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Total exposure to airborne particulate matter in cities: The effect of biomass combustion



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HIGHLIGHTS

- Exposure to PM increases significantly in wintertime due to biomass combustion for space heating
- · Biomass burning significantly increases the levels of indoor PM of lower diameter
- Meteorological parameters explain about 80% of the ambient PM concentrations variability
- · Peaks of intra-day PM intake rate do not correspond to intra-day ambient air concentrations
- Exposure assessment is greatly facilitated by coupling in silico tools to in situ measurements

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ABSTRACT

The study deals with the seasonal variability of PM exposure and the effect that biomass combustion has upon it in the urban environment. The study is based on measurements, chemical analyses and modeling results performed in Thessaloniki (Greece). The measurements campaign included the assessment of outdoor and indoor air quality and the evaluation of biomass use for domestic heating. The outdoor measurements highlighted a significant increase of PM10 (from 30.1 to 73.1 µg/m³) and PM2.5 (from 19.4 to 62.7 µg/m³) concentrations during the transition from the warm to the cold period of the year 2012 compared to 2011. The increase in ambient air PM during the winter was attributed to the use of biomass burning for space heating. The latter was verified by the presence of levoglucosan in the PM (concentrations up to $8 \,\mu g/m^3$), especially for samples taken from the urban background site. Outdoor PM concentrations were also modeled using an artificial neural network model taking into account major meteorological parameters; the latter explained more than 90% of PM10 and PM2.5 day-to-day variability. Indoor concentrations followed a similar pattern, while in the case of fireplace use, average daily concentrations rise to $10 \,\mu g/m^3$ and $14 \,\mu g/m^3$ for PM2.5 and PM10 respectively. Indoor air concentrations were affected the most by the ambient air particle infiltration. Indoor air quality went down after 3 h of open fire biomass combustion for space heating. Personal exposure was significantly determined by overall indoor air quality. Yet, dynamic exposure analysis revealed that peaks of intake do not correspond to peaks of ambient air PM concentrations altering thus total exposure patterns. Thus, cost-effective public health protection has to aim at reducing the exposure profile of susceptible population sub-groups combining awareness raising, emission reduction measures and financial incentives to influence the choice of space heating systems.

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1. Introduction

Particulate matter (PM) is currently one of the most serious urban environmental pressures on public health. PM can be directly emitted from a variety of sources, both natural and anthropogenic, or generated by atmospheric reactions, leading to a complex mixture of solid particles and liquid droplets with different size and chemical composition (Galindo et al., 2011), met in both outdoor and indoor environments (He et al., 2004; Lai et al., 2006).

Macro and micro environmental concentrations of PM are affected by seasonality, reflecting both changes in the prevalent meteorological conditions as well as PM emission patterns, traffic emissions and domestic heating being dominant among them. In fact PM levels in cities tend to be higher in the winter with respect to summertime especially when intensive emissions from human activities (e.g. traffic and spatial heating)

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are coupled with adverse weather, e.g. prevalence of anticyclonic conditions that limit long-distance transport of polluted air masses (Viana et al., 2008). Meteorology is a significant determinant of PM concentrations in ambient air since dispersion processes, removal mechanisms, and chemical formation of atmospheric particles depend on common meteorological descriptors such as wind speed, rainfall rate, and solar radiation. Increased levels of ambient air PM are generally associated with colder air temperature, especially when in tandem with lowering of the atmospheric mixing layer and atmospheric stability (Katsouyanni et al., 2001).

However, changes in prevalent meteorological conditions accompanying seasonality changes are also correlated to changes in emission patterns, although the latter are difficult to be dissociated from the direct effects of meteorology. Kinney et al. (2002) identified that negative correlation between PM10 and temperature was attributed to increased traffic density and domestic heating during winter. In fact, increased PM concentrations due to biomass combustion for space heating are a problem that mars different European regions (Puxbaum et al., 2007). Over the last five years this phenomenon has spread beyond the Scandinavian and alpine regions of Europe where wood biomass is used regularly for domestic heating (Fuller et al., 2014). A combination of two distinct drivers may be behind the observed trend. Biomass as heating fuel has been heralded as a valid CO₂ reduction alternative (Wagner et al., 2010) across Europe. Furthermore, the financial crisis in southern Europe over the last two to three years has brought about a significant increase in the market price of alternative fuels primarily due to exorbitant taxation-the increase on the non-solid fuel price was ca. 40-60%; the ultra-high market price together with the limited financial capacity of many households has resulted in an excessive use of biomass for space heating.

Indoor air PM concentrations are affected by the penetration of outdoor air as well as the presence of indoor sources (Karakitsios et al., 2014). Smoking and biomass use are the dominant among them (He et al., 2004; Lai et al., 2006). Irregular pollution peaks attributable to vacuuming, candle, or incense burning may also be observed. However, they are likely to contribute only a little to 24-h mean PM levels (He et al., 2004; Semple et al., 2012). Biomass-using households have two to four times higher PM10 and PM2.5 concentrations as measured from Banerjee et al. (2012) by a real-time laser photometer.

Exposure to PM is the result of aggregation of several components. It mainly reflects the outdoor and indoor concentrations, as well as the interaction between ambient and indoor air and the time spent by the exposed individuals in specific macro- and micro-environments. Refined aggregate exposure assessment reduces the uncertainties incorporated in exposure assessment with regard to specific exposure scenarios such as the contribution of individual indoor emission sources related to either different consumer behavioral patterns (Sarigiannis et al., 2012a) or accidental events that may take place indoors (Sarigiannis et al., 2012b). Computational tools supporting aggregate exposure assessment enable us to forecast population exposure to PM. However, due to the complexity of the processes that control formation, transportation and removal of particulate matter in the atmosphere, modeling and prediction of aerosol concentrations are considered to be more difficult and complex than forecasting of common gaseous pollutants. Thus, statistical techniques based on existing time-series are expected to be more reliable forecasting tools than deterministic approaches (Karakitsios et al., 2006). Among the several statistical techniques, artificial neural networks (ANN) are expected to show better aerosol forecasting performance when compared to the traditional ones such as regression models (Paschalidou et al., 2011; Vlachogianni et al., 2011). This is because they adapt better to fitting data describing highly non-linear physico-chemical processes (Grivas and Chaloulakou, 2006; Kassomenos et al., 2011a) or to assimilating different environmental data classes (Sarigiannis et al., 2009).

The study presented herein aimed at assessing in detail the population exposure to PM during wintertime in a large Greek metropolitan area (Thessaloniki) and how this exposure is affected by the use of biomass for domestic heating (use of open fireplaces). Exposure assessment was based on the computation of the total exposure of population sub-groups taking into account both ambient and indoor air quality, as well as the time-activity patterns of the respective individuals to account for physiological changes in the intake rate. Measurements and model results were fused to complete data gaps and to derive total exposure estimates.

2. Methodology

2.1. Study design

The study assesses overall personal exposure and actual population intake of PMx, taking into account both outdoor and indoor exposure patterns, the multiple interactions between outdoor and indoor particulate matter and interseasonal variability. The results are geared to support integrated health risk assessment for the urban population. Our methodology couples an extensive network of ambient air measurements across the urban canopy for a period of 6 months (from October 2012 to April 2013) to targeted indoor air PMx measurements in dwellings and exposure modeling. Field measurements are supported by laboratory analyses of the PM chemical composition focusing on the quantification of chemical markers revealing the effect of biomass combustion to the PM mass concentrations. PMx levels in the ambient air provide the input needed for the INTERA computational platform (INTERA, 2011), for the calculation of indoor concentrations, exposure and intake. The INTERA platform is linked to a database that provides the necessary data for estimating indoor PMx concentrations (emissions from various sources, indoor/outdoor air exchange rates, infiltration and deposition rates), as well as data related to exposure (time activity patterns, types of locations encountered) and intake (inhalation rates based on activities, gender and age group). The detailed time dynamics incorporated in the INTERA platform allow capturing in detail intra-day exposure variability related to changes in outdoor concentrations, selection of activities, and change in the indoor emission sources. This is very important, since different types of complex interactions (e.g. simultaneous changes in indoor emissions and outdoor concentrations) are translated into actual terms of human exposure and intake, thus transforming qualitative hypotheses into quantitative estimates. In order to support exposure scenario analysis an exposure forecasting model was built based on an artificial neural network (ANN) that uses the ambient air PMx measurement data from previous days and meteorological information to predict next day PMx levels in different types of urban environments and for different days in the week. Coupling the ANN to the INTERA platform allows us to predict actual total population intake of PMx and eventually assess intervention scenarios and risk management measures.

2.2. Field measurements

With regard to the outdoor measurements, PM10 and PM2.5 samplers were installed at 3.5 m above ground, located at two traffic and two background sites. Several streets with typical city heavy-traffic levels surrounded the traffic sites; in contrast the background sites were not exposed to direct traffic emissions. The measurements lasted for 6 months, covering the transition from the warm to the cold period (average daily temperatures 23 and 4 °C respectively) and vice versa.

Parallel measurements of indoor air quality (for PM10 and PM2.5) were carried out in 30 houses close to the traffic and the urban background station respectively, so as to validate the INTERA platform estimates. In order to identify the contribution of biomass burning in indoor air quality, targeted measurements for the effect of the open fireplaces operation on PM concentrations were carried out in a selected indoor location and under controlled conditions of air exchange rate (AER). More specifically, within a third floor apartment of 140 m², extensive measurements of PMx were carried out, while a Particle Number Count sizer was used for measuring temporal variation of PMx number and mass concentration.

Meteorological data needed for the study were provided by the local meteorological station from the traffic site.

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