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Electrical effects on droplet behaviour

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Abstract.
The effect of charge on water droplets modulates various aspects of their behaviour. These include the droplet stability, evaporation, and lifetime. Microphysical models have been developed such that a reasonably good understanding of these processes has been achieved. However, the specific effects of charge deserve further scrutiny as they are an intrinsic component of the factors controlling droplet characteristics. Describing the effects of these requires an understanding of the electrostatic pressure present in the droplet and its surface tension. One way to test these effects and assess droplet response to charge is to take an experimental approach to make observations directly. In this study, individual droplets are levitated in an acoustic wave to allow isolated measurements to be taken. The droplets are monitored using a CCD camera with a microscope objective lens. In some cases, with sufficient charge present, effects on droplet stability can be observed as Rayleigh explosions, where a sudden drop in mass is seen superimposed on the evaporation profile. These events also allow the charge on the droplet to be calculated, which is then compared with the droplet evaporation. Another factor that plays a part in droplet behaviour is droplet composition. Different substances have different surface tension, and this is explored by performing some experiments on sulphuric acid droplets. Theory predicts that the more highly charged a droplet is, the more resistant to evaporation it becomes. Experimental data collected during this study agrees with this, with more highly charged droplets observed to have slower evaporation rates. However, highly charged drops were also observed to periodically become unstable during evaporation and undergo Rayleigh explosions. Each instability of a highly charged drop removes mass, reducing the overall droplet lifetime regardless of the slower evaporation rate. The sulphuric acid droplets were observed to be much more resistant to evaporation and no Rayleigh instabilities were observed.

1. Introduction

Droplets are small, quasi-spherical volumes of a liquid, the molecules of which are held together by surface tension forces. They are commonly found suspended within another fluid, such as cloud droplets in the atmosphere or moving through another medium, e.g. rain, under the influence of other external forces. Throughout this paper, we will largely be considering water droplets in air although the effect of composition will also be addressed.

The microphysics of droplet behaviour with regard to their physical and electrical properties have been intensively studied for well over 100 years. The physical properties such as composition, viscosity, and surface tension all have effects on droplet behaviour and stability. In addition, the droplets are subjected to electrostatic effects, both naturally occurring and externally imposed, which modify and regulate droplet behaviour in a significant manner. This review summarises these effects and explores the impact of charge on droplet behaviour.

Forces acting on droplets are of a diverse interest for multiple reasons. For example meteorologists are interested in the microphysics of cloud processes; other applications include inkjet printing and electrospray ionisation mass spectrometry among others. The fundamental science underpinning this droplet science has been thoroughly covered in a broad section of the literature from Rayleigh (1879, 1882) to the present as discussed below in Section 3. In the intervening time, microphysical models have been developed to simulate droplet evaporation and stability; these have become quite advanced and incorporate the broad range of variables acting on the droplet.

The droplet properties that define its evaporative style are the vapour pressure and temperature gradient across the droplet-vapour/vacuum interface as outlined in existing models. Charge, however, constitutes an important factor determining droplet behaviour and is considered in relation to other droplet properties. The charge structure of a droplet will generate internal electrostatic pressure, which, when considered along with its surface tension, plays a role in the stability of the droplet. A droplet that becomes unstable either through evaporation or sufficient charging may undergo a Rayleigh explosion and lose a proportion of its mass and charge in the process (Doyle et al., 1964; Duft et al., 2003). The Rayleigh explosion event itself consists of the formation of fine jets which erupt from the droplet surface when internal electrostatic pressure exceeds the effects of surface tension, which acts to maintain the spherical shape.

Following this introduction, Section 2 outlines the processes that lead to droplets having a net charge. Section 3 provides detail on the various aspects of droplet charge such as droplet charge structure, composition, evaporation, their interactions and the resulting implications; this section also incorporates some new lab results that demonstrate these factors. Conclusions are provided in Section 4.

2. How does a droplet charge?

The mechanisms by which droplets become charged are varied and a number of distinct processes can be observed.

Diffusion charging occurs when there are free ions in the region around the droplet, which adsorb themselves onto the droplet interface in a process whereby a net charge is transferred to the droplet. The magnitude of the net charge is determined by the difference in the number of positive and negative ions that become attached. As a precursor to this process, a process causing ionisation of the local environment is required. This could be, for example, ionisation in the atmosphere resulting from galactic cosmic rays or the decay of radioactive isotopes which could then go on to charge aerosol droplets such as those constituting clouds and fog.

Field charging occurs when there is an electric field present in the region surrounding the droplet. For example where a droplet such as a rain drop or cloud drop is present in the Earth's fair weather electric field. Where the electric field is negative (a positive potential gradient), the droplet will be induced with a positive charge on its lower surface and a negative charge on its upper surface.

3. Charge and droplets

3.1. Droplet composition

The composition of the droplet is important as the chemical characteristics can affect the physical properties of the fluid such as the droplet viscosity and surface tension. Surface tension is a critical characteristic of droplets as it affects droplet behaviour during evaporation. For example, in terms of cloud droplets, this can have implications for other planetary environments such as Venus, where the clouds are composed of sulphuric acid. Therefore, when cloud processes

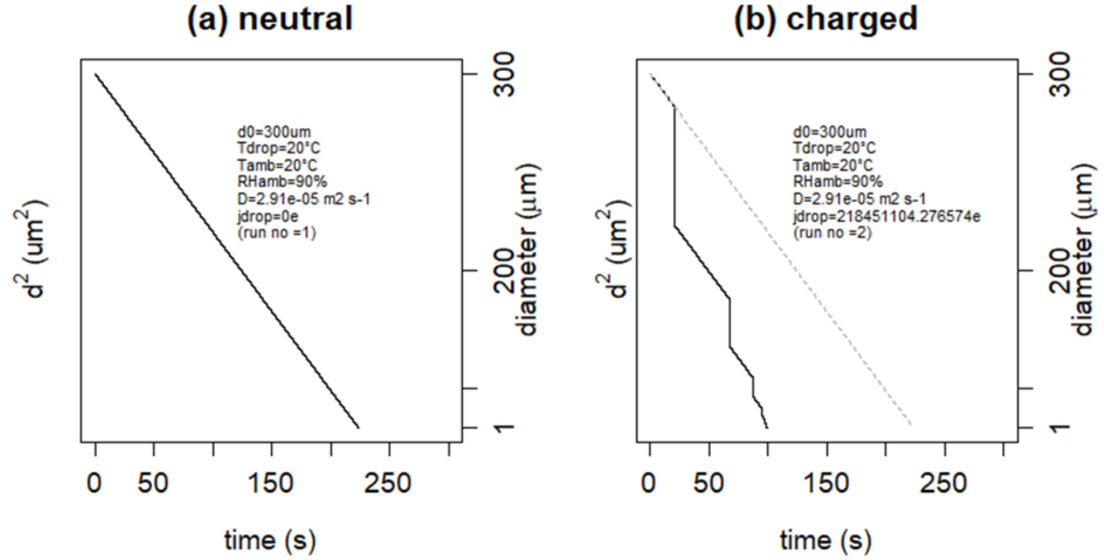

Figure 1. Models of evaporation for (a) a neutral drop and (b) a drop with 2×10^8 elementary charges. Initial droplet diameter is 300 μm, temperature is 20°C, and RH is 90%

are compared on different planets, charge is a parameter that must be incorporated into investigations.

3.2. Droplet charge structure

Regardless of the net charge on a droplet, the distribution of charge at the sub-droplet scale is not uniform (Kwan et al., 2019). A droplet contains positive and negative ions which may occur in particular locations within the droplet, such as at the centre or on the surface. Ion size is also important in determining its location, where modelling has been found to suggest that small ions prefer an inside position whereas larger ions, with higher mobility, show a preference to reside at the surface (Thaunay et al., 2017).

The interface between a droplet surface and its surroundings develops a charge structure known as the surface excess charge layer (SECL) (Kwan et al., 2019) consisting of a layer of ions attached to the droplet surface extending to 1.5-1.7 nm (Kwan and Consta, 2021). Kwan and Consta (2021) also note that the composition of the ions is important, with some ion species, for example I^- and H_3O^+, being present in much higher concentrations than others, for example Na^+ and Cl^-, for a droplet of the same size.

3.3. Droplet evaporation

The conditions surrounding a suspended droplet in a relatively less viscous fluid affect the transfer of material from the droplet to the environment or from the environment to the droplet. This is due to the vapour pressure at the interface between droplet and environment and further away from the droplet, surface tension, temperature gradient, and electrostatic forces within the droplet. Where droplet mass is lost to the surroundings, this is known as evaporation. Complex microphysical models have been developed to simulate these processes, including the addition of the effects of charge.

Dissolved salts in the droplet have a fundamental impact on droplet evaporation. Salt molecules at the surface of the droplet take up space that would otherwise be taken up by water molecules meaning that the concentration of water is reduced at the surface-surroundings interface, increasing the surface tension (Takhistov and Chang, 2002).

3.4. Experiments on evaporation

A study was conducted into how charge affects droplet lifetime. In order to do so, an experimental approach was taken using an acoustic levitator to isolate individual droplets and the droplet evaporation observed using a CCD camera equipped with an objective microscope lens. The process chamber is also equipped with an ioniser to introduce charge into the region surrounding the droplet, and two electrodes either side of the droplet between which an electric field can be applied.

In an initial run of experiments the ioniser was not used and droplets were suspended with no electric field applied. The relative humidity within the chamber was raised using water-soaked cotton wool to simulate a within-cloud environment. In each run, the droplet was observed to evaporate and the droplet diameter was measured at regular intervals to monitor the evaporation rate. In some of these runs, charge was sufficient to cause the droplets to undergo Rayleigh explosions, which could be observed in the data as sudden reductions in mass.

Figure 1 (a) shows the modelled evaporation of a neutral drop of diameter 300 μm in terms of diameter squared as a function of time. This relationship shows a linear trend as has been established previously in the literature. This relationship changes when charge is taken into consideration as can be seen in Figure 1 (b). Here it can be observed that when its diameter drops below the threshold of its Rayleigh limit an explosion occurs, resulting in a sudden reduction in mass thereby reducing the droplet diameter as seen in the plot.

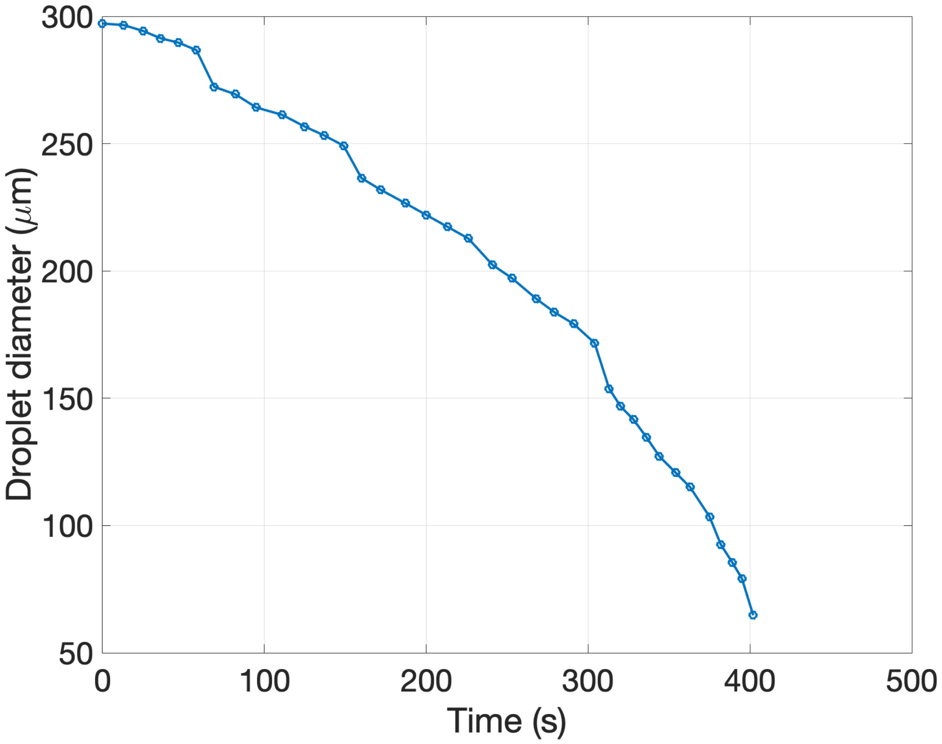

Figure 2. Levitated water droplet evaporation profile from droplet with sufficient charge to undergo Rayleigh explosions. Sudden drops in diameter are Rayleigh instabilities.

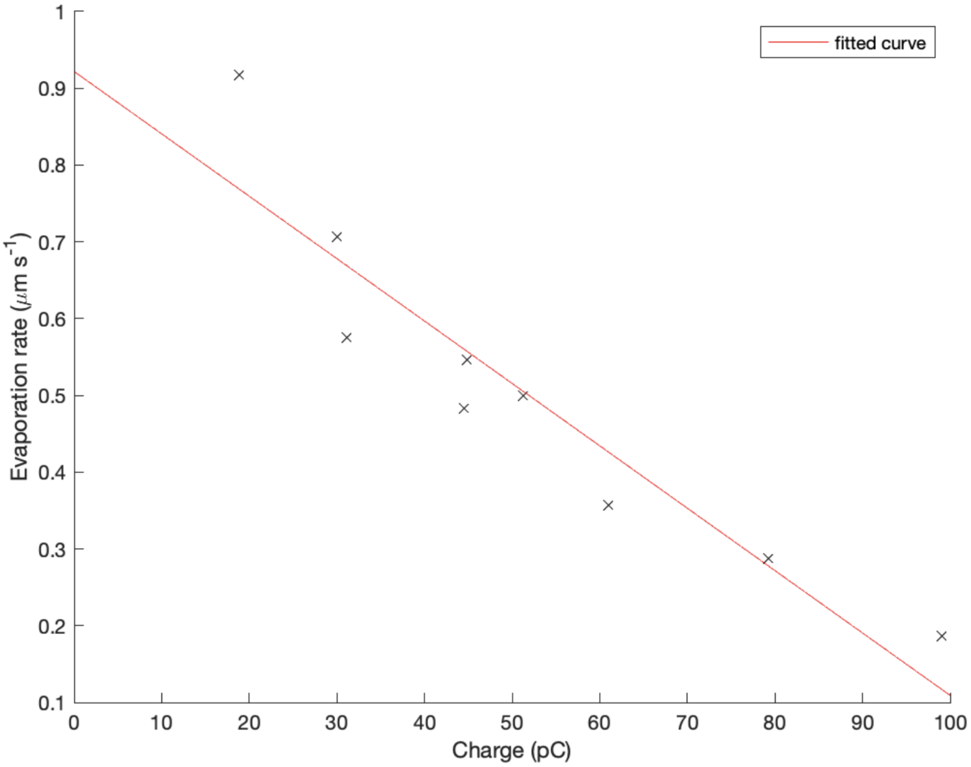

Figure 3. Droplet charge against evaporation rate for several drop evaporation experiments in which Rayleigh explosions occurred

These effects have been seen in the experimental data too. Figure 2 shows an experimental run where the droplet charge is sufficient to cause Rayleigh explosions as observed in the model results. It is possible therefore to determine the charge when explosions occur, as for an event occurring at a known droplet size the relationship in equation 1 can be used

$$X = Q^2/(64\pi^2\epsilon_0\sigma a_0^3) \tag{1}$$

where X = fissility (which, when X = 1 an explosion occurs), Q = charge, σ = surface tension, and a_0 = droplet radius (Duft et al., 2003). Using these data it is possible to observe the evaporation rate for a given droplet charge to investigate the relationship between these properties. Theory predicts that charge will stabilise droplets against evaporation, extending their lifetime. Given these experimental data it can be tested whether this is the case. Evaporation rate is plotted against charge for several droplets in Figure 3. The resulting relationship can be seen as increasing charge resulting in slower evaporation. However, high charge is known to result in increased incidence of Rayleigh explosions.

3.5. Droplet-droplet interactions

The electrical characteristics of droplets affect how they interact with one another in a number of ways. These include dynamical interactions as droplets move past each other and static effects as droplets interact due to the proximity of pairs of droplets. All droplets exhibit a certain amount of charge at the surface and within the droplet itself meaning that they are rarely truly neutral, and the effects of charge must be considered important in all aspects of aerosol science.

4. Conclusions

In this brief communication, a number of factors affecting droplet lifetime have been assessed and it can be seen that charge is a very important factor among them. Charge acts to stabilise droplets against evaporation, reducing the evaporation rate as has been observed both in model results and experimental observations. By comparing models and observations, one can be certain, when these agree, that scientific conclusions can be drawn confidently. This work provides that certainty and allows certain conclusions to be drawn with a degree of confidence.

It is apparent that charge does indeed act to stablilise a droplet against evaporation with, the higher the charge, the slower the evaporation rate. This effect however, is countered by the fact that highly charged droplets periodically experience Rayleigh explosions. This acts to make higher charge *reduce* the droplet lifetime. This combined effect of these two competing effects results in the key conclusion of this work that although charge increases a droplet's lifetime, the occurrence of Rayleigh explosions far outweighs this effect in order to reduce the overall droplet lifetime.

Acknowledgements

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