Summary of Research

for NASA Award NAG 2-1242:

Laboratory Testing and Calibration of the Nuclei-Mode Aerosol Size Spectrometer

Closing Date: 31 July 1999

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I. Introduction

This grant was awarded to complete testing and calibration of a new instrument, the nuclei-mode aerosol size spectrometer (N-MASS), following its use in the WB-57F Aerosol Measurement (WAM) campaign in early 1998. The N-MASS measures the size distribution of particles in the 4-60 nm diameter range with 1-Hz response at typical free tropospheric conditions. Specific tasks to have been completed under the auspices of this award were:

1) to experimentally determine the instrumental sampling efficiency,
2) to determine the effects of varying temperatures and flows on N-MASS performance, and
3) to calibrate the N-MASS at typical flight conditions as operated in WAM.

The work outlined above has been completed, and a journal manuscript based on this work and that describes the performance of the N-MASS is in preparation. Following a brief description of the principles of operation of the instrument, the major findings of this study are described.

II. Principles of operation

The N-MASS is composed of five, parallel CNCs based on an existing instrument, the ER-2 CNC (Wilson *et al.*, 1983). In each of the five CNCs, or channels, the aerosol from the inlet is separated into two streams (Fig. 1). Particles are removed with a filter from one branch of the airflow. This particle-free air passes through a temperature-controlled saturator. In the saturator, vapor from a perfluorinated organic compound (perfluorotributylamine, 3M's Fluorinert FC-43) maintained at 35°C diffuses into the airstream until the saturation vapor pressure at that temperature is reached. The vapor-laden flow is carried to a vertical cylinder, where the remaining aerosol flow, carried through a capillary tube, is introduced coaxially into the center streamline. After a short vertical section in which the Fluorinert vapor diffuses radially into the central aerosol flow, the combined flows pass into a cylindrical condenser maintained at a temperature below the saturator temperature. With the rapid drop in temperature, the vapor supersaturates and condenses on the particles. The particles grow by continued condensation and reach a size that is sensed by a simple optical detector. The instrument produces a count rate of particles that can be converted to concentrations by measuring the aerosol flowrate through the capillary.

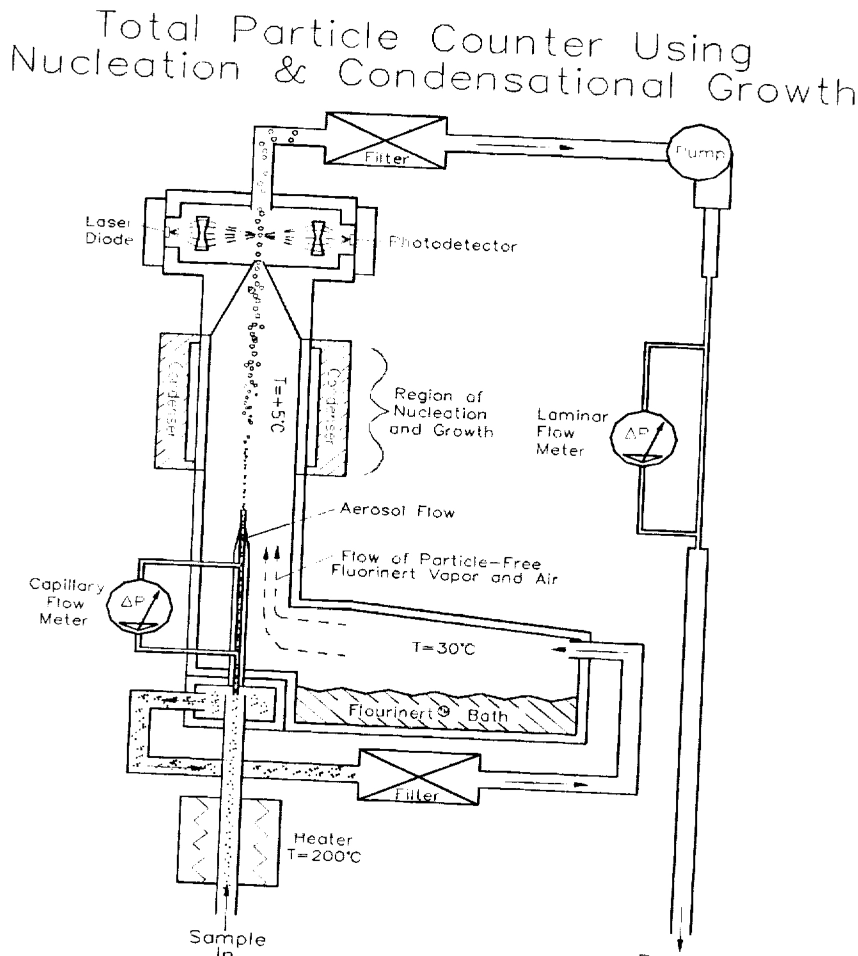

Figure 1. Schematic drawing of a condensation nucleus counter comprising a single channel of the N-MASS.

The minimum particle diameter detected by the each of the five CNCs (channels) within the N-MASS depends on the maximum vapor supersaturation encountered by the particles. At constant pressure and flow, this supersaturation is controlled by the absolute temperature of the saturator and the temperature difference between the saturator and condenser. By fixing pressure and flow and varying the temperatures of the saturators and condensers, each channel of the N-MASS can be tuned to provide a different minimum detectable diameter for particles with diameters <100 nm. These differences in detection efficiency among the five channels can be exploited to provide information on the size distribution of particles in the nucleation mode. A numerical inversion technique is used to account for the non-ideal response function of each channel and to provide a more continuous recovered size distribution over the range of interest.

The five CNCs comprising the channels of the N-MASS operate in parallel and sample from a common aerosol line (Fig. 2). To maintain a fixed internal operating pressure of 60 mb, one of three orifices is chosen depending on upstream, ambient pressure. The losses of particles during transit to and downstream of these orifices is the subject of a portion of this investigation.

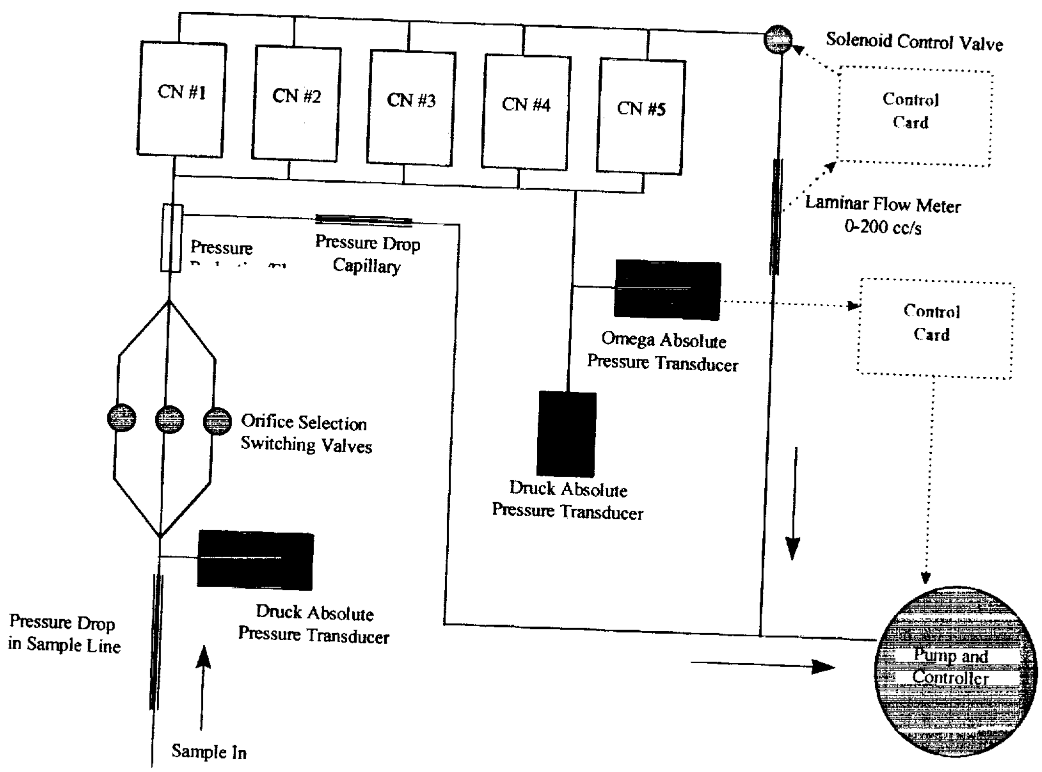

Figure 2. Schematic diagram of the flow system in the N-MASS.

II. Experimental evaluation of sampling efficiency.

The efficiency with which particles were transmitted through the orifice was studied in the laboratory. Particles were generated by vaporizing solid $(NH_4)_2SO_4$ in a quartz tube within a tube furnace, then nucleating and growing $(NH_4)_2SO_4$ particles by rapid cooling of the vapor at the tube furnace exit. The particles were then electrically neutralized by flowing them across a radioactive (Kr-85) source. A differential mobility analyzer (DMA) was used to classify the small fraction of the particles that were charged. The resulting calibration aerosol contained particles with a mean diameter known to within 3% and a diameter distribution width of ±5%.

The concentration of the generated calibration particles was determined with either a Faraday cup/filter assembly attached to a sensitive electrometer (Keithley Model 642) or with a TSI Model 3025A CNC borrowed from W. A. Cooper at the National Center for Atmospheric Research. The electrometer and TSI CNC agreed in concentration to within 8%. The N-MASS simultaneously measured the same calibration aerosol stream as the TSI CNC and/or the electrometer.

The counting efficiency of the first channel of the N-MASS--the channel capable of detecting the smallest particles--was near 100% over only a relatively narrow range of particle diameters, from 30 to 70 nm (Fig. 3). As expected in the design of the N-MASS, diffusion losses for particles with diameters <30 nm were significant. However, inertial losses of particles with diameters >70 nm were not expected, since the flow deceleration section immediately downstream of the orifice--where inertial losses are most likely--was designed following the criteria established by Pui et al. (1990) to minimize such effects. However, the losses observed--less than 10% for particles with diameters near 100 nm--are not expected to significantly affect the size distributions measured reported during WAM. This is because the FCAS, an optical particle counter, counts and sizes particles larger than 90 nm. These FCAS data are incorporated into the inversion algorithm, and dominate the portion of the size distribution with diameters exceeding 90 nm recovered from the inversion.

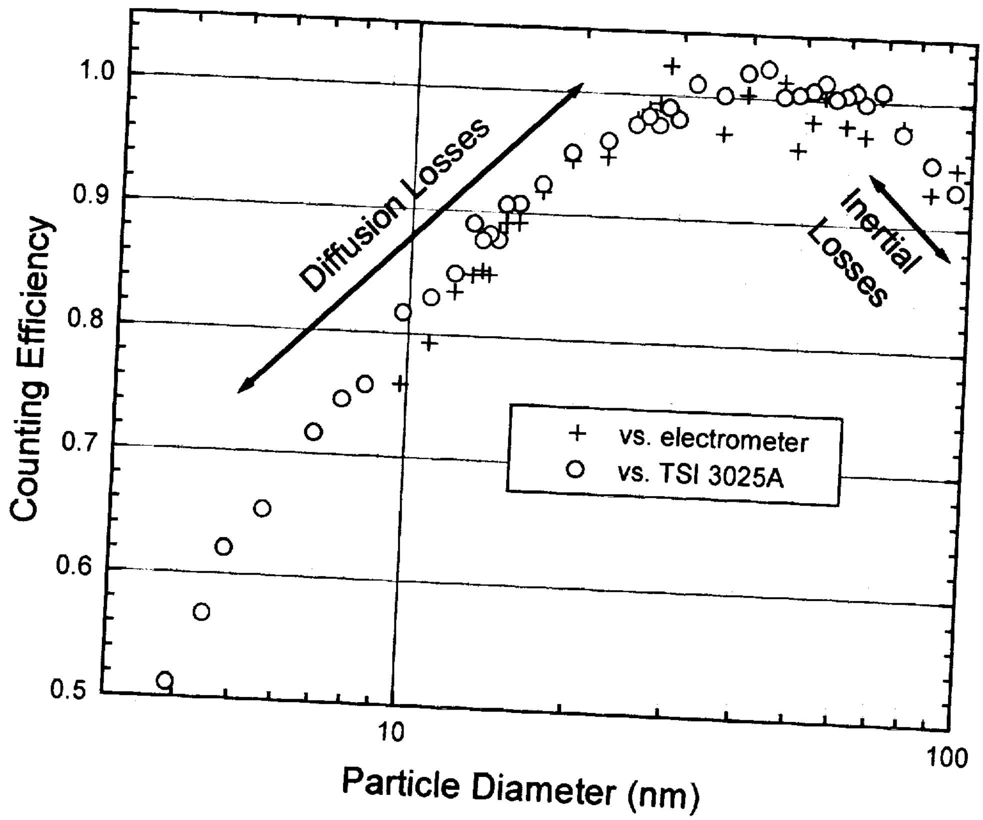

Figure 3. Counting efficiency of channel 1 of the N-MASS compared to and electrometer and to a TSI 3025A CNC. Particle size ranges over which diffusional and inertial losses occur are indicated.

III. Effects of varying temperatures and flows

Tests were made to determine the sensitivity of the size classification of the N-MASS. Preliminary results quickly showed that the sizing was insensitive to flow excursions of ± 20% from the standard value. Greater sensitivity was found to temperature, however. In fact, maintaining the temperatures of the saturator and condenser of each CNC is the principal control problem in the N-MASS. The diameter of nucleation, approximated by the Kelvin diameter, depends upon the maximum supersaturation encountered by aerosol particles flowing through the center section of the condenser. At relatively low values of supersaturation, small changes in temperature difference between the saturator and condenser can produce large excursions in supersaturation (Fig. 4). The result is that,

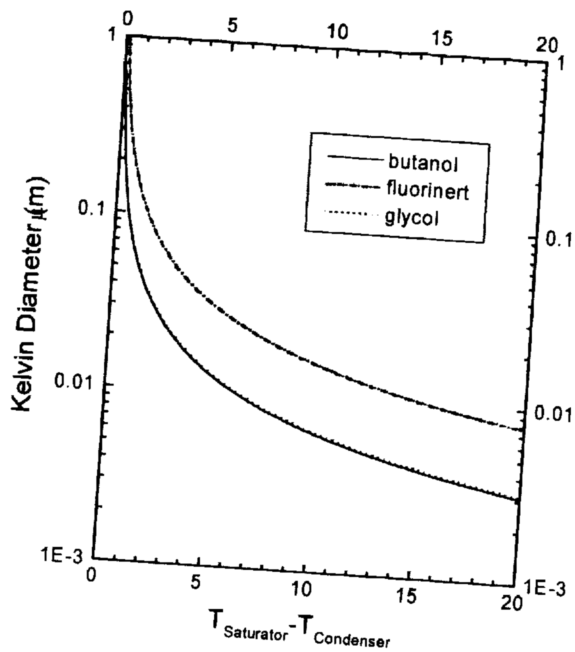

Figure 4. Calculated nucleation (Kelvin) diameter as a function of the temperature difference between the saturator and condenser for three fluids: n-butanol (used by commercial CNCs), ethylene glycol (used by the University of Wyoming's balloon-borne CNC), and 3M's Fluorinert FC-43 (used by the N-MASS). Note that for a given Kelvin diameter, the slope of the curve is smallest for the Fluorinert fluid.

to consistently nucleate relatively large (>30 nm) particles, the temperature difference between the saturator and condenser must be tightly controlled. In the N-MASS, the absolute temperature of each of the five saturators is monitored by two high-precision thermistors per channel and maintained by resistive heaters. The power to each heater is controlled by a custom rate-of-change proportional feedback control circuit. The temperatures of the condensers are maintained by thermoelectric (Peltier-type) coolers and governed by circuits that controls the difference in temperature between the saturator and condenser. With this system, the temperature of the saturator and the temperature difference between the saturator and condenser can be maintained to within ± 0.1 °C over a wide range of ambient thermodynamic conditions.

An experiment was performed to test the sensitivity of the diameter of nucleation for channel 5 (which nucleates the largest particles) to variations in the temperature difference between the saturator and condenser. As in the experiment studying sampling efficiency, a calibration aerosol was generated, and an electrometer and/or a TSI 3025 CNC was used to monitor the particle concentration. The saturator of channel 5 was maintained at a constant temperature, while the condenser was cooled by a varying amount. The diameter of the calibration aerosol was then adjusted until channel 5 reported 50% of the particle concentration measured by the other 4 channels. At a saturator-condenser ΔT of 7.5 °C, the diameter of 50% detection, Dp_{50}, was 54 nm (Fig. 5). Within ± 0.5 °C of this value, diameter, the system exhibited a sensitivity of approximately 20 nm/°C. Given the stated temperature control of ± 0.1 °C, Dp_{50} for channel 5 is 54 nm ± 2 nm, or 4%.

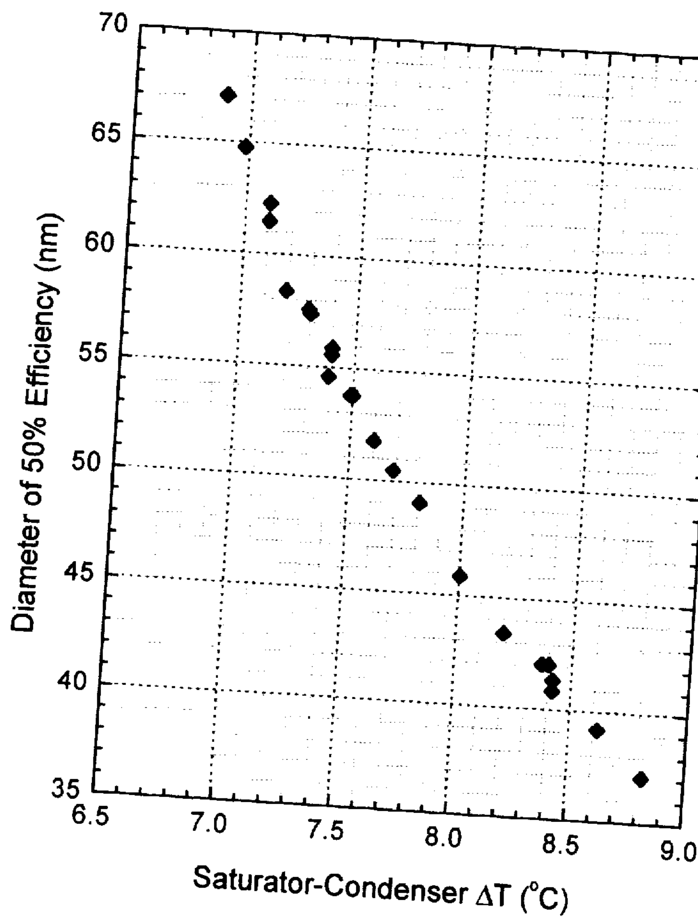

Figure 5. Diameter at which channel 5 of the N-MASS counts particles with 50% efficiency as a function of the difference in temperature between the saturator and the condenser. This temperature difference controls the supersaturation, which controls the Kelvin diameter (Fig. 4).

IV. Calibration of the N-MASS at flight conditions

The calibration of the N-MASS is expected to shift with changing pressure as the heat and mass transfer that control the vapor supersaturations in each of the condensers change. For stratospheric missions, the internal pressure of the N-MASS is maintained at 60 ± 5 mb regardless of flight conditions. As a result, only a single calibration, describing counting efficiency as a function of particle size, needs to be completed for each channel.

Using the same experimental setup as described in sections (II) and (III) above, each channel of the N-MASS was calibrated with a large number of laboratory-generated particles of known size, composition, and concentration while the instrumented operated at its flight-standard internal pressure of 60 mb (Fig. 6). When using the electrometer, particles with D_p<9 nm could not be measured because the low particle concentrations produced poor signal/noise for the electrometer current. Particles with D_p<4 nm could not be produced at measurable concentrations. Thus, the counting efficiencies for channel 1 for particles with D_p < 4 nm could not be determined. The commonly reported "figure of merit" for CNCs, the diameter of 50% counting efficiency, is ~4, 7, 14, 28 and 54 nm for the five N-MASS channels.

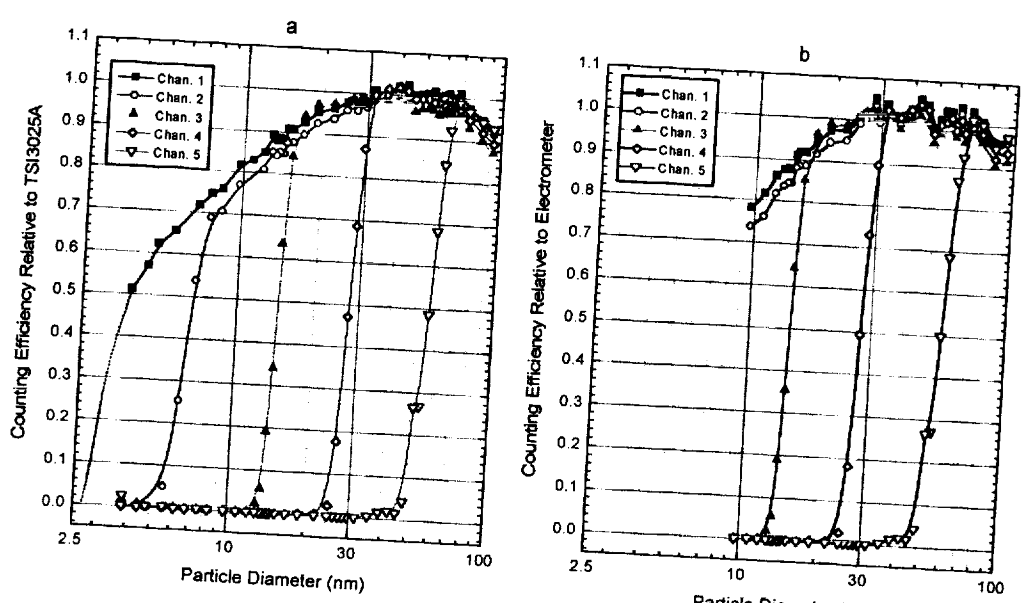

Figure 6. Counting efficiency of each channel of the N-MASS as a function of particle diameter as determined with (a) a TSI 3025A CNC and (b) an electrometer. The internal pressure of the N-MASS was 60 mb, its stratospheric flight-standard operating condition.

VI. Summary

The performance of a new nuclei-mode particle sizing instrument, the N-MASS has been studied. Inertial losses of particles are larger than was anticipated in the design phase of the instrument, but these losses--less than 10% for particles with diameter near 100nm--will not significantly affect the data quality when the measurements are combined with an optical particle counter. The worst-case sensitivity of the instrument to variations in the thermodynamic conditions in the condenser is relatively small. Given the precise regulation of the thermal control circuits, the expected variations in particle sizing should be <4%. Calibration of the N-MASS by two standard technique--a TSI 3025A condensation nucleus counter and a Faraday cup and electrometer assembly--show very good agreement. The diameters of 50% counting efficiency are ~4, 7, 15, 28 and 54 nm for the five N-MASS channels. This research effort has demonstrated that the N-MASS is a robust system that can be calibrated with relatively common laboratory techniques.

References

Pui, D. Y. H., K. L. Rubow, J.-K. Lee, and B. Y. H. Liu, Design of pressure reducing devices for high purity gas sampling, *Swiss Contamin. Control*, **3**, 408-412, 1990.

Wilson J.C., Blackshear E.D., Hyun J.H., The function and response of an improved stratospheric condensation nucleus counter, *J. Geophys. Res.*, **88**, 6781-6785, 1983.