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The Relationship Between Cloud Droplet Number Concentrations and Anthropogenic Pollution: Observations and Climatic Implications

W. R. LEAITCH, G. A. ISAAC, J. W. STRAPP, C. M. BANIC, AND H. A. WIEBE

Atmospheric Environment Service, Downsview, Ontario, Canada

Measurements of the concentrations of sulfate and nitrate in approximately 400 cloud water samples collected during four field studies carried out since 1982 are used with coincident measurements of cloud droplet number concentrations (CDNC) and liquid water content (LWC) to examine the relationship between CDNC and anthropogenic pollution, where sulfate concentration is used as the measure of the latter. The number of samples is compressed to 92 by averaging duplicates and multiple samples at similar altitudes during any particular flight, with 85 including CDNC measurements. Positive linear regressions between log (CDNC) and log (cloud water sulfate concentration) are determined for both stratiform and cumuliform cloud. Because of the number of factors affecting the CDNC, the coefficients of determination are only 0.30 and 0.49 for the respective cloud types. The LWC is relatively invariant with the cloud water sulfate concentration. The observed range of CDNC for the study region is 20–600 cm^{-3} (median of 59 observations is 210 cm^{-3}) for stratiform clouds and 170–1100 cm^{-3} (median of 26 observations is 400 cm^{-3}) for cumuliform clouds. The median CDNC for all sampled clouds is 250 cm^{-3}. CDNC are also determined for "clean-air" conditions. The latter is defined as cases for which the concentrations of both cloud water sulfate and cloud water nitrate are comparable to aerosol sulfate concentrations and aerosol nitrate plus HNO_3 concentrations, respectively, as reported for remote regions of the globe. For the clean-air clouds the observed range of CDNC for the study region is 20–250 cm^{-3} (median of 12 observations is 120 cm^{-3}) for stratiform clouds and 170–370 cm^{-3} (median of four observations is 240 cm^{-3}) for cumuliform clouds. The median CDNC for all clean-air clouds is 160 cm^{-3}. The median CDNC for the complete population is 56% greater than the clean-air CDNC; the hypothesis that the clean-air CDNC is not different from the median CDNC is rejected at a confidence level of >99.5%. The present-day climatic forcing due to cloud albedo change arising from increased CDNC is estimated from a rudimentary model at between -2 W m^{-2} and -3 W m^{-2} for eastern North America.

INTRODUCTION

It has been suggested that global warming could be offset by a chain of processes initiated with increasing anthropogenic sulfur emissions and culminating in increased cloud reflectance [*Twomey*, 1977; *Twomey et al.*, 1984]. The latter results from an increase in the cloud droplet number concentrations (CDNC), because of increased concentrations of sulfate particles which will act as nuclei for the cloud droplets. The cloud liquid water content (LWC) is assumed to be invariant, while the manner in which water is distributed (i.e., the numbers and sizes of droplets) is altered. This reduces the amount of short wave radiation reaching the surface of the Earth, thereby cooling the atmosphere. A hypothesis concerning the control of global climate via natural sulfur emissions (i.e., dimethyl sulfide (DMS)) and cloud reflectance has also been advanced [*Charlson et al.*, 1987], and it has been implied that the biosphere, through its influence on cloud albedo and precipitation, may respond to anthropogenically induced changes in the atmospheric sulfur cycle [*Meszaros*, 1988]. Although substantiation of these hypotheses is difficult [e.g., *Schwartz*, 1988; *Wigley*, 1989], the apparent ability of increasing CDNC to visibly increase cloud albedo has been demonstrated through satellite images of "ship tracks" in cloud [*Coakley et al.*, 1987; *Radke et al.*, 1989], and relationships between DMS and condensation nuclei have been measured [*Bates et al.*, 1987]. One estimate of the importance of natural sulfur emissions to global climate has indicated it to be small [*Foley et al.*, 1991]; however, it is possible that anthropogenic sulfur emissions may have already played an important role in climate via changing CDNC [*Wigley*, 1989]. *Kaufman et al.* [1991] suggest that the atmospheric cooling resulting from the effect of SO_2 emissions on cloud albedo presently dominates over the atmospheric heating from the greenhouse effect of CO_2 emissions. However, they also suggest that nonlinearities in relationships among cloud condensation nuclei, CDNC, and cloud optical depth may reverse that dominance in the future if the burning of fossil fuels continues to increase.

Much of the current discussion on this issue is quite speculative due to a lack of observational data describing how the CDNC are affected by changing pollution. Here, airborne measurements of CDNC, LWC, and cloud water sulfate and nitrate concentrations (multiplied by the LWC) in stratiform and cumuliform clouds performed over eastern North America from 1982 to 1988 are used to address this problem of pollution, CDNC, and cloud albedo. The relationship between the CDNC and cloud water sulfate, increases in CDNC attributable to anthropogenic sources, and the present-day climatic forcing for eastern North America due to the direct effect of increased CDNC on cloud albedo are discussed.

EXPERIMENT

Clouds were sampled during four intensive studies carried out in the summer of 1982, the winter of early 1984, the autumn of 1984, and the summer of 1988. All studies were conducted over central Ontario, Canada, with the exception of the autumn study which was conducted over upper New York State. Stratiform and cumuliform clouds were sampled under both warm and supercooled conditions. The individual

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studies are described in more detail by *Isaac et al.* [1986, 1990, 1991], *Leaitch et al.* [1986*a*], *Isaac and Daum* [1987], and *Strapp et al.* [1988].

The instrument platform was the Canadian Institute for Aerospace Research deHavilland Twin Otter aircraft. Measurements of LWC and CDNC were made continuously with devices mounted under the wings of the aircraft. Most of the LWC were measured using either a Johnson-Williams hot-wire probe or a particle measuring systems (PMS) King hot-wire probe. These probes have been routinely calibrated in icing tunnels [e.g., *Strapp and Schemenauer*, 1982], and the measurement error is estimated to be ± 0.05 g m^{-3} for most of the concentrations measured here. For a few samples it was necessary to use a PMS FSSP-100 optical particle counter to determine the LWC. The error in the FSSP-100 LWC is a combination of sizing and number uncertainties, and for the typical diameters and LWC here (i.e., about 10 μm and 0.2 g m^{-3}, respectively) it is estimated to be ± 0.10 g m^{-3}. The CDNC were measured with the FSSP-100. All CDNC were corrected for probe dead time and coincidence error following the method described by *Baumgardner et al.* [1985]. For the majority of the concentrations here, the CDNC measurement error is estimated at $\pm 10\%$; the uncertainty may be greater for the higher CDNC here.

Aerosol particles were sized and counted with a wing-mounted PMS active scattering aerosol spectrometer probe (ASASP-100X). The probe has been routinely calibrated using nearly monodisperse NaCl particles and latex spheres, details of which are described by *Liu et al.* [1992]. The lower detection limit of this probe is estimated at 0.17 μm.

Samples of the cloud water were analyzed for the concentrations of sulfate and nitrate by ion chromatography using a Dionex 2000i. Cloud water collections were made in warm cloud using the slotted-rod technique described by *Winters et al.* [1979]. One drawback of this technique is the potential for the measured chemistry to be weighted toward the solute contained in the larger cloud droplets owing to a reduced efficiency of the collector at small droplet sizes (i.e., <10 μm diam). In consideration of this problem, modified and standard versions of this collector were flown simultaneously during the 1988 study. The principal modification was to make the rods one half of the width of the standard collector, with the object being to increase the collection efficiency at smaller droplet sizes; a similar modification is described by *Huebert et al.* [1988]. Simultaneous measurements with the two collectors during the 1988 study indicated no systematic differences, and the concentrations of cloud water sulfate ($cwSO_4^=$) were found to be within $\pm 25\%$ of each other in 44 of 58 cases and to within $\pm 60\%$ in 57 instances. Comparison of simultaneous samples collected during the earlier studies, but with two standard-type collectors, yielded similar results. Although the modified collector has not been sufficiently evaluated to consider it an "absolute" collector, an error in the absolute values of $cwSO_4^=$ due to collection efficiency sufficient to impact on the following discussion is not indicated. Supercooled cloud collections were made using solid rods or meshes exposed to the airstream in a similar manner to the warm collectors. In this case, the rods or meshes collected rime. After exposure the rimed rods or meshes were placed into sterilized polyethylene bags. The meltwater was extracted and analyzed in the laboratory. Details of this process are described by *Isaac and Daum* [1987].

OBSERVATIONS

Approximately 400 cloud water samples were collected. LWC and CDNC were averaged for each individual or duplicate collection. The length of time required to collect a cloud water sample varies from 0.5 to 10 min, equivalent to a distance of 2–40 km. The cloud water chemical concentrations were multiplied by the measured LWC, prior to any statistical reduction, so that the concentrations are referenced to a volume of air. The $cwSO_4^=$ and cloud water nitrate concentrations ($cwNO_3^-$) are then directly comparable to concentrations of sulfate and nitrate measured in air. For each flight the median values of the concentrations of samples collected in each 300 m altitude interval were determined, compressing the total number of samples to 92 and eliminating unwarranted weighting of the distributions by flights on which a large number of samples were collected. Because of instrument difficulties there are only 85 CDNC averages. For this compressed data set, percentiles of pertinent quantities for the individual studies and the complete data set are given in Table 1. All concentrations, CDNC and LWC as well as $cwSO_4^=$ and $cwNO_3^-$, are expressed at 0°C and 1 atm. Histograms of these four quantities are shown in Figure 1 as the "all" distributions; the "clean-air" distributions also shown in Figure 1 are discussed later.

During the two summer studies, slightly more than half of the samples were collected in cumuliform clouds with the remainder taken from stratiform clouds. During the autumn and winter studies, most of the samples were collected in stratiform clouds. The CDNC and LWC in Table 1 are higher for the two summer studies than for the autumn and winter studies, as are the median collection altitudes. This is consistent with higher cloud bases and stronger updrafts found in the summertime. Conversely, the highest median $cwSO_4^=$ and $cwNO_3^-$ are found during the winter study. This result may reflect the fact that the median collection altitude is lowest for the winter, due to lower cloud bases, and the lower mixing heights during this season can lead to higher concentrations near the ground [*Isaac et al.*, 1990].

SULFATE-CDNC RELATIONSHIP

The CDNC are plotted as a function of the $cwSO_4^=$ for each study in Figure 2. The median CDNC for each $cwSO_4^=$ quartile (i.e., 0–25, 25–50, 50–75, and 75–100) are also shown (large circles). An upward trend in CDNC with increasing $cwSO_4^=$ is indicated for each study. In Figure 3 the median CDNC for each $cwSO_4^=$ quartile for the four projects are shown together in order to compare the results from the individual studies. The agreement between the two summer cases suggests a reproducible relationship. The separation between the different seasons is consistent with the above discussion of seasonal differences in CDNC and cloud type.

The data were divided between stratiform and cumuliform cloud: 59 points are stratiform cases and 26 points are cumuliform cases. In Figure 4 the CDNC plotted as a function of $cwSO_4^=$ are shown for the stratiform and cumuliform cloud. The median CDNC for each $cwSO_4^=$ quartile are again plotted (open circles), and regressions of the CDNC (units of cubic centimeters) onto the $cwSO_4^=$ (units of nEq m^{-3}) are also included (solid curves). The regression curves are given by

TABLE 1. Seasonal and Combined Percentile Data for Cloud Sampling

Project	Percent	Altitude km	LWC g m^{-3}	CDNC cm^{-3}	Temperature °C	nEq m^{-3} $cwSO_4^=$	nEq m^{-3} $cwNO_3^-$
Summer	25	1.5	0.18	260	3.7	17	8.0
1982	50	2.1	0.30	360	7.2	34	15
(19)	75	2.8	0.61	400	12	63	99
Winter	25	1.0	0.08	110	−10	17	4.9
1984	50	1.3	0.11	190	−7.4	36	23
(27)	75	1.8	0.17	250	−3.8	57	46
Autumn	25	1.2	0.14	200	−1.7	15	8.1
1984	50	1.5	0.23	240	5.7	22	14
(17)	75	1.8	0.33	270	8.0	56	34
Summer	25	1.5	0.18	240	7.6	7.6	3.6
1988	50	2.0	0.26	350	10.	16	10
(29)	75	2.3	0.37	510	14.	50	36
All	10	1.0	0.09	110	−9.8	4.0	2.6
(92)	25	1.2	0.12	170	−5.1	15	7.4
	50	1.8	0.19	250	4.5	25	15
	75	2.1	0.34	380	11	57	48
	90	2.7	0.53	510	14	130	100

The values are strictly percentiles; no relationships among the different quantities should be inferred from this table. The values in parentheses indicate the effective number of samples used. LWC is liquid water content, and CDNC are cloud droplet number concentrations.

$$\log (\mathrm{CDNC})_S = 0.257(\pm 0.052)\ \log (cwSO_4^=) + 1.95(\pm 0.21) \quad (1)$$

$$\log (\mathrm{CDNC})_C = 0.186(\pm 0.038)\ \log (cwSO_4^=) + 2.33(\pm 0.13) \quad (2)$$

where the subscripts S and C refer to stratiform and cumuliform, respectively, and the numbers in parentheses are the standard error. Values of the coefficient of determination (R^2) are 0.30 and 0.49 for (1) and (2), respectively. The log-log regression lines and the median CDNC for each $cwSO_4^=$ quartile are quite similar; however, the central tendency is toward slightly higher sensitivities of the CDNC to the $cwSO_4^=$. Regressions for CDNC-$cwSO_4^=$ and CDNC-log ($cwSO_4^=$) were also computed: for the stratiform case, the greatest variation was explained with the log-log regression (i.e., equation (1)); for the cumuliform case, the greatest variation was explained with the CDNC-$cwSO_4^=$ regression, with the log-log regression second. Inspite of the slightly lower R^2 for the cumuliform log-log regression, it is felt that (2) is the best representation, because the linear regression is strongly influenced by the highest $cwSO_4^=$ (i.e., >100 nEq m^{-3}). This is not desirable since sulfate values of <100 nEq m^{-3} are much more common in the atmosphere.

The relatively low values of R^2 serve to show that variables other than sulfate influence the CDNC. The more important of these variables are updraft speed, temperature, and aerosol composition. By differentiating between stratiform and cumuliform cloud, the more significant variations due to updraft speed have been considered. However, variations of updraft speed among the same cloud types will still be an important influence on the variability observed in Figure 4. The effects of the temperature and aerosol composition are now examined.

The effect of temperature on the CDNC is shown in Figure 5. Here the CDNC are plotted as a function of the cloud temperature. For the stratiform cloud the CDNC appears to be independent of the temperature. For the cumuliform data a positive trend is indicated. (The highest CDNC point in Figure 5*b* was sampled in a towering cumulus approximately 3.5 km above the cloud base [*Leaitch et al.*, 1991*a*]. Although the sampling temperature was 0°C, the cloud base temperature was about 25°C. Since the CDNC are primarily determined at cloud base, this point strictly belongs at a higher temperature. This problem may affect other points in this plot; however, this is by far the most significant change.) In contrast, cloud droplet growth theory indicates lower temperatures give higher CDNC for the same LWC, suggesting that negative trends are to be expected in Figure 5 if other factors do not dominate the control of the CDNC. One explanation for a positive trend with temperature in these cumuliform data is that for this study region, higher temperatures usually accompany southerly winds, which also bring higher aerosol sulfate concentrations [*Isaac et al.*, 1986, 1990]. If the true effect of temperature on the CDNC is as indicated by theory, then temperature is moderating the relationships in Figure 4.

The variability in the relative concentration of water-soluble species in the aerosol, of which sulfate species usually dominate, will also affect the CDNC [*Fitzgerald*, 1974]. This is examined here using the data from the summer 1988 study. In Figure 6*a* the aerosol sulfate concentration measured on airborne filter samples is compared with the aerosol mass estimated from the ASASP measurements, assuming an individual particle mass density of 2 g cm^{-3}. It can be seen from Figure 6*a* that on average the fraction of sulfate in the aerosol increases with increasing aerosol mass concentration. This is also a moderating factor for the relationships in Figure 4. In Figure 6*b* the total particle number concentration, measured with the ASASP, is plotted against the filter sulfate concentration. The similarity of this relationship and that of Figure 6*a* indicates that mean properties of the size distribution of the aerosol do not change dramatically with concentration; this is shown more directly by the observations of *Leaitch and Isaac* [1991].

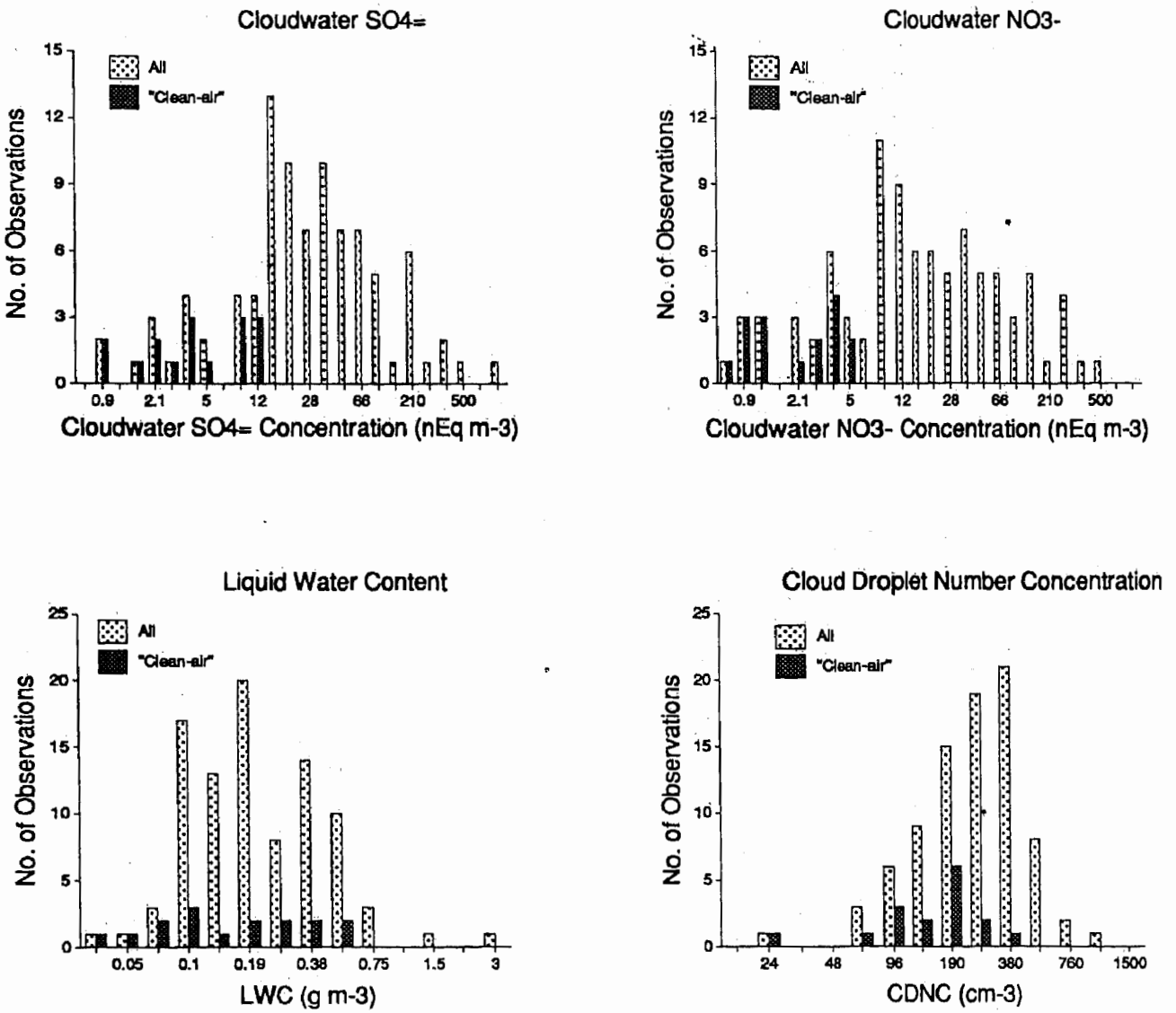

Fig. 1. Histograms of $cwSO_4^=$, $cwNO_3^-$, LWC, and CDNC for all samples and for the clean-air samples only.

Since the cloud condensation nuclei are a subset of these aerosol particles, the variability in the relationships between the aerosol mass concentration and sulfate mass concentration and between aerosol number concentration and sulfate mass concentration will contribute to the variability in the CDNC-$cwSO_4^=$ relationship. Note also that the range of aerosol particle number concentrations in Figure 6*b* is much greater than that of the CDNC in Figure 4. This may be explained in some part by the following factors: particles smaller than the detection limit of the ASASP are activated, particularly at lower number concentrations, while the increase in sulfate mass attributable to these particles is usually negligible; the fraction of sulfate in the aerosol is higher for higher concentrations; and the cloud nucleation scavenging efficiency is reduced at higher aerosol concentrations [*Jensen and Charlson*, 1984; *Leaitch et al.*, 1986*b*].

The aqueous phase oxidation of SO_2 (hereafter APOS) will increase the $cwSO_4^=$ and thus affect the CDNC-$cwSO_4^=$ relationship. Although numerous modeling studies have indicated the importance of APOS in the cycling of atmospheric sulfur, there have been relatively few estimates of the effectiveness of this mechanism from measurements. One of the reasons for this is the difficulty of making sufficiently accurate measurements [*Kelly et al.*, 1989]. *Hegg and Hobbs* [1986] indicated that an average of 60% of the $cwSO_4^=$ in samples collected in the western United States was attributable to APOS. Measurements by *Husain* [1989] in the northeastern United States showed this process to contribute <30% of the $cwSO_4^=$. Measurements pertaining directly to the present studies suggest the contribution of APOS was ≤50% during the summer of 1982 [*Leaitch et al.*, 1986*a*], very low (probably <10%) during the winter of 1984 [*Strapp et al.*, 1988], and ≤40% during the summer of 1988 [*Leaitch et al.*, 1991*a*]. APOS is apparently less significant to these observations during the wintertime and for many of the stratiform cloud samples is not considered important. For the cumuliform cloud, which is primarily summertime data, allowance for APOS will reduce the $cwSO_4^=$. Whether the sensitivity of the CDNC to $cwSO_4^=$ will change depends on how APOS varies with absolute concentration, and it is unclear whether the contributions of APOS are generally more or less important for higher $cwSO_4^=$.

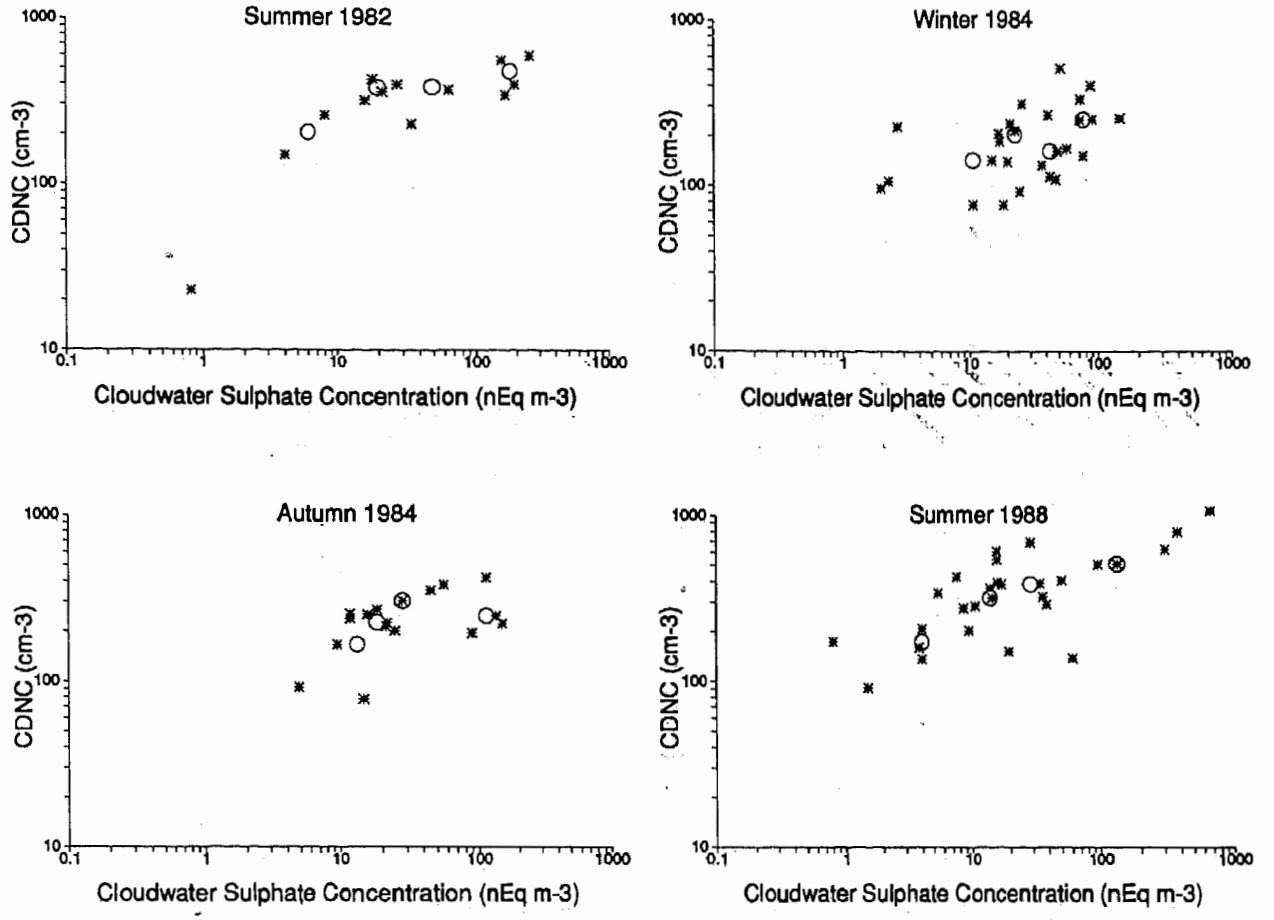

Fig. 2. CDNC plotted as a function of $cwSO_4^=$ for each of the four studies discussed. The circles represent the median CDNC for each $cwSO_4^=$ quartile.

The underlying assumption here is that the LWC is invariant with respect to the sulfate concentration. In Figure 7 the LWC are plotted against the $cwSO_4^=$, with the median LWC for each $cwSO_4^=$ quartile also included. These data do not indicate a dependence of the LWC on the $cwSO_4^=$ and therefore support the above assumption.

Equations (1) and (2) define relationships between CDNC and pollution as observed for this region: they should not be viewed as universal. It may be that these equations have some global application; however, there are too many uncertainties involved in such an extension at the present. This is an important area for further investigation.

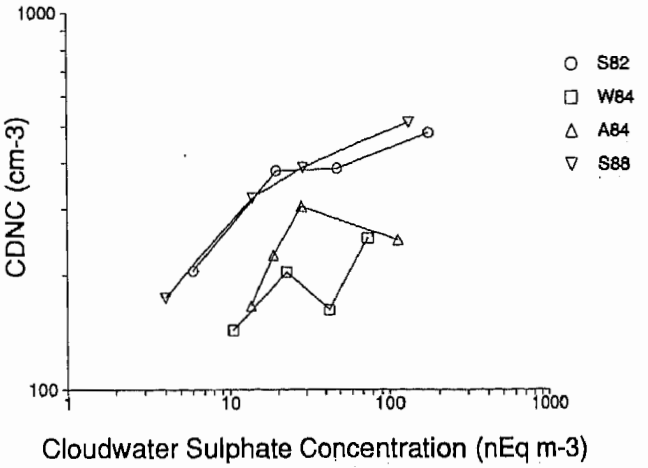

Fig. 3. The median CDNC for each $cwSO_4^=$ quartile plotted as a function of the quartile median $cwSO_4^=$ for each study.

CLEAN-AIR CDNC

Any assessment of the impact of increasing CDNC due to anthropogenic emissions on cloud albedo requires knowledge of the background CDNC (i.e., the typical CDNC present in the atmosphere when anthropogenic sources are absent). For the northern hemisphere it may be impossible to find regions which are typically free from some contribution by anthropogenic pollution. In the absence of background conditions, clean-air conditions for eastern North America are considered here. These clean air conditions are assumed to be present when the $cwSO_4^=$ and $cwNO_3^-$ are similar to measurements of these species in the air in remote areas of the globe. A potential clean air data set is identified below and the accompanying $cwSO_4^=$ and $cwNO_3^-$ are then compared with concentrations of particulate sulfate ($p - SO_4^=$) and particulate nitrate ($p - NO_3^-$) plus HNO_3 as reported for remote environments.

The modes of both the $cwSO_4^=$ and $cwNO_3^-$ distributions in Figure 1 are coincident with the 25th percentile, and the distributions drop sharply for concentrations below the

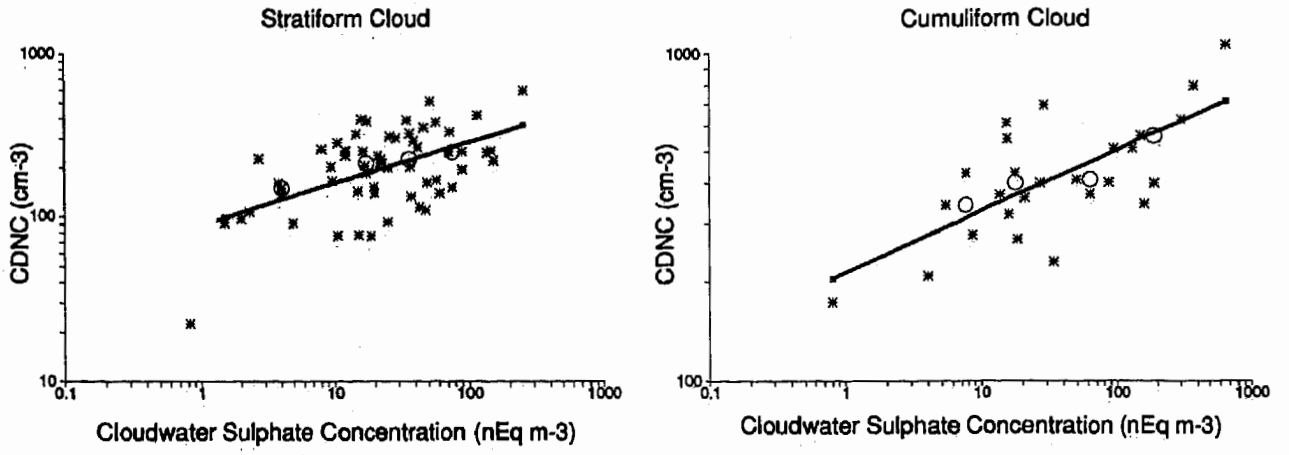

Fig. 4. The CDNC plotted as a function of the $cwSO_4^=$ for the data split between stratiform and cumuliform cloud. The circles represent the median CDNC for each $cwSO_4^=$ quartile and the solid lines represent regressions of the bulk data.

modes. For increasing concentrations above the modes, both distributions appear to decay gradually. Below the modes the observation frequency is more constant with changing concentration. These distributions appear to be unique relative to the LWC and CDNC distributions and may not be lognormal. If CDNC, LWC, as well as sulfate and nitrate were distributed purely randomly in the atmosphere, a Poisson distribution of their concentrations would be expected. If, however, there are multiplicative processes underlying their change, a lognormal distribution might be expected [*Aitchison and Brown*, 1969]. This behavior does not seem to be an artifact of the application of the LWC to the measurements, since the LWC appears to be lognormally distributed. It is probable that it is associated with precipitation scavenging and changing air trajectory bringing highly varying levels of sulfates and nitrates into the study region [e.g., *Isaac et al.*, 1990].

Regardless of the nature of the distributions the concentrations below the modes of the $cwSO_4^=$ and $cwNO_3^-$ distributions represent the lower 25% of the distribution concentrations and are the samples collected in the cleanest air. A subset of these samples is considered for the clean-air definition. The subset consists only of the samples with both $cwSO_4^=$ and $cwNO_3^-$ within the 25th percentile (i.e., $cwSO_4^=$ < 15 nEq m^{-3} and $cwNO_3^- < 7.4$ nEq m^{-3}); this eliminates strong anthropogenic contributions which might be influencing only one of the major anions. Details of this subset are summarized in Table 2 and are shown in Figure 1 by the shaded bars. The $cwSO_4^=$ range in these samples is 0.8–14 nEq m^{-3} and the median is 4.0 nEq m^{-3}. The $cwNO_3^-$ range is 0.6–5.3 nEq m^{-3} and the median is 2.6 nEq m^{-3}.

A selection of particulate sulfate concentrations ($p - SO_4^=$) measured at or over remote areas of North America and the Pacific Ocean are given in Table 3. The $p - SO_4^=$ for the northern hemisphere range up to about 65 nEq m^{-3}, but more typical values are in the 4–16 nEq m^{-3} range. Over the southern hemisphere the concentrations range up to about 9 nEq m^{-3} and are typically 2–7 nEq m^{-3} The $cwSO_4^=$ from Table 2 tend to be slightly lower than the northern hemisphere $p - SO_4^=$ and more similar to those in remote regions of the southern hemisphere. It should be noted that the comparison between $cwSO_4^=$ and $p - SO_4^=$ is affected by vertical gradients, incomplete nucleation scavenging, and APOS.

A selection of average particulate nitrate concentrations (i.e., $p - NO_3^-$) plus HNO_3 concentrations measured over remote areas of the globe is given in Table 4. These two nitrate species are the most common soluble nitrate species

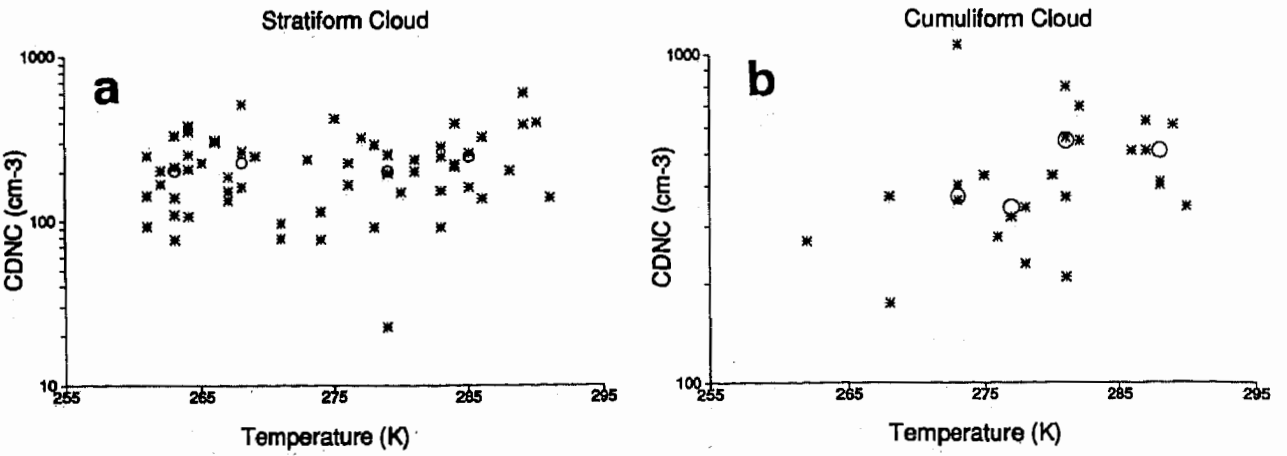

Fig. 5. The CDNC plotted as a function of the cloud temperature for the data split between stratiform and cumuliform cloud. The circles represent the median CDNC for each temperature quartile.

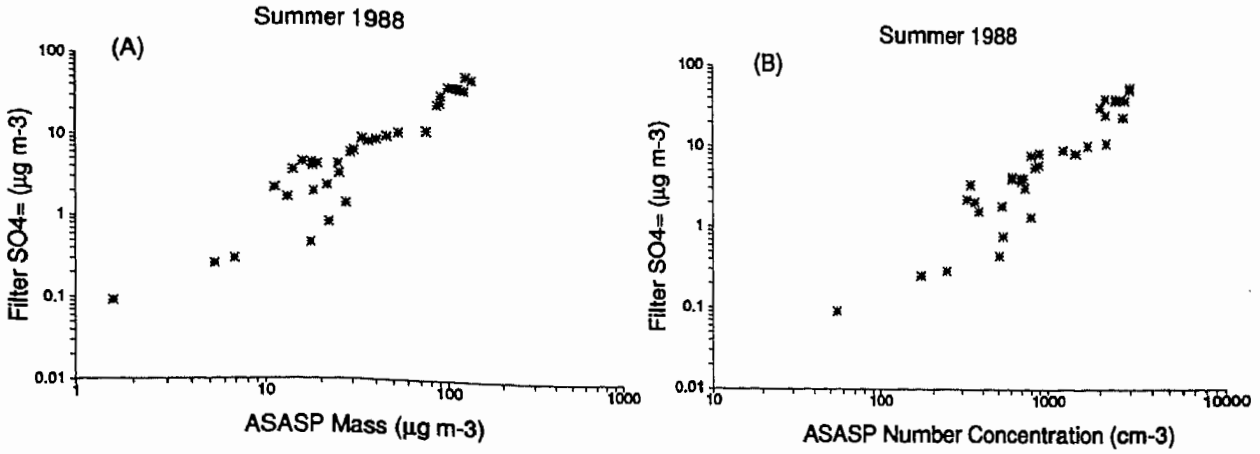

Fig. 6. (*a*) Airborne filter sulfate concentration plotted as a function of estimated aerosol mass in the 0.17–2 μm size range, assuming a mass density of 2 *g* cm^{-3}. (*b*) Airborne filter sulfate concentration plotted as a function of aerosol number concentration in the 0.17–2 μm size range.

in the atmosphere and are the greatest contributors of nitrate in cloud water. Most of the values in Table 4 fall in the 0–7 nEq m^{-3} range. Recent model results for surface HNO_3 concentrations in remote regions suggest variations of about 0.4–4 nEq m^{-3} [*Penner et al.*, 1991]. The range of $cwNO_3^-$ in Table 2 is very similar to both the measured and the modelled air nitrate concentrations reported by others.

The similarities of the $cwSO_4^=$ and $cwNO_3^-$ in Table 2 to those of $p - SO_4^=$ and $p - NO_3^-$ plus HNO_3 in Tables 3 and 4, respectively, confirm them as clean-air samples according to the above definition. As a further test, the samples in Table 2 are compared against the background $SO_4^=$ concentrations in precipitation suggested by *Vong* [1990]. His estimates, for precipitation collected on the west coast of North America, range between 2 μEq l^{-1} and 16 μEq l^{-1}. Vong suggests the LWC of the precipitating systems to have been 0.5–1.0 g m^{-3}. Using the conservative value of 0.5 g m^{-3}, then Vong's results give comparable $SO_4^=$ concentrations of 1–8 nEq m^{-3}. Ten of the 16 $cwSO_4^=$ in Table 2 are within the range given by Vong for this LWC, and all are within Vong's range if a LWC of 1 g m^{-3} is assumed. Thus the data in Table 2 are consistent with remote precipitation $SO_4^=$ concentrations as well as remote air $SO_4^=$ concentrations.

Assuming sulfate and nitrate are the major anthropogenic species affecting cloud condensation nuclei, then the CDNC in Table 2 represent a subset of clean-air CDNC for this region. In Table 5 the median, mean, and geometric mean cloud data are summarized for all samples and for the clean-air samples for each of all cloud, stratiform-only cloud, and cumuliform-only cloud. Note that in most cases the corresponding geometric mean and median CDNC are closer, suggesting the distributions may be lognormal. The clean-air median CDNC for all cloud (160 ± 40 cm^{-3}; the error is the sum of the experimental plus standard errors) is substantially lower than the all median CDNC for all cloud (250 ± 50 cm^{-3}). This is also true if means or geometric means are compared. Using the Student *t* test and assuming a lognormal distribution of the CDNC, the hypothesis that the "all-cloud clean-air" median CDNC is the same as the "all-cloud all" CDNC can be rejected at the 99.5% confidence level. The confidence levels are 98 and 95% for the stratiform-only and cumuliform-only cloud, respectively.

The clean-air CDNC are not definitive background measurements: some of the data are influenced by sulfate above background concentrations, and the data set is small. Still, these clean-air CDNC are approaching background concentrations. A few measurements of CDNC reported for more

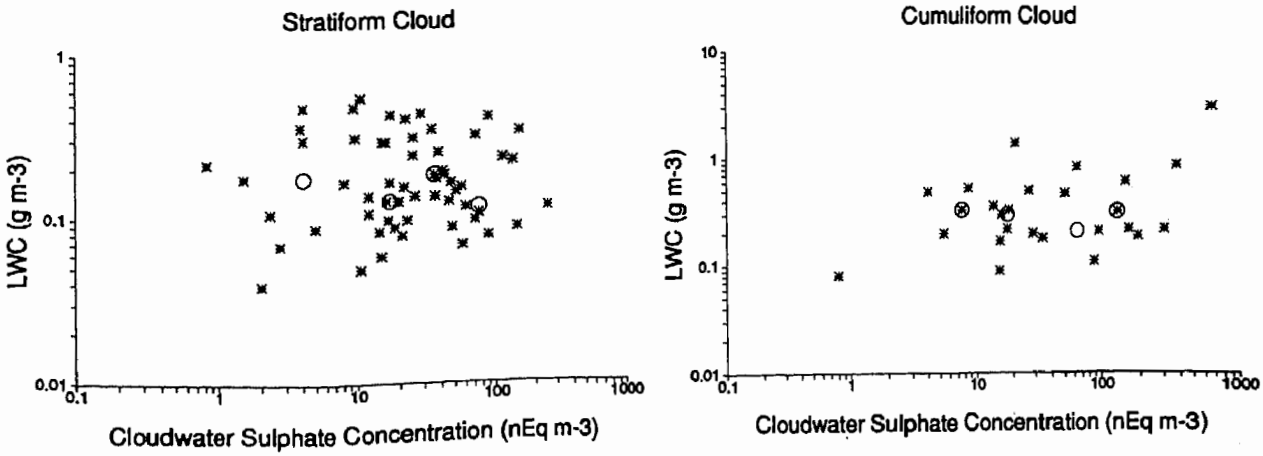

Fig. 7. The LWC plotted as a function of $cwSO_4^=$ for the data split between stratiform and cumuliform cloud. The circles represent represent the median LWC for each $cwSO_4^=$ quartile.

TABLE 2. Clean-Air Cloud Sample Data

Date	Altitude km	LWC g m^{-3}	CDNC cm^{-3}	Temperature °C	nEq m^{-3} $cwSO_4^{=}$	nEq m^{-3} $cwNO_3^{-}$
Jan. 12, 1982	1.1	0.22	15–30	5.7	0.8	0.6
Feb. 8, 1984	1.5	0.05	77	−9.6	10.5	4.9
Feb. 9, 1984	2.0	0.11	107	−8.8	2.3	0.9
Feb. 17, 1984	2.5	0.04	97	−2.3	2.0	1.1
Feb. 19, 1984	3.1	0.07	228	−7.6	2.7	1.3
Oct. 29, 1984	2.7	0.09	92	5.0	4.9	4.1
Nov. 9, 1984	1.7	0.14	254	5.7	12.0	2.0
Nov. 10, 1984	2.4	0.32	166	3.0	9.6	3.8
Aug. 15, 1988	1.5	0.37	161	12.3	3.8	3.5
Aug. 17, 1988	1.5	0.51	204	14.6	9.4	5.3
Aug. 17, 1988	1.8	0.31	137	13.3	4.0	3.6
Aug. 17, 1988	2.1	0.18	92	9.7	1.5	1.0
Aug. 26, 1988	1.4	0.12	209	8.1	4.0	0.8
Aug. 26, 1988	1.7	0.36	369	7.6	13.8	3.6
Aug. 26, 1988	2.1	0.53	278	3.4	8.6	3.3
Aug. 29, 1988	2.6	0.08	174	−4.5	0.8	1.1
25%	1.5	0.09	95	−3.4	2.2	1.1
Median	1.9	0.16	160	5.4	4.0	2.7
75%	2.5	0.34	220	8.9	9.5	3.7

remote locations are considered for comparison. The range of CDNC measured in cumulus at remote locations is 50–400 cm^{-3} [e.g., *Pruppacher and Klett*, 1980; *Paluch and Knight*, 1984; *Ryan et al.*, 1985; *Raga et al.*, 1988], with more typical values of 100–300 cm^{-3}. Thus the present clean-air cumulus samples are in the same range as other measurements of CDNC in remote cumulus. *Radke et al.* [1989] show CDNC in North Pacific stratocumulus between 30 and 50 cm^{-3} outside of ship tracks; *Albrecht* [1989] shows average CDNC between 30 and 100 cm^{-3} for stratiform cloud over the North Pacific; and *Isaac* [1991] gives median CDNC between 72 and 92 cm^{-3} for stratiform cloud over the Atlantic off the east coast of Canada. Thus measurements in marine stratiform cloud suggest somewhat lower concentrations than indicated for most of the clean-air continental stratiform cloud here.

CLIMATIC IMPLICATIONS

A few estimates of the effect of the direct increase of cloud albedo, due to increased CDNC, on atmospheric cooling have been made [e.g., *Twomey et al.*, 1984; *Wigley*, 1989]; however, it is still useful to make some first-order estimates using the present data, in order to place these results in perspective.

The effect of increased CDNC on the planetary albedo (A_p) can be estimated in the following manner. Considering multiple reflections between one cloud layer and the ground but neglecting scattering by other constituents, as well as all absorption, the planetary albedo can be estimated from the following:

$$A_p = A_c + [(1 - A_c)^2 A_s]/(1 - A_c A_s) \quad (3)$$

where A_s and A_c are the surface and cloud albedo, respectively. The cloud albedo is related to the CDNC through the cloud optical thickness (τ). The following relationship between A_c and τ from *Lacis and Hansen* [1974] is used:

$$A_c = \tau/(7.7 + \tau). \quad (4)$$

Further, assuming the scattering efficiency of cloud droplets is 2 [e.g., *Hansen and Travis*, 1974], then τ is related to the CDNC as follows:

$$\tau = H\{9\pi\ \mathrm{LWC}^2\ \mathrm{CDNC}/(2\rho^2)\}^{1/3} \quad (5)$$

where ρ is the density of water and H refers to the depth of the cloud layer. The manner in which (5) is used here assumes that the size distribution of the cloud droplets is constant, independent of height in the cloud. This assumption will have more importance for clouds of greater vertical extent where differences in droplet sizes are greater. For stratiform cloud the importance of this problem is relatively low [*Fouquart et al.*, 1990]. In terms of the present data the sampling was not designed to favor either cloud base or cloud tops and on average is a good representation of the cloud. A discussion of the uncertainty involved in the application of this simple approach can be found by Fou-

TABLE 3. Particulate Sulfate Measurements at Remote Locations

Location	p-$SO_4^{=}$ Concentration, nEq m^{-3}	Reference
Remote northern hemisphere	0.65–45.2, median 7.4	*Schwartz* [1988]
Remote southern hemisphere	0.3–5.3, median 2.1	*Schwartz* [1988]
North Pacific	9.4–14.6	*Savoie and Prospero* [1989]
South Pacific	4.8–8.8	*Savoie and Prospero* [1989]
Pacific, free Troposphere	4.7, average	*Huebert and Lazrus* [1980*b*]
Remote North America, free Troposphere	7.8, average	*Huebert and Lazrus* [1980*b*]
Northeast Pacific, <6 km	4–16	*Andreae et al.* [1988]
South Pacific, <5 km	1–5	*Berresheim et al.*, [1990]

TABLE 4. Particulate Nitrate and HNO_3 Measurements at Remote Locations

Location	$p\text{-}NO_3^-$ + HNO_3 Concentration, nEq m^{-3}	Reference
North Pacific, surface	2.5–7.0	*Prospero and Savoie* [1989]
South Pacific, ground	1.7–3.4	*Prospero and Savoie* [1989]
North Pacific, free troposphere boundary layer	<0.8–13, median 3.7 (HNO_3 only) 1.2–4.3, median 2.5 (HNO_3 only)	*LeBel et al.* [1990]
Western United States, free troposphere	<0.8–23, median 2.8 (HNO_3 only)	*LeBel et al.* [1990]
North Atlantic, surface	5.1 + 0.5, averages	*Qi et al.* [1990]
South Atlantic, surface	4.9 + 0.4, averages	*Qi et al.* [1990]
Pacific and remote North America	3.7–5.0, (HNO_3 only)	*Huebert and Lazrus* [1980*b*]
Remote North America	3, ($p\text{-}NO_3^-$ only)	*Huebert and Lazrus* [1980*a*]
North Pacific	2–4, ($p\text{-}NO_3^-$ only)	*Huebert and Lazrus* [1980*a*]
North Pacific	0–4, ($p\text{-}NO_3^-$ only)	*Berresheim et al.* [1990]

quart et al. For an increase in the CDNC (i.e., CDNC′) and a constant LWC, (4) becomes

$$A_c' = \tau'/[7.7 + \tau'] \quad (6)$$

where

$$\tau' = (\text{CDNC}'/\text{CDNC})^{1/3}\tau. \quad (7)$$

The fractional change in A_p (i.e., $[A_p' - A_p]/A_p$ or $\Delta A_p/A_p$) due to a change in A_c (i.e., $A_c' - A_c$), is derived from (3), (6), and (7), assuming a constant LWC. This is done here for a CDNC of 160 cm^{-3} (i.e., median clean-air CDNC: Table 5) and a CDNC′ of 250 cm^{-3} (i.e., the region's median CDNC: Table 5). The results of this analysis for 100% cloud cover are shown in Table 6 for a number of A_s and A_c to cover the range of surfaces and cloud optical thicknesses which may be encountered. They indicate increases in planetary albedo of a few percent depending on the underlying surface and the optical depth of the cloud.

The fractional coverage of cumuliform and stratiform cloud below 3 km (i.e., typical measurement altitude) is established from *Lelieveld et al.* [1989] at 20–30%, weighted by season, for continental regions between 40° and 50°N (i.e., the region of the present measurements). This estimate conforms with the cloud coverage distributions for low-to-middle altitude mid-latitudinal cloud presented by *Rossow and Lacis* [1990]. From the data of Lelieveld et al. it is estimated that the average depth of cloud is 1.0 km, and the corresponding cloud albedo is about 0.85. From the Rossow and Lacis analysis it is estimated that the typical optical thickness for low and middle cloud at mid-latitudes is about 20–25, which equates to an A_c of 0.72–0.76 using (4). Considering these estimates, an A_c of 0.80 is assumed for the present purpose. Further, an A_s of 20% is assumed, which is high according to the discussion of Rossow and Lacis, but conservative in terms of the present estimate. For these A_c and A_s the $\Delta A_p/A_p$ from Table 6 is about 2.5% for 100% cloud cover and 0.5–0.75% for a cloud fraction of 20–30%. According to *Ramanathan* [1988], an increase in the planetary albedo of 0.5% is sufficient to halve a warming of 4 W

TABLE 5. Median and Mean Cloud Data for Stratiform Cloud, Cumuliform Cloud, and All Cloud

	No.*	Altitude km	LWC g m^{-3}	CDNC cm^{-3}	Temperature °C	nEq m^{-3} $cwSO_4^=$	nEq m^{-3} $cwNO_3^-$
			Stratiform Cloud				
All median	(61)	1.5	0.16	210	1.8	22	12
All mean	(61)	1.6	0.20	220	1.4	38	28
All geometric mean	(61)	1.5	0.17	200	1.1	22	13
Clean-air median	(12)	1.9	0.16	120	5.4	3.9	2.8
Clean-air mean	(12)	2.0	0.20	140	3.4	5.3	2.7
Clean-air geometric mean	(12)	1.9	0.15	120	3.3	3.9	2.1
			Cumuliform Cloud				
All median	(31)	2.1	0.32	400	7.6	35	34
All mean	(31)	2.3	0.47	450	6.4	89	67
All geometric mean	(31)	2.1	0.34	410	3.6	37	27
Clean-air median	(4)	1.9	0.24	240	5.5	6.3	2.2
Clean-air mean	(4)	2.0	0.27	260	3.7	6.8	2.2
Clean-air geometric mean	(4)	1.9	0.21	250	6.3	4.4	1.8
			All Cloud				
All median	(92)	1.8	0.19	250	4.5	25	15
All mean	(92)	1.8	0.29	290	3.1	56	41
All geometric mean	(92)	1.8	0.21	250	2.8	26	16
Clean-air median	(16)	1.9	0.16	160	5.4	4.0	2.7
Clean-air mean	(16)	2.0	0.22	170	3.5	5.7	2.6
Clean-air geometric mean	(16)	1.9	0.16	140	3.4	4.0	2.0

*Indicates number of samples. For CDNC only, the number is 59 for stratiform cloud, 26 for cumuliform cloud, and 85 for all cloud.

TABLE 6. Percentage Change in Planetary Albedo for an Increase in CDNC From 160 to 250 cm^{-3}

					Percent	
A_s	A_c	A_p	A'_c	A'_p	ΔA_p	$\Delta A_p/A_p$
1.0	0.9	1.0	0.913	1.0	0.0000	0
0.8	0.9	0.929	0.913	0.935	0.0067	0.72
0.6	0.9	0.913	0.913	0.923	0.0097	1.1
0.4	0.9	0.906	0.913	0.917	0.0112	1.2
0.2	0.9	0.902	0.913	0.914	0.0120	1.3
0.0	0.9	0.900	0.913	0.913	0.0126	1.4
1.0	0.8	1.0	0.823	1.0	0.0000	0
0.8	0.8	0.889	0.823	0.896	0.0074	0.83
0.6	0.8	0.846	0.823	0.860	0.0138	1.6
0.4	0.8	0.824	0.823	0.841	0.0179	2.2
0.2	0.8	0.810	0.823	0.830	0.0207	2.6
0.0	0.8	0.800	0.823	0.823	0.0227	2.8
1.0	0.6	1.0	0.635	1.0	0.0000	0
0.8	0.6	0.846	0.635	0.852	0.0055	0.65
0.6	0.6	0.750	0.635	0.764	0.0142	1.9
0.4	0.6	0.684	0.635	0.707	0.0223	3.3
0.2	0.6	0.636	0.635	0.665	0.0293	4.4
0.0	0.6	0.600	0.635	0.635	0.0351	5.9
1.0	0.4	1.0	0.436	1.0	0.0000	0
0.8	0.4	0.824	0.436	0.827	0.0033	0.40
0.6	0.4	0.684	0.436	0.695	0.0103	1.5
0.4	0.4	0.571	0.436	0.590	0.0188	3.3
0.2	0.4	0.478	0.436	0.506	0.0276	5.8
0.0	0.4	0.400	0.436	0.436	0.0362	9.0
1.0	0.2	1.0	0.225	1.0	0.0000	0
0.8	0.2	0.810	0.225	0.811	0.0014	0.18
0.6	0.2	0.636	0.225	0.642	0.0052	0.82
0.4	0.2	0.478	0.225	0.489	0.0107	2.2
0.2	0.2	0.333	0.225	0.351	0.0174	5.2
0.0	0.2	0.200	0.225	0.225	0.0249	12

$\Delta A_p/A_p$ is zero for all $A_c = 0$ and 1.

m^{-2} due to a doubling of CO_2. Thus if no other cloud-related factors are countering the increase of cloud albedo due to increased CDNC, the latter is contributing a cooling of 2–3 W m^{-2} over eastern North America. Ice particles in mixed phase clouds and higher ice clouds (e.g., cirrus) will counter this effect (it is assumed that ice particles are unperturbed by the effects of pollution; however, this is largely an unknown area) by reducing the intensity of the light available for reflection by the liquid water cloud. Therefore this estimate of the effect must be considered an upper limit.

Since the anthropogenic influence is very strong over eastern North America, the present-day climatic forcing due to increased cloud albedo from increased CDNC for the entire northern hemisphere must be well below the above estimates. *Wigley* [1989] has shown that forcing much weaker than 2–3 W m^{-2} is consistent with observed trends in the northern hemisphere temperature relative to the southern hemisphere temperature for the last century. He suggests the effect of sulfur pollution on climate may already be important on the global scale. An alternative or at least supportive explanation based on aerosol scattering has recently been proposed [*Charlson et al.*, 1990].

Slingo [1990] recently used three versions of the National Center for Atmospheric Research (NCAR) community climate model to describe the changes in any of three quantities pertaining to low clouds necessary to offset the radiative forcing due to a doubling of CO_2 in the atmosphere. One of these quantities is r/CDNC, where r is the effective radius of the cloud droplets; a constant LWC is assumed. In Table 7 the factors by which r/CDNC must be multiplied to counteract the warming by CO_2 doubling as given by Slingo are compared with the present observations. An estimate of the present change in r/CDNC for eastern North America was obtained from the present data by taking the typical LWC and CDNC for each of the stratiform and cumuliform cloud cases and determining the mean volume radius (MVR) of the cloud droplets. The MVR and CDNC were determined for each cloud type as well as for the clean-air and complete data sets, and the difference in these quantities between the clean-air and the complete set of observations is shown in Table 7. The MVR/CDNC are very close to Slingo's r/CDNC, particularly for the cloud versions 2 and 3. The difference between the effective radius and the MVR can be expressed as

$$r = (k/0.62)\ \mathrm{MVR} \tag{8}$$

where k is a constant for a particular size distribution of cloud droplets, typically about 0.7 μm, but with a range of

TABLE 7. Comparison of the r/CDNC Factors Necessary to Counteract the Net Radiation Changes Due to CO_2 Doubling From *Slingo* [1990] With the Present Observations

r*/CDNC From Slingo Time of Year		CCM Cloud Version 1	2	3
January		0.81/1.90	0.86/1.60	0.85/1.65
July		0.79/2.02	0.85/1.65	0.83/1.72
Present Results: Clean-Air Assumptions	LWC g m^{-3}	$CDNC_c$† cm^{-3}	MVR_c μm	
Stratiform	0.16	120	6.8	
Cumuliform	0.32	240	6.8	
Eastern North America	LWC g m^{-3}	$CDNC_a$ cm^{-3}	MVR_a μm	$(MVR_a/MVR_c)/(CDNC_a/CDNC_c)$
Stratiform	0.16	210	5.7	0.84/1,75
Cumuliform	0.32	400	5.8	0.85/1.67

CCM is community climate model, and MVR is mean volume radius.
*The value of r is the effective radius.
†CDNC is the median value from Table 5.

0.62–1.02 μm [see *Fouquart et al.*, 1990]. The data of *Tsonis et al.* [1987] indicate this range is indeed appropriate for the present data. As a result the factors indicated by the present data in Table 7 are exacerbated by the conversion to effective radius. In any case, Table 7 gives further cause to suggest that the pollution over this region is already sufficient to offset the effect of CO_2 doubling via cloud albedo increases over eastern North America. Although this is more dramatic than the effect suggested from comparison with the discussion of *Ramanathan* [1988], the net reduction in upward radiation at the top of the atmosphere in Slingo's model due to CO_2 doubling is about 2.5 W m^{-2}, which is consistent in magnitude with estimates made here for cloud albedo change using the present rudimentary model.

Albrecht [1989] and *Liou and Ou* [1989] have considered the effect of changing cloud microphysics on precipitation efficiency. Liou and Ou showed that a reduction in the mean droplet radius of about 0.5 μm could cool the atmosphere and offset warming due to CO_2 doubling. This arises from increased cloud reflectance due to increased LWC, which is the result of a reduction in precipitation efficiency due to the smaller droplet sizes. In Table 7 the present data indicate a reduction in droplet radii of about 1 μm for eastern North America, indicating that this process is also a factor which must be considered.

Finally, this discussion has concentrated on the relationship between sulfate and CDNC. While it is widely accepted that sulfate aerosols are major sources of cloud nuclei, a relationship between CDNC and $cwNO_3^-$ very similar to that shown in Figure 4 is also observed in the present data [*Leaitch et al.*, 1991*b*]. This result may be fortuitous, because nitrate concentrations generally correspond with sulfate concentrations in the atmosphere over the large range of concentrations discussed here. On the other hand, some nitrate species may make significant contributions to the CDNC. It is important to make the distinction that the effect of sulfate on CDNC described here may include pollutants other than sulfate and should be regarded as a net effect of pollution, with sulfate serving as the measure of the pollution.

CONCLUSIONS

Measurements of the $cwSO_4^=$ and $cwNO_3^-$ in approximately 400 cloud water samples have been used with coincident measurements of CDNC and LWC to examine the relationship between CDNC and anthropogenic pollution, where sulfate is considered as the primary measure of the latter. The observations were made during four field studies carried out in the summer of 1982, the winter of early 1984, the autumn of 1984, and the summer of 1988 over a portion of eastern North America. The number of observations has been summarized to 92 through averaging for duplication. Positive linear regressions between log (CDNC) and log ($cwSO_4^=$) were determined for both stratiform and cumuliform cloud. Because of the number of factors affecting the CDNC the coefficients of determination are only 0.30 and 0.49 for the respective cloud types. The LWC is found to be relatively invariant with the $cwSO_4^=$.

The observed range of CDNC for the study region is 20–600 cm^{-3} (median is 210 cm^{-3}) for stratiform clouds and 170–1100 cm^{-3} (median is 400 cm^{-3}) for cumuliform clouds. The median CDNC for all sampled clouds is 250 cm^{-3}.

CDNC were determined for clean-air conditions. The latter was defined as cases for which the concentrations of both cloud water sulfate and cloud water nitrate are comparable to aerosol sulfate concentrations and aerosol nitrate plus HNO_3 concentrations reported for remote regions of the globe. For the clean-air clouds the observed range of CDNC for the study region is 20–250 cm^{-3} (median is 120 cm^{-3}) for stratiform clouds and 170–370 cm^{-3} (median is 240 cm^{-3}) for cumuliform clouds. The median CDNC is 160 cm^{-3} for all sampled clean-air clouds. It is cautioned that these clean-air values are not necessarily "background" CDNC for eastern North America. The hypothesis that the clean-air median CDNC are not different from the median CDNC for all samples can be rejected at confidence levels of 95, 98, and 99.5% for cumuliform, stratiform, and all cloud, respectively.

The median CDNC for all cloud represents an increase of 56% over the clean-air median CDNC. The corresponding potential increases in cloud albedo and the accompanying radiative forcing due to this increase were estimated using a rudimentary model. The increases are quite dramatic for this area of North America, indicating a cooling of up to 2–3 W m^{-2} due to the increased reflection of short wave radiation.

These observations offer support for the importance of the issue of pollution, cloud microphysics, and cloud albedo. They should underscore the need for more global measurements of cloud microphysics and chemistry, particularly in remote regions, to help address the issue of global climate change.

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C. M. Banic, G. A. Isaac, W. R. Leaitch, J. W. Strapp, and H. A. Wiebe, Atmospheric Environment Service, Environment CANADA, 4905 Dufferin Street, Downsview, Ontario, Canada M3H 5T4.

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