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Ambient particles at an urban, semi-urban and rural site in Central Europe: hygroscopic properties

G.A. Ferron*, E. Karg, B. Busch, J. Heyder

Institute for Inhalation Biology, GSF-National Research Center for Environment and Health, P.O. Box 1129, D-85758 Neuherberg/Munich, Germany

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Abstract

Measurements on hygroscopic properties of sub-micrometer atmospheric particles have been carried out using a tandem differential mobility analyser (TDMA). It measures the growth of initially dry monodisperse particles with sizes of 50, 100, 150 and 250 nm at a relative humidity (RH) of 0.85. Measurements have been carried out at an urban, semi-urban and rural location in Bavaria (Germany) each during 1 week in winter, summer and fall. Two fractions of particles are found, hygroscopic and non-hygroscopic particles with mean growth factors of 1.37 and 1.03, respectively. Growth factors are nearly independent of location and season, but increased significantly with initial particle size from 1.31 to 1.47 for 50 and 250 nm particles, respectively. The mean number concentration of non-hygroscopic particles is strongly dependent on location and increases by a factor of 2.4 for the semi-urban site and by a factor of 9 for the urban site, compared to the rural location. The corresponding differences for hygroscopic particles are less than a factor of three, respectively. Individual measurements strongly differ in number concentration from each other. Assuming that the hygroscopic material is ammonium sulphate, the non-hygroscopic particles and the hygroscopic particles contain about 3% and 54% ammonium sulphate, respectively. For the hygroscopic particles the mean increase of ammonium sulphate was 38–74% for 50 and 250 nm particles, respectively.

Keywords: Atmospheric particles; Hygroscopic particles; TDMA; Growth factor; Hygroscopic mass fraction

1. Introduction

Hygroscopicity of ambient particles play an important role in earths climate (Charlson and Wigler, 1994; Hougthon et al., 1996). On one hand sulphate particles promote cloud formation and reduce the amount of radiation of the sun that reaches the earth's surface. On the other hand soot particles absorb sun radiation and warm up the atmosphere. Both processes are about equal. The hygroscopicity also strongly influences the chemical reactivity of the particles and their deposition behaviour.

Sulphate and soot differ in their hygroscopic properties. So these properties enable a differentiation of both types of particles. The growth of ambient particles in situ can be determined with a tandem differential mobility analyser (TDMA: Lui et al., 1978; Rader and McMurry, 1986b; Winklmayr et al., 1991). Experiments with this instrument have shown the existence of both not-growing or non-hygroscopic (less-hygroscopic) and growing or hygroscopic (more-hygroscopic) particles, with a high variability in time (McMurry and

*Corresponding author. Tel.: +49 89 3187 2848; fax: +49 89 3187 2400.
E-mail address: george.ferron@gsf.de (G.A. Ferron).

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Stolzenburg, 1989, Los Angeles aerosol, Claremont; Svenningson et al., 1992, Po Valley; Busch et al., 1994, campus GSF and Hohen-Peissenberg; Pitchford and McMurry, 1994, Grand Canyon; Svenningson et al., 1994, Kleiner Feldberg; Svenningson et al., 1997; Great Dun Fell; Swietlicki et al., 1999, Great Dun Fell; Swietlicki et al., 2000, subtropical north-eastern Atlantic). None of these studies concern aerosols in cities.

Aim of this study was to measure hygroscopic properties of the atmospheric aerosol simultaneously at an urban, semi-urban and rural location in South Germany during a week in winter, summer and fall. Data on size distribution were measured simultaneously, which will be published separately. Results of both studies can be used in climate research and risk analysis.

2. Methods

2.1. Strategy

A TDMA and a particle size spectrometer (Karg et al., 2004) were installed ready for operation in a van. The van visited an urban, semi-urban and a rural site for 1 week during winter, summer and autumn in 1998.

2.2. Sampling sites

Measurements have been carried out at

- an urban site located in Munich at a crossroads on the main circle highway inside the city. It is one of the locations in Germany with the highest traffic density. It is located 5 km west of downtown Munich at an elevation of 500 m above North Sea level,
- a semi-urban site (GSF campus) located 1 km north of the city border of Munich, distance to two highways is larger than 200 m). It is surrounded by meadows and forest. It is located 14 km north of the urban site at an elevation of 490 m,
- a rural site (Meteorological Observatory Hohen-Peißenberg of the German Weather Service) located about 200 m above the surrounding small villages, meadows and forest at an elevation of 980 m, 52 km southwest of the urban site and 20 km north of the nearest alpine peak. It has a weather observatory and its meteorology is well documented. It therefore may serve as a reference.

Distances of Munich to the Adriatic Sea, Mediterranean and North Sea are 290, 460 and 670 km, respectively. Air pollution in Munich is not heavily determined by emissions of large industries. No heavy industry is near to the three sampling sites.

2.3. Instrumentation

The TDMA has been extensively described in literature (Lui et al., 1978; Rader and McMurry, 1986a). The main detectors of the TDMA are two differential mobility analysers (DMA) (Model EMS 1000, Haucke, Gmunden, A) (Winklmayr et al., 1991) and a condensation particle counter (CPC) (Model 3010, TSI, St. Paul, MA, USA). Before entering the TDMA the ambient aerosol is dried in a diffusion drier and particles larger than 1 μm are removed by a cascade impactor (EPI 3.0/1000, Haucke, Gmunden, Austria). The remaining particles are charged to equilibrium with a radioactive source (Am-241, 5 mCi, Amersham-Buchler, Brunswich, Germany).

The first DMA selected a nearly monodisperse fraction from the dried ambient aerosols with initial particle diameters of 50, 100, 150 or 250 nm. This fraction and the sheath air at the second DMA were stabilized at a relative humidity (RH) of 85%. This second DMA was used to determine the size distributions of the humidified monodisperse particles. The ratio of the aerosol flow and sheath flow in both DMAs was 0.1 resulting in a resolution of 0.1 (Reischl, 1991; Flagan, 1999). Data were stored and processed with EMS 1000 software (Haucke, Gmunden, Austria) and our own script software. The software cycles continuously measuring ambient particles of 50 and 100 nm, calibration with NaCl particles, measuring ambient particles of 150 and 250 nm, and again calibrating with NaCl particles. The growth factor for NaCl particles, whose growth is known for different RHs (Robinson and Stokes, 1965), was used to correct the growth factor of the hygroscopic particles for variations of the experimental relative humidity (for equations see next section).

The calculation method is described in the Appendix. For each growth spectrum the particle number density was derived from the CPC count rate including the flow rates through the CPC and DMA2 and a number distribution was determined. This distribution was separated in one cumulative distribution for non-hygroscopic and one for hygroscopic particles using a cut point of 1.15 of the initial particle size. Each cumulative distribution was characterized by a count median diameter (CMD) and a number concentration $\mathrm{d}N/\mathrm{d}\log(D_p)$, where N is particle number and D_p is particle diameter. The growth factor was defined as the ratio of the CMD and the original particle diameter selected by DMA1. The CMD of the hygroscopic particles was corrected for the RH of the measurement based on the calibration with NaCl. Mean cumulative distributions for non-hygroscopic and hygroscopic particles were determined for each site, season and initial particle size.

The different parameters of the atmospheric aerosol particles, time of the measurement, day of the year, are