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Variation in characteristics of ambient particulate matter at eight locations in the Netherlands – The RAPTES project

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A B S T R A C T

Numerous epidemiological studies have shown health effects related to short- and long-term exposure to elevated levels of ambient particulate matter (PM). It is not clear however which specific characteristics (e.g., size, components) or sources of PM are responsible for the observed effects.

The aim of RAPTES (Risk of Airborne Particles: a Toxicological–Epidemiological hybrid Study) was to investigate which specific physical, chemical or oxidative characteristics of ambient PM are associated with adverse effects of PM on health. This was done by performing experimental exposure of human volunteers to air pollution at several real-world settings that had high contrast and low correlation between several PM characteristics.

For this goal, eight sites in the Netherlands that differed in local PM emission sources were chosen for extensive air pollution characterization. Measurement sites included an underground train station, three different road traffic sites, an animal farm, a sea harbor, a site located in the vicinity of steelworks, and an urban background site. Five- to six-hours average concentration measurements at each site were made between June 2007 and October 2009. We measured PM_{10}, $PM_{2.5}$, particle number concentration (PNC), oxidative potential of PM, absorbance, endotoxin content, as well as elemental and chemical composition of PM, and gaseous pollutants concentrations. This paper presents a detailed characterization of particulate air pollution at the sampling sites.

We found significant differences in all PM characteristics between the sites. The underground train station, compared to each outdoor location, had substantially higher concentrations of nearly all PM characteristics. The average PM_{10} and $PM_{2.5}$ mass concentrations at the underground train station were 394 $\mu g\ m^{-3}$ and 137 $\mu g\ m^{-3}$, respectively, which was 14.1 and 7.6 times higher than the urban background. The sum of the concentrations of trace metals in fine and coarse PM was nearly 20 times above the outdoor levels. Elemental carbon (EC) was elevated at the underground site in the fine but also in the coarse mode, in contrast to the traffic sites where EC was predominantly found in fine PM. The highest concentrations and contrasts in PNC were at the traffic sites (between 45,000 and 80,000 particles cm^{-3}), which was several times higher than measured at any other site. Correlations of PNC with metals, PM_{10}, $PM_{2.5}$ and absorbance were low to moderate, while correlations between PM_{10}, $PM_{2.5}$ and the metals Cu and Fe were high. After excluding the underground train station data, correlations between PM10, EC and metals decreased whereas the correlation between PNC and EC increased.

We conclude that we were able to successfully identify and characterize real-world situations with very different particle characteristics. High contrast and low correlations between PM characteristics, as well as consistency of these differences across sampling campaigns, provide a good basis for identifying health relevant PM characteristics in the upcoming analysis.

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1. Introduction

In recent years numerous epidemiological studies have shown health effects related to short- and long-term exposure to elevated levels of ambient particulate matter (PM) (Brunekreef and Holgate, 2002; Pope and Dockery, 2006). These observations are supported by findings from controlled human exposure studies, animal toxicology and mechanistic *in vitro* studies (Maier et al., 2008; Lippmann and Chen, 2009).

Apart from regulated PM_{10} and $PM_{2.5}$ mass concentrations, only a small number of PM characteristics are measured. Health effects have been mostly associated with the PM mass concentration and in few studies with specific PM characteristics such as number concentration, surface area, elemental and chemical composition, or oxidative potential (WHO, 2007; Brunekreef, 2010). Additionally, in epidemiological studies high correlations are typically present between the constituents (particulate as well as gaseous) of the air pollution mixture, which makes it difficult to disentangle their independent effects on human health (Brunekreef and Holgate, 2002).

A recent World Health Organization workshop (WHO, 2007) on the health relevance of PM from various sources suggested that future research should, among other things, (1) explore “the role of various characteristics of ultrafine, fine and coarse thoracic particles that might be responsible for health effects, (2) consider “the contributions of different emissions sources to population exposure”, (3) consider “other biological effects in the studies, beyond classically measured mortality or respiratory function and symptoms”, (4) ensure “that the exposure levels used in toxicological studies correspond to the components and levels of exposure experienced by populations and evaluated by epidemiological studies”, and (5) promote “an integrated study approach, combining detailed characterization of exposure, epidemiological observation of effects in populations and toxicological or clinical evaluation of the effects”.

We attempted to address these recommendations in the RAPTES project (Risk of Airborne Particles: a Toxicological–Epidemiological hybrid Study). The overarching aim of RAPTES was to combine real-world exposure conditions with an experimental exposure design so that health effects could be associated with specific ambient PM characteristics. In order to achieve this, we performed an extensive exposure assessment and characterization of physical, chemical and oxidative properties of PM at sites which were selected to provide high contrast and low correlation between PM characteristics and between contributing sources. At selected sites health effects were investigated in a panel of volunteers. PM samples were also collected for *in vivo* and *in vitro* experiments to investigate the mechanisms behind the health effects.

In this paper we present a detailed characterization of air pollution at the selected sites focusing on particulate air pollution and the contrast and correlations between its physical and chemical characteristics. The associated health impacts will be presented in subsequent publications.

2. Materials and methods

2.1. Sampling site selection

We selected sites anticipated to provide a large contrast in exposure to specific characteristics of PM as well as a low correlation between PM characteristics, based upon previous studies of specific sources. The characteristics we took into account were mass concentration of particles less than 10 μm and less than 2.5 μm in aerodynamic diameter (PM_{10} and $PM_{2.5}$, respectively) as well as the mass concentration of coarse PM fraction ($PM_{2.5-10}$), particle number concentration (PNC) as a proxy for ultrafine particles, absorbance of PM, trace metal composition, secondary organic and inorganic components, and endotoxin content. These characteristics were chosen because of hypotheses that they may explain (part of) the observed associations between PM and health. The following sites, representing unique emission characteristics, were selected for the study (Fig. 1):

1. The *underground* site in Amsterdam is a train station where all the trains enter and exit the station through a 5.1-km long tunnel. Only electric trains are allowed in the tunnel, with the exception of maintenance trains (two per week). The tunnel has two tubes which accommodate four railway tracks with additional two tracks in the area of the platforms. We sampled on the middle of three platforms and the equipment was positioned approximately half way between the tunnel exits. There is no air conditioning or ventilation at the platforms (only passive exchange of air with the airport terminal building located above), but the tunnel is equipped with jet-fan longitudinal ventilation connected to several ventilation shafts. At the underground train station the abrasive braking and accelerating actions of the steel surfaces of wheels, gears, rail tracks, overhead lines, etc. and the high-velocity, turbulent air motion could cause aerosolization of metals in both coarse and fine PM fractions.
2. The *urban background* site was located in Rotterdam in a garden semi-enclosed by apartment buildings and a school. The site was not influenced by single, local sources such as traffic.
3. The *farm*, located nearby Utrecht, was a medium-sized organic farm (ca. 600 animals). Sampling took place outdoors in close vicinity to an open area where the animals were kept. Elevated endotoxin content could be measured at this site due to the ubiquity of biological material present.
4. *Continuous traffic* site was located at the exit of a motorway tunnel to monitor the increased concentrations of primary tailpipe vehicle emissions.
5. The *stop-and-go traffic* site was a major inner-city intersection where in addition to primary tailpipe emissions, non-tailpipe vehicle emissions (i.e., tire and brake wear material) contribute to exposure.
6. The *truck traffic* site was also located at a busy intersection, but on a road leading to the harbor terminals. Traffic there was dominated by 55% heavy diesel trucks, therefore resulting in an even higher proportion of ultrafine particles and increased PM absorbance than at the other traffic sites.

All traffic sites were located in or around Rotterdam.

7. The *harbor* site was located at a pier on the New Waterway (Nieuwe Waterweg) which is a primary shipping route between the busy Port of Rotterdam and the North Sea. At the site, shipping emissions were associated with high V and Ni concentrations.
8. The site in the vicinity of *steelworks* was located north-west of Amsterdam in a quiet residential area, which in previous studies showed a significant impact of the steel factory on the air pollution levels. The site was located ca. 1–2 km from the source to increase the chance of sampling the material emitted from the high stacks. PM around steelworks could be characterized by a high transition metal content.

At all sites, except the steelworks site, sampling was performed at the closest possible distance to the source (Table 1). Pictures are provided in Appendix A.