ICE NUCLEI MEASUREMENTS FROM SOLID ROCKET MOTOR EFFLUENTS

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INTRODUCTION

In this paper, a study of ice-crystal-forming nuclei (IN) measured in solid rocket motor (SRM) exhaust products is described together with the involvement of the Naval Weapons Center (NWC) in these measurements. Preliminary results from laboratory investigations and flight preparations for the March 1978 Titan launch are discussed. The final section consists of a statement of future work necessary to provide adequate measurements of IN and cloud condensation nuclei (CCN) in the stabilized ground clouds from SRM's.

BACKGROUND

In the summer of 1976, Dr. W. G. Finnegan of the NWC received a call from the State University of New York at Albany (SUNYA), on behalf of the Institute of Man and Science at Rensselaerville, New York. Dr. Finnegan was asked to burn some propellant in NWC's 24-cubic-meter cloud chamber, which as nearly as possible simulates the properties of the supercooled clouds found in the atmosphere. Dr. F. K. Odencrantz, who performed the burns, measured 5×10^{10} IN/g of Al_2O_3 from the SRM propellant in the laboratory at -20° C. This information was given to SUNYA.

The Space Shuttle will release 10^9 grams of propellant per launch; it may therefore release approximately 10^{19} IN per launch into the atmosphere. The hurricane modification program of the National Oceanic and Atmospheric Administration (NOAA) may release as many as 10^{19} IN per day. Thus, as a comparison, each Space Shuttle launch is potentially comparable to the world's largest intentional release of ice nuclei.

The National Aeronautics and Space Administration (NASA) called a meeting in the summer of 1977 at Estes Park, Colorado, of various atmospheric scientists knowledgeable in the art of nuclei counting. The consensus of that meeting was that measurements were needed to validate the magnitude of the effects of the propellant exhausts and to settle other questions that had arisen through the investigation performed by the Institute of Man and Science. The most critical parameters to be measured were the concentrations of ice nuclei and cloud condensation nuclei in stabilized ground clouds (SGC's) from the SRM's.

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In the fall of 1977, NASA launched a program, with the help of NOAA, NWC, and others, to instrument the NOAA C-130 aircraft at Miami to make measurements of the IN, the CCN, and the liquid particles in the SGC's from the Voyager/Titan III-C launches. Two flights were made, one in August 1977 and the other in September 1977.

In December 1977, the investigators involved met in Houston to report their preliminary findings. The final report of the investigations is being prepared by NWC; hence, the information here is subject to change. The contributions of NWC to the data collection and analysis included preliminary findings from IN and CCN measurements. Ice nuclei in the SGC's were measured using portable IN counters (INC's). The INC's consisted of a small chamber that contained a supercooled cloud similar to one that might exist in the atmosphere. If air that contains IN is injected into the cloud, ice crystals form and can be counted. An independent method of IN measurement, provided by NOAA and SUNYA, used filters to collect particles from the SGC. The filters were later exposed to a controlled, subcooled environment, where particles from the SGC that could serve as ice nuclei did so and were counted. The CCN measurements were made using a small cloud chamber of about 2 centimeters in diameter. That volume contains supersaturated air so that conditions during cloud formation in the atmosphere are simulated. When a sample of air containing CCN is introduced, droplets form and may be counted. The concentration of droplets is related to the concentration of CCN.

During a 5-hour flight, in which the SGC of the Titan launch of September 5, 1977, was observed, measurements were made using portable INC's, CCN counters, and filter devices. Air was captured in bags and taken to a laboratory at the end of the flight. Some 10 hours from launch, the air in the bags was sampled and found to contain active IN. The portable counters onboard the aircraft showed that ice nuclei were present in the cloud in amounts greater than found in the surrounding air, not contaminated by the cloud. The counters were not properly calibrated, and absolute counts were not possible. The filters exposed to the cloud were cut in half. The halves of the filters processed by NOAA personnel supported the data from the portable INC's, but the other half of the filters processed by SUNYA did not. The discrepancy between the results from the filters was not clarified. The CCN counters were saturated for the duration of the flight because the cloud was such an intense source of these nuclei. The consensus of the experts meeting in Houston in December was that the ice nuclei data were inconsistent and inadequate to improve on the analysis previously performed by the Institute of Man and Science.

The Voyager investigators decided on a course of action at that meeting. One item was to do basic laboratory studies of Al_2O_3 ice nuclei. Another item was to calibrate the portable INC's to be used in the airborne measurements following the March 1978 Titan launch at the NASA John F. Kennedy Space Center (KSC). The last item was to measure the concentrations of IN and the physical and chemical properties of the droplets in the SGC from the March launch.

LABORATORY MEASUREMENTS OF ICE NUCLEI FROM SHUTTLE PROPELLANT

Ice nuclei were measured from the effluent produced by burning Shuttle propellant (unpressurized) in the laboratory at Colorado State University (CSU) in Fort Collins, Colorado. Propellant was obtained from the Thiokol plant at Wasatch, Utah.

Objectives

A workshop at CSU was held on February 13 to 18, 1978. The objectives of the workshop were (1) to intercompare the portable INC's that were to be flown in the ground clouds; (2) to compare data from the portable INC's with data from the large, permanent isothermal cloud chamber (ICC) at CSU; (3) to acquire preliminary information on the IN activity of the Shuttle propellant; (4) to collect filter samples for IN and particle analysis similar to that done with the filters from the Voyager launches; and (5) to replicate ice crystals produced in the cloud chamber by nuclei from the propellant exhaust and to study them using a microprobe-equipped scanning electron microscope to determine nucleant composition.

Investigators and Their Instruments

The following investigators were present at CSU with their instruments.

1. Dr. F. Kirk Odencrantz from the NWC was present with a portable Mie INC.

2. Mr. Gerhard Langer from the National Center for Atmospheric Research (NCAR) was present with the NCAR portable INC that he had designed, a filter apparatus, and a portable CCN counter.

3. Dr. Farn Parungo from NOAA (Boulder, Colorado) took filter samples and made ice crystal replicas.

4. Mr. Paul Willis from NOAA (Miami, Florida) came with a portable Mie INC.

5. Dr. Dennis Garvey, director of the CSU facility, conducted the burning of the propellant, ran the ICC, and operated a Gardner condensation nucleus (CN) counter.

6. Dr. G. Gregory from the NASA Langley Research Center brought a portable HCl monitor.

Procedures

A schematic of the test setup in the Atmospheric Cloud Simulation and Aerosol Laboratory at CSU is shown in figure 1. Samples of the propellant, measuring 1 by 1 by 3 inches, were burned in the bottom of the wind tunnel (A). Because of the thrust developed during burning, it was necessary to immobilize the propellant in a small cage. The fan (B) drew air into the tunnel, mixing the exhaust products with the ambient air and diluting it by a known amount. A sample was taken from the top of the wind tunnel by means of a 4-liter syringe (C). A portion of the diluted sample was placed in a 770-liter aluminum storage tank (D), and the rest was put into the ICC (E). A measurement of the ice nuclei concentration was made in the isothermal chamber. Simultaneously, measurements of the concentration were made with the NCAR counter and the Mie counters, using air taken from the holding tank (D).

A supercooled water droplet cloud is formed in a 1-cubic-meter region inside the ICC. The exhaust material from the propellant is injected into the cloud. Ice crystals form and settle onto microscope slides. The slides are immediately placed on the cold stage of a microscope, and the ice crystals are counted. This counting procedure continues as long as crystals settle from the chamber. The crystals are counted on the cold stage and translated into numbers of crystals per liter. Finally, the number of crystals produced from the mass of propellant effluent in the cloud chamber is calculated.

The Mie and NCAR INC's are instruments in which ice crystals are grown in a cloud of supercooled water droplets. The chambers are about 2 feet high and 6 inches in diameter and are filled with a cloud of supercooled droplets. The crystals that are formed in the cloud chambers are drawn (because of the airflow) and settle to the bottom of the cloud chambers. In the Mie instrument, the crystals pass through a polarized light beam. Each crystal depolarizes the light and sends a signal of light to a photodetector. The signals detected by the photodetector are counted. In the NCAR instrument, the crystals pass through a capillary tube, one by one. Each crystal causes an acoustic wave, which is detected using a microphone. The "clicks" detected by the microphone are counted.

Results

Results from the experiments are preliminary. A final report is being prepared by NWC, and the results and conclusions reported here are subject to change.

<u>Cross-calibration of portable INC's</u>.- All the portable INC's measured ice nuclei concentrations from the Shuttle propellant that were above background concentrations; e.g., see the data from the NCAR INC (fig. 2). Counters operated at the same time produced measurements that often correlated; when one counter indicated low concentrations, the other two counters also indicated low concentrations of IN and vice versa (fig. 3). The differences between the numbers of IN measured by the three counters are due to the different response characteristics of the counters. These differences have not been resolved, and to do so would be a major research effort. The

important point is that the portable INC's can and do detect ice crystals produced by Shuttle propellant.

An unexpected phenomenon observed during the workshop was that ice nuclei concentrations first increased with time in the holding tank and then decreased (fig. 2). The IN concentrations should be high initially and then decay with time. When a few tenths of a gram of Shuttle propellant was wrapped with a Nichrome wire, dropped into the holding tank, and flashed off, the portable IN instruments determined the initial IN count to be quite low. Then, the IN counts started to increase with time and increased until finally they plateaued. Three days later, high concentrations of ice nuclei were still sampled out of the holding tank. This phenomenon indicates that HCl may be a factor in either the nucleation or the growth of ice crystals on the Al_2O_3 particles. The effect of HCl is not known; consequently, further work is needed.

Comparison of counters with the isothermal cloud chamber.- When the crystal concentrations were high, as measured by the NCAR counter, they were also high in the ICC and vice versa, as shown in figure 4. It appears from the results in figure 4 that there is a factor of about 1000 difference between the counts measured in the ICC and in the NCAR portable chamber. That is, the crystal concentrations in the ICC were a factor of about 1000 higher than the concentrations measured in the portable counter. Eight data points were collected from the NCAR counter and the ICC that cover a range of concentrations (fig. 4).

The NWC Mie counter produced one comparison point with the ICC (test 19). A difference in concentration of 10^3 between the portable counter and the ICC was observed (fig. 4). Similar results were obtained when the IN counts from the counter were compared with the counts from the NWC 24-cubic-meter cloud chamber. No data on the NOAA Mie counter were received for comparison purposes.

Preliminary ice nuclei activity.- No consistent set of ice nuclei activity figures were obtained, because of a lack of preparation time with the propellant. Nevertheless, the data obtained indicate that the results from the ICC (approximately 10^{11} IN/g of Al_2O_3) support the data obtained in the NWC 24-cubic-meter chamber (approximately 5×10^{10} IN/g of Al_2O_3). Both chambers were operated at -20° C. The most representative experiment run at the CSU workshop resulted in the production of 1.7×10^{11} IN/g of Al_2O_3 in the ICC (i.e., 5×10^{10} IN/g of propellant).

Filter samples.- Filter samples were collected at the workshop and reduced and analyzed at NCAR and at SUNYA. The results of the two analyses were consistent. Ice nuclei were detected in high concentrations on the filters; crystals grew all over the filters. The detection of ice nuclei on the filters supports the NOAA results from the filters exposed to the SGC's following the Voyager launches, but the result is inconsistent with the SUNYA results from the Voyager filters. The IN concentrations from the laboratory filters correlated with the IN concentrations from the NCAR counter.

Ice crystal replicas.- Ice crystal replicas that were collected in the bottom of the ICC showed particles containing aluminum in their centers when examined with the electron microscope. Dr. Parungo indicated that Al_2O_3 particles with traces of sulfur and calcium were responsible for the crystal formations.

CCN concentrations in propellant effluent.- Langer obtained such enormous concentrations of CCN from the Shuttle propellant that his instrument was saturated most of the time. This result is consistent with measurements in the ground clouds from the Voyager launches.

Preliminary Conclusions

All the portable INC's detected ice nuclei from the Shuttle propellant effluent; the results from the counters often correlated. The ice nuclei counts from the portable counters were a factor of about 1000 less than the counts from the laboratory ICC. The ice nucleus activity of the effluent (from unpressurized burns) was measured to be approximately 10^{11} IN/g of Al_2O_3 at -20° C as determined in the ICC. The laboratory results show ice nuclei activity at -15° C. Ice nuclei were detected in high concentrations on filters exposed to the effluent; this result was consistent with the results from the portable counters and the ICC. The concentrations of ice nuclei detected by the NCAR portable counter and by the filters correlated. The counts from the counter were higher than the counts from the filters. The ice crystals formed in the ICC were found with aluminum particles containing traces of sulfur and calcium in their centers; the crystals were probably nucleated by Al_2O_3 particles.

PROPOSED STEPS FOR LABORATORY WORK

The following steps for laboratory work are proposed.

1. Determine the cause of the differences in IN counts between the portable counters and the ICC.

2. Determine whether the ICC can be considered an "absolute" cloud chamber (compare results from seeding natural clouds and clouds in the ICC with Shuttle propellant effluent).

3. Determine the response of the ICC, portable counters, and filters to simulated SGC environments (various combinations of Al_2O_3, HCl, CCN, and water-vapor concentrations).

4. Determine ice nuclei activity of the propellant effluent from pressurized burns (micro-SRM's).

5. Complete the comparison of the Mie portable counter and the ICC.

6. Determine the uniqueness and significance of the traces of sulfur and calcium in the Al_2O_3 IN.

ANTICIPATED FLIGHT MEASUREMENTS FROM THE TITAN III LAUNCH

The objectives of the Titan III flight (to occur after the program review) are to determine the concentrations of ice nuclei in the SGC using the portable counters investigated at CSU and to determine the physical and chemical properties of liquid particles in the SGC.

Investigators and Their Instruments

The following investigators participated in the experiment.

1. Dr. E. E. Hindman of NWC exposed pH paper and obtained ice nuclei concentrations with the Mie INC.

2. Mr. G. Langer of NCAR operated the NCAR INC counter, CCN counter, and filters.

3. Dr. L. Radke of the University of Washington, Chief Scientist aboard the B-23 aircraft (fig. 5), and his crew determined the sizes and the concentrations of liquid particles by means of the particle measuring system (PMS) probes and operated Formvar and foil impactor devices. Standard meteorological variables were measured aboard the aircraft. A side camera was aboard for obtaining photographs, and onboard-navigation data were used to determine, at any instant, the positions of the aircraft and the ground cloud. A condensation nucleus counter, an electrical aerosol analyzer (EAA), optical particle counters (OPC's), and a mass monitor were aboard for obtaining aerosol particle data. A grab sampling system with dilution capability was aboard to obtain air samples for IN, CCN, and aerosol particle measurements. The configuration of the instrumentation aboard the aircraft is shown in figure 6.

Procedures

The flight is planned to be coordinated between an NASA aircraft and the B-23. The two aircraft are to make alternate cloud penetrations. The cloud was to be monitored for a maximum of 7 hours.

Anticipated Results

Anticipated results are concentrations of IN, CCN, liquid droplets, and dry particles in time and space within the SGC as it drifts away from the launch site. The pH of the droplets will be determined. Cloud volume data will be obtained, and the meteorological conditions in the vicinity of the SGC will be recorded.

FUTURE WORK TO PROVIDE ADEQUATE MEASUREMENTS OF CLOUD-FORMING NUCLEI FROM SRM EFFLUENTS

Future work to provide adequate measurements of IN and CCN in SRM effluents includes the following.

1. Complete the proposed steps for laboratory work as previously outlined.

2. Monitor the SGC's produced by Titan and Shuttle launches at Vandenberg Air Force Base and by SRM firings at Thiokol, Utah. Opportunities to collect data other than at KSC will increase our limited knowledge of the behavior of Al_2O_3 ice nuclei in different environments.

3. Determine the effects of the Shuttle propellant effluents on precipitation from atmospheric supercooled clouds.

SUMMARY

Laboratory measurements show conclusively that the Shuttle propellant (burned unpressurized) is a source of ice nuclei; preliminary estimates of the magnitude of the source were obtained. Flight measurements of ice nuclei in Titan-produced stabilized ground clouds are inconclusive; the flight of the University of Washington B-23 aircraft is intended to provide definite measurements. Deficiencies in the laboratory and field measurements are identified. Future laboratory and field measurements are specified to eradicate the deficiencies. These investigations should provide adequate measurements of Shuttle-produced ice and cloud condensation nuclei.

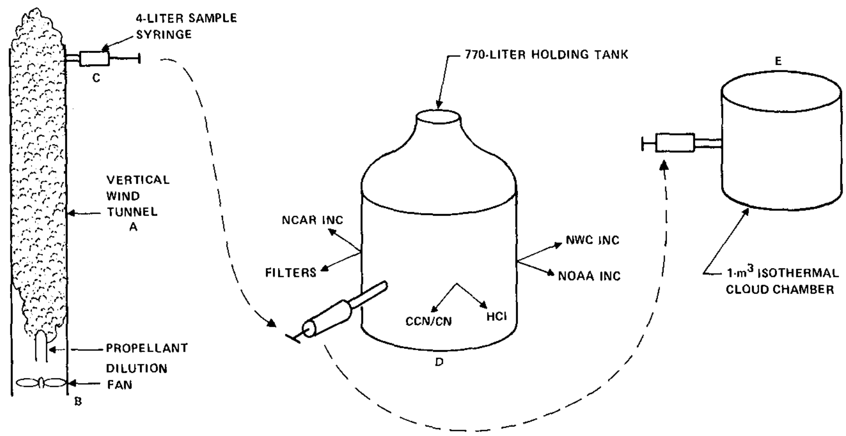


Figure 1.- Schematic of laboratory setup used to calibrate ice nuclei counters (INC's).

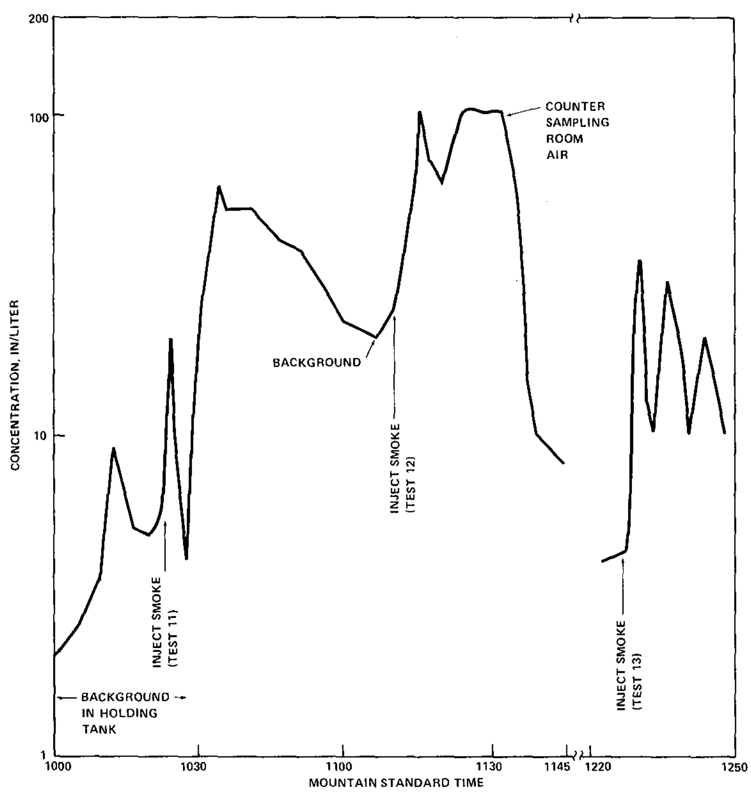


Figure 2.- Ice nuclei concentrations measured at -24° C with the NCAR counter on February 16, 1978. Note the curious increase-decrease-increase in IN concentration with time after injection of Shuttle propellant effluent on test 11.

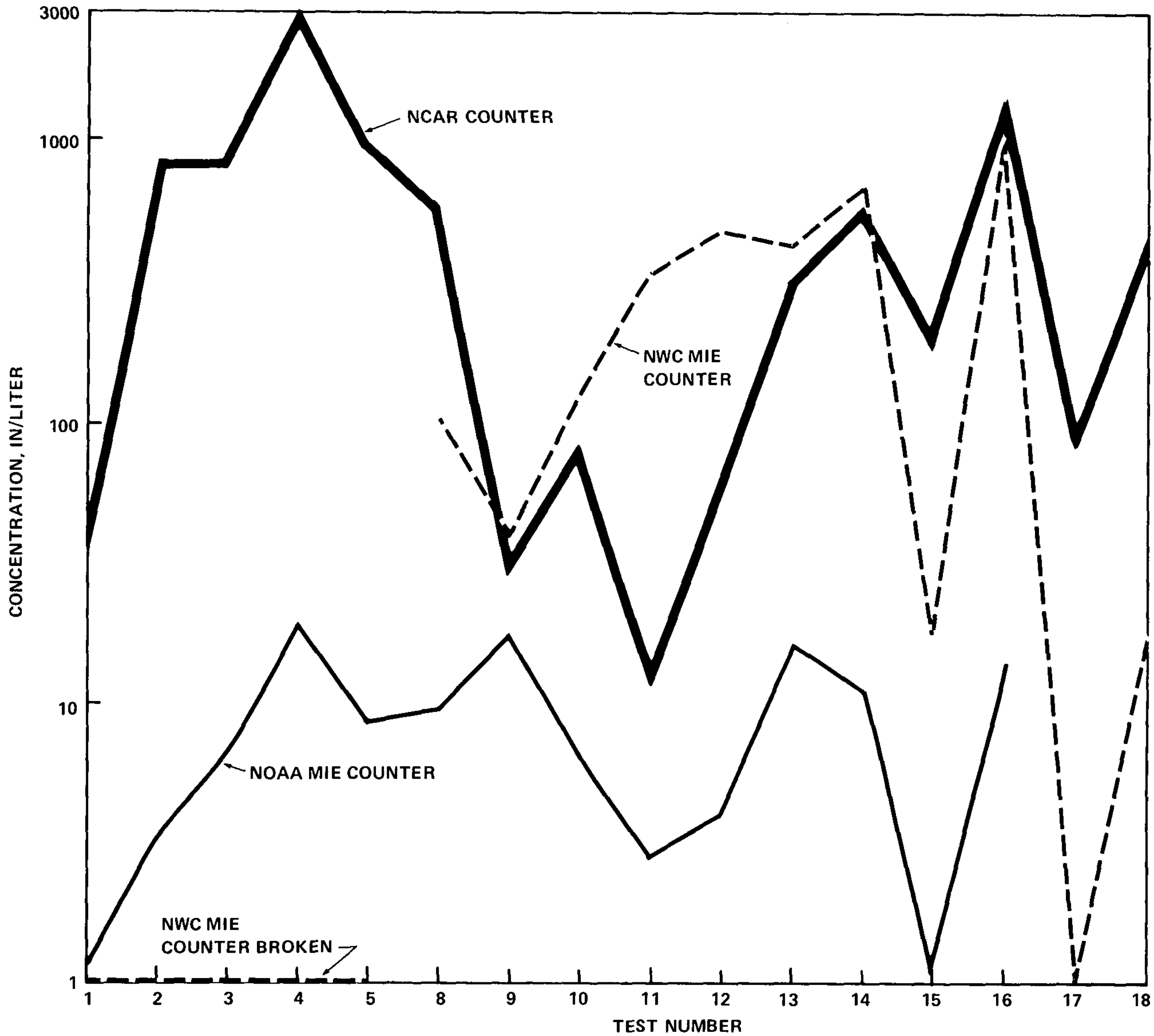


Figure 3.- Ice nuclei concentrations measured 2 minutes after injection of effluent from propellant burns into the holding tank. The values are instantaneous, not average.

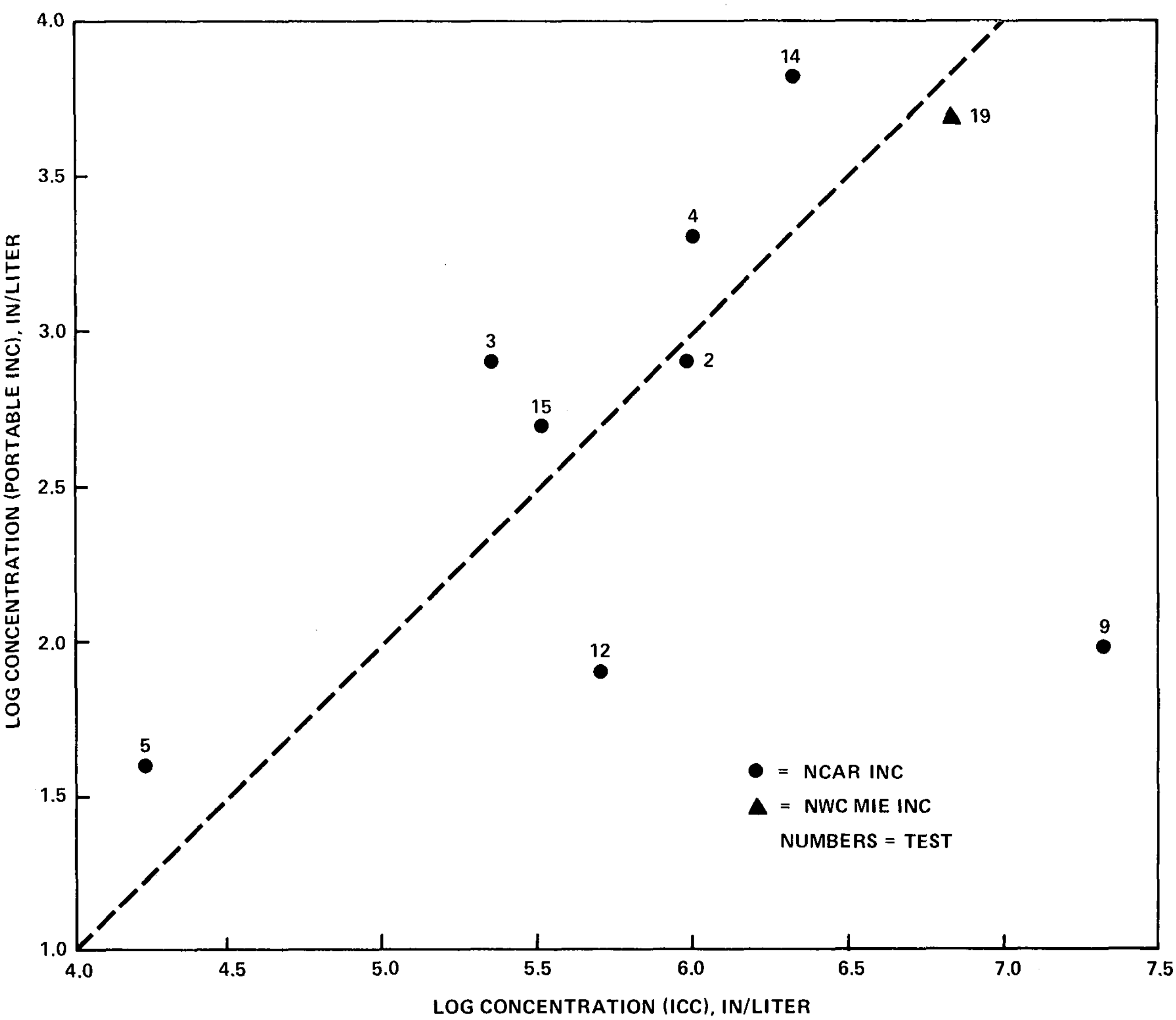
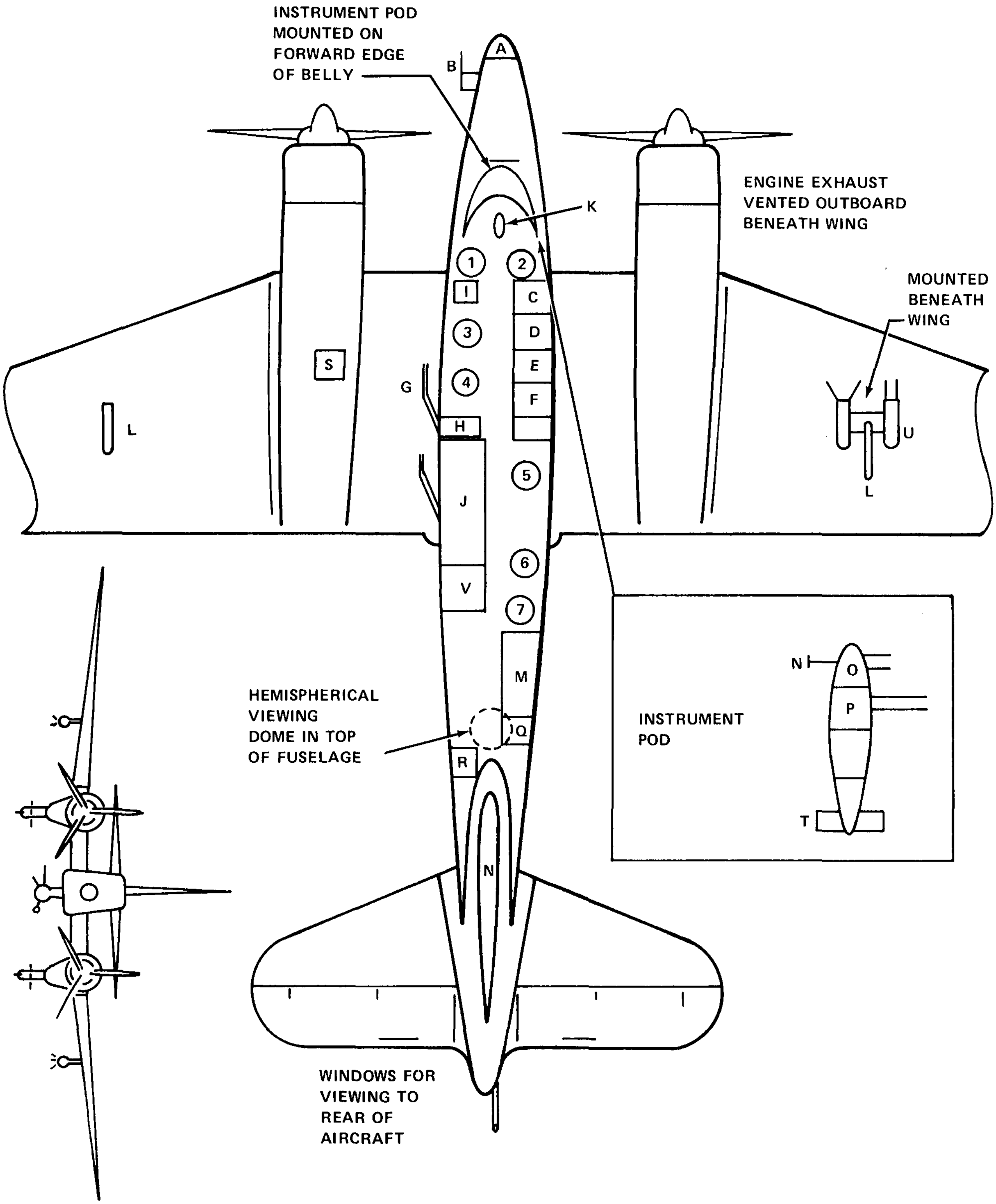


Figure 4.- Comparison of ice nuclei concentrations measured simultaneously with portable counters (NCAR and Mie) and the CSU isothermal cloud chamber (ICC). The dashed line is arbitrary and represents a difference between concentrations measured by the portable counters and the ICC of a factor of 1000.



(a) Detail.

Figure 5.- Plan view of instrumentation onboard the University of Washington Douglas B-23 aircraft.

LEGEND

Locations of crew and research instruments on the University of Washington's Douglas B-23 aircraft:

1-2 Pilot and copilot

3 Observer

4 Instrumentation engineer

5 Aerosol instrumentation monitor

6 Flight director

7 Gas instrumentation monitor

A 5-cm gyrostabilized weather radar

B Rosemount airspeed, pressure altitude and total temperature probes, MRI-turbulence probe and electronics, J-W liquid water probe, angle-of-attack sensor

C Slaved position plotter for the vhf omnirange, distance-measuring equipment, research power panel (3 kW, 110 V, 60 Hz; 1.5 kW, 110 V, 400 Hz; 150 A, 28 Vdc)

D Electronic controls for J-W liquid water indicator, reverse housing thermometer, electrical cloud particle counter and dew point thermometer, time-code generator and time display, WWV time standard receiver, TAS and T_{tot} analog computers, signal conditioning amplifiers, audio signal mixers, phase shift key time-share data multiplexers (63 channels), two-dimensional electric field and turbulence analog readouts, Doppler horizontal winds

E Minicomputer (16-bit word; 16K-word capacity), computer interface to instrumentation, remote analog-to-digital converter, keyboard and printer

F Hybrid analog-to-digital tape recorder (seven track; one-half inch) and high-speed six-channel analog strip chart recorder

G Inlet for isokinetic aerosol sampling

H Aircraft oxygen, digital readout of all flight parameters, dew point sensor, time-code reader and time display, heated aerosol plenum chamber, vertical velocity, millipore sequential filter system

I Controls for metal foil impactor and continuous particle replicator

J Aerosol analysis section, generally containing integrating nephelometer, sodium particle flame photometer, automatic CCN counter, vhf air-to-ground transceiver, electrical aerosol analyzer, optical particle counters, automatic condensation nucleus counter, automatic bag samplers (28-ℓ and 55-ℓ)

K PMS axially scattering spectrometer (small droplet probe) vertically mounted

L Bomb rack hard point suitable for small instrument pods

M NCAR INC

N Reverse flow static temperature probe

O Automatic ice particle counter and metal foil hydrometer impactor

P Continuous Formvar replicator

Q Radar repeater, side-viewing automatic camera real-time display of PMS data

R Radar altimeter, two-dimensional electric field mill electronics 20-channel telemetry transmitter

S Instrument vacuum system consisting of four high-capacity vacuum pumps connected individually to the cabin

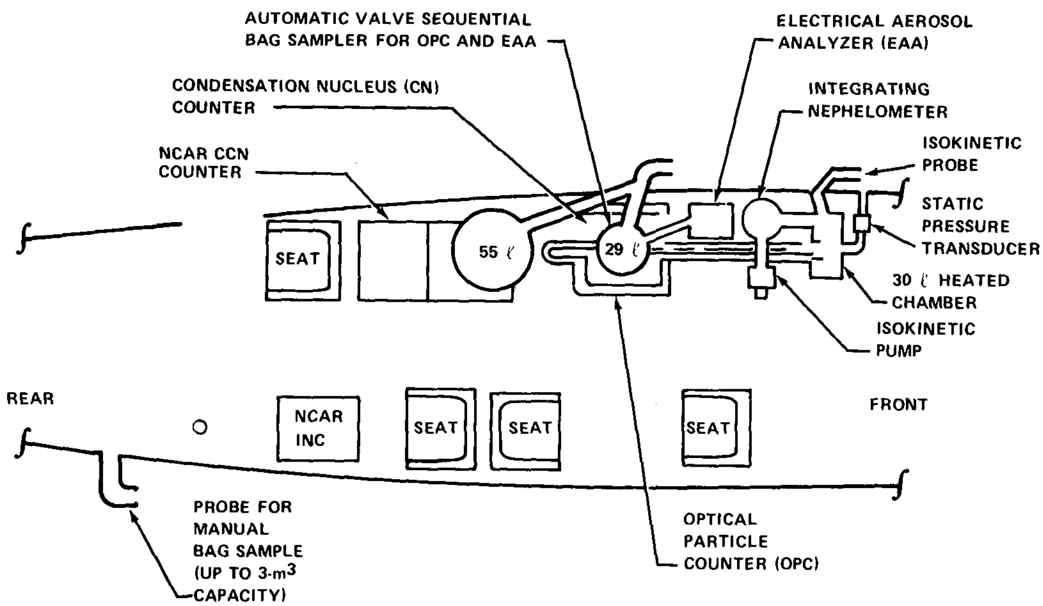
T Anisokinetic large-volume aerosol sampler

U PMS optical array precipitation spectrometer; PMS optical array colored droplet probe

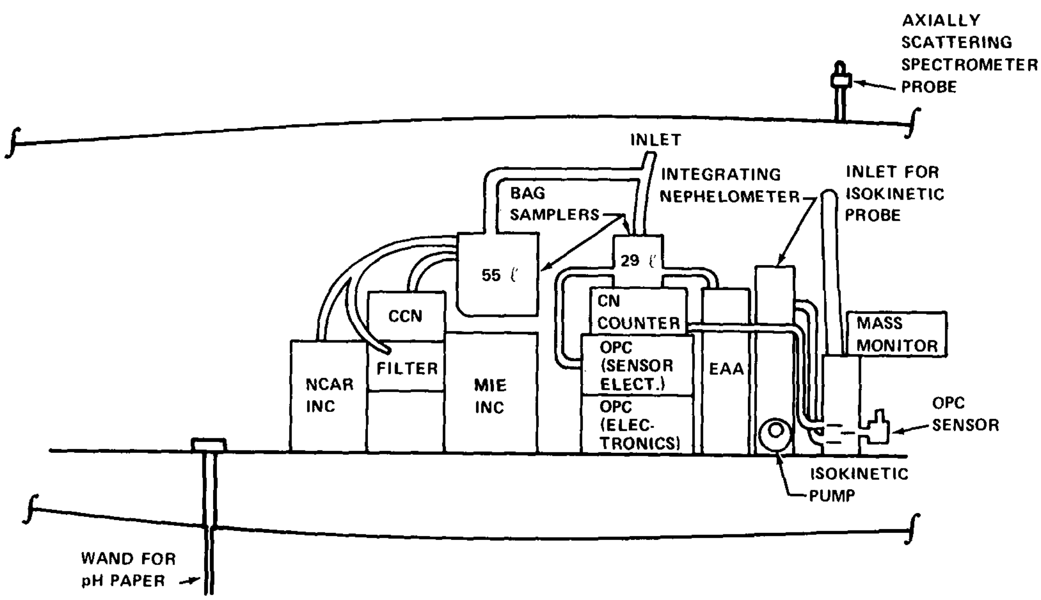
V Mie INC and filter holders

(b) Key.

Figure 5.- Concluded.



(a) Plan view.



(b) Side view.

Figure 6.- Configuration of University of Washington B-23 for March 1978 Titan III launch at KSC.