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Organic and elemental carbon concentrations in fine particulate matter in residences, schoolrooms, and outdoor air in Mira Loma, California

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Abstract

Indoor and outdoor elemental carbon (EC) and organic carbon (OC) concentrations were measured from September 2001 through January 2002 at 20 residential sites and a local high school in western Riverside County, CA. The correlation (R^2) between indoor vs. outdoor EC and indoor vs. outdoor OC were 0.63 and 0.47, respectively, while the correlation of EC to OC outdoors and indoors was 0.58 and 0.23, respectively. The average OC content of $PM_{2.5}$ was 0.25 and 0.55 for outdoor and indoor $PM_{2.5}$, respectively. It was concluded that there were no significant indoor sources of EC while indoor OC sources contributed significantly to indoor $PM_{2.5}$. Home with smokers had significantly higher TC and OC than homes without. Schoolrooms generally had less EC and OC due to the schools HVAC system.

Keywords: Organic carbon; Elemental carbon; Indoor air quality; Environmental tobacco smoke

1. Introduction

It is generally recognized that most people spend the majority of their time indoors (approximately 85% of one's time according to Jenkins et al., 1992), suggesting that a significant portion of total personal exposure to ambient particles occurs in the indoor environment. Indoor sources of $PM_{2.5}$ include environmental tobacco smoke (ETS), cooking, carpet cleaning, dusting and other combustion sources (Kamens et al., 1991; Wallace, 1996; Long et al., 2000; Jones et al., 2000), which may lead to higher indoor $PM_{2.5}$ levels compared to ambient air. Kamens et al. (1991) measured indoor particles in three homes without smokers in North Carolina, USA in November and December 1987. The authors concluded that the most significant indoor source of fine particles in all three of these non-smoking homes was cooking, while the most significant indoor source of coarse particles was vacuuming and sweeping. A study conducted by Leaderer and Hammond (1991) showed that use of a gas stove fueled by natural gas did not elevate total particulate matter indoor concentrations.

Epidemiological studies have shown strong associations between particulate matter in outdoor air and lung function parameters, respiratory symptoms and mortality. These findings were especially pronounced for fine

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particles (smaller than 2.5 μm) that are inhaled (Schwartz, 1993; Xu and Wang, 1993). In addition, epidemiological studies indicate that ETS causes lung cancer in adults and is classified as by US EPA as a group A human carcinogen (US EPA, 1992). The preliminary recommended potency value of ETS for use in determining the excess cancer risk for a person exposed over a lifetime to 1 $\mu g\,m^{-3}$ of ETS is estimated to be no greater than 28 in one million (ARB, 1997). Indoor and outdoor studies in US homes showed that the most important indoor source of fine and coarse particles was cigarette smoking (Sheldon et al., 1989; Leaderer and Hammond (1991); Wallace, 1996).

It has been known that organic carbon (OC) and elemental carbon (EC) particles exist mainly in aerodynamic particle diameters of $0.1 < d < 1$ μm (Kleeman et al., 2000; Funasaka et al., 2000). Thus, OC and EC are more enriched in the fine mode than in the coarse mode. A study conducted by Funasaka et al. (2000) showed that EC is more abundant in the fine mode than OC. According to a study by Abt et al. (2000) on contribution of outdoor and indoor particle sources to indoor concentrations, effective penetration efficiencies for 0.02–0.5 μm particles was higher than 0.7–10 μm by a factor of two. This suggests that EC can penetrate into the house easier than OC. Likewise, EC can more easily penetrate into the airways and lungs where it may produce harmful health effects such as the worsening of heart and lung diseases.

Other than indoor emission sources, indoor air is affected by outdoor air through infiltration. Therefore, it is important to: (1) look into the relationship between indoor and outdoor concentrations of particulate matter; (2) examine whether the quality of air inside a home is affected by changes in outdoor concentrations; and (3) determine which emission sources within the domestic environment are the most important. To accomplish this, we measured concentrations of $PM_{2.5}$ mass, OC, EC, trace elements, ionic species, carbonyls and hydrocarbon in indoor and outdoor air. Several studies have shown that the region in which Mira Loma lies has some of the highest levels of outdoor $PM_{2.5}$ measured in southern California (Allen et al., 2000; Kim et al., 2000; Na et al., 2004). In this paper, we focus on sources of only OC and EC.

2. Experimental methods

2.1. Sampling

The community of Mira Loma (33° 59′N, 117° 31′W) is located in western Riverside County, CA, approximately 90 km east of downtown Los Angeles. Twenty residences in Mira Loma were randomly chosen for this study. Table 1 summarizes the characteristics of each residence. All but one residence (3B) used natural gas for cooking, and all but two (1B and 3A) used natural gas for heating. Five of the residences (3B, 4B, 5A, 7A and 7B) had occupants who smoked. Of the five residences, only residence 5A had frequent indoor smokers (average of 40 total cigarettes per day). All residences except two (2A and 3A) had pets. A ''sampling period'' consisted of 12 calendar days with sampling occurring on alternating days (six 24-hour samples per period). For each sampling period, the residences were chosen in sets of two in the same neighborhood, normally within 400 m of each other. The two residences in each set were named ''A'' and ''B'' for convenience. One outdoor sampler (located in the backyard of one of the two residences) was used for each pair of residences.

The samplers were installed inside the houses based on the following criteria: convenience of residents, proportion of time spent by residents in particular areas of the house, availability of power and ease of accessibility. In the case of homes with small children or pets, child security gates were installed for safety and security. The equipment was electronically set to operate for 24 hours starting at 8 PM. Participating residents were given logbooks to record household activities such as cooking, cleaning, use of air-conditioning/heating, smoking (inside/outside), etc. Additionally, samples were collected from a total of six rooms within the local high school during different sampling periods (one per sampling period). These included a library, an administrative office, and four classrooms. Sampling at the school and at the residences began simultaneously.

All samples throughout this study were collected approximately 1.5 m above ground level at flow rates of 5 $L\,min^{-1}$. A particle trap impactor (Biswas and Flagan, 1988) removed particles larger than 2.5 μm aerodynamic diameter. $PM_{2.5}$ was collected on 47 mm Teflo™ (Pall-Gelman, Ann Arbor, MI, USA) substrates. Parallel substrates collected samples for anions, EC/OC, carbonyls, nitric acid and ammonia (Sawant et al., 2004). A 47 mm diameter QAT Tissuquartz quartz fiber filters (Pall-Gelman, Ann Arbor, MI, USA) was used for OC and EC sampling. The filters were pre-cleaned to remove carbonaceous contaminants by firing at 600 °C for 4 h. Quartz filters were stored in petridishes in the dark at 0 °C until use.

2.2. Organic and elemental carbon analysis

A Thermal/Optical Carbon Aerosol Analyzer (Sunset Laboratory, Forest Grove, OR, USA) was used to analyze organic carbon and elemental carbon (OC/EC). OC and EC were determined by thermal-optical transmittance (TOT) using the National Institute for Occupational Safety and Health (NIOSH) Method 5040 protocol. More details on the analysis can be found elsewhere (Birch and Cary, 1996; Na et al., 2004).

Table 1
Summary details of houses used as sampling sites

Sampling period	Site ID	Smoking	Pets/Livestock[a]		Cleaning frequency[b]			Cooking		Ventilation and climate control			
			Outdoors	Indoors	Vac	Dust	Solv	Type	Frequency[b]	Heating type	Frequency[b]		
											H	W	AC
17/9–28/9	1A	No	7	0	0.5	0.3	0.5	Gas	1.6	Gas	0	0.6	0.9
	1B	No	2	0	1.0	0.4	0.9	Gas	2.0	Electric	0.1	0.9	0
29/9–10/10	2A	No	0	0	0	0.3	1.0	Gas	1.8	Gas	0.1	1.0	0
	2B	No	3	0	0.5	0.5	0.7	Gas	1.7	Gas	0	0.9	0
11/10–22/10	3A	No	0	0	0.3	0.2	0.1	Gas	0.9	Gas	0	0	0.9
	3B	Yes	33	6	0.4	0.1	0.3	Electric	2.4	Electric	0	0.5	0
23/10–3/11	4A	Yes[c]	1	0	0.3	0.3	0.3	Gas	0.7	Gas	0	0.8	0
	4B	Yes	116	2	0.1	0.1	1.0	Gas	2.8	Gas	0.1	1.0	0
4/11–15/11	5A	Yes	0	5	0.5	0.5	0	Gas	2.5	Gas	0.5	0.9	0
	5B	Yes[c]	15	7	0	0.3	0.4	Gas	2.3	Gas	0.5	0.9	0
16/11–29/11	6A	No	2	0	0.5	0	0.2	Gas	1.2	Gas	0	0.4	0
	6B	No	0	4	0.2	0.3	0.1	Gas	2.0	Gas	0.9	0.3	0
30/11–11/12	7A	Yes	0	5	0	0.3	0	Gas	1.2	Gas	0.8	0	0.2
	7B	Yes	0	8	0.5	0.8	0.2	Gas	2.3	Gas	1.0	0.8	0
12/12–23/12	8A	No	2	2	0.5	0.8	0.8	Gas	2.5	Gas	1.0	0.1	0
	8B	No	1	0	0.2	0.2	0	Gas	1.3	Gas	0.6	0.1	0
3/1–14/1	9A	No	5	7	0	0.1	0.2	Gas	1.1	Gas	1.0	0.9	0
	9B	No	2	0	na[d]	na	na	Gas	na	Gas	na	na	na
15/1–26/1	10A	Yes[c]	8	0	0.3	0.1	0.1	Gas	1.0	Gas	0.9	0.4	0
	10B	No	15	0	0.3	0.2	0.2	Gas	0.6	Gas	0.8	1.0	0

Key to abbreviations: Vac: vacuuming; Dust: dusting; Solv: use of solvents; H: use of heater; W: opening of windows; AC: use of air-conditioning.

[a]The "Pets/Livestock" field shows the actual number of animals present.

[b]The "Frequency" field indicates the number of times an activity was performed per day. For example, a frequency of 2.0 under the "cooking" field indicates that on average two meals were cooked per day.

[c]Although the residents of these homes smoked, they did not do so inside their homes.

[d]na indicates that data were not available.

2.3. Filter artifact correction of organic carbon

Gas-phase OC may be adsorbed to the filter media or PM already collected during sampling leading to a positive sampling artifact. Negative sampling artifacts result from the volatilization of organic compounds that are adsorbed to the PM due to pressure drop through the sampler as well as changes in temperature during particulate OC sampling. The combined effect of positive and negative OC artifacts can be estimated by measuring OC concentrations on a "backup" quartz filter located behind a Teflon filter (Kim et al., 2001). Turpin et al. (1994) and Fitz (1990) concluded that positive sampling artifacts comprise the major sampling artifacts for OC. Fitz (1990) showed that OC on the back filter is primarily an artifact due to irreversible adsorption of gas-phase organics and suggested that the subtraction of weight of OC on the back filter from that on the front filter gave a reasonable correction to the sampling artifact. Consequently, in this study, final OC concentrations are estimated by subtracting the OC measured on backup filters from the OC measured on front filters. Sampling artifacts were calculated from samples measured both inside and outside the residences. Positive artifacts measured inside the residences made up $58 \pm 15\%$ of the OC concentrations, which is higher than $45 \pm 17\%$ of outside the residences. When *t*-test was applied to the two cases, the difference was significant ($p < 0.01$ at 95% confidence interval).

2.4. OC correction factor

Turpin and Lim (2001) suggested using OC correction factors of 1.6 and 2.1 for urban and non-urban aerosols, respectively. An OC correction factor is defined as the ratio of the molecular weight of an organic compound to the total molecular weight of carbon in an organic compound. OC concentrations given throughout this paper were estimated as 1.9 times the OC measured in order to account for oxygen, nitrogen and hydrogen associated with organic matter after correction for adsorption and evaporation of organic matter from

the filters. This number (1.9) was chosen because it explained the constructed total $PM_{2.5}$ mass well.

3. Results and discussion

3.1. Indoor and outdoor houses

3.1.1. Indoor and outdoor concentrations of OC and EC

Table 2 presents the average indoor and outdoor concentrations of OC and EC, together with indoor/outdoor concentration ratios (I/O ratio). The two residences in each set were named "A" and "B" for convenience. In the absence of indoor sources, the mass concentration I/O ratio must be less than or equal to 1 (Chao and Wong, 2002). The average indoor concentration of OC measured at 20 homes was higher than the average outdoor concentration by a factor of 1.4. In addition, higher I/O ratio fluctuation in the OC concentrations was observed compared to those in the EC concentrations. The variation of elevated OC and I/O ratio can be attributed to varying OC emissions due to indoor activities.

In contrast to OC, average indoor EC concentrations are generally lower than those of outdoor concentrations (with one notable exception observed for house 5A), with an average I/O ratio of 0.8. It suggests that indoor EC emissions are much less significant than indoor OC emissions. The EC and OC results are consistent with other results observed by Geller et al. (2002) and Jones et al. (2000). Table 3 lists results of recent studies accomplished indoors and outdoors. All results show that OC concentrations indoors are higher than those outdoors, while EC is more abundant in outdoor air than in indoor air. This leads to higher indoor OC/EC in comparison with outdoor OC/EC.

It was reported by Daisey et al. (1998) that cigarette smoking emitted a significant amount of $PM_{2.5}$ (8100 μg per cigarette). It indicates that cigarette smoking can significantly contribute to elevated $PM_{2.5}$ mass indoors. In this study, the highest average OC/EC concentration ratios were observed inside the house with frequent indoor smokers (57.3), and the lowest average concentration was observed outdoors (5.0). A study for fine organic aerosol conducted by Hildemann et al. (1991)

Table 2
Indoor and outdoor concentrations of OC and EC measured in 20 homes (μg m^{-3})

Residences	Organic carbon				I/O	Elemental carbon				I/O
	Indoor		Outdoor			Indoor		Outdoor		
	Mean	S.D.	Mean	S.D.		Mean	S.D.	Mean	S.D.	
1A	13.5	1.4				1.9	0.2			
1B	15.5	2.7	14.0	4.1	1.1	2.4	0.4	2.5	0.7	0.9
2A	8.9	1.7				2.4	1.0			
2B	10.4	2.5	9.9	4.5	1.1	2.1	1.0	2.6	1.3	0.8
3A	18.8	4.3				1.5	0.7			
3B										
4A	16.0	2.1	13.0	5.7	1.2	1.9	0.9	3.0	1.8	0.6
4B	9.3	1.6				2.2	1.0			
5A	229.0	67.6	12.6	7.7	18.2	4.0	1.5	3.0	2.4	1.3
5B	28.5	13.9				2.5	1.1			
6A	25.3	11.6	11.6	4.7	2.2	2.0	1.2	2.3	1.5	0.9
6B	18.4	15.4				2.0	0.8			
7A	24.7	7.4	10.4	3.2	2.4	1.7	0.7	2.3	1.0	0.7
7B	17.2	3.9				1.8	0.6			
8A	17.1	4.5	14.8	1.7	1.2	2.0	0.5	2.4	0.6	0.8
8B	11.4	4.0				2.3	1.1			
9A	6.5	2.8				1.6	0.8			
9B	8.7	2.6				2.0				
10A	12.2	5.0				1.7	0.5			
10B	4.6	0.6				1.6	0.9			
Mean[a]	14.8		12.3		1.4	2.0		2.5		0.8

Blank values represent not measured or not analyzed values due to failure of measurement.
[a]Data of 5A was excluded in the calculation of average value.

Table 3
Average organic carbon (OC) and elemental carbon (EC) concentrations at various indoor and outdoor sites

		Indoor ($\mu g\,m^{-3}$)			Outdoor ($\mu g\,m^{-3}$)			OC	EC
		OC	EC	OC/EC	OC	EC	OC/EC	*I/O*	*I/O*
This study	House	14.8	2.0	7.4	12.3	2.5	5.0	1.4	0.8
Funasaka et al. (2000)	House	6.4	5.5	1.1	5.9	6.8	0.9	1.1	0.8
Long et al. (2000)	House	7.7	0.9	9.1	3.1	1.0	3.2	2.5	0.9
Landis et al. (2001)	Apartment	9.7	0.4	24.3	5.4	0.5	10.8	1.8	0.8
Geller et al. (2002)	House							1.8	0.9
LaRosa et al. (2002)	House		0.4			0.7			0.6

showed that OC contributes approximately 95% to total fine particle mass in cigarette smoke. This supports that cigarette smoking results in a high OC/EC ratio inside the house.

3.1.2. Relationship between indoor and outdoor EC and OC concentrations

Correlation analysis was applied to indoor and outdoor OC and EC concentrations to examine the presence of indoor emission sources of OC and EC. Residences with smokers are not considered in the analysis. The results are illustrated in Fig. 1. The correlation between indoor and outdoor EC ($R^2 = 0.63$) is better than the correlation for indoor and outdoor OC ($R^2 = 0.47$). The R^2 value of 0.63 indicates that outdoor EC can explain 63% of the variation for indoor EC concentrations. A weaker correlation between indoor and outdoor OC concentrations is probably due to significant contributions by indoor sources to the indoor OC concentrations. When outdoor EC and OC concentrations are used as independent values for indoor EC and OC concentrations, their intercepts are 0.44 and 5.12, respectively, as shown in Fig. 1. Each intercept roughly represents OC and EC concentrations that originate exclusively from indoor emission sources because intercepts are the concentration value when outdoor OC and EC concentrations are zero. In other words, the higher intercept indicates that a greater portion of the indoor concentration is derived from indoor sources. The ratio of the OC concentration intercept to the average indoor OC concentration represents the contribution of indoor sources of OC to measured indoor OC concentrations. The same concept can be applied to EC concentrations. The ratios for OC and EC are 0.4 and 0.2, respectively. This suggests that a substantial fraction of indoor EC concentration can be influenced by outdoor EC concentration. This is consistent with results reported by Jones et al. (2000), who found that EC originates

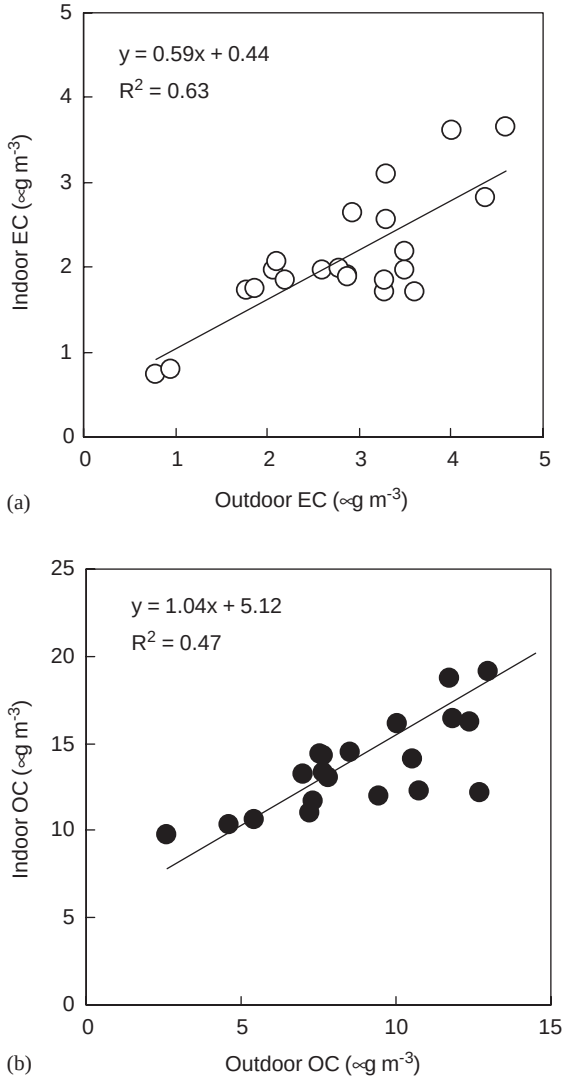

Fig. 1. Relationship between indoor and outdoor concentrations of OC and EC.

outdoors, mostly from vehicular emissions, except for homes with cigarette smokers.

To examine the similarity of emission sources between indoor and outdoor EC and OC, correlations between EC and OC in the indoor environment and correlation between EC and OC in the outdoor environment are compared in Fig. 2. The correlation between EC and OC indoors is weaker ($R^2 = 0.23$) than that of EC and OC outdoors ($R^2 = 0.58$). It implies that a majority of the outdoor OC comes from emission sources similar to those responsible of EC (e.g. combustion of fossil fuel). In addition, this result suggests that indoor emission sources of OC differ from those outdoors.

To observe the seasonal changes in OC I/O, we compared OC I/O between September and October (the warmer season) and December and January (the colder season). The overall I/O ratio for OC was approximately 1.4 times lower during the September and October period (1.4) than during December and January (2.0).

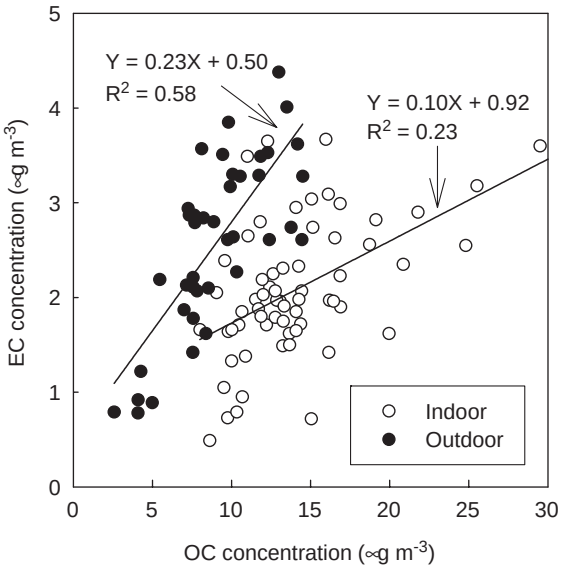

Fig. 2. Comparison between EC and OC concentrations for indoors and outdoors.

This suggests that the indoor OC concentration is influenced more by outdoor OC sources than by indoor OC sources during September and October. The more frequent opening of windows (refer to Table 1) during the September/October period may be responsible for a lower I/O ratio in OC concentration.

3.2. Concentrations of OC and EC inside the local high school

Table 4 shows average concentrations of OC and EC measured inside the school. On average, OC concentrations in the classrooms are slightly higher that those in the library and office. The I/O ratios for average OC and EC concentrations in the schoolroom were found to be 0.6 and 0.2, respectively. A ratio below 1.0 indicates that there are no significant emission sources of OC and EC and that there is an effective air cleaning system in the schoolroom. Each schoolroom in this study was equipped with a heating/ventilation/air-conditioning (HVAC) system with HEPA filters. It is likely that this played a role in removing particulate matter. The lower ratio also indicates that indoor air quality is more sensitive to influence by outdoor air. We thus believe that OC and EC concentrations in the schoolroom may be attributed mainly to resuspension of previously deposited OC and EC, and infiltration from outdoor sources.

To examine the impact of students' activities on indoor air quality, a comparison of OC and EC concentrations between samples taken on weekdays and weekends was made. Here, weekend and weekday samples were defined as ones measured from 8 PM Friday night to 8 PM Sunday night and from 8 PM Sunday night to 8 PM Friday night, respectively. During weekends, there was no activity and all doors remained closed. Therefore, indoor air on the weekends is expected to be affected much less by human activities at these times. The average OC concentrations are higher in the weekday samples ($8.8 \pm 4.7\,\mu g\,m^{-3}$) than in the weekend ones ($7.4 \pm 2.4\,\mu g\,m^{-3}$). No significant

Table 4
Organic carbon and elemental carbon concentrations inside the school (unit: $\mu g\,m^{-3}$)

	Organic carbon			Elemental carbon		
	Mean	Min	Max	Mean	Min	Max
Classroom-1	6.9	4.4	8.6	1.3	1.0	1.5
Classroom-2	9.7	5.1	18.7	0.9	0.7	1.2
Classroom-3	7.5	6.8	8.5	0.9	0.4	1.4
Classroom-4	6.2	5.8	7.3	1.4	0.3	3.4
Administrative office	7.0	6.7	7.8	1.0	0.6	1.8
Library	5.3	4.8	6.3	0.7	0.3	1.4

difference between EC concentrations in the weekday ($1.1 \pm 0.9\,\mu g\,m^{-3}$) and weekend ($1.0 \pm 0.5\,\mu g\,m^{-3}$) samples was found. OC and EC concentrations are not significantly different between weekday and weekend measurements. However, higher fluctuations in both OC and EC concentrations during the weekday is likely due to a combination of student activities and infiltration from outdoor sources. Since doors and windows are opened more frequently during the weekday, it is easier for outdoor sources of EC and OC to infiltrate the classrooms.

3.3. Comparison of OC and EC concentrations between the residences and the school

Average concentrations of OC and EC for total $PM_{2.5}$ mass measured inside the residences, outside the residences, and inside schoolrooms are compared in Fig. 3. Current legislation focuses on outdoor air quality as a marker for exposure of individuals to pollutants. A standard for $PM_{2.5}$ has not been established for indoor environments. Therefore, the current California 24-hour $PM_{2.5}$ standard and the annual $PM_{2.5}$ standard were applied to the indoor $PM_{2.5}$ mass for the purpose of air quality assessment. In this figure, the component "others in the fine particles" represents particulate matters (e.g., NO^-, SO_4^{2-}, NH_4^+, trace elements, etc.) other than OC and EC.

Average total $PM_{2.5}$ mass concentration inside the house than is lower outside the house excluding the frequent smoker's house. This is consistent with the results of other studies (Jones et al., 2000; Geller et al., 2002). However, the fraction of OC to total $PM_{2.5}$ mass concentration is significantly higher inside the houses than outside the houses. Especially, the fraction in the house with frequent smokers is 3.9 times higher than that of outside the house. Houses with smokers showed a higher $PM_{2.5}$ mass concentration when compared to those of inside the houses without smokers and outside the house. This is consistent with results reported by other researchers (Lebret et al., 1987, Quackenboss et al., 1991). Lebret et al. (1987) estimated that smoking one cigarette per day added $0.8\,\mu g\,m^{-3}$ to the 24-hour $PM_{2.5}$ concentration indoors. These findings show that cigarette smoking is a significant contributor to $PM_{2.5}$ mass concentration. In this study, high concentration of $PM_{2.5}$ in the residence with frequent smokers is because of OC.

It is noted that indoor OC concentrations for residences exceeded the current outdoor California annual $PM_{2.5}$ standard ($12\,\mu g\,m^{-3}$) without consideration of other particulate matter. A house with frequent indoor smokers violates both California 24-hour and annual $PM_{2.5}$ standards. This suggests that a reduction in OC concentration is a very important factor to reduce the total $PM_{2.5}$ mass concentration for an indoor environment. Classrooms in the school showed the lowest EC, OC and total $PM_{2.5}$ mass concentrations. The school had an air-conditioning system with HEPA filters that were cleaned on a regular basis, while many residences had no filtering systems. The lower levels of OC and EC inside the school, in comparison with those inside the houses, may be attributed to a lower net indoor infiltration due to the heating/ventilation/air-conditioning system.

4. Summary

The relationship between EC and OC concentrations inside a number of residences and classrooms relative to outdoor concentrations has been thoroughly investi-

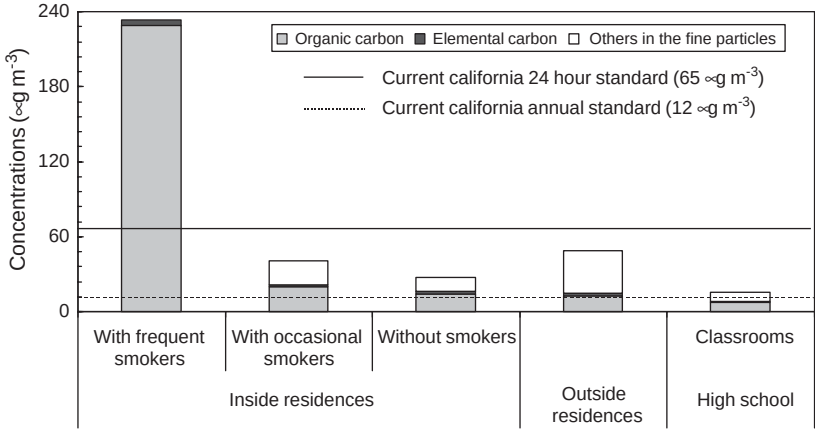

Fig. 3. Comparison of average OC and EC concentrations between inside the residences, outside the residences, and inside the classrooms.

gated. It was determined that indoor $PM_{2.5}$ was significantly influenced by indoor OC sources while indoor EC sources were predominantly of outdoor origin. OC levels were significantly higher inside the homes (and often exceeded annual outdoor fine particle air quality standards by OC alone) than outside for all homes with the most significant differences observed for the home that had a frequent indoor smoker. Overall, OC and EC levels were lower inside the classrooms than inside the residences due to the efficient HVAC system at the school. Comparison of weekday/weekend data demonstrated that school activity did increase OC loading but did not affect EC loading.

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Disclaimer

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