Atmos. Chem. Phys. Discuss., 15, C2767–C2774, 2015
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating collision efficiencies from contact freezing experiments" *by* B. Nagare et al.

Anonymous Referee #1

Received and published: 20 May 2015

General comments

The authors introduce a new method to study the collision efficiencies between aerosols and cooling water droplets at low temperatures, of interest for ice nucleation in contact mode. They derive the collision rate from experimental contact freezing data obtained with the ETH Collision Ice Nucleation Chamber CLINCH. They note that phoretic forces are possible factors affecting the scavenging of aerosol. The experimental collision efficiency is slightly higher than estimations given by theoretical expressions reported in literature. The paper brings new elements to the experimental determination of collision efficiency at low temperatures and is of interest for ACP readers. In general, the manuscript is well written, and the authors did a good job in showing its importance for contact freezing and for atmospheric aerosol scavenging. However,

in the current form, the paper is missing some important details and clarifications related to measurements, uncertainties involved, role of phoretic effects, and comparison with theoretical estimations. For these reasons, I recommend publication in ACP after major revisions, as suggested below.

Major revisions

1) The principal concern is related to the way the paper addresses its main goal, expressed in Introduction, last paragraph "This study therefore provides experimental data to validate theoretical formulations exactly in this least explored parameter space". The key experimental finding is summarized in Fig 6. It describes a single data point of efficiency collision (E) for water droplet of 80 microns diameter (D) with an aerosol of 0.2 microns diameter (d) and compares it with estimations for theoretical formulations. The paper will benefit if authors show experimental data for a range of aerosol diameters of interest and water droplet of different diameters. The size range will be constrained by the available instrumentation, and details should be described in the revised version.

The scavenging of aerosol by water droplet and particularly the phoretic effects depend strongly of aerosol and droplet sizes. For details, see Wang and Pruppacher, 1977, Fig 2, page 1667; Wang et al., 1978 (W78) , Figs 1-5, with focus on Fig 5. For instance, Wang and Pruppacher (1977) used water radius in the range 150 – 2500 microns, and aerosol radius of 0.25 microns. See also their Table 1 (page 1665) with a summary of what other researchers used as size ranges for water droplets and aerosol in their experiments until 1974. Recently, Ladino et al (2011), operating the same nucleation chamber CLINCH, used water droplets having radii between 12.8 and 20.0 microns and aerosol particles having radii between 0.05 and 0.33 microns. In their Fig. 1, Page 1854, they give the range of aerosol and droplet size used by many investigators in previous work. Thus, it seems possible and important to have more measurements for aerosol diameters in the accumulation mode and compare them with theoretical estimations.

2)The experimental data point shown with a E of 0.13 in Fig. 6, should be presented with an error bar, which can be represented similar to Ladino et al (2011) in their Fig. 7, page 1859, where "solid error bars represent the average uncertainty and the dashed error bars are the maximum and minimum range for E at a specific a and r" (in their work a is droplet radius and r is aerosol radius). Also note, in their conclusions, page 1860: "The collection efficiencies for our experimental data agree within one order of magnitude with the theoretical calculations." Since the current study uses the same nucleation chamber and similar instrumentation as Ladino et al, the E value will probably compare with theory estimations in a similar fashion, after both the experimental errors and the uncertainties in the theoretical estimations are shown and discussed. The E values estimated from the theoretical work have their own uncertainties and differences between different schemes. Such differences are at least one order of magnitude in the accumulation mode, as noted by previous work (see Wang et al., 2010; 2011 for comparisons and discussion of various formulations and comparisons with experiment). In the comparison from Fig 6, authors can display the uncertainty ranges from both experimental and theoretical estimations. In assessing the uncertainty of E from a theoretical expression, one can consider the possible variations of parameters in the experiment due to uncertainties of aerosol and water droplet sizes, electric charge, temperature, relative humidity (RH), etc. Then, use these variations to calculate a range of values for each aerosol size, and plot E versus particle diameter. This method will give some good idea about the uncertainties in the theoretical estimation of E.

3)The description of the experiment (Section 3) needs to be more detailed such that others can reproduce the results. A list of instruments and references can be included for this specific setup. In addition, the parameters of interest and their uncertainties need to be described quantitatively. For example, give the time evolution of temperature (T), relative humidity (RH), droplet diameter (D), size distribution of aerosol, charge on aerosol and droplet, estimations of loss of aerosol on walls. Is any significant variability of these parameters during individual experiments? . How many experiments were done to produce the data point from Fig. 6, and what is the variability of chamber

parameters during those experiments?

The experiment starts with water droplet of diameter 80 microns. Give the histogram of diameters of generated droplets (not all droplets have exactly 80 microns, there is a range of variation, and most likely, a narrow normal distribution with the maximum at 80 microns). Once a droplet starts falling, it can evaporate in time since the chamber is subsaturated with respect to water. What is the droplet diameter versus time in chamber and what values do you use in the theoretical estimations of E?. For comparisons of experimental E with theoretical E, it may be useful to calculate E for the observed/estimated range of diameter of the water droplets. Aerosol size is taken to have the average diameter of 0.2 microns, but how about standard deviation (STD)? How long do aerosol particles stay in the chamber? Is the aerosol size increased due to water condensation, coagulation or electrical interaction?.

The electric charge on water droplet is measured at beginning, but some statistics will be helpful, such as a distribution of charge, average, STD, min and max values. These values will be useful to estimate the maximum possible effects of electric charge on E. Does the charge on droplet stay the same during experiment? Can it change during experiment? Any assumption should be explained in more detail. The electric charge on aerosol is assumed "only one elementary charge" (page 12189). There is no indication that these charges are measured in this study. Are they based on some references about the instrument. Please clarify and provide specific references. If some particles have more than one charge (even a few elementary charges), it can increase the variation of the estimated E from electrophoretic forces (Fig 5). If some particles have opposite charges they can coagulate more efficient. Is this effect large enough to make a difference in E?

4)Phoretic effects. One concern is that using one data point makes it difficult to assess the role of phoretic effects. Measurements done at various aerosol sizes can give a better idea about behavior of E in the accumulation mode. Moreover, measurements are done with all factors acting in the same time (from Brownian diffusion, impaction,

and phoretic forces) and this makes it difficult to separate experimentally the role of various forces. Prodi et al (2014) show that one way to isolate the phoretic forces is to do experiments in microgravity conditions. In their study they find that diffusionphoresis predominates, contrary to the statement from this paper that "The combined description of thermophoretic and diffusiophoretic forces indicate that for our experimental conditions of evaporating droplets in the presence of rather small aerosol particles, thermophoresis should exceed diffusiophoresis" (page 12188). This needs more analysis and clarification, to provide experimental evidence of strength of each phoretic factor. For example, in the case of electric effects, the effects of image charges are not included and can be discussed in detail.

Specific comments

In abstract, "Freely falling 80, microns water droplets. . ." should indicate that 80 microns is the diameter of the water droplet.

In the statement "The chamber is kept at ice saturation in the temperature range from 236–261K leading to slow evaporation of water droplets giving rise to thermophoresis and diffusiophoresis." must say something about the values and evolution of relative humidity (RH) during experiment.

The statement "Droplets and particles bear charges inducing electrophoresis" can contain more quantitative information about the charge on aerosol and water droplet.

The reported collision efficiency of 0.13 can be given with plus/minus standard deviation or the range observed values (min – max). This can be done after revisiting issues as mentioned at point (1) above.

The statement "This discrepancy is most probably due to uncertainties and inaccuracies in the description of thermophoretic and diffusiophoretic processes acting together." does not have much support with the available data from this experiment, but it can be improved by adding data, and presenting it with error analysis when com-

pared with model estimation with their uncertainties, as mentioned in (1) above. The discrepancy you mention can be due at least in part to the experimental uncertainty or variability (please see the major comments above).

Introduction

In the last paragraph (page 12172), "The present study investigates collision efficiency of 200nm diameter silver iodide (AgI) particles with 80 microns droplets at low temperatures" need to specify that 80 microns is the diameter of water droplet.

Experimental setup

In section 3.2, the authors must explain and justify why "The particles have a charge of 1e after the size selection by the DMA which was operated at 1 liter outflow and 10 liter sheath flow air." Can aerosols charge change after they are generated, and how much?. Is charge affecting the aerosol coagulation and how much? These details can be important for the electrophoretic effects (see comments at points 2 and 3 above)

In section 3.3, "The mean charge on the droplets was about 65 fC (39 000e$\pm$20 000 e)". Does the charge on droplets remain constant after generation? Please see comments at point 3 above and include a more detailed characterization of the charge distribution on water droplets.

4 Experimental results.

Is any change in the size of water droplet during experiment, and how this impacts the estimation of E? How is the droplet diameter changing in time due to evaporation?

The method to estimate E from the frozen fraction (eq 19) should be described in more detail. Does Kgeo change in time? What are the errors in this calculation? Discuss the number of experiments conducted to get the reported E value? The possible errors associated with this procedure need to be analyzed and presented in more details.

8 Summary and conclusions

The range of values of the main parameters in chamber should be described. Instead of the statement “More importantly, comparisons of different theoretical formulations show considerable differences.” I suggest, “. . .comparisons of different theoretical formulations show differences within one order of magnitude in the accumulation mode” which seems to be seen in many reported comparisons.

The statements “There are large differences between the formulations for thermophoresis from A06 and W78 regarding size and temperature dependence. Calculated collision efficiencies for impaction in the size range from 0.5–2 microns strongly depend on whether a critical Stokes number is assumed. The temperature dependence of W78 is much larger than the one of A06.” do not appear as significant conclusions since they refer to comparison of two schemes, which can be done in sections 5 and 7, in connection with Figs 3- 5. Also, it seems that A06 and W78 agree within one order of magnitude for the range of parameters used in this experiment. Instead of “. . .W78 is much larger than the one of A06”, I suggest to compare them in a more quantitative manner, otherwise it could be misleading for someone reading only the conclusions. Reasons for such differences are given on page 12187, related to different regimes assumed, specific assumptions, and the comparison statements should be in that section. In the conclusion section, I think is more important to compare the experimental E with theoretical evaluations. A statement like “Our measurements agree with theoretical evaluations within one order of magnitude. . .” might be accurate (after you add the error bars in the experimental data and the estimated E).

One short paragraph at the end can give more details and specifics about what measurements are needed in the future to improve the experimental data for characterization of phoretic effects and/or useful for ice contact nucleation studies. Such paragraph will put this work in perspective and show its potential, which is otherwise well illustrated in the main sections.

Technical recommendations

Page 12204. Table 2 can be eliminated. Its content can be added in Table 1 at the end.

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