NOTES AND CORRESPONDENCE

Feasibility of Retrieving Cloud Condensation Nucleus Properties from Doppler Cloud Radar, Microwave Radiometer, and Lidar

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ABSTRACT

This paper explores the possibilities of using K_a-band Doppler radar, microwave radiometer, and lidar as a means of retrieving cloud condensation nucleus (CCN) properties in the stratocumulus-capped marine boundary layer. The retrieval is based on the intimate relationship between the cloud drop number concentration, the vertical air motion at cloud base, and the CCN activation spectrum parameters. The CCN properties that are sought are the C and k parameters in the $N = CS^k$ relationship, although activation spectra based on the lognormal distribution of particles is also straightforward. Cloud droplet concentration at cloud base is retrieved from a Doppler cloud radar combined with a microwave radiometer following a previously published technique. Cloud base is determined from a lidar or ceilometer. Vertical velocity just above cloud base is determined from the vertically pointing Doppler cloud radar. By combining the simultaneous retrievals of drop number and vertical velocity, and assuming theoretical relationships between these parameters and the subcloud aerosol parameters, the C parameter can be derived, under the assumption of a fixed k. If a calibrated backscatter lidar measurement is available, retrieval of both C and k parameters is possible. The retrieval is demonstrated for a dataset acquired during the Atlantic Stratocumulus Transition Experiment using a least squares minimization technique. Sensitivity to assumptions used in the retrieval is investigated. It is suggested that this technique may afford the acquisition of long-term datasets for climate monitoring purposes. Further investigation with focused experiments designed to address the issue more rigorously is required.

1. Introduction

Nearly four decades ago, Twomey (1959) explored the relationship between cloud condensation nucleus (CCN) parameters, the production of supersaturation due to adiabatic expansion in a vertically moving parcel, and the number of droplets activated from the CCN phase. The resulting relationship, called an activation spectrum, is of the form

$$N_d = CS^k, \tag{1}$$

where N_d is the number of droplets activated, S is the maximum supersaturation produced near cloud base, and C and k are parameters related to a power-law size distribution of aerosol particles. During the course of the intervening decades, (1) has become a standard in CCN measurements. The determination of C and k provides a critical link between aerosol and droplet microphysics and important information for studying the aerosol indirect effects whereby aerosol particles affect incoming solar radiation by allowing droplets to condense upon them. Thus, these parameters are central to the prediction of the impact of CCN on clouds and future climate states.

Although we present here a method that uses (1) as its representation of the activation spectrum, other rep-

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resentations, such as those based on a lognormal representation of CCN, are equally feasible, provided they do not have more than two parameters. Thus a lognormal distribution of CCN can be used, if the breadth parameter is assumed constant. The advantage of the lognormal function is that there is copious evidence of its ability to match the aerosol population with fidelity, and that it allows for the curvature frequently observed in the log–log graphical representation of activation spectra (e.g., von der Emde and Wacker 1993). However, to demonstrate the fundamental concepts of this work, we prefer to use (1) at this point. In section 4 we explore use of the lognormal function further.

CCN measurements at various geographical locations generally distinguish between continental spectra having high C ($\approx$1000 cm^{-3}) and high k ($\approx$1), and maritime spectra with low C ($\approx$100 cm^{-3}) and low k ($\approx$0.5). However, C and k vary greatly depending on a number of factors, such as the source of the atmospheric aerosol, its age, whether or not it has been processed by clouds, the altitude of the measurement, etc.

Values of C and k are derived from measurements in thermal gradient diffusion chambers or isothermal haze chambers. These measurements are performed in situ either at the earth's surface (e.g., Twomey and Wojciechowski 1969) or on board aircraft flying in the vicinity of cloud base (e.g., Hudson 1989). Because of the importance of these parameters in the aerosol–cloud–climate puzzle, long-term monitoring of CCN parameters at different geographical locations is essential. Unfortunately, the in situ measurement of CCN is limited in that it provides only a point measurement (or line measurement, in the case of aircraft) with limited spatial resolution. Surface measurements suffer from the additional limitation that they may not be representative of the properties at cloud base. On the other hand, a remote measurement, if successful, could supply long-term datasets with fine temporal resolution and valuable information for climate monitoring studies. Feingold and Grund (1994) explored one such avenue by using multiwavelength lidar measurements to retrieve CCN parameters. That study indicated the strong sensitivity of the retrieval to measurement errors, as well as assumptions about the chemical composition of the particles. In this study, we explore a different technique. By utilizing a combination of Doppler K_a-band radar, microwave radiometer, and lidar, a method is developed whereby both C and k can be retrieved. The technique builds on the previously published technique of Frisch et al. (1995) where K_a-band radar and microwave radiometer were used to retrieve the number concentration of cloud droplets in stratocumulus. The retrieval assumes that drop number is invariant with height—a good assumption in nondrizzling stratocumulus with negligible cloud-top shear. In this work, that same retrieval of N_d is then combined with simultaneous Doppler vertical motion w, and using known relationships between w, supersaturation production, number of droplets activated, and the C and k parameters, an inversion of the value of C is performed for fixed k. If, in addition to the aforementioned measurements, a calibrated backscatter lidar measurement is available, it is shown how both C and k can be retrieved using a least squares minimization technique.

The method is outlined in section 2. Thereafter it is applied to data acquired during the Atlantic Stratocumulus Transition Experiment (ASTEX; Albrecht et al. 1995). A discussion of the results is presented in section 4, followed by a summary.

2. Method

The well-known equations for droplet activation and growth in a parcel rising at a constant velocity (see, e.g., Squires 1958; Twomey 1959; Pruppacher and Klett 1978) are used. The equation in supersaturation S is given by

$$\frac{dS}{dt} = \psi_1(T, P)w - \psi_2(T, P)\frac{dr_l}{dt}, \tag{2}$$

where w represents the updraft velocity and r_l is the liquid water mixing ratio. Here, ψ_1 and ψ_2 are functions of temperature (T) and pressure (P); their exact formulations can be found in the aforementioned references.

The growth equation for a droplet of radius r_d is given by

$$r_d\frac{dr_d}{dt} = G(r_d, T, P)[S - y(r_d, T)]. \tag{3}$$

Here, $G(r_d, T, P)$ determines the rate of condensational growth of a droplet, and $y(r_d, T)$ represents the surface tension and solute correction terms to the saturation field around the droplet. Using (3), dr_l/dt can be written as

$$\frac{dr_l}{dt} = 4\pi\frac{\rho_l}{\rho_a}\int G(r_d, T, P)[S - y(r_d, T)]r_d n(r_d)\, dr_d. \tag{4}$$

Twomey (1959) solved (2) by assuming that droplets grow from an initial radius of zero, ignoring kinetic effects (i.e., the dependence of G on r_d) and neglecting y. He then approximated $\int r_d n(r_d)\, dr_d$ through $\int S(t)\, dt$. This, together with (3), yielded a maximum supersaturation of the form

$$S = \left[\frac{A(T, P)w^{3/2}}{CkB(3/2, k/2)}\right]^{1/(k+2)}. \tag{5}$$

Here, $A(T, P)$ is given by Twomey (1977) and B is the complete beta function. Equation (1) is then used to determine N_d. Feingold and Heymsfield (1992) offered an alternative, more accurate formulation based on empirical relations from detailed parcel model calculations simulating the growth of droplets formed on completely soluble ammonium sulfate particles. The essence of the parameterization is not substantially different from the

Twomey formulation and therefore will not be repeated here.

Based on (1) and (5) we may write

$$N_d = f_1(C, k, w, T, P), \tag{6}$$

where f_1 is a known function. The first step in the retrieval of C and k involves measurement of N_d and w at known T and P. Then using (1) and (5), or an alternative formulation represented by (6), the parameter C can be retrieved for an assumed value of k. If an additional measurement is available, for example, a calibrated lidar backscatter measurement, the method can be extended as follows. The lidar backscatter is defined as

$$\beta_\pi(\lambda) = \int_{r_{\min}}^{r_{\max}} Q_\pi(r, m, \lambda)\pi r^2 n(r)\, dr, \tag{7}$$

where λ is wavelength, $Q_\pi(r, m, \lambda)$ is the backscatter efficiency at particle radius r, complex index of refraction m and λ, and $n(r)$ is the size distribution of aerosol particles. In accordance with (1) we assume that $n(r)$ can be described by a Junge (1952) power-law distribution

$$\frac{dN}{d\ \ln r} = C_j r^{-\beta}, \tag{8a}$$

or

$$n(r) = C_j r^{-\beta-1}, \tag{8b}$$

although β is not fixed as in the aforementioned work. It is then straightforward to show (see appendix) that the two free parameters in (8), C_j and β, are related to C and k as follows:

$$C_j = 1.5kCD^{k/2}; \qquad \beta = 1.5k, \tag{9}$$

and D is a function of particle chemistry (see appendix). A further interesting relation is that between the Ångström coefficient α, which describes the backscatter dependence on wavelength, and k

$$\alpha = \beta - 2 = 1.5k - 2. \tag{10}$$

Substituting (8b) into (7), and using (9), (7) may be written in the general form

$$\frac{\beta_\pi}{f_2(C)} = f_3(k). \tag{11}$$

Thus, a measurement of β_π enables determination of k provided C is known. Also, if a measurement of the Ångström coefficient α is available, (10) can be used to determine k. Unfortunately, measurements of α are usually performed at the surface so that their utility in this retrieval may be limited, unless the boundary layer is well mixed and the settling velocity of particles is small enough so that vertical gradients in the size distribution are minimal.

A retrieval algorithm for C and k has been formulated based on the above discussion. It consists of using (6)[1] and (11) together with a least squares minimization technique to determine the free parameters. The technique assumes that the CCN distribution changes on timescales longer than the sample period for which C and k are sought. The vertical velocity w, on the other hand, changes rapidly and generates local activation of new droplets. Finding C and k from the observations is equivalent to a problem that has two unknowns (C and k) and a number of observations that link C and k through nonlinear equations (6) and (11). This problem can be solved using the least squares method. Specifically, optimal values of C and k can be obtained by minimizing the squared difference between the observed drop concentration and backscatter and their theoretical values calculated for a given C and k. This difference is termed a cost function, defined as

$$J(C, k) = \sum_i [w_1(N_o^i - N_d^i)^2 + w_2(\beta_o^i - \beta_\pi^i)^2], \tag{12}$$

where i indicates the ith sample; N_o and β_o represent the observed drop concentration and lidar backscatter, respectively; N_d^i and β_π^i represent the theoretical values of drop concentration and backscatter calculated according to (6) and (11) from the observed w, T, and P; and w_1 and w_2 are the weightings for drop concentration and backscatter observations, and are assigned to the reciprocals of the variances of their observational errors. Here, $J(C, k)$ is minimized using the quasi-Newton conjugate-gradient algorithm (Liu and Nocedal 1989). It is worth noting that C and k should be properly scaled in order to obtain a convergent solution. In this paper, C and k are scaled as

$$C' = C/C_s, \qquad k' = k/k_s, \tag{13}$$

where C_s and k_s are the scales of C and k, respectively. Here, $C_s = 500\ \text{cm}^{-3}$ and $k_s = 0.5$. Numerical tests have shown that J converges to its minimum as long as $C \in (50, 5000)$ and $k \in (0.1, 5.0)$.

3. Results

The algorithm described in section 2 is applied to a dataset acquired by the National Oceanic and Atmospheric Administration's (NOAA) Environmental Technology Laboratory on the island of Porto Santo in the Madeiras during ASTEX. The primary instruments were the NOAA K_a-band (8.6-mm) Doppler radar (Kropfli et al. 1995), 10.59-μm lidar (Pearson 1993), and microwave radiometer (Hogg et al. 1983), all collocated on the island. Radar data were recorded at 3-s temporal resolution and 37.5-m vertical resolution; radiometer data, yielding cloud integrated liquid-water path, were recorded at 1-min intervals. The lidar recorded range

[1] Equation (6) will be represented in the form given by Feingold and Heymsfield (1992).

measurements of power and Doppler velocity at a temporal resolution of 1 s. The lidar system was still in its infancy and unfortunately was not calibrated. Therefore, we have had to apply a calibration factor to these data in order to yield reasonable retrievals. Sensitivity to this calibration factor will be investigated in section 4. A ceilometer run by Colorado State University was also available and provided data on cloud base at 1-min resolution.

A look-up table of the normalized lidar backscatter (11) as a function of k was built a priori assuming a Junge distribution defined over the range 0.01 μm–1 μm, and a relative humidity of 95%. Aerosol particles were assumed to be composed of pure ammonium sulfate and at their equilibrium sizes. Sensitivity to these assumptions will be explored in section 4.

Data are conditionally sorted into updrafts and downdrafts, with only the updrafts considered, since it is they that are responsible for activation of new droplets. No other sorting of the data has been performed, although in principle, the technique should only be applied to active growth regions of cloud and not to decaying cloud. The value of w at one radar range gate (37.5 m) above cloud base (determined by lidar or ceilometer) is used. Data are also thresholded so that the retrieval is not attempted when the maximum radar reflectivity factor is larger than -15 dBZ, to avoid contamination by drizzle. The presence of drizzle drops would bias the w measurement and mean that the measured w is some indeterminate mix of vertical air motion and drop terminal velocity. The retrieval is also not attempted when updrafts are less than 5 cm s^{-1}.

The data are ingested into the retrieval algorithm (12) for a short period of time for which simultaneous vertically pointing radar and lidar returns were available. These data spanned the period 1035–1135 UTC on 22 June 1992 during which there were no indications of a shift in air mass. Although copious amounts of data were collected by these instruments during ASTEX, the lidar was still in its prototype design phase and we are uncomfortable with applying the lidar data beyond this period. Fortuitously, the U.K. C-130 instrumented aircraft flew over the measurement site, albeit over 4 h later (1558 UTC). Nevertheless, the CCN measurements from that flight provide some in situ measurement against which to evaluate this remote sensing technique.

Table 1 indicates the retrieved C and k parameters for the sample period. Other values reported relate to the sensitivity studies discussed in section 4. The retrieved C and k parameters for the baseline simulation are $C = 175$, $k = 1.55$. These values are quite reasonable for typical marine boundary layers (e.g., Hudson 1980). The log reports of the Meteorological Research Flight A215 (Johnson et al. 1992) indicated that on this day there existed a ''continental airmass [with] several very distinct haze layers'' and ''very sharp concentration transitions from 100 to 2000 cm^{-3} measured by the PCASP above cloud.'' [2] The data from this day are not ideal for intercomparison, first, because they were not collocated and simultaneous, and second, because of the degree of variability in the vertical. The comparison of the remote sensing data with the in situ data is therefore of limited value but is nevertheless included in Table 1. It is seen that the observed C is significantly (4.5 times) larger than the base case retrieval; however, the observed value of $k = 1.30$ is fairly close to the retrieved value of 1.55. The only way that the retrieval technique can match this high C would be if the retrieved N_d were significantly higher. As noted earlier, the Frisch et al. (1995) technique is the source of the N_d measurement.

TABLE 1. Retrieved C and k parameters for base case and sensitivity tests. The base case assumes an aerosol composition of ammonium sulfate, $r_{min} = 0.01$ μm and $r_{max} = 1$ μm. Relative humidity at the height of the backscatter measurement is assumed to be 95%.

Experiment	C	k	Details
Base case	175	1.55	See text
S0	677	1.84	$\sigma = 1.43$ in N_d retrieval
S1	175	1.80	$r_{max} = 2.2$ μm
S2	164	1.00	$\beta_\pi + 10$ dB
S3	172	2.06	$\beta_\pi - 10$ dB
S4	218	1.40	T + 5°C
S5	175	1.78	RH = 98%
In situ	789	1.30	Aircraft measurement 4.5 h later

By increasing the assumed breadth parameter of the drop size distribution used in that retrieval from a geometric standard deviation of 1.20, or relative dispersion of 0.18 (as used here) to one of 1.43 (relative dispersion of 0.37), one achieves larger N_d and $C = 677$, $k = 1.84$ (indicated by S0 in Table 1). Although no in situ measurements of drop spectra are available for this event, we do not believe the breadth of the drop size distribution was this large in the nonprecipitating clouds analyzed here. An alternative explanation for the discrepancy might lie with measurement uncertainty associated with the CCN spectrometer.

4. Discussion

a. Sensitivity to assumptions used in the retrieval

A number of experiments have been performed to establish the sensitivity of results to assumptions used in the retrieval. These appear in Table 1 and are labeled S1–S5. Here, S1 assumes that the upper cutoff of the aerosol distribution is an order of magnitude larger in mass, or a factor of about 2.2 larger in radius. This does not affect the retrieved value of C but increases k to 1.8 compared to the base case value of 1.55. The reason is that the presence of larger particles tends to produce enhanced backscatter, but under the constraint of equal

[2] The PCASP, or passive cavity aerosol spectrometer probe, measures particles in the size range 0.1–3 μm.

β_π, the algorithm converges on a larger k or smaller number of large particles [see (8) and (9)]. Experiments S2 and S3 were performed bearing in mind the uncertainties in the lidar backscatter calibration. An increase in β_π of +10 dB (a factor of 10) results in a drop in C from 175 to 164, and a more significant decrease in k from 1.55 to 1.00. This increase in β_π has to be provided by a larger fraction of large particles or smaller k. A decrease in β_π of −10 dB has little effect on C, but k is increased to 2.06. Again, this decrease in β_π has to be accounted for by a much smaller number of large particles, or larger k. Note that these are rather extreme values of uncertainty; typical β_π errors are about 3 dB, or a factor of 2.

Experiment S4 assumes that the cloud-base temperature is 5°C higher than used for the base case. Here, there is a strong increase in C (218 vs 175), and a drop in k to 1.4. This emanates from the sensitivity of the function f_1 to T and the fact that supersaturations are lower at higher T (all other fields being equal). According to (1), a lower S must result in a higher C if N_d is constant. This results in a higher C for a fixed number of N_d (assuming constant k); clearly the situation is a little more complex when the assumption of fixed k is relaxed. Finally, S5 indicates the sensitivity to the base case assumption that particles are in an environment of 95% relative humidity by increasing this value to 98%. Here C remains unaffected by this change, while k increases from 1.55 to 1.80. The explanation for this again relates to the fact that a higher RH of 98% tends to produce enhanced backscatter, but if β_π is held constant, a reduction in the relative number of large particles (or larger k) must follow. The result for S5 is very similar to that of S1 (where $r_{\max}$ is larger), which is consistent with the fact that a higher RH results in enhanced deliquescence growth of particles.

In general, the k parameter is more sensitive than C. The reason is that with the exception of S4, the experiments all affect β_π, which is a strong control on k [see (11)]. In the case of S4, the change in T mainly affects C through the close relation between N_d, S, and C. Experiments S2 and S3 form the lower and upper bounds on k and indicate that lidar backscatter must be well calibrated if this technique is to be applied. If an acoustic sounder is collocated with the other instruments, measurement of cloud base T is straightforward and any errors incurred should be small. Errors in P have almost no effect on the retrieval. On the other hand, assumptions about RH, $r_{\max}$, and particle chemistry are more difficult to constrain. An average value of RH of 98% may be more reasonable than 95%, considering that only updraft regions are considered. This assumption will also depend on the lidar range gate used as a source of the β_π data. In these retrievals, a single range gate was used because cloud base was fairly constant; however if cloud base varies and crosses from one range gate to another, it is quite conceivable that there will be some level of variation in RH associated with the vertical resolution of the lidar. The $r_{\max}$ assumption is a difficult one to constrain, although meteorological conditions may be of some help. For example, if the atmosphere is stably stratified, there should be no large particle source from the surface, and given that residence time decreases with increasing size, the subcloud aerosol should be devoid of giant particles. On the other hand, in a well-mixed marine boundary layer, breaking waves could be a source of giant CCN, some fraction of which make their way up to cloud base.

Finally, some information on particle chemistry would be valuable. It is noted that the general form of the relationship between wet and dry aerosol radius is given by

$$r_{\text{wet}} = \delta r_{\text{dry}}^{\gamma}, \tag{14}$$

(e.g., Fitzgerald 1975). The coefficient δ varies with chemical composition and RH, whereas γ is a function of RH only. For example, sodium chloride has a value of δ that is about 35% larger than that for ammonium sulfate (Fitzgerald 1975), so that incorrect assumptions about chemistry would be equivalent to a 35% enhancement in particle size.

b. Dependence on cloud droplet number retrieval

The CCN activation spectrum retrieval technique depends on a good measure of drop number N_d. In this work, that parameter has been derived following Frisch et al. (1995). A few words in this regard are in order. The method retrieves profiles of cloud water and median drop size, under the assumption of a constant-in-height number concentration and spectral breadth. To the extent that these conditions are met in real clouds, the scheme will perform well (Feingold et al. 1995). However, in many cloud scenarios these conditions may not be met and alternative techniques for retrieving cloud base N_d should be considered. One such possibility is the combination of radar and lidar. It has been shown (Eberhard et al. 1997) that by taking the ratio of radar and lidar backscatter, and assuming a functional form of the drop spectrum, two parameters of the spectrum (number and size) can be retrieved. Because lidars suffer from attenuation as they penetrate clouds, this retrieval is only of use in the region of cloud base, but since this is the region of interest in our case, it could provide a useful alternative to the Frisch et al. (1995) scheme.

c. Relative role of C and w in determining N_d

Twomey (1977) discusses how for a particular T and P, (1) and (5) yield a dependence of N_d on C and w that varies with k:

$$N_d \propto C^{1-k/(k+2)} w^{1.5k/(k+2)}. \tag{15}$$

For $k = 0.5$, $N_d \propto C^{0.80} w^{0.30}$, indicating that drop number is primarily determined by C, and much less so by w. With increasing k, w takes on an increasingly

important role in determining N_d so that for $k = 1.5$, $N_d \propto C^{0.57}w^{0.64}$, and for $k = 2$, $N_d \propto C^{0.50}w^{0.75}$. The k values listed in Table 1 are all greater than unity, and thus w is an important factor. It is interesting too that in recent years, the advent of a new generation of in situ instruments (e.g., Hudson 1989) that measure down to low S (on the order of 0.01%) indicate larger k than some of the earlier instruments that could not measure at very low S. Because the low S measurements measure the larger CCN particles, this could be indicative of a variable slope in the activation spectrum (plotted on a log–log plot) as has been measured by Hudson and Frisbie (1991). When converting this spectrum to a size distribution for an assumed particle chemistry, this translates to a curvature in the size distribution (when plotted on log–log ordinates) and deviation from (8). A variable k across the size spectrum could also be indicative of a size-dependent chemical composition of particles.

d. Limitations of the C, k representation

As pointed out by Twomey (1977), the Junge power-law distribution (8) and the closely associated C and k parameters (9) is limited in that it requires a minimum radius for a closed solution to the particle concentration. In addition, various other physical parameters do not exist for specific values of β. Perhaps more importantly, measurements of particle size spectra indicate that the Junge distribution is only approximate and that a lognormal distribution is more appropriate. In fact, many workers (e.g., Jaenicke 1988) have suggested a bi- or even trimodal lognormal distribution. Clearly, for the purposes of climate monitoring, it makes little sense to use parameterizations that would require the retrieval of up to nine parameters (for the case of a trimodal lognormal) from remote measurements. Nevertheless, it is feasible to perform the same retrieval presented here under the assumption of a single lognormal distribution of CCN with fixed breadth

$$n(r) = \frac{N_a}{(2\pi)^{1/2}\ \ln\sigma r} \exp[-\ln^2(r/r_g)/(2\ \ln^2\sigma)], \quad (16)$$

where N_a is the total CCN concentration, r_g is the median size of the CCN particles, and σ is the geometric standard deviation is a constant. Equation (6) is then written as

$$N_d = f_4(N_a, r_g, w, T, P), \quad (17)$$

where f_4 is a function given by Ghan et al. (1993). Von der Emde and Wacker (1993) have also used the lognormal form for the CCN spectrum and shown that instead of the characteristic straight line on a log–log display predicted by (1), the activation spectrum has a concave downward curvature. The proposed algorithm is easily extended to the lognormal function using (7) with $n(r)$ represented by a lognormal function. Equation (11) then has an analog of the following form:

$$\frac{\beta_\pi}{N_a} = f_5(r_g). \quad (18)$$

By analogy to the proposed method [(6) and (11)], retrieval of N_a and r_g is performed with the aid of (17) and (18). The lognormal form is thus a viable alternative to the C, k representation and at this point it appears a matter of preference which form is more desirable. Clearly both are limited in that they simplify the form of CCN distribution. The large body of data on C and k at different locations in the world might indicate a preference at this stage for the C, k representation. On the other hand, some general circulation models (e.g., Ghan et al. 1993) include prognostic equations based on a lognormal distribution.

e. Multiple-sensor issues

The proposed technique requires a combination of three remote sensors, each having a different field of view. For example, the beam divergences for the K_a-band radar, microwave radiometer, and lidar are, respectively, 0.5°, 2.5°, and 0.004°. Note that the radar/radiometer pair have beam divergences with the least disparity and therefore sampling volumes are not grossly different within the boundary layer. To the extent that this is a problem, retrievals of N_d will be affected. One method of alleviating it is to temporally average the data, and this is currently under investigation as part of a field experiment that includes in situ measurements of N_d to verify the technique. The disparity in volumes sampled by the instruments is especially important when a combination of the measurements is used to derive a physical property of the particles in partially overlapping sample volumes. However, here the difference in sample volumes between the radar/radiometer pair and the lidar is not a serious problem because the former is used to retrieve *in-cloud drop spectra*, while the latter is used to retrieve *subcloud aerosol properties*. In other words, different physical properties are retrieved by the radar/radiometer and lidar, each in their respective sampling volume.

5. Summary

A technique for retrieving CCN parameters using a combination of a Doppler K_a-band cloud radar, a microwave radiometer, and a calibrated lidar has been presented. The retrieval relies on the technique of Frisch et al. (1995) to retrieve cloud droplet number concentration. Using theoretical relationships between drop number, radar-measured vertical velocity, supersaturation production, and the CCN parameters, together with a calibrated backscatter measurement, the C and k parameters in the $N_d = CS^k$ relationship proposed by Twomey (1959) are derived. A least squares retrieval has been applied here to a dataset acquired during ASTEX. Unfortunately, lidar backscatter measurements

were not calibrated and so a calibration has been assumed; sensitivity studies have been performed by varying this calibration over a broad range.

Results show that retrieved values of C and k are quite reasonable, although there is no validation for these retrievals other than an in situ aircraft measurement that was taken 4 h later. Analysis indicates sensitivity of the k parameter to assumptions about the backscatter calibration, upper truncation of the distribution, and cloud-base RH. For this technique to be useful at some level, a well-calibrated lidar system will be important. In addition, because the cloud droplet number retrieval relies on radar and radiometer measurements, these instruments will also have to be well calibrated.

It is stressed that the various assumptions made in this retrieval are simplifications of the real world; the assumption of the Junge power-law size distribution is perhaps the most severe. Nevertheless we are of the opinion that the advantages of being able to provide copious amounts of data with a level of detail that is appropriate for analysis of first-order aerosol effects, as well as for use in general circulation models, are important enough to warrant pursuing measurements of this kind. Focused experiments that collect data specifically with this problem in mind will be required, including in situ verification with thermal gradient diffusion chambers. If a greater understanding of the remote sensing retrieval is achieved by intercomparison with in situ measurements, the true utility of this technique will be clearer. Thus, it is not suggested that this technique replace the detailed in situ measurement, but rather augment it. Given the importance of CCN measurements in the aerosol–cloud–climate problem, we feel this is a worthy pursuit. The alternative—namely, a reliance on surface measurements of CCN, or the occasional airborne campaign—seems inadequate.

Further testing of this technique could be performed at the Atmospheric Radiation Measurement Cloud and Radiation Testbed site in Oklahoma where all instruments required by this retrieval are located. In situ measurements of activation spectra during intensive observation periods will establish the ''ground-truth'' activation spectra. Two lidar systems, the NASA micropulse lidar (Spinhirne et al. 1993) and Raman systems (Goldsmith et al. 1996), once calibrated, will provide important information on both backscatter and relative humidity (in the case of the Raman lidar) that will be essential for successful implementation of this algorithm.

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APPENDIX

Derivation of Eq. (9)

The approximate Köhler relation is

$$S = 1 + \frac{a}{r} - \frac{b}{r^3}, \tag{A1}$$

where

$$a = \frac{3.310^{-5}}{T} \quad \text{and} \quad b = \frac{4.3im}{M_s}. \tag{A2}$$

Here, i is the van't Hoff factor (approximately equal to 2), m is the aerosol mass, and M_s is the molecular weight of the solute. When $dS/dr = 0$, we obtain the relation between S and particle radius:

$$S = \left(\frac{4a^3}{27b}\right)^{1/2}. \tag{A3}$$

Substituting (A3) into (1), we obtain after some rearranging

$$N = CD^{k/2}r^{-3k/2}, \tag{A4}$$

where

$$D = \frac{3a^3M_s}{27(4.3i\pi\rho_s)}, \tag{A5}$$

and ρ_s is the aerosol density. Now, since

$$N = \int_r^\infty n(r)\, dr = \int_r^\infty C_j r^{-\beta-1}\, dr, \tag{A6}$$

we have

$$N = \frac{C_j}{\beta}r^{-\beta}. \tag{A7}$$

Comparing (A4) and (A7), we obtain (9):

$$C_j = 1.5kCD^{k/2}; \qquad \beta = 1.5k. \tag{A8}$$

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