We appreciate the thoughtful comments by referee #2. For discussion purposes we would like to respond to the general and detailed points raised.

This work describes the use of a plasma torch as an aerosol neutralizer. The work measures and compares the charging probability of the new source with commercial available other aerosol chargers. The charging probabilities were measured for positive and negative particles (Ag, and NaCl) and at different aerosol flow rates. In addition, the plasma torch has been evaluated at operation with different working fluids (He, Ar, N2). The mobility distributions and mass graphs of the charger ions were also measured in order to get an information on the properties of the charger ions. The work includes a very thorough investigation of the charger source and the charger ions. However, there are a few general concerns about the work, that should be addressed before publication:

1. The plasma torch itself, is not described at all in the current manuscript, a paragraph on the working principle should be added to the manuscript.

There is indeed not much information given about the charger itself and we acknowledge that this is unusual for a research paper. However, we have to point out that there is still a patent pending and, thus, we cannot reveal all the technical details of how the charger works. Broadly speaking the atmospheric pressure plasma charger consists of a gas flow that is shielded by another gas flow from the surrounding atmosphere. The plasma is ignited inside the inner flow while the aerosol is administered through the outer gas stream. The main source of the plasma is a high-frequency copper electrode that is situated on the central axis of those two gas streams.

We will add the following description to the experimental section:
"The atmospheric pressure plasma charger consists of a gas flow that is shielded by another gas flow from the surrounding atmosphere. The plasma is ignited inside the inner flow while the aerosol is administered through the outer gas stream. The main source of the plasma is a high-frequency copper electrode that is situated on the central axis of those two gas streams."

2. In the measurement with different gases, one would expect that using a DMA and CPC in a helium-air mixture would result into changes in the instrument performance. The voltage mobility relationship in the DMA is gas dependent, and in the CPC the flow calibration would change when adding helium to the system, also the supersaturation profile would change and the detection limit would shift to smaller particles (e.g. Thomas et al. 2018, Journal of Heat and Mass Transfer). These are points that should be addressed in this manuscript.

We will add the following statement to the manuscript:
"The additional flow rate from the working gas was at max 1/9 of the aerosol flow. According to Thomas et al. (2018) a cutoff drift to lower sizes for helium mole fractions below 0.67 was found for butanol-based CPCs. However, the used CPC in this study was operated with reduced temperature settings and thereby a lower detection efficiency was established (Tauber et al. 2019a). As a result, the recorded cutoff drift would therefore only influence the charging efficiency measurements conducted at < 3 nm. The resulting error is already covered for this particle sizes by the measurement uncertainties of nDMA and CPC."

3. The charger ion mass and mobilities were measured, however, no qualitative thoughts were included in how and why that would result into the observed changes of charging probability. It has been stated correctly that the charger ion composition plays an important role to the final charge distribution. But what is missing is to apply the information found in this work to existing theories and see if the trend agrees with the observations. Simulation results of charge distributions considering different ion mass and mobility of charger ions have been performed in the past, see for example Maisser et al, 2015, Journal of Aerosol Science

We compared our results to approximations given by Wiedensohler (1988) and Tigges et al. (2015) and performed calculations for different ion masses and mobilities. The results of the calculations are posted in the review comment #1 and will be added to the manuscript.

4. The results of the optical emission spectroscopy seem very isolated from the rest of the publication. It is not clear how these experiments were performed. This is a bit confusing, is this supposed to be part of the experimental setup description, or already an experimental results section? If it is experimental results, then the procedure of how these measurements were done should be added in a bit more detail in the experimental section. Was this a completely separate measurement, or did you do that while aerosol generation and charging was happening as well? This would require also a description of the source itself, which was already mentioned above. Was the optical emission spectroscopy done only in pure helium environment, and how would that be relevant to the rest of the measurement?

The optical emission spectroscopy was conducted as a separate experiment with the flow rates stated in the supplemental material but without aerosol generation. The emission spectra yield additional information about the charging mechanism and the plasma itself. The former encompasses the discovery that the charging of the aerosol particles is achieved via electrons that originate from the central electrode of the charger. The latter point includes, for example, the detection of singly charged He particles, which have a lifetime that is so short that they recombine before reaching the detector in the ioniAPi-TOF. Furthermore, the OES measurements led to a better understanding of the plasma behavior and, thus, the way of charge transfer from the plasma to the molecules or aerosols. The electrons are detached from the high-frequency copper electrode as those atoms are easily ionized. After leaving the electrode the charges attach themselves onto preexisting molecules or aerosol particles. As a result, based on the ioniAPi-TOF and mobility measurements it was shown that the different charging mechanism lead to comparable measurement results.

We will add the following to the experimental section:
“The optical emission spectrometer was located at the nozzle of the plasma charger and used to record spatially averaged optical data along the axis of the plasma source.”

Some more detailed comments:
Ad Section 2) Experimental Setup: No description, schematic or anything on the charger!

We will add the following description to the experimental section:
“The atmospheric pressure plasma charger consists of a gas flow that is shielded by another gas flow from the surrounding atmosphere. The plasma is ignited inside the inner flow while the

aerosol is administered through the outer gas stream. The main source of the plasma is a high-frequency copper electrode that is situated on the central axis of those two gas streams.
According to Kallinger et al. (2012), the used radioactive 241Am charger has a cylindrical geometry with an axial flow direction. The radioactive source is mounted on the inner wall. The chamber has an inner diameter of about 30 mm and a length of 120 mm. Furthermore, the soft x-ray charger is composed of a stainless-steel tube and a photo ionizer. The aerosol particles are directed along the tube towards the soft x-ray source and leave the charger via an outlet, that is oriented perpendicularly to the axis of the tube. The tube has an inner diameter of 30 mm and a length of 200 mm."

Page, Fig. 2: It seems like the mobility distributions were measured in an air Helium, Argon, or N2 mixture. But I don't see any discussion of the influence of this gas mixture on the mobility measurements. If the DMA has been operated in a closed loop this has to be considered. The mobility of THAB in a helium air mixture would not be the same, so how was the calibration done in this case? If this was considered and found to be negligible a discussion and reference has to be added. If it has not been considered, then this needs to be done.

The mobility distributions were measured with air as carrier gas and only for the plasma charger an additional working gas (Air, N2, He) was added. This working gas flow was between 40 and 280 cc/min. The mobility spectrum / calibration measurement with THAB was always recorded with air as carrier gas. After the calibration the charger was mounted to the setup and the experiments with different working gases was conducted. So, there was no helium air mixtures during the calibration runs in the sheath air of the UDMA.

Section 2.1. I think this should be numbered 3, not 2.1, since it does not seem to be part of the experimental setup

Thank you for making us aware of the wrong numbering. We will separate the results and discussion section from the experimental section.

Page 5, line 94, 95: What is the copper antenna for?

We will add the following sentences on P5 L89 to explain the antenna and its usage:
"Thereby the plasma jet is shielded by another gas flow from the surrounding atmosphere. The plasma is ignited inside the inner flow while the aerosol is administered through the outer gas stream. The main source of the plasma is a high-frequency copper antenna/electrode that is situated on the central axis of those two gas streams."

Page 8, line 144 says that the different masses of charger ions created in the plasma torch and the other charger sources might result in the observed differences. Can this be quantified. Is the mean mass, and mobility higher or larger than in the other case. How does an increase or decrease of mass and mobility affect the final result. Why did you not apply the measured mass and mobility to the theory?

We would like to thank the reviewer for his thoughtful comments and make him aware of the performed calculations for different ion masses and mobilities which are posted in the review comment #1 and which will be added to the manuscript.

Page 9, line 163, why would it charge better in air than in helium? And how can the large difference of 50% be explained?

We will add the following paragraph to the manuscript:
"According to Maisser et al. (2015), nitric acid has an anomalously high gas phase acidity for its mass and can persist in the gas phase in higher concentrations than other low mass species. By using helium as working gas the concentration of nitrate ions in the gas phase is lower than in air or N2 and therefore charge transport decreases. This is contrary to using N2 as working gas where an increased charging efficiency up to 50% was measured."

Page 10, line 185, polarities wrong, also, you should mention that it was the y-axis that was normalized

We will change it in the manuscript accordingly:
"The negative mass spectra were normalized to the nitrate ion (NO^-_3) peak at an integer mass of 62 Th and the positive mass spectra to the $(H2O)2{\cdot}H3O^+$ water cluster at an integer mass of 55 Th."

Page 11, Fig. 7, for negative mass graphs the chemical equations are mentioned but not the mass, while for the positive ions it's the other way around. Is there a reason for that? What are the rectangles in the positive Am-241?

The dashed square box marks unidentified masses in the positive 241Am mass spectrum and the solid square box shows the silicone compounds that are listed in Table 2 in the Manuscript. For space reason we mentioned only the chemical equations for the negative masses but in Table 3 all chemical equations and masses are listed.

References:

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5. Tigges, L., Wiedensohler, A., Weinhold, K., Gandhi, J., and Schmid, H.-J.: Bipolar charge distribution of a soft X-ray diffusion charger, Jour-nal of Aerosol Science, 90, 77-86, https://doi.org/https://doi.org/10.1016/j.jaerosci.2015.07.002, 2015.
6. Wiedensohler, A.: An approximation of the bipolar charge distribution for particles in the submicron size range, Journal of Aerosol Science, 19, 387 – 389, https://doi.org/https://doi.org/10.1016/0021-8502(88)90278-9, 1988.