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Analytical electron microscopy of combustion particles: a comparison of vehicle exhaust and residential wood smoke

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Abstract

Particulate matter has been associated with a number of adverse health effects. Since combustion particles from vehicle exhaust and wood smoke are common constituents of ambient air, the morphology and elemental composition of particles from these two sources were analysed and compared using single particle analysis. Ambient air particles were collected in locations dominated by vehicle exhaust or residential wood smoke. To verify the source contributions to the ambient air samples, particles were collected directly from the combustion sources. All particulate samples were analysed on carbon extraction replica by transmission electron microscopy (TEM) and X-ray microanalysis (XRMA). The particles were classified into four groups based on morphology and elemental composition. Carbon aggregates were the only particles identified to originate from combustion sources and accounted for more than 88% of the particle numbers in the ambient air samples from both sources. The carbon aggregates were therefore further analysed with respect to morphology and elemental composition on germanium extraction replica. Carbon aggregates from vehicle exhaust were characterised by higher levels of Si and Ca compared to wood smoke aggregates that contained higher levels of K. The S content in aggregates from both sources was probably caused by interaction with gases in the air. Furthermore, the diameters of primary particles from vehicle exhaust were significantly smaller (27 ± 7 nm) than the diameters for wood smoke (38 ± 11 nm). The observed differences in elemental profiles and primary particle diameters for vehicle exhaust and wood smoke may influence the health effects caused by these particles.

Keywords: mCarbon aggregates; Wood combustion; Vehicle exhaust; Transmission electron microscopy; X-ray microanalysis; Elemental composition

1. Introduction

Combustion particles from vehicle exhaust and wood smoke are common constituents of urban particulate matter. The reported number concentrations for combustion particles in urban environments

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range from 30% to 80% (Katrinak et al., 1995; Paoletti et al., 2002; Shi et al., 2003). While epidemiological studies have demonstrated adverse health effects of exposure to particulate matter in general (Brunekreef and Holgate, 2002; Pope et al., 2002), the application of source apportionment methods have also associated combustion particles from motor vehicles with increased hospital admissions (Janssen et al., 2002) and increased mortality (Laden et al., 2000; Mar et al., 2000). Similarly, studies of subjects living near major roads have associated traffic emissions with increased asthma (Brauer et al., 2002) and mortality (Hoek et al., 2002). For residential wood smoke, a recent review reported a significant association between acute asthma and increased levels of wood smoke particles (Boman et al., 2003). The particle characteristics responsible for the adverse health effects reported in epidemiological studies are, however, still widely debated (Harrison and Yin, 2000). A number of in vivo and in vitro toxicological studies have examined particles derived from different sources, such as ambient air, combustion and natural sources, as well as laboratory-derived surrogate particles. These studies have suggested certain particle characteristics like size and contents of transition metals and organic compounds as possible casual properties for PM-associated health effects (Dreher, 2000).

While particulate matter with diameters up to 100 μm are inhalable, only particles with aerodynamic diameters below 10 μm (PM_{10}) are considered to be relevant for health effects. The PM_{10} fraction is commonly divided into a coarse fraction containing particles with diameters >2.5 μm ($PM_{2.5-10}$) and a fine fraction with diameters <2.5 μm ($PM_{2.5}$). The probability of particle deposition and the deposition areas within the respiratory tract vary considerably with the aerodynamic size of the particles. Particles smaller than 4 μm can penetrate deeply into the lung to the bronchial and alveolar regions, while the larger particles are trapped in the upper airways (Phalen, 2002). The size distribution, morphology and elemental composition of combustion particles vary with combustion source (Lighty et al., 2000; Hedberg et al., 2002). Spherical carbon particles (diameters ≈30–50 nm) found as individual particles, aggregates or large complex agglomerates are observed most frequently in vehicle exhaust (Berube et al., 1999; Dye et al., 2000; Paoletti et al., 2002). Carbon aggregates have also been observed in wood smoke collected from fireplaces and single-stage wood stoves during high-temperature combustion (Evans et al., 1981; Dasch, 1982; Tesfaigzi et al., 2002). In contrast, particles emitted from other wood combustion appliances (e.g., boilers for wood chips or pellets) have been reported with morphologies different from carbon aggregates (Hueglin et al., 1997; Osan et al., 2002; Mavrocordatos et al., 2002).

Winters in Norway are cold, with long periods of temperatures below 0°C. In urban areas, the principal sources of pollutant emissions are vehicular traffic and residential wood combustion. These two sources account for more than 65% of the total particulate matter emissions in Norway (Skogesal et al., 2001). The levels of combustion particles may be particularly high on cold days with little wind when low-lying inversion layers cause little exchange of air (Namork et al., 2004). It is of special interest to characterise particles from vehicle exhaust and wood smoke since morphology, surface area and chemistry may be of importance for the particle potency and their health effects (Lighty et al., 2000; Dreher, 2000; Donaldson et al., 2001). The objective of this study was to analyse and compare combustion particles from vehicle exhaust and residential wood smoke from a single particle perspective, using transmission electron microscopy (TEM) and X-ray microanalysis (XRMA). Particles were collected from both ambient air and directly from the combustion sources. The particles were studied on carbon extraction replicas, and the combustion particles were recognised by morphology and analysis of elemental composition. To relate the elemental analysis to the carbon content of the particles, the combustion particles were also analysed on germanium extraction replicas using a thin window XRMA detector.

2. Materials and methods

2.1. Sample collection and sites

The Respicon® particle sampler (TSI, USA) is a virtual impactor collecting three particle fractions, $PM_{2.5}$, $PM_{2.5-10}$ and PM_{10-100}. In this study, the $PM_{2.5}$