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

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REFEREE 3 recognises that this paper does not conform to the style used to report new experimental results, but instead describes a new interpretation of observations that began with Lenart in the 19th century and include those reported just now in 2016. The paper argues that the waterfall effect, the presence of clusters of water molecules and droplets less than 30 nm in diameter with negative charges consequent on the splashing at a waterfall, is one specific example of the general formation of a double layer with a negative inner charge that occurs spontaneously when water forms an interface with a low dielectric hydrophobe. The thickness of the double layer in water at the natural pH of 5.6 is of the order 200 nm. Hence fragments of a larger drop formed by shear of the surface will have a net negative charge. Charge neutrality requires that the remainder of the larger drop and the bulk water formed by these must be positively

charged. Ultimately condensation of the smaller negative drops with the larger positive drops or pools restores the system to its original neutral state.

Zilch and colleagues have given a detailed account of how the rupture of a large drop with an excess of negative charge at the surface leads to a population of small, negatively charged droplets (Zilch et al., 2008). They attribute the accumulation of negative charge to the attraction of hydroxide ions to oriented water dipoles, but their account can be simply re-expressed in terms of the fluctuation-correlation explanation. The mechanics of the droplet formation remain the same.

The author is not aware of a theoretical model that predicts the size distribution of the negative ions. The distribution is likely to be very sensitive to the conditions in which it is formed, being a dynamic process. For example, a large difference is expected between the size distribution found in fresh water and that in sea water measured with identical parameters, because the properties of the water are significantly different, in pH, viscosity, ionic strength, etc. In response to specific comments of Referee 3:

References to Tammet et al and to Kamira et al added.

The names describing the different sized charges species has been adopted.

As discussed above, the size distributions under different conditions are expected to differ.

The added Figure from Zilch contributes to clarification of the process.

The last sentence referring to Laakso and Tammet has been replaced by several commenting on the generality of the concept presented in the paper.

âĂČ Referee 4 is disturbed by the departure of the paper from the conventional format used to describe new experimental results. As discussed above, this paper examines to consequences of a new idea, the origin and application of the spontaneous adsorption of hydroxide ions to nanoscopic droplets formed in a waterfall and by other means. The only 'new result' is the description of the effect of the adsorbed hydroxide

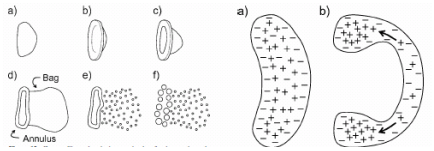

Fig. 1.