

A Comparison of the Langer, Rosen, Nolan-Pollak and SANDS Condensation Nucleus Counters

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ABSTRACT

Laboratory comparisons have been made of aerosol concentrations indicated by four different types of condensation nucleus counters. Three of these counters, the Langer, Rosen, and General Electric SANDS instruments have been used to measure Aitken nuclei concentrations in the upper troposphere and the stratosphere, and the fourth, a Pollak counter, had been carefully calibrated to serve as a standard. Except for the smallest particles employed, quite good agreement was experienced among the Rosen, SANDS and Pollak counters, and the tests served to calibrate the Langer instrument.

1. Introduction

Particles in the stratosphere scatter and absorb solar radiation, thus affecting the earth's climate; possibly they catalyze stratospheric chemical reactions such as the decomposition of ozone. However, our knowledge of the concentrations of stratospheric particles in the Aitken size range ($<0.1 \mu\text{m}$ in radius) is fragmentary. Condensation nucleus counters can be used to estimate the concentrations of Aitken particles in the stratosphere since although they count large particles as well as Aitken particles, the concentrations of the latter greatly exceed the concentrations of the former. Three Aitken particle (condensation nucleus) counters which

are being flown on aircraft or balloons in the stratosphere are the Langer counter, the Rosen counter, and the SANDS counter built at the General Electric Co. and calibrated at the University of Missouri at Rolla. These instruments are based on somewhat different principles. Therefore, a laboratory comparison has been undertaken of the particle concentrations indicated by these instruments and by a calibrated Nolan-Pollak counter when sampling the same aerosols. This paper reports the result of that comparison, held during 19–21 August 1974, at the National Center for Atmospheric Research (NCAR).

2. The nucleus counters

The Langer instrument is a mixing-type counter built at the National Center for Atmospheric Research. A moist sample is mixed with and cooled by cold air. It

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also differs from other counters in that individual droplets are counted acoustically as they settle through the bottom of the instrument. Each droplet produces a click as it passes through a glass capillary when the linear flow velocity in the capillary is between 10 and 100 m s⁻¹ and the droplet exceeds 20 μm in diameter (Langer, 1968–69, 1972). Each click is converted to an electrical signal by a microphone. The relative humidity produced by the Langer instrument is estimated to be 102%.

The Rosen device, built at the University of Wyoming, uses a thermal gradient diffusion cloud chamber and the condensing vapor is ethylene glycol instead of the water vapor used in the other instruments. Under normal operating conditions the relative humidity is about 110%. The droplets produced by condensation are individually counted by a photoelectric particle counter.

The Stratospheric Aitken Nuclei Detection System (SANDS) was developed for the U. S. Department of Transportation, Transportation Systems Center, to measure the concentration and size characteristics of Aitken particles at altitudes to 20 km. Particle detection is accomplished by cyclic adiabatic expansion of sample air, producing a relative humidity of about 330%. Condensation of water droplets on the nuclei in a dark-field optical chamber produces a scattered light signal which is detected by a photomultiplier tube. The system consists of two units, a pressurization unit and an instrument unit, designed to mount in the nose cone of a WB57-F aircraft.

Measurement of the size characteristics of Aitken nuclei is provided for by incorporation of an electrostatic cylindrical condenser (drift tube) preceding the Aitken nuclei counter cloud chamber. A sequentially increasing series of precipitating voltages may be applied on command, and size data subsequently derived from the fractional loss of particles.

The Nolan-Pollak (1946) counter was of the 1957 type (Metnieks and Pollak, 1959) using an unglazed ceramic lining and a slightly convergent light beam. The aerosol laden air is brought into the moist-wall chamber, and pressurized with filtered air. The sample rests within the chamber for 60 s, becoming saturated and attaining temperature equilibrium with the ceramic wall. The overpressure is suddenly released, producing a relative humidity of 260% and causing fog droplets to grow on the particles. The optical density of this fog is sensed photoelectrically and the number concentration of the particles is determined from the transmission, using the calibration table of Pollak and Metnieks (1960).

This particular Pollak counter had been compared with an electrostatic aerosol detector at the University of Minnesota in December 1973 (Liu *et al.*, 1975) on several monodisperse aerosols generated by the technique of Liu and Pui (1974). This independent comparison showed the 1960 calibration to indicate aerosol

concentrations to within 9–20% of those indicated by the electrostatic technique. As this discrepancy is within sampling error, the 1960 calibration is assumed to be quite accurate, and useful as a reference standard.

The Langer, Rosen and SANDS counters were operated at the supersaturations at which they have been operated in the stratosphere, as indicated above, instead of attempting to compare them for the same supersaturation, since a major purpose of the comparison was to facilitate the interpretation of the result of measurements by these counters in the stratosphere. Similarly, including the effect on the results obtained of using ethylene glycol instead of water as the condensing vapor in the Rosen instrument was an important aspect of the comparison.

3. The experiment

Most of the experiments were conducted with sulfuric acid aerosols or with room air. Sulfuric acid aerosols are especially appropriate, since most of the time most of the particles in the stratosphere consist of impure sulfuric acid (Lazrus *et al.*, 1971; Cadle and Grams, 1975). Also, essentially monodisperse aerosols with particle sizes in the Aitken size range are readily prepared from dilute sulfuric acid. Room air was also used because of its availability, the wide size distribution of the airborne particles, and the widely varying composition of the particles. A few comparisons were made with aerosols prepared from the minute residues from the evaporation of droplets of distilled water. Thus the comparisons were made with three very different types of aerosols.

Sulfuric acid aerosols and aerosols formed from distilled water were prepared with an evaporation-condensation type generator similar to that described by Horvath (1969). First a polydisperse aerosol of a sulfuric acid solution was produced by a glass atomizer (Fig. 1) which removed the larger droplets by forcing the aerosol to pass through small holes over which the solution flowed. The removable lid of the atomizer was provided with a ground-glass interface with the remainder of the generator which was sealed with a film of the sulfuric acid solution. The cap containing the small holes was supported on three glass legs two of which are shown in the figure. The atomizer was essentially cylindrical in cross section. It was filled to a depth of 2–3 cm with the solution to be dispersed, and filtered air was introduced through the tube shown at the left of the figure at a pressure of 3.5 psi. The opening in the lid was usually closed, but could be attached to a pressure gage or used to add diluting air. The atomizer was attached by the ground glass joint, at the right in Fig. 1, to the vertical glass U-tube shown in Fig. 2. The left leg of the tube was heated with Nichrome ribbon covered with glass insulating tape and connected to a variable transformer. The aerosol produced by the atomizer was completely evaporated in the left leg of

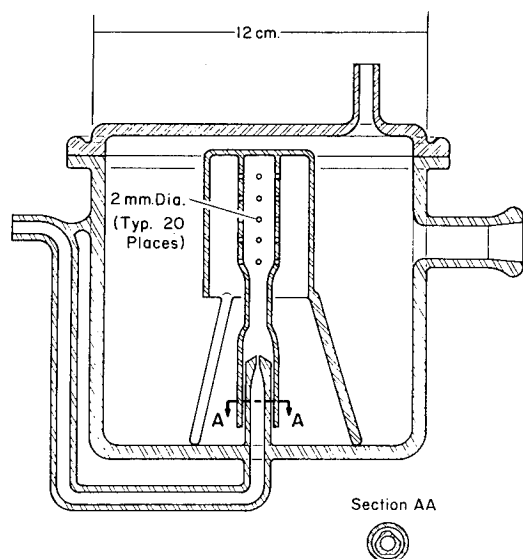


FIG. 1. Atomizer.

the U-tube and recondensed to form an essentially monodisperse aerosol in the right leg. When the liquid atomized was dioctyl phthalate or an 80% by volume solution of sulfuric acid, the reformed aerosol exhibited two Tyndall spectra, corresponding to a diameter of about $0.4 \mu\text{m}$. Electron micrographs of the aerosol droplets collected with an electrostatic precipitator indicated mean diameters of about 0.3 and $0.5 \mu\text{m}$, respectively, uncorrected for any spreading on the collecting surface (Formvar shadowed with carbon).

A 0.8% by volume aqueous solution of sulfuric acid was also dispersed in order to obtain smaller droplets. The mean diameter estimated from the electron micrographs was about $0.04 \mu\text{m}$, again uncorrected for any spreading. When distilled water alone was dispersed, a much wider size distribution was obtained than when dioctyl phthalate or sulfuric acid was dispersed. The diameters were mainly in the range $<0.04 \mu\text{m}$ to about $0.16 \mu\text{m}$.

Numerous comparisons were made using air of the room in which they were conducted. Electron micrographs of particles collected from this air indicated a mean diameter between 0.1 and $0.2 \mu\text{m}$.

Three dilution systems were used, all feeding into a 3 m long by 10 cm diameter Pyrex manifold within which a slight positive pressure was maintained (Fig. 3). The aerosol produced by the evaporation-condensation generator was much too concentrated to be used undiluted for comparison of the nucleus counters and even the two-stage dilution with filtered compressed air was inadequate. Two methods for achieving additional dilution were used. One involved inserting a Gelman Fiberglass Type E filter with or without a minute hole pricked in the center. This system (Fig. 3a) was used to obtain the data of Tables 1 and 2. The second system, which avoided the undesirable use of

the filter, pumped air into the manifold with a stainless steel bellows pump as shown in Fig. 3b. The particle concentration in room air was also too large, so it was pumped into the manifold as shown in Fig. 3c.

Operating the dilution systems so that all four counters could be compared was found to be difficult, and a few sets of data were of questionable significance, especially when obtained when the aerosol concentrations were changing rapidly. Such data have been omitted from this paper but are available from the senior author.

4. Results

The nucleus counters gave the following concentrations (number of particles per cubic centimeter) for the filtered compressed air alone: Langer, 0.2; Pollak, 0; Rosen, 2; SANDS, 5. The results of the comparisons are shown in Tables 1-4. The Langer counter, unlike the others, had never been carefully calibrated although some preliminary tests suggested that the direct calculations from the particle counts should be multiplied by 3-4 to correct for wall losses of the droplets. The ratios of the Pollak to the Langer counts for sulfuric acid droplets (Table 1) and room air (Table 3) provide a rough calibration. The average ratio was 4.2 for sulfuric acid and 3.4 for room air.

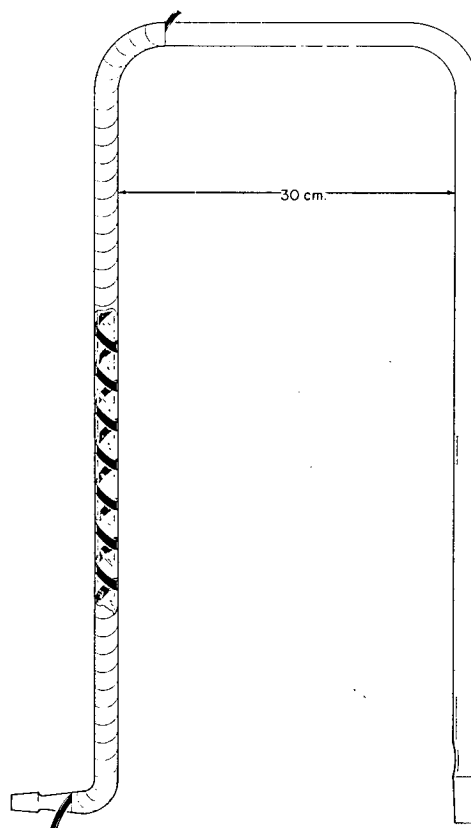


FIG. 2. Evaporator and condenser.

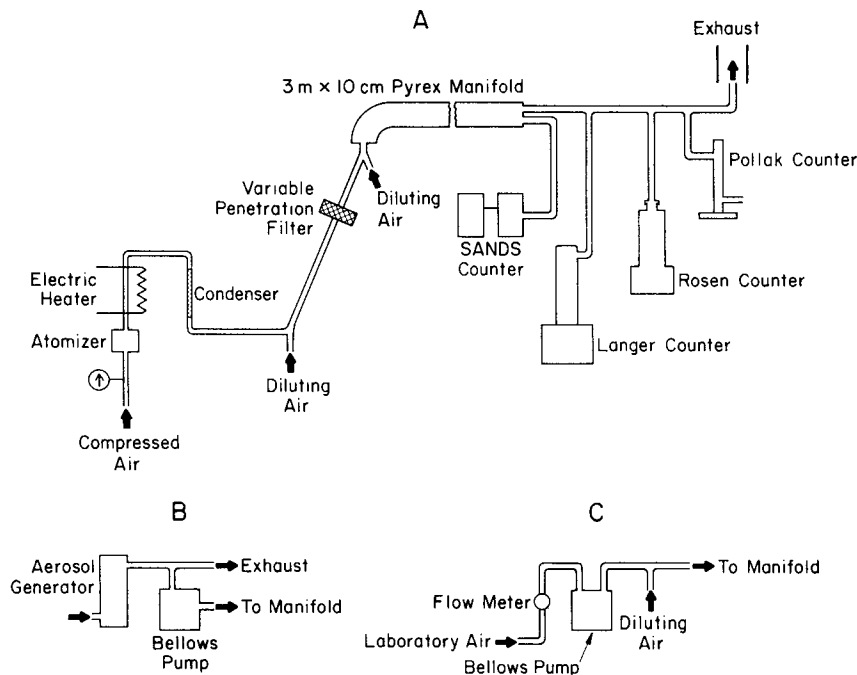


FIG. 3. Systems for aerosol dilution.

The Pollak counter gave generally higher particle concentrations than did the Rosen or SANDS counters for the aerosol prepared from 0.8% sulfuric acid and for six out of ten counts of diluted room air. Arithmetic

TABLE 1. Comparison of nucleus counters with aerosol prepared from 0.8% H₂SO₄. Units: number of particles per cubic centimeter; number mean particle diameter: ~0.04 μm.

Experiment	Counter				B'/A'
	Langer A'	Pollak B'	Rosen C'	SANDS D'	
A	—	4900	—	7000	—
B	—	5300	—	7000	—
C	—	3500	—	3000	—
D	—	5200	—	5000	—
E	275	1150	875	900	4.2
F	400	1400	1000	1000	3.5
G	250	1300	790	950	5.2
H	300	1250	875	925	4.2
I	320	1079	918	850	3.4
J*	10	46	31	—	4.6
K*	9	39	31	—	4.3
L*	10	39	33	—	3.9
M*	15-20	65	57	45	—
N*	15-20	70	63	50	—
Arithmetic mean (E-I)					
mean (E-I)	309	1240	892	925	
σ(E-I)	50	113	68	50	
Relative σ	16%	9.1%	7.6%	5.4%	

* Aerosol passed through a Gelman Type E fiberglass filter, without hole.

means, standard deviations (σ), and relative standard deviations (100 σ/arithmetic mean) were calculated for those concentration values in Tables 1 and 3 obtained when the aerosol concentrations were nearly constant. The Langer counter had the largest relative σ for the aerosol prepared from 0.8% H₂SO₄, but the lowest for room air. There was somewhat closer agreement between the SANDS and Rosen counters than between either of these and the Pollak.

The Pollak counter was not available while the results of Tables 2 and 4 were obtained. The relative counts for the aerosol prepared from 80% sulfuric acid with the evaporation-condensation generator (Table 4) were similar to those of Tables 1 and 3.

The Langer counts for an aerosol prepared with the evaporation-condensation generator from distilled water (which presumably contained impurities which became the aerosol particles) were only about one-tenth the Rosen and SANDS counts, as shown in Table 2.

TABLE 2. Comparison of nucleus counters with an aerosol prepared from distilled water with evaporation-condensation generator. Units: number of particles per cubic centimeter; particle diameters: <0.04 to ~0.16 μm.

Langer	Rosen	SANDS
75	740	900
95	800	825
80	880	775
100	920	825
100	900	875

TABLE 3. Comparison of nucleus counters with diluted room air. Units: number of particles per cubic centimeter; number mean particle diameter: 0.1–0.2 μm .

Experiment	Counter				B'/A'
	Langer A'	Pollak B'	Rosen C'	SANDS D'	
A	25	88	70	86	3.5
B	25	97	86	75	3.8
C	25	97	84	78	3.8
D	30	100	88	80	3.3
E	25	91	88	78	3.6
M	120	560	490	500	4.7
N	150	490	490	500	3.3
O	85	195	280	220	2.3
P	80	190	250	220	2.4
Q	60	190	250	220	3.2
R	40	—	140	130	—
S	45	—	140	120	—
T	40	—	135	120	—
Arithmetic mean (A–E)					
	26	95	83	79	
σ (A–E)	2.7	4.4	6.8	3.7	
Relative σ	1.0%	4.6%	8.2%	4.7%	

5. Discussion

Statistical analysis of five members of a population of values cannot be considered to have much significance, and the relative standard deviations should not be taken very seriously. However, these values do provide a rough idea of the scatter to be expected when using such instruments.

Although the ratios of the Pollak to the Langer counts were somewhat greater for the sulfuric acid aerosol than for room air, the scatter was sufficiently great that this difference has little significance. The average ratio for the combined data of Tables 1 and 3 is 3.7, which can be used as a calibration for the Langer counter. Multiplying the Langer counts by this ratio brings them much closer to those obtained with the Rosen and the SANDS instruments. The correction factor 3.7 probably largely represents the deposition of condensed droplets on the inner wall of the instrument before they can be counted, as mentioned above.

TABLE 4. Comparison of nucleus counters with an aerosol prepared from 80% sulfuric acid. Units: number of particles per cubic centimeter; number mean particle diameter: $\sim 0.5 \mu\text{m}$.

Langer	Rosen	SANDS
17	80	50
24	75	40
170	410	400
110	340	300
100	295	250
80	240	230

Once this correction factor has been applied to the Langer counts, the differences among the values obtained with the various counters are rather difficult to explain because of the large differences among the counters themselves. Probably the difference between the Langer counts (after correction) on the one hand, and the Rosen and SANDS on the other for the aerosol prepared from distilled water was in part at least the result of the low supersaturation used in the Langer instrument. This aerosol probably had a higher relative concentration of very small particles than any of the other aerosols included in the comparison. True, the Rosen counter used ethylene glycol instead of water, and judging from the Kelvin equation alone, for a given relative humidity water should be nucleated by smaller particles than is ethylene glycol. But the Kelvin equation is not directly applicable to heterogeneous nucleation since its derivation does not consider the contact angle. And the SANDS, like the Langer counter, employs water vapor.

The fact that somewhat higher values were obtained with the Pollak than with the SANDS counter was somewhat surprising since the latter had been calibrated with another standardized Pollak counter. This result prompted a comparison of Pollak counters at the Atmospheric Science Research Center (ASRC) in Albany on 6 September 1974 (Haberl, personal communication). Three counters were used in the tests: the ASRC counter included in the NCAR series, a GE 1946 model Pollak counter, and a GE 1957 model Pollak counter which had been built and calibrated at the University of Dublin. The 1946 model had been automated with electrically operated solenoid valves. It had been calibrated periodically with the 1957 model, and was used to initially calibrate the SANDS counter. The three counters yielded very similar results, leaving unexplained the differences observed at NCAR. Haberl suggested the probable cause was losses in the pressurization portion of the SANDS counter.

6. Conclusions

Three recently developed condensation nucleus counters were compared with a Pollak counter, on several types of aerosol. The SANDS and Rosen instruments generally indicated concentrations within $\pm 20\%$ of the Pollak counter, except when extremely small particles were generated. This is good agreement among field instruments, and facilitates the comparison of atmospheric aerosol concentrations measured by these several techniques.

Calibration was established for the NCAR instrument during these comparisons.

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