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Short Communication

Indoor and outdoor suspended particulate matter and associated carbonaceous species at residential homes in northwestern Portugal

Danilo Custódio, Isabel Pinho, Mário Cerqueira *, Teresa Nunes, Casimiro Pio

Department of Environment & Centre for Environmental and Marine Studies (CESAM), University of Aveiro, 3810-193 Aveiro, Portugal

HIGHLIGHTS

- Indoor/outdoor PM_{10}, OC and EC concentrations were examined in Portuguese residences.
- Indoor PM_{10} levels were higher than those reported previously in central and northern Europe.
- Outdoor dust was the most likely source of the PM_{10} excess in Portuguese residences.
- OC and EC were the major and minor contributors, respectively, to indoor PM_{10}.
- Smoking, cooking, biomass burning and movement of people were the likely sources of indoor OC.

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ABSTRACT

Particulate matter with an aerodynamic diameter equal to or less than 10 μm (PM_{10}), organic carbon (OC) and elemental carbon (EC) concentrations were measured simultaneously in the indoor and outdoor air of 4 residences located in urban and sub-urban areas in northwestern Portugal. The residences were studied with occupants. One residence was also studied without any indoor activity, taking advantage of the fact that the occupants had moved into a new home. First, 48-h aerosol samples were collected on quartz fiber filters with low-volume samplers equipped with size selective inlets. The filters were weighed and then analyzed for OC and EC using a thermal–optical transmittance method. The average indoor and outdoor PM_{10} concentrations in the occupied residences were 71.9 ± 38.3 μg/m^3 and 54.0 ± 13.3 μg/m^3, respectively. Despite the higher concentration of PM_{10} indoors, outdoor sources were found to be a significant contributor to indoor concentrations. An estimate based on data from the residence studied under different occupancy conditions indicated that outdoor sources can account for 68% of the indoor PM_{10} mass concentration. Average indoor to outdoor (I/O) ratios for OC ranged from 1.7 to 5.6 in occupied residences, showing that indoor sources, such as cooking, smoking, biomass burning and movement of people, strongly influenced indoor OC concentrations. In contrast, I/O ratios for EC were close to 1, except for a smokers' residence, suggesting that indoor concentrations were mainly controlled by outdoor sources, most likely from vehicular emissions and biomass burning.

1. Introduction

Exposure to particulate matter in ambient air has been linked to diverse health effects, particularly within the respiratory and cardiovascular systems (Brunekreef and Holgate, 2002; Pope et al., 2002). As people in industrialized countries spend 80 to 90% of their time indoors and most of that time is spent at home, personal exposure to particulate matter mainly occurs in the indoor residential environment. Particles in the indoor air of residences originate from various sources. Common indoor sources include cooking, smoking, cleaning, biomass burning in fireplaces and stoves, handling of fiber containing materials and people's movements (Abt et al., 2000a; Cerqueira et al., 2010; Nasir and Colbeck, 2013; Polidori et al., 2007; Ward et al., 2008). Another important source of indoor particulate matter is air transported from the outside by infiltration and ventilation processes (Abt et al., 2000b; Riley et al., 2002).

Many studies have been conducted in Europe to assess human exposure to residential aerosol particles and to describe their sources, physical properties and chemical composition (e.g. Fischer et al., 2000; Jones et al., 2000; Monn et al., 1997; Stranger et al., 2007). However, as these studies focused on residences from central and northern Europe, little or no information is available for residences located in the southern part of the continent. This lack of information is of particular concern given the current knowledge about the aerosol spatial distribution in Europe. Exceedances of the EU daily limit for PM_{10} are common in southern European countries (Mitsakou et al., 2008; Moreno et al., 2005; Pederzoli et al., 2010; Querol et al., 2008), due to local dust resuspension and mineral dust intrusions from African desert regions, thus suggesting a significant contribution of outdoor sources to residential indoor PM_{10}

* Corresponding author. Tel.: +351 234 378134; fax: +351 234 370309.
E-mail address: cerqueira@ua.pt (M. Cerqueira).

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levels. This contribution can be even more important in the warm climates of southern Europe because windows are frequently open to provide natural ventilation.

This paper reports the first quantitative description of particulate matter (PM_{10}) concentrations and associated carbonaceous fractions (OC and EC) in the indoor air of residences in Portugal. The aims of this study were to: (1) assess exposure to particulate matter in urban and sub-urban residences in Portugal; (2) investigate the relationships between indoor and outdoor concentrations of PM_{10}, OC and EC concentrations; and (3) describe relevant sources of particulate matter in the indoor air of investigated residences.

2. Experimental

2.1. Sampling sites

The study was conducted in four homes located in the cities of Aveiro (population = 45 000) and São João da Madeira (population = 21 000), both located in northwestern Portugal. The homes were selected to reflect different indoor sources (cooking, smoking, presence of pets, fireplace utilization, etc.) and outdoor environments (urban density, traffic intensity, etc.). Three homes were located in blocks of flats, and one home was a terraced house. All of the homes were studied with occupants. One of them was also studied without any indoor activity, taking advantage of the fact that the occupants had moved into a new home. Doors and windows provided natural ventilation in all cases. These doors and windows were normally closed and were opened only for short time periods (a few hours per day) according to the occupants' requirements. The exception was home 2, with a door linking the kitchen to a roof terrace that was frequently opened. A summary of the main characteristics of the sampling sites used in this study is presented in Table 1.

Indoor sampling was conducted in the kitchen because of the known diverse sources of airborne particles and because occupants spend much of their time inside this room. In many Portuguese homes, the kitchen is a communal area that is used not only for cooking but also for eating, relaxing and socializing. Outdoor sampling was conducted at the same height above the ground as indoor sampling by means of a balcony (a terrace was used in home 2) at the front of the building.

2.2. Sampling method

Indoor and outdoor PM_{10} measurements were performed simultaneously with two MiniVol™ TAS samplers from AirMetrics (Eugene, Oregon, USA). The MiniVol is a low-volume portable sampler equipped with an air pump, a rotameter, a flow rate controller and a programmable timer. In operation, air is drawn through a size selective inlet and then through a filter pack fitted to the top of the sampler to selectively collect aerosol particles in the PM_{10} size range. The size-selective inlets were located approximately 160 cm above the floor to obtain samples within the occupants' breathing air zone. Aerosol samples were taken at a flow rate of 5 L min^{-1} for 48 h. The filters used for the concentration of particles were made of quartz fiber (Whatman QMA, 47 mm diameter). These filters were previously combusted at 550 °C for 4 h to remove organic contaminants. Before the start of the study in the residences, duplicate sampling runs were performed to compare the response of the low-volume aerosol samplers. A total of 9 pairs of samples were gathered (5 indoors and 4 outdoors), and the mean ratio found for PM_{10} was 1.00 ± 0.05, showing insignificant mass concentration bias between the samplers.

2.3. Analysis

The mass concentration of particulate matter was determined by gravimetry. Before sampling, the filters were conditioned for at least 24 h in a room with constant humidity (50%) and then weighed on an electronic microbalance with a sensitivity of 0.01 μg. After collection, the filters were re-conditioned, re-weighed and stored at −20 °C until they could be chemically analyzed.

The EC and OC particulate fractions accumulated in the filters were measured by the thermal optical method previously described by Castro et al. (1999). The system comprises a quartz tube with two heating zones, a laser and a nondispersive infrared (NDIR) CO_2 analyzer. A punch of the filter sample is placed vertically inside the quartz tube within the first heating zone, which is then heated to 600 °C in a nitrogen atmosphere to vaporize the organic fraction of particles. EC is determined by sequential heating at 850 °C in a nitrogen/air atmosphere. The second heating zone is filled with cupric oxide and maintained at 850 °C during the entire analysis process to guarantee the total oxidation of the volatilized carbon to CO_2, which is quantified continuously by the NDIR analyzer. Correction for the pyrolysis contribution to EC from OC is achieved by monitoring the transmission of light through the filter with the laser beam. The OC/EC split is set when the transmittance returns to the value measured at the beginning of the analysis. Pyrolytic carbon (PC) is calculated from the mass of CO_2 emitted during the second heating phase, under the gas flow containing O_2, until the recovery of filter light transmittance.

3. Results and discussion

3.1. Mass concentrations

A total of 7 pairs of filter samples were collected per residence and analyzed for mass and carbon fractions. A summary of PM_{10} measurements is shown in Table 2. Exposure to particulate matter in occupied residences was higher indoors than outdoors (an average of approximately 30%). The highest indoor concentrations (average of 116.4 ± 57.2 μg/m^3) were found in home 3. The presence of 3 smokers and poor ventilation conditions seems to explain these results. Significant differences in PM_{10} concentrations between residences of non-smokers and smokers have also been described previously (e.g. Breysse et al., 2005; Chao and Wong, 2002; Stranger et al., 2007).

The lowest concentrations, on the other hand, were found in home 1B (average of 35.7 μg/m^3), a result of the absence of indoor activities. Data from this residence show an average PM_{10} decrease of 32% from the occupied to the unoccupied condition. This outcome is comparable to the 50% decrease in an unoccupied living room in a UK suburban location reported by Nasir and Colbeck (2013). Average concentrations from home 1 measured during periods without indoor activity (condition B) can be used as an indicator of the contribution of outdoor sources

Table 1
Summary details of sampling sites used in this study.

Site	Study period	Location	Site condition	Home type	Floor	Occupants	Smokers	Pets	Cooker type	Fireplace use	Ventilation
Home 1A	Feb.–Mar. 2011	Aveiro/city center	Near road with medium traffic flow	Apartment	4	5	No	No	Gas	No	Natural
Home 1B	Jan.–Feb. 2012	Aveiro/city center	Near road with medium traffic flow	Apartment	4	0	No	No	Not used	No	Natural
Home 2	Apr. 2011	Aveiro/suburb	Near road with medium traffic flow	Apartment	6	3–5	No	Dog	Gas	No	Natural
Home 3	May 2011	Aveiro/city center	Near road with high traffic flow	Apartment	4	4	3	Cat	Gas	No	Natural
Home 4	Mar. 2011	São João da Madeira/suburb	Far from busy roads	Terraced house	–	3–4	No	Dog & cat	Gas	Wood burning	Natural