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Urban background levels of particle number concentration and sources in Vilnius, Lithuania

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ABSTRACT

This study presents results of research on urban aerosol particles with a focus on the aerosol particle number concentration (PNC) and the particle size distribution. The real time measurements of aerosol PNC (>4.5 nm) and number size distributions (9–840 nm) were performed. The seasonal variations essentially comprised the minimum monthly mean in October 2010 (3400 ± 3000 cm^{-3}) and the maximum in April 2011 (19,000 ± 15,000 cm^{-3}). The mean annual PNC was 10,000 ± 8000 cm^{-3} with an average mode size of 30–50 nm. The presence of strong diurnal patterns in aerosol PNC was evident as a direct effect of three sources of aerosol particles (nucleation, traffic, and residential heating). Hybrid receptor modeling potential source contribution function (PSCF) and concentration weighted trajectory (CWT) were used by incorporating 72-h backward trajectories and measurements of PNC in Vilnius. The results of trajectory clustering and the PSCF method demonstrated that possible additional source areas contributing to the elevated particle number concentration in Vilnius could be industrial areas in central Europe. Principal component analysis (PCA) revealed highest loadings for PNC, PM10, NO_x, NO, NO_2 and SO_2 concentrations, indicating combustion processes occurring in vehicle engines and use of sulfur-containing fossil fuels for residential heating.

1. Introduction

The adverse impacts of nanoparticles on the human health (Brook et al., 2010) in urban environment have stimulated the scientific community to research urban air quality. The European Union, in common with many other administrations, has established tough targets for air quality in order to limit adverse effects upon human health. Although the mass of airborne particulate matter (PM) in the ambient atmosphere is the subject to regulation, there continues a debate about which PM size fractions have a more harmful effect on human health. This is important from a public health point of view because recent research has indicated that ultrafine particles (UFP: particles with diameters <100 nm) may be more toxic per unit mass than the larger size fractions of PM (Sioutas et al., 2005; Pope et al., 2002) and they sooner deposit in the deepest alveolar portions of the respiratory tract as a function of size (Daigle et al., 2003), induce oxidative stress (Araujo et al., 2008), and translocate into secondary organs and tissues (Oberdörster et al., 2004). The European Union (EU) set two limit values for PM10 concentrations: annual average — 40 μg m^{-3}, 24-hour limit value — 50 μg m^{-3} not to be exceeded more than 35 times a calendar year (2008/EC/50). However, the implementation of the EU air-quality directives and introduction of more stringent vehicle emission standards were deferred since some countries were unable to meet the pre-existing limit value. In consequence fact, the decreased PM mass concentration has increased the concentration of UFP in Western countries (Morawska et al., 2002). Unavoidably, the legislative pressures arising

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from air quality standards turn attention to identification of PM source categories and their geographic distribution.

Previous studies have suggested that the major influence on contributions to the PNC is caused by the included vehicle exhaust emissions during the traffic peak hours and photochemical nucleation events (Ketzel et al., 2004; Holmes et al., 2005; Jeong et al., 2006; Cyrys et al., 2008; Pey et al., 2008; Perez et al., 2010), the residential heating during winter periods (Hussein et al., 2004) and new particle formation (NPF) by photochemical reactions (Perez et al., 2010). Typically, the formation process occurs over large areas (Plauškaitė et al, 2010). Therefore, it may significantly contribute to changing the regional climate (Spracklen et al., 2008).

In recent years, these events have been studied extensively in background (O'Dowd et al., 2002; Rodriguez et al., 2005) and also urban (or polluted) areas (Tuch et al., 2006; Kulmala et al., 2005). It was found that the anthropogenic emission sources such as manufactories and the power plants (Köhler et al., 2008), the tire and road surface wear (Stocker and Carruthers, 2007), burning biofuel (Karvosenoja et al., 2008; Kumar et al., 2010), the aircraft activity (Hu et al., 2009), the natural sources (Holmes, 2007), the biomass burning (Simmonds et al., 2005; Wardoyo et al., 2007) and the long-range transport (Beverland et al., 2000) were important contributors to the PNC mostly in the <300 nm size range (Kumar et al., 2008). The particles from the residential combustion can sometimes grow in size in the urban environments mainly due to condensation of gases (Park et al., 2008). The exhaust emissions from the gasoline and diesel-fuel vehicles remain the dominant source in polluted urban environments (Harrison et al., 2011; Kumar et al., 2011). The size range for the road traffic emissions is consistent with the particle number size distributions for the gasoline direct injection and the diesel engines with the majority of particles in the size diameter interval of 20–60 and 20–130 nm, respectively (Maricq et al., 1998; Morawska et al., 1998, 2008). These can alone contribute up to about 90% of the total PNCs (Perez et al., 2010) reaching magnitudes of 10^4–10^5 cm^{-3} during the nucleation events (Cheung et al., 2010).

In the case of the Lithuania, NPF has been studied by Plauškaitė et al. (2010) in a relatively clean background marine area site. These studies revealed NPF taking place more frequently in conjunction with high levels of solar radiation. In spite of its importance, no long-term continuous measurement campaigns studying the PNC dynamics have been carried out at urban sites in Lithuania so far. As NPF was found to be a regional phenomenon it was expected that NPF would also occur in the polluted urban areas. The current work is aimed at studying the source areas and processes affecting or contributing to PNC at an urban site for the period June 2010–September 2011. To reach this objective, the work focuses on the seasonal evolution of the source apportionment of PNC based on back-trajectories clustering techniques to identify the regional area sources by the concentration weighted trajectory (CWT) and the potential source contribution function (PSCF). Additionally some nucleation events have been identified and characterized to find out that the most likely conditions lead to high PNC concentration appearance in the measuring site.

2. Data set and methodology

2.1. Description of the study region

The investigations of particle size distribution and PNC are presented within this paper. The aerosol PNC was analyzed during the period from 1 June 2010 to 30 September 2011 in Vilnius. The continuous aerosol measurements were taken at the top floor of the academic building of the Center for Physical Sciences and Technology campus located in Vilnius. The inlet of sampling system was placed on the top floor about 20 m above the ground level, 12 km southwest of downtown area. The location can be described as an urban background (Fig. 1).

Under the normal meteorological conditions, the potential for accumulation of the vehicle emissions is limited by the site location. The urban sampling site was relatively far from the local sources of the primary particles. The nearest highway was 0.24 km to the southwest; on the opposite side a low traffic road was 0.6 km away.

2.2. Instrumentation

The aerosol PNC was continuously measured using a Condensation Particle Counter (CPC, custom built) UF-02 (Mordas et al., 2005). The CPC was designed to detect the particle number concentration from 0.002 up to 100,000 cm^{-3}. The detection efficiency reaches a value of 1 at large particle sizes and it is smaller than 0.9 for particles smaller than approximately 5 nm. The maximum observable number concentration (accuracy 20%) is 100,000 cm^{-3}. The detection efficiencies of CPC were carefully measured in the calibration set-up presented by Mordas et al. (2005). The design of the instrument is based on the swirling flow generated inside the saturator (43 °C)-condenser (10 °C). The instrument uses a high flow rate (1 l min^{-1}) of the carriers. From the carrier flow, the aerosol flow (0.27 l min^{-1}) is extracted by a capillary. This aerosol flow is divided into two flows. The first one (0.03 l min^{-1}) is directed to the condenser. The second flow (0.24 l min^{-1}) is circulated through a HEPA filter and a saturator block, in which the flow is saturated with respect to n-butanol and then mixed with the aerosol-laden air before cooled condenser. This mixing generates a supersaturated region with respect to n-butanol. The n-butanol vapor condenses on the particles which act as condensation nuclei. This process increases the size of each individual nanoparticle. Such large droplets can be conveniently detected by light scattering. The lower cut-off size of the CPC, i.e. the limiting size when 50% of the particles are successfully accounted for, is determined to be ≥4.5 nm (Mordas et al., 2005). The instrument is fitted with an impactor (laminar flow, nozzle diameter = 7.4 mm) to reduce the influence of the large particles. The yearly maintenance included the CPC calibration and thorough cleaning.

A Magee Scientific Company Aethalometer™, Model AE40 Spectrum, manufactured by Optotek, Slovenia was deployed at the Vilnius site and provided realtime, continuous measurements of the BC mass concentrations. The optical transmission of carbonaceous aerosol particles was measured sequentially at seven wavelengths λ (0.37, 0.45, 0.52, 0.59, 0.66, 0.88 and 0.95 μm). The 0.88 μm is considered as the standard channel for BC measurements as at this wavelength BC is the principal