (Qiu and Zhang, 2012)

Response: The double citation has now been removed.

Anonymous Referee #2: 5. I wonder why there is no data point for MMAS and TMAS from _0.7 to _0.8 aw in Fig. 3.

Response: We have responded to this issue in our response to the comments made by Referee #1. Measurements were taken for a large number of systems over a wide range of conditions and it became apparent only later that measurements were not available for this small range of conditions for two compounds. We concluded that the trends were sufficiently clear that additional measurements were not essential.