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Atmospheric Measurement Techniques Discussions

Interactive comment on "Characterization of a Non-Thermal Plasma Source for the Use as a Mass Spec Calibration Tool and Non-Radioactive Aerosol Charger" *by* Christian Tauber et al.

Anonymous Referee #1

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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.

- 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.

- 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?

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

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

- P1 L9, "enhance" compared to what?

- P1 L10, "increased down towards", reformulate

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

opposite

- 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.

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

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

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

- 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?

- P2 L56, which kind of ion trap?

- 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?

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

- 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?

- P6 L103, “reduced temperature settings”, what were the settings?

- P6 L107 “In Tauber et al. (2019b) the particle counting efficiency of the CPCs used here was determined, and the results obtained were corrected for the CPC detection efficiency”, it is not clear what are used settings, since in the reference there are three different settings for the CPC.

- P6 L100-110, the description of the experiment should be in section 2 where the setup is described. How what about losses in the dilution, how they were corrected? The second DMA scans the voltage, what is the CPC2 signal that is compared to the CPC1?

- P6 L120, what the particle concentration has to do with charging efficiency? At extremely high concentrations concentration may affect the charging efficiency but not at low concentrations. Which CPC records the concentration that is mentioned? It is stated that data between 2-4 nm is 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

- P7 L129, agrees with what?

- P7 L131, what is dependent on what?

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

- 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?

- P8 L147, “the ions can form in a pure nitrogen environment almost like in the men-

tioned case of Wiedensohler and Fissan (1991)", what do you mean?

- 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.

- 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?

- 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 Âů $H3O^+$ water cluster at an integer mass of 55 Th", the polarities do not match

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

- P12 L211, "(NH4)14SO^-_4", is it really a cluster of 14 NH4 ammonium molecules?

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

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

- P15 L219, increased compared to what?

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