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Calibration of a photoelectric cloud condensation nucleus counter

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Abstract

Calibrations were peformed for the photodetector output of a static thermal-gradient diffusion chamber by comparing the detector signals with visual counts of the number of droplets developing in the chamber. The calibrations cover the supersaturation range 0.3% to 1%. For each supersaturation, the CCN concentration was found to be proportional to the detector output. During the activation and growth of droplets, the time of peak signal corresponds well to the time at which droplet count is a maximum within the sample volume. The use of an average output signal over a period bracketing the mean position of the peak is a slightly better measure of droplet concentrations than the peak value. The calibration equations here derived are specific to the CCN counter used to obtain the data. Also, the calibration constants can be expected to be dependent on the type of aerosol which serve as CCN; the limited data so for available show only weak variations in the constants.

1. Introduction

Cloud droplet concentrations are of fundamental importance in determining the radiative properties of clouds and in the evolution of precipitation in them. Measurements of cloud condensation nuclei (CCN) have a parallel importance, as there is a reasonably clear, though not always simple link between these nuclei and droplet concentrations. CCN measurements thus provide considerable predictive capability about cloud characteristics. Major research efforts to understand the sources and roles of CCN are being organized within the framework of the International Global Atmospheric Chemistry (IGAC) program, and the International Global Aerosol Program (IGAP), among others. A recent review of cloud condensation nuclei observations is that of Hudson (1993). Many aspects of CCN

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measurements are covered in the summary of the Third International Cloud Condensation Nuclei Workshop (July–December 1981 issue of the *Journal de Recherches Atmosphériques*, pp. 177–370).

CCN measurements can be obtained by direct activation of the nuclei in a chamber of controlled supersaturation, or CCN activity can be deduced from measurements of particle size and composition. Direct CCN measurements can be obtained with a variety of instruments, the static thermal-gradient diffusion chamber (SDC) being perhaps the most widely used since its introduction by Twomey (1963). Continuous-flow devices and those providing measurements at several supersaturations simultaneously have many advantages over the SDC's, but tend to be more complex (Hudson and Squires, 1976). The SDC devices are useful for measurements in the supersaturation range 0.2% to 1%. The supersaturated zone forms in the middle of the space between two wet parallel plates which are at different temperatures. Aerosol particles exposed to the supersaturation become cloud droplets, grow, and fall out of the zone of supersaturation. The volume concentration of the droplets so generated is the required measurement; concentration as a function of supersaturation is the CCN spectrum.

Accurate determination of the concentration of droplets forming in the middle of the thermal-gradient diffusion chamber is not a trivial problem, and this determination is the limiting factor in the accuracy of these devices. Two approaches are used: direct counting of droplet images, or the measurement of light scattered by the droplets. Direct counting is done on a photographic or video image, the sample volume being defined by the illuminating beam and the depth-of-field of the imaging system. This method has the advantages of being independent of the sizes of the droplets that develop, or of the intensity of the illumination (beyond that required for creating a usable image). On the other hand, the counting is laborious if done simply; alternatively it requires sophisticated image analysis. Therefore, the indirect approach of measuring the light scattered by droplets has considerable attraction, and has been used in a number of different forms (Lala and Jiusto, 1977; Bartlett and Ayers, 1981). These methods lead to more convenient automation of the measurement, at the cost of introducing the additional uncertainty associated with the relationship of scattered light signal to droplet count in the chamber. That relationship is determined by empirical calibration.

The signal from a photodetector which collects some of the light scattered by the droplets in the chamber is time-dependent, in response to the activation, growth and fall-out of droplets. In direct counting, the assumption is made that the highest count during the cycle of droplet activation and growth is the best estimator of the true concentration of CCN (some limitations of this procedure are pointed out by Hoppel and Wojciechowski, 1976, 1981). Accordingly, the peak signal from the photodetector was usually assumed to provide the best measure of the droplet concentration. Thus, the calibrations of Lala and Jiusto (1977), Lala (1981), and Bartlett and Ayers (1981) relied on comparisons of the peak signal with the count of droplets at the time of the peak signal. A common problem in these calibrations is that the scattered signal is proportional to the integral of droplet number times scattering cross-section, not just droplet number. Since droplet sizes also vary with time, the interpretation of peak signal as maximum droplet concentration is subject to question.

In this paper we examine the reliability of using the scattered light for determination of droplet concentration, and suggest an improvement in the use of the method. Our evaluations

are based on the comparison of the direct, visual count of droplets with the photodetector output. The results do not constitute an absolute calibration of the instrument, but focus on increasing the reliability of the photodetector output for determination of CCN concentrations.

2. Description of the equipment

The calibrations here described were developed with the use of the University of Wyoming cloud condensation nucleus counter (UW-CCNC, 1991). This instrument was built for use in pressurized aircraft. Its main component is a static thermal-gradient diffusion chamber. Controlled supersaturations are created in the chamber by cooling the bottom plate, while allowing the top plate to be at ambient temperature. Both plates carry water-soaked paper. The interior of the chamber is visible through three windows. One is used for illumination by a 5 mW He–Ne laser tube. One window allows visual inspection, or viewing by a video camera; for video recording the window has lenses added which provide a four times magnification. A photodetector and lens arrangement is placed at the third window. The arrangement of these components is shown in Fig. 1. The basic design of the chamber follows that of Lala (1981). The CCNC is controlled by a Motorola MC68010 microprocessor which provides digital temperature control, data manipulation, read-out and manual device control via a flat panel graphics touch screen (UW-CCNC, 1991). Video data were recorded with a standard video tape recorder. Plate temperatures and the output of the

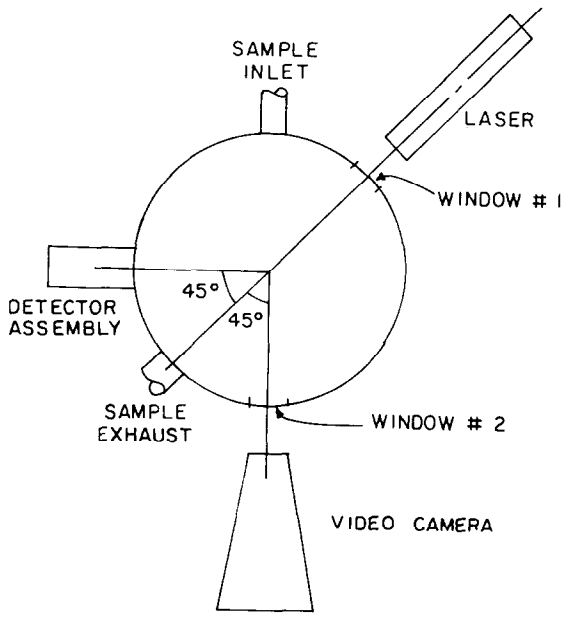


Fig. 1. Schematic diagram of the main components of the detection system (plan view).

photodetector were recorded at 10 Hz by a separate computer-based data system. The status of the control valves were also recorded. The output of the photodetector is adjusted to compensate for changes in laser output, and the baseline for the output is checked before each cycle of measurements.

The sample volume seen in the video images was determined by placing a semi-transparent millimeter scale in the chamber. The diameter of the laser beam could be directly read off the scale. The length of beam over which drops were to be counted was arbitrarily restricted to a region in which droplets appeared well focussed. The width of this region was marked on the CRT display on which the counting was performed; the width of this region was determined from the image of the millimeter scale. This procedure yielded 0.060 ± 0.005 cm^3 for the video sample volume. The volume seen by the photodetector was determined by moving a thin wire along the laser beam and noting the distance where a signal appeared on the photodetector; the sensitive volume was found to be 0.073 cm^3.

The photodetector used is a Devar 509-50, with built-in amplifier. This sensor has a sensitivity of 10^{-12} W, and a responsitivity of 2.0×10^7 V/W at the He–Ne laser radiation of 632.8 nm wavelength. After 200-fold amplification, and using the calibration presented below, the counter output is 25 mV/droplet at 0.5% supersaturation. The scattered light power corresponding to this is 6.25×10^{-12} W/droplet (4.25×10^{-9} lumen/droplet).

3. Data collection and results

The primary calibration data were collected over a two-week period in November 1991, in Laramie, Wyoming, a small town of 25,000 inhabitants located in a broad valley of 2300 m altitude. Air was drawn through a large metal duct from above the building where the instrument was housed. Samples to the CCN counter were taken from the duct, with a steady airflow maintained at the intake of the instrument. A small pump inside the counter flushed the instrument by at least ten times the chamber volume, before the sample was scaled in the chamber for processing. Sequences of measurements were made in the following order: 3 samples at 0.3% supersaturation, 2 at 0.5% and one each at 0.8% and 1%. One such sequence was obtained approximately every 4 minutes. The data set used for the calibration amounted to approximately five hours of sampling.

3.1. Time delays to maxima

The first step of data processing was to determine the maximum count of droplets and the time of that maximum from the video records. The video tapes were played back on a recorder which could hold individual still images; the tape was scanned back and forth until the maximum was located. A count was then obtained within the boundaries indicated on the screen for definition of the sample volume. The clock time of the frame with maximum count was read off the time encoded onto the video record. The lapse of time from the instant of closing the chamber to the time of maximum count, t_v, was obtained from the difference of clock time on the video and the recorded time of valve closure at the chamber air outlet. Similarly, the lapse of time from closing the chamber to time of the maximum detector voltage, t_p, was obtained for each measurement from the digital records.

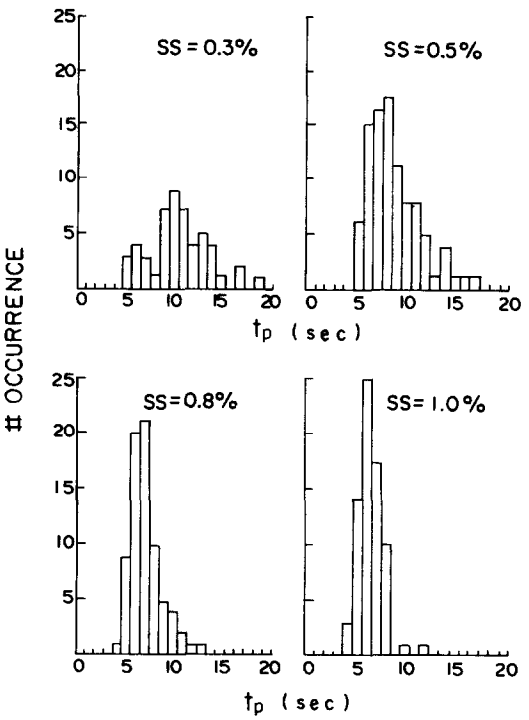


Fig. 2. Frequency distributions of the lapse of time from closing the chamber to peak output from the photodetector.

Frequency distributions of t_p values are shown in Fig. 2 for each supersaturation. The dominant trends are for shorter times and less spread with higher supersaturations. One contribution to the large spread of times at low supersaturations, is that the detected signals are generally smaller and hence more subject to noise superimposed on the signals to produce a spurious peak reading. To guard against this possibility, the data were screened rejecting runs with $t_p < 5$ s and with $|t_p - t_v| \geq 3$ s. These restrictions removed 40%, 20%, 10% and 10% of the data points for supersaturations of 0.3%, 0.5%, 0.8% and 1.0%, respectively.

Mean values of the lapse times $\bar{t}_v$ and $\bar{t}_p$ and of their standard deviations σ_v and σ_p are given in Table 1. Also indicated are the number of data points included in each average, and values for Student's t-test for difference in the averages obtained in two different ways.

Table 1
Summary of time delays to maxima

Supersaturation	Video records			Photodetector output			*t*-test
%	$\bar{t}_v$ (s)	σ_v (s)	N_p	$\bar{t}_p$ (s)	σ_p (s)	N_p	t
0.3	10.2	3.2	59	10.5	3.0	39	0.46
0.5	8.1	2.2	77	7.8	2.4	69	078
0.8	6.8	1.2	65	6.3	1.4	62	1.71
1.0	6.2	1.1	65	5.9	1.2	58	1.44

The null-hypothesis tested using the Student's t-test is: "The two populations (lapse times, t_v and t_p) are equals". The result of the t-test is true, at 95% confidence, if $t < 2.576$ for samples sizes greater than 30. According with Table 1 it is seen that the two methods in fact yield equivalent results. This is reassuring from the point of view of using the photodetector output for concentration determinations, and appears to indicate that the growth of the droplets, and the corresponding increase in total scattering cross-section is fortuitously compensated for by changes in the angular distribution of the scattered light, and by the reduction of droplets in the sample volume by sedimentation.

3.2. *Normalized output curves*

In order to generate a representative depiction of the time variation of the detector output, independent of the concentrations being measured, the signals were normalized to the peak value of the particular sample and an average of all samples for given supersaturations obtained. The resulting curves were then readjusted in magnitude to the mean magnitudes of the peak voltages to produce Fig. 3. The horizontal bars in the figure correspond to values of $\bar{t}_p \pm \sigma_p$. This figure gives a good indication of the reduction in t_p and in the narrowing of the curves with increasing supersaturations.

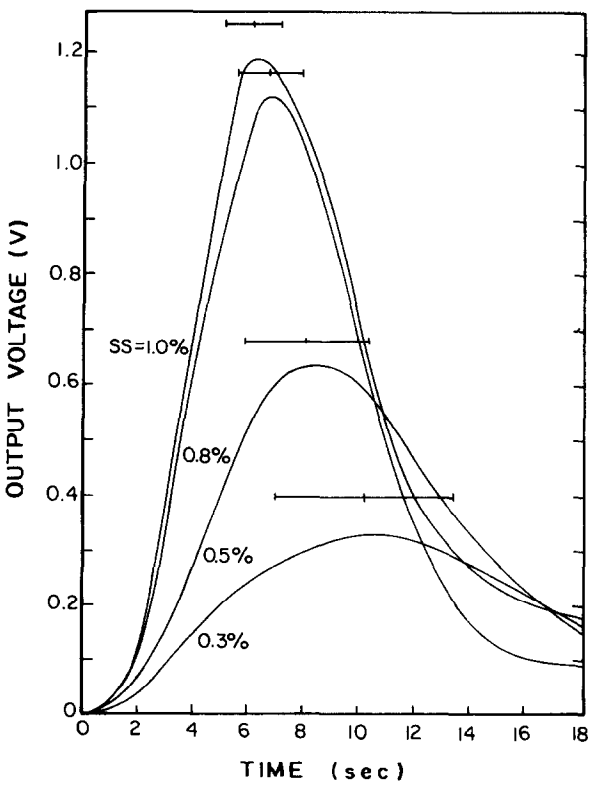


Fig. 3. Variation of photodetector output with time.

Table 2
Constants for the calibration equations

Data set	α	β	γ	δ
Laramie, WY – November 1991	0.20	0.11	0.17	0.19
Broomfield, CO – February 1993	0.17	0.14	0.25	0.09

3.3. Calibration equations

Two different measures of the photodetector output were used for expressing the observed CCN concentrations derived from the visual counts: the peak voltage V_m and an average V_a to be defined below. The peak voltage was taken to be the maximum recorded value in the 10 Hz samples of photodetector output, with the restriction that this peak occur within the time interval $\bar{t}_p \pm \sigma_p$ for the given supersaturation. The average voltage was defined in order to provide some immunity from the noisiness of the output. Rapid variations in the output arise from the "flickering" of scattered light as droplets move in and out of the laser beam,

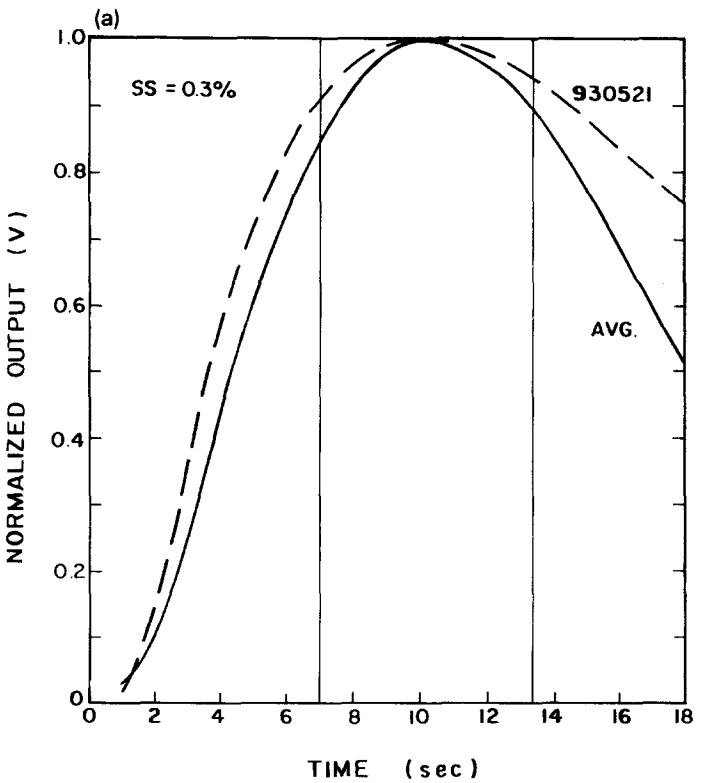
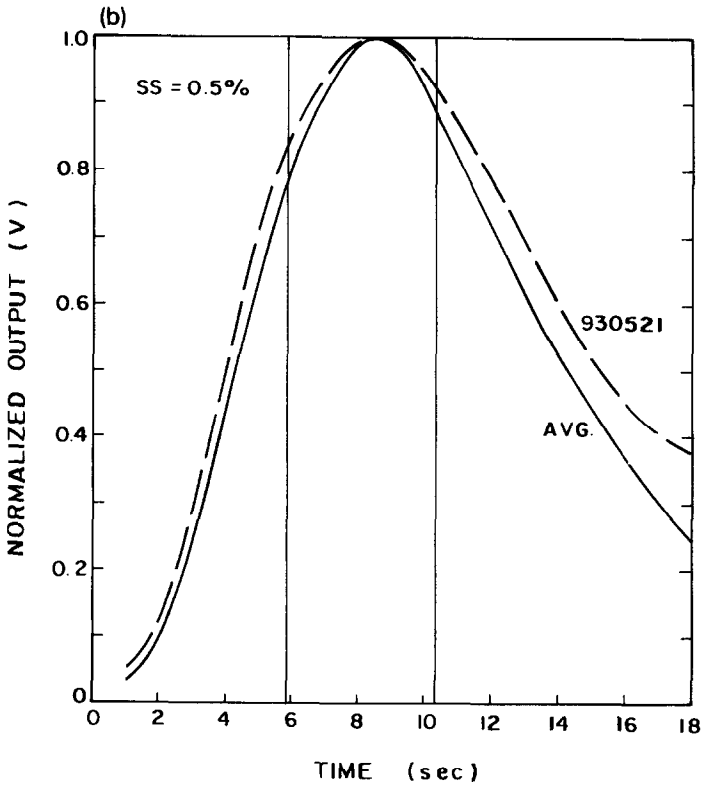


Fig. 4. See caption on p. 246.



and from electronic circuit noise. The effects of these fluctuations can be greatly reduced by using an averaged output over the time interval $(\bar{t}_p - \sigma_p)$ to $(\bar{t}_p + \sigma_p)$.

Denoting the concentration derived from counting (equal to count divided by sample volume) by C_v, the calibration equations are expressed in the form

$$C_v = V_m\left(\sigma + \frac{\beta}{SS}\right) \tag{1}$$

and

$$C_v = V_a\left(\gamma + \frac{\delta}{SS}\right) \tag{2}$$

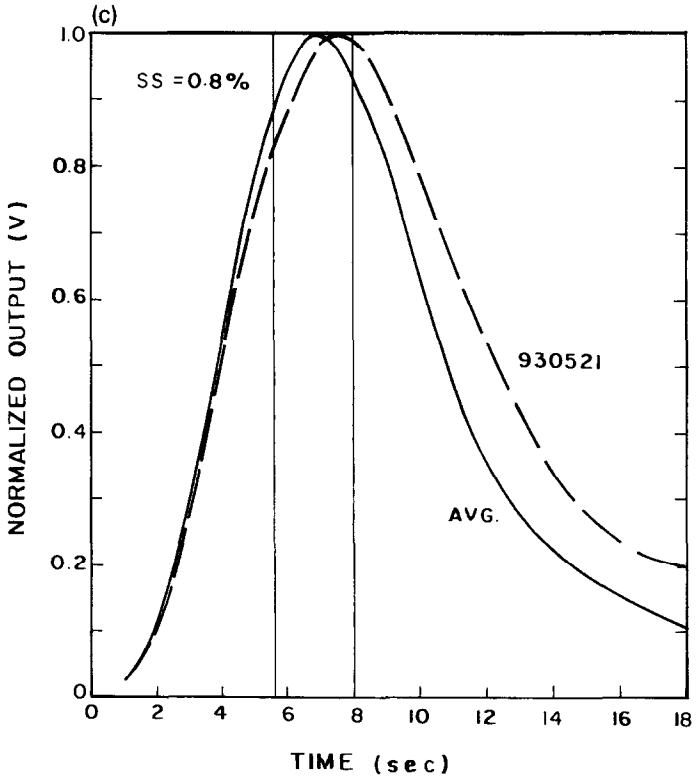
were C_v is in (cm^3), the supersaturation SS is in $(\%)$, and V_m and V_a are in millivolts. The constants of these equations were determined by first obtaining a linear best-fit relationship between C_v and V_m or V_a, restricting the fit to pass through the origin, and then obtaining a linear relationship between the constant of that proportionality and the inverse of the supersaturation SS.

The linear relationship here given between voltage output and concentration is in accord with the finding of Lala (1981). A numerical comparison of the two sets of calibrations is not possible because of different gains in the two instruments (200 for the Wyoming device, unstated in the Lala instrument).

For the data described above, the constants were found to have the values shown in the first line of Table 2.

3.4. *Range of applicability*

The theory of condensation on aerosol particles indicates that the size and rate of growth of the droplets depend on the dry size and chemical composition, or surface properties, of the nuclei. The magnitude of this dependence is less clearly established, primarily because of the difficulties of full characterization of surfaces. The importance of these factors diminishes as the droplets grow. Empirical evidence for these effects is scant, but there are clear indications that they are not negligible. For example, Hoppel and Wojciechowski (1981) report that the time-dependence of droplet counts in a CCN chamber can have quite large variations, and that those variations are contrary to expectations for different aerosol



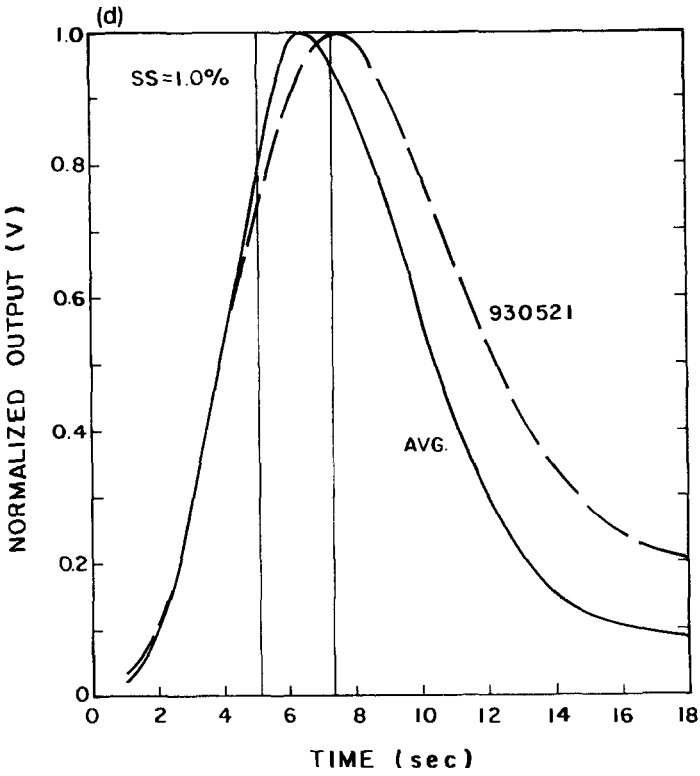


Fig. 4. Comparison of the detector output on May 21, 1993 with the average of many runs.

types. The times for maximum concentrations to develop varied from 3–8 s in their observation at 0.7% supersaturation. Longer times were associated with monodisperse aerosol of well-defined composition than with mixed aerosol of atmospheric origin. Clearly, such variations put limits on the validity of the calibrations here described.

To examine the range of applicability of the calibrations described in the foregoing, additional data were collected from different locations. Calibration data were obtained from one flight over the Florida peninsula on August 4, 1991, from one flight near the island of Santa Maria, Azores on June 26, 1992, and from samples taken inside a well-ventilated hangar at Broomfield, Colorado in February 1993. Only the Broomfield data were extensive enough to calculate new values for the calibration constants. These values are shown in Table 2. In the other data sets, comparisons were made, by comparing concentrations derived from visual counts for the field data, with the value predicted by the calibration equation with the constants for the Laramie data. The difference between the two sets of constants in Table 2 has the greatest impact at the lowest supersaturation, where the difference amounts to 32%, whereas at 1% supersaturation the difference is only 6%. The comparisons with the Florida and Santa Maria data — where larger discrepancies might have been expected

due to rather different, maritime aerosols — revealed maximum differences of only 12% and 9%.

These comparisons indicate, that even beyond the inherent problems of the SDC type of instrument (transient supersaturation, internal air motions, etc.), and the problems associated with translating droplet counts in the supersaturated region with CCN concentrations, further errors can arise from inadequate calibration of the photodetector output versus the visual count. There is some possibility that the light-scattering signal is in fact superior to the visual counting method; reasons for this might be in the larger sample volume possible and the elimination of subjectivity in counting unevenly illuminated droplets. However, the dependence of the light scattering on droplet size introduces a factor in addition to the basic measurement requirement, which is the number concentration. Our observations do not indicate that the light scattering signal should be preferred to the visual count. As a practical matter, the results do indicate that the calibration relationship should be established for each aerosol type. Until some generalizations are developed in that regard, frequent calibrations are desirable. The use of the constants here reported to other situations may be associated with errors of 30% or even larger.

The variability in the shape of the voltage output versus time has not been systematically examined so far. The general impression is that variations from run to run with a given sample are comparable to the changes seen in different data sets. One exception was noted: the activation appeared to proceed slower than usual on one day, May 21, 1993. A comparison of the pulse shapes for this day with the averages shown in Fig. 2 is given in Fig. 4. The pulses are notably wider for the lower supersaturations, and the peak is shifted slightly for the higher supersaturations. Nonetheless, the effects of these changes on the calibration equation were minor; values of V_a differed by 4–7% from the standard curves, after normalization to eliminate differences in CCN concentrations.

4. Summary

The experiments here reported are in essential agreement with previous work about the use of the light-scattering signal from droplets in a static diffusion chamber for obtaining a measure of the number of droplets developing in the chamber. The method yields reliable results, but the calibration relationship between scattered light and droplet count has to be determined with great care. Using an averaged signal intensity over a period of time bracketing the peak signal provides better results than using the peak value. The sensitivity of the calibration to aerosol type needs further examination.

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