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In situ measurements of particle number concentration, chemically resolved size distributions and black carbon content of traffic-related emissions on German motorways, rural roads and in city traffic

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Abstract

In-situ measurements of various properties of traffic-related aerosol particles have been performed with a mobile laboratory. The measured aerosol quantities include particle chemical composition (sulfate, nitrate, total organic matter, ammonium, black carbon) as well as particle size distributions covering diameters from 10 to 300 nm, and total particle number density. Additionally, gas phase emissions (CO_2, NO, NO_2) were monitored. We performed six measurement drives in the vicinity of the city of Aachen (population ca. 260,000) at 50.8°N, 6.1°E in Germany on motorways, rural roads, and in the inner city of Aachen in June 2005.

The results indicate that the main influence of traffic on the aerosol properties results in soot particles coated with organic matter, having a modal diameter around 100 nm (“soot mode”). The abundance of these particles was found to be highest in the inner city traffic. Nucleation mode particles (around 30 nm) have been observed occasionally during truck chasings on motorways. These particles consisted mainly of organic compounds but included possibly also a sulfuric acid core. Data observed under “motorway background” conditions were similar to the rural regional road data. Highest number concentrations have been observed during truck chasings. Comparison between non-refractory (here with respect to 600 °C) and total particle volume indicated a higher contribution of refractory material under all traffic-influenced conditions compared to rural road data. Fuel specific emission ratios were derived for a subset of 18 truck-chasing experiments, yielding $(8.3 \pm 5.8) \times 10^{15}\,kg^{-1}$ for particle number, $224 \pm 136\,mg\,kg^{-1}$ for black carbon, $125 \pm 125\,mg\,kg^{-1}$ for organic matter, $17 \pm 12\,g\,kg^{-1}$ for NO and $18 \pm 14\,g\,kg^{-1}$ for NO_x (mean values and standard deviations).

Keywords: Diesel particles; Traffic emissions; Urban pollution; Aerosol mass spectrometry

1. Introduction

Particle emissions from traffic play an important role in urban particle pollution. The adverse health effects of coarse and fine particles (with diameters

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$>2.5\,\mu m$ and $<2.5\,\mu m$, respectively) have been studied in detail (Brunekreef and Forsberg, 2005; Pope and Dockery, 2006; Schlesinger et al., 2006). Furthermore, there is evidence that especially ultrafine particles ($d<100$ nm) have adverse health effects (Kreyling et al., 2006; Oberdörster, 2001). In contrast, little to no data are available on the health effects of nucleation particles from diesel exhaust, which are formed by gas-to-particle conversion of sulfuric acid and water and may contain a significant amount of semi-volatile organic species due to uptake during particle growth. Although sulfuric acid aerosol has been shown to have a toxicologically significant effect, it is likely that in human airways sulfuric acid is rapidly neutralized to ammonium sulfate, which has turned out to have no significant health effect in rat lungs (Cassee et al., 2002).

Still it is uncertain what amount of the ambient particle mass concentration (PM10, PM2.5) or particle number concentration can be attributed to traffic emissions. However, since particle emission control strategies are under discussion both in the U.S. and in the E.U., this question is to be answered by current scientific research. Current European legislation prescribes that the threshold value for PM10 (particulate matter with aerodynamic diameter below $10\,\mu m$) of $50\,\mu g\,m^{-3}$ may only be exceeded on 35 days per year.

During recent years, many studies of engine particle emissions have been performed on engine test sites (Sakurai et al., 2003; Shi and Harrison, 1999; Tobias et al., 2001) or chassis dynamometers (Kleeman et al., 2000; Schneider et al., 2005). In order to obtain information about the emissions of traffic exhaust gases and particles into the atmosphere under realistic conditions, these laboratory measurements have to be complemented by field measurements. Stationary measurements (Allen et al., 2001; Charron and Harrison, 2005; Ketzel et al., 2004; Kirchstetter et al., 1999; Pakkanen et al., 2006; Rose et al., 2006; Shi et al., 1999; Virtanen et al., 2006; Vogt et al., 2003a; Wehner et al., 2002, 2004) are well suited to provide data relevant to a particular measurement site, but are less suited to study the contribution of individual vehicles. Thus, on-road experiments under real cruise condition are crucial experiments that enhance the scientific understanding of traffic emissions.

Recently, several research facilities have used a mobile laboratory and demonstrated the feasibility of mobile measurements of traffic emissions with special emphasis on aerosol particles (Bukowiecki et al., 2002, 2003; Canagaratna et al., 2004; Kittelson et al., 2004b; Pirjola et al., 2004; Vogt et al., 2003b; Weijers et al., 2004; Yli-Tuomi et al., 2005). These studies yielded emission indices for pollutants as CO, NO_x, and particulate matter, but chemically resolved particle measurements with an appropriate time resolution, especially with respect to size-segregated chemical composition with a mobile laboratory, have so far been reported only by Canagaratna et al. (2004). Here we report on a study with the Ford Mobile Laboratory (FML) equipped with a real-time aerosol mass spectrometer (Aerodyne Q-AMS) and with additional aerosol and gas phase measurement systems. The measurements were performed under various traffic scenarios in June 2005 in the vicinity of Aachen, Germany. While the focus of this study was on the emission measurement of in-use trucks under real-world conditions, the data set also includes data representing conditions of motorway background, rural roads and inner city traffic.

2. Experimental

The measurements took place between 7 and 10 June 2005 in the vicinity of Aachen, Germany (population ca. 260,000, location: 50°48′N, 6°06′E), mostly on motorways (“Autobahn” A4, A44, A61), on regional roads in a rural environment, in the inner city of Aachen, and partly in the suburban areas or smaller towns (Fig. 1). The city of Aachen and its surroundings contain a number of emission-intensive industries like glass and ceramic production, asphalt and concrete production, basic precious and non-ferrous metal production, etc., while the region contains surface mining sites for soft coal and larger power plants.

The FML of the Ford Forschungszentrum Aachen (FFA) (Schneider et al., 2005; Vogt et al., 2003a) was instrumented as detailed in Table 1. Specifically for this study, the Aerodyne Quadrupole aerosol mass spectrometer (Q-AMS, see Canagaratna et al., 2007; Jayne et al., 2000; Jimenez et al., 2003b) and the AVL micro soot sensor (Model 483), which is the commercially available version of the photoacoustic soot senor (PASS) (Beck et al., 2003; Petzold and Niessner, 1996), were integrated into the FML. The micro soot sensor detects black carbon with 1 Hz time resolution and a detection limit of $5\,\mu g\,m^{-3}$. The Q-AMS is equipped with an aerodynamic lens that focuses particles with