Laboratory Studies of Scattering Properties of Polluted Cloud Droplets: Implications for FSSP Measurements

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ABSTRACT

Laboratory experiments were conducted in the Mainz vertical wind tunnel to study the effects of pollutants dissolved or suspended in cloud droplets on the droplet size measurements of a Forward Scattering Spectrometer Probe (FSSP). The FSSP is a widely used instrument to derive microphysical properties of atmospheric clouds. Individual droplets of different well-defined sizes were freely falling at their terminal velocities in the wind tunnel while the intensity of radiation emitted by the He–Ne laser of the FSSP and scattered by the droplets was measured. For this purpose, the FSSP was adapted and mounted to the wind tunnel. The intensity of radiation scattered by the droplets in the FSSP measurement is principally used to derive the droplet size. The droplets contained soluble ammonium sulfate or suspended absorbent graphite particles as pollutants in concentrations that were higher than usually found in atmospheric cloud droplets. The results of the measurements and corresponding simulations indicate that for high pollutant concentrations, the scattered laser radiation detected by the FSSP depends significantly on the refractive index of the droplet (i.e., on the concentration of soluble or insoluble pollutants). However, for the lower pollutant concentrations usually observed in atmospheric cloud droplets, the need for correcting the droplet sizes measured with the FSSP for the effects of the pollutions can be avoided.

1. Introduction

Clouds in general and their radiative properties in particular, substantially affect the earth's radiative balance and, therefore, the global climate (Kiehl and Trenberth 1997). The radiative implications of clouds are strongly determined by their microphysical properties such as the droplet effective size and liquid water content. These quantities can either be retrieved from indirect, remote sensing techniques (e.g., radar or microwave radiometers; see, e.g., Crewell et al. 2004) or measured in situ with optical methods. A very common in situ instrument for measuring cloud droplet microphysical properties is the Forward Scattering Spectrometer Probe (FSSP; Knollenberg 1976, 1981; Dye and Baumgardner 1984), modified by Brenguier et al. (1998) and Schmidt et al. (2004).

The effects of soluble and insoluble pollutants on cloud radiative properties are described by a number of model investigations (e.g., Grassl 1975; Twomey 1977; Chylek et al. 1984; Kondratyev and Binenko 1987; Heintzenberg and Wendisch 1996). However, significant effects of pollutants on cloud radiative properties could not be verified experimentally (e.g., Twohy et al. 1989). One possible explanation for this missing experimental confirmation is presumably the fact that the model simulations mostly assume a homogeneous mixture between pure water and the pollutant, which may lead to an underestimation of the pollution effects when insoluble components are present in the droplets. Another hypothesis is that the droplet size measurements in heavily polluted clouds are biased because of the neglect of effects of the pollutants on the size mea-

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surements of the FSSP. This instrument uses the scattering intensity in the near forward scattering range to derive the droplet size. In the data analysis, pure water droplets are assumed. Thus far it is unclear if pollution of the water droplets impacts the droplet size measurements with the FSSP. Experimental investigations considering both soluble and insoluble pollutants in a droplet were carried out by Chylek and Hallett (1992). These authors generated droplets coated with soot layers; however, the droplet sizes were much larger than typical cloud droplet diameters. In contrast, the present paper deals with cloud droplets around 30 μm in diameter containing pollutants in various concentrations. The scattering properties of the droplets were measured with a modified FSSP while they were freely falling in a vertical wind tunnel; in this way, the optical effects of the pollutants on the measuring characteristics of the widely used FSSP are determined.

2. Experimental methods and Mie simulations

The laboratory experiments were carried out in the Mainz vertical wind tunnel (Pruppacher 1988; Vohl 1989). This tunnel allows investigations of freely floating droplets in the size range from micrometers to millimeters. The droplet sizes are determined from their terminal velocities. The airflow remains laminar over the entire range of wind speeds required for suspending the droplets. The droplets were produced by means of a commercial droplet generator (manufactured by Microdrop, Norderstedt, Germany). The nozzle used for the present experiments generated droplets around 30 μm in diameter, which is well in the range of atmospheric cloud droplets (Pruppacher and Klett 1997). With an FSSP-100 (manufactured by Particle Measuring Systems, Inc., Boulder, CO), the intensity of the laser radiation emitted by the He–Ne laser of the FSSP and scattered by the droplets in the near-forward direction was measured while the droplets were freely suspended with terminal velocity in the wind tunnel. For details of the FSSP calibration, the reader is referred to Wendisch et al. (1996). To mechanically adjust the FSSP to the wind tunnel, a segment was designed so that the droplets moved within the measuring volume of the FSSP.

The intensity of the forward scattered laser radiation was derived from the electrical analog pulse height, which was directly obtained from the two preamplifiers of the FSSP photodetectors. The pulse signals were observed with a digital storage oscilloscope and recorded. The detector signals were evaluated by a Gaussian fit function to determine the size of the droplets passing through the laser beam. The droplets were observed and recorded using a video film camera. The droplet generator pulse was used to synchronize a stroboscope with a light-emitting diode to allow the observation of the droplets. The recordings show the picture of the droplets together with the glares that result from reflection and refraction of the FSSP laser beam at the air–water interface (see Fig. 1). The position of the refracted beam depends on the refractive index of the solution (Wendisch et al. 1996). The video camera was connected to a video capture card that digitized the pictures in real time and recorded them on a computer. The droplets were viewed simultaneously on a monitor. From the droplet pictures, the horizontal diameter of the droplets (d-true) and the distance between the two glares (d-glare) were determined.

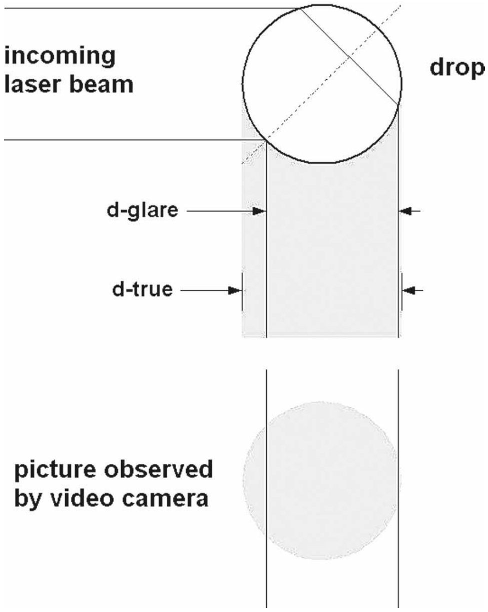

FIG. 1. Recording of droplet pictures.

In one series of experiments, the droplets contained ammonium sulfate in various concentrations up to 100 g L^{-1} as a soluble pollutant. These concentrations are orders of magnitudes larger than those found in atmospheric cloud droplets, which are less than 1 g L^{-1} (Warneck 1988: 6–25 mg L^{-1}; Pruppacher and Klett 1997: 2–330 mg L^{-1}). The high sulfate concentrations in the experiments were used to induce maximum effects. If no serious impact could be observed for these unrealistically high ammonium sulfate concentrations, then the pollution of atmospheric cloud droplets by ammonium sulfate would not needed to be considered in FSSP data analysis. In a second series of experiments the droplets contained suspended absorbing particles. A commercially available suspension (Aquadac, from Acheson Industries) was used; it contained approxi-

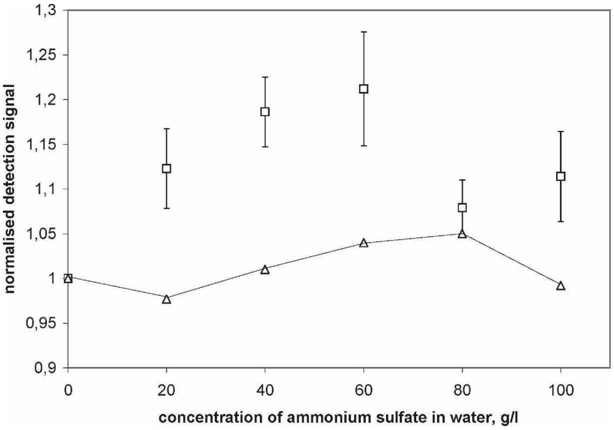

FIG. 2. Detection signal as a function of ammonium sulfate concentration: experimental data in comparison to Mie calculations normalized to the value of pure water with a droplet diameter of 22.8 μm. The solid line with triangles represents simulations; squares with error bars show measurements.

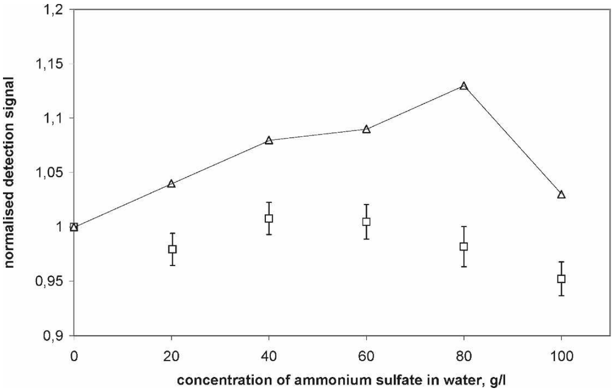

FIG. 3. As in Fig. 2, but for a droplet diameter of 30.4 μm.

mately 18% of colloidal graphite. In fact, the graphite particles are both absorbing and scattering particles: both effects were considered in the experiments. The size of the graphite particles was up to 5 μm, with a modal diameter around 1.5 μm. Suspensions with concentrations of 900 and 1800 mg L^{-1} graphite were produced. This is again significantly higher than the concentrations found in atmospheric cloud droplets (e.g., 0.3 to 3.5 mg L^{-1}; Hitzenberger et al. 2001); however, these concentrations were examined for distinct effects.

The simulations of the FSSP signal were performed with a Mie program in an accelerated iteration scheme according to Seltman (1985). In these calculations, the spectral (i.e., at the wavelength of the He-Ne laser beam of the FSSP) radiant intensities scattered in different directions by a spherical droplet were simulated. They were integrated over a scattering angle range between 3° and 13°, which is relevant for FSSP measurements. These Mie simulations were performed for pure and polluted droplets by modifying the refractive indices of the droplets.

3. Results

a. Ammonium sulfate

Results from the experiments using ammonium sulfate as droplet pollution as well as those from Mie calculations are given in Figs. 2 and 3 for two different droplet diameters (22.8 and 30.4 μm). Both figures show the variation of the normalized detection signal from the FSSP (i.e., the detection signal divided by the signal for pure water, either measured or simulated by Mie theory) as a function of the ammonium sulfate concentration in the droplet. The solid lines with triangles show the results of the simulations; squares with error bars indicate the measurements. The experimental values are based on approximately 20 individual measurements; the error bars reflect the standard deviation of the data. For droplets with a diameter of 22.8 μm, Fig. 2 indicates that the measured relative FSSP detection signal increases for increasing solute concentrations up to 60 g L^{-1} by roughly 20%. With further increasing solute concentrations above 60 g L^{-1}, the measured relative FSSP signal mostly decreases. For the larger droplets with a diameter of 30.4 μm (Fig. 3), the measured changes of the normalized detection signal are smaller (clearly less than ±5%) compared to the 22.8 μm droplets. However, the data show a similar pattern, with a tendency to increase until 60 g L^{-1} solute concentration and decrease with further increasing concentrations. This nonmonotonic behavior is a clear experimental verification of the nonlinear ripple structure of the theoretical Mie scattering efficiency curve. Typically, one would expect a gradual increase of the relative FSSP scattering signal with increasing solute concentration because the real part of the refractive index (which mainly determines the scattering intensity of droplets) increases for higher ammonium sulfate concentrations. Instead, the normalized scattering signal does not monotonically increase with solute concentration.

The Mie computations (solid lines with triangles in Figs. 2 and 3) show deviations from the measured normalized FSSP scattering signal that are outside of the measurement standard deviation (error bars). This is explained by considering that in the FSSP instrument, the intensity of the scattered radiation measured by the detector also depends on the exact position of the droplet inside the optical sampling volume of the FSSP. This may effectively smooth variations that are obvious in the simulated FSSP detector signal for different solute concentrations when the signal from many droplets is accumulated. In other words, in a "real instrument simulation," the expected variations will be of lower magnitude.

It can be concluded from Figs. 2 and 3 that the scattering signal measured by the FSSP may significantly depend on solute concentration for very high ammonium sulfate concentrations in droplets, especially for smaller droplets (22.8 μm). Thus, a correction of the FSSP signal as a function of solute concentration might be appropriate in particular cases. However, a closer look into the maximum observed deviations of the relative scattered signal strength (compared to the pure water droplet case) reveals that the maximum deviation translated into droplet size variation leads to 2.8 μm for a droplet size of 22.8 μm and 2.5 μm for a droplet size of 30.4 μm (the measurement errors, similarly translated into droplet sizes, give about ± 0.5 μm). Because the width of the most useful FSSP channel is 3 μm, the scattering of solution droplets could shift the FSSP signal by at most one channel. This size channel shift would occur only for extremely high solute concentrations, namely 40 g L^{-1} and 60 g L^{-1}, respectively. Therefore, for realistic ammonium sulfate concentrations in cloud droplets (e.g., less than 1 g L^{-1}; Pruppacher and Klett 1997), the possible shift of size channels in FSSP measurements due to ammonium sulfate contamination can safely be neglected. This extrapolation to smaller, more realistic ammonium sulfate concentrations in cloud droplets is justified by Mie calculations additionally carried out for the two investigated drop sizes with small solute concentrations down to 0.01 g L^{-1} (atmospheric level concentrations). These results indicate that the strength of the scattering signal for solute concentrations of less than 10 g L^{-1} gradually decreases for decreasing solute concentrations, and thus does not show strong fluctuations for smaller concentrations.

b. Suspended absorbing particles

Figure 4 shows the measured relative FSSP signals for droplets of 30.4-μm diameter containing suspended absorbing particles. The variation of the normalized detection signal from the FSSP (i.e., the detection signal divided by the signal for pure water) is given as a function of the graphite concentration in the droplets. Each data point was based on approximately 20 individual measurements; the error bars reflect the standard deviation of the data. The measured relative FSSP detector signal continuously decreases with increasing concentrations of suspending particles in the droplets. This is indeed not an unexpected result because Mie absorption efficiency does not show fluctuations but changes continuously. The maximum deviation of scattering translated into droplet size variation leads to 2.5 μm for the measured droplet size of 30 μm for high graphite concentrations (i.e., 1800 mg L^{-1}). Because observed black carbon concentrations in droplets are much lower (less than 10 mg L^{-1}; Hitzenberger et al. 2001), it is obvious that these deviations are negligible. Thus, a correction of FSSP measurements is not necessary even for unrealistically high concentrations of absorbing material suspended in the droplets.

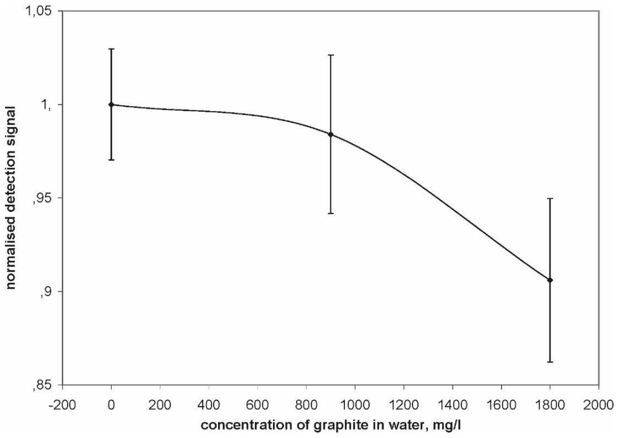

FIG. 4. Detection signal as a function of graphite concentration in droplets of 30.4-μm diameter. Experimental data are normalized to the value of pure water; measurements are shown with error bars.

4. Conclusions

The presented results yield solid experimental evidence that for high pollution concentrations, the FSSP detection signal significantly depends on the refractive index of the droplet—that is, on the respective concentrations of soluble and insoluble pollutants. However, for common atmospheric concentrations of soluble or insoluble material in droplets, a correction of the droplet size distributions measured with an FSSP can be neglected, at least for droplets in the investigated size range of 20–30 μm. An extrapolation of these results to larger and smaller droplet sizes could not be carried out because the scattered signal is widely fluctuating and does not show a well-defined tendency. Even for the investigated two droplet sizes, the theoretically determined signal shows the maximum value for different solute concentrations. An extrapolation also would require a complete model simulation of the FSSP instrument. Such a modeling is out of the scope of the present short note.

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