“The influence of surface charge on the coalescence of ice and dust particles in the mesosphere” by Joshua Baptiste et al.

Authors response to the interactive comments of anonymous referee 1.

Referee 1: “*The treatment here of the effects of ‘polarization of surface charge’ is equivalent, for conducting spheres and dielectrics of ε > 80, treatments of collisions in terms of ’image charges’.*”

Authors response: Indeed, modelling interactions between conducting spheres that carry a charge using ‘image charges’ model has been the subject of many numerical and analytical studies over a number of decades. In contrast, comparable theoretical studies of interacting dielectric spheres began only quite recently. An image solution to the problem of a point charge outside a conducting sphere at zero potential was first proposed in 1845 by Thomson later Lord Kelvin who interpreted the Legendre series expressing the potential due to the actual charge on the sphere as the potential due to an imaginary point charge. Since then the classical Kelvin image theory for a charged sphere was successfully generalized by Lindell (J. C.-E. Sten and I. V. Lindell, *J. Electromagn. Waves Appl.* (1995) 9: 599); I. V. Lindell, G. Dassios, and K. I. Nikoskinen, *J. Phys. D: Appl. Phys.* (2001) 34: 2302) and extended to dielectric spheres (I. V. Lindell, J. C.-E. Sten, and K. I. Nikoskinen, *Radio Sci.* (1993) 28: 319; I. V. Lindell and K. I. Nikoskinen, *J. Electromagn. Waves Appl.* (2001) 15: 1075; D. V. Redžić and S. S. Redžić, *J. Electromagn. Waves Appl.* (2003) 17: 1625; D. V. Redžić, *J. Phys. D: Appl. Phys*. (2005) 38: 3991).

Many models based on the image charge theory exist today, and these include a variety of boundary conditions suitable for describing some aspects of experiment. However, at close separation image charge methods require increasing numbers of images leading to convergence problems for a series expansion of the electrostatic force. The main additional advantage of the methods developed in our group is that, unlike any ‘image charges’-based models, they provide an accurate quantitative analysis of surface charge density thus allowing us to study the physical effects underpinning electrostatic interactions. An instantaneous mutual polarisation of charge on the interfaces depends on the geometry, composition, charge, size, solvent, external fields, and is particularly strong close to the point where the particles make contact, i.e in the region where ‘image charges’ models are often unreliable.

Referee 1: “*That particles with the same amount of charge ‘should’ have dissimilar sizes for size dependent attraction is not a new result from Bichoutskaia et al. It has been known for decades from work on conducting spheres that only for large charge differences or large size ration can there be a significant attractive force due to image charges to oppose the Coulomb force. From the 1964 work of Davies (Quart. J. Mech. and Appl. Math. 17, 490-511) and 2004 work of Khain et al. (J. Appl. Met., 43(10), 1513-11529) it follows that for equal charges on equal sized spheres the forces due to the image charges induced by the spheres on each other exactly cancel out the Coulomb force as the separation of the spheres goes to zero.*”

Authors response: We do not claim novelty of this conclusion in the paper not do we use “new results” as a phrase. In all appropriate cases in the manuscript, we will make sure we use “our calculations confirm” rather than “our calculations indicate” and also add references to suitable results obtained with ‘image charges’ models.

Referee 1: “*Line 164. Yes, a uniform charge distribution definitely would be more appropriate. Why is an inappropriate distribution used?*”

Authors response: As we point out in the manuscript, the uniform charge model is not meaningful in the limits of very small particles and/or charges of the order of one-two elementary charges that we consider in this paper. However, as particle size and the amount of charge on its surface grow the uniform charge distribution becomes more appropriate. Having said that, it would seem that, for the chemical scenario of molecular-size, singly- or doubly-charged ions, the difference between the two approaches is negligible (see Figure 4 of the paper).

Referee 1: “*The air ions are produced by the cosmic ray flux in the mesosphere and lower thermosphere, and are present in essentially equal numbers (see comment below), giving rise to approximately equal numbers of positively and negatively changed aerosol particles. So most of the aggregation there will be due to oppositely charged or charged and neutral particles, and the same-sign encounters will be quite a minor contribution.*”

Authors response: The latter statement is certainly true. We have discussed this point on lines 49-54 of the manuscript in the following way: “The presence of negative, positive and neutral particles in the MLT region implies that Coulomb forces between oppositely charged objects are the main attractive component of any electrostatically-driven dust agglomeration process. However, in addition to the strong attractive interaction between oppositely charged particles, our predictions indicate that in some instances, attractive interactions between particles of the same sign of charge can also take place at small separation distances, leading to the formation of stable aggregates.”

As for stating the proportion of charged and neutral aerosol particles, we will clarify this in the revised manuscript. Indeed, the galactic cosmic rays are the main source of ionisation in the troposphere. The focus of our study is on particles present in the mesosphere and lower thermosphere where extra-terrestrial matter is deposited from the meteor ablation. Meteor ablation has its maximum and generates particles at roughly 80 to 120 km altitude. Since we are interested in the formation of ice particles, we only consider the atmosphere at mid and high latitudes (approximately greater than 60 degrees) where ice clouds can be formed when the summer temperature minimum around the mesopause is sufficiently low. At those latitudes, the particles that originate from the Sun and from Sun-magnetosphere interactions cause ionisation. (See, for example, figure taken from I. A Mironova et al. *Space Sci Rev* (2015) 194:1). In comparison to *e.g.* the solar photon flux this flux is more time-variable.

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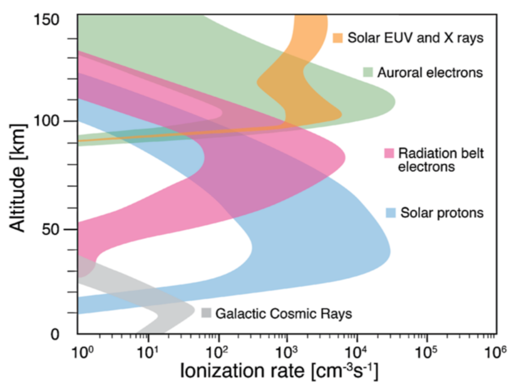
Fig. 1 Instantaneous ionization rates of EPP, Solar EUV and X rays in Earth’s atmosphere. The figure is modified from Baker et al. (2012)



It is also of interest to consider a possible influence of these ionisations because observations show that these particle precipitation events are often observed at the same time as polar mesospheric summer echoes, which are radar echoes that form in the presence of charged ice particles. In other words, it is interesting to study whether the conditions of particle precipitation can influence the growth of dust/ice particle. We will address this more clearly in the modified manuscript and we will also modify the title of this paper to include the conditions of lower thermosphere, i.e. "*The influence of surface charge on the coalescence of ice and dust particles in the mesosphere and lower thermosphere*".

Referee 1: "*with reference to the Figure 3 and the charges of - 2e on the oxides used for the calculations, the second charge is in the same location as the first charge. This is highly unlikely, and its use in this location negates the value of the calculations on this assumption*."

Authors response: We never actually put a charge of -2e on the oxide particles; the charge on metal oxides is kept at -1e, 0, +1 e. The charge of -2e corresponds to the ice particles. Figure 4 (line 4) indicates that in the cases involving ice particle with the charge of -1e or -2e the exact location of the surface charge does not affect the energy barrier (see a comparison between the point charge model and uniform distribution), which is the only contribution to the aggregation percentage in these cases. The difference begins to emerge as the charge on ice is raised to -5e (and higher) for which the uniform surface charge model is more appropriate.

Referee 1: "*Line 154. The coefficient of restitution 'CR' is taken as 0.9. For CR = 1.0 the aggregation probability would go to zero. What is the justification for this apparently arbitrary value?*"

Authors response: A. I. Ayesh et al. *Physical Review B* (2010) 81: 195422 gives a wide range of values for the coefficient of restitution for bouncing nanoparticles and shows that the coefficient of restitution is dependent on number of variables the angle and velocity of impact. We have now tested collisions scenarios where the coefficient of restitution was varied from 0.01 (extremely sticky, inelastic collisions) to 0.98 (almost elastic case) to show that values for the aggregation percentage remain the same in the entire range. Only for the purely elastic cases (>0.99), the aggregation percentage goes down by a very small degree.

The only set of data that might be affected, to a small degree, corresponds to the neutral - charged particle attraction presented in Table 4. However, the main point of this comparison is to investigate the importance of particle composition and the effect that dielectric constants and densities have on the aggregations. The value of 0.9 for the coefficient of restitution allows us to explore these differences for a wide range of particle size combinations.