Reply on RC1
Anonymous Referee #2

Referee comment on "Surface charge of environmental and radioactive airborne particles" by Gyoung Gug Jang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-417-RC2, 2021

The charge of different aerosol particles is determined by means of an electrodynamic balance using optical microscopy for particle monitoring. From the comparison of sink velocities in free fall and migration velocities in an applied electric field, statements about density, size and charge of the particles are derived. Besides mineral dust powders and agglomerated engineered particles, radioactive uranium oxide particles are also investigated, for which self-charging is suspected as a result of radioactive beta and alpha decay.

While the manuscript is sound in the parts related to radioactivity aspects and the objectives become clear, it contains many inaccuracies and the conclusions made are not supported by the data presented. In particular, key aspects of particle characterization and charging remain unexplained or are not even addressed. Therefore, the manuscript cannot be recommended for publication in the present form.

Particular comments:

Page 3: The determination of the electrical charge is explained using the Milikan setup in Fig.2. This is fine for dense spherical objects like oil droplets. But the particles used sometimes deviate considerably from dense and spherical morphologies. Here, effective densities and especially shape factors can be used, but this is later only marginally taken up for densities (tagged density for GO agglomerates) and not at all with respect to shape (e.g. in the manner of sphericity or dynamic shape factors).
P 4: The abbreviations "ICDD", "GSASII" and "CIF files" are not explained anywhere.

P 5, middle: “Before blowing the particles into the balance chamber, the pipette was vigorously shaken for 1 min to induce charging though the triboelectric effect.” This is one of the main points of criticism. Triboelectric charging of particles is an extremely complex process that depends not only on the material (e.g. work functions of particle and walls), but also on the type and number of collisions, on adsorbates and on environmental conditions. Good review articles on this can be found, for example, in Matsusaka et al. (2010), Mirkowska et al. (2016), Zou et al. (2019) and Lacks & Shinbrot (2019). Even for particles of the same size from the same material, bipolar charge distributions are observed and the charge polarity can even switch from negative to positive with increasing number of collisions. Therefore, the type of particle charging presented here is completely undefined and is not suitable for drawing conclusions about the charging behavior of different particles (varying in material, shape, internal structure, surface roughness, surface conductivity, etc.).

P 5: “From among the particles that were in view, a number of particles were selected, and their velocities were monitored under various electric fields to determine their approximate size, density, and charge.” If I understand the procedure correctly, sink velocity and migration velocity in the electric field were determined on individual particles using microscopy. Only if $v_f$ and $v_r$ were measured for each individual particle, the presented evaluation makes sense. Otherwise, one has only average values, which would not allow a representation of charge vs. size, as shown in Fig. 5. Do the light spots shown in Fig.3 really come from UO2 particles with diameters of 0.5 µm?

P 6, Eq.(6): Since the pressure-dependent term in Eq.(6) is an approximation of the Cunningham correction, it would be helpful to explain for which Knudsen numbers this approximation is valid. In addition, as mentioned above, the shape influence of the particles was not taken into account in the derivation of this equation.

P 7: The densities of UO2 and U3O8 are reversed.
P 7: Where does the “tapped density” of the GO particles come from?

P 8: “Combining the size and free-fall velocities determined previously with the rise velocities determined through levitation experiments, Eq. 6 was used to calculate the charge q of the observed particles.” Here it sounds as if the free-fall-sink velocities were measured first and then the migration velocities in the electric field were measured on other particles (of the same material). As explained above, this makes no sense at all and leads to unmeaningful results, as can be seen from the huge standard deviations (in part significantly larger than the mean value).

P 8/P 9: “The average charge carried by the urban dust particles was just over twice that amount at $5.12 \times 10^{-18}$ C, with SD = $1.12 \times 10^{-18}$ C, or approximately 32 e, with SD = 7 e., though the dust particles were also approximately 2.5 times larger.” The comparison of uranium oxide with urban dust makes no sense. For one thing, only a vague average value of the diameter is 2.5 times larger, and for another, different materials charge triboelectrically in a completely different way. Therefore, apples and oranges are compared here. Also the significant charging difference between uranium oxide and Arizona dust is, except for one point, not recognizable. The statement that UO2 is substantially more charged cannot be recognized in view of the enormous ranges of the standard deviations.

P 9/P 10: “Based on the size and charge on the observed particles, the surface charge density for each of the particle populations examined in this study was determined and the results are shown in Figure 6.” The derivation of Fig.6 contains some doubtful assumptions. Although in Tab. 1 the surface area of uranium oxide was not determined (N/A), a value must be assumed here to calculate the surface charge density. However, this value is not given. On the other hand, for SiO2 and GO, the primary particle sizes are used to determine the specific surface area. This reduces the surface charge, although the structure of the agglomerated particles has not been characterized and therefore no information about the charge distribution over these particles is available. Especially hydrophobic SiO2 has a very low surface conductivity, so that charges accumulate only on the outer surface and the surface specific charge is significantly higher. On the other hand, in Fig. 6, agglomerate diameters are used for the size of SiO2 and GO particles. This is not correct.
P 10/P 11: Self-charging simulation results: The discussion is very lengthy and ends with the uranium oxide particles unlikely to have developed a significant charge during the experiment. Therefore, it remains incomprehensible why in the conclusions it is said that “Self-charge from radioactive decay of uranium was believed to be the origin of these higher charging characteristics.”

It seems that the presented method is not suitable to quantify the amount of particle charge induced by radioactive self-charging. For this, other techniques such as Kelvin Probe Force Microscopy seem to be better suited.