

Traffic-related PM_{2.5} aerosol in residential houses located near major highways: Indoor versus outdoor concentrations

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ABSTRACT

PM_{2.5} mass concentration and its elemental composition were measured inside and outside of six houses located at a distance of approximately 30–300 m from major highways. To investigate the amount of traffic particles penetrating from indoor to the outdoor (I/O) environment, the data were analyzed by three-way factor method PARAFAC. The PM_{2.5} I/O ratio ranged from 0.5 ± 0.2 to 2.9 ± 1.2 in spring and from 0.7 ± 0.1 to 4.7 ± 6.9 in fall. The traffic component was very pronounced in the ambient air of the six houses. Although the distance from the highway and traffic intensity are generally important for assessing the indoor concentration of traffic-related aerosols, the data collected in this study suggest that – specifically for houses located in a close proximity to major highways – these two factors may not necessarily play the most important role in differentiating exposure levels between houses.

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1. Introduction

Airborne particulate matter (PM) has been associated with various adverse health effects (Schwartz et al., 1996; Brunekreef et al., 1997; Duhme et al., 1998). The fine fraction of the ambient and indoor aerosol, defined as PM with the aerodynamic diameter less than 2.5 μm (PM_{2.5}) is generated by various sources. Fuel combustion processes in transportation and energy production are the primary sources of the outdoor PM_{2.5}, while cooking, smoking, and cleaning activities contribute primarily to the indoor PM_{2.5} levels. Traffic-related fine particles are recognized as an important contributor to outdoor PM_{2.5} concentrations (Gertler et al., 2000).

The US population spends a majority of their time indoors (Klepeis et al., 2001). Recently, there has been an increasing interest towards the estimation of the

contribution of outdoor particles to the indoor particle concentration levels (Abt et al., 2000; Morawska et al., 2001; Vette et al., 2001; Riley et al., 2002; Franck et al., 2003; Cyrys et al., 2004; Maston, 2005; Meng et al., 2005; Hoek et al., 2008). The concentrations of traffic-related particulates and their elemental composition have been reported by several authors (Funasaka et al., 2000; Fischer et al., 2000; Kingham et al., 2000; Jones et al., 2000; Hänning et al. (2004); Ho et al. (2004); Martuzevicius et al., 2004; Hu et al., 2006). Many researchers have addressed the phenomena of particle penetration through the building structure into the indoor environments (see, e.g., Wallace, 1996). Some investigators determined penetration factors for specific particle size fractions but did not aim at obtaining information about chemical/elemental composition of the collected aerosol. For instance, Zhu et al. (2005) deployed real-time aerosol particle counters for measuring ultrafine particles indoors and outdoors of four apartments located within 60 m from the centerline of the 405 Freeway in Los Angeles, USA.

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Most of the available studies have relied on the particle transport modeling, determining or estimating parameters such as air exchange rate, particle deposition and re-suspension rate (Schneider et al., 2004). The transport model examines particle penetration and transformation as a function of time through a building structure. In addition to the highly time-resolved particulate number or mass monitoring, the air exchange rate and particle deposition rate may be important variables for consideration.

The utilization of a dynamic particle penetration model for the time-integrated (12, 24 h and longer) particulate mass measurements requires several approximations. Since such studies aim at assessing a long-term exposure of residents to the particulate matter and its elemental constituents, measurement campaigns usually span over several seasons. In such cases, ventilation conditions are difficult to control (frequent openings of windows, automatic HVAC system regulation), thus suggesting a continuous monitoring or the approximation of the air exchange rate in the residences. The air exchange measurement procedure requires injection of an inert trace gas into the building ventilation system (ASTM, 2001). Such procedures are often not feasible (or difficult to implement), particularly over extended time periods and in occupied homes. As a tool for evaluating the time-integrated pollution sources in indoor and outdoor air, receptor modeling techniques can be used. Kopperud et al. (2004) have utilized a Chemical Mass Balance (CMB) technique to estimate the contribution of outdoor sources and indoor re-suspension activities to indoor particulate matter. The CMB receptor model consists of a solution to linear equations that express each receptor chemical concentration as a linear sum of products of source profile abundances and source contributions (US EPA, 2001). The authors also compared the CMB to the air exchange-based mass balance model, and found good agreement in the contribution of outdoor and indoor sources to the indoor PM levels. The application of the CMB model is somewhat limited in cases of a large variance of the indoor sources, and when there is a lack of reliable source signatures. Factor analysis methods, such as principal component analysis (PCA), however, have been used by Biswas and coauthors (Biswas, et al., 1989; Tian et al., 1989, 1990) to unravel particle sources of Class 10 to Class 1000 clean room environments. Conventional two-way receptor models, such as Unmix (Henry, 2000) and PMF (Paatero, 1997), were not appropriate in this study due to limited number of samples collected at each site (Table 1) and because the indoor source categories varied from house to house. The latter made it impossible to run those models on the mixture of all indoor or outdoor samples.

Alternatively, the parallel factor analysis (PARAFAC) (Harshman and Lundy, 1984), a generation of PCA model, was introduced to solve two or more data arrays simultaneously. The advantage of PARAFAC is the uniqueness of the solution, i.e. less rotation ambiguity. The paired indoor and outdoor PM levels can form a three-way array assuming that these are attributed to similar sources, and only the strength of those sources varies between indoor and outdoor measurements. The PARAFAC models have been utilized to apportion indoor, outdoor and personal

PM_{2.5} (Yakovleva et al., 1999; Hopke et al., 2003; Larson et al., 2004) with the non-negative constraints.

The above-mentioned studies focused on penetration of outdoor particulate matter and aimed at apportioning various sources of indoor and outdoor aerosols. Few investigations, however, were specifically designed for the evaluation of traffic component infiltration to buildings. The purpose of this study was to determine the contribution of traffic-related particulate matter to the indoor aerosols by using receptor modeling based on a PARAFAC model.

2. Experimental methods

2.1. Sampling houses

The characteristics of the six houses used in the study are outlined in Table 1. These were selected from the cohort of the Cincinnati Childhood Allergy and Air Pollution Study (CCAAPS), which has recruited about 800 households in the Greater Cincinnati and Northern Kentucky area (Ryan et al., 2005). The following factors were reviewed when selecting houses for this study: the distance of a house to a highway (30–300 m), age (older than 10 years), window type and material (conventional), ventilation system (central HVAC), presence of indoor sources (allowed, including cooking and smoking), and the willingness of the residents to participate in an extensive air sampling (refusal rate was about 40%). Among six selected units, five (1, 2, 3, 5, and 6) were single-family homes located in residential neighborhoods of the Greater Cincinnati area. House 4 – originally built as a large single-family home and presently used as a small-scale patient clinic – was located in the Cincinnati downtown area. This study was concerned specifically with houses located in a close proximity to major highways. The CCAAPS study design identifying “exposed” families as those living in close proximity to highways utilized 400 m as the limit and defined a major highway as a road with a heavy-duty diesel vehicle traffic of >1000 vehicles per day. House 3 was positioned the closest to an interstate highway (~30 m), with a sound barrier separating it from the traffic lanes. House 1 was the farthest with the distance of approximately 300 m.

The age of the selected houses ranged from 24 years (House 3) to approx. 115 years (House 4). The residents performed their normal activities during the entire study period. The most prevalent indoor source was cooking. Most of the residents cooked at least once per day. Other significant indoor aerosol sources included smoking (House 1) and dusting by furniture spray (House 2). The residents were asked to keep an indoor event diary, marking activities such as ordinary cooking, cigarette smoking, window openings, dusting, and vacuuming.

2.2. Sampling and analysis of PM_{2.5}

There were two aerosol sampling campaigns performed inside and outside of each of the six selected houses during five days starting Monday and ending Saturday. The spring sampling campaign was conducted from March 30 through May 14, 2004 and the fall campaign was from September 13 through October 22, 2004. Each day the 24-h sampling

Table 1

Main characteristics of houses used in the study

House number	Year built	Total area, m ²	No. of floors	Ventilation; heating	Prevalent sources of indoor pollution	Distance from highway, m	Average traffic vehicles/day	No. of samples taken
1	1949	71	1 + Basement	HVAC, frequent window opening, gas forced air heating	Smoking (15 cigarettes/day, gas cooking (1–2/day)	300	48,870	10
2	1945	~150	2 + Basement	HVAC, frequent window opening, gas forced air heating	Cooking (2/day), furniture cleaner spray (one day)	180	93,210	10
3	1981	190	2 + Basement	HVAC, rare window opening, gas forced air heating	Cooking (1/day)	30	103,620	10
4	1890	580	2 + Basement	HVAC, no window openings, no heating	No	200	148,590	10
5	1972	150	2 + Basement	HVAC, frequent window opening, gas forced air heating	Cooking (1–2/day)	160	119,930	10
6	1957	190	2 + Basement	HVAC, frequent window opening, gas furnace heating	Cooking (1/day), heating, renovating, latex painting	180	48,870	10

periods started between 8 am and 12 pm, depending on the inhabitants' activities. Four Harvard-type PM_{2.5} impactors (MS&T Area Sampler, Air Diagnostics and Engineering, Inc., Harrison, ME, USA) were used for the aerosol sampling. The impactors were configured with a 2.5 μm cut-off size nozzles and operated at an air flow rate of 20 Lpm, which was calibrated with a flow meter (DryCal DC-Lite, BIOS International Corporation, Butler, NJ, USA) before each 24-h measurement period. The outdoor sampling setup consisted of two impactors mounted on a tripod under rain-protective covers, as well as a meteorological station (Vantage Pro, Davis, CA, USA) mounted on the same tripod. The indoor setup included another pair of impactors positioned on the top of a noise-insulated enclosure containing sampling pumps. The samplers were placed near a wall in the primary activity room, and the sampler's inlet was at ~1 m above the floor. The PM_{2.5} aerosols were collected on two types of filters. For gravimetric and elemental analysis, two samplers (one indoors and one outdoors) operated with 37-mm Teflon membrane filters (pore size = 1 μm) with a support ring (Pall Corporation, Ann Arbor, MI, USA). For carbon analysis, 37-mm quartz filters (Whatman Inc., Clifton, NJ, USA) were utilized. The polyester "Drain disk" pads (Whatman Inc., Clifton, NJ, USA) were used with the filters to maintain integrity. Teflon filter preparation before and after the sampling included conditioning in a humidity chamber at a relative humidity of 30–40% with a temperature of 22–24 °C for at least 24 h before weighing and labeling (McDonald, 2003). Quartz filters were pre-baked in a temperature of 550 °C for at least 24 h before sampling (Schauer and Cass, 2000; Bae et al., 2004).

Once the PM_{2.5} mass concentration was determined, Teflon filters were sent for X-Ray Fluorescence (XRF) analysis (Chester Labnet, Tigard, OR, USA) for a total of 38 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Hg, Pb). Quartz filters were analyzed for elemental and organic carbon (EC and OC) with the Thermal–Optical Transmittance (TOT) technique using NIOSH-5040 method (Sunset Laboratory Inc., Hillsborough, NC, USA).

2.3. Data analysis and receptor modeling

The descriptive statistical parameters, such as arithmetic mean and standard deviation were used to present

the data. Non-parametric statistical analyses (Spearman correlation coefficient, Wilcoxon Mann–Whitney test, Kruskal–Wallis test) were utilized to examine the significance of variation of the PM concentrations among houses, indoors versus outdoors, and spring versus fall seasons, as well as to determine the relationship between certain elements of the PM_{2.5} composition.

Three-way factor method has been utilized as a tool to retrieve the source contribution estimates (SCE) simultaneously to different types of the data, such as indoor and outdoor samples (Hopke et al., 2003; Yakovleva et al., 1999; Paatero, 1999). The general equation of the three-way factor method can be expressed as:

$$x_{ijk} = \sum_{r=1}^p a_{ir} b_{jr} c_{kr} + \varepsilon_{ijk} \quad (1)$$

where a_{ir} is the r th source contribution (in our case, r th source contribution for each i th sampling day), b_{jr} is the concentration of j th species in the r th source profile, c_{kr} is the weight of source r on k th sample type (indoor or outdoor), ε_{ijk} is the residual term.

The loss function of Eq. (1) can be expressed as follows:

$$\min_{A,B,C} Q(X, \sigma, A, B, C) \quad (2)$$

where

$$Q = \left\| \frac{(X - ABC)}{S} \right\|_F^2 = \sum_i \sum_j \left(\frac{e_{ijk}}{s_{ijk}} \right)^2 \quad (3)$$

with

$$e_{ijk} = x_{ijk} - \sum_{h=1}^p a_{ir} b_{jh} c_{kh} \quad (4)$$

and $a_{ir} \geq 0$, $b_{jr} \geq 0$, $c_{kr} \geq 0$ for $r = 1, \dots, p$, and s_{ijk} is the uncertainty for the individual data point x_{ijk} .

Several algorithms have been developed to solve Eq. (1), such as alternating least squares (ALS) (Bro, 1997), Gauss–Newton curve fitting and Conjugate Gradient curve fitting (Paatero, 1999). Determining the number of sources is critical for the multivariate receptor model (Henry et al., 2000; Hopke et al., 2003; Paatero, 1999), however, there are still no well-developed methods for solving the three-way factor analysis (Harshman and Lundy, 1984; Yakovleva

et al., 1999). In our study, N-way tools developed by Andersson and Bro (2000) were utilized to solve Eq. (1) with non-negative constraints on all the three factor modes. The fitting number of sources was determined by both split-half experiments (Harshman and Lundy, 1984) and core consistency diagnostic (Bro and Kiers, 2003). Then, the determined sources were further checked with prior knowledge of source types in Cincinnati region (Hu et al., 2006) and indoor sources during the sampling.

3. Results and discussion

3.1. Seasonal variation of indoor and outdoor concentrations

Out of 40 analyzed components of the PM_{2.5}, several of those (EC, OC, Si, S, Mn, Fe, Zn, Br, and Pb) were selected for data analysis purposes as the tracers of traffic-related emissions such as from diesel engine exhausts (HEI, 2002; Hu et al., 2006). Some of these elements, such as EC, Mn, Zn, Br, and Pb, directly represent traffic-related particulate matter (Gertler et al., 2000). Others, such as OC, Si and S, have multiple sources, including both gasoline and diesel engine exhausts. In indoor environments, a major portion of the OC is generated by indoor activities (primarily, cooking and cleaning) and some from infiltration of aerosols generated by outside sources (combustion sources including gasoline and diesel-powered engines). The OC present in the form of volatile and semi-volatile organic compounds may be converted to particles by physical and chemical means. Sulfur particles in the form of sulfate (usually as ammonium sulfate) aerosol in the ambient air, has been proved to be a good marker for outdoor particles found in indoor air (Sarnat et al., 2002). Si represents re-suspended dust material indoors and road dust or soil dust outdoors (Eldred et al., 1987).

The average PM_{2.5} concentrations and its compositions measured indoors and outdoors during five-day periods in two seasons are presented in Fig. 1. The outdoor PM_{2.5} concentrations ranged from $7.3 \pm 0.7 \mu\text{g m}^{-3}$ (House 1) to $23.2 \pm 9.5 \mu\text{g m}^{-3}$ (House 6) in spring and from $14.1 \pm 4.4 \mu\text{g m}^{-3}$ (House 3) to $22.1 \pm 11.8 \mu\text{g m}^{-3}$ (House 5) in fall. The indoor PM_{2.5} concentrations ranged from $9.1 \pm 3.3 \mu\text{g m}^{-3}$ (House 4) to $29.0 \pm 16.6 \mu\text{g m}^{-3}$ (House 2) in spring and from $10.4 \pm 2.6 \mu\text{g m}^{-3}$ (House 3) to $30.8 \pm 5.1 \mu\text{g m}^{-3}$ (House 1) in fall. These values, as well as the elemental concentration data, fall within the range previously reported for the Greater Cincinnati area (Martuzevicius et al., 2004). The PM_{2.5} concentrations measured inside and outside of houses in the vicinity of traffic sources are comparable to those found by other investigators. Kingham et al. (2000) reported mean values of 18.9 and $17.8 \mu\text{g m}^{-3}$, respectively, as measured in Huddersfield, UK. Fischer et al. (2000) reported mean values of 25.0 and $27.0 \mu\text{g m}^{-3}$ measured in Amsterdam, The Netherlands in high traffic area homes. From their study performed in Baltimore, MD, USA, Landis et al. (2001) reported mean values of 6.7 and $18.9 \mu\text{g m}^{-3}$.

There was insignificant seasonal variation for the outdoor PM_{2.5} concentration ($p > 0.05$). Slightly higher concentrations measured in the fall, especially for House 1, can be attributed to the ambient meteorological

conditions. During the first week in spring (end of March), the average ambient temperature was 7.4°C , with freezing temperatures occurring at night time, which prevented the temperature-dependent secondary sulfate aerosol formation. While EC and OC concentration levels were rather similar during both seasons, the concentrations of Si and S were significantly different for two seasons. The outdoor EC/TC ratio (TC = total carbon, calculated as EC + OC) depended on the distance from a highway and was the highest near Houses 4 and 3 (0.18 ± 0.03 and 0.17 ± 0.03 in spring and 0.17 ± 0.02 and 0.16 ± 0.07 in fall). The traffic-related trace metals showed high variability during both seasons, however, the concentrations were not statistically different. There were several days in the fall season when elevated Zn and Br concentrations were detected outdoors of Houses 2 and 6 (for unknown reasons). Three days of elevated Pb concentration resulted in high standard deviation for the data collected at House 2 in spring. Such high concentrations have been identified as outliers in a detailed outdoor factor analysis study (Hu et al., 2006).

The indoor PM_{2.5} concentration was mostly overruled by the variation indoor OC concentration caused by indoor events such as smoking, cleaning, and cooking, and thus does not indicate seasonal variation *per se*. The indoor concentrations of outdoor source-related elements, such as EC, S, Mn, Fe, Zn, and Br demonstrate lower values and lower variation compared to the outdoor concentrations, and insignificant differences between the two seasons.

Table 2 presents the five-day averaged indoor/outdoor (I/O) ratio of PM_{2.5} and its elemental concentrations. The values are presented for each of the six houses during spring and fall seasons. The PM_{2.5} I/O ratio ranged from 0.5 ± 0.2 (House 6) to 2.9 ± 1.2 (House 1) in spring and from 0.7 ± 0.1 (House 3) to 4.7 ± 6.9 (House 2) in fall. Both Houses 1 and 2 had significant indoor aerosol sources (smoking and cleaner spray, respectively). However, high I/O ratio in House 1 was observed during each day of sampling, which is indicated by low standard deviation, while House 2 had one day of cleaning in the five-day measurement campaign during both seasons (high standard deviation). The I/O ratio for PM_{2.5} was lower at Houses 3–6 during spring compared to fall. The OC I/O ratios exhibited the same pattern as that of PM_{2.5}. As expected, the averaged I/O ratio was higher than one in the houses with strong indoor activities (cooking, smoking, and cleaning). This result is somewhat surprising for House 4 (I/O = 1.4 ± 0.9 for spring and 1.2 ± 0.1 for fall) since the sampler was kept in the upper floor, where no human activities were taking place, and no forced ventilation (heating or air conditioning) occurred during the sampling period. The possible explanations include the accumulation of OC inside the house (due to presence of chemical materials, since the premises were utilized as a clinic), transportation from the lower floor, or the formation of OC particulate matter from gaseous compounds. The I/O for EC displayed controversial behavior from house to house. The highest value was measured for House 1 (I/O = 2.5 ± 1.4 for spring and 1.3 ± 0.7 for fall), which can be attributed to the influence of cigarette-emitted particles.

Due to a “tight” building structure (characterized by age, ventilation system, window type and material) the EC I/O values were below 1 in Houses 2 and 3. We were intrigued to observe rather high I/O values for Houses 4 and 6 during the spring season. As mentioned before, there were no indoor activities generating EC in both houses, and low I/O values for S confirm that the high EC I/O values did not result from penetration of the outdoor particles. The averaged S I/O values were lower than 1 for all houses and sampling seasons. During the fall season, Houses 4–6 had the I/O close to 1. While the questionnaires filled by the residents do not unequivocally confirm this, we believe

that a small difference in indoor versus outdoor concentrations should be attributed to frequent window openings, at least in Houses 5 and 6. House 4, located in the most polluted area of the city, showed consistent values of I/O close to 1, indicating relatively “loose” structure of the building envelope due to its age. The I/O for the trace metals was below 1 for almost all cases, except House 1 in spring and House 6 in fall. High levels of Si, Fe, Br and Pb in House 1 most likely indicate high content of fine household dust and cigarette smoke particles. High levels of Si, Mn, Fe, Zn and Pb in House 6 found during fall measurements indicate intensive re-suspension of

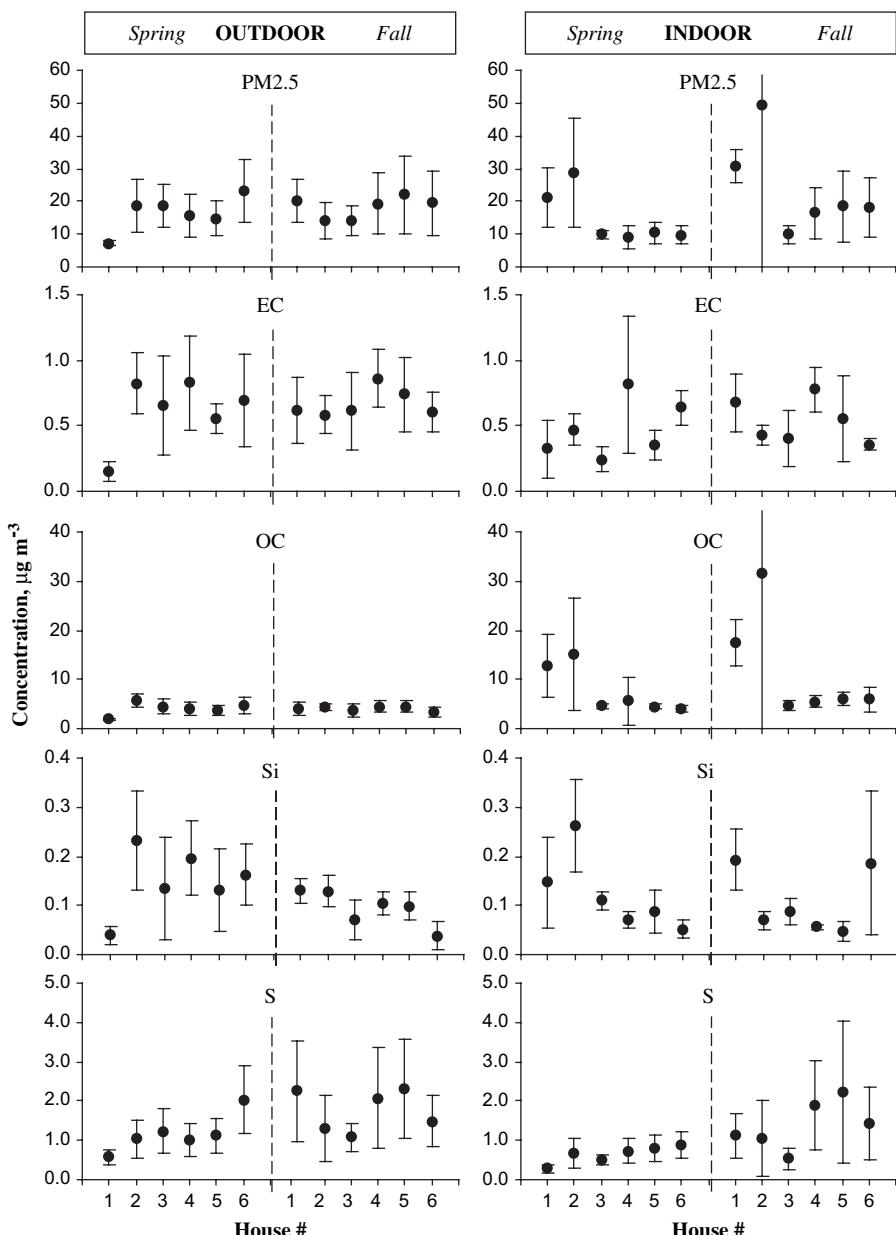


Fig. 1. Five-day averaged PM_{2.5} mass and elemental concentrations measured inside and outside six homes. The symbols represent the averages and the error bars the standard deviations.

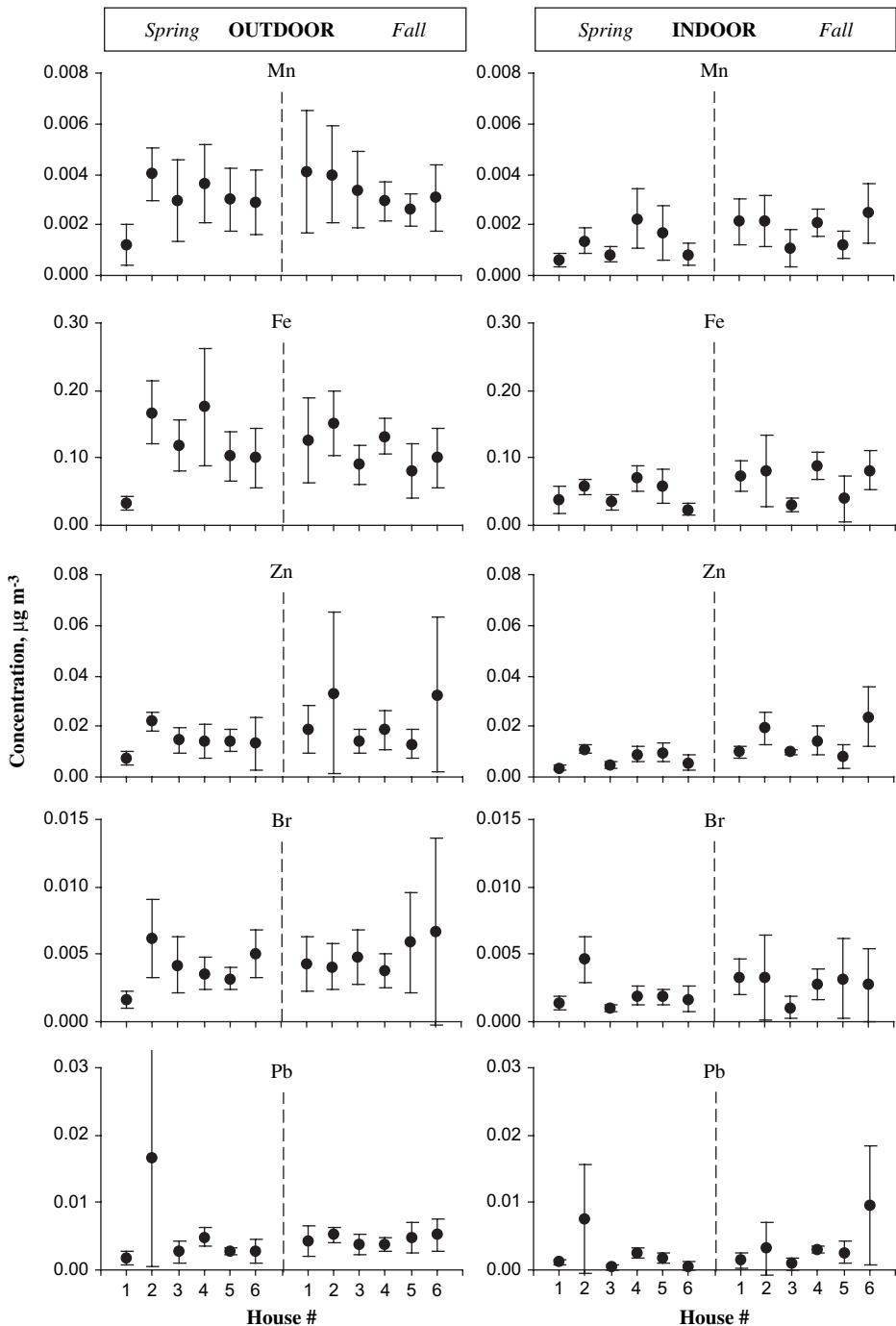


Fig. 1. (continued)

household dust due to renovating process and painting activities (latex paint).

The results obtained in this study agree with those found by other investigators who performed PM measurements in the roadside houses. Jones et al. (2000) reported the I/O of 1.6–1.7 for $\text{PM}_{2.5}$, 0.6–0.9 for sulfate, 0.5–0.7 for lead, 0.1–0.8 for zinc, 0.6–4.3 for elemental carbon, and 2.2–7.7 for organic carbon.

Similarly to our case, I/O ratios for carbonation compounds and zinc were higher in the smoking houses, while presence of sulfur and lead were attributed solely to the outdoor sources. The elemental carbon I/O ratio of less than 1 was observed in few houses and attributed to the outdoor source, most likely to the primary exhaust emissions from road traffic.

Table 2Five-day averaged (mean and standard deviation) I/O ratio of PM_{2.5} mass and elemental concentrations

House #	PM _{2.5}	EC	OC	Si	S	Mn	Fe	Zn	Br	Pb
Spring										
1	2.9 ± 1.2	2.2 ± 1.7	6.9 ± 3.3	5.1 ± 3.9	0.5 ± 0.1	0.8 ± 0.6	1.3 ± 0.8	0.6 ± 0.3	1.0 ± 0.5	0.9 ± 0.6
2	1.8 ± 1.2	0.6 ± 0.2	3.0 ± 2.9	1.3 ± 0.7	0.6 ± 0.1	0.4 ± 0.1	0.3 ± 0.0	0.5 ± 0.1	0.8 ± 0.3	0.4 ± 0.3
3	0.6 ± 0.2	0.3 ± 0.1	1.1 ± 0.3	1.6 ± 1.3	0.4 ± 0.1	0.4 ± 0.4	0.3 ± 0.1	0.3 ± 0.1	0.3 ± 0.1	0.3 ± 0.3
4	0.6 ± 0.2	1.0 ± 0.2	1.4 ± 0.9	0.4 ± 0.1	0.7 ± 0.1	0.8 ± 0.6	0.4 ± 0.1	0.7 ± 0.1	0.6 ± 0.2	0.5 ± 0.2
5	0.7 ± 0.0	0.6 ± 0.2	1.3 ± 0.3	0.8 ± 0.3	0.7 ± 0.1	0.5 ± 0.1	0.6 ± 0.1	0.6 ± 0.1	0.6 ± 0.2	0.7 ± 0.3
6	0.5 ± 0.2	1.9 ± 2.5	1.0 ± 0.7	0.4 ± 0.3	0.5 ± 0.1	0.3 ± 0.2	0.3 ± 0.2	0.7 ± 0.6	0.3 ± 0.1	0.4 ± 0.4
Fall										
1	1.7 ± 0.6	1.3 ± 0.7	4.9 ± 2.9	1.5 ± 0.2	0.5 ± 0.1	0.6 ± 0.2	0.7 ± 0.3	0.6 ± 0.2	0.9 ± 0.6	0.3 ± 0.3
2	4.7 ± 6.9	0.8 ± 0.3	7.6 ± 10	0.6 ± 0.2	0.7 ± 0.5	0.7 ± 0.4	0.6 ± 0.3	0.8 ± 0.5	0.7 ± 0.5	0.6 ± 0.7
3	0.7 ± 0.1	0.8 ± 0.8	1.4 ± 0.5	1.6 ± 1.1	0.5 ± 0.2	0.3 ± 0.2	0.4 ± 0.1	0.8 ± 0.4	0.2 ± 0.1	0.2 ± 0.2
4	0.9 ± 0.0	0.9 ± 0.1	1.2 ± 0.1	0.6 ± 0.2	0.9 ± 0.1	0.7 ± 0.1	0.7 ± 0.0	0.8 ± 0.2	0.7 ± 0.1	0.9 ± 0.3
5	0.9 ± 0.2	0.7 ± 0.2	1.4 ± 0.1	0.5 ± 0.1	0.9 ± 0.4	0.5 ± 0.1	0.4 ± 0.1	0.6 ± 0.1	0.5 ± 0.2	0.6 ± 0.3
6	0.9 ± 0.3	0.6 ± 0.1	1.8 ± 0.6	5.3 ± 4.5	0.9 ± 0.3	1.0 ± 0.7	0.9 ± 0.6	1.1 ± 0.8	0.5 ± 0.2	1.9 ± 2.3

3.2. The relationship between traffic-related PM_{2.5} components in the indoor/outdoor air

The relationships between traffic-related PM_{2.5} constituents in the outdoor air are presented in Table 3 as Spearman coefficients of correlation. This table gives a fair indication that the traffic component is very pronounced in the ambient air of the six houses, suggested by relatively high values of Spearman coefficient of correlation between traffic-related elements. We also observed elevated levels of certain elements, including EC, Zn and others, which seems to be in agreement with our previous measurements at the Cincinnati inner-city area (Martuzevicius et al., 2004).

Higher overall correlations were observed in spring compared to fall. In spring, PM_{2.5} showed significant and high correlation with OC ($R = 0.7$), S (0.84), EC (0.79) and several other elements. The lowest correlation with the PM_{2.5} concentration among tracers was identified for Zn (0.6) and Pb (0.59). These values are still considerably

high considering the origin of particles and very low concentrations of these elements. EC and OC had high correlation (0.86) and correlated well with the trace metals. High correlation of PM_{2.5} with S confirms an important role of secondary sulfate in the PM_{2.5} mass formation, as shown in our earlier study (Martuzevicius et al., 2004). The medium to low correlation with all remaining elements suggests that S content in particles cannot be attributed to traffic exhaust, but rather to regional coal combustion products. There was particularly high correlation among Si, Fe, Mn, and Zn. While the first two represent crustal material, Mn and Zn represent traffic-related particulate matter, which is either generated by tire wear and brake linings (and contributes to the road dust) or emitted by the vehicle exhausts (HEI, 2002).

During the fall season, the correlations in all cases were considerably lower and showed higher variability between species. PM_{2.5} was still relatively highly correlated with OC (0.71), but PM_{2.5} versus EC revealed only medium correlation (0.50), as well as EC versus OC (0.56). S was still highly correlated with PM_{2.5} (0.93) and showed consistent correlation with OC (0.63 in fall versus 0.61 in spring). This suggests that a significant portion of OC represented the transport of coal combustion aerosol (primary energy production source in the region). Similarly to the spring dataset, Mn, Fe and Zn from the fall samples revealed relatively high correlation among each other, although lower than those measured in spring. Mostly insignificant correlations were obtained for Pb in the fall season. This can be attributed to the overall level of Pb that in many cases was as low as the limit of detection.

The indoor versus outdoor correlations of PM_{2.5} and elements are presented in Table 4. This analysis serves as another indication of the influence of outdoor aerosols to the indoor particle composition and concentration. Higher correlation is expected for the elements of solely outdoor origin (S, heavy metals, Br) while indoor source-related elements should show lower or no correlation. As seen from the table, the values are rather low for all combinations. The indoor and outdoor PM_{2.5} was moderately correlated in fall, but showed little correlation in spring. The highest value is achieved by outdoor S versus indoor S, which consistently revealed high correlation in both

Table 3Correlations between PM_{2.5} elemental components in the outdoor air

	EC	OC	Si	S	Mn	Fe	Zn	Br	Pb
Spring									
PM _{2.5}	0.79	0.86	0.78	0.84	0.78	0.75	0.60	0.71	0.59
EC		0.86	0.68	0.53	0.76	0.86	0.72	0.53	0.66
OC			0.84	0.61	0.86	0.84	0.65	0.68	0.72
Si				0.56	0.89	0.82	0.53	0.69	0.73
S					0.53	0.41	0.28	0.62	0.40
Mn						0.91	0.71	0.64	0.71
Fe							0.79	0.57	0.75
Zn								0.56	0.63
Br									0.51
Fall									
PM _{2.5}	0.50	0.71	0.4	0.93	0.27	0.27	0.09	0.68	0.03
EC		0.56	0.44	0.39	0.27	0.40	0.16	0.48	0.06
OC			0.63	0.63	0.35	0.50	0.18	0.57	0.10
Si				0.43	0.43	0.63	0.17	0.27	0.02
S					0.14	0.22	-0.04	0.57	-0.02
Mn						0.77	0.64	0.41	0.48
Fe							0.71	0.37	0.28
Zn								0.25	0.44
Br									0.08

The values denote Spearman coefficients of correlation ($n = 30$). Significant correlations ($p < 0.05$) are marked bold.

Table 4Correlation between PM_{2.5} elemental components in indoor and outdoor environments

		Outdoor									
		PM _{2.5}	EC	OC	Si	S	Mn	Fe	Zn	Br	Pb
Indoor											
Spring	PM _{2.5}	0.21	0.04	0.24	0.05	0.33	0.08	0.00	0.14	0.24	0.16
	EC	0.47	0.50	0.47	0.41	0.33	0.39	0.52	0.47	0.33	0.46
	OC	-0.07	0.09	0.11	0.27	-0.11	0.14	0.14	0.40	0.3	0.25
	Si	0.26	0.28	0.49	0.18	0.20	0.22	0.23	0.25	0.25	0.21
	S	0.77	0.37	0.51	0.55	0.87	0.46	0.38	0.26	0.56	0.37
	Mn	0.49	0.32	0.37	0.39	0.40	0.39	0.38	0.38	0.39	0.31
	Fe	0.56	0.49	0.56	0.44	0.39	0.46	0.56	0.45	0.31	0.38
	Zn	0.62	0.56	0.65	0.53	0.42	0.63	0.67	0.77	0.54	0.58
	Br	0.41	0.18	0.47	0.45	0.40	0.37	0.30	0.34	0.61	0.43
	Pb	0.41	0.37	0.48	0.45	0.36	0.41	0.40	0.33	0.20	0.59
Fall											
PM _{2.5}	PM _{2.5}	0.52	0.27	0.47	0.42	0.51	0.11	0.25	0.04	0.27	0.14
	EC	0.38	0.42	0.53	0.42	0.33	0.21	0.31	0.21	0.19	-0.05
	OC	0.29	0.07	0.26	0.53	0.29	0.18	0.25	0.12	0.02	0.18
	Si	0.21	0.03	-0.01	0.24	0.20	0.31	0.29	0.23	0.07	0.07
	S	0.85	0.47	0.57	0.25	0.86	-0.04	0.15	-0.05	0.51	-0.03
	Mn	0.32	0.29	0.26	0.14	0.26	0.37	0.43	0.30	0.18	0.35
	Fe	0.32	0.31	0.34	0.29	0.34	0.31	0.65	0.42	0.23	0.09
	Zn	-0.07	-0.04	0.04	0.04	-0.11	0.36	0.57	0.69	0.11	0.33
	Br	0.68	0.47	0.63	0.45	0.61	0.13	0.32	0.05	0.64	-0.09
	Pb	0.31	0.23	0.22	0.03	0.37	0.13	0.21	0.06	0.13	0.36

The values denote Spearman coefficients of correlation ($n = 30$). Significant correlations ($p < 0.05$) are marked bold.

seasons (0.87 for spring and 0.86 for fall). The findings agree well with the value of 0.91 obtained by Long and Sarnat (2004) in Boston area, USA. The higher correlation was also obtained for Zn (0.77 for spring and 0.69 for fall), as well as Fe (0.56 and 0.65, respectively) and Br (0.61 and 0.64, respectively). Outdoor OC, S, Si, Mn, and Fe correlated moderately with the traffic-related elements (Mn, Fe, Zn, and Br) identified indoors, suggesting their infiltration.

The data should be interpreted with caution. For some elements, the relationships presented in Table 4 may not have an obvious physical meaning and therefore may seem to be of insignificant value. However, it is clearly seen that those correlations are rather low, while, in contrast, the elements exhibiting clear origin-related relationships have higher correlation values. Putting these values together allows comparing the significant and insignificant values. Consequently, conclusions are drawn on the physical relationship between the appropriate elements.

3.3. Estimation of the contribution of traffic-induced particulate matter to indoor environment

Simultaneously collected indoor and outdoor data were analyzed as different sets in three-dimensional data matrix for each house. Only "strong" species (signal/noise > 2 , Paa-toro and Hopke, 2003) were retained in model calculations. For each species, the source contribution estimate (SCE) – expressed in $\mu\text{g m}^{-3}$ – represented a portion of the total PM_{2.5}.

The estimated indoor and outdoor source contributions to indoor air samples are presented in Table 5. The indoor PM_{2.5} aerosols measured in Houses 1, 2, 3, 5 and 6 originated mostly from indoor activities. For House 1, the indoor source contribution was 6.3 times higher than outdoor

contribution in spring and 3.1 times higher in fall. The corresponding numbers for House 2 were 3.1 and 2.3, for House 3 – 2.6 and 3.2, for House 5 – 1.5 and 2.4, and for House 6 – 1 and 2.8. This finding is confirmed by the high I/O ratio determined for Houses 1 and 2 (Table 2).

For House 4, either the contributions of indoor and outdoor aerosols were comparable (spring) or the outdoor contribution was higher (2.3 times in fall). This house had no or very minor human activity at the time of sampling as well as a relatively loose building structure.

Contributions of two dominant outdoor sources – traffic- and regional combustion-associated sulfate – are shown in Table 6. The traffic-related sources contributed more to indoor samples, compared to regional sulfate source, in Houses 2–4 and 6 (spring); on the other hand, regional sulfate source showed greater contribution in Houses 1, 5 and 6 (fall). The competition between traffic- and regional combustion-associated sources does not seem to be solely dependent on the distance from highway

Table 5Indoor and outdoor source contributions to indoor air samples by PARAFAC ($n = 10$)^a

House no.	Mass contributions ($\mu\text{g m}^{-3}$)			
	Spring		Fall	
	Indoor origin	Outdoor origin	Indoor origin	Outdoor origin
1	12.1 ± 6.9	1.9 ± 0.3	15.3 ± 5.5	4.9 ± 1.8
2	12.7 ± 12.1	4.1 ± 1.3	9.9 ± 9.1	4.3 ± 1.7
3	4.2 ± 0.7	1.6 ± 0.5	4.5 ± 1.4	1.4 ± 0.6
4	4.2 ± 5.0	3.4 ± 1.0	2.7 ± 0.2	5.7 ± 2.4
5	3.5 ± 1.0	2.4 ± 1.0	6.5 ± 2.6	2.7 ± 0.9
6	2.9 ± 1.2	3.0 ± 0.9	6.5 ± 3.9	2.3 ± 1.2

^a PARAFAC model applied to each house individually (two seasons, five days).

Table 6

Traffic and sulfate source contributions (average \pm standard deviation) to indoor samples by PARAFAC ($n = 10$)^a

House no.	Mass contributions ($\mu\text{g m}^{-3}$)			
	Spring		Fall	
	Traffic	Sulfate	Traffic	Sulfate
1	0.2 \pm 0.1	1.7 \pm 0.4	0.7 \pm 0.5	4.1 \pm 1.6
2	3.3 \pm 6.8	0.9 \pm 0.7	2.1 \pm 0.3	2.1 \pm 1.6
3	1.4 \pm 0.73	0.14 \pm 0.09	1.3 \pm 0.74	0.14 \pm 0.07
4	1.8 \pm 0.6	1.2 \pm 0.7	2.1 \pm 0.6	3.3 \pm 2.1
5	0.5 \pm 0.2	0.4 \pm 0.4	0.6 \pm 0.3	1.3 \pm 0.9
6	1.8 \pm 0.9	1.1 \pm 0.3	0.6 \pm 0.6	1.7 \pm 1.7

^a PARAFAC model applied to each house individually (two seasons, five days).

and needs to be further examined. While House 1, which was the farthest distance (300 m) had one of the lowest contributions of traffic sources to indoor concentration for both seasons ($0.2 \mu\text{g m}^{-3}$ in spring and $0.7 \mu\text{g m}^{-3}$ in the fall), the remaining houses did not show appreciable differences, especially when averaged over the two seasons.

Generally, the traffic-related sources characterized by the model utilized in this study represent the mixture of diesel and gasoline powered engines. Due to the relatively high volume of the diesel engine vehicles on the highways near the tested houses (Table 1), we believe that the findings are primarily representative of the diesel engine emissions. Detailed differentiation seems possible resulting from more specific, advanced chemical analysis (Schauer et al., 1996; Kim and Hopke, 2004), which is rather expensive to process the samples generated in this study.

One of the advantages of the PARAFAC model is that it can provide the average estimation of the I/O ratio representing contributions from specific sources. This allows inter-comparing the houses in terms of penetration of traffic-induced particles. As presented in Fig. 2, I/O ratios representing contribution of traffic sources varied from 0.09 (House 3) to 0.86 (House 6). I/O ratios that represent contribution of regional combustion sulfate varied from 0.23 (House 6) to 0.91 (House 4), which agreed with the

I/O ratio measured for sulfur (Table 2). The I/O ratios of regional sulfate sources were higher than those of the traffic-related sources for all the houses except House 6. This can be attributed to the composition of the two sources. Sulfur is the most dominant species in regional pollution source, while OC, EC and sulfur are the dominant species generated by the traffic. These elements may exhibit different losses during the outdoor–indoor transport due to the difference in (a) particle size distributions and (b) tightness of the houses.

4. Summary and conclusions

The PM_{2.5} concentration was measured inside and outside of six residential houses located at 30–300 m from major highways during spring and fall sampling campaigns. The aerosol concentration determined inside the houses was dependent primarily on the presence of indoor pollution sources. Activities such as smoking (House 1), cooking and cleaning (House 2) resulted in I/O ratio >1 for PM_{2.5}. Sulfur proved to be a good marker of the penetration of outdoor pollution indoors, with the I/O between 0.4 and 0.9. The I/O for EC displayed varying behavior from house to house during spring season with an I/O >1 determined at Houses 4 and 6, which had no major indoor sources. The I/O ratio for the trace metals was below 1 for almost all cases, except House 1 (smoking and dusting) in spring and House 6 (dusting and painting) in fall.

The elemental correlation analysis for the outdoor and indoor aerosols confirmed the high influence of traffic component in the outdoor environment. A higher correlation between PM_{2.5} and trace elements was observed in spring. The outdoor versus indoor correlation analysis revealed that the highest value was observed for outdoor S versus indoor S, which consistently revealed high correlation in both seasons (0.87 for spring and 0.86 for fall). The indoor and outdoor PM_{2.5} concentrations were moderately correlated in fall and showed little correlation in spring. Higher correlations were also obtained for Zn (0.77 for spring and 0.69 for fall), as well as Fe (0.56 and 0.65) and Br (0.61 and 0.64), possibly indicating the penetration of traffic-related aerosols to indoor environment.

The receptor modeling based on three-way factor method with non-negative constraints allowed the quantification of contribution of the traffic-related aerosol to indoor aerosol level. The traffic component was distinctly pronounced in the ambient air of the six houses. Generally, traffic sources contributed more to indoor PM_{2.5} than sulfate sources, in the houses located close to a highway. Infiltration factors of traffic-related source varied from 0.09 to 0.86. Infiltration factors of regional sulfate source varied from 0.23 to 0.91. The method utilized in this study was proven to be suitable for the quantitative assessment of the contribution of traffic-related aerosol to the indoor environment. Furthermore, this assessment was made without involving the air exchange parameters (the latter are conventionally characterized using tracer gas methods, which are of limited application in occupied test houses). The data suggest that – for the houses located in a close proximity to major highways – the distance from the highway and traffic intensity factors may not necessarily

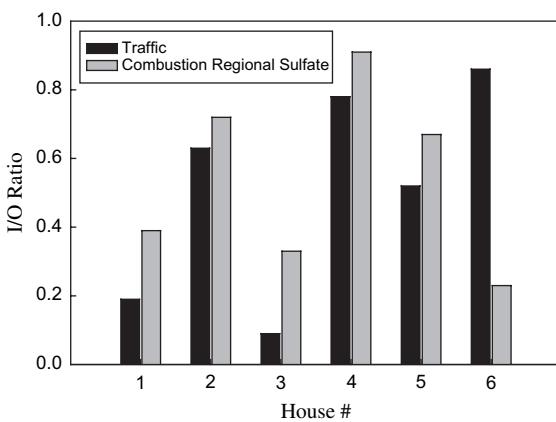


Fig. 2. Average I/O ratios of contributions from the traffic sources and the combustion regional sulfate sources, as determined by PARAFAC.

play the most important role in affecting the indoor concentrations of the traffic-related aerosol. The more important factors seem to be the structure of the house envelope and ventilation pattern, although these were not quantitatively assessed in this study.

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