10.1 REAL-TIME PROCESSING AND CHARACTERIZATION OF ATMOSPHERIC PARTICULATES

John Hallett*, James G. Hudson, Douglas H. Lowenthal, and Rick Purcell
Desert Research Institute, Reno, Nevada

1. INTRODUCTION

From an historical perspective, real time processing of atmospheric particles is not new. The Aitken counter is over a century old, Aitken (1923); it involves an immediate expansion of aerosol containing moistened air and counting of resulting particles as they grow and fall out under gravity on a plate having an engraved scale. Labor intensive? Yes. Difficult to obtain long data sets? Yes. A controlled expansion was possible and in principle could lead to a supersaturation characterization. But practicality here prevailed and a new age and a new technology was to be developed before such control and large data sets, to be set against the extreme variability of atmospheric aerosol in a turbulent atmosphere, could become reality. Yet the early instrument, while clearly demonstrating the need for such technology, did provide insight into physical properties of aerosol at a given point and a given time.

2. CRITERIA FOR INSTRUMENT DESIGN AND DATA ANALYSIS

2.1 *Aerosol*

Knowledge of the properties of individual atmospheric particles is important for understanding their origin, evolution, and fate in clouds and precipitation. Instruments are evolving that enable specific properties to be measured in near real time. Such approaches are advantageous because the particle mixing state is stable and chemical reactions are minimized and phase changes can be controlled. Processing may be achieved by particle loss through volatilization and oxidation of organic and inorganic compounds (e.g., water, sulfuric acid, ammonium sulfate, sodium chloride) as a processing furnace increases sample temperature followed by optical or other detection, Hudson and Da (1996). It may be achieved by controlled increase of supersaturation at controlled temperatures and measurement of hygroscopic particle growth, as in a water vapor diffusion chamber, both for ice and water nucleation, Hussein and Saunders (1984), Hudson (1989), Rogers (1994). Irreversible preprocessing of aerosol from its environment as it reaches the detection site of the instrument must be avoided, as in the heating or drying of ice nuclei which could destroy ice preactivation which exists at lower temperature and higher relative humidity.

Corresponding author address: Dr. John Hallett, Desert Research Institute, Division of Atmospheric Sciences, 2215 Raggio Parkway, Reno, NV 89512; e-mail: hallett@dri.edu.

2.2 *Ultra Giant Aerosol and Precipitation*

Larger particles, drizzle, rain or ice, are first captured on a preheated optical surface. This can be accomplished by collection on a forward facing optical flat under flight conditions, or in field or laboratory under controlled ventilation. Subsequent evaporation and/or melting of the particle are video-recorded for immediate or subsequent analysis. This is the basis of the cloudscope (Figs. 1-3), an airborne instrument, Arnott et al. (1995), Hallett et al. (1998); simultaneous forward and side viewing provides a record of the changing geometry. Alternatively, growth by controlled cooling (to produce a supercooling or solute supersaturation) and freezing may be examined. It is convenient to process particles following collection at or near the forward stagnation point of the forward facing optical flat. The dynamic temperature increase is directly related to airspeed and can be computed and also measured directly. Ice particles are imaged from behind and from the side and the change in volume by evaporation and melting can be tracked. An ambient mixing ratio, measured or assumed, provides the additional boundary condition necessary to compute mass loss from the particle and enables the density of the particle to be inferred (less dense particles evaporate more quickly). Changes of density for a particle can readily be observed as layers of ice are progressively stripped from the upper surface during evaporation. A parallel application is seeking the presence of insoluble and soluble components of ice and liquid particles. The former remain as a residue after complete evaporation; the latter are to be identified in terms of its hygroscopicity (defined in terms of the activity per ion) through solute crystallization following evaporation, Meyers and Hallett (2001). This gives the opportunity to seek the nature of supersaturated solutions through their rate of crystallization as evaporation occurs. As an alternative strategy, large hygroscopic aerosol captured dry may be induced to grow by moistening or active cooling rather than heating of the window and their hygroscopicity thus determined from a knowledge of the ambient relative humidity at the stagnation point. Results suggest the universal presence of hygroscopic particles between the tail of the CCN spectrum, beyond 0.2μm dry diameter and the optical resolution of the cloudscope >1μm dry diameter that could be important in the initiation of precipitation. At present such particles are not well measured or characterized. Present measurements show that both CCN and ultra giant aerosol are highly variable in concentration as is their occurrence in drizzle and cloud elements collected under maritime, continental, and anthropogenically-influenced environments.

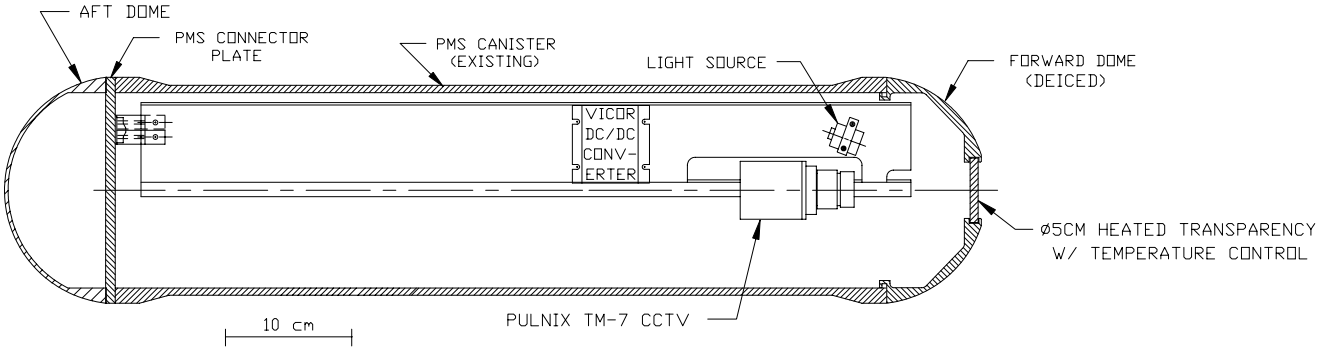

Figure 1. Large Format Cloudscope with 2.5 cm forward facing sapphire window. Sample volume 5 – 50 liters per second.

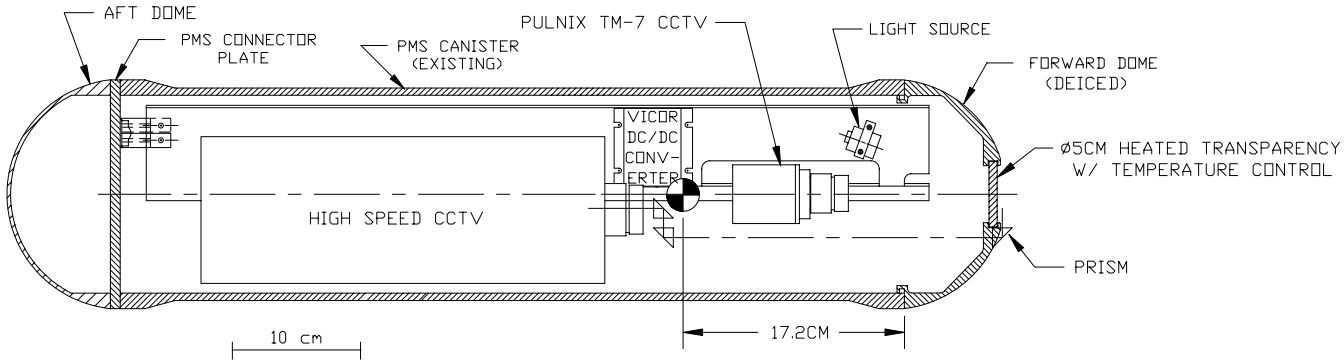

Figure 2. Large Format Cloudscope with high speed side looking CCTV.

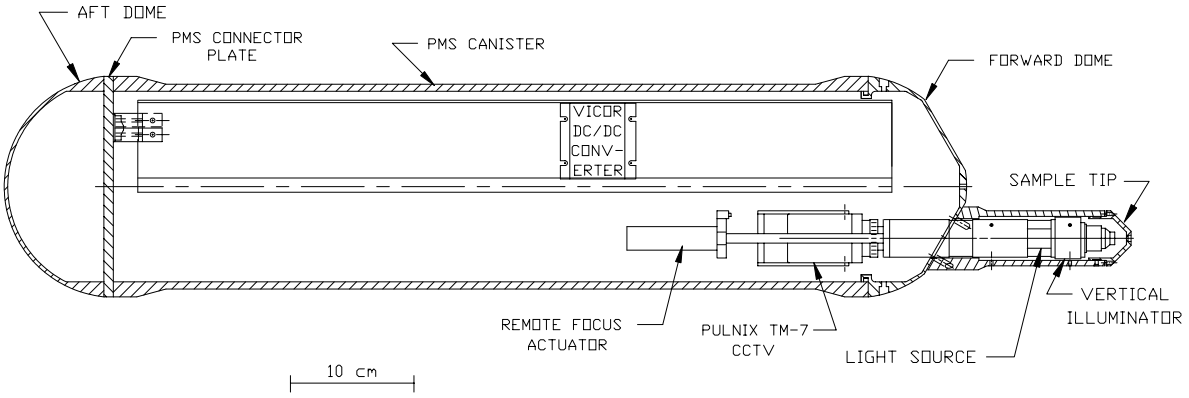

Figure 3. Small Format Cloudscope with 0.3 X 0.3cm sapphire window field of view and sample volume 25 - 50 cm^3 s^{-1}. Instruments are readily exchanged in a standard aircraft-mounting pod.

Examples of such analyses from field observations suggest measurement strategies to yield information on hygroscopicity, nucleus concentration, and solubility.

3. PRACTICAL CONSIDERATIONS

Collection of particles poses several problems. A cylinder may form the leading section of a collector system from aircraft, facing into the airstream and driven by the dynamic pressure, the flow controlled at the exit at the rear of the aircraft. The system may or may not approach isokinetic conditions. A ground system driven by a down stream low pressure pump serves the same purpose and needs to meet the same requirements. The systems may or may not approach isokinetic conditions as collection conditions vary. A short cylinder (as an FSSP) may have organ pipe resonances driven by the passage time of the air through the tube, and be highly susceptible to oscillating reverse flow at the leading edge at low angles of attack.

Collection on the leading edge of the cloudscope optics favors larger particles as the flow divides on approach to the stagnation point and smaller, lower density particles are diverted around the collector. Thus the collection efficiency depends not only on the ambient velocity, particle density and size but also on the aerodynamic drag of the particles. An advantage of the cloudscope is that the particles shape is determined through the image. Large particles may be deformed as they impact, Hallett and Isaac (2002) and the derivation of density may need adjustment should such squishing or breakup occur.

Perhaps the most difficult problem for assessment of the larger, rarer, particles is the uncertainty introduced by Poisson statistics. The sample volume is key here and any determination that particles are randomly distributed in space must assess this uncertainty for a minimum number - say 100 - for a 10% uncertainty in each size interval of interest. This implies a longer sampling time for the larger, rarer particles and provides major uncertainty to the low concentration tail of any particle spectrum (Figure 4).

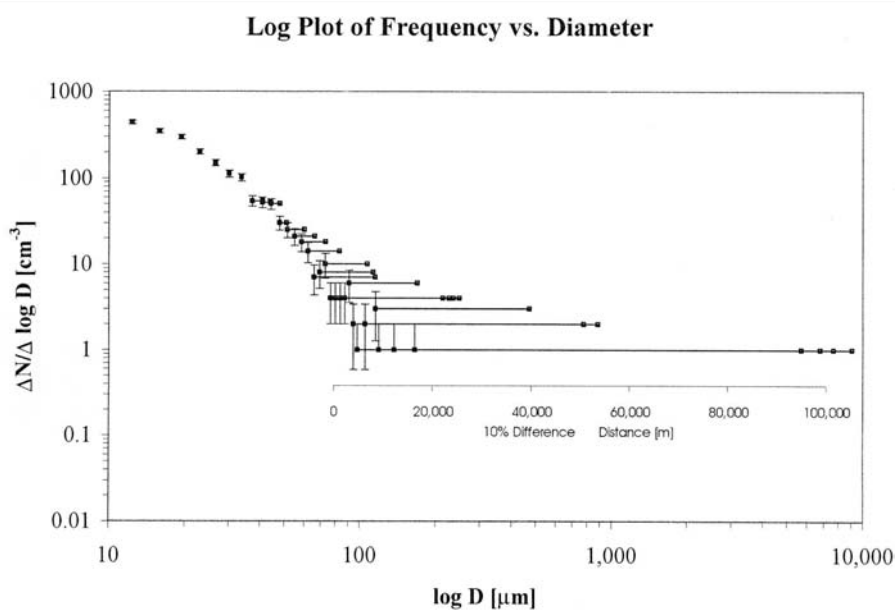

Figure 4. Analysis routine for an ice particle size distribution collected by an airborne cloudscope (an instrument which images particles collected on a forward facing optical flat), sample volume about 5 liters per second. The protocol is set initially by the selection of the number of size bins on a logarithmic scale. It is further set by the level of uncertainty which can be tolerated - for example ± 10%. The uncertainty in the number of particles actually counted in each bin (N) is given by Poisson statistics as $N^{1/2}$, represented by the **vertical** error bars on each point. The uncertainty is obviously large for the small concentrations of large particles. The **horizontal** line from each point represents a flight distance necessary to sample 100 ± 10 particles (10% uncertainty) at the concentrations observed using the instrument of designated sample volume. The physical domain over which any set of measurements must be analyzed is then selectable. This could be (as this case) 10 km of a hurricane outflow for a specific size, but would be something like 1000 km for marine stratus or the whole Northern Pacific Ocean for frontal systems for larger rarer particles. In order to reduce the sampling uncertainty, it would be necessary to use an instrument with a greater sampling volume or sample for a longer time/distance in regions of interest delineated by other considerations - for example energy dissipation rate or radiance at a given wavelength.

Data collected on the NASA DC-8 in outflow of hurricane Earl approaching the Louisiana coast, 2 September 1998.

Cloudscope design can vary from a sample area dimension of 0.5 mm to 10 cm; resolution obviously degrades with increased sample area. The compromise leads to a lower limit of a few μm to a few 100 μm over this range.

Other things are also sometimes not so simple. Furnace processing of aerosol to high temperature >900°C suggests the production of nanometer-sized particles under non-steady-state conditions during processing. This has implications for the techniques used to characterize such particles. The use of preheating as an analytical tool has provided a simple approach to identifying the composition of aerosol and seeking the critical temperature at which a significant decrease in aerosol concentration as measured by a CN counter occurs, Jennings and O'Dowd (1990), Jennings et al. (1994). A recent finding has demonstrated that under some conditions nanoparticle concentrations substantially increase in association with such heating particularly above 300°C. This suggests that the outlet region of the furnace is cooling at such a rate that the diffusion of the vaporized material to the exit walls is slow enough that in situ nucleation occurs should the lateral temperature gradient be sufficiently large and is confirmed in several different geometries (Matt Meyers, student project; private communication A. Strawa).

The detail of the image analysis for these measurement strategies needs to be carefully worked out and employed since manual analysis, although initially necessary, becomes impractical for large data sets. The advantage of such approaches are considerable in as far as the detailed microphysics becomes much clearer and leads the way to assembly of data sets through judicious modeling approaches.

4. CONCLUSION

- It is suggested that real time processing of atmospheric particles can yield information on their properties which determine their behavior in the atmosphere and which cannot be obtained in other ways.

- For aerosol, the measurement of changes following heating, supersaturation or drying yields criteria for their subsequent behavior as the particle trajectory follows similar changes in relation to cloud geometry and their importance in precipitation.

- For larger ice particles, density can be inferred through the rate of change of volume which determines the particle total mass and, through an inferred fall speed, the vertical mass flux.

- The challenge lies in design of instrumentation to capture and modify the particles in a prescribed manner related to both specified atmospheric processes and changes for diagnostic purposes. The challenge also lies in developing data systems to record these changes in order to be capable of automating the analysis and its presentation.

ACKNOWLEDGEMENT

Work supported in part by grant ATM-9900560, Physical Meteorology Program, National Science Foundation, Arlington, Virginia.

REFERENCES

Aitken, J., 1923: Collection of Scientific Papers. Cambridge Univ. Press, London and New York.

Arnott, W.P., Y. Dong, R. Purcell and J. Hallett, 1995: Direct airborne sampling of small ice crystals and the concentration and phase of haze particles. *9th Sym. on Meteorological Observations and Instrumentation*. Charlotte, North Carolina, pp. 415-420.

Hallett, J. W. P. Arnott, R. Purcell and C. Schmidt, 1998: A Technique for characterizing Aerosol and Cloud Particles by Real Time Processing. *$PM_{2.5}$: A fine particle standard, Proceedings of an International Specialty Conference,* Sponsored by *EPA Air & Waste Management Association,* Ed. By J. Chow and P. Koutrakis, 1998, **1**, 318-325.

Hudson, J.G., 1989: An instantaneous CCN spectrometer. *J. Atmos. & Ocean. Tech.*, **6**, 1055-1065.

Hudson, J.G. and X. Da, 1996: Volatility and size of cloud condensation nuclei. *J. Geophys. Res.,* **101**, 4435-4442.

Hussein, K. and C.P.R. Saunders, 1984: Ice nucleus measurements with a continuous flow diameter. *Q. J. Royal Met. Soc.*, **110**, 75-84.

Jennings, S.G., and C.D. O'Dowd, 1990: Volatility of Aerosol at Mace Head, on the West Coast of Ireland. *J. Geo. Res.*, **95(D9)**, 13,937-13,948.

Jennings, S.G., C.D. O'Dowd, W.F. Cooke, P.J. Sheridan, and H. Cachier, 1994: Volatility of elemental carbon. *Geo. Res. Ltrs.*, **21(16)**, 1719-1722.

Meyers, M.B. and J. Hallett, 2001: Micrometer-sized hygroscopic particles in the atmosphere: Aircraft measurement in the Artic. *J. Geo. Res.*, **106 (D24)**, 34,067-34,080.

Rogers, D.C., 1994: Detecting Ice Nuclei with a Continuous-Flow Diffusion Chamber—Some Exploratory Tests of Instrument Response. *J. Atmos. and Ocean. Techn.* **11**, 1042-1047.