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Total and size-resolved particle number and black carbon concentrations near an industrial area

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HIGHLIGHTS

- Ultrafine particles are a factor 3 elevated 5 km downwind industrial area.
- The size-distribution of these particles is dominated by particles of 10–50 nm.
- 70,000 addresses exposed to annual average additional 5–10,000 #/cm^3.

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ABSTRACT

Total and size-resolved particle number and black carbon concentrations were investigated in urban areas of the city of Rotterdam (the Netherlands) situated near an industrial area. Several monitoring campaigns were conducted in the period 2011–2014 at three local locations and at a regional background site. Black carbon levels showed minor elevation due to industrial emissions. In contrast, particle number concentrations (PNC) increased during periods with wind directions from the industrial area, by 1000 to 23,000 particles per cm^3 depending on the distance to the area from 1 to 40 km. The size distribution of elevated PNC was characterized by two modes: 10–20 nm (nucleation particles) and 20–100 nm (Aitken particles). Five dominant industrial sources were identified and used as input for dispersion modelling of PN in 2012. The results showed that in Rotterdam about 70,000 addresses were exposed to an additional annual PNC of 5000–10,000 particles per cm^3 and about 55,000 addresses to additional PNC of 10,000–20,000 particles per cm^3 for 39% of the time. More measurements of PNC up- and downwind of the industrial area are recommended to identify more accurately the PN emission sources and to validate the dispersion modelling.

1. Introduction

The association between exposure to particulate matter (PM) in ambient air and adverse health effects has been found in many epidemiological studies. Various physical and chemical fractions in PM have been proposed as the causative mechanism (Cassee et al., 2013). Submicron particles are a suspect fraction as they may contain potential toxic species and can enter deep into the respiratory system (Oberdörster et al., 2005). Submicron particles may be distinguished into three size modes, with distinctive sources and chemical composition: 1–20 nm (*nucleation* particles), 20–100 nm (*Aitken* particles) and 20–300 nm (*accumulation* particles) (Kumar et al., 2010). Particles smaller than 100 nm are generally referred to as *ultrafine* particles. Nucleation particles in ambient air, mostly consist of secondary sulphates, nitrates and organic compounds (Chow and Watson, 2007). Nucleation particles are also directly emitted from combustion sources and formed by cooling of semi-volatile components (Maricq, 2007). Aitken particles originate from the growth and coagulation of nucleation particles and are also emitted directly from primary combustion processes. Accumulation particles in combustion emissions are mainly composed of *black carbon*: a mixture of elemental carbon (EC), organic compounds (OC) and condensed material, such as sulphuric acid, lubricating oil and metals (Lingard et al., 2006). In combustion emissions, the *mass* of particles is dominated by accumulation particles, while the *number* of particles is dominated by the ultrafine particles (Kittelson et al., 2004). In ambient air the number of particles is dominated by ultrafine particles and therefore PNC is

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considered to be representative for the concentrations of ultrafine particles (Chow and Watson, 2007).

Road traffic is the main source of ultrafine particles emissions in urban areas (Kumar et al., 2010). Other sources are combustion emissions from airports, shipping, industry, power plants and refineries, and photochemical formation (González and Rodríguez, 2013; Keuken et al., 2015; Reche et al., 2011). While these studies conclude that a significant number of ultrafine particles are emitted from these non-traffic sources, their contribution to ambient PNC in nearby urban areas is less investigated (Kumar et al., 2013). This paper presents a study on the emission of total and size-resolved PNC and black carbon from an industrial and harbour area, and their dispersion to the city of Rotterdam area in the Netherlands. Refineries were identified the main emitters of PNC in the study area. In view of the limited studies on industrial emissions of PN (Al-Dabbous and Kumar, 2014; Fernández-Camacho et al., 2012), the findings and conclusions from this study may also be relevant for other refineries.

2. Methodology

2.1. The study approach

In selected periods between 2011 and 2014, total and size-resolved PNC and black carbon (BC) were measured at four sites in the Netherlands, including a regional background site. In 2011, total PNC and BC were measured at an urban background site in Rotterdam. In 2012 total PNC was measured at another urban background site in Rotterdam and size-resolved PNC and BC were measured at the regional background site. It is noted that these measurements were part of other studies, but when their results were combined an industrial area west of the city of Rotterdam was identified as a source of PN emissions. To further investigate these emissions, in 2014, a monitoring site was set up north of this industrial area. From April to July 2014, sulphur dioxide (SO_2), total and size-resolved PNC and BC were measured at this site.

Meteorological data were retrieved from the National Meteorological Monitoring Network at the Rotterdam Airport and Cabauw sites (www.knmi.nl). In view of the distance less than 2 km to the study area, the hourly average wind direction (which is an important parameter in this study) at the Rotterdam Airport is considered to be representative for the urban background and the industrial monitoring sites. Emission data from the industrial area were available from the emission inventory managed by DCMR Environmental Protection Agency Rijnmond (DCMR, www.dcmr.nl). The meteorological and emission data were used as input for a dispersion model to estimate the contribution of industrial-related PNC in urban areas in Rotterdam in 2012. Finally, the annual average (long-term) exposure of the population to PNC in these areas was estimated and compared to (short-term) exposure during periods in which the wind was coming from the industrial area (short-term exposure).

2.2. Monitoring sites

The monitoring sites were the national regional background site at *Cabauw*, an industrial site on the north bank of the Nieuwe Maas river (*Vlaardingen*) and two urban background sites in Rotterdam (*Zwartewaalstraat* and *Statenweg*). Cabauw is at a distance of more than 20 km from Rotterdam without emission sources in the direct vicinity of this site. The sampling inlet at this site is at 80 m height. Vlaardingen is 1 km north of the investigated industrial area where PN emissions were identified and the sampling inlet was at 2.5 m height. The first urban background site, Zwartewaalstraat, is part of the DCMR monitoring network in the Rijnmond area with a sampling inlet at 2.5 m height, while the second urban background site of Statenweg was set up at 15 m height on the rooftop of a building along the Statenweg in the centre of Rotterdam. The four sites are shown in Fig. 1 and the measurements at these sites are summarized in Table 1.

2.3. Monitoring equipment

Black carbon concentrations were measured with a multi-angle absorption photometer – the MAAP model 5012 (Thermoscientific, Germany). The sampling flow rate was 16.5 l per minute and the measurement frequency was two minute average. The black carbon concentrations were converted to elemental carbon (EC) concentrations to facilitate comparison with thermographic measurements of EC (Keuken et al., 2013). A factor of 0.75 was applied to convert the MAAP data to elemental carbon concentrations. The detection limit of the MAAP is 0.1 μg EC per m^3. Total PNC was measured using a CPC 3025A (TSI Inc., USA) with a 50% cut-off at 3 nm and a concentration range of up to 10^5 particles per cm^3. The sampling flow rate was 1.5 l per minute and the measurement frequency was 1 s. Size-resolved PNC was measured with a Scanning Mobility Particle Sizer (SMPS 3080; TSI Inc., USA). The SMPS consists of a differential mobility analyser (DMA) covering a size range of 10–480 nm and a CPC 3775 (TSI Inc., USA) with a 50% cut-off at 4 nm and a concentration range up to 10^4 particles per cm^3. The sampling flow rate was 2.5 l per minute and a measurement scan takes about 3 min. Sulphur dioxide measurements were conducted by a 100E UV fluorescence monitor (Teledyne, USA) with a detection limit of 1 ppb SO_2. The sampling flow rate was 0.6 l per minute and the measurement frequency was 1 min.

2.4. Dispersion modelling

Dispersion modelling in this study was not conducted for BC as the contribution from industrial sources to BC was relatively too small (see: section 3.3). The contribution of PN emissions from the industrial source area to hourly average PNC in the urban areas of Rotterdam was modelled with a Gaussian plume model (*SRM3*). This concerned both the monitoring period from 9th April till 15th July 2014 and for the whole year 2012. SRM3 is the regulatory model used in the Netherlands to calculate the dispersion of air pollutants from point and surface area sources to a maximum distance of 25 km. The dispersion of PN was approximated as an inert gas. Thus, processes other than dilution were ignored beyond 1 km from the emission sources in the modelling domain of about 10 km^2 in this study. The modelling procedure concerns a number of steps which are described in more detail in Keuken et al. (2015). Based on observed concentrations in different wind directions at the monitoring site in Vlaardingen five potential point sources were identified in the industrial area and their relative source strength was estimated. Next, a conversion factor between modelled and measured contribution to PNC at the Vlaardingen site was established for the monitoring period April–July 2014. Finally, the 2012 contribution to concentrations of PNC in urban areas of Rotterdam was modelled using the meteorological data from 2012. The grid size of the modelling output was 12*8 km and the grid resolution was 0.5*0.5 km. The spatial resolution of the output was increased to 50*50 m by interpolation between the modelled PNC. The interpolation was performed with the 'natural neighbour' algorithm from the spatial analyst tool in ArcGIS software (www.esri.com). The exposure of the population to industrial-related PNC was determined by matching the postal addresses in the urban areas from the national cadastral survey (www.kadaster.nl) with the calculated spatial distribution of PNC.