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Measurements of the size-dependence of solute concentrations in cloud droplets

By J. A. OGREN, J. HEINTZENBERG, A. ZUBER, K. J. NOONE, *Department of Meteorology*[1], *University of Stockholm, S-106 91 Stockholm, Sweden* and R. J. CHARLSON, *Department of Atmospheric Sciences AK-40, University of Washington, Seattle, Washington 98195, USA*

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ABSTRACT

Measurements of the particulate material and water vapor released when cloud droplets evaporate are used to determine the amount and concentration of material contained in the droplets. Droplets are sampled with a counterflow virtual impactor (CVI), which extracts them from the surrounding air and evaporates them in a flow of warm, dry, filtered air. The resulting concentrations of water vapor, particle number, and particulate light scattering coefficient are determined, allowing calculation of average droplet and particle radii, as well as the aqueous-phase concentration of non-volatile, soluble and insoluble material ("solute concentration"). By changing the internal flows of the CVI, the minimum radius of droplets sampled is varied within the interval 4–15 μm. Measurements obtained in central Sweden in the summer of 1986, near the base of stratocumulus clouds, show that solute concentrations increase with droplet size by roughly a factor of 3 over the radius range 5–9 μm. This observation is consistent with the hypothesis that condensational growth controls the droplet size distribution near cloud base.

1. Introduction

It has been observed by Noone et al. (1988a) that the solute composition and concentration in marine cloud droplets varies with droplet size. In a companion paper, Twohy et al. (1989) describe a simple model of the initial stages of condensational growth of cloud droplets, which results in the larger droplets nucleating on the larger aerosol particles. Because the smaller droplets have a larger surface area to volume ratio, they are calculated to increase in radius and become dilute more quickly than the larger droplets. In turn, this model predicts that the systematic difference of the chemical composition of the cloud condensation nuclei (CCN) with size will result in the sulfate-dominated particles growing into smaller cloud droplets and the supermicrometer, e.g. sea salt and soil dust, particles becoming the larger droplets.

Coalescence of the cloud droplets should tend to mix the composition of the large end of the droplet size spectrum while leaving the small end unaffected. The theory of inhomogeneous mixing proposed by Telford and Chai (1980) in the extreme case should totally randomize the composition of droplets with size. A combination of mixing and coalescence has been predicted to yield the highest solute concentrations in the smallest droplets (Flossman et al., 1985). Hence, more observations of the actual dependence as given by Noone et al. (1988a) should help to elucidate which processes dominate the different stages of cloud evolution.

Besides allowing an improved understanding of the fundamental physical processes involved in cloud formation and droplet dilution, there are chemical reasons for wanting to know the drop-size dependence of cloud-water composition. Chemical reactions in cloud droplets depend on the aqueous concentrations of the reactants, and sulfur dioxide solubility in cloud water is pH-

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dependent. If solute concentration or composition leads to a size dependence of acidity, oxidation state IV sulfur will not be uniformly distributed with size (Hegg and Hobbs, 1979). Deposition from the atmosphere by rain or by direct impaction may not involve the smallest droplets, such that the composition of the largest droplets alone may account for observed fluxes to land biota and materials.

Many models of the liquid phase chemical reactions in clouds assume that all the droplets have the same composition (e.g., Jacob and Hoffman, 1983; Chameides, 1984; Tremblay and Leighton, 1986; Hough, 1987). While this assumption may simplify the required computations, evidence is needed to establish its validity. If the assumption is not always valid, it would be useful to know how sensitive the model predictions of e.g., reaction rates, are to differences in the chemical composition of the individual droplets.

Thus we have refined and extended the measurements of Noone et al. (1988a) to a continental site and included a longer series of observations. We will conclude that the size dependence of composition in the stratocumulus clouds studied is as discovered earlier in a marine setting, i.e., that the largest droplets have the highest solute concentrations, and therefore must have nucleated on the largest cloud condensation nuclei.

2. Experimental approach

A detailed description of the sampling site, along with a summary of the ionic composition of bulk cloud-water sampled there, is given in Ogren and Rodhe (1986). Briefly, the site is located above the timberline of the Swedish peak Åreskutan (63°26′N, 13°6′E) at an elevation of 1250 m asl, and there are no significant local sources of pollution. An electrically powered aerial tramway serves the site, and allows regular observations of the height of cloud base. The clouds encountered at the sampling elevation are generally widespread, indicating that their formation is due to lifting (frontal or convective) and boundary layer mixing, rather than orographic forcing. Moderate levels of air pollution, as reflected in the bulk cloud-water composition, are encountered in air arriving from the southerly sector, while extremely clean air, comparable to that found in the Arctic arrives at the site from the northwest (Ogren and Rodhe, 1986).

The results described here were obtained during a field campaign in July and August of 1986. During this period, measurements were made of the chemical and physical properties of evaporated cloud droplets. To aid interpretation of these measurements, temperature, pressure, wind speed and direction, and submicrometer aerosol particle number and light scattering coefficient were determined continuously, while cloud base, cloud type and amount of coverage, wet bulb temperature, and visibility were determined on an hourly basis (Johansson, 1987). Bulk cloud-water was collected roughly every half hour, using the same sampler and analytical methodology as described in Ogren and Rodhe (1986). The average, bulk cloud-water composition for the period 22 July to 8 August 1986, corresponding to the period for which results are presented below, is shown in Table 1. The average composition is that which would result if the CCN were mainly ammonium bisulfate, with smaller amounts of nitric acid and sea salt added. Airmass trajectories for this period were predominantly from the south and southwest (Johansson, 1987), and the high solute concentrations are consistent with previous samples collected under similar conditions (Ogren and Rodhe, 1986).

Table 1. *Average bulk cloud-water composition (μeq/l) during the sampling period (22 July to 8 August 1986)*

H^+	188	Cl^-	11
NH_4^+	151	NO_3^-	61
Na^+	17	SO_4^{2-}	264
K^+	3		
Mg^{2+}	5		
Ca^{2+}	8		

Cloud droplets were sampled from a vertical wind tunnel, which accelerated the air to ca. 100 m s^{-1} (Noone et al., 1988b; Ogren et al., 1986). Different speeds were used in some cases, and the actual speed was determined by measuring the static pressure in the cylindrical test section of the wind tunnel. The droplets were

separated from the surrounding air into a quiescent flow of warm, dry, particle-free air with a counterflow virtual impactor (CVI) (Ogren et al., 1985, 1986; Noone et al., 1988a, b). By adjusting the internal flows in the CVI, the radius of the droplets that were sampled with 50% efficiency (R_{50}) could be adjusted within the range 4–15 μm. The R_{50} values are based on a Stoke's law analysis of the flow around and into the CVI, assuming that curvature of streamlines past the tip of the CVI adds 1 cm to the effective stop distance of the droplets (Noone et al., 1987b). The size selectivity of the CVI is fairly broad (Noone et al., 1988a). For example, with the internal flows set to give an R_{50} of 13 μm, the radii for 10 and 90% sampling efficiency were 9 and 16 μm, respectively.

Droplets sampled by the CVI quickly evaporated in the internal air stream (a 10 μm radius droplet typically evaporates in 0.1–0.4 s, depending on the droplet temperature), yielding an aerosol which was analyzed for its water vapor content, particle number concentration, and particulate light scattering coefficient. Water vapor concentration was determined using a Lyman-alpha hygrometer, which measured the extinction of 121 nm radiation in the sample air stream relative to that in a dry reference cell (Zuber and Witt, 1987). This water vapor concentration yielded the liquid water content (L) of the sampled droplets. The number concentration of residual particles was determined with a condensation nucleus counter (Thermo Systems Inc. (TSI) model 3020). Assuming that evaporation of one cloud droplet produces exactly one residual particle, the number of cloud droplets (N) sampled by the CVI was calculated. We presently have no proof that one droplet produces one particle; experiments are planned to explore this point.

Light scattering coefficient (σ_{sp}) was determined with an integrating nephelometer similar to that described by Heintzenberg and Bäcklin (1983). The mass of particulate material was calculated from the light scattering coefficient using an assumed light scattering efficiency (β) of 4 m^2 g^{-1} (Waggoner et al., 1981). This mass corresponds to the mass of non-volatile material dissolved or suspended in the droplets. For convenience, this will be referred to as solute mass in the following discussion, although it must be realized that it includes insoluble particles contained within the cloud droplets (e.g., elemental carbon), and excludes volatile substances that return to the gas phase when the droplets evaporate (e.g., SO_2, NH_3, HNO_3, H_2O_2). The residual particles may or may not be in vapor pressure equilibrium with these gases, depending on the kinetic factors governing transfer to the gas phase from the evaporating droplets. We will assume that the solute mass is controlled mainly by non-volatile substances like SO_4^{2-}, consistent with the bulk cloud-water composition observed on Åreskutan (Ogren and Rodhe, 1986). Admittedly, some volatilization of HNO_3 probably occurs inside the CVI, causing a loss of up to 25% of the solute mass.

The value of the light scattering efficiency (β) depends on the size distribution of the particles, which may vary as different fractions of the cloud droplet population are selected with the CVI. Measurements of the size distribution of the residual aerosol particles in the radius range 0.04–0.6 μm were obtained with an optical particle counter (OPC, Particle Measuring Systems model LAS X-HS). The OPC was calibrated with monodisperse ammonium sulfate particles provided by a system consisting of an atomizer (TSI 3076), differential mobility classifier (TSI 3071), and condensation nucleus counter (TSI 3020). Many more determinations of solute mass concentration are available from the nephelometer observations, so the OPC results are only used in the present work to evaluate the sensitivity of the results to the assumption that β is constant.

From the three primary measurements (L, N, σ_{sp}), the solute mass (M), aqueous-phase solute mass concentration ($[M]$), mass–mean droplet radius (R_d), and mass–mean residual particle radius (R_p) can be calculated as follows:

$$M = \sigma_{sp}/\beta, \tag{1}$$

$$[M] = M/L, \tag{2}$$

$$R_d = \left(\frac{3L}{4\pi\rho_w N}\right)^{1/3}, \tag{3}$$

$$R_p = \left(\frac{3M}{4\pi\rho_p N}\right)^{1/3} \tag{4}$$

where ρ_w is the density of liquid water and ρ_p is the density of the particles (assumed to be

1.5 g cm^{-3}). The mass-mean residual particle radius corresponds to the mean size of the cloud condensation nucleus (CCN) associated with the droplets, assuming that the amount of non-volatile material produced by chemical reactions after the droplets formed is small compared to the mass of the CCN. This is expected to be the case at a clean, rural site such as Åreskutan (Hegg, 1983).

The dependence of both the primary and derived parameters on droplet size was determined by setting the CVI flows for a particular value of R_{50} ($R_{50,1}$) and obtaining an ~5-min average of L, N, and σ_{sp}. The flows were then readjusted for another size fraction ($R_{50,2}$) and another set of averages was obtained. The difference between the two averages corresponded to the parameter value for the droplets in the size interval $R_{50,1}$–$R_{50,2}$. In Figs. 2, 4, below, the droplet size reported for the differential measurements is the geometric mean of $R_{50,1}$ and $R_{50,2}$.

It must be kept in mind, however, that clouds are not homogeneous, and that major variations in cloud properties (e.g., liquid water content) can occur during the course of a measurement (Ogren et al., 1986). This is not a serious problem for analyses of a single size fraction (i.e., with a single R_{50} setting), as inhomogeneities in the cloud can be seen directly in the data. When analyzing sequential measurements made on different size fractions, it was impossible to determine with certainty whether any observed variations are due to a size-dependence of the measured parameters or to inhomogeneities in the cloud. This limitation of the method was addressed by repeating measurements at the lowest setting of R_{50} (~4 μm) several times during a sequence, thereby obtaining a measure of the variability of the measured parameters during the 30–60 minutes required to complete a sequence. This variability was often as large as a factor of two.

3. Results

As a test of the consistency of the results, solute mass concentrations determined with the CVI are compared with the composition of simultaneous bulk cloud-water samples in Fig. 1. For this comparison, the instantaneous values of $[M]$ determined from eq. (2) were averaged over the time during which the bulk cloud-water samples were collected (typically 3–5 min). Not all bulk cloud-water samples are included, as the CVI was not always operating when cloud-water samples were obtained. Only CVI measurements with R_{50} less than 5 μm are included in the averages, as the cut size for the bulk cloud-water collector is calculated to be ~5 μm for the wind speeds typically encountered on Åreskutan (Ogren and Rodhe, 1986).

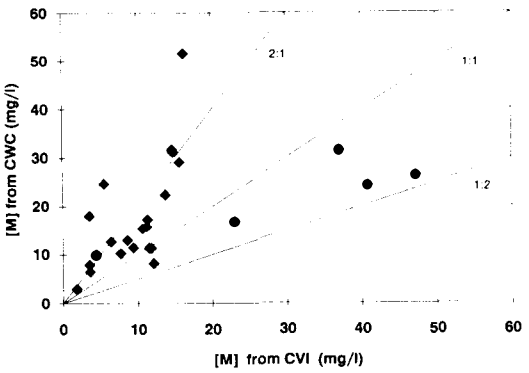

Fig. 1. Comparison of solute mass concentration (mg/l) determined from CVI and samples from bulk cloud-water collector (CWC).

The data presented in Fig. 1 show that the CVI provides meaningful measurements of cloud-water solute concentrations that are within a factor of two of those obtained with a conventional cloud-water sampler. Factors which could contribute to the observed differences include partial evaporation of droplets collected by the bulk cloud-water sampler, volatilization of nitric acid and ammonia from the evaporating droplets in the CVI, the possible presence of insoluble particles in the droplets, and differences in the collection characteristics of the CVI and the bulk cloud-water sampler.

More evidence indicating that the CVI performs as expected is shown in Fig. 2, where the calculated average droplet radius (eq. (3)) is plotted against the geometric mean of the two R_{50} values used to obtain each point. As each point in Fig. 2 represents a difference measurement, the caveat above regarding the inhomogeneity of clouds applies to this analysis. The droplet

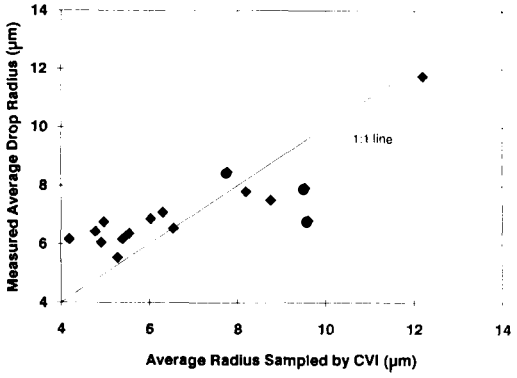

Fig. 2. Comparison of average measured droplet radius, determined from measurements of L and N (eq. (3)), with the average droplet radius sampled by the CVI.

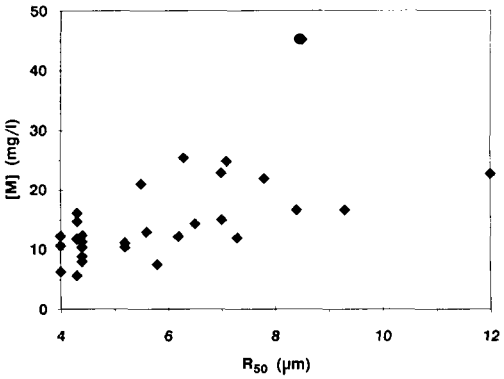

Fig. 3. Dependence of solute mass concentration on CVI cut size (R_{50}). Samples with N less than 1 cm^{-3} were excluded from the analysis.

sizes calculated from measurements of water vapor and particle number concentrations in the CVI are seen to be consistent with the sizes determined independently from the CVI sampling characteristics, although there is a tendency for the measured droplet size to be greater than the expected value for the smallest droplets.

The calculated, five minute average, aqueous-phase solute mass concentrations ($[M]$) are shown in Fig. 3 as a function of the droplet radius for which 50% of the droplets were sampled (R_{50}). To eliminate cases where very little material was sampled, only those cases are shown where the droplet number concentration was greater than 1 cm^{-3}. As small droplets are excluded from the sample (moving to the right in Fig. 3), solute concentrations are seen to increase markedly. That this systematic increase is observed over the four sampling days included in this analysis provides convincing evidence that the smaller droplets have lower solute concentrations, on the average, than the larger droplets. As each point in Fig. 3 was obtained with a single value of R_{50}, the effects of cloud inhomogeneity are not hidden within each point but rather are explicitly represented by the spread of the data.

The analysis of the dependence of solute mass concentration on droplet size can be carried one step further by evaluating the differences between sequential measurements made with different values of R_{50}. This amounts to calculating the difference between points in Fig. 3 corresponding to sequential measurements, so that the abscissa value becomes the geometric mean of the two cut sizes, and the ordinate value is the difference between two 5-min averages. Each line in Fig. 4 results from a sequence of at least three 5-min averages, which were obtained on two days. Being difference measurements, the effects of cloud inhomogeneity are implicit in each point. In spite of this source of variability, solute mass concentrations are seen to increase monotonically with droplet radius by a factor of ~ 3 over the range 5–9 μm. This size dependence of solute mass concentration implies that the largest droplets are associated with the largest CCN, and the calculated values of R_p (eq. (4)) show this to be the case.

For one of the sequences of measurements in Fig. 4 (marked with a bold line), OPC data are available to evaluate the sensitivity of the results to the assumption that the light scattering efficiency of the particles (β) is constant. An approximation to the Mie scattering efficiency curves (Heintzenberg and Baker, 1976) was used to calculate the scattering efficiency (β_{OPC}) from the measured size distributions, assuming particles with a refractive index of 1.54–0.05i and a density of 1.5 g m^{-3}. The results (Table 2) show that β_{OPC} decreased from 4.1 to 2.5 m^2 g^{-1}, due to an increase in the volume mean radius of the residual particles from 0.08 to 0.14 μm. Residual particles associated with the largest droplets (7–13 μm radius) are calculated to have a lower scattering efficiency than was assumed, resulting in even higher calculated solute concentrations in

Table 2. *Effect of changing particle size on solute concentrations calculated from nephelometer measurements*

$R_{50,1}$ (μm)	$R_{50,2}$ (μm)	β_{OPC} ($m^2\,g^{-1}$)	$[M]$ $\beta = 4.0$ (mg/l)	$[M]$ $\beta = \beta_{OPC}$ (mg/l)
4.4	5.6	4.1	6.1	6.0
5.6	7.1	3.4	9.6	11
7.1	13	2.5	24	37

the large droplets if the value of β derived from the OPC is used. Thus, it is likely that solute concentrations for all the sequences illustrated in Fig. 4 have an even stronger dependence on drop size than is evident from the figure.

4. Discussion

The results described above are qualitatively consistent with the results of models of the condensational growth of cloud droplets (Howell, 1949; Jensen and Charlson, 1984; Twohy et al., 1989; Seidl, 1989). However, the observed difference of a factor of three in solute mass concentrations over the droplet radius range of 5–9 μm is much lower than the factor of ~ 1000 calculated by the models. Differences in CCN composition, size distribution and updraft velocity, as well as turbulent mixing in the vertical direction, are likely contributors to this discrepancy, but the characteristics of the sampler are probably also a major cause. The results of Twohy et al. (1989) show that a calculated factor of 1000 difference between 4.3 and 8.4 μm radius droplets is reduced to a factor of 9.8 when the solute concentrations in the droplets in the radius interval 4.3–8.4 μm are integrated over two classes separated at 6.6 μm. The broad transmission curve of the CVI evens out the differences between adjacent size classes, which will reduce the difference between the calculated and observed results even more.

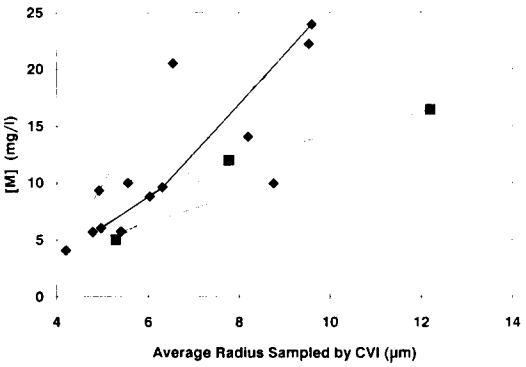

Fig. 4. Dependence of solute mass concentration on mean droplet size sampled by the CVI, as observed on 24 July (—■—) and 1 August (—◆—) 1986. The bold line marks the measurements described in Table 2.

The results presented here were obtained within 300 m of the bases of non-precipitating, stratocumulus clouds. Under these conditions, it is unlikely that coalescence is a major factor controlling droplet growth. Even under conditions where coalescence controls the droplet size distribution, the size-dependence of solute mass concentration in the radius range 5–9 μm is expected to show the same tendency as that reported here. Although coalescence removes droplets from this size interval and transfers their water and solute to larger droplets, it cannot change the composition of the small droplets which remain.

In contrast to coalescence, changes in the droplet size distribution caused by mixing processes are expected to alter the size-dependence of solute concentrations in the small droplets (Telford and Chai, 1980; Baker et al., 1984). Entrainment of subsaturated air into the cloud will cause some or all of the droplets to evaporate partially or completely, depending on the event. As the air continues to rise and cool, some of the evaporated droplets will reactivate and grow. It has been suggested that, in the extreme case, repeated cycles of activation/evaporation/reactivation could totally eradicate the size-dependence of solute concentration (Telford and Chai, 1980). This does not appear to be the case for the clouds studied in the present work, as solute concentration shows a clear dependence on the size of the droplets. A similar result was obtained in stratus clouds by Hudson and Rogers (1986), who found that the largest droplets were associated with the largest CCN. It should be noted, however, that 20–30 minutes were typically required with our technique to obtain one measurement of the size-dependence of solute mass concentrations, corresponding to spatial scales of ~ 10 km. It is possible that a different result

would be obtained if measurements could be obtained over smaller spatial scales.

Two major improvements in the CVI are warranted by the results of the present study. First, a reference system is needed to eliminate the uncertainties associated with inhomogeneities in the cloud. The reference system could be a CVI operated with a constant R_{50} value, while the other system could sequentially scan through a series of R_{50} values. In this fashion, instantaneous difference measurements would be obtained. The other major improvement could be to sharpen up the separation characteristics of the CVI and reduce the minimum size that the CVI can sample from ~ 4 μm to below 2 μm.

5. Conclusions

Within the lowest 300 m of the stratocumulus clouds studied, solute mass concentrations were observed to increase by a factor of three over the droplet radius interval 5–9 μm. This observation is consistent with the hypothesis that condensational growth of the cloud droplets controls solute concentrations near cloud base. The implications of this size-dependence of solute concentrations in cloud droplets need to be considered in models of cloud-water chemistry and deposition.

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