We thank Referee#3 for their comments regarding our manuscript. Below we provide our answers (shown in Blue) to their comments (shown in Black), and where changes were made to the manuscript, the modified text is given (**Blue Bold**).

Answers to Anonymous Referee #3, [Report #2]

1.) When developing the Caltech nano-RDMA, Brunelli et al. (2009) reported that elecrtrospray generated tetra-alkyl ammonium ions including THAB ions were measured by the Caltech nano-RDMA with good resolutions. During their experiments, the electrospray chamber was grounded and connected to the nano-RDMA directly. However, this manuscript claims that the spectra of electrosprayed THAB solution can not be detected with the nano-RDMA when the ES chamber is grounded (Fig. 4). More information needs to be added in the manuscript to **explain this discrepancy**.
The authors claim that the THAB monomer can be detected, but with a low efficiency, when the ES chamber is grounded; not that the spectra of electrosprayed THAB solution cannot be detected at all with the nano-RDMA.
In the study of Brunelli et al., 2009 no data on transmission are presented and when using an electrospray low transmission is not a problem, as the concentration of ions is very high. Therefore, the authors think that the good resolution of the nRDMA reported in the detailed characterization by Brunelli et al., 2009, using tetra-alkyl ammonium ions is not a discrepancy with respect to our results.

Did the authors use the same nano-RDMA aerosol inlet to Brunelli et al. (2009)? Is the nano-RDMA the same? Are there any differences in experimental setup and conditions which may cause the above discrepancy?
The DMA used in this study is the new version of the DMA used in Brunelli et al., 2009. The main change was a larger diameter in the exhaust pipe line to allow larger sheath flow rates to be used without causing turbulence.
We added in Section 2.1 at Line 18 the sentence: **"The DMA used in this study is the new version of the DMA used in Brunelli et al. (2009). The main change was a larger diameter in the exhaust pipe line to allow larger sheath flow rates to be used without causing turbulence."**

In addition, **an illustration of the original nano-RDMA aerosol inlet needs to be added in Fig 2** to complement the comparison between chamber results reported in Figs. 3 and 10.
We added a schematic picture of the original inlet in Fig. 2.

2.) Terms such as "Transmission", "Penetration", "Transfer function", and "Resolution" are used frequently through the manuscript without proper definitions. The reviewer found them confusing in several places. Please clearly define them in the manuscript.
In addition, the authors made mistakes in estimating the nano-RDMA transfer function (section 3.3). Similar mistakes occur in Brunelli et al. (2009). Please refer to the second paragraph in Page 484 of Jiang et al. (2011a) for details

To clarify the terminology we replaced the word “penetration” with “transmission” throughout the manuscript and we added the definition of transmission and resolution in the paragraph at page 11 from line 25, where we cited Jiang et al., 2011 as suggested by the Referee.

3.) The reviewer has some reservations on the qualitative explanation illustrated by Fig 5. When the ES chamber is grounded (left and right sub-figures), the electric potential at ES chamber is zero rather than several thousand volt shown in these figures. When the ES chamber is floating, **how to justify that the electric potential at ES chamber is several thousand volt?** High voltage of several thousand volt was applied on the liquid. If the ES chamber is at several thousand volt, **one may not have the cone-jet ES**.

The authors think that the reservations of the Reviewer are a misunderstanding generated by the inadequate labels of Figure 5. In fact, the labels are too small. Therefore, the Reviewer couldn’t see that the scale was logarithmic, hence the apparent non-linear decrease in the right sub-figure. The decrease in Figure 5 is in fact linear and the electric potential at ES chamber is only 500 Volts, not several thousands. The liquid of the electrospray was kept at 5.5 kV, therefore there was enough potential difference to generate a Taylor cone at the tip of the electrospray.

Once this mistake is corrected, **the authors need to rethink how to explain the high transmission when the ES chamber is floating.** Does this have something to do with the high space **charge density at the outlet of electrospray**? In addition, the non-linear (vs. linear?) **decrease in the electric potential in the right sub-figure needs to be justified**.

We changed Figure 5 in order to make it more intuitive.

4.) Although the improvement in CLOUD chamber measurement was obvious when comparing Fig 3 and Fig 10, **the reason why the Caltech nano-RDMA with the original aerosol inlet can not measure sub 3 nm particles from the CLOUD chamber was not clearly summarized in the manuscript**. The authors imply that this is due to bipolar ions are present when sampling from the CLOUD chamber, while the good performance of Brunelli et al. (2009) and Jiang et al. (2011a) are achieved with unipolar ions, e.g., electrosprayed tetraalkyl ammonium ions. The charge saturation effect may explain the observed high ion transmission. If this is the case, will clear signal be observed for ES gnd (Fig 4) when running the system for a long time since they are also unipolar ions?

We added the following sentence in the conclusions at line 14 to summarize our explanation.
“We attribute the improvement of the transmission of the nRDMA to a mitigation of the adverse electric field in the inlet region.”

As explained in the answer to Referee#3’s first comment, the results of Brunelli et al. (2009) are not directly comparable to this work, as they do not show transmission values. However, the results of the comprehensive work by Jiang et al. (2011a) do. The discrepancy between the transmission measured in our work and the one shown in detail by Jiang et al. (2011a) is explained in the paper with the saturation of the insulators shown in Fig. 7 (Line).

Concerning the signal from the ES, the referee is correct. In principle, running the ES for a long time with the setup shown in Fig. 4 should result in a high transmission. Unfortunately, we were not able to show this effect experimentally with the setup as in Fig. 4, for reasons that we cannot explain. Nevertheless, we were able to demonstrate the saturation with monodisperse ions. One could speculate that the presence of solvent in the ES experiments might hinder the saturation of the insulator. The molecules of solvent might change the surface properties of the insulator, allowing the deposited charge (deposited by the electrosprayed ions on the walls by diffusion) to move and dissipate.
The authors think that, despite the saturation was not measured in the electrospray case with the setup as in Fig. 4, their explanation of the improvement in transmission in the measurements in the CLOUD chamber is sound.

5.) More information is needed to explain how ion size distributions were estimated from the nRDMA-PSM measurement (as reported on Fig 10). **Which inversion method was used? What penetration efficiencies and transfer functions were used during the inversion?** In addition, the inlet design with the core sampling may **bias nanoparticle sampling (especially for highly mobile ions) from the inlet flow with the presence of the electric potential gradient. An analogy is the iso-kinetic sampling in a flue gas system.** Please discuss how to address this possible bias during size distribution measurement.
We added a more detailed explanation in paragraph 3.4 about how we estimated the size distribution.
The authors tend to exclude the possibility of a bias in the sampling, mainly because of the use of the core sampling. As shown in the calculations reported by Tammet (2015) the disturbances of the electric field lines (i.e., losses of particles) occur in the region next to the walls of the segmented tube. This effect is cancelled, or at least minimized by discarding the flow in the region closest to the walls and sampling from the center via the core sampling probe giving a sampling efficiency close to 1.

A few minor comments are given below,
a) Lines 9-11 in page 5856: what is discussed here applies to a bipolar charger only. Please be precise.
Correct. We added to the line "**in case of a bipolar charger is used**" .
b) Line 24 in page 5858: was this a mistake? i.e., I anticipate that you put the electrometer upstream from the insulator (rather than downstream from it as described here)
Yes, it is a mistake. Corrected with "**downstream**"
c) Line 13 in page 5860: high ion transmission was observed not only by Jiang et al. (2011a), but also explicitly by Brunelli et al. (2009).
Here, we do not think adding reference to Brunelli et al. (2009) would be correct, since in their study the ion transmission was not measured.
d) Lines 4-8 in page 5856 are repeating Lines 24-27 in page 5855.
We removed the lines at page 5855.
e) Figure 7 can be improved to make it self-explaining. For instance, which data is from electrometer? Which y-axis?

We updated the figure making it clearer. We changed the right-hand side y-axis label to green and added a legend.

f) Figure 10 can be improved. For instance, what is the y-axis in the right figure? What is the difference between (b) and (d)?

We improved the figure following the Reviewer's suggestions by clarifying the y-axis labels to panels b and d.

Cited literature:

Tammet, Hannes. 2015. "Passage of Charged Particles Through Segmented Axial-Field Tubes." *Aerosol Science and Technology* 49 (4): 220–28. doi:10.1080/02786826.2015.1018986.