

## Comments on "Ion-Drop Interaction during Drop Evaporation"

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Takahashi and Endoh (1983) have described experimental results from which they conclude that water drops acquire a positive charge when they evaporate in an ion-rich environment. There is a long history of contradictory evidence on drop charging during evaporation, some of which is mentioned in the paper. Because of this and because such charging would have important implications with regard to the electrification that may occur when bodies of water or clouds evaporate, the evidence for evaporative charging must be carefully scrutinized. In particular it is necessary to demonstrate that the observations are not the result of some other effect.

Takahashi and Endoh's published account of their experiment is unclear in many respects and this raises a number of questions about the experiment and about the validity of their conclusions. Some questions and comments are as follows:

1) One major question is whether the charging that they observed was caused by the collection of spurious charges from the ventilating airstream. Apparent charging was observed only in an ion-rich environment when the relative humidity was low (experiment C). Section 2 of their paper states that the small ions for such an environment "were supplied by radioactive polonium and a small electric potential was applied to control the ion concentration." This technique for controlling the ion concentration should cause an unequal number of positive and negative ions to enter the airstream, resulting in a net space charge that would be collected by the drop. This obviously important possibility is not discussed in the paper, nor does the paper mention that the space charge density of the airstream was monitored.

2) The authors appear to have relied upon the results of a separate experiment to infer that the charging was due to evaporation. This experiment,

called "A," was similar to experiment C except that it was conducted in a "very humid" ion-rich environment obtained by setting up a "large hot bath" in a closed room with the experiment (and with the measuring and recording electronics). No charging was observed during this experiment, which presumably led the authors to conclude that the charging in experiment C was due to evaporation. However, no mention was made concerning how the electronics, insulators and ion-source functioned in the saturated or nearly saturated environment. Our experience is that the very small charges of such an experiment (less than 100 fC) are difficult if not impossible to measure in conditions of high humidity. At the very least, such measurements would require elaborate precautions and testing, none of which are mentioned.

3) The procedure adopted during the crucial experiment C in which charging was observed is not clear. The description of this experiment first states that "the ion concentration was initially adjusted to have no effect on the drops as in process A." No mention is made of this procedure or of the need for the procedure in the description of experiment A. However, the statement implies that some other adjustment of the ion concentration affected the drop charge. Next it is stated that "small ions were then introduced," presumably by adjusting the control potential at the polonium probe. It would appear that small ions were already present as a result of the first procedure, and that the second procedure would cause a net space charge to be introduced into the airstream. For these and the above reasons, it is not at all clear that the high-humidity experiment A was a control for experiment C.

4) Another experiment, "B," was conducted in which drops were evaporated in "natural air" having low relative humidity and no introduced ions. Not surprisingly, no charging could be detected. If this is

considered to be the only meaningful control experiment, the charging of experiment C is associated only with the introduction of ions and not with the occurrence of evaporation.

5) Not only did ions need to be present for charging, but ion-rich air was used actually to remove or reduce an initial negative charge on the drop at the beginning of an experiment. The authors have presumed that the charge reduction was due to the increased conductivity of neutral air, and they used the reduction rate to crudely estimate the ion concentration. But the charge reduction could have been aided or dominated by net positive charge in the airstream.

6) Other questions arise in reading the authors' account of their experiment. One concerns the charge measurements themselves. The charge signals corresponded to the small wiggles on the drifting waveforms in their Fig. 4, which were reportedly caused by motion of the drops in and out of the Faraday cage used for the measurements. Proper interpretation of the wiggles requires that they be correlated with observations showing when the drop entered and left the Faraday cage, but there is no indication that this was done. In the absence of such correlating information, it is conceivable that the wiggles were due to the presence of spurious charges or charged particles in the airstream. Presuming that the wiggles were caused by charge on the drop, the authors state that "only the maximum value of positive electrification" has been reported 'to eliminate those cases involving ion repulsion from positive drop charges in later stages of evaporation and cases where the drop does not float in the center of the Faraday cage.' Why would ion repulsion necessitate selecting the largest values, and on what basis were the data selected?

7) Another question concerns the effect of aerosols upon the experiment. The authors state that "the large variations observed probably reflect changes in the contents of the natural aerosol on different days." In addition, they state that "most experiments were carried out during the night or on weekends when traffic is at a minimum." These statements suggest that aerosols substantially affected the experiment, which is not surprising. In this case it would have been helpful to have some measure of the aerosol population. If exchanges of charge occurred between the drop and the surrounding air, they may have been produced not as a result of evaporation but by the attachment of charged aerosol particles to the drop.

In conclusion, we comment that the authors have the responsibility to make a critical evaluation of their own measurements and to communicate this evaluation to the readers. This has not been done in the paper and does not appear to have been done in the experiment. In the absence of such evaluation, we believe that the experimental results may have alternative explanations, as discussed above, that are at least as plausible as the interpretation that the drops acquire charge when they evaporate.

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#### REFERENCES

- Takahashi, T., and T. Endoh, 1983: Ion-drop interaction during drop evaporation. *J. Atmos. Sci.*, **40**, 463-468.