Atmospheric Environment 62 (2012) 82–88

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Environmental tobacco smoke aerosol in non-smoking households of patients with chronic respiratory diseases

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HIGHLIGHTS

- We analyzed indoor and outdoor PAHs and alkanes in the residencies of asthmatic and COPD patients.
- ETS tracers in non-smoking households were due to penetration of outdoor air.
- We identified the contribution of ETS, indoor activities and ventilation on indoor concentrations.

ARTICLE INFO

Article history:
Received 3 April 2012
Received in revised form
24 July 2012
Accepted 30 July 2012

Keywords:
Urban aerosol
Indoor aerosol
Environmental tobacco smoke
iso/*anteiso*-alkanes

ABSTRACT

Fine particulate matter samples were collected in an urban ambient fixed site and, outside and inside residencies in Athens greater area, Greece. *n*-Alkanes, *iso*/*anteiso*-alkanes and polycyclic aromatic hydrocarbons (PAHs) were identified by gas chromatography and mass spectrometry. The values of concentration diagnostic ratios indicated a mixture of vehicular emissions, fuel evaporation, oil residues and environmental tobacco smoke (ETS) in outdoor and indoor samples. Particulate *iso*/*anteiso*-alkanes, specific tracers of ETS, were detected in both non-smoking and smoking households. The indoor-to-outdoor ratios of particulate *iso*/*anteiso*-alkanes and unresolved complex mixture (a tracer of outdoor air pollution) in non-smoking households were comparable to the measured air exchange rate. This suggested that penetration of outdoor air was solely responsible for the detection of tobacco smoke particulate tracers in indoor non-smoking environments. Overall, residential outdoor concentrations accounted for a large fraction (from 25 up to 79%) of indoor aliphatic and polyaromatic hydrocarbons. Open windows/doors and the operation of an air condition unit yielded also in higher indoor concentrations than those measured outdoors.

1. Introduction

Associations of urban fine particulate matter ($PM_{2.5}$) with respiratory and cardiovascular adverse health effects leading to increased morbidity and mortality are computed using particle mass measurements in one or multiple ambient fixed sites (Katsouyanni et al., 2001; Peng et al., 2005). Personal exposures have shown weak cross-sectional correlations with ambient levels (Williams et al., 2003). This difference is partially attributed to indoor sources and sinks of particulate matter including ventilation, and personal activity patterns (Kousa et al., 2002).

$PM_{2.5}$ is composed of a mixture of chemical substances originated from natural and man-made sources. Carbonaceous (organic and elemental carbon) aerosol frequently accounted up to 50% of $PM_{2.5}$ mass (Putaud et al., 2004). Natural sources include the epicuticular waxes of terrestrial plants and secondary organic compounds formed through the oxidation of biogenic hydrocarbons (Kavouras et al., 1998b). Large quantities of organic compounds are released during the combustion of fossil fuels and fresh biomass, including fires (Rogge et al., 1993a, 1993b, 1993d; 1997a, 1997b). Cigarette smoke is recognized as a source of fine

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http://dx.doi.org/10.1016/j.atmosenv.2012.07.086

aerosol (Rogge et al., 1994; Kavouras et al., 1998a), particularly indoors due to smaller dilution volumes, poor ventilation and higher emissions (Singer et al., 2002).

Exposures to environmental tobacco smoke (ETS) are related to asthma, bronchitis, pneumonia and other respiratory disorders in both adults and children (Gilliland et al., 2001). ETS is emitted into the atmosphere from the smoke exhaled from the smokers (i.e. mainstream) and the smoke between puffs during tobacco products smoking (i.e. side-stream). The chemical fingerprint of organic aerosol from fossil fuel combustion is different from that of fresh biomass burning and of naturally emitted organics allowing for the reconciliation and apportionment of their sources (Schauer et al., 1996).

The RUPIOH (Relationship between Ultrafine and fine Particulate matter in Indoor and Outdoor air and respiratory Health) examined the exposures to outdoor and indoor particle mass (PM_{10}, $PM_{2.5}$), ultrafine particle number concentrations and their effects on the respiratory health of asthmatic and chronic obstructive pulmonary disease (COPD) patients in European cities. Ambient (in a fixed urban background site), residential outdoor and indoor PM_{10} mass, $PM_{2.5}$ mass, particle number concentrations and respiratory health (i.e. respiratory symptoms–restriction of activities, spirometry, exhaled breath condensate, Clara cells 16) of study participants were concurrently measured. A strong spatial concentration gradient was observed for coarse ($PM_{10-2.5}$) and ultrafine particles and to a lesser extend for $PM_{2.5}$ mass (Lianou et al., 2011). Traffic at the nearest street and the location of the residence (center/suburban) were the most important determinants of the observed spatial variation of outdoor particulate matter. Similar conclusions were drawn for indoor particle mass and number concentrations (Hoek et al., 2008). Strong correlations were observed between NO_3^- levels in the exhaled breath condensate of asthmatic/COPD patients and $PM_{10-2.5}$ mass concentrations (Manney et al., in press); while an increase of 10 μg m^{-3} of $PM_{10-2.5}$ mass concentrations was associated increases of 0.6–0.7% in shortness of breath, wheezing, cough and limitation in walking. However, no statistically significant correlations between lung function and particle levels were computed probably due to the high use of respiratory medication (de Hartog et al., 2010).

A subset of ambient and residential $PM_{2.5}$ samples collected in Athens, Greece was analyzed for specific organic compounds. The objectives of this effort were to characterize the aliphatic hydrocarbons and PAHs contents of urban, residential outdoor and indoor $PM_{2.5}$ aerosol and reconcile their sources using diagnostic ratios and molecular markers (Kavouras et al., 2001). The relationships of indoor concentrations with indoor activities and residential outdoor concentrations were also examined. This analysis improves the understanding on how indoor air levels are affected by outdoor air and typical activities in real living conditions including ETS.

2. Material and methods

2.1. Study area and sampling

$PM_{2.5}$ aerosol sampling was conducted in an urban fixed site and thirty-five residencies in the Athens greater area for eighteen months (from October 2002 to April 2004) (Puustinnen et al., 2007; Hoek et al., 2008). 24-h aerosol samples (from noon to noon) were collected continuously at the fixed site and for a period of seven days sequentially in each residence (one home monitored per week). The $PM_{2.5}$ Harvard Impactor was used to collect fine particles on a 37-mm Teflon filters. Fig. 1 shows the fixed site, the residencies, the major roads and population density (inhabitants km^{-2}) in Athens area. The fixed site was at Goudi air pollution monitoring site (37°59′04″, 23°46′04″), which is located centrally to the metropolitan area. Residential sampling was done

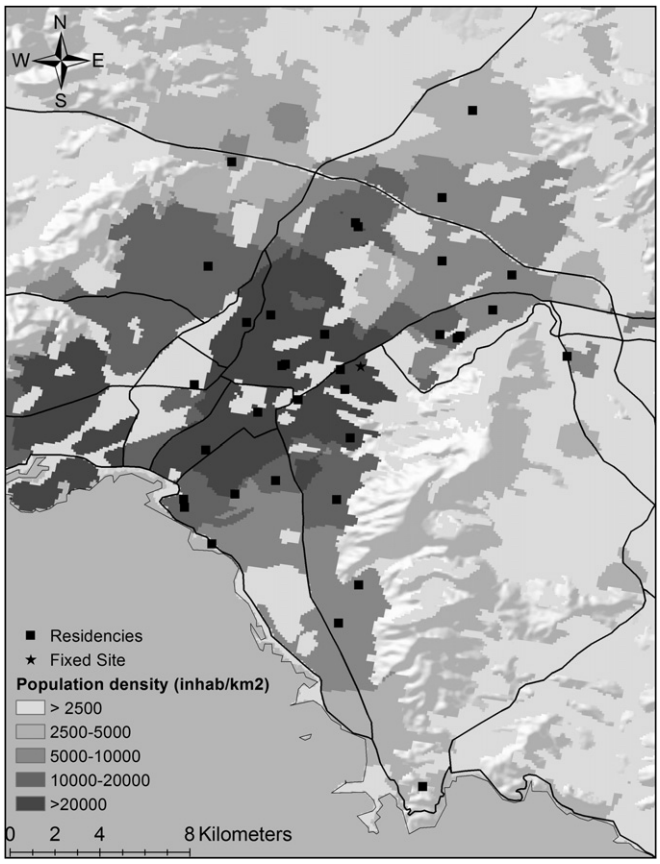

Fig. 1. Locations of the ambient fixed site and 35 residencies, major arterial roads and population density in Athens, Greece.

in and outside the living room, 1.8 m above the floor and at least 0.5 m from vertical surfaces. Particle mass was determined by gravimetric analysis. Information on traffic, residential features and indoor activities was obtained using questionnaires. In addition, the air exchange rate was measured in each residence.

The selected residences extended through a large fraction of the urban area (and within 20 km of the fixed site) with different population densities and traffic characteristics and typical of the urban plan of Athens (Fig. 1). Briefly, there were mostly apartment buildings (33 of 35) built after 1960 and adjacent (>10 m) to streets carrying on average 5000 vehicles day^{-1}. About half of them were within 100 m of a major street carrying, on average, 43,000 vehicles day^{-1}. For all residencies, the kitchen was adjacent to the living room (with 12 of them having an open floor plan). In all cases, there was at least one outside door in the living room leading to a balcony and an outside window (or a door) in the kitchen. Air condition units were installed in twelve residencies. Cooking was done almost exclusively in electrical stoves and ovens with an overhead fume hood usually on. Because of the underlying health conditions of the study participants, the initial study design excluded smoking households to eliminate potential confounding from smoking. However, a limited number of smoking households was included in the study to achieve a sufficient number of subjects for the epidemiological analysis (de Hartog et al., 2010).

2.2. Chemical analysis

A total of 96 ambient samples and 192 paired residential outdoor and indoor samples (at least three paired samples from 32