Modification of a Commercial Integrating Nephelometer for Outdoor Measurements

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ABSTRACT

One of the major challenges in atmospheric aerosol research is the quantification of aerosol optical properties at ambient relative humidities (RHs) and their attribution to natural and anthropogenic aerosol sources. As an essential tool for this task, a commercial single wavelength integrating nephelometer built for dry indoor aerosol measurements was modified and complemented for outdoor operating conditions. The temperature difference $\Delta T$ between air inlet and outlet is the crucial parameter indicating the effect of the instrument on RH of the sampled aerosol. By removing all heat sources from the optics compartment and by doubling the length of the light pipe between lamp and sensing chamber, $\Delta T$ became smaller in the laboratory than the uncertainty of the temperature sensors. After mounting the instrument in a protective housing an average $\Delta T = 0.55^\circ C$ ($0.22^\circ C$ standard deviation) was maintained under nonoptimized ventilation conditions for one week of unattended outdoor operation parallel with an unmodified unit indoors. Data evaluation of the field test provided promising results concerning the temporal variability of the humidity dependence of aerosol optical properties.

1. Introduction

For the measurement of volumetric scattering coefficients, the integrating nephelometer with closed sensing volume is one of the backbones of climate- and health-related atmospheric aerosol research (Anderson et al. 1996; Heintzenberg and Charlson 1996). This prominent role largely depends on the design feature of its closed optical chamber, which ensures both high sensitivity [down to $10^{-8} \text{ m}^{-1}$ at green wavelengths, that is, 0.1% of Rayleigh scattering of particle-free air (Heintzenberg and Bücklin 1983)] and ease and stability of calibration (Bhardwaja et al. 1973; Bodhaine 1979). Unfortunately, these positive design features have negative side effects that limit the atmospheric applications of this instrument. Among these, the effect on relative humidity (RH) often is the most severe.

The size of atmospheric aerosol particles and thus their optical properties are strongly dependent on relative humidities of the carrier gas (Junge 1950). The RH of the aerosol is strongly reduced by the heating of the sample air during its passage through the nephelometer. The most important heat source in the sensing volume is the lamp, which illuminates the aerosol and much more so the optically black inner surfaces of the chamber. Signal and control electronics attached to the sensing chamber also contribute to aerosol heating, leading to a combined warming of about 5$^\circ C$ between aerosol inlet and outlet.

In studies aimed at physical and chemical aerosol properties, it may be desirable to dry the sample aerosol in order to segregate the effect of condensational growth on the nephelometer signal from that of particle size distribution and chemical composition. For that kind of application, it would be desirable to replace the uncontrolled aerosol heating due to above-named heat sources by a well-defined and controlled heating upstream of a sensing chamber that is unaffected by additional (uncontrolled) instrumental heating.

In the past, several attempts have been made to avoid or overcome the RH sensitivity of the nephelometer signal. The original design (Beuttell and Brewer 1949) employed an open chamber to avoid this problem. This approach was taken up by the open chamber design of Ruppersberg (1964) for a visibility meter, the commercial version of which had a detection limit of $10^{-4} \text{ m}^{-1}$ for the sum of molecular scattering and particle scattering coefficients. The design also induced significant systematic measuring errors because of the truncated range of scattering angles over which the optical integration was made (Heintzenberg and Quenzel 1973; Quenzel 1969).

Closed chamber nephelometers can be calibrated easily and their background signals can be characterized by filling the instruments with gases of known optical
properties, temperature, and pressure. This elegant procedure is not easily accomplished with instruments that cannot be hermetically sealed. To overcome this problem, calibration disks with known scattering properties have been inserted into the light path of open instruments (Ruppersberg 1964). For such calibrations, it was necessary in some devices to cover the sensing chamber with a black cloth (Garland and Rae 1970). Another design employs a sensing chamber that can be closed automatically for calibrations (Malm et al. 1996). The U.S. National Park Service has been using a commercialized version of this instrument for a number of years at some 25 sites as part of the Interagency Monitoring of Protected Visual Environments (IMPROVE) program.

The aerosol transition from haze to fog, with its inherent visibility change (Eldridge 1969; Heintzenberg et al. 1998), and the strongly humidity-dependent direct climate forcing of atmospheric aerosols (Charlson et al. 1999; Nemesure et al. 1995) are two important issues in atmospheric aerosol research. Understanding these issues requires accurate measurements of aerosol optical properties, such as the scattering coefficient, which must be as close as possible to ambient conditions as part of so-called closure experiments (Quinn et al. 1996). Ultimately, empirical models will be developed from such closure experiments, which allow the prediction of ambient optical aerosol properties from size distribution, chemical composition, and the state of the mixture of the atmospheric aerosol. Development and testing in different atmospheric settings of these particle growth models require the existence of aerosol-optical reference data that are taken as close as possible to ambient conditions. The present report describes the development of an aerosol scattering sensor, which is meant to fill this function besides other devices, such as aerosol path extinction measurements (Plentje et al. 1997).

The present approach of modifying a nephelometer maintains all advantages of the closed chamber design, including a high-sensitivity, self-calibrating commercial realization while minimizing the RH problem due to self-heating of the instrument. After a description of the modifications and subsequent tests of the nephelometer (Model 3551, TSI Inc., St. Paul MN), a discussion of a week-long atmospheric time series is given in which the modified nephelometer is compared to an unmodified unit of the same manufacturer at varying levels of ambient RH.

2. Modifications of the instrument

The objective of the modifications of the TSI Model 3551 integrating nephelometer was to minimize the self-heating of the instrument. The base instrument for the present study has been described in detail by Anderson (1996). Here, we focus on the parts of the unit that were modified.

The 75-W quartz halogen lamp is the dominating heat source of the instrument. A parabolic dichroic mirror focuses most of its radiant output through a protective glass, which is an integral part of the lamp, on to a 57 mm × 12.25 mm diameter quartz light pipe leading to the opal glass through which the diffuse illumination of the sensing chamber is accomplished. While some of the heat output is kept from the chamber through this light guide, much of the heat transmitted to the metal environment of the lamp is conducted to the sensing chamber through good heat contact between the aluminum tube of the optical chamber and the lamp housing. Additional heat sources arise from the fact that all signal and control electronics except for the main power supply are located within the casing of the optical unit. The cooling fins of a switching power supply within this electronic unit have direct heat contact with the optical chamber. For long-term operation outdoors, additional radiative heating of the body of the whole instrument has to be taken into account.

Without modification of the instrument, most of the heating of the sample air can be avoided by minimizing its residence time in the instrument, that is, by maximizing the volume flow through the sensor. The manufacturer recommends volume flows larger than 10 L min⁻¹. The temperature difference between inlet and outlet is the crucial parameter to describe the RH effect of the nephelometer on the sample aerosol. We measured the reduction of temperature difference for flows between 10 and 100 L min⁻¹ and calculated an average rate of heat carried away by the air flow. With that value, a sample flow of 290 L min⁻¹ would be required to bring the remaining temperature difference to zero. Such high flows through the sensor are out of the question because of the high impaction losses they would cause, particularly for large haze particles at high RH.

Consequently, other ways had to be sought to minimize sample heating. Our basic approach in doing this was to implement reversible modifications only, that is, at any point, the original instrument could be reinstalled. As a first and major step, the heat input from the lamp to the sensing chamber was reduced by the following measures. By means of a 131-mm-long quartz pipe optically cemented onto the original light guide, the distance between the lamp and the optical chamber was more than doubled. When extending the lamp housing accordingly, its heat contact with the chamber was reduced by attaching it through Teflon washers. A 50 × 50 × 3 mm³ bandpass filter (KG4, Schott Glaswerke AG, Mainz, Germany) was placed between the lamp and the extension of the light guide. At central wavelength 550 nm of the present nephelometer, this filter has a transmission of 89% (also down to 380 nm). Even at the longest wavelength (750 nm) of the three-wavelength model of the instrument, the filter transmission is still 60% (less than 10% transmission at 1100 nm). Because of the expected high signals, the ensuing reduction in sensitivity is negligible for haze and fog studies with a modified nephelometer.
The cooling unit of the lamp was modified by replacing the original cooling fan with a stronger model (Comair Rotron Galaxy 24VDC, RS Components Corby, United Kingdom) and covering all four sides of the lamp housing with cooling fins that were vented by the fan. The aluminum casing of the extended light guide was cooled by an additional fan (Slim-line fan 83CFM 24VDC, RS Components Corby, United Kingdom), providing an airstream through the extended lamp housing perpendicular to the direction of the light guide.

The power consumption in the signal and control electronics leads to an additional heat source near the sensing chamber, which was eliminated by moving all electronics except the photo detector unit and the environmental sensors to a separate box that was connected electrically to the optical unit.

For outdoor deployment, the optical unit was mounted vertically in a protective reflective aluminum housing with vents for convective cooling in the bottom and around the top (cf. Fig. 1). The separate electronics unit was mounted in a water-tight aluminum carrying case.

All subsequent tests of the modified instrument relied on the built-in temperature sensors in inlet and outlet of the optical chamber. For these sensors, the manufacturer TSI gives a tolerance of ±0.3°C. After the modifications of the nephelometer, temperature differences between inlet and outlet were held in the laboratory within the combined uncertainty of the two temperature sensors (±0.5°C). At relative humidities around 90%, this temperature uncertainty translates into a considerable uncertainty in RH of about ±3%. Because of the common exponential condensational growth of atmospheric particles at these humidities, even larger uncertainties in the related scattering coefficients of up to 20% can be expected, according to published growth curves. These uncertainties should be kept in mind in the interpretation of the high humidity results of the atmospheric test discussed below. Clearly, further developments of the modified sensor should include better temperature sensors. Individually calibrated accuracies ±0.1°C are achievable, which would reduce the uncertainty in humidity changes through the instrument to values below 1%.

Even a very good knowledge and close similarity of relative humidities at inlet and outlet of the instrument do not remove all uncertainties in the interpretation of the scattering data. Temperature gradients within the sensing volume would still be possible and are likely, at least on the timescale for thermal equilibration of the sensing chamber to ambient thermodynamic changes.

3. Atmospheric deployment

In April/May 1999, the modified prototype instrument was tested on a roof platform of the institute, where it was mounted in its protective housing on a 1-m-high
FIG. 2. Time series of meteorological parameters measured in the nephelometer inlets and at an automatic weather station, both on roof platforms of the Institute for Tropospheric Research, Leipzig. Zero hours corresponds to 28 April 1999, 0000 local time. The time series end on 4 May 0527 local time. RHw = RH in the outdoor nephelometer, RHd = RH in the indoor nephelometer, RHmet and Tmet are RH and dry bulb temperatures measured at a weather station (40 m horizontal distance, 2 m above outdoor nephelometer), respectively.

scaffolding as close to the outer edge of the platform as possible. Through a 15-m-long, 3-cm inner diameter hose, sample air was drawn from a point right beside the inlet of the modified nephelometer to a second nephelometer (Model TSI 3556) that was operated indoors. After setting the flows to 30 L min⁻¹, the units were operated without further adjustments for one week, with 1-min time resolution of the output and 1-h intervals between automatic air calibrations. The stability of the automatic air calibration in the modified unit was checked with the data on the first and last complete days of the experiment. The corresponding measured daily averages of Rayleigh scattering coefficients in the 0.55-μm channel degraded from 1.145 × 10⁻⁵ m⁻¹ (±7.56 × 10⁻⁸ m⁻¹) to 1.12 × 10⁻⁵ m⁻¹ (±2.16 × 10⁻⁸ m⁻¹). The standard deviations in parentheses correspond to 0.66% in the beginning versus 1.9% at the end of the experiment. No flow adjustments and no changes in ventilation of the modified nephelometer were made to minimize the temperature difference between inlet and outlet.

For the data evaluations, time series of meteorological variables were available, which were recorded routinely on another roof platform of the building. The horizontal distance to the outdoor nephelometer was 40 m. Their elevation was about 2 m higher than the nephelometer input. The humidity sensor of the routine weather data was a conventional hair hygrometer with a strong nonlinear response above 90% RH. From the manufacturers data, the combined uncertainty of the two independent humidity sensors [outdoor nephelometer (±5%) and weather station (±2%)] is estimated to be ΔRH ≈ 6 percent units. Within this uncertainty, the two RH sensors track each other during most of the test period. Toward the end of the test period, though, the deviations became larger during the sunlit part of the day because of some radiative heating of the housing of the outdoor nephelometer. In the cooler early parts of the test period, limited heating capacity in the indoor laboratory led to rather high RH in the indoor nephelometer.

In Figs. 2 and 3, the measured parameters are displayed as time series beginning on 28 April at 0000 LT and ending on 4 May at 0527 LT. The time period had been chosen because of relatively stable weather conditions with strong diurnal variations in RH covering a range between 25% and values slightly over 90%. Nevertheless, due to changing pollution levels, particle scattering coefficients at 550-nm wavelength varied between 1.4 × 10⁻⁵ and 3.6 × 10⁻⁴ m⁻¹ in the indoor (dry) nephelometer.

Figure 4 shows the frequency distribution of the temperature differences between outlet and inlet of the out-

FIG. 3. As in Fig. 2 but for the ratios of relative humidities (RHw/RHd) and particle scattering coefficients at 550-nm wavelength (WN/DN) in outdoor and indoor nephelometer.

FIG. 4. Frequency distribution of the temperature differences between outlet and inlet of the modified nephelometer during the field test. The data have a digitizing uncertainty of 0.1°C.
Fig. 5. Scatterplot of the ratio of scattering coefficients (outdoor/indoor, corrected for sampling losses) vs RH in the outdoor nephelometer (RHw). Only data with RHw values > 50% are plotted.

Door nephelometer. The 4557 data points of the series yield an arithmetic mean ΔT of 0.55°C with a standard deviation of 0.22°C. Because of the nonoptimized sample flow and ventilation of the instrument, the temperature difference is positively biased. Thus, with an optimized design, we can expect considerably smaller temperature differences.

The long inlet hose upstream of the indoor nephelometer led to particle losses, which can be quantified in terms of resulting change in scattering coefficient. No significant RH dependence of the ratio of the readings of the two nephelometers was observed below an outdoor RH of 50%. Consequently, the particle loss was calculated from the average ratio of the scattering coefficients of the two sensors below 50% RH, yielding a value (±1 standard deviation) of 1.12 (±0.06). This factor was used to correct the reading of the indoor nephelometer in the scatterplot of Fig. 5. After this correction, scattering ratios of outdoor to indoor, up to 1.5 at relative humidities above 90%, remain. The measured scattering ratios around 90% RH are considerably lower than the scattering ratios from comparisons of dry and humidified nephelometer data given in the literature for urban aerosols over the United States, where values between 2 and 2.5 have been reported frequently (Charlson et al. 1978, 1974; Covert et al. 1972, 1980). However, they are similar to the only humidified nephelometer data available for urban locations in Germany (Winkler et al. 1981), which for January 1973, yield 50%–90% RH growth factors for scattering coefficients at 0.55-μm wavelength of 1.6.

For the geographical region of the test site, the annual average relative humidity is 60% (Peixoto and Oort 1996). For this RH level, and at times up to 80% RH, no consistent increase in light scattering with RH beyond the level measured indoors was recorded during the test period (cf. Fig. 5). There are, however, two branches in the scatter plot of Fig. 5, which indicate relatively strong condensational growth a) around 70% and b) above 80% RH. The two branches do not stem from the same time period. With the exception of a few outliers (7 one-minute data points during hour 52), branch a was measured during the last night of the experiment (hour 145). Branch b was recorded during the first two nights of the test. As the highest ratios of outdoor to indoor nephelometer were recorded during the period with the highest RH values in the indoor nephelometer (branch b), we exclude a varying dry baseline RH level in the indoor nephelometer as an explanation for the observed differences in light scattering. Instead, we hypothesize different hygroscopic growth of the aerosol during the respective time periods.

4. Conclusions

One of the major remaining challenges in atmospheric aerosol research is the quantification of aerosol optical properties at ambient relative humidities (RHs). As one step in this direction, a commercial single wavelength integrating nephelometer built for dry indoor aerosol measurements was modified and complemented for outdoor operating conditions. The temperature difference between air inlet and outlet is the crucial parameter, indicating the effect of the instrument on RH of the sampled aerosol. All heat sources were removed from the compartment of the nephelometer optics, and the light pipe between the lamp and the sensing chamber was doubled in length and cooled. With these relatively
small and reversible modifications, the temperature difference was reduced in the laboratory from 4° to 50°C in the unmodified instrument to less than the uncertainty of the temperature sensors. After mounting the instrument in a protective housing, one week of unattended and nonoptimized outdoor operation parallel with an unmodified unit indoors proved the validity of the modification. A stable temperature difference of +0.5°C was maintained between outlet and inlet under conditions from 25% to 95% RH over a temperature range of 5°-20°C. Data evaluation of the field test provided promising results concerning the temporal variability of the humidity dependence of aerosol optical properties.

For the usage of the sensor in atmospheric closure studies (Quinn et al. 1996), further refinements of the modifications will be necessary, primarily more accurate temperature measurements in the sensing chamber. For that purpose, the humidity response of the sensing volume to ambient thermodynamic changes also needs to be established, and the humidity distribution inside the chamber will need to be characterized.

The instrumental development reported above primarily addressed the correct measurement of the optical effects of condensed water on atmospheric aerosols. The reported sensor improvements are equally important for assessments of the optical effects of other volatile aerosol species such as nitrate or organic components, which also may exhibit losses from the condensed phase in heated nephelometers.

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REFERENCES


