

Authors' response to Referee #1 comments

We would like to thank Referee #1 for the valuable comments that aided us to improve the manuscript. In this post, we will provide our response to the Referee's comments. In our replies, we provide original comment from the Referee and our response followed by the changes made to the manuscript.

General comment:

(1) More details on the physical aspects of the ADIc may need to be reported. For example, it would be helpful to know the dimensions of the ADIc growth tube and the residence time of particles for a certain flow rate. Also, for the sake of clarity, consider to add on Figure 1 references to the parameters reported in Table 1. In mentioning the importance of minimizing the time the particle being a droplet inside the growth tube (line 113), it would be useful to quote the approximate time scale. In addition, the issue whether the ADIc modifies the shape or phase of particles should be addressed, at least briefly. Such changes could significantly affect aerosol quantification by the AMS.

Response and author's changes in manuscript:

Dimensions: The dimensions of the growth tube are already stated in the manuscript (see the text starting on line 126 (page 5) "The conditioner, initiator and moderator are 140 mm, 51 mm and 102 mm long, respectively, separated by 7.5 mm thick insulator sections. In both prototypes the growth tube was lined with a 9 mm-ID, ~1.5 mm-thick wick formed from rolled membrane filter."

Residence Time: We added to the manuscript: "For particles along the centerline of the flow, the calculated residence time from the point of activation to the inlet of the focusing nozzle is 200-300 ms, depending on the point of activation. Along the flow trajectory that encompasses 50% of the flow, the residence time is as long as 400 ms."

Operating temperatures for conditioner, initiator, moderator and focusing nozzle have been added to Fig. 1.

Particle Shape: Discussion of particle shape has been added to manuscript (see details in response to comment 6).

Detailed comments:

(2) Line 18, change "ultrafine" to "fine" since ADIc can clearly concentrate particles beyond the ultrafine mode.

Response and author's changes in manuscript: "ultrafine" has been changed to "fine"

(3) Figure 2 shows the size dependent concentration factors for particles only up to 400 nm in mobility diameter. What are the concentration factors for larger particles? Also, the blue circles appear to show in two different shades. Are these from two separate sets of experiments? If so, explain the differences.

Response and author's changes in manuscript: Unfortunately we were not able to investigate the concentration factors for the particles larger than 400 nm (in mobility diameter) in the laboratory due to the instrumentation available. However, based on the ambient size distribution data measured by the SP-AMS at SMEAR III (Fig. 5), the CFs were rather stable until 1 μm in vacuum diameter (dva) corresponding to ~600 nm in mobility diameter (see figure below).

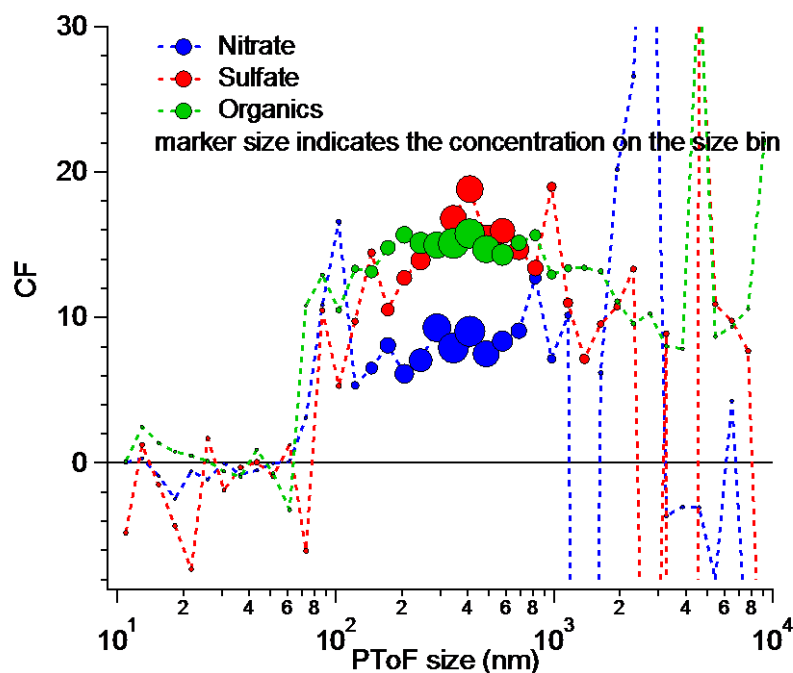


Fig. R1. Size dependent concentration factor for nitrate, sulfate and organics during the field measurements at SMEAR III in Helsinki.

In contrast, during the field measurements at ARI, the size distributions for organics and m/z 57 from the Q-AMS+ADIC were missing mass above $d_{va} \sim 700$ nm that was measured by the HR-AMS without the ADIC. This difference can be at least partly explained by a difference in the cutoff of the aerodynamic lenses in the two AMS instruments. The difference in the size distributions is discussed also in comment (7) for Referee #2.

All blue circles are from the same experiments so they should appear in same color.

No changes were made to manuscript based on this comment.

(4) Line 201, the sentence “. . . measured size distributions were normalized to the mass spectra” is vague. Consider to revise.

Response and author’s changes in manuscript: The sentence was modified to: “The measured size distributions were normalized to the mass concentrations measured in the mass spectrum mode.”

(5) Line 237, how often was SP-AMS switching between laser-on and laser-off?

Response and author’s changes in manuscript: Laser was switching between on and off every 1.5 minutes. Switching period has been added to manuscript.

(6) For the evaluation of ADIC’s influence on aerosol composition and size, Figure 4 is presented to compare the average high resolution mass spectra for organics and rBC from an SP-AMS downstream and bypass the ADIC. The measured-CF for Cx was significantly higher than for the other ions. Could it be due to change in particle shape, thus particle collection efficiency in the laser beam? It would be also interesting to see an evaluation of the ADIC’s influence on bulk PM composition, including both inorganics and organics.

Response and author’s changes in manuscript: The influence of ADIC on particle shape has been added to the text: “One possible explanation is that the ADIC altered the shape of the rBC-containing particles. The effect of the condensation/evaporation process on particle shape was not explored in

this study; however, others have found changes in the shape of aggregates. In a study using a condensation system similar to that employed here, Ma et al (2013) reported collapse of the aggregate structure of laboratory-generated soot in the evaporation process. Regarding the SP-AMS, the morphology of the particles had been demonstrated to affect the collection efficiency since it affects the overlap of the particle beam and the laser beam (Willis et al., 2014)."

The bulk PM composition measured by the SP-AMS with and without the ADIc has been added to Supplemental material (Fig. S4).

References

Ma, X., Zangmeister, C. D., Gigault, J., Mulholland, G. W., & Zachariah, M. R. (2013). Soot aggregate restructuring during water processing. *Journal of Aerosol Science*, 66, 209-219.

Willis, M.D., Lee, A.K.Y., Onasch, T.B., Fortner, E.C., Williams, L.R., Lambe, A.T., Worsnop, D.R., Abbatt, J.P.D., 2014. Collection efficiency of the soot-particle aerosol mass spectrometer (SP-AMS) for internally mixed particulate black carbon. *Atmos. Meas. Tech.* 7, 4507–4516.

(7) Line 311 – 313, this sentence is a bit confusing. Consider to revise.

Response and author's changes in manuscript: That sentence has been modified as well as few other sentences related to it.

(8) Line 355 – 358, does it mean that the Q-AMS and the SP-AMS report different ammonium concentration for the same air mass? Won't this discrepancy correctable through proper relative ionization efficiency calibration and fragmentation table adjustment (e.g., for better ammonium quantification)?

Response and author's changes in manuscript: The referee is correct that we did not have good agreement in bypass for ammonium between the Q-AMS and the HR-AMS, even with several RIE calibrations and adjustments to the fragmentation tables. Part of the problem is that ammonium concentration was low ($< 0.4 \text{ ug m}^{-3}$), and it was often close to the detection limit for the Q-AMS during the bypass periods. We think that this is an indication of how hard it is to get two instruments to agree for all species.

Lines 355-358 were revised:

"Another possibility is that the RIE for ammonium was incorrect for one or both of the instruments, even though it was measured before and after the ambient sampling period with pure AN particles. The CF during bypass periods was 1.3 ± 0.4 (Table 3) indicating that the two instruments did not agree well for ammonium even when the Q-AMS was bypassing the ADIc. However, the ammonium mass loading was low ($< 0.4 \text{ ug m}^{-3}$) and often close to the detection limit for the Q-AMS during the bypass periods, leading to a large uncertainty in the bypass CF."

(9) Fig 8, the ammonium measurement after ADIc shows more spikes. Is this an artifact induced by the ADIc?

Response and author's changes in manuscript: Ammonium spikes in the time series of the ACSM are not induced by the ADIc since similar spikes are seen in Fig. 9a when the ACSM was used in bypass without the ADIc. We think that these spikes are likely related to the detection of small air bubbles in the ACSM that affect the measured ammonium concentration. The spikes may be either negative or positive if the air bubble is released during the filter or aerosol measurement phase.

We added to figure caption 8: “Spikes in the time series of ammonium in the ACSM are likely related to the detection of small air bubbles in the ACSM that affect the measured ammonium concentration.”