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UNIVERSITY OF NEVADA SYSTEM

INTERIM REPORT
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TITLE OF GRANT:	Stratospheric CCN Sampling Program
RESEARCH INSTITUTION:	Desert Research Institute Atmospheric Sciences Center University of Nevada System P.O. Box 60220 Reno, NV 89506
PRINCIPAL INVESTIGATOR:	C. Fred Rogers
REPORT FOR THE PERIOD:	April 1, 1981 - October 1, 1981
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AUTHOR(S): C. Fred Rogers/James G. Hudson

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I. INTRODUCTION

A. Work Prior to Inception of Grant

The Desert Research Institute has had a history of involvement in cloud condensation nuclei (CCN) measurements which began in the mid-1960's. In the period from about 1971 to the present, a series of state-of-the-art instruments for the measurement and characterization of CCN have been built at the Atmospheric Sciences Center of DRI. When Mt. St. Helens produced several major eruptions in the late spring of 1980, there was a strong interest at DRI in the characterization of the CCN activity of the material that was injected into the troposphere and stratosphere. The scientific value of CCN measurements is two-fold: first, CCN counts may be directly applied to calculations of the interaction of the aerosol (enlargement) at atmospherically-realistic relative humidities or supersaturations. Such enlargement may be important in calculations of the scattering of solar radiation by the stratospheric aerosol. If the aerosol is actually transported across the tropopause into the troposphere, then of course CCN measurements are also of value in considerations of the modification of cloud microstructure (several references relevant to these points are cited in Appendix A). A second application of CCN measurements is that if the chemical constituency of the aerosol can be assumed, the number-versus-critical supersaturation spectrum may be converted into a dry aerosol size spectrum covering a size region not readily measured by other methods.

Telephone inquiries in the days following the important May 18, 1980 eruption led to the initial contact with Mr. Bill Page of NASA Ames Research Center, who encouraged us to discuss our interests with the Ames research team headed by E.C.Y. Inn and J.F. Vedder. These investigators offered to share one-liter grab samples of stratospheric air collected when their own trace gas sampling experiment was flown on the U-2 aircraft. The one-liter containers presented various problems when adapted to CCN sampling; these will be discussed below. On the other hand, the offer represented a unique opportunity since there were previously no measurements of stratospheric CCN *per se*). Four samples were thus obtained in June of 1980, and two in December of 1980. Results will be discussed below.

Additional activities included participation by the Principal Investigator (F. Rogers), J.G. Hudson, and W.C. Kocmond in the "Workshop on Mt. St. Helens Volcano", July 11, 1980, and in the Aerosol Climate Effects (ACE) Program Workshop, October 1-2, 1980.

A letter proposal for further CCN sampling utilizing the existing one-liter grab samples was sent to NASA Headquarters on September 22, 1980 and, with revised budgetary estimates, was resubmitted on November 20, 1980.

B. Work Following Inception of Grant

Following the award of Grant NAG 2-114 on April 22, 1981, plans were immediately implemented to refurbish a DRI instrument trailer to be the base of operations at NASA Ames. The trailer remained there while two samples were provided by a U-2 flight on May 12, 1981 and four samples were provided by two flights on July 13 and 14, 1981. Following the two July flights, the one-liter sample bottles were taken back to DRI-ASC for repeated flight simulation experiments.

Activities in the period from April 1, 1981 to October, 1981 also included attendance at the ACE Workshop of May 27-28, 1981 by F. Rogers and J. Hudson. At that time, Dr. Tom Ackerman of the ACE Program had solicited inputs concerning desirable activities of ACE as the program grows to include tropospheric as well as stratospheric aerosol studies. A letter from DRI to Dr. Ackerman was prepared, and is included as Appendix B of this report.

II. STRATOSPHERIC AEROSOL SAMPLING METHOD

A. One-liter Containers

The grab sample containers available on the cryogenic trace gas sampling U-2 pallet of Inn and Vedder are modifications of an "off-the-shelf" Whitey one-liter stainless steel sample cylinder having a length of 20 cm, radius of 5 cm, and a wall thickness of 0.47 cm; part number 304-HDF4-1000. A Varian "mini-conflat" vacuum flange has been welded onto one end of each cyliner, and a Cajon vacuum fitting for 1/4 inch tubing has been welded to the other end. The Varian fitting connects directly to a Varian vacuum valve via a solid copper gasket; the Cajon fitting similarly utilizes a solid nickel gasket. In the design application of trace gas sampling, a pilot-actuated motor opens and closes the Varian valve, admitting samples to the previously evacuated cylinders from a supply duct through which stratospheric air flows under ram pressure. Two such cylinders may be flown on each mission.

We slightly modified these containers after the June, 1980 flights and before the December 1980 U-2 flights. Small stainless steel valves were welded to a matching Cajon fitting, and outfitted with 1/8 inch stainless steel tube probes. The lengths of the probes were calculated to allow their inlet ends to be located at the geometric center of each container when the matching Cajon fittings were joined. Later withdrawal of the stratospheric samples was thereby facilitated; the aerosol could then be withdrawn with (in theory) the least effect of depletion of particles by diffusion to the inside walls of the containers.

For the purposes of the June 1980 flights, time had not allowed pre-flight attachment of a probe system for withdrawing the samples and, in a very difficult and risky operation, probes were inserted into the sample bottles after each flight.

Four different one-liter container and valve sets have been utilized to date. These are labelled "4 FWD", "4 AFT", "9 RT", and "9 LFT"; Table 1 summarizes the U-2 missions and ground simulations to date, and which container was used in each case.

B. Sampling Procedure

The objectives vary from one U-2 mission to another but, in all cases, we have been carefully advised by responsible parties at NASA Ames as to the flight plan, the stratospheric wind conditions, and the expected stratospheric aerosol vertical and horizontal distribution (as well as that can be specified) before each mission. Table 2 summarizes the broad objectives of the U-2 missions in which we have been involved. Our preferences for the vertical and horizontal coordinates of the two

TABLE 1

Date	Container No.		Comments
14 JUN 80	4 AFT		High count, 45 K ft.
	4 FWD		Low count, 61.5 K ft.
16-17 JUN 80	9 RT	Simulation:	Checked dilution, decay factors
17 JUN 80	4 AFT		Low count, 62 K ft.
	4 FWD		Low count, 49 K ft.
19 NOV 80 - 1 DEC 80	9 RT	Simulation:	Revised decay factor
5 DEC 80	9 LFT		High count, 52 K ft.
	9 RT		Low count, 46 K ft.
12 MAY 81	9 LFT		High count, 60 K ft.
	9 RT		Low count, 50 K ft.
13 JUL 81	9 LFT		High count, 55 K ft.
	9 RT		Failed to open
14 JUL 81	9 LFT		42 K ft.
	9 RT		Very low count, 57.5 K ft.
15 JUL 81	9 LFT	Simulation:	First noticed spurious CCN production
	9 RT		
16-28 SEP 81	9 LFT	Simulation:	Study of spurious particle production
	9 RT		

TABLE 2

Date	Objective
14 JUN 1980	Intercept plume of St. Helens 13 June eruption
17 JUN 1980	Intercept plume of 18 May eruption
5 DEC 1980	Study St. Helens aerosols several months after major eruptions
12 MAY 1981	Look for plume of Atlasova volcano
13 JUL 1981	Study remaining volcanic aerosol north of jet stream
14 JUL 1981	Rendezvous with University of Wyoming balloon

grab samples of each flight have then been given to Dr. Vedder, who then incorporates the required pilot action (actuating the motor on the Varian valve) into the cryo pallet pilot log sheet. Preceding each sample, air flow through the cryo pallet sample supply duct must be established if not already done.

Previous to each flight, the sample containers are evacuated by the vacuum system used for the cryo pallet, to a pressure of about 40 millitorr. This pressure is maintained while the containers are heated with a heat gun, and the vacuum gauge is monitored for evidence of outgassing. The procedure no doubt could be refined but, to date, there has been no evidence of outgassing; on a few occasions, leaks have been detected but were easily repaired.

The one-liter samples have been taken off the U-2 aircraft as quickly as possible at the end of each mission to minimize losses of CCN through Brownian diffusion to the walls of the containers. Typically, about three hours elapse between the time the sample bottles are filled and the time the CCN samples are analyzed on the ground.

The sample containers are quickly taken to the DRI instrument trailer, where the contents must be brought from stratospheric pressure to ambient pressure with particle-free air. This "backfill" air has also passed through a bed of fresh activated charcoal and, on certain occasions, through an oxalic acid trap to remove ambient ammonia. These latter two traps are always placed upstream from the particle filter, and are repeatedly checked to be sure they do not in themselves generate spurious CCN. The backfilling with particle-free air continues as the aerosol content of the sample containers is drawn into a CCN spectrometer built at DRI, through the previously-described sample probe at a flow rate of order one $cm^3 sec^{-1}$. Table 3 is a copy of a checklist used when the sample containers are backfilled and connected to the CCN spectrometer. A description of the CCN spectrometer can be found in Appendix C.

The CCN count is then taken as the cumulative number of particles active at supersaturations over the approximate range 0.1% to 1.0% (i.e., 100.1 to 101% R.H.). Results to date will be shown in Section III.

C. Corrections to Data and Estimates of Error

The end result of these measurements is a number density of CCN active at supersaturations in the range 0.1% to 1.0%. Several major factors affect the measurement:

(1) Depletion of the stored aerosol by Brownian diffusion to the walls reduces the concentration by 25% ± 5% per hour, when the contents are at 850 mb pressure and laboratory temperature, as determined by experiments utilizing the actual flight bottles. An early measurement of this loss rate, with a mock container, yielded a higher value, 35% ± 5%, but we believe this figure to have been an overestimate.

We have not yet experimentally checked the CCN diffusion loss rate under the actual temperature and pressure conditions

TABLE 3

Preparation of Grab Sample Containers for Connection to CCN Spectrometer

1. Flush backfill air line with particle-free air
2. Tape thermistor to container to record temperature
3. Attach backfill air line connection flange (Varian "mini conflat") to container
4. Flush connection flange with particle-free air
5. Shut off pressurized flush air, make connection of backfill air line to container
6. Valve off backfill line
7. Open Varian valve to connect container to Magnehelic pressure gauge
8. Slowly open valve on backfill line, allow particle-free air to dilute contents of container
9. Open valve between internal container sample probe and sample inlet to CCN spectrometer.

found in U-2 missions, and this remains an important task yet to be done. To a first approximation, however, the diffusion coefficient, D, of submicron particles in air is proportional to the ratio of kT to the viscous retarding force, or

$$D = \frac{kT}{6\pi\eta r},$$

where η is the viscosity of air (independent of density) and r is the radius of an aerosol particle. Since η is independent of density, but proportional to (air temperature) $T^{\frac{1}{2}}$,

$$D \propto T^{\frac{1}{2}}.$$

If the grab sample containers were as cold as ambient air, T would be about 223 K (minimum) and D would be reduced by 13% over the value at ordinary conditions. The diffusion loss rate would be reduced in proportion. The sample containers are, however, quite massive, and it seems unlikely that they ever cool to actual stratospheric values. One attempt was made to measure both the temperature of a container and to detect temperature gradients between top and bottom on one of the sampling missions, but examination of the data revealed a problem with the calibrations of the thermocouples used which renders the results inconclusive.

A typical total value, then, of the fraction of aerosol lost over 3 hours at 25% ± 5% per hour is 58% ± 9%, implying a

TABLE 4

Flight of	Container No.	Aerosol Decay Factor	Dilution Factor
14 JUN 80	4 AFT	2.9	4.98
	4 FWD	3.2	8.10
17 JUN 80	4 AFT	4.7	10.2
	4 FWD	1.5	4.73
5 DEC 80	9 LFT	1.8	7.65
	9 RT	2.4	6.07
12 MAY 80	9 LFT	1.6	9.67
	9 RT	2.0	6.61
13 JUL 81	9 LFT	2.7	8.20
	9 RT	Accidental Blank	
14 JUL 81	9 LFT	4.2	4.52
	9 RT	4.4	9.09

(4) The statistical counting error applicable to CCN counts at any given setting of the CCN spectrometer is proportional to the square root of the magnitude of the count. Typically at least 50 cm^3 of the diluted sample air are passed through the CCN spectrometer to obtain the CCN counts, and a minimum count would be of order 2 CCN cm^{-3}. Therefore, statistical counting error accounts for a maximum error bar of about ±10% in these results. (Usually, the error bars due to the uncertainty in the diffusional decay factor completely dominate statistical counting errors, as well as uncertainty in the dilution factor.)

III. RESULTS TO DATE

A. Flights of June and December, 1980 and May, 1981

The results of the measurement in the CCN spectrometer is a "raw" count of CCN active at a given supersaturation. To obtain a number density, the count must be divided by the volume of air sampled by the instrument; that number must then be multiplied by the decay and dilution factors as shown in Table 4. Figures 1 and 2 display data from the flights of June 14, 1980, December 5, 1980, and May 5, 1981. The discussion to follow will include the data of June 17, 1980 but, for reasons to be given, the graphs of these data are not included in the main series of Figures 1 and 2; the reader will find the June 17, 1980 data graphed in the *Science* paper in Appendix A. Figures 3 and 4 display the data from the most recent flights of July 13, 1981 and July 14, 1981.

The samples over the period June 14, 1980 to May 5, 1981 were taken at altitudes ranging from 13.6 km to 18.6 km. The data are shown roughly segregated into two groups: four samples taken between 13.6 km and 15.8 km and two samples taken at the higher altitudes of 18.2 km and 18.6 km.

Figure 1 shows the lower altitude data set, with measurements from June 14, 1980 (over western Montana), December 5, 1980 (both over northern California), and May 5, 1981 (over northern California). The error bars represent the uncertainty in the measurement of the aerosol decay rate in the sample containers; as mentioned above, the statistical counting error is much smaller.

The June 14, 1980 sample was taken at the northern edge of the June 12 eruption plume, in freshly-injected debris. Simultaneous SO_2 measurements were not particularly elevated (Inn, *et al.*, 1981a); thus these CCN may be largely composed of material which originated in the volcano itself, rather than H_2SO_4 generated by gas-to-particle conversion.

We do not show results taken out-of-plume over central California at the same altitude of 13.6 km. The CCN counts were comparable to the 13.9 km sample of December 5, 1980 (Figure 1), and indicate the concentration that can be expected from the unperturbed lower stratosphere.

The next two CCN spectra in Figure 1 were taken on December 5, 1980 at 15.8 km and 14.0 km. The 15.8 km results (upper curve) are comparable in concentration to the 13.6 km results of June 14, 1980, which is surprising in view of the relative dormancy of Mt. St. Helens in the fall of 1980. This result does, however, correspond with an observation of enhanced SO_2 (Inn, *et al.*, 1981b), and suggests that gas-to-particle conversion was still a significant producer of aerosol mass at 15.8 km in December.

The lower altitude (14 km) result of December 5, 1980 may be an indication of the normal background CCN concentration at that level as the Mt. St. Helens perturbation was dying away.

Finally, in Figure 1, we show results at 15.1 km taken on May 12, 1981 during a flight intended to intercept the then recent plume of the volcano Atlasova (Kamchatka Peninsula). The CCN spectrum is similar in both concentrations and slope to the 13.9 km December 5, 1980 result, further supporting the suggestion that this approximate magnitude of concentration is the normal background in the 14-15 km regions.

Figure 2 shows two CCN spectra taken between 18 km and 19 km on June 14, 1980 (over central California) and on May 12, 1981 (over northern California). The June 14, 1980 sample was intended to intercept debris of the May 18, 1980 Mt. St. Helens eruption after it had passed once around the world, but we are unable to confirm (e.g., by trajectory analyses) that this is the case. The counts observed are low enough to make us question whether or not the sample was actually taken in-plume.

The flight of May 12, 1981 was, again, intended to intercept the plume of the volcano Atlasova. Figure 2 shows a very elevated count for this sample, which almost certainly cannot be attributed to Mt. St.

Helens. In Section IV we discuss a problem with sample container 9 LFT which was used for this measurement; spurious particle production in this container was first observed on July 15, 1981 in simulation experiments, and may have been present on May 12.

Not shown in Figure 2 are results from a sample taken at 18.8 km over central Colorado, within the May 18, 1980 Mt. St. Helens eruption plume on its second pass around the world. Since this sample was 5.3 hours old when analyzed, the longest elapsed time of any of our samples, the loss rate correction was uncomfortably large. The dilution factor was also quite large, and the total multiplication to convert raw data into final values was a factor of 48. The counts from this sample after this large correction are comparable to the 18.6 km results of June 14, 1980.

Before leaving Figures 1 and 2, it should be noted that the results of June 14, 1980 were previously reported in *Science* (see Appendix A) as computed from the then-estimated aerosol decay rate of 35% ± 5% per hour. Figures 1 and 2 show that data as recomputed on the basis of the more accurate decay rate of 25% ± 5% per hour.

The data shown in Figures 1 and 2 were also presented at the Symposium on the Role of Volcanic Emissions in Atmospheric Chemistry, IAMAP General Assembly, August 21-22, 1981, Hamburg, Germany.

B. Flights of July, 1981

The U-2 sampling missions of July 13 and 14, 1981 had the important goals of looking at the remaining Mt. St. Helens aerosol north of the jet stream, and a rendezvous with a balloon launch from the

University of Wyoming. Figures 3 and 4 show our results from both of these flights.

The data from July 13 show a high count at 12.7 km; we presently question this result, as it was obtained with container 9 LFT. This container may have been subject to spurious CCN production as discussed in Section IV of this report, and first observed on July 15.

We also intended to open the remaining sample container, 9 RT, at 19.7 km, but this run provided an unintended blank sample when the motor-driven Varian valve failed to open. When later connected to the CCN spectrometer, this sample showed only the expected background count.

The flight of July 14 gave very similar results in the sense that on that day the 12.7 km sample, again from container 9 LFT, showed several hundred CCN cm^{-3} active at 1% supersaturation. The count in container 9 RT, from 17.4 km, was much lower, indicating only about 52 CCN cm^{-3} active at 1% supersaturation. Note in Table 4 that both these samples had large aerosol decay factors, due to the distance at which the rendezvous with the University of Wyoming balloon had to be accomplished. We have not yet had the opportunity to compare these CCN counts with the results of the Wyoming instrument from July 14.

C. Concluding Remarks

It is noteworthy that the CCN concentrations we have observed, both in fresh plumes and long after eruption events, have generally been comparable in magnitude or larger than condensation nuclei (CN) counts obtained by Rosen and Hofmann at the University of Wyoming (e.g. Rosen and Hofmann, 1980). In general, CCN counts are a subset of CN counts, but if all aerosol particles being sampled are mostly soluble and exceed the size needed to register in the CCN counter (critical supersaturation less than about 1%), then the two types of counters should yield equal concentration values. In measurements made soon after the major eruptions of Mt. St. Helens, it did indeed seem to be the case that aerosol sizes met this criterion (Chuan *et. al.*, 1981). It may be that spurious CCN production in the sample containers (Section IV) can account for some of this difference, but it would be instructive at some point in the future to compare our CCN instrument to the CN counter used by Rosen and Hofmann, when both are sampling monodisperse aerosols which should register the same number in both devices. Such comparisons were made with numerous other particle measuring devices from around the world at the recent Third International CCN Workshop held at DRI, October 6-17, 1980.

On the other hand, we can identify two reasons for our counts to be underestimates of the true CCN concentration:

(1) We believe our value for aerosol decay rates in the one-liter sample bottles is more likely to be an underestimate rather than an overestimate. (It has been difficult to perfectly simulate the actual experimental situation in every respect, including stratospheric temperatures).

(2) We expect that CCN composed mostly of sulfuric acid would evaporate as they are brought from stratospheric temperatures to surface ambient temperatures.

CCN spectral measurements take on added utility if one can assume the CCN to be mostly or entirely composed of a pure soluble salt. Through the use of the standard Kohler relationship between the critical supersaturation and the dry aerosol radis (number of moles of solute), a CCN spectrum may be transformed into a cumulative size distribution. This has been done for two of our cases, and the result is shown in Figure 5; it was assumed that the CCN were pure ammonium sulfate, but the curves would be shifted only slightly along the size axis if the aerosol composition were assumed to be sulfuric acid. (We have actually attempted to determine whether or not our CCN samples are sulfuric acid by performing our measurements with and without oxalic acid ammonia traps on the backfill air; results to date have been inconclusive, whereas we would expect a small increase in critical supersaturation if sulfuric acid CCN were being converted to ammonium sulfate CCN). The size distributions of Figure 5 illustrate the potential value of CCN measurements in providing size information over the radius interval of about 10^{-6}cm to 10^{-5}cm, where few other methods of aerosol sizing are of much value and where the aerosol products of gas-to-particle conversion may be found.

IV. PROBLEM WITH SPURIOUS PARTICLE PRODUCTION IN SAMPLE CONTAINERS

Simulations of the stratospheric sampling procedure with actual flight versions of the one-liter containers have been carried out on four occasions, as shown in Table 1. Generally, there has been a protocol of leaving these containers at NASA Ames should they be needed for the cryogenic gas sampling experiment, and it has been necessary to conduct the simulations while visiting the Ames facility. Following the July 1981 flights, however, it was arranged to bring containers 9 RT and 9 LFT back to DRI for more extended tests.

The simulations of 16-17 June 1980 utilized container 9 RT. Of particular interest to this discussion is one type of simulation performed, wherein the container was flushed with room air, then evacuated with a vacuum pump to typical stratospheric pressures. The bottle was then backfilled in the usual manner, and connected to the CCN spectrometer to see if the resulting count was the proper fraction of the original, room CCN count. Several tests of this type showed that the dilution was indeed as would be calculated on the basis of the initial and final pressures in the sample container. No evidence of spurious particle production due to these operations was seen.

On July 15, 1981, a different sample container, 9 LFT, was being similarly tested in the DRI instrument trailer at Ames Research Center. This time, however, a spurious CCN count of order 300 CCN cm^{-3} was found to be generated when this container was evacuated, then backfilled with particle free air. Container 9 RT was tested again and found to exhibit a very small spurious count (more than a factor of ten less than 9 LFT). Time did not allow further experiments, except to check and confirm that the spurious count in the containers did decrease with time, as though the particles were all generated during the backfill operation.

Containers 9 LFT and 9 RT were taken back to the DRI laboratory in Reno, where simulation experiments were resumed on September 16, 1981.

In these tests, a "total" or condensation nucleus (CN) counter was being used as the detector of spurious particle production. This device, a standard Nolan-Pollak counter, applies a supersaturation of 160%, and hence detects small nuclei which would not register in the CCN spectrometer. The results are, therefore, probably overestimates of the numbers of spurious CCN produced. Laboratory simulations to determine the cause of the spurious counts are continuing at the time of this report, but the following preliminary conclusions can be stated:

(1) Production of spurious CN when backfilling with particle-free room air generates concentrations of 200 to 300 cm^{-3} in container 9 LFT, and 20 to 60 cm^{-3} in container 9 RT; there is a consistent and reproducible difference between the two containers;

(2) The spurious particle concentration decreases with time following the backfilling operation; there is no indication of ongoing particle production in either container;

(3) The spurious particle concentration is unaffected by movement of the mechanisms of either of the two valves on each container; this shows, for example, that the spurious CCN are not generated by rubbing of the valve bodies against the valve seats, or by flexing of the stainless steel bellows in the Varian vacuum valve;

(4) Simply blowing particle-free air through a sample container with opened valves does not produce spurious CCN;

(5) If the backfill air used in tests on container 9 LFT is replaced with tank nitrogen which is passed through an activated carbon filter, the spurious CN production seems to be reduced by a factor of two to three.

Again, we note that container 9 RT provided an unintentional "blank" in the flight of July 13, 1981, when it properly showed a zero count on the CCN spectrometer.

Tests in the near future will address two basic hypotheses:

(1) Spurious particle production is a function of contamination in the containers; for example, container 9 LFT was opened in a region of high SO_2 concentration in the December 5, 1980 flights, and may have been exposed to significant amounts of H_2SO_4 aerosols; contrary to this explanation is the preflight preparation procedure wherein each container is heated while being evacuated and while a vacuum gauge is being monitored for evidence of outgassing;

(2) Spurious particle production is a function of the expansion of particle-free gases into the containers during the backfilling operation, with some gas-to-particle conversion process reponsible for at least part of the observed count.

A test of hypothesis (1), for example, would involve baking container 9 LFT while continuously evacuating it; it is desired, however, to identify the source of the spurious particles before destroying the generating mechanism or circumstances. A test of hypothesis (2) would involve an attempt to collect a sufficient deposit of the spurious particles for elemental analysis in the University of Nevada Physics Department proton-induced X-ray (PIXE) apparatus. Presently, the latter course of action seems the most productive.

To conclude this section, several broad statements can be made. First, this kind of spurious particle production was quite unexpected in containers which are almost entirely fabricated from stainless steel (there is a copper insert on the sealing face of the Varian vacuum valve, a copper gasket where the Varian flanges seal, and a nickel gasket at the opposite end of each container where the Cajon fitting seals). Even an expansion from one atmosphere into moderate vacuum was not expected to produce long-lived particles as large as CCN (of order 10^{-6} to 10^{-5} cm) when clean gases are the expanding medium. Second, if spurious particle production is often present, there may be significant mass conversion from the gaseous to the solid phase. It would seem likely that the trace gas investigators who originally designed the sample containers would have noticed such a perturbation in their measurements. Third, until we have positively identified the sources of the spurious particle production, it seems unreasonable to reject previous data collected before July of 1981.

Containers 4 AFT and 4 FWD will be tested as soon as practicable, but it should be noted that, in the results of June 17, 1980, both of these containers gave a low count. It would appear that, at least, spurious CCN were not generated in significant numbers during the post-flight particle-free air backfilling operation.

V. CONCLUSIONS AND RECOMMENDATIONS

In the fifteen months since June 1980, two one-liter grab samples of stratospheric aerosol have been returned from each of six U-2 sampling missions. CCN spectra from each sample have been obtained by instrumentation taken to and set up at NASA Ames Research Center.

The measured concentrations of CCN have varied from about 800 cm^{-3} (December 5, 1980, 15.8 km, coincident with enhanced SO_2) to about 38 cm^{-3} (December 5, 1980, 14 km, over northern California) active at 1.0% supersaturation. If these CCN were composed of sulfuric acid, their radii would be in the range of about $1.2x10^{-6}$cm to $6x10^{-6}$cm, and observations of them would be relevant to comparisons with models of gas-to-particle conversion in the stratosphere.

The CCN concentrations deduced from these measurements are often as large as, or even exceed, the CN measurements performed by the balloon-borne CN counter of Rosen and Hofmann. They are also significant with respect to tropospheric measurements, as for example quoted by Twomey and Wojciechowski (1969) who found a value of about 100 CCN cm^{-3} active at 1% for maritime air below cloud base. Uncertainties in the CCN measurement are due to aerosol decay in the sample containers, dilution necessary to bring the samples from stratospheric to surface pressure, CCN spectrometer background count, and statistical counting error, but each of these factors have been accounted for, based upon the best information available. Overestimation of the aerosol decay rate and evaporation of sulfuric acid particles as the grab samples are warmed upon their return to the surface would cause these results to be an underestimate of the true stratospheric CCN count.

In July 1981, spurious particle generation was found to be a serious problem in container 9 LFT and a much lesser problem in container 9 RT. The spurious count seems to be generated at the time particle-free air enters an evacuated container; container contamination and gas-to-particle conversion will be investigated in the laboratory as possible generation mechanisms.

Recommendations for improvements in the sampling technique will be an important part of our final report (due January 1, 1982). At the present time, it appears that two approaches are possible. The present containers would be better suited to the purpose of aerosol, rather than trace gas sampling if they were replumbed for flow-through flushing by stratospheric air. The present method of filling involves expansion into the evacuated container when the motorized Varian valve is opened; it is proposed to add a second motorized Varian valve to the outflow end of each container. Sample air would then be supplied from the present duct as before, but would flush the grab sample container and return to the outflow end of the duct. The two Varian valves would be opened simultaneously after some ten or more container volumes of air had been flushed through the bottle. This modification to the cryo pallet has not been discussed with the trace gas investigation team, but seems physically possible and would avoid exposing the CCN samples to an expansion during the collection process. In addition, we are designing and building a new CCN spectrometer which will be able to operate at an internal pressure of 300 mb. Typically, then, stratospheric samples would have to be diluted only enough to bring their pressures up from 100 mb or 150 mb to 300 mb, greatly reducing the dilution factor.

The second and preferred approach is to design new grab sample containers of larger internal radius, with the flow-through flushing provision mentioned above provided by an improved sample supply duct. The larger size of the containers would reduce the aerosol decay factor.

VI. PLANS FOR THE NEXT REPORTING PERIOD

Study of the production of spurious CCN in containers 9 LFT and 9 RT will proceed during the remaining months of this grant. We will continue the experiments described in Section IV, with emphasis on elucidating the spurious particle generation mechanism. Simple experiments using the Nolan-Pollak counter as the detector of particles will include studies of the effect of the rate at which the Varian value is opened (on container 9 LFT). The Varian values will also be exchanged between the two containers, to further isolate the phenomenon.

Additional experiments will include examination of a filter collection of the spurious CCN by a PIXE apparatus, and deliberate exposure of the sample container to a sulfuric acid aerosol of concentration in the range of several hundred cm^{-3} to see if the spurious production is increased.

A representative from DRI will attend the fall ACE Workshop, presently scheduled for the first week of December, 1981, at which time recommendations for continued, improved sampling efforts will be available.

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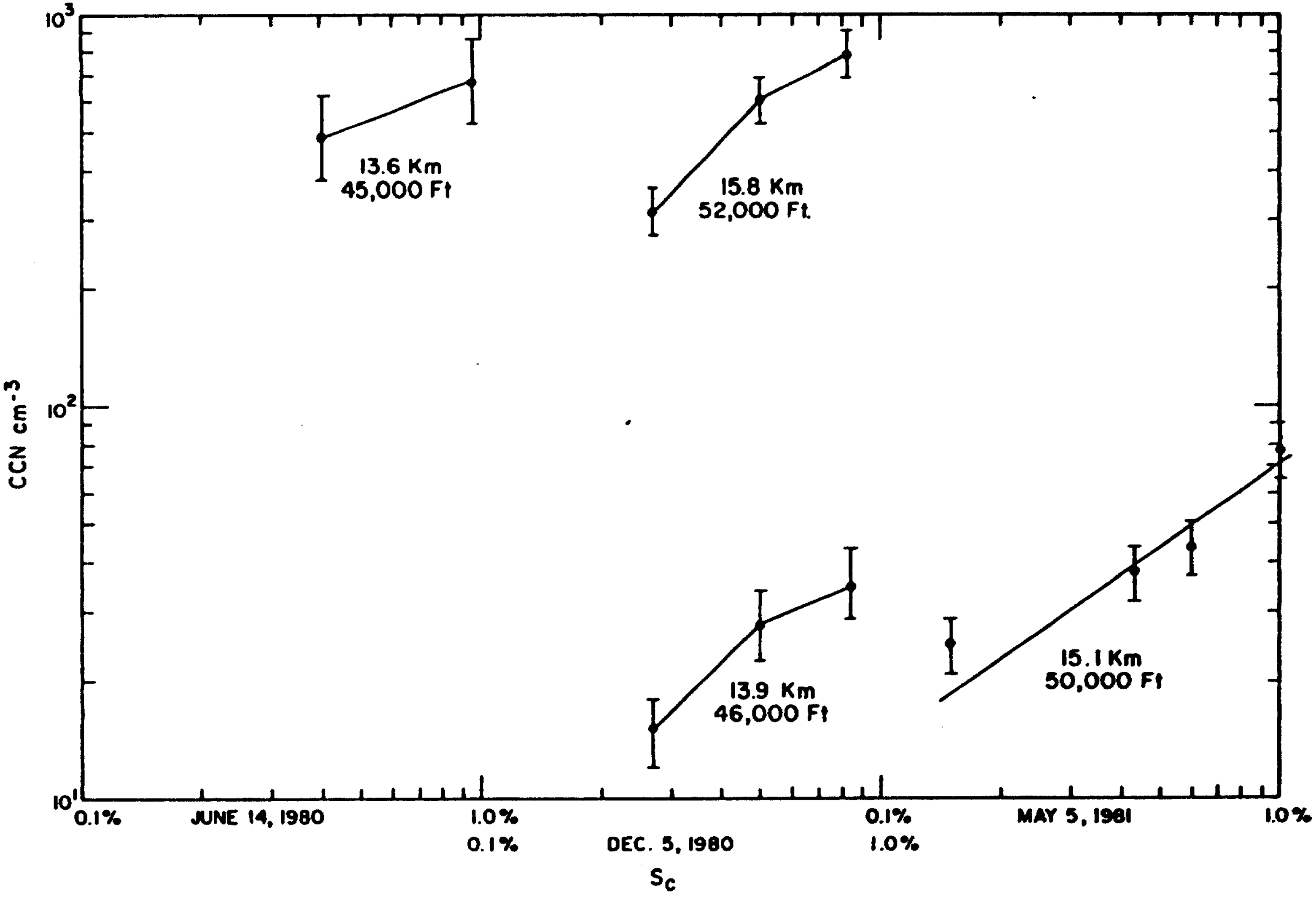

FIGURE 1. LOWER ALTITUDE DATA SET

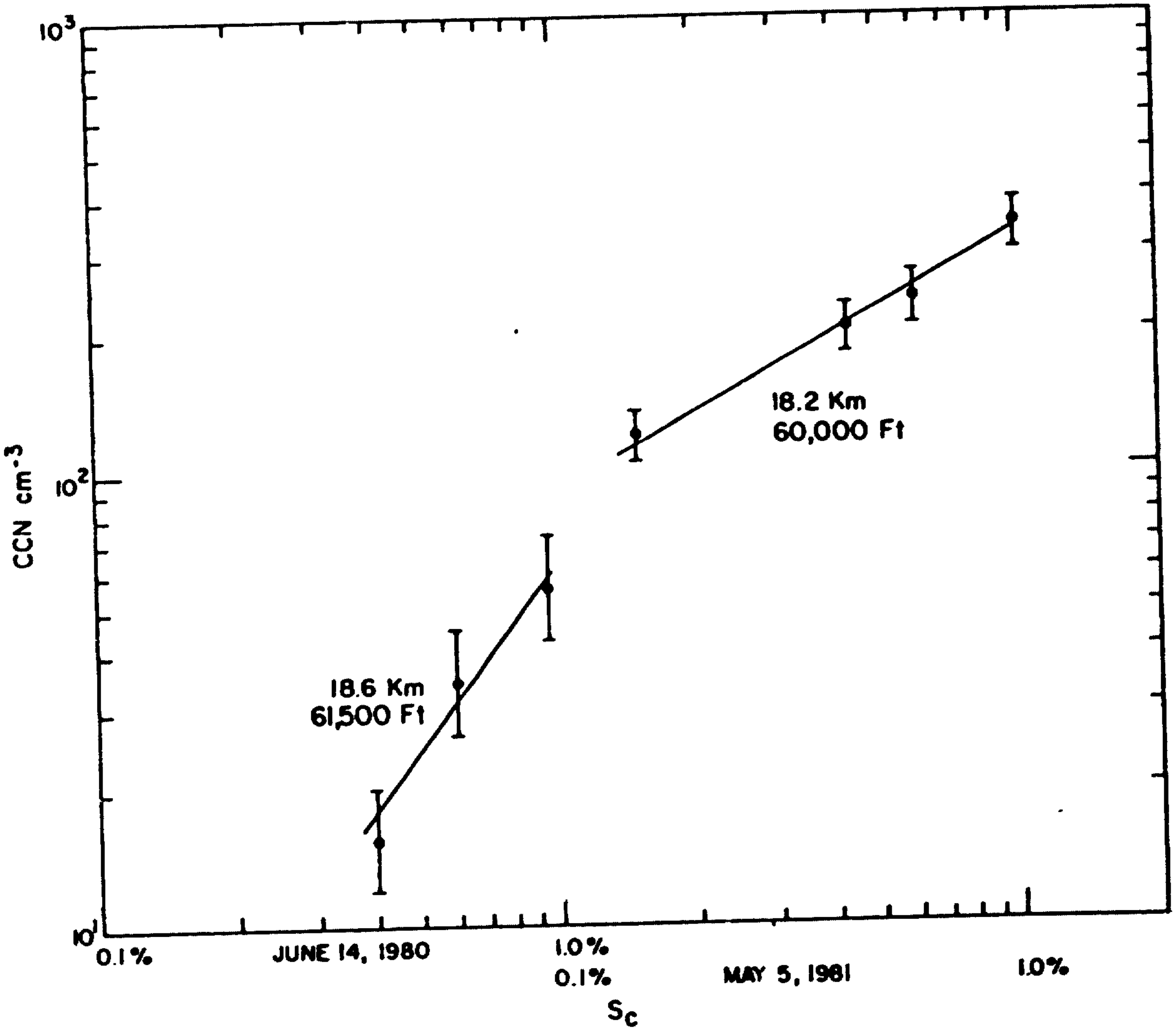

FIGURE 2. HIGHER ALTITUDE DATA SET

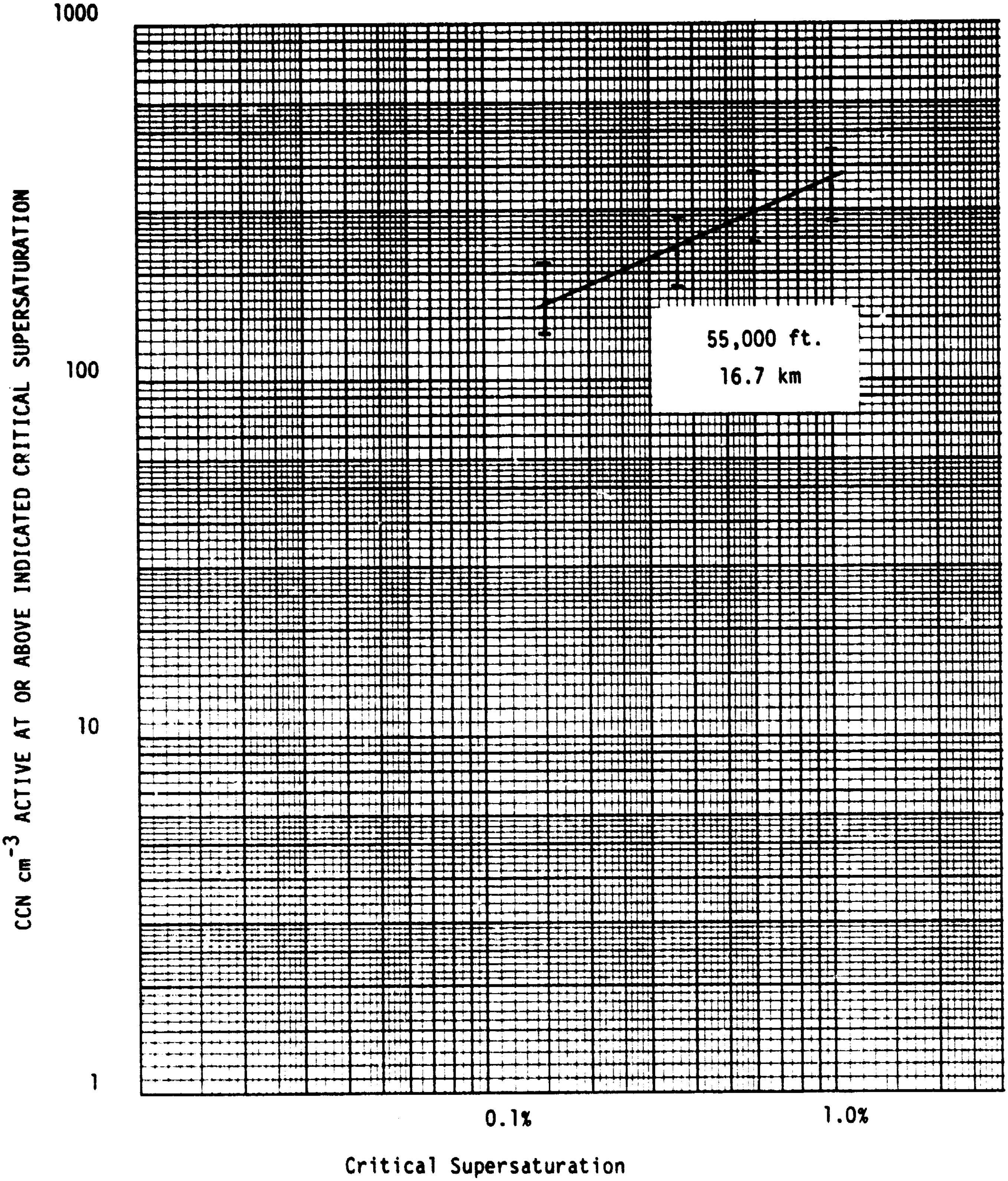

FIGURE 3. FLIGHT OF 13 JULY 1981

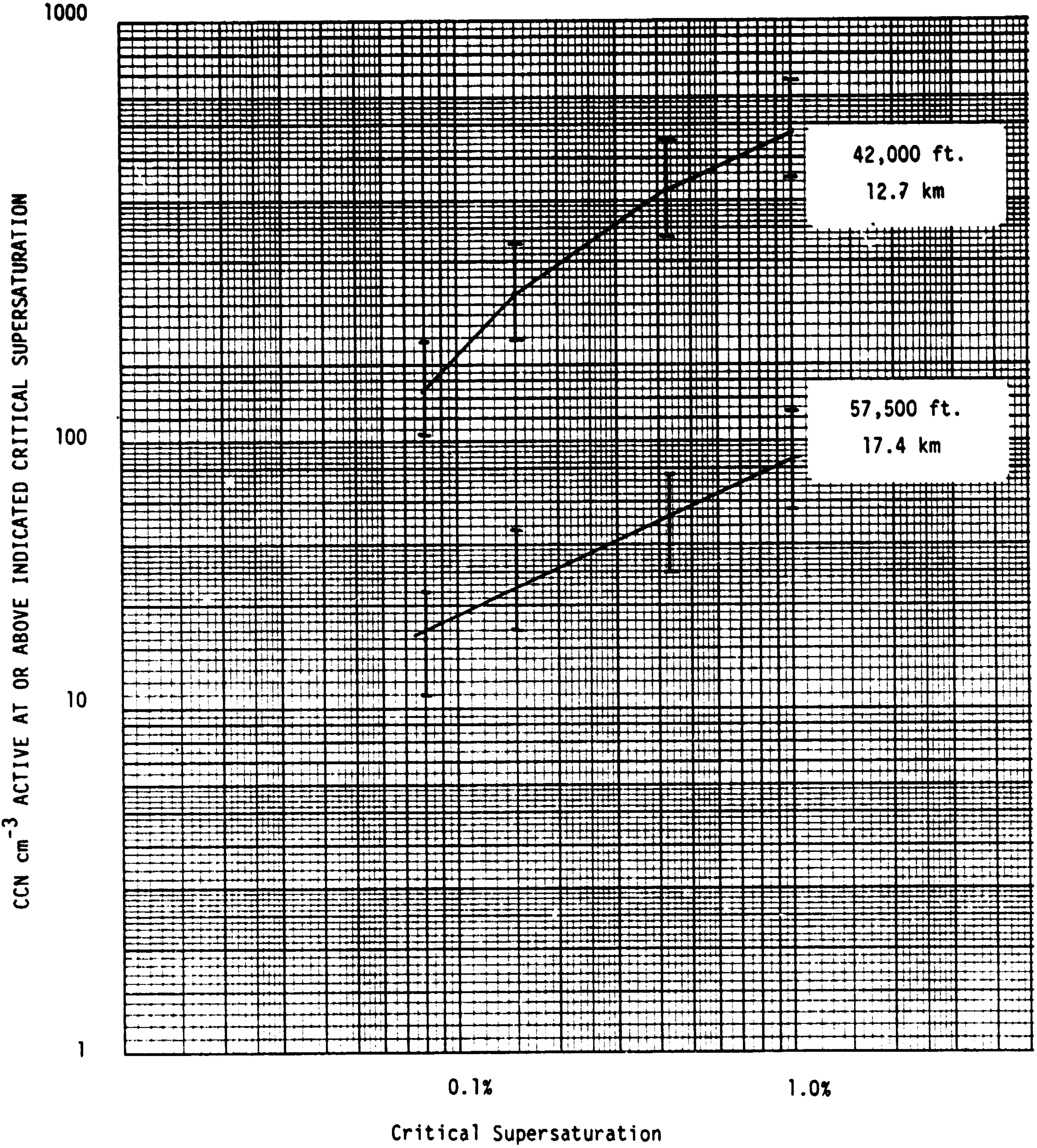

FIGURE 4. FLIGHT OF 14 JULY 1981

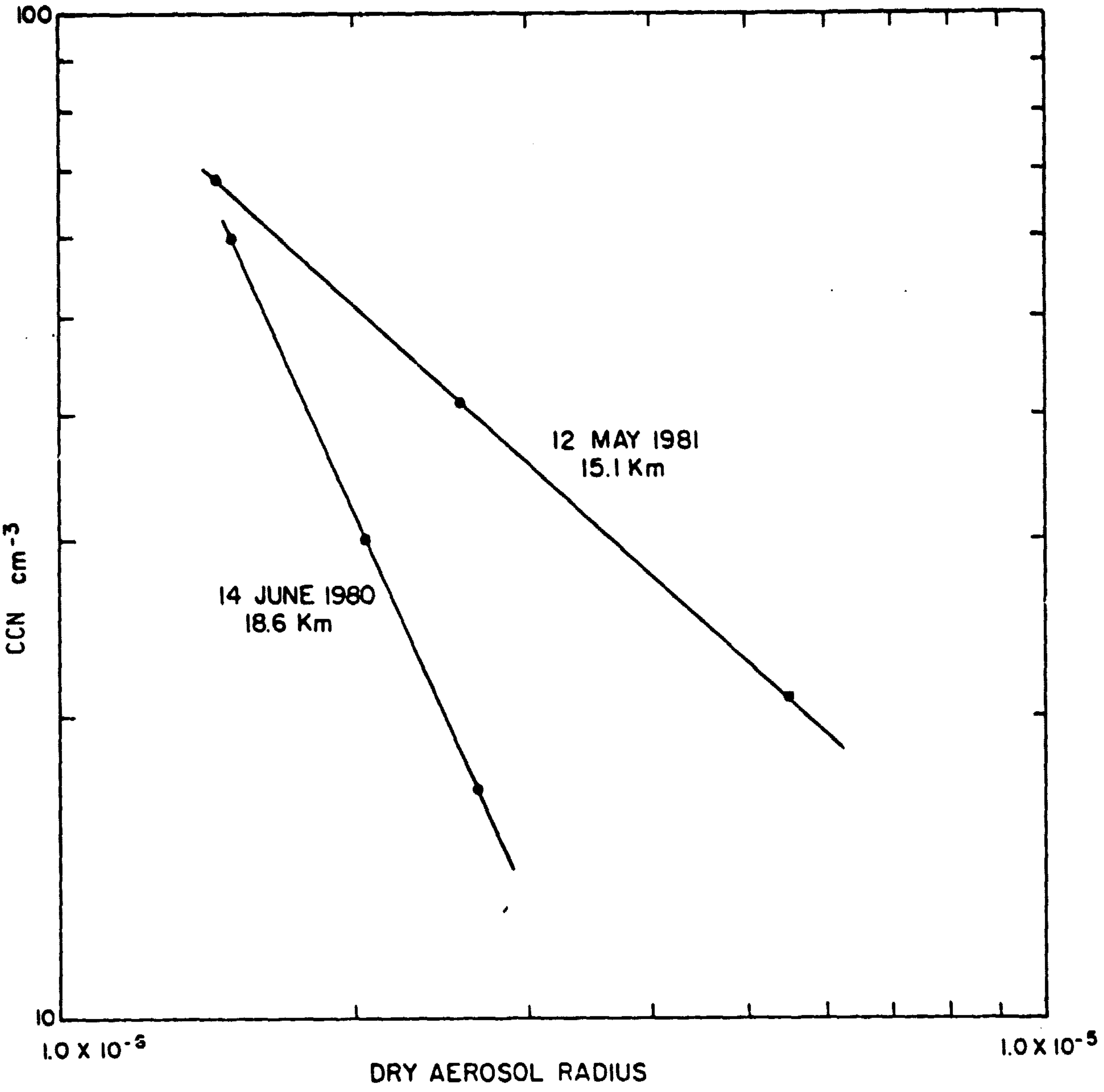

FIGURE 5. CONCENTRATION OF DRY AEROSOL OF RADIUS ≤ GIVEN VALUE

APPENDIX A

ARTICLE BY ROGERS, ET AL., FROM

SCIENCE, 211, 824-825, 1981

Measurements of Cloud Condensation Nuclei in the Stratosphere Around the Plume of Mount St. Helens

Abstract. *Measurements of cloud condensation nuclei were made from small samples of stratospheric air taken from a U-2 aircraft at altitudes ranging from 13 to 19 kilometers. The measured concentrations of nuclei both in and outside the plume from the May and June 1980 eruptions of Mount St. Helens were higher than expected, ranging from about 100 to about 1000 per cubic centimeter active at 1 percent supersaturation.*

The eruptions of Mount St. Helens in May and June 1980 injected significant amounts of gases and particles into the stratosphere. We measured cloud condensation nuclei (CCN), the part of the aerosol capable of nucleating water vapor condensation at supersaturations of the order of 1 percent (relative humidity of 101 percent).

Rather elaborate projections of the effects of volcanic aerosol on the earth's climate have been made in recent years, such as that of Pollack *et al.* (*1*). Whether stratospheric CCN are important in the total picture of global weather depends on (i) the numbers of CCN available in the stratosphere, (ii) the rate at which they enter the troposphere, and (iii) the way in which they affect weather systems in the troposphere. Our measurements relate to the first of these three aspects. The second may involve any of eight mechanisms described by Shapiro (*2*). Once in the troposphere, CCN of stratospheric origin could modify cloud microstructure, leading to two possible effects upon climate: alteration of precipitation processes and alteration of the scattering and absorption of solar radiation by clouds (*3*).

To the best of our knowledge, our CCN data are the first reported from altitudes above the local tropopause. An indication of the CCN count, however, can be gained from the measurements of Rosen and Hofmann (*4*) taken between 10 and 20 percent supersaturation before June 1980, and more recently at 200 percent supersaturation, in numerous balloon ascents over Laramie, Wyoming. These investigators reported evidence of both anthropogenic and volcanic increases in stratospheric sulfates (*5*). Our measurements are taken as a function of two to three supersaturations within the range of those found in actual clouds; if compared to tropospheric counts, they should help to resolve questions of whether or not the stratosphere can ever be a significant source of CCN.

We analyzed four 1-liter samples of stratospheric aerosol collected by a NASA U-2 aircraft. These samples, although well suited for their original purpose of trace gas analysis, presented a serious concern with respect to our CCN measurements. It was expected that losses due to Brownian diffusion to the walls might cause unacceptably rapid depletion of the CCN present in the small sample containers, which were stainless steel cylinders with rounded ends (radius, 5 cm; length, 20 cm). Laboratory simulations of the experiment with similar containers showed that the loss of CCN active at 1 percent supersaturation was a rather consistent 35 ± 5 percent per hour. (No attempt was made to establish stable thermal stratification of the container contents.)

The 1-liter sample containers were cleaned and evacuated before each flight and were opened by pilot activation of motor-driven valves at the specified horizontal and vertical coordinates. All samples were obtained from a sample entry system designed for gas analysis. Diffusion losses of CCN were probably neg-

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ligible, but bends and restrictions in the sample entry plumbing may have allowed impaction losses of some of the largest CCN. Both kinds of losses would cause our final results to be underestimates of the actual stratospheric concentrations of CCN. A range of 1.2 to 5.3 hours elapsed between the time the sample bottles were filled and their CCN counts were measured.

Sample 1 (Fig. 1A) was taken over western central Montana (46°18'N, 112°25'W) at an altitude of 13.6 km. Although trajectory analyses (6) place this sample slightly outside the northern boundary of the plume from the 13 June eruption, it is difficult to attribute the very high CCN count of this sample to a source other than the volcano. We suggest that the boundary of the plume injected by this eruption may be diffuse enough so that our sample 1 was actually volcanic material. Samples 2 (Fig. 1A; 38°00'N, 120°30'W) and 3 (Fig. 1B; 39°00'N, 106°30'W) were both taken between 18 and 19 km, over central California and central Colorado, respectively. Sample 2 was apparently taken within the widely scattered debris of the 18 May eruption, but trajectory analyses and other data are unable to confirm this. The coordinates of sample 2 were chosen to intercept the 18 May plume after it had passed once around the world. Sample 3 was taken within a portion of the 18 May plume, as judged by supporting lidar and aerosol data. Sample 4 (Fig. 1B; 37°50'N, 120°35'W), which was taken at an altitude of 13.6 km over central California, provides a CCN background count near, but above, the tropopause; volcanic aerosol was not involved.

A continuous-flow diffusion chamber (7) was used as the detection apparatus in all experiments. Sample containers were unloaded as soon as possible after each U-2 flight, and were immediately connected to the diffusion chamber. Precautions, including test runs on blank and mock-up samples, were taken to avoid contaminating the samples with room air. Each sample container was backfilled with particle-free air to bring its internal pressure up to the ambient value required for operation by the continuous-flow diffusion chamber; backfilling continued as each sample was withdrawn from its container through a stainless steel probe at the container's geometric center, at a flow rate of 1 cm^3 sec^{-1}. By varying the operating settings of the diffusion chamber, spectra of CCN active at various supersaturations were taken for each sample.

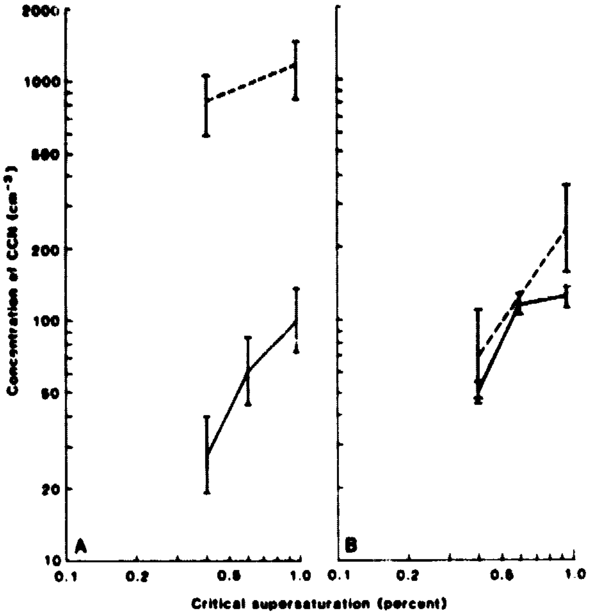

Fig. 1. (A) Measurements of CCN active at critical supersaturation from samples taken on 14 June 1980. The dashed line is sample 1 from 13.6 km; the solid line is sample 2 from 18.6 km. (B) Measurements of CCN active at critical supersaturation from samples taken on 17 June 1980. The dashed line is sample 3 from 18.8 km; the solid line is sample 4 from 13.6 km.

For the CCN spectra obtained from these four samples (Fig. 1), the primary source of error is the uncertainty in the aerosol depletion rate. The background count of the diffusion chamber and residual amounts of room air in the very short lengths of connecting tubing contributed a basic sensitivity level, or background count, conservatively estimated to be 20 to 30 CCN cm^{-3} or less. This background has been subtracted from the results shown in Fig. 1.

The results from samples 1 and 4, both taken at 13.6 km, suggest that volcanic eruptions may temporarily cause the CCN count near, but above, the tropopause to be in the range of 100 to 1000 cm^{-3} active at 1 percent supersaturation, a higher value than that measured in samples from below the tropopause (8). Samples 2 and 3, taken above the tropopause at 18.6 and 18.8 km, respectively, appear to be consistent with data of Rosen and Hofmann (9). The results of samples 2 and 3 differ qualitatively from those of sample 1 because they were taken in volcanic plumes whose aerosols have had a chance to age, and perhaps to coagulate. The CCN counts of samples 2 and 3 are about one order of magnitude above the counts obtained by Rosen and Hofmann (4) after the eruption of Volcan de Fuego in Guatemala in 1974. Our measurements are, however, about equal in magnitude to the tropospheric CCN counts thought typical of maritime conditions below cloud level by Twomey and Wojciechowski (10). Sample 4 represents what may be the background count at altitudes much lower than those of samples 2 and 3.

More measurements are needed to establish the normal CCN background count at these altitudes and the spatial and temporal dependence of the volcanically caused CCN count. Aerosol losses in the sample entry lines are only one mechanism that would cause our results to be an underestimate of true stratospheric CCN counts; further underestimation would result if some of the stratospheric CCN are sulfuric acid particles, which partially evaporate when the sample bottles are warmed as they are brought to the laboratory. Our results, higher by an order of magnitude than anticipated, suggest that volcanoes may be an important source of CCN in the lower stratosphere, both by direct injection of CCN and by contributing precursors for homogeneous nucleation of CCN.

C. F. ROGERS
J. G. HUDSON
W. C. KOCMOND

Desert Research Institute,
Reno, Nevada 89506

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11. We thank W. Page, E. C. Y. Inn, J. F. Vedder, D. O'Hara, and E. Condon for providing laboratory space and considerable assistance at NASA Ames Research Center and for dedicating the four samples to our experiment. We thank E. F. Danielsen for his early analysis of the trajectories of the 18 May and 12 June plumes.

22 September 1980; revised 16 December 1980

APPENDIX B

DRI LETTER TO DR. T. ACKERMAN
NASA AMES RESEARCH CENTER

DESERT RESEARCH INSTITUTE
University of Nevada System
Atmospheric Sciences Center

P.O. Box 60220
Reno, Nevada 89506
(702) 972-1676

May 20, 1981

Dr. Tom Ackerman
Atmospheric Experiments Branch
Building 245
NASA-Ames Research Center
Moffett Field, CA 94035

Dear Dr. Ackerman:

We are writing you in connection with the Aerosol Climatic Effects (ACE) program in which we are involved. For a number of years, our group at the Desert Research Institute (DRI) has been engaged in condensation nuclei studies in the troposphere, and last year we were fortunate to obtain a few grab samples through the efforts of Ed Inn and Jim Vedder for such studies at stratospheric levels. At the suggestion of Dr. Pollack, we'd like to forward a suggestion relative to the discussions scheduled for the next ACE Workshop (May 27 and 28) on possible tropospheric directions for the program. (At least one of the undersigned will attend the Workshop, should you wish further information).

Our work here, specifically, has included over a decade of design and development of instrumentation which can detect, characterize, and count cloud condensation nuclei (CCN), i.e., nuclei active at supersaturations of order 1% and below, typical of natural clouds. Our feeling that good, reliable chambers have evolved was reinforced last year when DRI hosted an International Workshop on CCN detection instruments; absolute accuracies have now improved to the less-than-10%-error range; precision of order 1% has been demonstrated on many occasions.

The idea that ACE would devote some attention to the troposphere is especially interesting to us since most of our work has been in the lower atmosphere where CCN are an important component of the atmospheric aerosol. You are probably familiar with some of the arguments in favor of CCN measurements; we generally view CCN data as having a triple relevance in models of climate change. The most obvious consideration is, of course, that the CCN distribution often determines the initial cloud droplet size spectrum, a direct input to precipitation processes.

Next, we consider the cloud droplet size distribution (or "cloud microstructure") to be a determining factor in calculating the cloud albedo. And finally, another contribution to calculations of the earth's radiation balance comes from the aerosol in cloudless situations, when knowledge of the CCN activity spectrum gives an estimate of how the aerosol absorbs water and enlarges, even in subsaturated conditions. In the paragraphs to follow, we discuss these three effects in sequence.

Cloud droplet number-versus-size distributions result from an interaction between the initial CCN distribution and air motions (updraft, entrainment, etc.). The precise effect of the resulting droplet size distribution on precipitation efficiency is still the subject of research (at our laboratory and many others). However, some aspects of the problem seem to remain clearly in focus, such as the views that high CCN concentrations lead to high cloud droplet concentrations which can then inhibit precipitation formation, and that the observed wide variations in cloud droplet concentrations, both high and low, can largely be accounted for by corresponding variations in CCN concentrations as suggested some time ago by Squires (1958). Thus, CCN are of special relevance because of the wide variations in their concentration (two orders of magnitude).

The relationship between cloud microstructure and cloud albedo has been considered by Twomey (1974, 1977) and Charlock and Sellers (1980). Cloud albedo is, of course, a very important component - perhaps 70% to 90% - of the planetary albedo, and the latest-cited authors concluded that the total influence of all aerosols on the radiation budget is divided roughly equally between the CCN contribution (which influences cloud microstructure which, in turn, determines cloud reflectivity) and all the rest of the non-cloud-forming aerosol.

Finally, we reiterate that analysis of a given aerosol in terms of its CCN activity, gives a measure of the deliquescence (enlargement) of that aerosol as the ambient relative humidity approaches 100%; for pure soluble salts, characterization of the critical supersaturation is totally sufficient, and only slight corrections are needed for mixed soluble/insoluble particles. DeLuisi, et al. (1976) and others have recognized that liquid water condensed on aerosols in subsaturated conditions should be taken into account in radiation balance calculations (this may be more important at higher altitudes where humidities may be elevated though not high enough to produce clouds). CCN measurements are, by definition, taken at standard and well-characterized humidities, allowing the modeler to separate those optical effects due to variations in concentration of the aerosol in subsaturated conditions from those effects due to enlargement of that same aerosol as relative humidities change, again at subsaturations.

In conclusion, then, we are suggesting that the ACE Program consider CCN measurements as part of a tropospheric aerosol measurement program. Chambers for the characterization of CCN (number active at a given applied supersaturation, or critical supersaturation spectrum) have reached the engineering state where it is feasible either to fly them on research aircraft or to install them at field stations on the earth's surface. (Most of our own field experience over the past seven years has been in surface measurements, which are economical and sometimes can be related to measurements aloft, but we are presently constructing for NASA an airborne version of an instantaneous CCN spectrometer.) We suggest that both surface and airborne measurements could be useful in a program concerned with climate change; a similar recommendation was already reached by NOAA's Global Monitoring for Climatic Change Program (Bodhaine, 1979).

Thank you for considering this input; we'd be glad to expand upon it if you'd like. We are looking forward to your presentation on May 28.

Sincerely,

C. Fred Rogers
Assistant Research Professor

James G. Hudson
Assistant Research Professor

CFR/JGH/swr

xc: W. Kocmond
J. Pollack

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APPENDIX C

DESCRIPTION OF INSTANTANEOUS SPECTROMETER

INSTANTANEOUS SPECTROMETER

A. Equipment Description

This instrument was built along the same lines as the DRI continuous flow diffusion (CFD) chamber (Hudson and Squires, 1976). The most important feature of this instrument is that it uses the sizes of the drops detected by the optical counter to deduce the critical supersaturations of the nuclei. Since several size thresholds can be used, this allows the possibility of simultanously determining the number, N, vs. critical supersaturation S_c, for several S_c's. This is difficult in a conventional CFD where the drops usually achieve a nearly monodisperse size distribution regardless of the range of S_c's in the sample aerosol (Hudson, 1976).

B. Theory of Operation

The instantaneous spectrometer, however, contains three supersaturation steps which disperse the drop spectrum over a wider size range. This range is further widened since the sample is exposed to these supersaturations in ascending order. The device, which is shown in Figure 1, is a series of three CFD's inside one chamber. It contains a sequence of three pairs of temperature controlled plates so that a sample aerosol can be exposed to three separate supersaturations (S_1, S_2, S_3). This means that in the first zone only the largest nuclei become activated drops. That is, only those nuclei with S_c's below S_1 grow into droplets while the remaining nuclei remain as unactivated haze drops. After being exposed to this constant supersaturation, these drops approach a monodisperse distribution.

In the next zone nuclei with $S_1<S_c<S_2$ become activated and grow into cloud droplets with similar sizes. In the meantime the drops which were already activated in the first zone grow even larger in the second zone. In fact, their growth rate is speeded up due to the higher driving supersaturation in zone two. Thus, the nuclei with $S_c<S_1$ grow even larger and somewhat more monodisperse and at the end of the second zone a bimodal drop distribution should result. Finally, the third zone activates the smallest nuclei (largest S_c) with $S_2<S_c<S_3$ and a trimodal distribution should result.

The most significant result is not the trimodal distribution but the fact that the drop size spectrum has a wider spread than it has in a CFD. In the spectrometer the drop concentration is less sensitive to drop size and it is easier to discriminate nucleus S_c's based on drop sizes. Therefore, a small change in the drop size thresholds results in a smaller change in apparent concentration than would be the case with the monodisperse distribution in a CFD. Thus it is much more feasible to relate drop sizes to S_c and to establish size thresholds which correspond to certain S_c's. If there were no other factors than S_c affecting drop size, then a trimodal drop distribution with clear separations between modes would always result. In that case, size discrimination could be made between the modes and a definite N vs. S_c spectrum could be made which would correspond to the three supersaturations used in the chamber. In such a case, a cumulative distribution would have three plateaus where the number concentration would be constant over a range of sizes. In the CFD there is one drop size plateau which ensures that all nuclei are activated but that none fall to the floor so that a direct determination of N vs. S_c can be made with S_c being the applied supersaturation in the chamber. There are some situations when the instantaneous spectrometer has three drop size plateaus which then allow direct determinations of N vs. S_c for the three S_c's. However, in most situations the modes are not completely separated (Fig. 2) and instead of plateaus in the cumulative distribution, we find decreases in the slope of N vs. r (Fig. 3). Although this is a much better situation than in the CFD, the lack of a plateau limits the accuracy of direct measurements of N vs. S_c. Accuracy can be increased by setting the voltage thresholds so that the number concentration in the spectrometer matches that in a CFD monitoring the same sample at a specific supersaturation.

Although this can also be done with two CFD's (Hudson, 1976) (where one of the CFD's takes the role of the spectrometer and the other one is used to calibrate the first CFD), the process works much better with the instantaneous spectrometer where there is nearly a constant concentration over some parts of the size range. This considerably reduces the requirement for stability of the various operating parameters. With the instantaneous spectrometer it has been possible to keep the operating parameters constant enough that voltage settings can be used for many days or weeks with continued good accuracy.

C. Description

Figure 1 shows most of the dimensions of the instantaneous spectrometer. The plates are separated by 1.6 cm while the plate width is 29 cm. This chamber was also operated with the plates vertical and sample moving horizontally. The main flow through the chamber was 50 $cm^3 sec^{-1}$ throughout the Workshop. This resulted in a particle velocity of v = 1.62 cm sec^{-1} so that the sample spent about 31 sec in the chamber; 18.6 sec at S_1, 7.4 sec at S_2, and 5 sec at S_3.

As with the DRI CFD, a Royco 225 optical particle counter is used as the detecting device for the instantaneous spectrometer. In addition a 512 channel analyzer (MCA) (Northern Scientific, Inc.) is also interfaced to the Royco to increase particle size resolution so that greater detail in the concentration vs. size spectrum can be displayed.

The plate temperatures were roughly the same for the entire Workshop so that the supersaturations were nearly constant at S_1 = 0.30%, S_2= 0.55%, and S_3 = 0.90%. The droplet size thresholds were set by matching the number concentrations in the instantaneous spectrometer with the concentrations measured with the CFD set at the three different plate temperatures in the instantaneous device. The largest drops corresponded with the lowest supersaturations, etc. All of the drops which could be detected down to the smallest sizes (∿0.2 μm radius water drops corresponded to the number of CCN active at the highest supersaturation in the spectrometer.

Channel 2 was set for about 1.42 μm radius water drops while Channel 3 was set for 1.75 μm radius water drops. A slight number vs. drop size plateau was observed here and the concentration of CCN in the CFD at 0.55% supersaturation (which was S_2 in the spectrometer) was found to be always less than the number of drops in Channel 2 but more than that found in Channel 3 of the instantaneous spectrometer. This meant that nuclei with S_c of 0.55% produced drops within the size range of 1.42 μm and 1.75 μm radius in that particular configuration of the instantaneous spectrometer. Thus, the average of Channels 2 and 3 were used to deduce the number of CCN active at 0.55% in the spectrometer. Channel 4 was set at 2.77 μm radius water drops and Channel 5 was set for 3.0 μm water drops. In a similar fashion, these corresponded to 0.30% S_c. It was found necessary to make a small adjustment in the size thresholds only once during the Workshop.

The sample flow rate was usually the same as the CFD, 0.60 $cm^3 sec^{-1}$, although it was at times as low as 0.1 $cm^3 sec^1$. The plates were also controlled by the same regulator baths and the same types of thermisters were embedded in the plates. This chamber differed from the CFD in three other respects: (1) There were no flows of particle-free air around the backside of the plates. Instead, a diffuser screen was used to eliminate any turbulence; (2) A metal mesh screen was used instead of filter paper for the moist plate surfaces; and (3) Instead of dripping water onto the plates as in the CFD's, water was fed to the metal screens by capillary action from a reservoir of distilled water.

D. Operation

Several tests can be performed to check the performance of the instantaneous spectrometer. When the upstream lowest supersaturation, S_1, is increased, the larger sized droplet peak increases and becomes larger as it should. When the higher downstream supersaturation, S_3, is increased, the magnitude of the smaller sized peak is increased and there is an increase in its size. Under these conditions, the larger sized peak is only shifted to a slightly larger size. These observations are all in keeping with the operating principles. Thus, sizes which allow separations between the peaks can be chosen. Moreover, the Royco voltage thresholds can be set so that certain size channels can be used to monitor the concentration at specific supersaturations. The size channels can be adjusted so that an individual drop size plateau can be obtained for each supersaturation (see Hudson and Squires, 1976). This assures that all drops which should have been activated at a certain supersaturation were activated and counted. Changes in the downstream supersaturation, S_3, do not affect the detected concentration active for instance at S_1 or S_2.

The spectrum of three supersaturations was available simultaneously as soon as the OPC counted and printed out the numbers. Agreement with the DRI CFD was very good and consistent throughout the Workshop as shown by the results.

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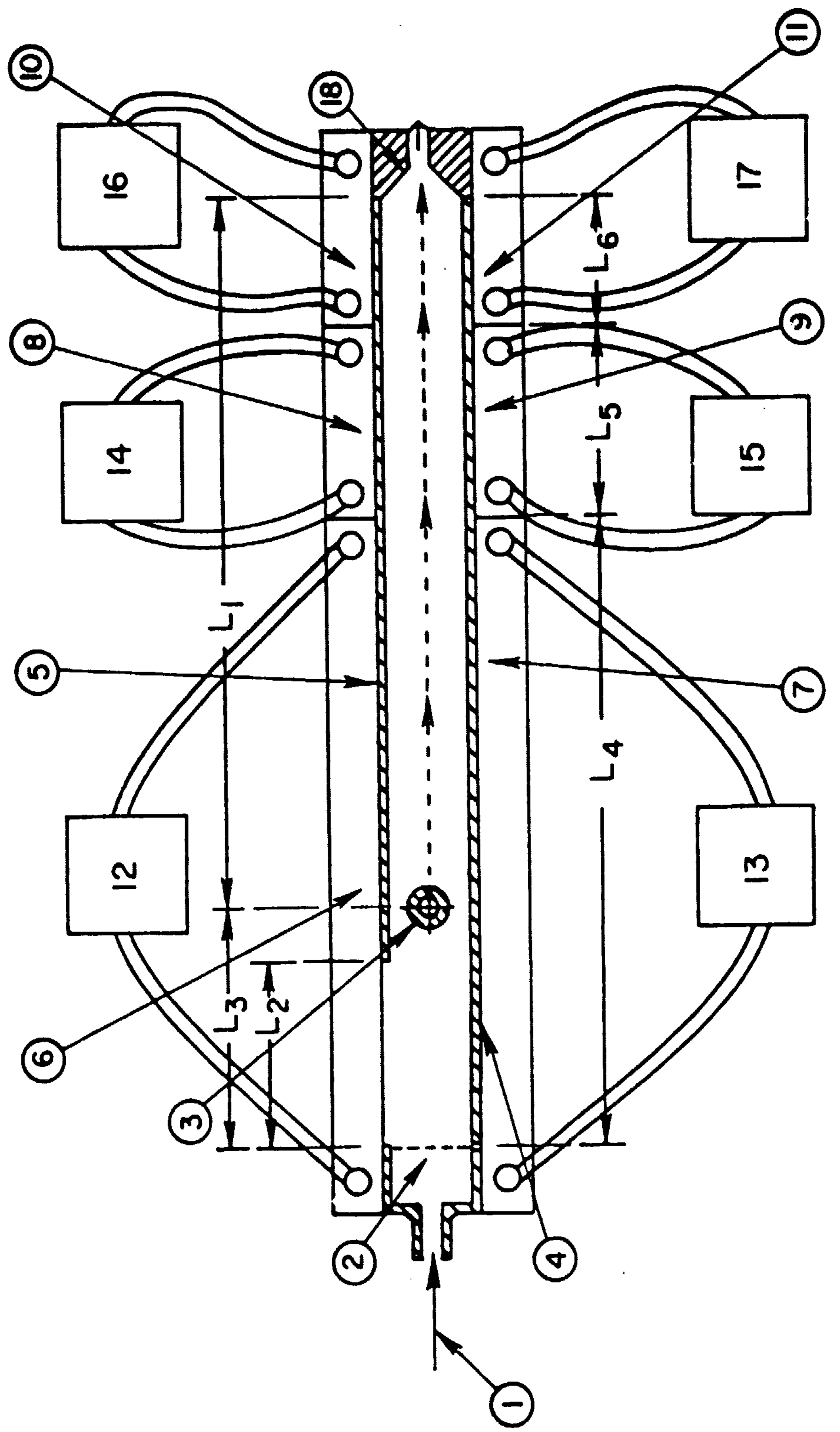

(1) Carrier flow entrance; (2) Diffuser screen; (3) Sample injection tube; (4) Cold plate wicking surface; (5) Warm plate wicking surface; (6) First warm section, T_4; (7) First cold section, T_3; (8) Second warm section, T_5; (9) Second cold section, T_2; (10) Third warm section, T_6; (11) Third cold section, T_1; (12) Temperature bath at T_4; (13) Temperature bath at T_3; (14) Temperature bath at T_5; (15) Temperature bath at T_2; (16) Temperature bath at T_6; (17) Temperature bath at T_1; (18) Exhaust to OPC.

Figure 1. Schematic of the instantaneous CCN spectrometer.

Legend: L_1 = 48 cm; L_2 = 8.4 cm; L_3 = 10.4 cm; L_4 = 38.4 cm (1st supersaturation zone - S_1, T_3, T_4); L_5 = 12 cm (2nd supersaturation zone - S_2, T_2, T_5); L_6 = 8 cm (3rd supersaturation zone - S_3, T_1, T_6).

Supersaturation: $S_1<S_2<S_3$

Plate Temp: $T_1<T_2<T_3<T_4<T_5<T_6$

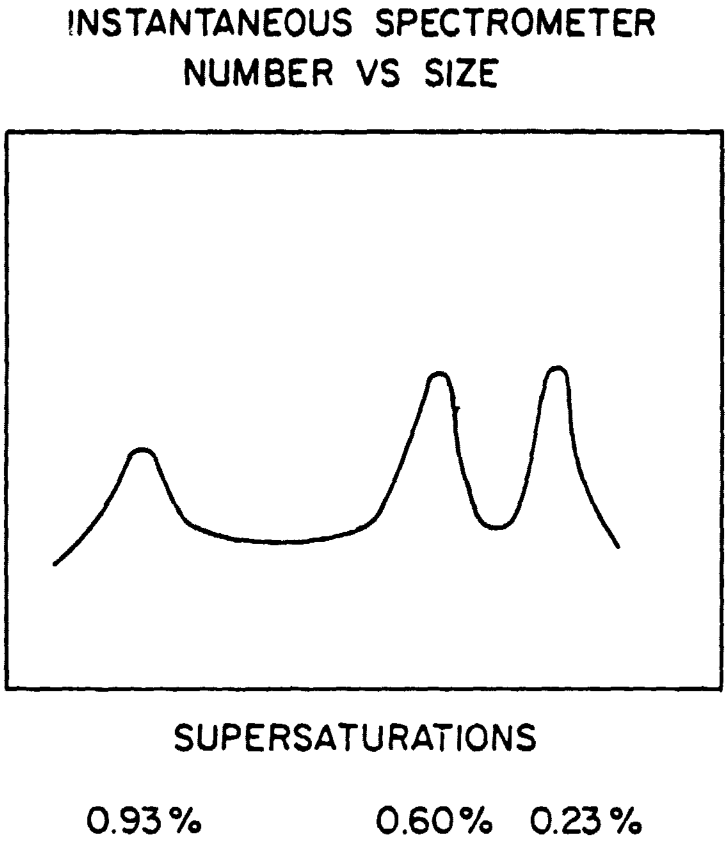

Figure 2. Relative number of drops vs. relative sizes (voltages) for the instantaneous spectrometer. Note that this is a differential and not a cumulative plot.

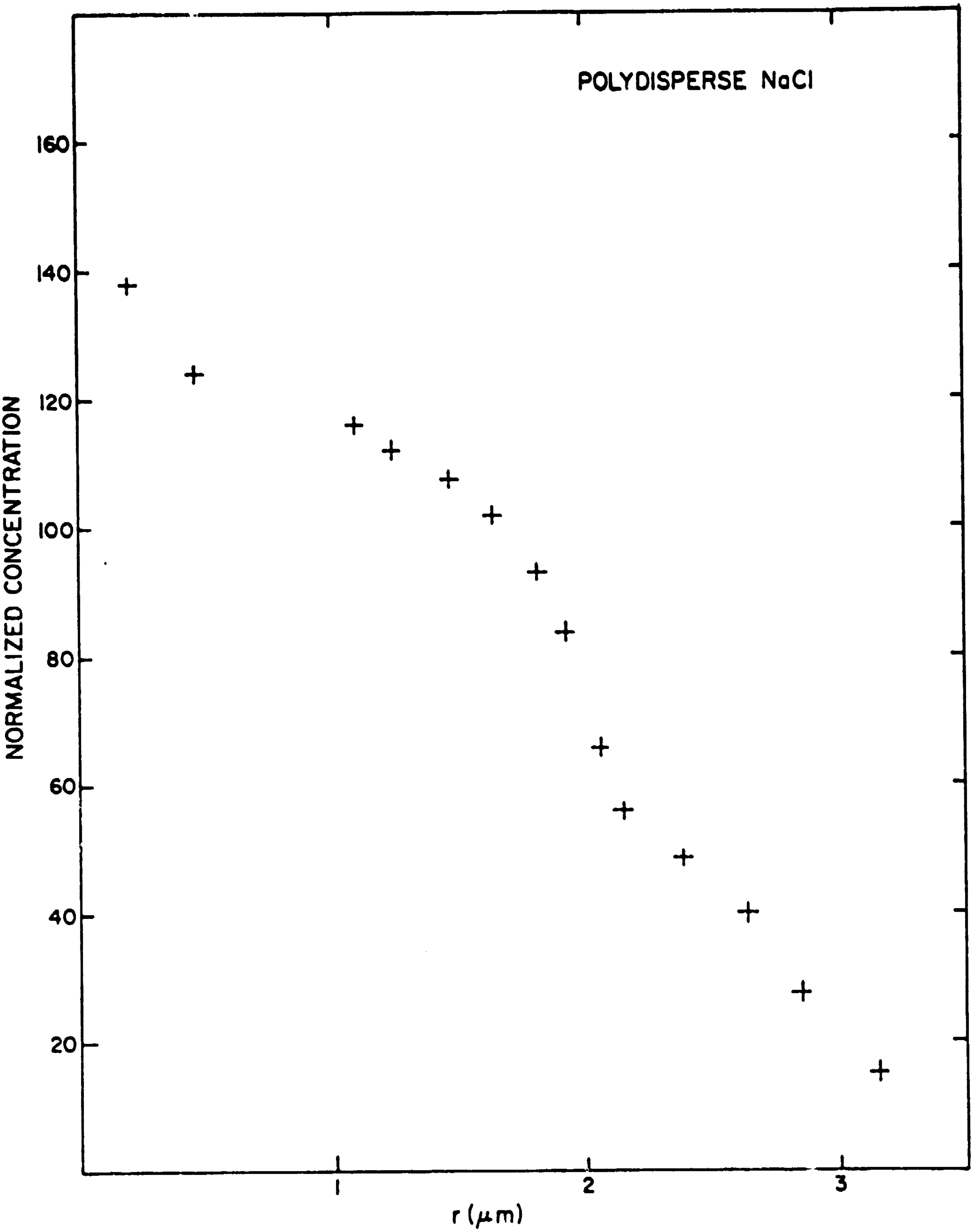

Figure 3. Relative cumulative number of drops vs. size in the instantaneous spectrometer. Applied supersaturations were 1%, 0.4% and 0.15%.