The study by Poulain et al. presented a comprehensive evaluation of the ACSM measurements based on the inter-comparisons with other collocated measurements for more than 5 years. The robustness, the limits, and the potential sources of uncertainties of the ACSM measurements for different aerosol species were well discussed. The results are very important for broad ACSM users to understand the long-term measurement uncertainties. The manuscript is well written, and I recommend it for publication.

We would like to thank the referee for his/her constructive comments and suggestions made to improve and clarify our manuscript. Our responses are given below. For clarity, comments from the referee are in black, our responses in blue, and change on the text of the manuscript in **bold blue**.

The authors would like to mention here that two additional co-authors were added to the initial list regarding to their contributions on the revision of the manuscript.

I have a few comments:

R1. The ACSM was calibrated twice at ACMCC, and also could be several times at the TROPOS research station Melpitz. How robust of the relative ionization efficiencies of ammonium and sulfate were. This is also an important information for long-term measurements.

A1: Thanks for bringing up this interesting and important point. To consider this, the following text has been added to the ACSM description (section 2.2) that discussed calibration and stability of the ionization efficiency (IE) and relative ionization efficiency (RIE) of ammonium and sulfate:

"**The ACSM was regularly calibrated according to the manufacturer's recommendations at that time with 350 nm monodispersed ammonium nitrate and ammonium sulfate particles selected by a DMA and using the jump scan approach. It is important to note that since recently, the recommended calibration method has changed to a full scan approach (Freney et al., 2019). The total particle number concentration was systematically set below 800 # cm^{-3} to limit the artefact due to multiple charged particles. An overview of the ionization efficiency (IE) and relative ionization efficiency (RIE) for ammonium and sulfate can be found in Figure SI-1. On average, all performed calibration provides a mean IE value of 4.93 (± 1.45) 10^{-11} (mean ± std. dev.) and mean RIEs for ammonium and sulfate were 6.48 ± 1.26 and 0.68 ± 0.13, respectively. These values are very close to the ones used for the data evaluation as indicated in Figure SI-1. Overall, no clear trend for IE and RIE of sulfate can be observed over the period, while a small decrease in the RIE of ammonium can be reported. The lowest RIE of ammonium was reported just after the replacement of the filament indicated a possible need for degassing and stabilization period. However, it is difficult to conclude if these tendencies could be associated with a possible aging effect of the instrument since it corresponds to a single instrument. Similar observations on various other individual ACSMs would be needed to allow for stating such a conclusion and a more systematic investigation of potential trends should then be performed with a large number of ACSM.**"

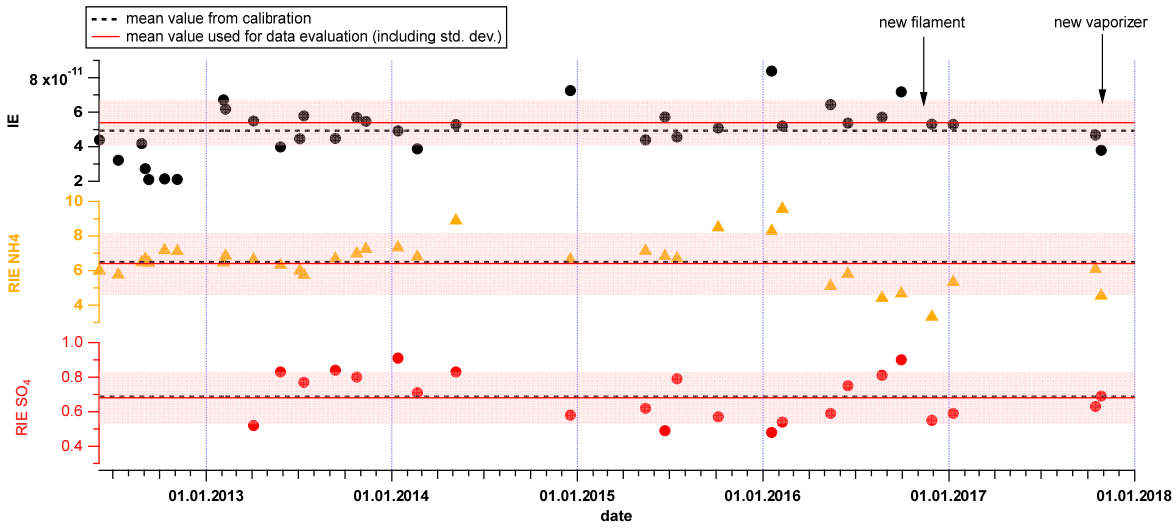

Figure SI-1: Time variation of the IE and RIE for ammonium and sulfate. The single points correspond to calibration, the dashed black line to the mean value from the calibration, and the full red line the mean value from the data analysis (shaded area corresponds to the standard deviation). Major maintenance (change of filament and vaporizer) are including.

R2. The format of ions should be consistent throughout the manuscript, e.g., page 7, line 15 – 25, use "+" for all ions.

A2: Charge was added to all mass spectra fragments.

Following correction were made:

P7, L 15-27: "[…] same m/z (for example, $\mathbf{C_6H_8^+}$ and/or $\mathbf{C_5H_4O^+}$ at m/z 80 for $\mathbf{SO_3^+}$, or $\mathbf{C_6H_9^+}$ and $\mathbf{C_5H_5O^+}$ at m/z 81 for HSO_3^+) […] change of SO_3^+/SO^+ and HSO_3^+/SO^+ […]"

P 9, L 11: "[…] at m/z 30 ($\mathbf{NO^+}$) and m/z 46 ($\mathbf{NO_2^+}$), as well as on a minor contribution of N^+ and HNO_3^+ ions […] "

P11, l31: "[…] a possible artefact on the CO_2^+ signal itself."

R3. Page 15, line 25 – 26, "ACMS" to "ACSM"

A3 Corrected

References

Freney, E., Zhang, Y., Croteau, P., Amodeo, T., Williams, L., Truong, F., Petit, J.-E., Sciare, J., Sarda-Esteve, R., Bonnaire, N., Arumae, T., Aurela, M., Bougiatioti, A., Mihalopoulos, N., Coz, E., Artinano, B., Crenn, V., Elste, T., Heikkinen, L., Poulain, L., Wiedensohler, A., Herrmann, H., Priestman, M., Alastuey, A., Stavroulas, I., Tobler, A., Vasilescu, J., Zanca, N., Canagaratna, M., Carbone, C., Flentje, H., Green, D., Maasikmets, M., Marmureanu, L., Minguillon, M. C., Prevot, A. S. H., Gros, V., Jayne, J., and Favez, O.: The second ACTRIS inter-comparison (2016) for Aerosol Chemical Speciation Monitors (ACSM): Calibration protocols and instrument performance evaluations, Aerosol Sci. Technol., 1-25, 10.1080/02786826.2019.1608901, 2019.