Atmospheric Environment 99 (2014) 175–182

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

A quantitative estimation of the exhaust, abrasion and resuspension components of particulate traffic emissions using electron microscopy

Stephan Weinbruch [a, *], Annette Worringen [a], Martin Ebert [a], Dirk Scheuvens [a], Konrad Kandler [a], Ulrich Pfeffer [b], Peter Bruckmann [b]

[a] Institute of Applied Geosciences, Technical University Darmstadt, Schnittspahnstr. 9, D-64287 Darmstadt, Germany
[b] Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen, Wallneyer Str. 6, D-45133 Essen, Germany

HIGHLIGHTS

- New quantitative approach for determination of traffic components of PM_{10}.
- High fraction (58%) of resuspension component at kerbside station.
- Low fraction (27%) of exhaust component at kerbside station.
- High significance of resuspension component for observed adverse health effects.
- High potential of resuspension component for mitigation measures.

ARTICLE INFO

Article history:
Received 28 February 2014
Received in revised form
12 September 2014
Accepted 17 September 2014
Available online 30 September 2014

Keywords:
Particulate matter
Urban air pollution
Traffic
Electron microscopy

ABSTRACT

The contribution of the three traffic-related components exhaust, abrasion, and resuspension to kerbside and urban background PM_{10} and PM_1 levels was quantified based on the analysis of individual particles by scanning electron microscopy. A total of 160 samples was collected on 38 days between February and September 2009 at a kerbside and an urban background station in the urban/industrial Ruhr area (Germany). Based on size, morphology, chemical composition and stability under electron bombardment, the 111,003 particles studied in detail were classified into the following 14 particle classes: traffic/exhaust, traffic/abrasion, traffic/resuspension, carbonaceous/organic, industry/metallurgy, industry/power plants, secondary particles, (aged) sea salt, silicates, Ca sulfates, carbonates, Fe oxides/hydroxides, biological particles, and other particles. The traffic/exhaust component consists predominantly of externally mixed soot particles and soot internally mixed with secondary particles. The traffic/abrasion component contains all particles with characteristic tracer elements (Fe, Cu, Ba, Sb, Zn) for brake and tire abrasion. The traffic/resuspension component is defined by the mixing state and comprises all internally mixed particles with a high proportion of silicates or Fe oxides/hydroxides which contain soot or abrasion particles as minor constituent. In addition, silicates and Fe oxides/hydroxides internally mixed with chlorine and sulphur containing particles were also assigned to the traffic/resuspension component. The total contribution of traffic to PM_{10} was found to be 27% at the urban background station and 48% at the kerbside station, the corresponding values for PM_1 are 15% and 39%. These values lie within the range reported in previous literature. The relative share of the different traffic components for PM_{10} at the kerbside station was 27% exhaust, 15% abrasion, and 58% resuspension (38%, 8%, 54% for PM_1). For the urban background, the following relative shares were obtained for PM_{10}: 22% exhaust, 22% abrasion and 56% resuspension (40%, 27%, 33% for PM_1). Compared to previous publications we have observed a significantly lower portion of exhaust particles and a significantly higher portion of resuspension particles. The high abundance of resuspension particles underlines their significance for the observed adverse health effects of traffic emissions and for mitigation measures.

* Corresponding author.
E-mail address: weinbruch@geo.tu-darmstadt.de (S. Weinbruch).

http://dx.doi.org/10.1016/j.atmosenv.2014.09.075
1352-2310/

1. Introduction

Traffic is an important to dominant source of the atmospheric particle load in urban areas (e.g., Lenschow et al., 2001; Querol et al., 2001; Marcazzan et al., 2003; Salvador et al., 2004; Mossetti et al., 2005; Furosjö et al., 2007; Viana et al., 2008; Gietl and Klemm, 2009; Belis et al., 2013). Particulate traffic emissions may be divided into an exhaust, abrasion (brake, tire) and resuspension (road dust) component (e.g., Abu-Allaban et al., 2003; Quass et al., 2008; Thorpe and Harrison, 2008; Denier van der Gon et al., 2013). In addition, gaseous emissions from traffic (e.g., NO_x, SO_2, volatile organic compounds) lead to formation of secondary aerosol particles.

Air pollution from traffic is associated with a number of adverse health effects including increased mortality (e.g., Hoek et al., 2002; Finkelstein et al., 2004) and respiratory symptoms/diseases in children (e.g., Brunekreef et al., 1997; Gehring et al., 2002; Janssen et al., 2003; Kim et al., 2004; Brauer et al., 2007; Gauderman et al., 2007). In a recent review (Denier van der Gon et al., 2013), the importance of non-exhaust traffic-induced particles for the observed adverse health effects is emphasized.

It was shown in several publications (e.g., Harrison et al., 2001; Lenschow et al., 2001; Sternbeck et al., 2002; Abu-Allaban et al., 2003; Gehrig et al., 2004; Hueglin et al., 2005; Mossetti et al., 2005; Thorpe et al., 2007; Quass et al., 2008; Amato et al., 2009; Bukowiecki et al., 2010) that the contribution of non-exhaust primary particles to the total traffic-generated primary particles in urban areas is significant. Furthermore, the ratio of non-exhaust to exhaust particles is strongly increasing in the last two decades, due to exhaust emission reductions (Denier van der Gon et al., 2013).

The increasing significance of non-exhaust particles clearly underlines the need for analytical methods which allow discrimination of the different traffic components (Denier van der Gon et al., 2013). Previous estimates of the abundance of the different traffic components are mainly based on (a) bulk chemical analysis combined with receptor modelling (e.g., Abu-Allaban et al., 2003; Mossetti et al., 2005; Quass et al., 2008; Amato et al., 2009; Bukowiecki et al., 2010), or (b) analysis of particle mass concentrations (e.g., Harrison et al., 2001; Lenschow et al., 2001; Gehrig et al., 2004; Charron and Harrison, 2005; Hueglin et al., 2005; Thorpe et al., 2007).

In the present paper, an approach for the quantitative determination of the different traffic components is presented, which is based on the analysis of individual particles by scanning electron microscopy (SEM) and energy-dispersive X-ray microanalysis (EDX). Exhaust and abrasion particles are recognized by their morphology and chemical composition, resuspension particles additionally by their mixing state.

2. Experimental

2.1. Sampling

In the period from February to September 2009 particle sampling was performed at two state monitoring stations within the Ruhr area, the largest agglomeration in Germany (approximately 5.1 million residents). The kerbside station Essen – Gladbecker Straße (VEAE; latitude: 51° 28′ 45″ N; longitude: 7° 0′ 21″ E; 55 m asl) is located in a street canyon next to a major four lane street, the urban background station Mülheim – Styrum (STYR; latitude: 51° 27′ 17″ N; longitude: 6° 51′ 56″ E; 37 m asl) within a residential area close to a sports field. In 2004, the daily traffic volume at the kerbside station was ≈40,000 vehicles with ≈5.2% heavy-duty vehicles. Both sampling sites lie about 10.5 km apart. Sampling was performed 1.5 m above ground.

Particles were collected on boron substrates using a two-stage micro inertial cascade impactor (50% cut of diameters: 0.1–1 μm and 1–10 μm) at a flow rate of 0.5 l/min. At each location, sampling of the two different size fractions was performed successively with different sampling times in order to obtain a coverage of the substrate with particles optimal for automated analysis (2–5% of the area). Depending on the particle concentrations, the sampling time was 45–240 s for the fine particle fraction, and 90–1000 s for the coarse particle fraction. Sampling at the two different locations was generally performed within 2 h. In total, 160 samples (40 sets of two samples from each location) were collected at 38 days. After screening of all 160 samples in the scanning electron microscope, 80 samples (20 days with two size fractions from both locations) were selected for detailed investigation. The selection criterion was that all four samples of the same day are suited for automated analysis in the scanning electron microscope. Sampling day, sampling time and duration, PM_{10} during sampling, and flow direction of the air masses derived from 96 h back-trajectories are listed in the Supplementary data (Table S1).

2.2. Back-trajectories

Ninety six hours air mass back-trajectories were calculated with the software 'Ready Hysplit 4' (Draxler and Rolph, 2003; Rolph, 2003) using starting heights of 0 m, 100 m and 200 m (above ground), respectively. The air mass back-trajectories for the 20 days investigated in detail are given in the Supplementary data (Fig. S1).

2.3. Electron microscopy

The size, shape, morphology, chemical composition, and mixing state of 111,003 individual particles were determined by high-resolution scanning electron microscopy (using secondary and backscattered electron imaging) and energy-dispersive X-ray microanalysis. All measurements were carried out with a field emission gun environmental scanning electron microscope (FEI ESEM Quanta 200 FEG, Eindhoven, The Netherlands) operated at 20 kV acceleration voltage. The particles were studied without coating in the high vacuum mode of the instrument ($\approx 5 * 10^{-6}$ mbar sample chamber pressure). The particle size (equivalent projected area diameter) was obtained from secondary and backscattered electron images. The spatial resolution obtained with secondary electrons was approximately 10 nm, and 15–20 nm for backscattered electrons (both on atmospheric particles). This spatial resolution was sufficient for identification of soot, fly ashes and biological particles by manual inspection (the particle groups where morphology is used as classification parameter). The chemical composition (elements with $Z \geq 5$) of the particles was measured with an energy-dispersive X-ray detector (EDAX, Tilburg, The Netherlands). Size and chemical composition of the individual particles were determined automatically using the software Genesis 5.11 (AMETEK, Wiesbaden, Germany). The automated analysis was restricted to particles with an equivalent projected area diameter above 0.2 μm. The morphology and mixing state of all 111,003 particles were determined manually. Net X-ray count rates were used for particle classification (see Chapter 3), i.e., matrix and geometric effects were not corrected for. It should be emphasized here, that net X-ray count rates are sufficient for classification of particles into the different groups according to the present scheme. Quantification of the particle composition prior to classification would not lead to different particle groups.

Particles of known composition and a geometric size between 0.7 and 10 μm are routinely measured in our lab (at least once a week), and the results with and without ZAF (atomic number, absorption, fluorescence) correction as well as with and without