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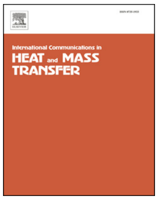

Experimental investigation of the evaporation rate of supercooled water droplets at constant temperature and varying relative humidity☆

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ABSTRACT

Supercooled water droplets are found in clouds at high altitude. They are exposed to very low temperatures and high relative humidity. The phase change of supercooled water droplets is an interesting heat and mass transfer problem. It is of paramount interest to understand droplet dynamics in clouds and hence, rain, snow and hail generating mechanisms. Therefore, in this work freely suspended supercooled water droplets are investigated experimentally. We present the evaporation rate at a constant temperature of 268.15 K and six different relative humidities (28 % − 89 %). It is found, that the evaporation rate is linear dependent on the relative humidity.

1. Introduction

Water droplets in clouds at high altitude are exposed to very low temperatures and high relative humidity as noted by Pruppacher and Klett [1]. Despite subzero temperatures, the droplets can still be liquid and are then called supercooled. The phase change of supercooled water droplets (SWDs) and the interaction between liquid and already frozen droplets is of paramount interest to understand rain, snow and hail generating mechanisms. For this purpose, freely suspended SWDs are investigated experimentally. This has been done by using a levitation technique, where the droplet is trapped in a test chamber. In literature different levitation techniques can be found. SWDs at different subzero temperatures were investigated by Roth et al. [2] with optical levitation and recently by Tong et al. [3] in an electrodynamic trap. The latter present results of evaporating droplets in mostly dry ambient. In this work we present optically levitated single water droplets. The optical levitation is a stable trap first investigated by Ashkin [4]. During the levitation the scattered light, also known as Mie scattering, in the forward hemisphere of SWDs is observed. A benefit of the optical levitation is that no further laser needs to illuminate the droplet. With this non-intrusive measurement technique, described by Roth et al. [2] and Wilms [5] the droplet diameter and the evaporation rate can be derived. The investigated size of SWDs in our study is around 50 μm and is in the magnitude of droplet diameters appearing in clouds mentioned by Pruppacher and Klett [1]. In the experiments carried out in this work, the influence of the relative humidity on the evaporation of SWDs is systematically investigated. To the best of the authors' knowledge, the only experimental observation of evaporating SWDs with known relative humidity is given by Tong et al. [3]. However, their experiments were done in dry ambient and at different subzero temperatures and so, the experimental results cannot be compared directly. Therefore, our paper is focused on the effect of humidity on the evaporation of SWD.

2. Material and methods

This chapter begins with a description of the experimental setup for trapping SWDs, as well as the measurement setup. It is followed by a section describing the evaluation methods and ends with a brief comment on how to calculate the evaporation rate.

2.1. Experimental setup

A schematic of the experimental setup is shown in Fig. 1. The observation chamber, with its inner dimensions $9 \times 14\ \mathrm{mm}^2$, is cooled down with a cryostat (*Huber, Unistat 815w*). A controlled flow (*Bronkhorst, El-Flow-MFC*) of almost dry nitrogen (*Air Liquid, 99.999 %*), provided by a pressurised gas cylinder, passes from the top to the bottom of the chamber. It is important to have an aerosol-free flow, because otherwise freezing could be triggered. The humidity of the nitrogen flow can be adjusted with an arrangement of precise mass flow meters and an evaporator (*Bronkhorst, CEM*). The flow rate in the presented work is about $\dot{V} = 40\ \mathrm{ml_n/min}$, leading to a velocity of $u = 0.0055$ m/s in the chamber. This results in very low Reynolds numbers in the chamber of $Re_D = 0.02$. This flow is only needed in order to guarantee that the chamber conditions are always similar and evaporated mass is transferred out of the chamber, preventing saturated chamber conditions. A single water droplet is generated with an in-house build droplet-on-demand generator [6]. The droplets in this study have an initial

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Nomenclature

B_Y	Spalding's mass transfer number, $B_Y = \frac{Y_{1,\infty} - Y_{1,s}}{Y_{1s} - 1}$ [–]
D, D_0	Diameter [m]
c_p	Specific heat capacity [J/(kg K)]
$D_{12,R}$	Diffusion coefficient at reference temperature [m²/s]
f	Focal length [m]
n	Real part of refractive index [–]
Nu	Nusselt number, $Nu = \frac{\alpha\, D}{k}$ [–]
M_1, M_2	Molar mass of droplet liquid and ambient gas [mol/kg]
p	Ambient pressure [Pa]
$p_{1,s}, p_{1,\infty}$	Pressure at droplet surface and far away from droplet [Pa]
p_v	Saturation vapour pressure [Pa]
Re	Reynolds number, $Re = \frac{\rho\, u\, D}{\eta}$ [–]
Sh	Sherwood number, Sh $= \frac{\beta\, D}{D_{12}}$ [–]
t	Time [s]
T	Temperature [K]
T_W, T_∞	Wet-bulb temperature and ambient temperature [K]
T_R, T_s	Reference temperature and temperature at droplet surface [K]
u	Velocity [m/s]
$\dot{V}$	Flow rate at standard conditions $T = 268.15$ K, $p = 1013.25$ hPa [l_n/min]
$X_{1,s}, X_{1,\infty}$	Molar fraction at droplet surface and far away from droplet [–]
$Y_{1,s}, Y_{1,\infty}$	Mass fraction at droplet surface and far away from droplet [–]
Greek symbols	
β	Evaporation rate [m²/s]
Δ	Accuracy, range [–]
$\Delta\theta$	Angular fringe distance [°]
θ	Mounting angle of camera [°]
λ	Laser wavelength [m]
ρ_R, ρ_l	Density gas vapour mixture and density of liquid droplet [kg/m³]
φ	Ambient relative humidity, $\varphi = \frac{p_{H_2O}}{p_v} \cdot 100\ \%$ [%]

diameter $D_0 = 50$ μm ± 6 μm. To avoid freezing of the droplets due to nucleation by impurities, water for laboratory analysis (*Merck LiChrosolv®, spec. conductance* ≤1 μS/cm) is used. With a *Nd:YAG* laser (*Laser Quantum, OPUS 532 nm, 5 W*), which is directed through the chamber, single droplets are optically levitated. Therefore, a lens with a focal length of $f = 0.1$ m, which can be driven in height (*PI, Linear Positioning Stages*), focusses the laser. Thus, the droplet can be positioned within the chamber by moving the focusing lens. It is important, that the light is not absorbed by the investigated substance. So the corresponding absorption coefficient of water for the chosen laser wavelength is low [7]. In fact, it is almost the minimum in the absorption spectrum of water. The droplet should not heat up notably when considering additionally the small size. In order to get a droplet levitated it has to fall along the centre of the laser beam. Therefore, two precise motion stages (*PI, linear positioning stages*) adjust the position of the droplet generator. Typically, the laser power in the presented experiments is about 2.4 W ± 0.2 W. To avoid a heating of the observation chamber, it is necessary to cool down the laser trap. For the investigations, the ambient conditions in the chamber are controlled as follows. A thermocouple (*Omega, Type K, unsheathed, diameter* 0.125 mm, *measurement accuracy* $\Delta T \pm 0.1$ K) is driven into the chamber to measure the temperature along the height. A U-shaped temperature profile develops, which has a constant temperature region with a length of about 10 mm. Here, a SWD is trapped and its position is monitored by a Position Sensitive Device (*PSD*). A humidity sensor (*Sensirion SHT-71,* $\Delta\varphi \pm 3$ %, $\Delta T \pm 0.5$ K) in the observation chamber controls the temperature and relative humidity φ at the entry. In addition, the relative humidity is measured at the levitation position. Therefore, a second equal humidity sensor is driven into the chamber before and at the end of a measurement series. In doing so, it can be assured that the ambient conditions are stable during the experiment. While the droplet is levitated, it scatters the laser light. As shown in Fig. 1 the Mie scattering is recorded by a CCD line camera (*E2V, AVIIVA SM2, 2048 pixels*) in the forward hemisphere under an angle of observation of $\theta = 60°$ in a range of ± 10°. Therefore, the light is collected by a cylindrical lens ($f = 0.08$ m), which is adjusted right in the focal plane of the camera in order to provide a Fourier transformation. This means, rays scattered under a certain angle will nearly be independent of the position of the droplet in the chamber and hence always illuminate the same camera pixel. Then, with a previous made calibration, the obtained recordings of intensity over pixel are transformed to intensity over scattering angle. According to Fig. 1, the camera is connected to a frame grabber card (*Matrox, Helios CL*) and triggered externally (*LabSmith, LC880*) with 5000 Hz. Once a droplet reaches the observation position, an oscilloscope, which samples the PSD, starts the recording. Thereby, a defined, arbitrary delay of 150 ms is introduced, so that the drop is trapped stable and cooled down to ambient temperature. A current recording contains 60,000 lines, which corresponds to a measurement duration of about 12 s. Besides the Mie scattering, also the droplet position is saved. This is necessary for the evaluation, to ensure that the droplet is exposed to a constant temperature.

2.2. Evaluation methods

The following section describes the evaluation of the scattered light in the forward hemisphere. This method for droplet sizing is of advantage, as it is non-intrusive. In the forward hemisphere the scattered light has a regular pattern, consisting of bright and dark fringes as shown in Fig. 2. According to Glantschnig and Chen [8], this fringe spacing is mainly dependent on the droplet diameter, while being less dependent on the refractive index. Hence, based on geometrical optics they derived a relation for the diameter

$$D = \frac{2\,\lambda}{\Delta\theta} \left(\cos(\theta/2) + n \sin(\theta/2) \cdot \left[1 + n^2 - 2\, n \cos(\theta/2)\right]^{-1/2} \right)^{-1}. \quad (1)$$

Here the variables are the mounting angle of the camera θ, the laser wavelength λ, the angular fringe distance $\Delta\theta$ and finally the refractive index n. Duft and Leisner [9] measured the refractive index for a wide range of supercooled temperatures. According to their data the refractive index at $T = 268.15$ K is $n = 1.333$. Note, that only the real part of the refractive index is considered, as absorbance is neglected. Next, the implemented evaluation algorithm is briefly explained. To remind, the saved data is a matrix with the size 2048 (pixel) and 60,000 (amount of lines). Each single line contains the intensity distribution over the observed range of scattering angle. At first, every line is filtered and the angular distance $\Delta\theta$ between two neighbouring maxima is determined. As there are several maxima, an average angular distance $\Delta\theta$ is calculated. Then, Eq. (1) is applied to calculate the droplet diameter. It should be pointed out, that the time, at which the line is grabbed, is well known, because the camera is triggered externally. The accuracy of the diameter for the current evaluation is ± 2 % in the range 20 μm < D < 60 μm. For D < 20 μm the accuracy starts to decrease. This is due to less bright maxima and therefore increasing noise. Furthermore, the amount of maxima in the observed angular range decreases, leading to more uncertainties in the angular distance $\Delta\theta$. Fig. 3 shows the diameter evolution over time, having apparently a parabolic trend. It can be noted that the diameter oscillates. This oscillating of the diameter is not physical, but is related to the appearance of morphology