

Concerning the research article:

**Methane sulfonic acid enhanced formation of molecular clusters  
of sulfuric acid and dimethyl amine**

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Reply to referee #1:

We thank the referee for the positive review and constructive comments. Here, we present a point-to-point response to all comments (blue colored text).

In this paper, the authors quantify how MSA impacts H<sub>2</sub>SO<sub>4</sub>+DMA nucleation rates using quantum calculations and the ACDC model. They show that MSA may increase nucleation rates by up to 300% in cold regions. These results are useful for the aerosol microphysics community. The paper is very well written and is commendably concise yet complete. I recommend publication after some minor comments are addressed; however, I am not an expert on the quantum chemical methods presented here, so other reviewers will be necessary to judge these methods.

P18681 L13: “MSA concentrations \*were\*”. In this sentence are you referring to gas- phase or particle-phase concentrations?

This sentence refers to gas phase MSA which will be clarified.

Figure 4: Would it be possible to make the arrow width proportional to the percentages in the table? I realized this would require 2 panels in order to get the 2 MSA concentrations, but this would be useful for visualizing the growth. It's taken me some time to mentally map the table numbers onto the plot.

We agree that Figure 4 is somewhat demanding and we did find this suggestion interesting. However, we were unable to construct a visually appealing 2-panel plot without significant loss of data, wherefore we were forced to abandon this idea. In stead, the original version of Figure 4 was modified to highlight the main growth pathways, thereby assisting the reader in mapping the numbers onto the plot. The figure caption will be modified as well, as seen below.

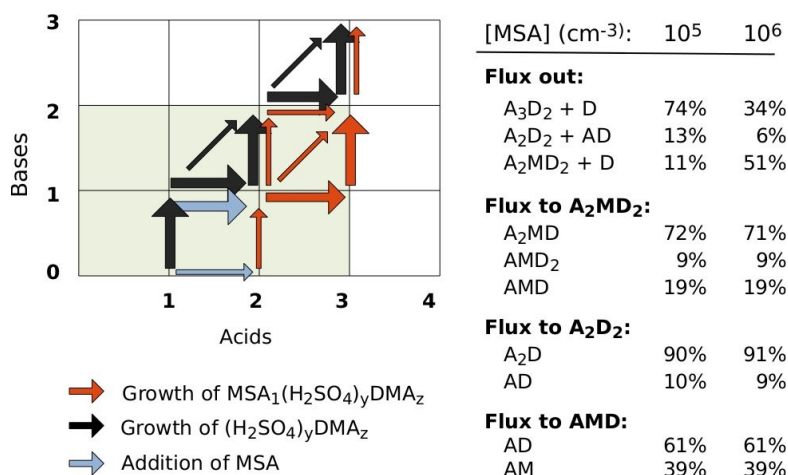


Figure 4. Main cluster formation pathways at  $T = 258 \text{ K}$  and  $[\text{H}_2\text{SO}_4] = 10^6 \text{ molecules cm}^{-3}$ ,  $[\text{DMA}] = 10^8 \text{ molecules cm}^{-3}$  and two representative MSA concentrations. Dominating growth pathways are indicated by larger arrows. Fluxes to clusters formed via several different pathways are indicated in the side table where A, M and D is shorthand  $\text{H}_2\text{SO}_4$ , MSA and DMA, respectively.

P18690 L22 and throughout: “DMA in most locations is in large excess compared to acid”. Is this representative of the remote marine boundary layer or the remote free troposphere (where DMS, the MSA precursor, may have been transported out)? Are the assumed DMA concentrations in the paper representative of these locations? The authors should be sure they are not mixing continental DMA concentrations with remote MSA concentrations.

We are fully aware that, typically, continental DMA concentrations are significantly higher than marine DMA concentrations due to emissions from livestock and industry. We mainly base our choice of DMA levels on two reviews by Gibb et al (Global Bio-geochem. Cy., 13, 161-178, 1999) and Ge et al (Atmos. Environ., 45, 524-546, 2011), where typical marine concentrations of  $10^7$  to  $10^9$  molecules per  $\text{cm}^{-3}$  are reported. We acknowledge that DMA concentrations could be very different in the free troposphere but, to the best of our knowledge, no altitude resolved DMA measurements have been published. In the revised manuscript we will stress that conditions in the free troposphere are poorly known and that insight into atmospheric processes from our study is limited by this.

On behalf of all authors,

Nicolai Bork