**Review on "Hygroscopicity and CCN potential of DMS derived aerosol particles"**

We would like to thank the editor for his constructive comments. We have addressed the comments in a point-by-point fashion below and revised the manuscript accordingly. Our answers to the comments are given below in blue letters, while the editor comments are given in *black italics*. Additionally, we added the changes we made in the revised manuscript in **blue bold** letters.

**(1)** *Both reviewers pointed out the issue of having 0.5-1 ug m-3 non-MSA OA detected by the AMS and its unclear influence on hygroscopicity of the generated particles. The mass fractions of such OA in some experiments are high. If the OA was from contamination, a high mass fraction of it may affect the measured hygroscopicity, e.g., when the OA is insoluble. If it was from analysis bias, should this part of mass be included into the MSA mass? I think what was added in Line 377 remains unclear and is insufficient to address reviewers' concerns. The key question is whether this mass should be added or excluded in the analysis herein. I would like to see more discussion with additional evidence on it. For example, the mass spectra of OA should provide some hints on the source and properties of OA.*

We agree with the editor that the OA signal is in some cases quite high and that the determination of the origin would be very helpful to understand its importance in the experiments. In order to discuss this further we present here some additional data for Exp. 5 to give an example.

Fig. R1 and Fig. R2 illustrate the organic mass spectra before DMS was injected into the chamber and when the particle mass reached its highest concentration, respectively. From Fig. R1 it is evident that the chamber is clean with respect to organic particle mass before DMS is introduced. The $CH_4O^+$ and the negative $CH_2^+$ signals are both artifacts of peak fitting in normal AMS data processing. m/z indicated in orange in Fig. R1 and R2 denote peaks that were also found during the MSA calibration (MSA calibration spectrum is shown in Fig. R3). In Fig. R2 some other ions appear to have negative concentrations pointing to the fact that too much of the signal of those specific ions was attributed to MSA rather than organics. This shows that the MSA fragmentation or ion transfer during the calibration was slightly different from MSA fragmentation and ion transfer under chamber conditions. This is within normal uncertainty for AMS. Looking closer at the other ions in the spectrum we found that $CH_4SO_2^+$ (indicated in blue), could stem from MSIA. There is also a potential contribution from dimethyl sulfoxide (DMSO) on ion $C_2H_6SO^+$. The signal is though so low it not clearly visible in Fig. R2. $C_6H_5^+$ and $C_7H_7^+$ are typical markers for aromatic compounds but represent only a small contribution of the overall organic mass. Finally, the organic mass signal is composed of the sum of many low-intensity fragments that we cannot attribute to a specific source. In comparison Fig. R4 shows the residual organic mass signal recorded during the MSA calibration, after the verified MSA ions (shown in Fig. R3) are subtracted. The signal intensities are clearly lower compared to during DMS oxidation experiments in the chamber and fewer peaks are visible (Fig. R3 vs. R4). Nonetheless, this also illustrates that even during calibration some organic fragments appear that we cannot clearly assign to a specific source.
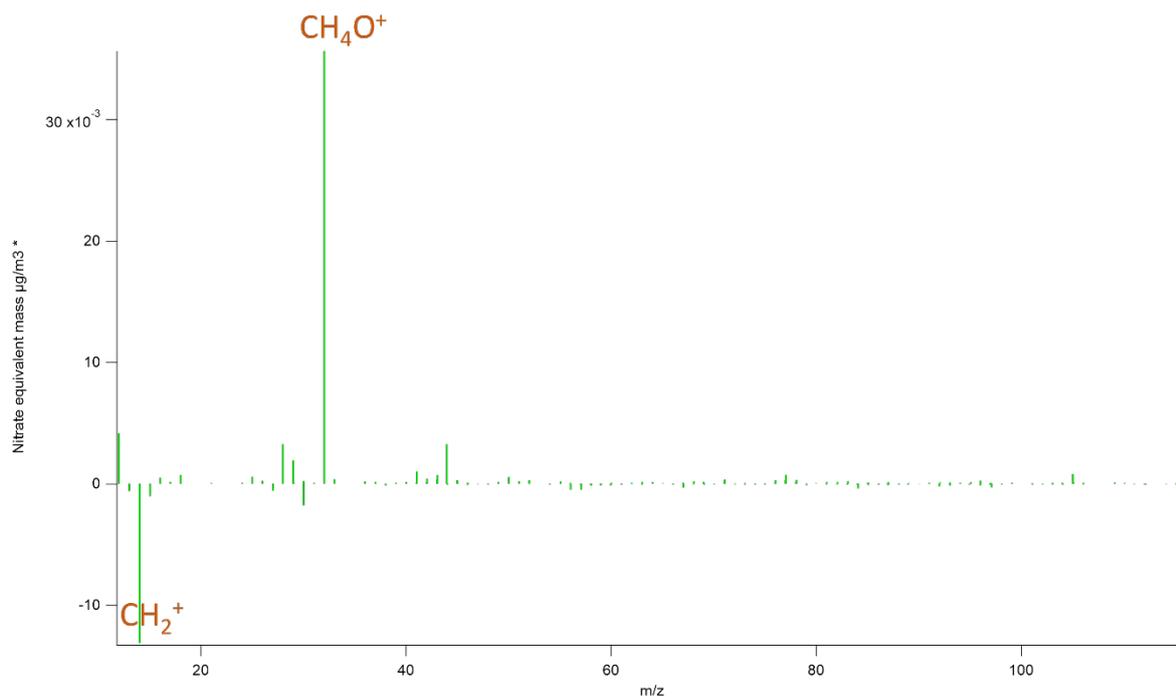
Fig. R1: High resolution mass spectrum of the organic signal measured during Exp. 5 before DMS was injected into the chamber. The ions indicated in orange color denote ions also found during the MSA calibration.
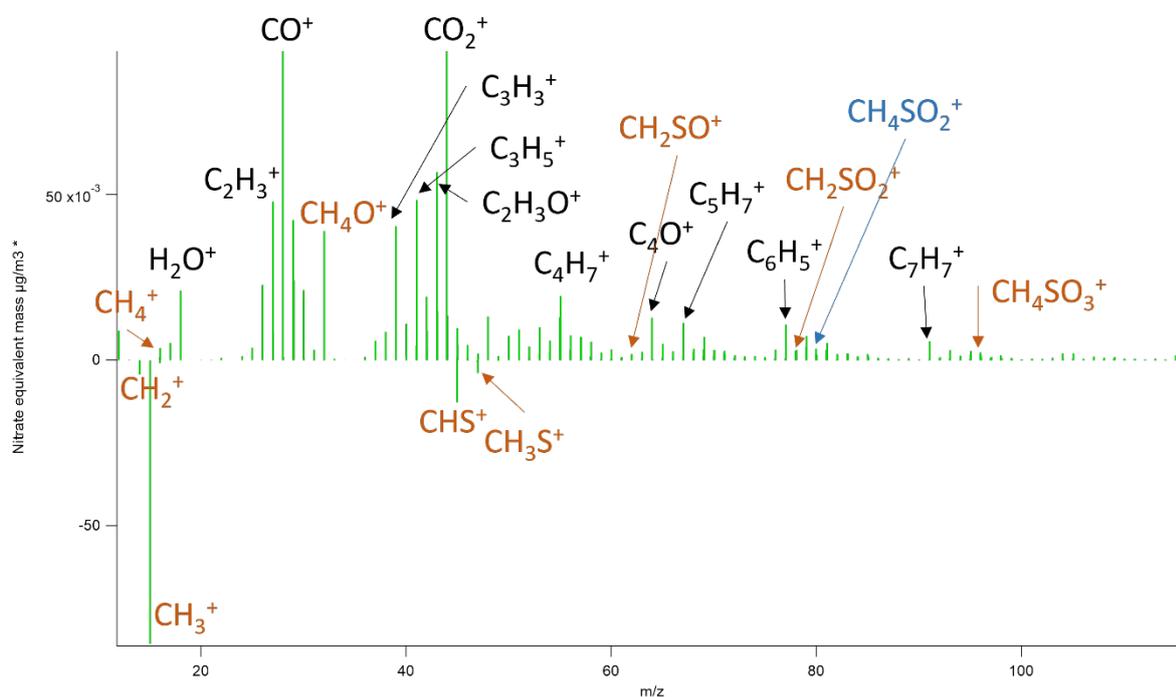


Fig. R2: High resolution mass spectrum of the organic signal measured during the time period where the particle mass peaked in Exp. 5. The ions indicated in orange color denote ions also found during the MSA calibration. Ions denoted in black color have an unknown origin. The ion denoted in blue could stem from MSIA.
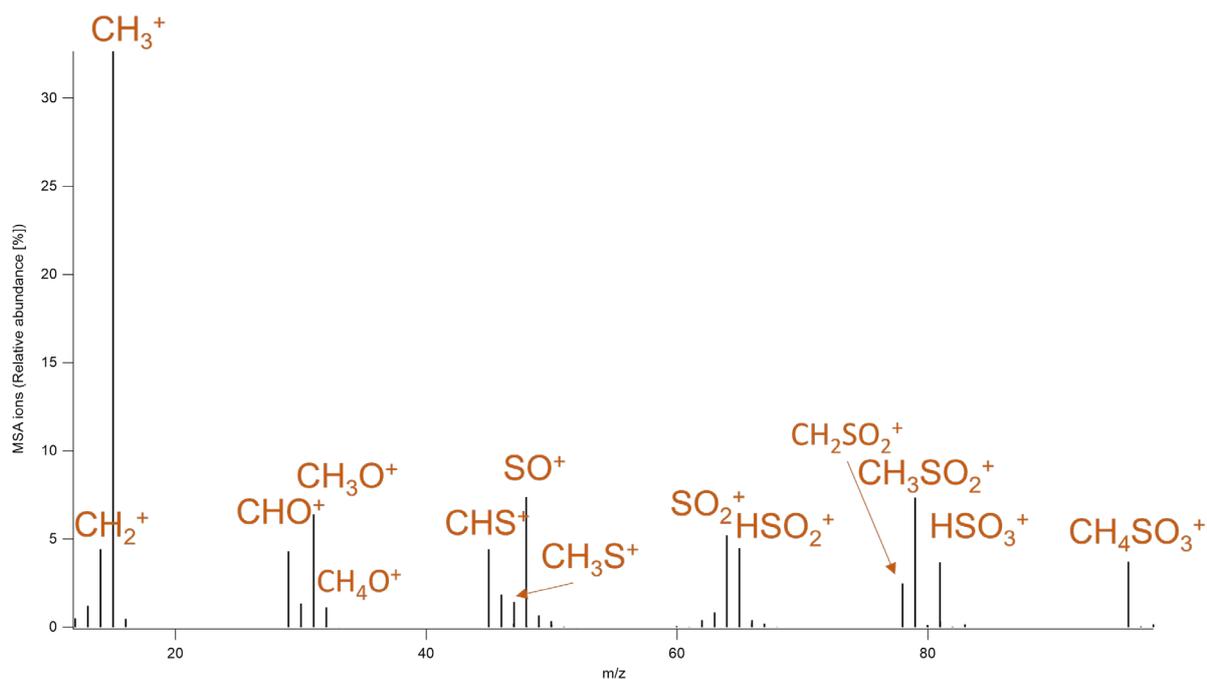
Fig. R3: High resolution mass spectrum of MSA from calibration including only verified MSA ions (Hodshire et al., 2019).
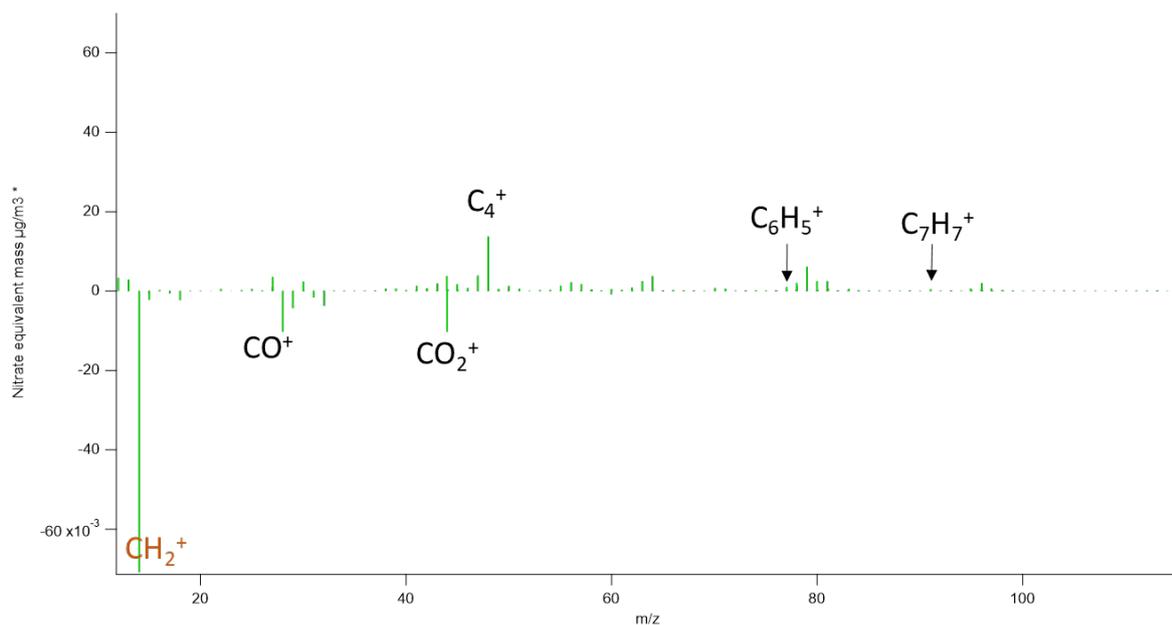


Fig. R4: High resolution mass spectrum of residual organics, i.e., unaccounted organics after subtracting verified MSA signal, at about 1.5 μg/m$^3$ MSA during calibration.

In order to assess the potential role of organic compounds for hygroscopicity we have performed a κ-sensitivity calculation:

According to Petters and Kreidenweis (2007) the simple volume weighted mixing rule can be applied to retrieve the mixed hygroscopicity parameter κ from organic and inorganic compounds. In order to

assess the importance of a potential organic contribution to the measured κ ($\kappa_{meas}$) we can use the following equation (mixing rule):

$$\kappa_{meas} = \kappa_{org} \cdot \varepsilon_{org} + \kappa_{mix,in} \cdot \varepsilon_{mix,in} \qquad (1)$$

Where $\kappa_{org}$ is the κ of the organic compounds, $\kappa_{mix,in}$ is the effective κ of the mixture of all inorganic contributions and $\varepsilon_{org}$ and $\varepsilon_{mix,in}$ are the volume fractions of the organics and inorganics, respectively. The volume fractions of organics and inorganics can be calculated from the mass concentrations measured by the HR-ToF-MS and assuming densities of the organics and inorganics, respectively. For organic compounds density values of approximately 1.2-1.4 g cm$^{-3}$ have been suggested (Nakao et al., 2013). For the expected inorganic compounds (ammonium sulfate, MSA and ammonium-MSA) Fossum et al. (2018) suggest densities of approximately 1.7, 1.5 and 1.4 g cm$^{-3}$ for ammonium sulfate, MSA and ammonium-MSA, respectively. By assuming a combination of these three inorganic compounds we estimate an average inorganic density of approximately 1.6 g cm$^{-3}$. Additionally, we assume the κ of the organics ($\kappa_{org}$) to be between 0 and 0.2 based on Jimenez et al. (2009). By using these inputs and rearranging Eq.1, the actual κ of the inorganic compounds during the measurements ($\kappa_{mix,in}$) can be found as:

$$\kappa_{mix,in} = \frac{\kappa_{meas} - \kappa_{org} \cdot \varepsilon_{org}}{\varepsilon_{mix,in}} \qquad (2)$$

For Exp.5, where mass spectra are presented above, $\kappa_{mix,in}$ in the range 0.6 – 0.7 are obtained when using $\kappa_{meas}$=0.52 (mean value for dry 80 nm particles) and $\kappa_{org}$ between 0 – 0.2. This would represent a κ of pure ammonium sulfate particles. For Exp.1, where organic contribution is highest, a $\kappa_{mix,in}$ in the range 0.7 – 0.8 is found when using $\kappa_{meas}$=0.45 (mean value for dry 80 nm particles). These values exceed the κ-range of pure ammonium sulfate. Both cases contradict the results from the HR-ToF-MS that did not only find an ammonium sulfate contribution but a considerable fraction of MSA in all experiments (see Fig. S7a and b; MSA fraction always above 37 %). This MSA fraction is expected to lower the overall inorganic κ, as its predicted κ value is 1.5 (Fossum et al., 2018).

These calculations thus point towards organics rather being a measurement artefact. Our data and the assumptions for the above presented hygroscopicity calculations do not make it possible to conclude on whether the organic mass should be added or excluded in the analysis. Nevertheless, we can conclude that the presented $\kappa_{meas}$ values represent lower limits of κ-values as any contribution of organic compounds (0< $\kappa_{org}$< 0.2) would imply a higher κ-value from the inorganic fraction.

We have revised the added text in Line 377 as follows:

**Absolute concentrations show that in general the aerosol mass loading was small and that the organic contribution was comparable during all experiments. As previously discussed in Rosati et al. (2021) and Wollesen de Jonge et al. (2021) we cannot conclude on the exact origin of the organic signal. A thorough analysis of the organic mass spectra revealed that the organic mass signal is composed of the sum of many low-intensity fragments that we cannot attribute to a specific source (see example organic mass spectrum for Exp. 5 in SI). It cannot be entirely excluded that it is due to contamination from previous experiments. It could however also derive from biases due to the complex fragmentation during ionization and uncertainties arising from the calibration with MSA. Additionally, instrumental memory effects have previously been documented for the AMS (e.g. Drewnick et al., 2015; Drewnick et al., 2009; Pieber et al., 2016). Two ionization efficiency calibrations before and after the experiments could neither exclude nor completely prove memory effects. If the organic signal stems from uncertainties in the analysis, it**

will not influence hygroscopicity. In the case that the organic signal is real, the herein presented hygroscopicity results represent lower estimates of the inorganic aerosol hygroscopicity as κ-values for organics in the range 0-0.2 (Jimenez et al., 2009) can be expected, which would lead to a decrease of the κ-values measured for an organic-inorganic mixture.

The abstract has been modified to read:

"We show that the hygroscopicity parameter for particles of 80 nm in diameter is in the range 0.48 - 0.52 or higher, at 293 K as measured at both sub- and supersaturated water vapour conditions."

The conclusion has been modified to read:

The average hygroscopicity parameters, κ, calculated from measurements performed during the main particles' growth mode at sub- and supersaturated water vapour conditions at 293 K, ranged from 0.48 to 0.52 for dry 80 nm particles. These values may represent lower limits and are well comparable with modelled values for MSA or mixtures of MSA and ammonium sulphate (Fossum et al., 2018).

**(2)** *The caption of Figure 1 stated that the fluctuations (I guess after 0.4 h should also be stated) are caused by scanning-mode operation of PSM. Are there any reference to support? Also, Sect. 2.3.3 is confusing because it said the instrument was operated in a fixed mode. The question is if the scanning mode is problematic, can we trust the data in Fig. 1 for that part? Please update Sect. 2.3.3 and describe PSM operation as well as its problem in details.*

The fluctuations caused by setting the PSM to scanning mode after about 0.4 h for the temperature of 273 K, do not have any effect on the calculations of the nucleation rates (described in Sect. 2.1.1). There is no problem or issue of the PSM in scanning mode, this is the normal way it behaves in this mode (Lehtipalo et al., 2014). This part of the measurement was shown purely to illustrate that the PSM had reached a maximum. The data when the PSM was measuring in scanning mode are not used in any calculations in this manuscript. To make this point clearer we have adapted the figure in the following way:
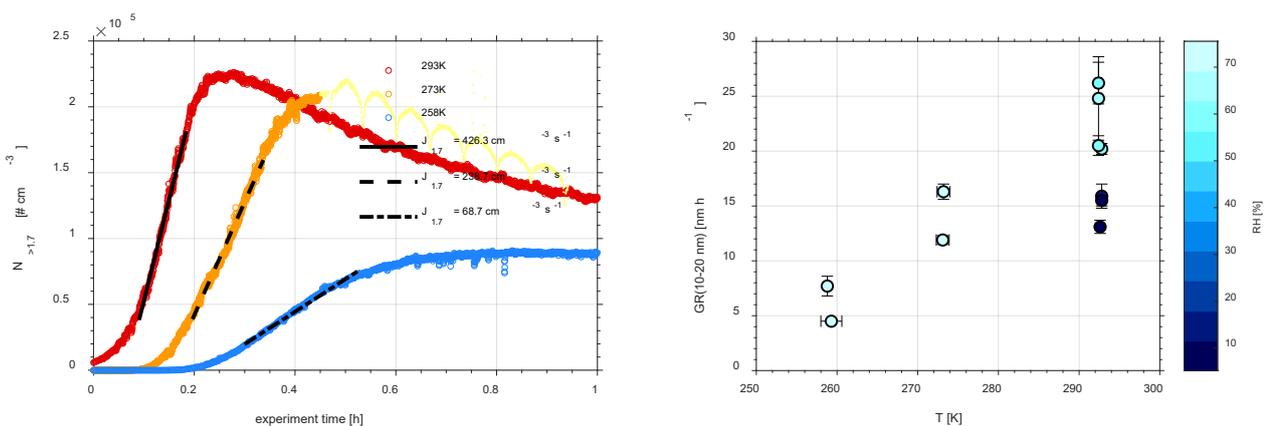


Additionally, we have added this sentence to the figure caption:

**The fluctuations seen in at T=273 K (light yellow dots) are due to the fact that the PSM was set to measure in scanning mode during this period (Lehtipalo et al., 2014). This data was not used for the**

**calculation of $J_{1.7}$ and is only illustrated to show the reader that the maximum in number concentration had been reached.**

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