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Concentration levels and source apportionment of ultrafine particles in road microenvironments

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HIGHLIGHTS

- Mobile measurements of UFPs were performed on various road microenvironments.
- PMF was applied to particle size distribution data for source apportionment.
- Two distinct vehicle exhaust emission patterns are the dominant sources of UFPs.
- Background photochemical processes also contribute significantly in the warm period.

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ABSTRACT

A mobile laboratory unit (MOBILAB) with on-board instrumentation (Scanning Mobility Particle Sizer, SMPS; Ambient NOx analyzer) was used to measure size-resolved particle number concentrations (PNCs) of quasi-ultrafine particles (UFPs, 9–372 nm), along with NOx, in road microenvironments. On-road measurements were carried out in and around a large Greek urban agglomeration, the Thessaloniki Metropolitan Area (TMA). Two 2-week measurement campaigns were conducted during the warm period of 2011 and the cold period of 2012. During each sampling campaign, MOBILAB was driven through a 5-day inner-city route and a second 5-day external route covering in total a wide range of districts (urban, urban background, industrial and residential), and road types (major and minor urban roads, freeways, arterial and interurban roads). All routes were conducted during working days, in morning and in afternoon hours under real-world traffic conditions. Spatial classification of MOBILAB measurements involved the assignment of measurement points to location bins defined by the aspect ratio of adjacent urban street canyons (USCs). Source apportionment was further carried out, by applying Positive Matrix Factorization (PMF) to particle size distribution data. Apportioned PMF factors were interpreted, by employing a two-step methodology, which involved (a) statistical association of PMF factor contributions with 12 h air-mass back-trajectories ending at the TMA during MOBILAB measurements, and (b) Multiple Linear Regression (MLR) using PMF factor contributions as the dependent variables, while relative humidity, solar radiation flux, and vehicle speed were used as the independent variables. The applied data analysis showed that low-speed cruise and high-load engine operation modes are the two dominant sources of UFPs in most of the road microenvironments in the TMA, with significant contributions from background photochemical processes during the warm period, explaining the reversed seasonal variation of UFP concentrations, compared to those observed in cities across Northern Europe. It was also demonstrated that town planning exerts a profound effect on the mitigation of traffic emissions.

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List of abbreviations

ART	Arterial Roads
CEMENT	Cement Plant
CPF	Conditional Probability Field
F1	Factor 1: Acceleration-High Vehicle Speed
F2	Factor 2: Cruise
F3	Factor 3: Idle-Creep
F4	Factor 4: Background-Photochemical
FREE	Freeways
GDAS	Global Data Analysis System
GMD	Geometric Mean Diameter
GPS	Global Positioning System
H/W	The Ratio of Building Height (H) to Canyon Width (W)
HYSPLIT	Hybrid Single Particle Lagrangian Integrated Trajectory
I	Industrial Complexes
IB	Residential Areas between Industrial Complexes
INTR3	3rd Class Interurban Roads
MLR	Multiple Linear Regression
MOBILAB	Mobile Laboratory
NOAA	National Oceanic and Atmospheric Administration
RH	Relative Humidity
SR	Solar Radiation
PMF	Positive Matrix Factorization
OIL	Oil Refinery Plant
PNC(s)	Particle Number Concentration(s)
SMPS	Scanning Mobility Particle Sizer
TMA	Thessaloniki Metropolitan Area
U1	Urban Area-City Center
U2	Urban Area-Eastern Part
UB	Residential Area-Elevated Position
UI	Urban-Industrial Area
UFP(s)	Ultra-Fine Particle(s)
US EPA	United States Environmental Protection Agency
USC(s)	Urban Street Canyon(s)
V	Vehicle Speed

1. Introduction

Ultra-fine particles (UFPs), i.e. particles less than 100 nm in diameter, have been on the focus of air pollution research in recent years, due to increased health effect potential, deriving from their special interaction pattern in the human airways (Donaldson et al., 1998; Jaques and Kim, 2000). UFPs may constitute less than 10% of particulate matter (PM) mass in the atmosphere, but they overwhelm particles larger than 100 nm in number concentrations (Yao et al., 2005). UFPs can efficiently penetrate deep into the human respiratory system, where they cannot be effectively removed by macrophage clearance mechanisms, causing mechanical damage to lung tissue (Donaldson et al., 1998; Jaques and Kim, 2000). Moreover, they rapidly pass the blood circulation system and can be transported to the heart, brain and other organs (Nemmar et al., 2002). Also, when in contact with skin, there is evidence of UFP penetration to the dermis followed by translocation via lymph to regional lymph nodes (Oberdörster et al., 2005).

UFPs can be directly emitted from combustion processes or indirectly formed by gas-to-particle conversions in the atmosphere. There are several atmospheric processes leading to sub-micron aerosol formation even in remote areas, which have not been fully resolved yet. In the continental boundary layer, the most probable formation mechanisms seem to be the homogenous binary water-sulphuric acid nucleation, the homogenous ternary water-sulphuric acid-ammonia nucleation, and the ion-induced nucleation of inorganic or organic vapors (Kulmala and Kerminen, 2008; Yao et al., 2005). In coastal and marine environments, another possible mechanism could be the oxidation of iodine emitted by microalgae. Recent studies have shown that coastally formed UFPs also include a significant fraction of secondary organic products, apart from iodine oxides, especially in spring and summer. The origin of these secondary organic compounds has been attributed to the biosynthetic production of alkenes from marine flora and their subsequent oxidation, during efficient solar radiation (Eleftheriadis et al., 2006; Kalivitis et al., 2015; O'Dowd et al., 2004; Vaattovaara et al., 2006).

In urbanized regions, the major sources of UFPs are combustion processes related to road traffic, with smaller contributions from residential heating or aircraft take-off, landing and taxiing activity (Bukowiecki et al., 2003; Hsu et al., 2014; Kumar et al., 2014; Pey et al., 2009). UFPs can be formed by vehicles either primarily, as soot particles, during combustion in the engine, or secondarily, from the nucleation of gaseous precursors during dilution and cooling of the exhaust. In the area adjacent to emissions, these two distinct pathways of UFP formation are usually reflected by a bimodal size distribution, characterized by a peak in the nucleation mode (D_p < 20 nm) and a peak in the Aitken's mode (20 < D_p < 100 nm), or in the accumulation mode (100 < D_p < 1000 nm), being strongly dependent on engine type, fuel quality, vehicle speed, and power demand, as well as on the background aerosol and the prevailing meteorological conditions (Domínguez-Sáez et al., 2012; Huang et al., 2013; Li et al., 2013). So far, diesel vehicles were considered to be more important sources of UFP than gasoline vehicles, although remarkable reductions (ranging from 90% up to over 99%), have been observed in PM mass emissions from modern diesel vehicles, with advanced emission control technologies (Biswas et al., 2008; Bergmann et al., 2009).

Once formed, UFPs may show huge number concentrations near the area of emission, but they generally have a limited life-time (in the range of minutes), since they are rapidly transformed by coagulation, vapor adsorption and secondary particle formation. As a consequence, UFP number concentrations have a much larger spatial and temporal variability than fine particle mass concentrations in the urban scale (Bukowiecki et al., 2003; Westerdahl et al., 2005). Recent studies have also shown that various urban microenvironments in modern cities, such as the interior of vehicles, side-walks, or even entire regions inside urban street canyons (USCs), could pose an increased exposure risk to the people that live or work there, due to elevated concentrations of UFPs that are not reflected in stationary measurements (Diapouli et al., 2008; Habilomatis and Chaloulakou, 2013; Kaur et al., 2005; Zhou and Levy, 2008; Zwack et al., 2011).

The aim of the present study was to further existing knowledge on the concentration levels and size distributions of UFPs and identify major influential factors in road microenvironments. A mobile laboratory unit (MOBILAB) was employed for on-road measurements, in and around a large Greek urban agglomeration, the Thessaloniki Metropolitan Area (TMA). Size-resolved particle number concentrations (PNCs) of quasi-ultrafine particles (UFPs, 9–372 nm) were measured simultaneously with a regulated gaseous pollutant, NOx, which is typically related to road-traffic emissions.

Spatial classification of measurements involved the assignment