We appreciate the thoughtful comments by referee #1. For discussion purposes we would like to respond to the general and detailed points raised.

Manuscript by Tauber et al. reports characterization of a new bipolar non-thermal plasma type charger for charging sub-15 nm aerosol particles. The main results of the study are measured size dependent charging efficiencies that are strongly biased to negative polarity, and mass and mobility characterization of the charger ions. Experimental data on sub-10 nm charging efficiencies are in the general interest in aerosol measurement techniques, while the current manuscript requires elaboration on several places prior to consideration for publication. General and specific comments are listed below

General:

- Experimental section: there is absolutely no description of the new plasma charger which is being characterized. This undermines the reproducibility of the most important results of the whole manuscript. The operation principle and general characteristics of the charger have to be described prior to publication. Otherwise a serious scientist in another lab cannot reproduce the potentially interesting results obtained with the charger. This applies also to the other used chargers.

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.
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."

- P8 L136 onward and P15 L223 onward, the authors discuss the effect of ion mass on the charging probability. They have even measured the ion masses and mobilities, while still in the theoretical prediction they use presumably "wrong" ion masses and mobilities. Why? Why not predicting the charging efficiencies with the measured ion mobility and mass? This is also speculated in the conclusions with statements that the charging efficiencies biased to negative polarity are explainable with the ion mobilities, while not a single ion mobility value (which should be actually weighed with the ion concentration) is reported even if they are measured.

We would like to thank the reviewer for making us aware of that and add the following to results section:
"In Table 1R the measured and calculated mean ion charger mobilities, mobility equivalent diameters, masses and ion mobility ratios are listed. For comparison the values found by Reischl et al. (1996) are also listed. The results were used to calculate the charge distribution with Fuchs theory as shown

in Figure 1R. Negatively charged particles in the size range from 4-12 nm by 241Am or X-Ray neutralizers agree well with the parameters derived by Reischl et al. (1996) and an ion mobility ratio of 1. For positively charged particles the charging efficiency is below the measurement results for particles between 4-10 nm. By correcting the charge distribution with the parameters derived by Reischl et al. (1996) with an ion mobility ratio of 0.8 the negatively charged particles with a size below 4 nm fit perfectly to theory. The measurement results of this work reveal an increased charging efficiency for both polarities as shown in Figure 1R. For mobility equivalent diameters between 4 and 12 nm and positive polarity the charging efficiency fits with theory for 241Am, X-Ray and the plasma torch with air as working gas. In contrast to negatively charged particles where the results of the plasma torch with nitrogen or air as working gas above 7 nm is higher and below 7 nm is lower than expected by theory. Also, for 241Am, X-Ray and the plasma charger with helium the theory exceeds the measured charging efficiency. By correcting the theory with the acquired ion mobility ratio, a good agreement between theory and measurement can be found for negatively charged particles above 7 nm and for positively charged particles for the plasma torch with nitrogen as working gas. Although the effect for diameters > 7 nm can be explained, there is still a deviation for the smaller diameters from theory. The reported discrepancy can therefore not solely be attributed to the ion mobilities. There are other effects which should by investigated in further studies. Especially the charging effects below 5 nm which cause deviations from the charging model."

Table 1R. Comparison of ion cluster properties: polarity, mobility diameter Dp calculated from mean ion mobility Z, mean ion mass M and ion mobility ratio Z^-/Z^+.

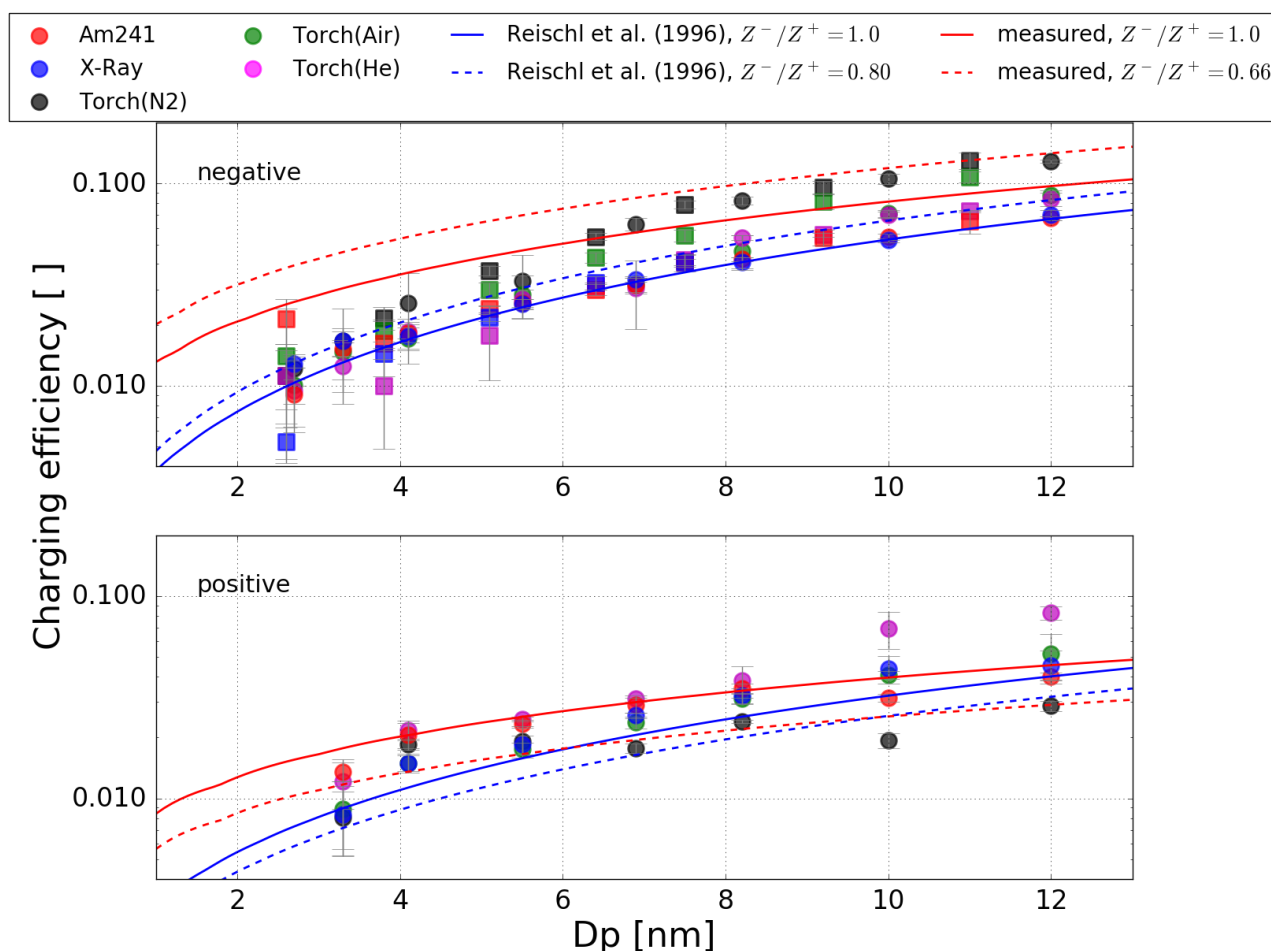|	Polarity	Dp [nm]	Z [cm2/Vs]	M [Da]	Z^-/Z^+	
Reischl et al. (1996)	+	1.32	1.15	290	1.0	0.80
Reischl et al. (1996)	-	1.19	1.43	140	1.0	0.80
measured	+	1.07	1.76	356	1.0	0.66
measured	-	0.87	2.66	116	1.0	0.66

Figure 1R. Measured charging efficiencies for the different aerosol chargers for negatively and positively charged Ag particles (dots) and negatively charged NaCl particles (squares) with mobility diameters less than 12 nm. The lines represent the charge distribution according to Fuchs theory, the parameters for the ion mobilities, ion masses and ion mobility ratio are listed in table 1. The collision probability of ions was calculated following Hoppel and Frick (1986).

- Generally there are several experimental details that are not reported in sufficient detail, or statements of which interpretation is ambiguous. Some of these are picked below.

Specific:

- P1 L5, "increased charging efficiency", compared to what?

The sentence will be corrected:
"A comparison of the different neutralization methods revealed an increased charging efficiency for negatively charged particles using the non-radioactive plasma charger with nitrogen as working gas compared to a radioactive americium neutralizer."

- P1 L7, "charging mechanism", are you referring to the different neutralizers? Charging mechanism is a different thing

Yes, we refer to the different neutralizers.
The sentence will be corrected:
"The mobility and mass spectrometric measurements show that the generated neutralizer ions are of the same mobilities and composition independent of the examined neutralizer."

- P1 L8, in which sense the TSI X-ray is "standard" neutralizer? I would say commercial

The sentence will be corrected:
"It was the first time that the Gilbert Mark I plasma charger was characterized in comparison to a commercial TSI X-Ray (TSI Inc, Model 3088) and a radioactive americium neutralizer."

- P1 L9, "enhance" compared to what?

The sentence will be corrected:
"We observed that the plasma charger with nitrogen as working gas can enhance the charging probability for sub-10 nm particles compared to a radioactive americium neutralizer."

- P1 L10, "increased down towards", reformulate

The sentence will be corrected:
"Consequently, the limit of detection of differential or scanning mobility particle sizers can be increased to nanometer sized particles with the Gilbert Mark I plasma charger."

- P1 L20, "below 10 nm in diameter are typically difficult to neutralize", I would say the opposite

The sentence will be corrected:
"Aerosol particles below 10 nm in diameter are typically difficult to charge and carry only one electrical charge at maximum (Wiedensohler, 1988)."

- P1 L21, "Quantitative particle detection in this size range is extremely challenging due to high diffusional losses. Hence, a higher charging efficiency is of importance to improve the signal intensity in the sub-10 nm regime", the first sentence implies challenges in determining the diffusion losses, i.e. systematic uncertainty, while the second one deals with detection limit and poisson type random uncertainty. Improvements in the second does not help the first one.

We thank the reviewer for making us aware of this unclear formulation and the sentences will be reformulated:
"Quantitative particle detection in this size range is extremely challenging due to high diffusional losses, which results in low number concentrations. Therefore, a higher charging efficiency is of importance to increase the detectable number concentration in the sub-10 nm regime."

- P2 L33, it will be good to define what is non-thermal plasma, if it is the operation principle of the new charger

We fully agree with the referee on this point and added the following sentences to the manuscript:
"The term non-thermal plasma is usually used to describe a discharge in which the electrons are in thermal non-equilibrium with the ions. This means that the average temperature of the gas in such a discharge is far lower than the temperature of a thermal plasma (i.e. some hundred K compared to several thousand K in the latter case)."

- P2 L39, "mobility spectrum of . . .", it is the mobility spectrum of the charger ions, not of the chargers.

The sentence will be corrected:

"In the past, various studies have characterized the charging probabilities and mobility spectra of the charger ions produced by AC-corona, X-Ray or alpha-radiation based chargers (Wiedensohler et al., 1986; Steiner and Reischl, 2012; Kallinger et al., 2012; Kallinger and Szymanski, 2015)."

- P2 L46, "steady-state", earlier you talked about equilibrium, which one it is?

We measured the charging probability and assumed that the charge equilibrium inside the charger leads to a well-known size-dependent charge distribution. The chargers were measured during steady-state operational conditions. We will reformulate the sentence:
"Here we report on size-dependent charging probability measurements of a non-thermal plasma source (Gilbert Mark I plasma charger, Gruenwald Laboratories GmbH, Austria), an americium 241 (241Am) charger and of a TSI Advanced Aerosol Neutralizer 3088 by means of a tandem DMA (Differential Mobility Analyzer) setup as depicted in Figure 1."

- Figure 1, what is the sheath the flow rate and size resolution of the nDMAs? What about the furnace output distribution? How much the size resolution of the first DMA affects the results? Based on the plot, the DMA2 was operated with aerosol flow of 8 lpm at some point, can it really handle such high flow? Is it so that only the Torch requires the working gas? It is not clear from the figure. If the torch mixes helium with the sample flow, it changes the gas composition. Does it affect sizing of the DMA or the flow rates of the CPC? If yes, how was this accounted for?

The ratios for sheath flow and aerosol flow for the nDMA are listed below:
sheath flow 19.5 lpm / aerosol flow 2.5 lpm = 7.8
sheath flow 33.0 lpm / aerosol flow 5.0 lpm = 6.6
sheath flow 41 lpm / aerosol flow 8.0 lpm = 5.1

The furnace output distribution is constant because the 3 lpm through the furnace were not changed. The additional flow was introduced after the aerosol size classification and the aerosol distribution was checked before a measurement run to reach the desired concentration for the analyzed particle size. Both nDMAs are of identical construction and the classifier (nDMA 1) was always operating at the same sheath flow ratio (19.5 lpm / 3 lpm = 6.5). Consequently, we assume negligible effect of the size resolution of the first nDMA.

The geometric standard deviation of the particle size for the used nDMAs was evaluated by Winkler et al. (2008) for a sheath flow of 25 lpm and an aerosol flow of 4.6 lpm and is below 1.05 for particles with a mobility diameter down to 2 nm. The resulting flow ratio (25 / 4.6 = 5.4) is close to our measurement with 8 lpm aerosol flow and the signal-to-noise ratio was comparable to the measurements with lower aerosol flow rates.

Only the Torch requires an additional working gas, we will add the following sentence to the figure caption: "The additional working gas flow is only needed for the plasma charger."
The additional flow rate from the working gas is at max 1/9 of the aerosol flow. According to our measurements no sizing effects were observed. Since the generated charger ions are comparable, one can assume that the charged particles are comparable, and the sizing of the DMA is not affected. In addition, helium is an inert gas and therefore non-reactive and combine rapidly with the surrounding substances.

We will add the following paragraph in the experimental setup section:
"The geometric standard deviation of the particle size for the used nDMAs was evaluated by Winkler et al. (2008) for a sheath flow of 25 lpm and an aerosol flow of 4.6 lpm and is below 1.05 for particles with a mobility diameter down to 2 nm. The resulting flow ratio (sheath / aerosol = 5.4) is close to our measurement with 8 lpm aerosol flow and the signal-to-noise ratio was comparable to the

measurements with lower aerosol flow rates. The different flow settings for the nDMAs are listed in Table 2R."

Table 2R. Flow rates for the aerosol and sheath flow for the used nDMAs with the calculated sheath flow ratio.

nDMA	Aerosol flow [L/Min]	Sheath flow [L/Min]	Ratio
1	3.0	19.5	6.5
2	2.5	19.5	7.8
2	5.0	33.0	6.6
2	8.0	41.0	5.1

- P2 L56, which kind of ion trap?

The ion-trap used consists of two symmetrical half shells separated by an isolator. The charged ions and particles were removed by an ion-trap which was set to +/- 500 V. The capturing efficiency of the electrostatic precipitator was found to be > 99% in the size range between 1 and 10 nm (Brilke et al. 2020; Tauber et al., 2019b).

- P5 L93-94, "It can be concluded from the OES spectra that there is no ionization of aerosol particles facilitated by the carrier gas, since only neutral helium emission lines have been recorded." It is not clear what is the experimental setup during this experiment. It is only mentioned that helium is fed to the charger, there should be no particles in helium in the first place?

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.

- P5 L94, "atoms from the copper high frequency antenna", what is this antenna?

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."

- Section 2.1.1, what is the purpose of this section, or what we should conclude in terms of aerosol charging? Are the emission spectra comparable to the conditions of the charging efficiency experiments, or to the recorded mass spectra?

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 origin 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. On the other hand, the optical emission spectroscopy (OES) measurements were done in order to compare this novel charging mechanism to established ones like the X-ray or Americium chargers. 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 the preexisting molecules or aerosol particles. As a result, based on the ioniAPi-TOF and mobility measurements it can be shown that the different charging mechanism lead to comparable measurement results.

We will change the following sentence and include the used temperature settings:
"Two butanol-based CPCs (TSI 3776 UCPC) with reduced temperature settings (Condenser 1.1°C, Saturator 30.1°C, Optics 31.1°C) compared to factory settings to increase the particle counting efficiency were used (Barmpounis et al., 2018; Tauber et al., 2019a)."

We will move P6 L100-110 to the experimental description section and exchange Figure 1 with the Figure R2 because of a misleading plot. The number concentration measurements were done after the dilution and not before as shown in the previous plot. The number concentration measurements of CPC 2 were inverted to calculate the number size distribution from the mobility distribution according to Petters et al. (2018), which was then compared to the average particle concentration measured by CPC 1.
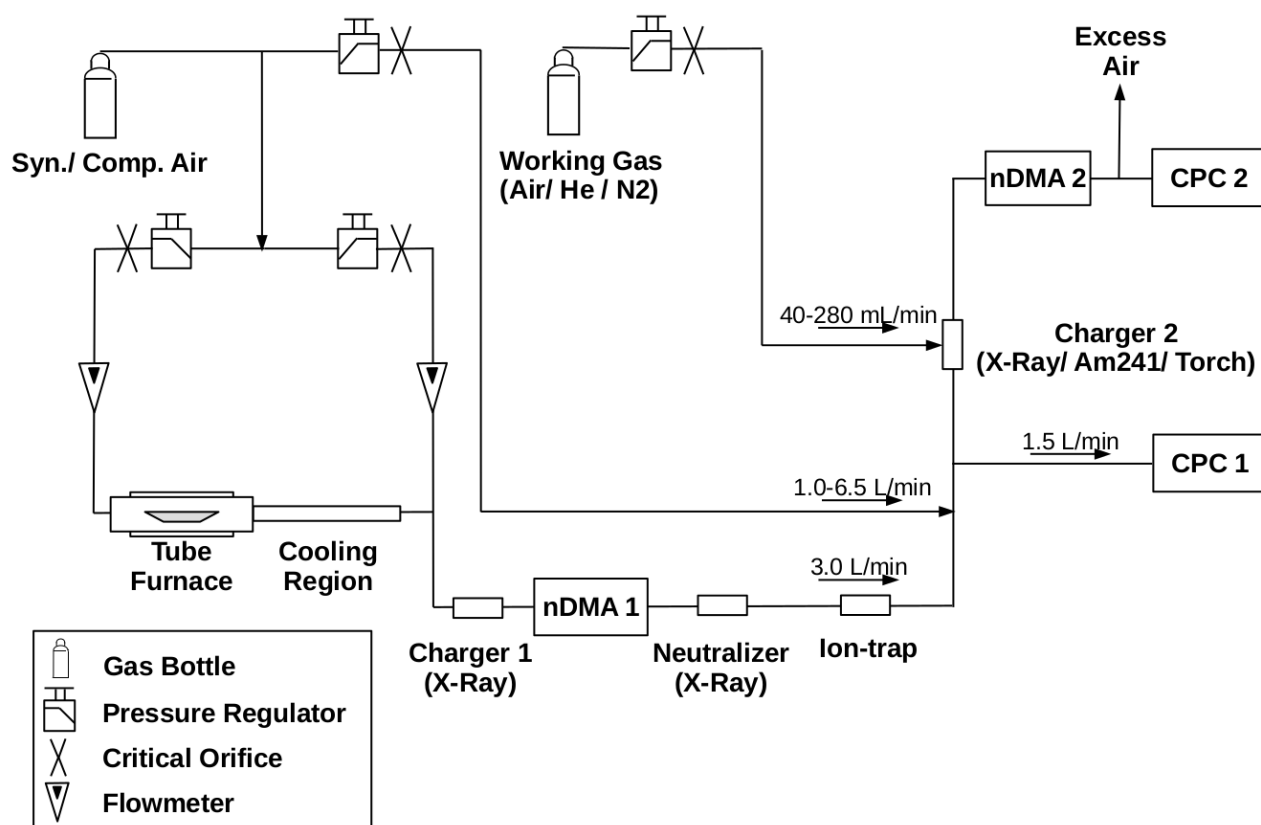

Figure 2R. Schematic of the experimental setup for the charging probability and particle size distribution inversion measurements. The additional working gas flow is only needed for the plasma charger.

in agreement with theory. Then the next sentence states this is not true for NaCl particles. Which one is true? Please elaborate the whole paragraph

We will change the following paragraph accordingly:
"As the particle concentrations for very small diameters between 2-4nm are below 10000 #/ccm, the charging efficiencies of the different devices vary strongly at these particle sizes. The nDMA in this size range has a low transmission efficiency ranging from 20-55% and therefore the signal at the CPC 2 is very low. In addition, small temperature-fluctuations in the tube furnace lead to bigger uncertainties for the low total number concentration of the selected particle size. Especially for NaCl particles a big variation in the charging efficiencies of the different charging devices was observed. Those variations almost certainly are caused by the low number concentrations of NaCl particles at these sizes compared to Ag particles. However, except for NaCl particles at particle diameters between 2-4nm the associated data points for the 241Am neutralizer agree well with the approximations by Tigges et al. (2015) and Wiedensohler (1988)."

- P7 L129, agrees with what?

We will modify this sentence accordingly:
"Evidently, the data using helium agree well with theoretical approximations from Tigges et al. (2015) and Wiedensohler (1988) in the size regime between 4 and 10 nm."

- P7 L131, what is dependent on what?

We will modify this sentence accordingly:
"The data suggest no dependency of the charging efficiency on the charger ion polarity, when helium is used as working gas in the plasma torch."

- P8 L137 onward, also charger ion mobility should affect the charging probability.

Thank you for making us aware of this inconsistence, we change this sentence as follows:
"Wiedensohler and Fissan (1991) have shown that the predicted charging probabilities of NaCl and Ag particles strongly depend on the used carrier gas and the ion mass and mobility."

- P8 L143 "As Figure 3 shows, the plasma torch forms copper ions and free electrons which charge aerosol particles in the carrier gas." Why these are not observed in the mass spectrum?

The ions and free electrons are not observed in the mass spectrometer because their life time is far too short to reach the detector. For example, the transition probability of the ionization state represented by the copper emission line at 775.40 nm is 9 x 10^6 /s (Kramida et al., 2017). Thus, the mean lifetime of this ion is 1/(9 x 10^6) ~ 0.1 µs. Even if we assume a close to sonic flow velocity of the gas as an upper limit, the maximum range of the copper ion would be ~ 300 m/s x 10^{-6} s = 0.03 mm before it recombines.

- P8 L147, "the ions can form in a pure nitrogen environment almost like in the mentioned case of Wiedensohler and Fissan (1991)", what do you mean?

We will modify this sentence accordingly:
"As the working gas flow is exposed to a high frequency electrical field before it mixes with the aerosol flow, the ions can form in a pure nitrogen environment. Hence the charger ions form in a nitrogen atmosphere like in the case of Wiedensohler and Fissan (1991) where silver and sodium chloride particles were charged with a Kr 85 source in a pure nitrogen atmosphere. "

- Figure 5. It is not described how the normalization is done for the right hand side plot. It is hard to believe that the differences in the distributions are only due to charging. At 8-10 nm the charging efficiency of the X-ray is about 10 times larger than of the torch which is opposite to what is reported in Fig4. On the other hand, at 3 nm the torch shows charging efficiency of a factor of 5 higher than the X-ray, which is also contradictory to fig4.

At this point we want to mention that Figure 5 is rather a qualitative representation than a quantitative one, because of the rotated periodical measurements which were conducted in multiple cycles. The discussed shifts in the number concentration especially for the small sizes, for example 3 nm particles might also be a result of the temperature-fluctuations inside the tube furnace. Furthermore, the deviation at 8-10 nm for the X-Ray charger might be a result of the ammonium sulfate contaminations measured with the mass spec and by the ion mobility measurements where an increase of about 0.25 nm for the negative ions was found. The comparison of the mobility equivalent diameters from the classifier nDMA to the scanning nDMA did not show a significant difference for the 241Am neutralizer and the plasma torch.

We will change the following paragraph accordingly:
"Figure 5 depicts the recorded negatively charged aerosol size distribution averaged over numerous measurements, as well as the charging efficiency of the individual chargers normalized to the 241Am charger recordings. The normalization was done by comparing the recorded total number concentration of the individual neutralizers with the recorded total number concentration of the 241Am charger. The inversion of the size distribution data was performed according to Petters (2018). These diagrams permit a qualitative descriptive comparison of the different charging devices. The left plot reveals a shift of about 1 nm in the maximum of the recorded size distribution between the X-Ray and the plasma charger, which might be a result of small shifts in the size distribution originated from the tube furnace because of small temperature-fluctuations. Furthermore, contaminations of the X-Ray charger from previous experiments with ammonium sulfate might also lead to an increased particle diameter for the observed neutralizer (Steiner and Reischl, 2012)."

Furthermore, will change the Figure 5 caption:
"The plot on the left side represents the same recorded negative Ag aerosol size distribution with a UCPC, depending on the mobility diameter for the plasma torch (N2), X-Ray and americium neutralizers. On the right-hand side all conducted measurements with the same aerosol distribution are normalized to the 241Am results for a direct comparison."

- Figure 6, the intro discusses with one sentence the nT product. Is that product sufficient in the chargers to reach the steady state charge distribution?

According to the manufacturer the applied X-Ray neutralizer can be used for aerosol flow rates up to 5 L/Min and the 241Am charger used in this study was characterized by Kallinger et al. (2015) for flowrates up to 5 L/Min. Hence, we used the 241Am neutralizer as a reference which reaches steady state charge distributions and compared it the other measurements. The higher charging efficiency might be a result of the high ion concentrations of ~10^{13}/cc which can be achieved in a cold plasma (Kurake et al., 2016). That is approximately 6 orders of magnitude above the ion concentration inside a conventional Am241 aerosol charger (Steiner et al., 2014).

- P10 L185, ". The positive mass spectra were normalized to the nitrate ion (NO−3) peak at an integer mass of 62 Th and the negative mass spectra to the (H2O)2 H3O+ water cluster at an integer mass of 55 Th", the polarities do not match

We would like to thank the reviewer for making us aware, 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·H3O+ water cluster at an integer mass of 55 Th."

- Section 2.1.3, previously there was discussion on the ionization of copper, why there is no copper signal in the charger ions?

Due to the low mean lifetime of this copper ions (about $1/(9 \times 10^6) \sim 0.1$ µs) it cannot be detected with the mass spec or with the UDMA setup.

- P12 L211, "(NH4)14SO− 4", is it really a cluster of 14 NH4 ammonium molecules?

We will change the following sentence:
"According to our mass spec analysis, this is due to ammonium sulfate contaminations from previous experiments."

- P15 L214, I would not say the are the same. Especially in positive there seem to be differences

We will reformulate this sentence:
"The presented measurements conducted with a non-thermal plasma source have shown that helium, nitrogen and air as working gases lead to the same ion species. According to the mobility and mass spec measurements the comparison of the plasma charger with the americium neutralizer indicates the same negative ion species. Whereas for the positive ions the measurements reveal a slight deviation."

- P15 L218, it was shown nowhere that the torch produces ozone.

Preliminary measurements were already conducted with an ozone monitor which will be included in the manuscript to support the statement in the conclusion section.

- P15 L219, increased compared to what?

We will change the following sentence accordingly:
"By switching the working gas to nitrogen an increased charging efficiency could be recorded for negatively charged particles compared to 241Am neutralizer."

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