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SCIENCE DIRECT®
Atmospheric Environment 39 (2005) 6696–6706
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Emissions of fine particles, NO_x, and CO from on-road vehicles in Finland

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Received 9 December 2004; received in revised form 12 May 2005; accepted 26 July 2005

Abstract

Real-time particle number size distributions and NO, NO_2, NO_x, CO, and CO_2 concentrations were measured with a mobile laboratory van in October 2003 on the streets and highways of the Helsinki metropolitan area, Finland. A bimodal particle size distribution was observed with about 85% of the particles being smaller than 29 nm. Real-time fuel-based emission factors for size-resolved particle numbers, CO, and NO_x were determined. Wide distributions of emission factors were obtained for all pollutants. In addition, $PM_{2.5}$ samples were collected and the elemental compositions were analysed. Relative to fixed site urban $PM_{2.5}$, street air $PM_{2.5}$ concentrations of Cu, BC, Fe, and Zn were elevated. Weather and road conditions influenced PM concentrations more than the differences between the city and highway traffic environments.

Keywords: Traffic; Particles; Carbon monoxide; Nitrogen oxides; Emission factors

1. Introduction

Exposure in traffic contributes considerably to the total human exposure to air pollutants (Duci et al., 2003; Adams et al., 2001; Gee and Raper, 1999), and knowledge of this exposure is important for the assessment of health effects of pollutants. In traffic environments, the concentrations of traffic-related pollutants are higher than in other environments and a considerable amount of time, on average from 4% to 8% of total hours of the day, is spent in traffic in developed countries (Eurostat, 2004; Jantunen et al., 1998; Jenkins et al., 1992). In-vehicle exposure to $PM_{2.5}$ has been reported to cause cardiovascular effects in healthy young patrol officers in North Carolina, US (Riediker et al., 2003). Most health endpoints were associated to a $PM_{2.5}$ source factor that reflects speed-changing traffic conditions with a high loading of copper, aldehydes, and sulphur (Riediker et al., 2004).

The most widely used methods for evaluating vehicle tailpipe emissions are dynamometer tests, which involve measurements of emissions from selected vehicles using standardized driving cycles under controlled conditions. A key concern with these tests is that they do not fully represent real-world driving conditions and emissions.

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1352-2310/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2005.07.049

In city traffic, the driving cycle, fleet age, and type distributions, as well as dilution conditions are highly variable compared to the fixed test cycles. Also, the non-tailpipe emissions such as those from the wear of brake linings, tyres and asphalt, and resuspension of road and soil dust are not covered by this test method. Alternative approaches to determine the emissions from motor vehicles are tunnel studies, remote sensing techniques, and model calculations combined with monitoring results. Recently, mobile laboratories have emerged as useful tools for real-world emission measurement (Kittelson et al., 2004; Pirjola et al., 2004a; Vogt et al., 2003; Bukowiecki et al., 2002).

Most of the on-road motor vehicle emission inventories have been travel based ($g\,km^{-1}$). A major disadvantage of this method is that the travel-based emission factors vary much more than the fuel-based emission factors ($g\,(kg\,fuel)^{-1}$) as driving modes change. Fuel-based methods have been used in on-road emissions measurements such as those from remote sensors (Pokharel et al., 2002; Singer and Harley, 1996, 2000), tunnel (Kirchstetter et al., 1999, 2002), and near-road (Shi et al., 2002) studies, as well as measurements with mobile laboratories (Kittelson et al., 2004). In this study, the fuel-based approach with CO_2 variation as a tracer of exhaust dilution in real conditions is used on mobile laboratory measurements.

In Finland, winter tyres are compulsory in motor vehicles during the winter months and approximately 90% of cars and 0% of buses and trucks have winter tires equipped with metal studs. Also, sand and de-icing salt are used to prevent road slipperiness. These factors have a considerable effect on the wear of road surfaces and resuspension of particles from them (Kupiainen and Tervahattu, 2004).

In 2003 in the Helsinki metropolitan area street traffic volume, the share of private cars was 84% of the total mileage and the shares of vans, trucks, and buses were 9%, 5%, and 2%, respectively. The share of private cars with three-way catalysts was 70% of the mileage of gasoline driven cars. Diesel vehicles corresponded to 18% of the mileage of private cars and 92% of the vans (VTT, 2005). Gasoline and diesel fuels sold in Finland are technically sulphur-free (<10 ppm). In Finland, 100% of the total automobile gasoline consumption has been unleaded since 1995.

The objective of this study is to determine the emission factors for CO, NO_x, and particle numbers in different size classes for the car fleet and traffic conditions in the Helsinki metropolitan area. Also, the particle number concentrations and the chemical composition of fine particles in the midst of traffic in relation to urban ambient levels are determined. Size-fractioned data has not been published before although it is urgently needed for dispersion modelling and exposure assessment.

2. Methods

A mobile laboratory called "Sniffer" was utilized to measure real-time traffic emissions in actual traffic conditions. The design of the Volkswagen LT35 diesel van laboratory has been described in detail elsewhere (Pirjola et al., 2004a, b). The mobile laboratory provides measurements of particle total number concentration and size distribution, some gaseous species (CO, CO_2, NO, NO_2, and NO_x), meteorological (T, RH, and wind speed and direction at the roof of the van) and geographical parameters (by a GPS system) with high spatial and time resolutions. Two different inlet systems opening towards the driving direction were constructed, one above the windshield at a height of 2.4 m for general traffic monitoring and the other above the bumper at a height of 0.7 m for chasing a single vehicle. In this study, the sample air was drawn through an inlet above the windscreen. Separate sampling lines were used for particles and gases. In addition to the air pollution measurement devices, the van was equipped with a video camera that recorded the driving conditions through the windscreen.

The concentrations of air pollution components were measured in traffic for 3 days on streets of the Helsinki city centre (22–24 October 2003) and for 3 days on highways in the Helsinki metropolitan area (27–29 October 2003). On both environments, the total 12.5 h measurement time included three morning peak periods (between 7:13 and 10:38) and two afternoon peak periods (between 14:33 and 17:22) (Table 1).

Particle number concentration and size distribution were measured by the Electrical Low Pressure Impactor (ELPI, Dekati Ltd.) with a flow rate of $10\,L\,min^{-1}$. ELPI (with the electrical filter stage) enables real-time particle size distribution in the size range of 7 nm–10 μm (aerodynamic diameter) with 12 channels. The particles are charged, size-classified by inertial impaction, and electrically detected (Keskinen et al., 1992). The offset currents were always checked before and after the measurement. In this work, results of sub-micrometre particles are presented.

LI-6262 CO_2/H_2O Analyzer (LI-COR, inc.) was used for real-time CO_2 monitoring. The detection range for CO_2 was 0–3000 ppm and accuracy ± 1 at 350 ppm. The monitor was calibrated against a new factory-calibrated GM70 (Vaisala hand-held CO_2 meter). Model CO12M (Environnement S.A.) CO gas monitor had a detection range of 0.05–20 ppm. In the NO_x analyser (Model APNA 360, Horiba) the measuring cycle is composed of three different parts: NO-, NO_x-, and zero level measurements. The NO_x detection limit is about 0.5 ppb and the measurement range reaches up to 4 ppm. CO and NO_x analysers were calibrated with five different certified standard gas concentrations. Zero and span checks were performed a couple of times per day for both analysers.