Comments on Comparisons of Condensation Nucleus Counters

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ABSTRACT

The recent article by Cadle *et al.* (1975) describes a comparison of different types of condensation nucleus counters. Some general remarks are made here on the terminology and calibration of the counters and on the interpretation of the results. A comparison of one of the calibrated instruments (SANDS) with the UMR Absolute Aitken Nuclei Counter (UMR-AANC) in Rolla just after the described workshop showed steadily lower counts by SANDS (about 30% lower). This enables one to compare indirectly the UMR-AANC with the other counters.

1. Introduction

The purpose of this note is to comment on some general problems related to the comparison of instruments counting small aerosol particles and to make some specific remarks on the workshop held in Boulder in August 1974. These remarks should complement the report on the workshop rather than criticize its valuable and difficult task.

2. General remarks on counting small particulates

In their introduction Cadle *et al.* (1975) reveal that they had some difficulty finding a common name for all the different types of counters compared during the workshop. The authors conclude that "three Aitken particle (condensation nucleus) counters (ANC) 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 Company." One might wonder if it is really necessary to find a common name for all these instruments operating at different supersaturations and based on different principles.

Taking a conservative attitude, we can say that an Aitken nuclei (AN) counter should count all airborne particles possessing critical supersaturations slightly smaller than small ions. The latter are defined by their mobility $k \gtrsim 1.0$ cm^2 V^{-1} s^{-1}, corresponding roughly to $r \approx 6.6 \times 10^{-8}$ cm. However, an AN counter should count a substantial portion of small intermediate ions (with mobilities $1.0 > k \gtrsim 0.01$ cm^2 V^{-1} s^{-1}) and all larger ions and uncharged particulates. Under these conditions the AN counter should operate at a supersaturation $>200\%$ relative to the equilibrium vapor pressure over a plane water surface and not larger than 350% [270% according to White and Kassner (1971)]. This requirement seems to be of importance mainly in the case of a mixture of nuclei activated simultaneously in the counter. For instance, all aerosol particles generated by the burning of a silver iodide pyrotechnic material mixture were not completely activated even at a supersaturation of 230% (Wegrzyn and Podzimek, 1975). We also usually assumed that the percentage of large and giant nuclei is negligible compared to the concentration of particulates $<r=0.1$ μm (AN). For this reason, the Nolan-Pollak counter was frequently used to calibrate the AN counters operated at an expansion ratio of 1.21. At this value, it is assumed that the physical-chemical nature of AN plays a relatively insignificant role in initiating the condensation of vapor on individual aerosol particles due to the extreme metastability of the gas-vapor system. Under these circumstances, the ANC becomes essentially a total particle counter. By operating the ANC at lower supersaturations, only a fraction of the total AN is observed.

Based on the foregoing definitions of AN and ANC (which have been widely accepted and used for decades in atmospheric physics), the Nolan-Pollak and SANDS counters are clearly AN counters while the other instruments tested during the Boulder workshop are not. Since the other counters operate at supersaturations substantially below 350%, the physical-chemical nature of the nuclei affects the probability of condensation and causes the counter to discriminate between particles of different characteristics yielding discrepancies between the counts of the various instruments. Such discrepancies were most readily apparent in the case of Langer's counter. The question arises whether such instrumental discrepancies can be accommodated by employing a certain correction factor which can be assumed to be valid for all nuclei size distributions, concentrations and physical-chemical characteristics.

Applying a simple theoretical model, which calculates the deviation in supersaturation from the theoretical maximum (supersaturation decrement) due to heat conduction from the walls (i.e., Kassner *et al.*, 1968),

one readily concludes that unless counters of similar geometry, operating at the same supersaturation, and nuclei of similar overall size distribution are employed in the comparison of the counters, it is not reasonable to expect to obtain an applicable correction factor. In other words, a universally applicable correction factor does not exist because the nature of the variable in question possesses an unreliable nonlinearity. This rather pessimistic statement can be made more palatable if one limits the use of a given correction factor to aerosols of some relatively narrow range of size distributions and concentrations. One should bear in mind that all of the existing theoretical models of fast droplet growth, applicable to conditions prevailing in expansion chambers used as ANC, describe only to a very limited extent the real physical process. This makes it difficult to extend calibration factors over wide ranges of aerosol concentration.

Looking at the comparison of the counters in Tables 1 —4 in Cadle *et al.*, one can agree with several conclusions made by the authors, particularly those pertaining to Langer's counter. However, it seems inappropriate to make any claims about the universality of the correction factors derived from the data obtained in these comparisons mainly for the reason that many of the described experiments were performed using aerosol sizes greater than that corresponding to the AN and where the size distribution probably did not extend smoothly down to the smallest sizes appropriate to AN. Under such circumstances an instrument which is not a total particle counter can give results which are similar to those of a true total particle counter. However, a calibration factor so obtained constitutes little more than a comparison of the efficiencies of the optical detection systems and says nothing about the counter's ability to detect the total number of fine particles when a continuous distribution prevails.

The calibration of AN counters suffers from another deficiency which is common to all the existing counters and which might explain some inconsistencies in measurements. This is the problem of residual nuclei, denoted and treated in the classical German monograph on meteorological instruments by Kleinschmidt (1935) as "Nachzüglers." These residual nuclei remained in the sensitive volume of the expansion chamber of the counter. Such nuclei either were not activated during the first expansion due to their physical-chemical nature as a result of insufficient expansion (supersaturation) or they may result from the reevaporation of droplets due to the rapid thermal recovery of the chamber. Several studies made on this subject, mainly with the Nolan-Pollak and Scholz counters (i.e., Pollak and Metnieks, 1959; Podzimek *et al.*, 1961), showed that the residual nuclei (not activated during the first expansion) might amount to several tens of percent of the nuclei counted at the first expansion. In manually operated counters, activated nuclei were still observed even after five successive expansions. This phenomenon, probably related to the "memory effect" of nuclei, was studied in detail by Smith *et al.* (1968) in the UMR-AANC. However, there are no available measurements of residual nuclei with the instruments compared in Boulder. Therefore, one might expect that due to this effect all counted AN concentrations will be lower than those which are actually present in the counter.

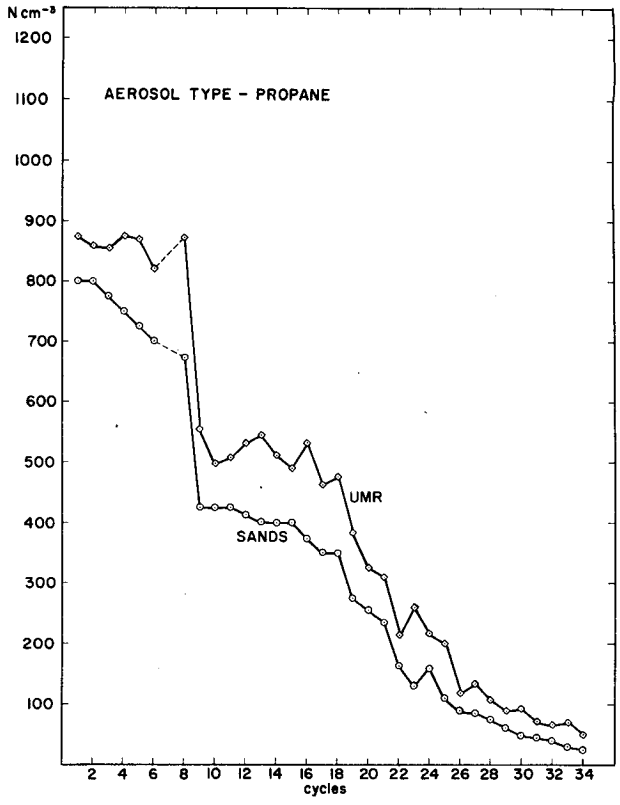

FIG. 1. Comparison of the UMR-AANC with the SANDS using residual nuclei after the evaporation of the propane droplets. One cycle means one data point taken by the UMR-AANC.

3. Comparison of the SANDS with the UMR-AANC

Because the SANDS was calibrated with the UMR-AANC just after the 1974 workshop (Cadle *et al.*, 1975), it might be useful to compare both measurements and in this way obtain information on the comparability of the instruments tested in Boulder with an absolute AN counter[1] with a sensitive volume of almost 15 liters. The detailed description of the laboratory arrangement of the comparison of both counters has been published elsewhere (Wegrzyn and Podzimek, 1975). The SANDS was compared at this time without the pressurization unit which itself caused particle losses of ~12% for aerosol particles in the upper size range of AN (estimated during a

[1] The term "absolute counter" is used here in accordance with long-established usage (e.g., Pollak, 1959).

separate experiment.) Fig. 1 shows the result of a comparison for residual AN after the evaporation of propane droplets and Fig. 2 for NaCl particles (in the upper range of AN and above it). NaCl aerosol was prepared by vaporization of NaCl crystals in a tungsten boat and by passing the aerosol through a series of low-efficiency filters before storage. Several other runs were made with the room aerosol, with residual nuclei of propane drops, and with NaCl and AgI aerosol. They showed a very similar trend in the plotted data, as in Figs. 1 and 2.

During the laboratory comparison of both instruments in Rolla in September and October, 1974, the SANDS measured consistently lower AN concentrations than UMR-AANC. The mean difference amounted to more than 30% and was higher for larger and hygroscopic particles (over 40%) and for smaller nuclei concentrations (below 300 nuclei cm^{-3}). As a result of these measurements, all the AN counts made with SANDS, mainly in the stratosphere in the first half of 1974, were corrected. After the calibration in the fall of 1974 in Rolla, the SANDS was readjusted so that the new calibration at the beginning of 1975 showed good correspondence between SANDS and UMR-AANC (Fig. 3). All observations made with the SANDS subsequent to November 1974 were taken with the adjusted SANDS in order to match with the UMR-AANC except for the slight correction for the pressurization unit (Podzimek *et al.*, 1975a,b).

These facts will influence the intercomparison of the counters made in Boulder. If the AN counts by UMR-AANC are correct, the data published by Cadle *et al.* should be lower by at least 30%.

4. Conclusion

The authors of these comments support the former division of nuclei counters into AN counters, cloud condensation nuclei counters, etc. They recognize well the inconsistency of this division mainly related to cloud condensation nuclei counters where the nuclei activity should have preference over nuclei size. In all cases, it would be very worthwhile for the users of the counters to know about the most important parameters of the counters, such as supersaturation related to the water surface and its duration.

In the authors opinion, the nuclei concentrations published in the Cadle *et al.* article are in mean too low. Moreover, there is little chance that nuclei of different physical-chemical nature and size distribution can be covered by a universal correction factor by which the data measured by Langer's counter could be multiplied in order to obtain the correct count.

The calibration of SANDS in Rolla indicates that the Nolan-Pollak counter measures lower AN concentrations than UMR-AANC (if we increase the data on the curve corresponding to the SANDS by 30% in the Cadle *et al.* diagrams). That was discussed

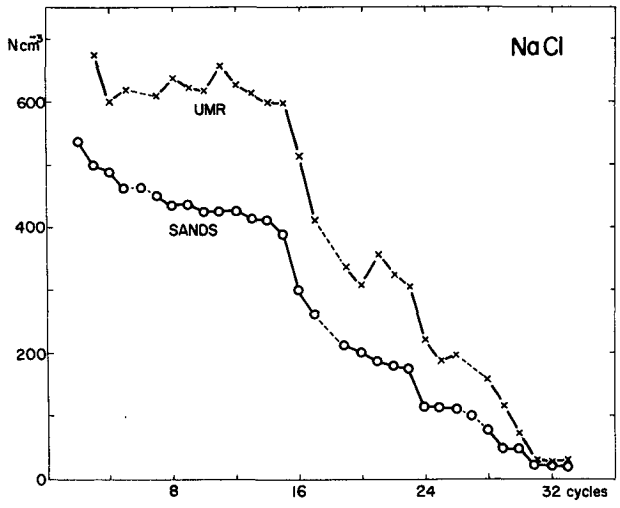

FIG. 2. Comparison of the NaCl aerosol concentration measured by the UMR-AANC and by the SANDS.

several times in the literature (i.e., Kassner *et al.*, 1968; Saxena *et al.*, 1972). However, there are no direct systematic measurements on this subject except some comparisons made in Rolla (Mansell, 1969) and during the Fort Collins International Workshop on Condensation and Ice Nuclei in 1970 (Grant, 1971; Saxena *et al.*, 1972) which are, unfortunately, not very conclusive.

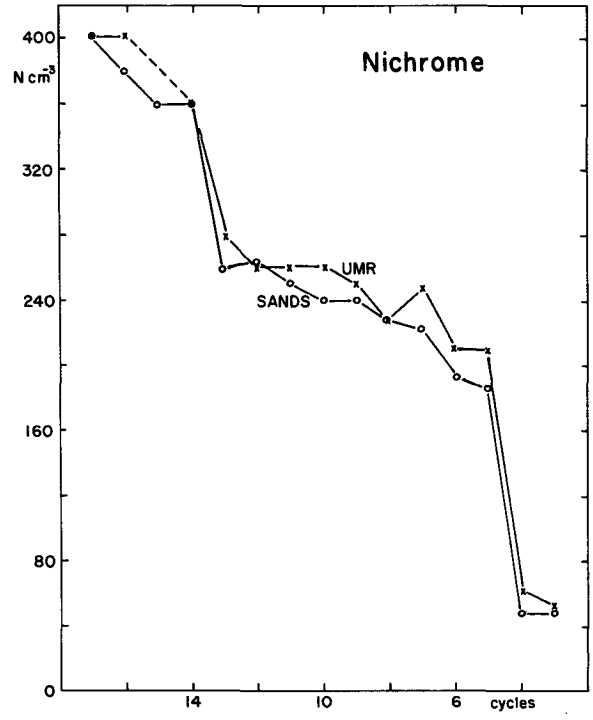

FIG. 3. Comparison of the UMR-AANC with the SANDS using nichrome aerosol.

For this reason, a direct comparison of the Nolan-Pollak counter used by several research centers and the AN counters manufacturered in the United States with the UMR-AANC would be very worthwhile. This comparison is further supported by the fact that the Nolan-Pollak counter was originally calibrated by an especially adapted Aitken counter (Pollak and Metnieks, 1959). In other words, the calibration of the Nolan-Pollak counter with the UMR-AANC reduces the problem to the comparison of two absolute counters, one with a large volume (UMR-AANC with approximately 15 liters volume) and the other with a small volume (Aitken type counter of 0.22 liters volume) if we disregard the different way of counting the drops. In our opinion, there can be no substitution of the suggested calibration by a comparison of the Nolan-Pollak counter with a differential mobility analyzer using AN of a size greater than 0.025 μm (Liu *et al.*, 1975).

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