Measurements of Ultragiant Aerosol Particles in the Atmosphere from the Small Cumulus Microphysics Study

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ABSTRACT

Ultragiant aerosol particles (UGA) are potentially important for warm rain formation because of their ability to initiate coalescence immediately upon entering a cloud, so it is desirable to obtain local estimates during any field campaign that studies warm rain. Estimates of UGA in clear air from a one-dimensional optical array probe averaged over long time periods from the Small Cumulus Microphysics Study have been published in the literature, but further analysis and comparisons to other probes, presented here, show that the data on which these estimates were based were probably contaminated by noise. A possible explanation for the noise in the probe is given, as are new upper limits, based on few or no particles detected by a two-dimensional optical array probe.

1. Introduction

Giant (2–20-μm diameter) and ultragiant (>20-μm diameter) aerosol particles are of particular interest in the warm rain process. Deliquesced, soluble giant particles and any ultragiant particles[1] can enter a cloud and initiate coalescence quite quickly; if they are larger than 50-μm diameter, their collection efficiencies are greater than 20%. Although their importance to warm rain production has not been firmly established, numerous modeling studies have shown that they can produce radar echoes ≥10 dB*Z* in less than twenty minutes (e.g., Ochs and Semonin 1979; Johnson 1982), and more recent studies that incorporated radar observations have discussed their potential for dominating the early radar echoes (Lasher-Trapp et al. 2001; Knight et al. 2002).

Woodcock (1952, 1953) first presented size distributions of giant salt particles (deliquesced at 99% relative humidity) collected off the coasts of Hawaii and Florida. Particles were collected by impaction on coated glass slides extended from an aircraft and were later sized under a microscope in a temperature- and humidity-controlled environment. Other studies have documented giant and ultragiant aerosol particles, both soluble and insoluble, over continents and oceans (e.g., Reitan and Braham 1954; Okita 1955; Nelson and Gokhale 1968; Noll and Pilat 1971; Mészáros and Vissy 1974; Johnson 1976), with maximum concentrations ranging from over 10^{-2} cm^{-3} at the smaller sizes to less than 10^{-8} cm^{-3} at the larger sizes. These studies have shown that concentrations of these particles in the atmosphere may vary by several orders of magnitude,

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[1] For the duration of this note, particles with $D > 50$ μm will be called ultragiant, since the probes used here cannot distinguish the size of the original particle before it was deliquesced, or even if it has deliquesced.

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depending on meteorological processes, local sources, altitude, and numerous other factors. Thus, it appears critical to take measurements of the sizes and concentrations of ultragiant aerosol particles (UGA) during field campaigns where their influence on radar echoes and precipitation needs to be quantified.

2. Some estimates of ultragiant aerosol particles from the SCMS

a. *The SCMS*

The Small Cumulus Microphysics Study (SCMS) was a field campaign conducted near Cape Canaveral, Florida, to collect observations on small warm cumuli and the initial stages of the precipitation process. A dual-wavelength radar and three different instrumented aircraft were deployed for this purpose. The Florida coastal region can be influenced by air over the continent and ocean, depending on the prevailing wind direction. Thus, special patterns (long, overlapping ovals at low levels) were flown by the National Center for Atmospheric Research (NCAR) C130 aircraft each day of the project to characterize the aerosol in and above the boundary layer.

b. *Estimates from the 260X probe*

The 260X is a one-dimensional optical array probe (manufactured by Particle Measuring Systems, Inc.) that records the cross-flight dimension of shadows of particles passing through its laser beam. For the SCMS, the probe was modified to measure particle sizes 51–1054-μm diameter at 17-μm resolution. The size-dependent sample volumes range from ~30 to 4000 cm^3 (100 m)$^{-1}$ of flight. (These values were determined by calibrations conducted after the SCMS.) This probe is thus capable of providing estimates of the number of UGA from the long periods of clear-air flight described above. For example, if the UGA concentration is ~10^{-4} cm^{-3} for 51-μm particles (~10^{-6} cm^{-3} for 204-μm particles), an averaging time of 4000 s is needed to measure this concentration with an uncertainty of ~30%, for a flight speed of 100 m s^{-1}. Some examples of size distributions computed from the SCMS 260X data are shown in Fig. 1. The size distributions appear quite realistic and are not a result of a few anomalously high values over short time intervals.

Laird et al. (2000) compiled composite size distributions of UGA over the entire duration of the SCMS using clear-air data from the 260X probe. Their analysis showed that UGA were present in number concentrations similar to the giant particles (deliquesced at 99% relative humidity) measured by Woodcock (1953) in weak-to-moderate winds off the coasts of Hawaii and Florida. Laird et al. (2000) concluded that the data were not contaminated by noise because the resulting size distribution from the particle counts was exponential, not flat, and not limited to one channel.

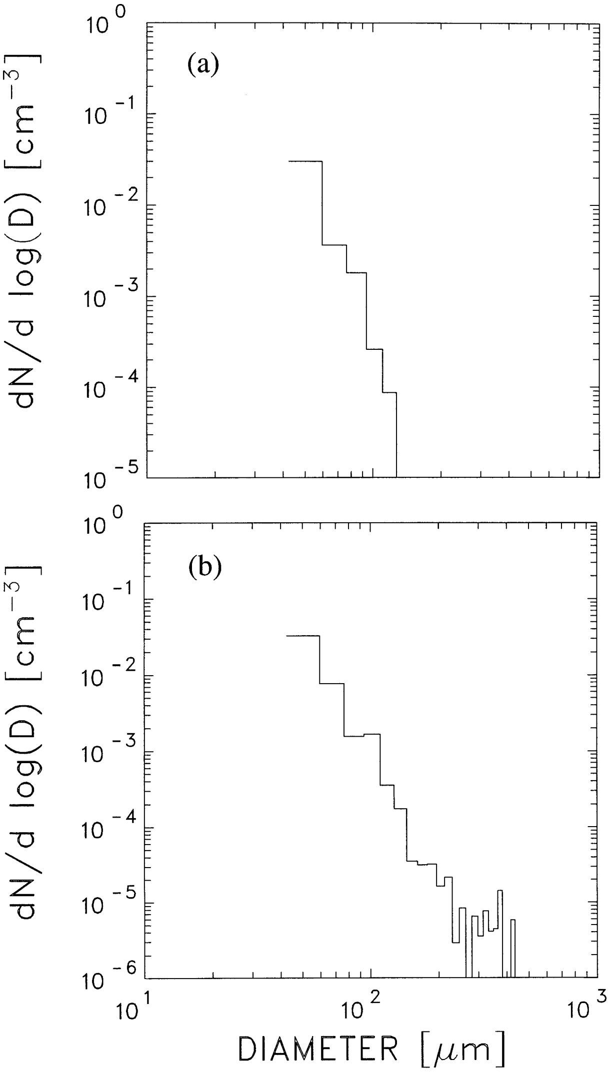

FIG. 1. Size distributions from 260X data collected in clear air for (a) 1442–1450 UTC 22 Jul 1995 and (b) 1403–1440 UTC 24 Jul 1995.

However, comparisons of the 260X data with a 2DC probe mounted on the aircraft suggest that the particles recorded by the 260X were not always real. The 2DC is a two-dimensional optical array probe (manufactured by Particle Measuring Systems, Inc.) that records two-dimensional images of particles passing through its laser. During the clear-air periods when the 260X recorded particles, very few or no images appeared on the 2DC, nor were there many instances that the probe triggered from particles too small to record images. The usable size ranges of the 260X (51–1054 μm) and 2DC (50–800 μm) overlap well, and the sample volume of the 2DC (5 L maximum for 100-m flight) is even greater than that of the 260X (3–4 L maximum for 100-m flight). In addition, the 2DC diodes are only required to be shadowed 40% when particles pass through the laser, versus the 70% shadowing required by the 260X to record a particle. Thus, when the 260X records particles, the 2DC should as well. The 2DC probe appears to be working correctly during the SCMS, because the 260X and 2DC size distributions from in-cloud penetrations agree well.

Additional evidence indicates that the 260X data are contaminated by noise. When the aerosol legs and aircraft soundings are partitioned into periods during which the aircraft was below 1500 m MSL (which included the aerosol legs) and periods during which it was above 1500 m MSL (during the sounding, with maximum height over 3 km MSL), the concentrations of UGA measured by the 260X fail to decrease with height, as expected of these large particles due to their relatively large settling velocities. Figure 2 shows size distributions measured by the 260X in clear air below and above 1500 m on three different days; the concentrations are nearly identical. Other days investigated in the same way show similar results.

Perhaps the most convincing evidence that a problem is occurring with the 260X comes from data collected toward the end of the SCMS. At this time, two short flights were conducted to compare data from the three different one-dimensional optical array probes flown on the three different aircraft during the project. For the comparison, all three probes (NCAR's 260X, the University of Wyoming's 1DC, and Météo-France's 200X) were flown on the NCAR C130. Results show that the 260X often recorded counts in clear air when both of the other 1D probes had none (Fig. 3). The 2DC, also present on the C130, also showed no images or counts from particles too small to record images.

We thus conclude that the distributions of UGA given by Laird et al. (2000) are contaminated by noise. The spurious counts recorded by the 260X are most likely not a problem in a cumulus cloud penetration because the time interval of interest is so short, but when averaged over long intervals in clear air, they dominate the measurements.

c. Estimates from the 2DC probe

Particle images from the 2DC, and lack thereof, can be used to provide some insight into potential concentrations of UGA from the boundary layer in the SCMS area. Six flights having the longest clear-air periods flown by the NCAR C130 at low altitudes are used; the time periods, altitude, relative humidity and 2DC images are listed in Table 1. Two of the legs (one being as long as 3000 s) had no 2DC images; the others had very few images. The occurrence of images does not appear to be related to the ambient relative humidity.

Because of the scarcity of images, concentrations derived directly from those images are misleading because of the poor counting statistics. In addition, it is impossible to determine if the noise affecting the 260X also may have been affecting the 2DC, creating false particle images. As a result of these uncertainties, the 2DC data listed in Table 1 are used to derive upper limits on the UGA size distributions, rather than actual measured size distributions, for UGA $50 < D < 200$ μm, at a 95% confidence level.

The computation of the upper limits for each flight proceeds as follows. It is assumed that the UGA are distributed randomly in space, so that counting of these particles by the 2DC is a Poisson process (e.g., Taylor 1982):

$$P(c) = \frac{\overline{c}^{c} e^{-\overline{c}}}{c!}, \tag{1}$$

where $P(c)$ is the probability of counting c particles during the entire sample, and $\overline{c}$ is the underlying mean number of particles over the sample volume. For each of the six cases in Table 1, c is set to the number of particles observed, and $\overline{c}$ is calculated as a function of the probability that the number of images or less will be recorded by the 2DC. The probability P in (1) is set to 0.05, and the solution for $\overline{c}$ is sought iteratively. A value of P of 0.05 has been chosen so that, at a 95% confidence level, the true underlying concentrations are not greater than those computed here. Once $\overline{c}$ is known, the concentrations within each size bin are also found in an iterative manner by requiring that their products with their sample volumes sum to equal $\overline{c}$. An additional constraint is placed on the concentrations, by requiring that the estimated concentration in each bin exponentially decreases from the previous bin, at a rate consistent with past observations.

The upper limits calculated in this manner for the six flights in Table 1 are shown in Fig. 4. The size distributions all fall within a relatively narrow region, regardless of the number of particles and the size of the accumulated sample volume. The maximum concentration of particles over the 100–200-μm size range shown is $\sim 6 \times 10^{-6}$ cm^{-3}.

Overlaid on Fig. 4 for comparison are the size distributions from Woodcock (1953), evaluated at 60% and 90% relative humidity, for data collected in weak wind (W) and tropical storm–force wind (*S*). The 60% and 90% relative humidity cases are meant to bracket the relative humidities observed for the SCMS cases listed in Table 1. It is important to note that the sizes of particles measured by Woodcock off the coast of Florida are only greater than 20-μm diameter when their deliquesced sizes at very high relative humidities are considered; otherwise they are all smaller, and undetectable by the optical array probes used for the SCMS.

The UGA project-composite distribution for 85%–90% relative humidity based on the 260X data from Laird et al. (2000) is shown in Fig. 4 as well, corrected for an error that made the concentrations an order of magnitude too low as reported in the original paper (Laird et al. 2000, corrigendum). It is quite clear that the scarcity of 2DC images implies that the concentrations of UGA present in the boundary layer are much

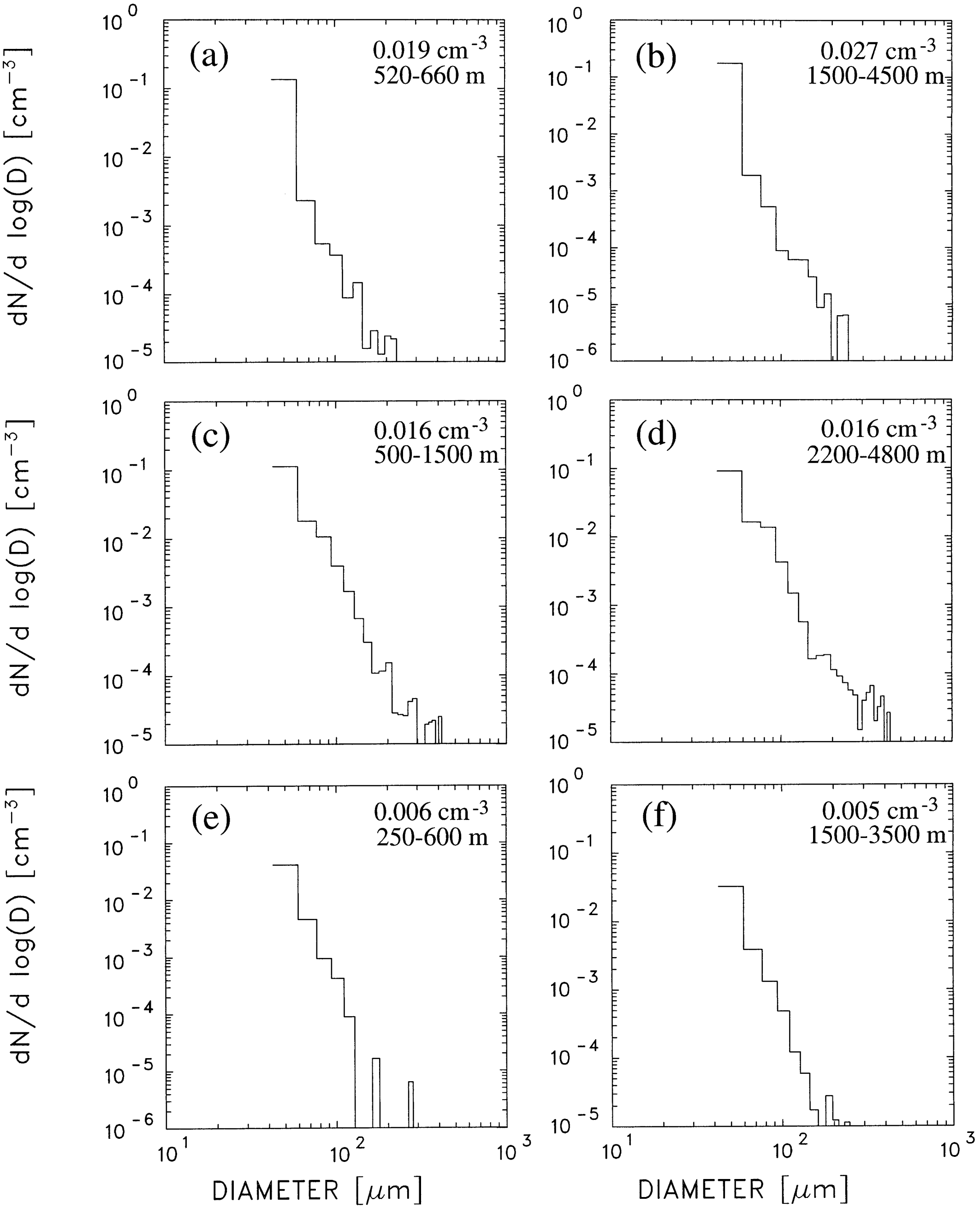

FIG. 2. Size distributions from the 260X in clear air (all times UTC): (a) 1513–1527 19 Jul, (b) 1530–1540 19 Jul, (c) 1415–1430 22 Jul, (d) 1430–1440 22 Jul, (e) 1323–1332 11 Aug, and (f) 1342–1359 11 Aug 1995. Total concentration and aircraft altitude are shown for each plot in upper right-hand corner.

less than the composite distribution derived from the 260X data, by at least three orders of magnitude. Figure 5 compares the upper limits derived from the 2DC data with the 260X distributions for some of the individual cases listed in Table 1 (the 260X was not operating on 24 July and 6 August, so those cases are not plotted). The individual 260X distributions are again at least three orders of magnitude greater than the upper limits derived from the 2DC data, and much more at the larger sizes.

3. A possible source of the 260X noise

The source of noise to the NCAR 260X data from the SCMS is at this time unknown, although it does not appear to be attributable to radio frequency interference.

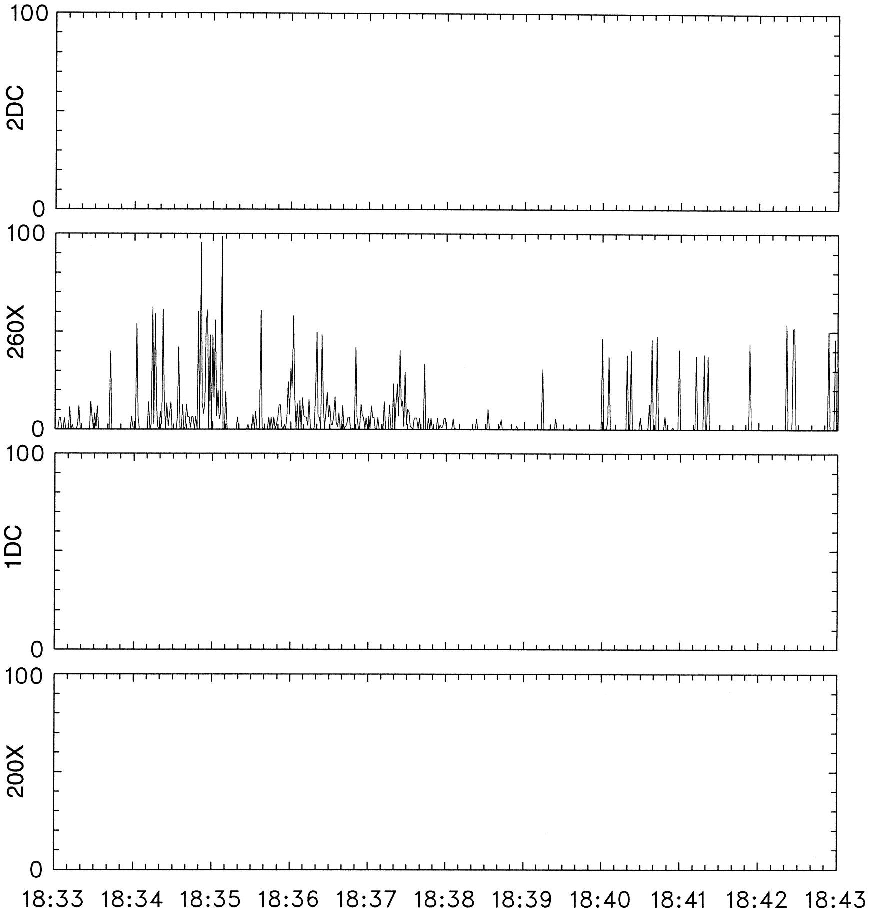

FIG. 3. Time series of total concentrations (L^{-1}) of particles recorded by four different optical array probes mounted on the NCAR C130 during flight 20 of the SCMS. No particles were recorded by the 2DC, the 1DC, or the 200X.

Two temperature sensors mounted near the nose of the C130 were known to be affected by use of the aircraft radio, and when these data are compared to those from the 260X it is clear that the noise affecting the 260X occurred equally as often when the radio was and was not being used.

The source of the noise does appear to be consistent with the effects of electromagnetic interference on one-dimensional and optical array probes studied by a group at the GKSS Research Center at the Institute for Atmospheric Physics in Hamburg, Germany (D. Nagel 2000, personal communication). In an unpublished study, they found that electromagnetic radiation from the 28-V aircraft generators or the aircraft inverters

TABLE 1. Description of six clear-air flight segments by the NCAR C130 during the SCMS in 1995.

Date	Time (UTC)	Total time (s)	Altitude (m)	Relative humidity (%)	2DC images (sizes) (μm)
21 Jul	1723–1750	2640	800	90	100, 150
21 Jul	1812–1829		500	70	75, 75, 175
24 Jul	1335–1420	4440	200–600	70–80	50, 100, 100, 175
24 Jul	1451–1520		550	80–95	75, 75, 150
6 Aug	1325–1340	3000	200–500	80–90	None
6 Aug	1345–1420		600–1800	60–80	None
7 Aug	1333–1351	2400	550	60–90	None
7 Aug	1355–1401		550	60–95	75
7 Aug	1452–1508		550	60–95	None
8 Aug	1430–1500	1800	200–800	70	125, 150
12 Aug	1400–1423	1680	200–1200	60–90	None
12 Aug	1435–1440		600	80–99	None

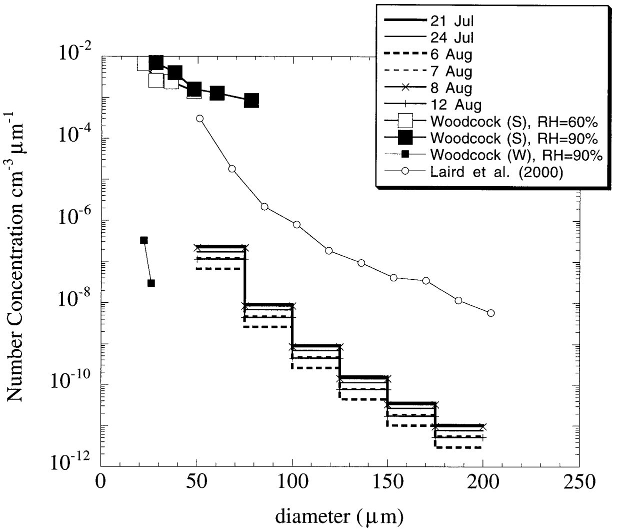

FIG. 4. Comparison of size distributions of UGA from different studies. Squares denote data from Woodcock (1953) for strong (S) and weak (W) wind cases at relative humidities of 60% or 90%. Open circles denote composite distribution from 85% to 90% relative humidity during the SCMS from Laird et al. (2000) using 260X data. Solid and dashed lines denote upper limits on UGA size distribution based on SCMS 2DC data for the six cases listed in Table 1.

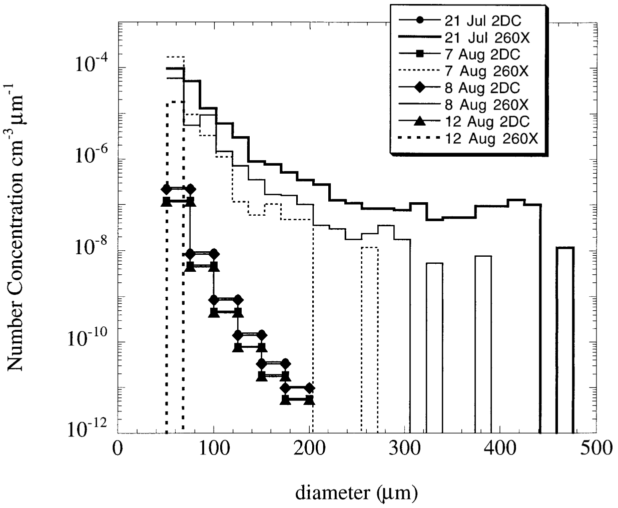

FIG. 5. As in Fig. 4, except size distributions of UGA from 260X data for four individual days are compared to the corresponding upper limits derived from 2DC data. The plot is truncated at a diameter of 500 μm, but counts at sizes as large as 884 μm were recorded by the 260X on 21 Jul.

(which convert the raw 28-V dc from the aircraft generators or alternators to 110-V ac for the probe electronics) could trigger the forward-scattering spectrometer probe FSSP-100 and cause narrow streaks on the 2D probe images. They also found that 400-Hz inverters produced much more noise than 50-Hz inverters. This noise occurred as counts in any channels of an FSSP-100 or 2DC and could produce an exponential size distribution. It is impossible to distinguish this kind of noise from real particle counts. More than one frequency of interfering radiation was required to replicate the variety of noise observed on the probes, and certain probes were sensitive to certain frequencies, while other probes were completely immune to those frequencies. This could explain why the 260X during the SCMS was subject to noise while the 2DC was not.

The GKSS group solved this noise problem by adding filters at the output of the aircraft inverters, as well as shielding and filtering all the data and power lines from within the probe to the data-acquisition system. Further study is needed to determine if this problem, and this solution, are relevant to the 260X noise on the NCAR C130.

4. Summary and discussion

Previous estimates of UGA derived from the SCMS 260X data have been shown to be most likely contaminated by noise. The 260X data are inconsistent with data from other 1D probes flown during the project, as well as the 2DC, when flown in clear air. The possibility of electromagnetic noise producing artificial particle counts has been discussed. New upper limits of UGA have been presented here, based on data from a 2DC probe. These upper limits are three orders of magnitude lower than the estimates from the 260X data.

Much higher concentrations of UGA than those estimated from the 2DC data have been used to explain the appearance of early radar echoes in Florida cumuli from the SCMS, with reasonable results (Lasher-Trapp et al. 2001). The reason for this apparent discrepancy is at this time unknown. One possibility is that the UGA concentrations in the boundary layer fluctuate over much smaller spatial and temporal scales than those represented here. (This cannot be investigated with the SCMS dataset because such long time-averaging is required to attain adequate sample volumes.) Of course, another equally likely possibility is that UGA are not the source of the large drops responsible for the early radar echo in these clouds. Both possibilities are topics for future work.

The noise contamination of the NCAR 260X data in clear air may not be limited to the SCMS. Investigation of a few flights from the Indian Ocean Experiment (INDOEX) showed similarities to the SCMS data: the 260X recorded very large particles in clear air, with no corresponding images on the 2DC.

Finally, based on the limitations experienced during this field campaign, it has become clear that better instrumentation is urgently required to measure ultragiant particles in the atmosphere. Upper limits of UGA concentrations in clear air calculated here with the SCMS 2DC data still vary by an order of magnitude, limiting their usefulness for any future studies quantifying their effect in clouds. Because of the large sample volumes required (several to hundreds of cubic meters at a minimum, for UGA as large as 200-μm diameter), the present electronic optical array probes are inadequate, especially if quantification of temporal and spatial variability is required. An additional desirable aspect in any new instrumentation would also be the ability to measure the solubility of the UGA, in order to include it in microphysical calculations of their growth.

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