

Interactive comment on “The mechanism of spray electrification: the waterfall effect” by James K. Beattie

Anonymous Referee #3

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Referee report on “The mechanism of spray electrification: the waterfall effect” by J. K. Beattie

General recommendation

The author investigated mechanism of water spray electrification, so-called balloelectric effect or Lenard effect. It is generally known that the breaking or splashing of water drops, as well as the bursting of bubbles in a water-air interface can generate large amounts of negatively charged particles and, therefore, a negative space charge in the atmosphere during rainfall and close to e.g. natural waters, the sea waves and waterfalls (Adkins, 1959; Gathman and Hoppel, 1970; Levin, 1971; Reiter, 1994). The Lenard effect (Lenard, 1892) occurs when drops of pure water break up into numerous fine negative droplets. The Lenard effect has been observed in several location when

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measuring small and intermediate ambient ions (Tamm et al., 2009, Laakso et al., 2007; Kolarz et al., 2012; Kamra et al. 2015) which I can personally confirm.

What makes this study to stand out is the attempt to actually explain theoretically the Lenard effect on a molecular level. Author offers an alternative explanation to Lenard's original explanation. These small negative ions may have a role in atmospheric particle formation in small scale. The initial formation processes, i.e. nucleation, which occur in the particle diameter range well below 10 nm are the subjects of extensive research and some of these processes favor the negative charge. Usually, the atmospheric newly formed charged particles and the bursts of small and intermediate ions, which are generated during the rain, on the sea waves or at the base of a waterfall, are considered as separate phenomena. On the other hand, the recent airborne studies have shown that small charged particles are observed as an outflow from the clouds. Interesting question is whether those small ions are nucleated or formed due to by the splashing of water (water droplet breaking in air).

There are some issues to be resolved, mainly related to some corrections and providing more details, which help understanding the methods and the results. How well does the experiments support existence of these negative ions generated from splashing water? In the light of the field and laboratory observations, is the theoretical size distribution of these charge carriers in agreement with the measured/observed size distribution? What is still unclear to me is the large drops and/ or the bulk with positive net charge?

Specific comments

Page 3, line 29-34: To add recent studies by Tamm et al. (2006) and Kamra et al. (2015 and references therein) on the direct observations of small balloelectric ions.

Page 3, line 32-22: In atmospheric science, the small clusters usually refer to air ions smaller than 3 nm and larger ions are called intermediate ions (3-7 nm) and large ion or charged particles (larger than 7 nm). Therefore, “. . .clusters of water molecules less than 30 nm in diameter. . .” should be replaced with “clusters of water molecules and

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water droplets less than 30 nm in diameter”.

Page 6, lines 89-95: What is the actual size distribution of these charge carriers? Does it agree with the atmospheric observations? E.g. Tammet et al. (2007) observed balloelectric ions in at ~ 7 nm whereas Kolarz et al. (2012) observed that the most abundant sizes were nanometer sized ions at 2 nm and submicrometer ions at 120 nm.

Page 6, lines 96-98: “The waterfall effect is hence the consequence of the spontaneous negative charge of the air/water interface followed by the shear rupture of the overall neutral drop into nanoscopic negative fragments from the surface with a net positive charge in the core.” Please illustrate this as well in a figure (e.g. Fig. 2 or add to Fig 1). The figure would be a great help to show what is the negative “surface” and positive “core” of the droplet.

Page 7, lines 115-119: Laakso and Tammet are not the only ones studying this effect. Therefore, consider modifying the sentence.

Page 7, lines 115-119: In addition, could you comment how is this theory applied? Can it also be used to explain other bursts of intermediate ions generated by the splashing of water (e.g. generated by rainfall and sea-spray) so called the balloelectric ions?

Technical correction

Type check manuscript, e.g. references in the text, and modify the manuscript to match the ACP guidelines: www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html

Page 8, line 125: Reference list is missing following citations which are mentioned in the text: Tammet et al. 2009 and Kamra et al. 2015.

Page 5, line 71: Fig. 1 is missing caption, or? The figure was improved well from the previous version. The caption should explain the steps introduced in the figure.

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References missing:

Kamra, A. K., A. S. Gautam, and D. Siingh: Charged nanoparticles produced by splashing of raindrops, *J. Geophys. Res. Atmos.*, 120, 6669–6681, doi:10.1002/2015JD023320, 2015.

Tammet, H., Hörrak, U., and Kulmala, M.: Negatively charged nanoparticles produced by splashing of water, *Atmos. Chem. Phys.*, 9, 357-367, 2009.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2015-892, 2016.

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