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TECHNICAL PAPER

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Source Apportionment of Indoor, Outdoor, and Personal $PM_{2.5}$ in Seattle, Washington, Using Positive Matrix Factorization

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ABSTRACT

As part of a large exposure assessment and health-effects panel study, 33 trace elements and light-absorbing carbon were measured on 24-hr fixed-site filter samples for particulate matter with an aerodynamic diameter <2.5 μm ($PM_{2.5}$) collected between September 26, 2000, and May 25, 2001, at a central outdoor site, immediately outside each subject's residence, inside each residence, and on each subject (personal sample). Both two-way (PMF2) and three-way (PMF3) positive matrix factorization were used to deduce the sources contributing to $PM_{2.5}$. Five sources contributing to the indoor and outdoor samples were identified: vegetative burning, mobile emissions, secondary sulfate, a source rich in chlorine, and a source of crustal-derived material. Vegetative burning contributed more $PM_{2.5}$ mass on average than any other source in all microenvironments, with average values estimated by PMF2 and PMF3, respectively, of 7.6 and 8.7 μg/m^3 for the outdoor samples, 4 and 5.3 μg/m^3 for the indoor samples, and 3.8 and 3.4 μg/m^3 for the personal samples. Personal exposure to the combustion-related particles was correlated with outdoor sources, whereas exposure to the crustal and chlorine-rich particles was not. Personal exposures to crustal sources were strongly associated with personal activities, especially time spent at school among the child subjects.

IMPLICATIONS

Total personal exposures to $PM_{2.5}$ can differ from the levels measured at outdoor monitoring sites. Therefore, the sources that contribute to personal $PM_{2.5}$ exposures also differ. Understanding the sources contributing to these exposures is important to interpreting health studies and also to setting relevant control policies. The source apportionment technique used in this study, positive matrix factorization, appears to be a useful approach when applied to filter-based particle measurements of both trace elements as well as light-absorption coefficients.

INTRODUCTION

Airborne particulate matter (PM) is a mixture of many different chemical species. Elevated morbidity and mortality have been associated with outdoor PM mass concentrations in many epidemiological studies.[1] However, an individual's exposure to PM derives from indoor and outdoor source emissions as well as PM generated by personal activities. The identification of sources and the assessment of their relative contribution to total indoor and personal exposures can provide valuable information for epidemiologists and regulatory agencies. Only recently have researchers possessed the tools and technologies to be able to examine the health effects of individual PM components[2–11] and source types.[12–17] These efforts

have been based on either traditional factor analysis or a priori source profiles.

Assessment of human exposure to various PM sources is an important step in understanding how specific outdoor PM components or sources contribute to the observed associations between PM concentrations as measured at an outdoor monitoring site and adverse health effects. Koutrakis et al.[18] applied a simple physical model to estimate the relative contribution of indoor and outdoor aerosol sources to indoor PM concentrations. Yakovleva et al.[19] used three-way positive matrix factorization (PMF3), rather than the more widely used two-way model (PMF2), to simultaneously analyze indoor, outdoor, and personal data from the 1991 Particle Total Exposure Assessment Methodology study in Riverside, CA, to identify major sources and their contributions to personal exposure to PM with aerodynamic diameter <10 μm (PM_{10}). More recently, Hopke et al.[20] have also simultaneously analyzed indoor, outdoor, and personal data from the U.S. Environmental Protection Agency's (EPA) 1998 Baltimore, MD exposure panel study using the multilinear engine.

In this paper, both PMF2 and PMF3 are applied to apportion source contributions on a subset of central site, outdoor, indoor, and personal PM samples collected from the Seattle, WA, exposure and health-effects panel study of high-risk subpopulations.[21] Major sources of $PM_{2.5}$ in Seattle are motor vehicles and wood combustion, with additional contributions from secondary sulfate (SO_4^{2-}), sea salt, oil combustion, resuspended soil, and relatively small contributions from specific industrial sources.[22] Here, attenuation of various sources from outdoor to indoor and personal environments is examined in the context of total exposure to $PM_{2.5}$. The effects of personal activities on the personal source estimates are also examined.

METHODS

Sampling and Analysis

Data consisted of 24-hr Teflon filter measurements collected as part of a large panel study in Seattle investigating the health effects of acute exposure to $PM_{2.5}$ in susceptible populations.[21,23] Figure 1 shows the study region and sample locations. Filter samples were simultaneously collected at several fixed locations: at a central outdoor site, immediately outside each subject's residence, and inside each subject's residence. An additional filter was collected with a sampler worn by each subject. Samples were taken from 20 subjects, including 10 asthmatic children, 9 elderly subjects with coronary heart disease, and 1 elderly subject with chronic obstructive pulmonary disease. The fixed-location samples were collected with a 10-L/min Harvard Impactor for $PM_{2.5}$ ($HI_{2.5}$; Air Diagnostics, Inc.), and the personal samples were collected with a 4-L/min Harvard Personal Environmental Monitor for $PM_{2.5}$ ($HPEM_{2.5}$; Harvard School of Public Health). Subjects reported their activities in a formatted time-activity diary (TAD) with 15-min resolution. Samples were collected between September 2000 and May 2001. Each filter was analyzed for $PM_{2.5}$ mass[24] and for 33 elements by X-ray fluorescence. Table 1 summarizes the measured values used in the PMF model described later. The highest mass found for all samples was sulfur, while personal and indoor samples also had high chlorine (Cl) mass. Nickel (Ni) and vanadium (V) are below the detection limit on more than 75% of the personal samples. In addition, a subset of the Teflon filters from home indoor, home outdoor, and central site locations were extracted with ethyl acetate, and then levoglucosan concentrations were determined by GC/MS, as recently described.[25] Levoglucosan was detected in >95% of samples analyzed, with a mean value of 204 ng/m^3 (range <4 to 1280 ng/m^3). Precision and accuracy of the $HI_{2.5}$ and $HPEM_{2.5}$ measurements were reported in a previous paper.[21]

Additional fixed 4-L/min $HI_{2.5}$ samplers were collocated both outdoors and indoors to measure particulate organic (OC) and elemental (EC) carbon on quartz filters via thermal optical transmittance using a modified version of the National Institute of Occupational Safety and Health method.[26,27] The particle light absorption coefficient (b_{ap}) on the Teflon filters was also measured using an integrating plate method.[28–30] It is important to note that personal quartz filter samples were collected but suffered from various analysis problems, mainly a large positive sampling artifact for OC.[26] Therefore, measurement of b_{ap} on the personal Teflon filters had the additional, important advantage of allowing us to quantify personal exposure to light-absorbing carbon (LAC) and, therefore, to combine all three types of samples (personal, indoor, and outdoor) in the PMF analyses. The PMF2 results for the outdoor samples using LAC were compared with that using EC and OC because the positive OC artifact is much less pronounced on the outdoor filters.[26]

The particle light-absorption coefficient as determined by the integrating plate method is defined as

$$b_{ap} = \frac{A}{V} \ln\left\{\frac{I_{blank}}{I_{sample}}\right\} \tag{1}$$

where b_{ap} is particle absorption coefficient, m^{-1}, V is volume of air drawn through the filter (14.4 m^3 nominally for the HI samples and 5.76 m^3 nominally for the HPEM samples, with ±5% error), and, for the integrating plate instrument, A is area of the instrument sample spot, (6.45×10^{-4} m^2), I_{blank} is intensity of light at 525 nm

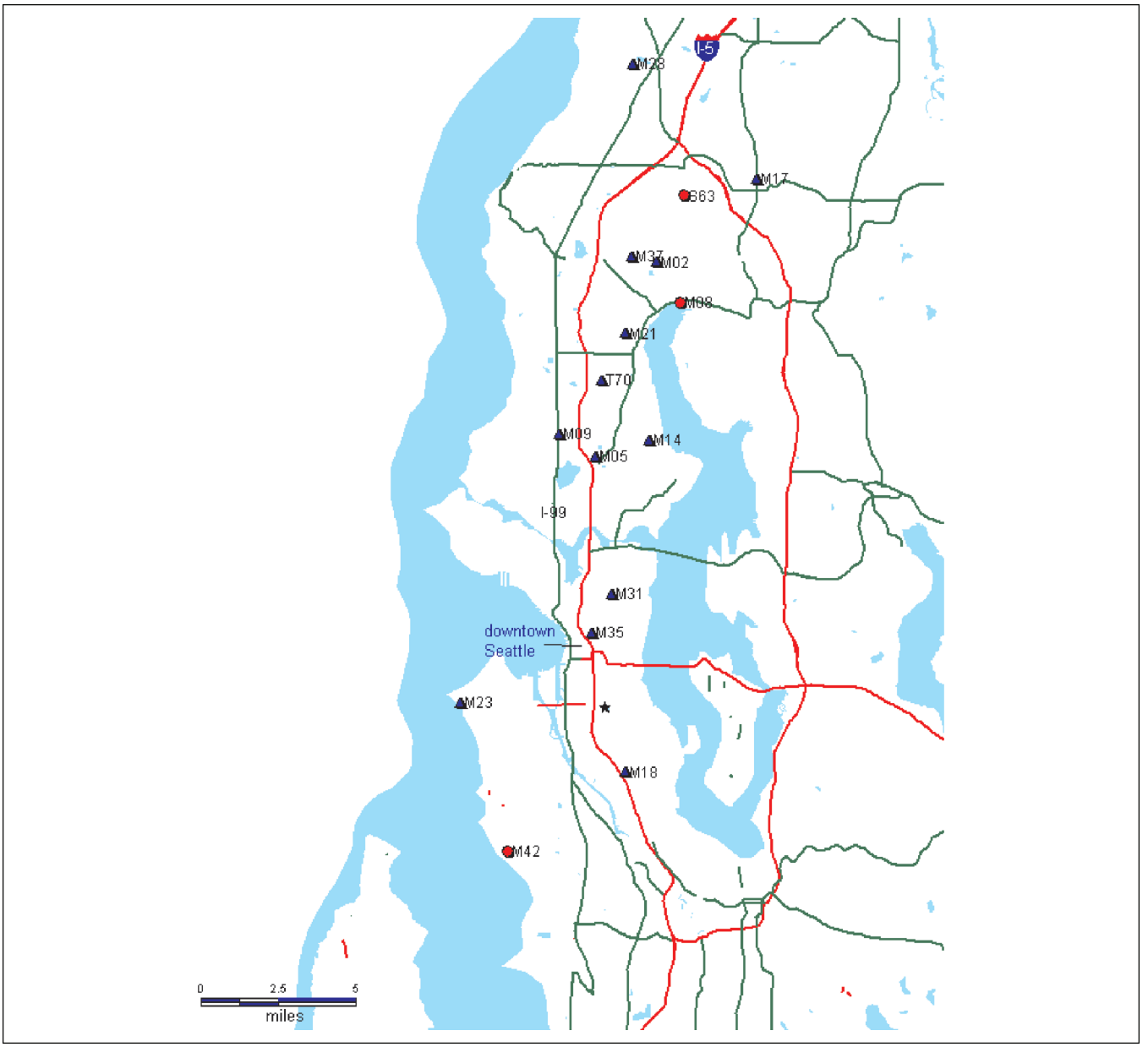

Figure 1. Map of study region, including the location of the central site and the subject's residences. Triangles mark residences with both fixed-site and personal samples, circles are residences with only fixed-site samples, and the asterisk sign denotes the central site.

transmitted through the unloaded, blank filter relative to a known initial reference light intensity, I_0, I_{sample} is intensity of light at 525 nm transmitted through the loaded sample filter relative to an initial reference light intensity of I_0.

Ideally, the values of I_{blank} would have been measured on each filter prior to sampling. Instead, a number of values from blank filters taken from each batch were averaged, and this average value was used in all subsequent calculations of b_{ap}. The variation in the blank values is relatively small (SD = 2.7×10^{-6} m^{-1}) compared with the average blank reading (2.9×10^{-5} m^{-1}). Regressing EC on the corresponding value of b_{ap} measured on the quartz filters, a particle light absorption efficiency of 7.1 m^2/g of EC was used (see Figure 2). We also regressed b_{ap} on EC to quantify the amount of LAC for use in subsequent analyses (LAC in μg/m^3 = $(1.09 \times 10^5)\, b_{ap} - 0.2$; R^2 = 0.77). Table 1 also summarizes the LAC results.

To examine the sources of $PM_{2.5}$ mass, the data were analyzed using PMF. PMF3 was used with sample type as the third dimension (paired indoor, personal, and outdoor samples on a given day). However, because of the limited number of personal samples relative to the indoor and outdoor samples, PMF2 also was used to separately analyze each sample set, allowing substantially more indoor and outdoor measurements to be included.

Table 1. Summary of XRF trace element and LAC analysis results (ng/m^3).

	Personal (n = 83)					Home Indoor (n = 161)					Home Outdoor (n = 198)					Outdoor Central Site (n = 96)				
Species	Mean	Median	Min	Max	% BDL	Mean	Median	Min	Max	% BDL	Mean	Median	Min	Max	% BDL	Mean	Median	Min	Max	% BDL
Mass	10,500	8860	1090	46,400	0	10,250	7780	2590	65,340	0	12,690	10,640	2020	43,000	0	11,970	10,480	1100	29,660	0
Al	32	23	1	114	1	19	15	0	131	3	21	15	0	218	12	31	19	0	220	5
As	1	1	0	11	50	1	1	0	9	52	2	2	0	16	31	2	1	0	8	32
LAC	1439	1246	0	5650	9	1105	971	0	3558	3	1830	1532	0	5914	12	1741	1599	0	4784	6
Br	3	2	0	26	23	2	2	0	25	11	3	3	0	25	2	3	3	0	11	2
Ca	72	50	11	381	0	46	35	9	647	0	36	28	5	398	0	50	38	9	383	0
Cl	248	34	0	2504	8	173	34	0	4218	5	75	29	0	1167	17	78	19	0	1818	19
Cr	2	1	0	13	45	2	1	0	68	30	1	1	0	5	51	2	1	0	12	23
Cu	3	2	0	40	30	4	2	0	237	23	2	2	0	18	18	3	3	0	12	7
Fe	63	47	11	385	0	35	30	5	101	0	61	58	6	264	0	95	82	13	557	0
K	57	44	2	420	0	54	45	19	219	0	78	58	8	326	0	67	60	4	167	0
Mn	2	2	0	26	43	2	1	0	15	27	3	2	0	48	21	6	4	0	32	8
Ni	0	0	0	4	95	0	0	0	5	73	1	0	0	5	58	1	1	0	13	41
Pb	2	1	0	25	70	2	2	0	11	54	5	4	0	36	25	5	4	0	16	21
Si	109	82	11	477	0	65	53	10	347	0	66	50	4	511	0	95	62	9	638	0
S	289	237	42	1835	0	289	238	82	1038	0	468	410	133	1526	0	492	448	83	1309	0
Ti	4	3	0	24	15	3	3	0	12	8	3	3	0	16	8	6	4	0	41	4
V	0	0	0	5	76	1	1	0	5	42	2	2	0	9	22	3	2	0	16	14

PMF Model Description

The general problem[31,32] can be stated in terms of modeling the contribution from p independent sources to all chemical species in a given sample, as follows:

$$\hat{x}_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} \tag{2}$$

where for airborne particles x_{ij} is the jth species concentration (μg/m^3) measured in the ith sample, g_{ik} is the particulate mass concentration (μg/m^3) from the kth source contributing to the ith sample, and f_{kj} is the jth species mass fraction (μg/μg) from the kth source.

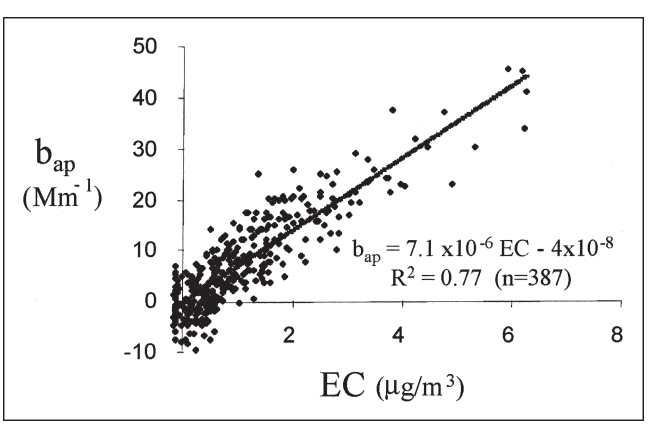

Figure 2. Scatter-plot of light absorption coefficient vs. EC (see text for details).

Eq 2 can also be solved without prior knowledge of f_{kj}. PMF provides only one of an infinite number of possible solutions to eq 2 (i.e., only one of many combinations of the g and f matrices).[33] PMF provides a solution that minimizes an object function, $Q(E)$, based upon the value of each observation and its corresponding uncertainty, subject to nonnegativity constraints.[34] Specifically, this function is defined as

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \hat{x}_{ij}}{u_{ij}} \right]^2 \tag{3}$$

where $\hat{\chi}_{ij}$ is the value predicted by PMF and u_{ij} is an uncertainty estimate in the jth element measured in the ith sample. The results of PMF modeling are scaled to the measured mass concentration by a scaling constant, s_k. Specifically, for PMF2

$$\hat{x}_{ij} = \sum_{k=1}^{p} (s_k g_{ik}) \left(\frac{f_{kj}}{s_k} \right) \tag{4}$$

where s_k is determined by regressing measured total $PM_{2.5}$ mass concentration against g_{ik}.

PMF3 solves the tri-linear equation

$$x_{ijh} = \sum_{k=1}^{p} a_{ik} b_{jk} c_{hk} + e_{ijh} \tag{5}$$

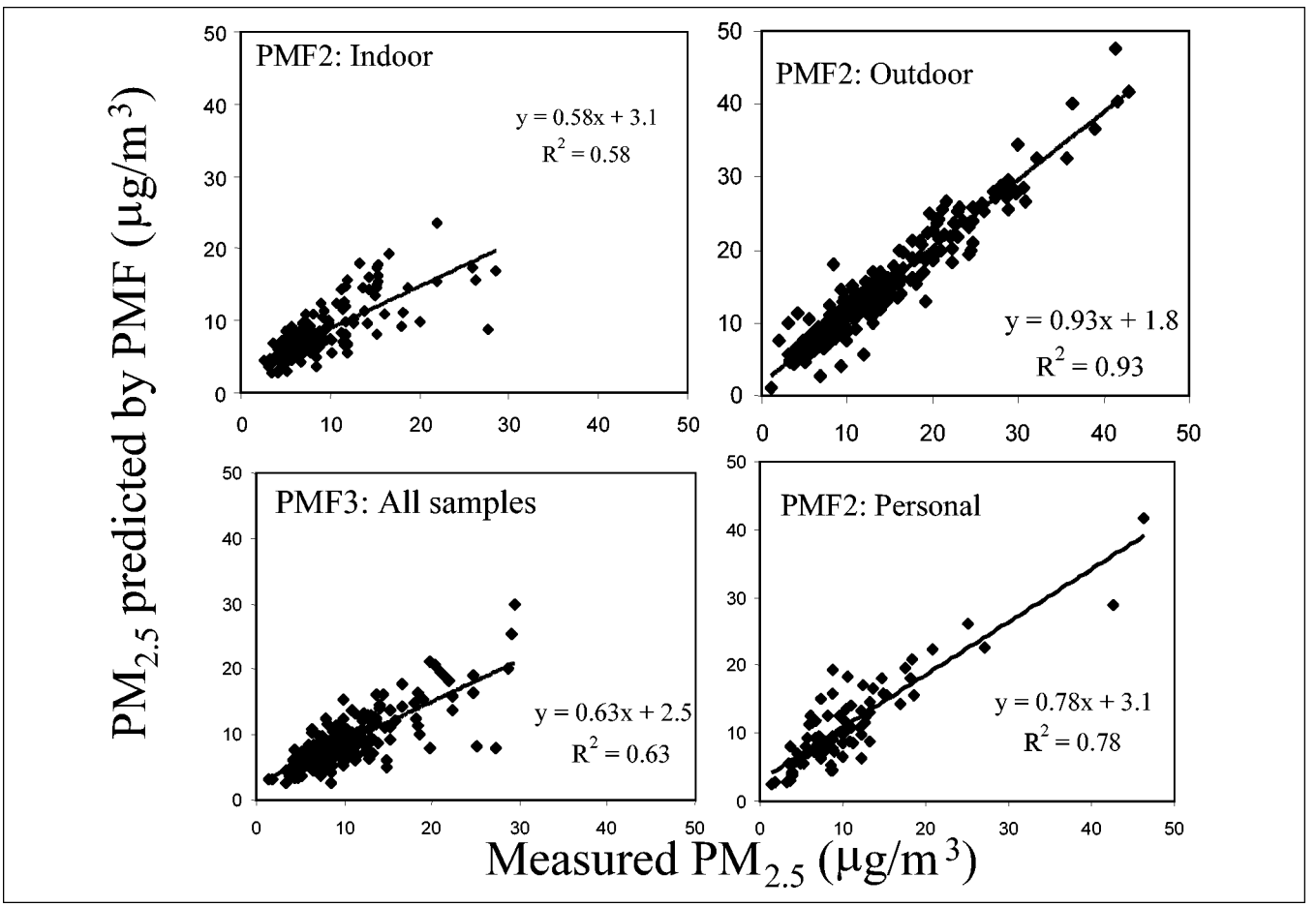

Figure 3. Scatter-plots of measured $PM_{2.5}$ vs. that predicted by PMF2 for indoor, outdoor, and personal samples, and for samples from all locations by PMF3.

where a_{ik} is an element of a matrix of factor scores related to the source contributions, b_{jk} is an element of a matrix of normalized factor scores related to the source profiles, and c_{hk} is an element of the matrix of the fractional contributions of each of h planes. In this case, $h = 3$ corresponding to the three sample modes: outdoor, indoor, and personal samples. PMF3 minimizes Q and reports one set of source profiles applicable to all three sampling environments. Unlike PMF2, there is no ability to rotate the solution after the fact; that is, there is no function equivalent to FPEAK, and thus, there is less rotational ambiguity. The values a_{ik} and b_{jk} are scaled via eq 4 the same as g_{ik} and f_{kj}, respectively.

PMF Model Implementation

PMF requires measurement uncertainties as well as the measurements themselves. The procedure of Polissar[35] was used to assign overall uncertainties to each measurement based upon reported one-sigma analytical uncertainty values and minimum detection limits. Specifically, the sum of the reported analytical uncertainty and one-third of the detection limit value was used as the overall uncertainty assigned to each measured concentration. Values below the detection limit were replaced by half of the detection limit values, and their overall uncertainties were set at 4 times the detection

Table 2. Average source contributions via PMF2 and PMF3 (μg/m^3).

	Outdoor	Indoor	Personal
Via PMF2	$n = 294$	$n = 161$	$n = 83$
Veg burn	7.6 (0.4)	4 (0.2)	3.2 (0.6)
Mobile	1 (0.06)	0.3 (0.02)	—
Fuel oil	0.9 (0.07)	—	—
S, Mn, Fe	—	—	0.9 (0.1)
Secondary	2.8 (0.1)	2.9 (0.2)	—
Cl-rich	0.5 (0.05)	0.3 (0.08)	1.1 (0.5)
Crustal	0.6 (0.04)	0.9 (0.05)	2.8 (0.6)
Crustal 2	—	—	3.1 (0.8)
Via PMF3	$n = 61$	$n = 61$	$n = 61$
Veg burn	8.7 (1.4)	5.4 (0.8)	3.4 (0.5)
Mobile	1 (0.3)	0.4 (0.1)	0.6 (0.2)
Secondary	2.2 (0.5)	1.3 (0.3)	1.6 (0.4)
Crustal	0.3 (0.06)	2.3 (0.5)	2.7 (0.6)

Note: Parentheses denote standard error.

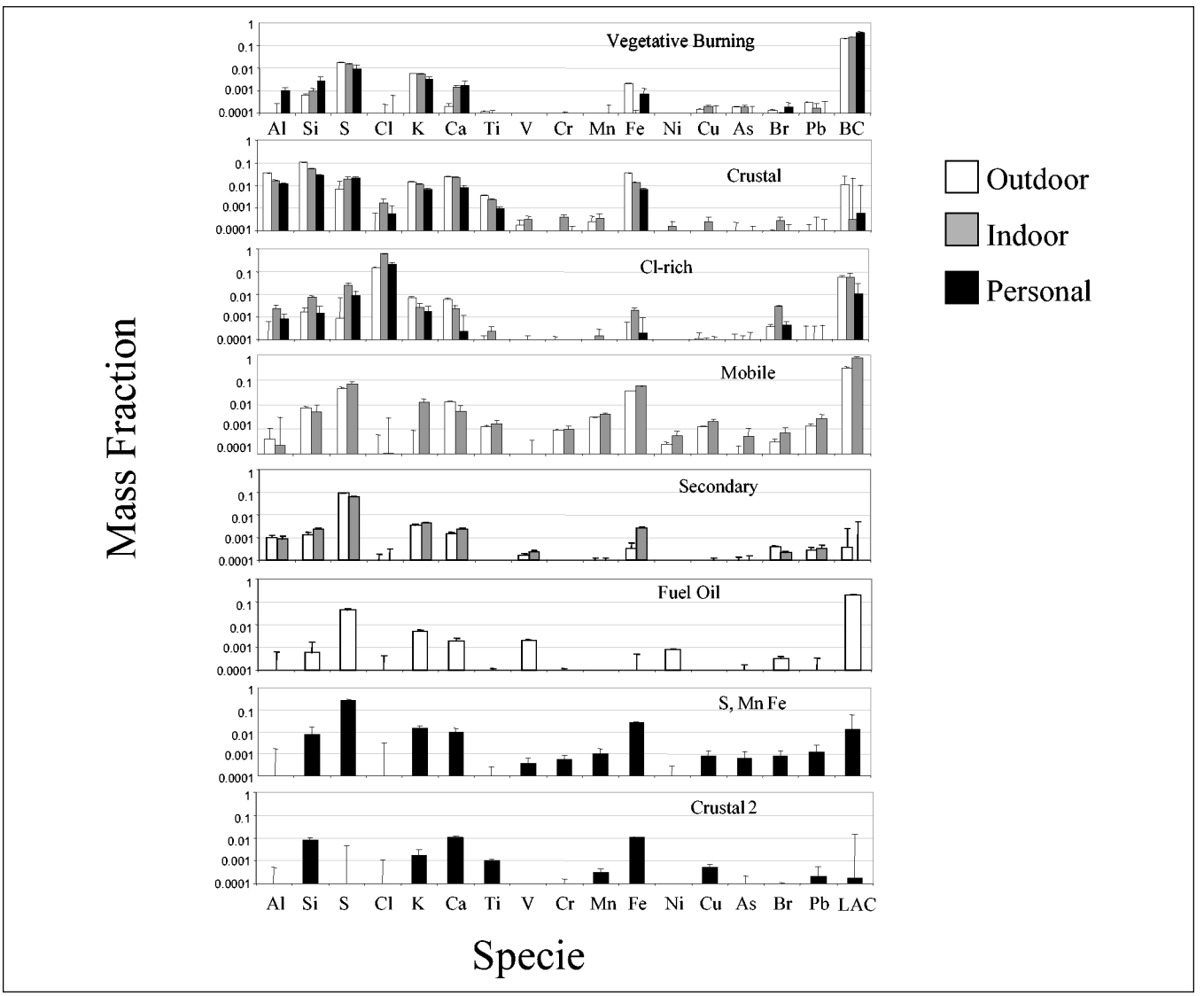

Figure 4. Source profiles derived from PMF2. Error bars reflect the 95% confidence intervals.

limit values. A number of species were not used in the PMF analyses because of the high percentage of values below the minimum detection limit. These included cobalt, gallium, germanium, selenium, barium, tin, rubidium, strontium, yttrium, zirconium, molybdenum, lead, silver, cadmium, indium, lanthanum, and mercury.

The model was run in the default robust mode to minimize the effect of outliers (outlier distance = 4). As mentioned earlier, even if the number of features is correctly chosen, the PMF2 results have some rotational ambiguity. A user-defined parameter, FPEAK, allows some fine-tuning of the derived rotations. To find the optimal PMF2 solution with the most physically reasonable results, it was necessary to run PMF2 with different numbers of sources and different FPEAK values. A value of FPEAK = 0 provided the most physically reasonable source profiles for all data sets. In addition, the global optimum of each model with FPEAK = 0 was tested. Finally, changing the seed of pseudo-random values for both the PMF2 and PMF3 models produced nearly identical source contributions and source profiles.

The PMF models were applied to a subset of the indoor and personal samples, excluding those whose mass concentration was 20% greater than the corresponding outdoor concentration. This was necessary to obtain any reasonable solution via PMF, because the PMF model cannot handle large indoor and personal contributions from sources that are variable in composition. This screening step eliminated ~15% of the indoor samples and ~30% of the personal samples. The number of simultaneous samples of outdoor, indoor, and personal PM limited the PMF3 input even more, resulting in running PMF3 with only 61 sample periods.

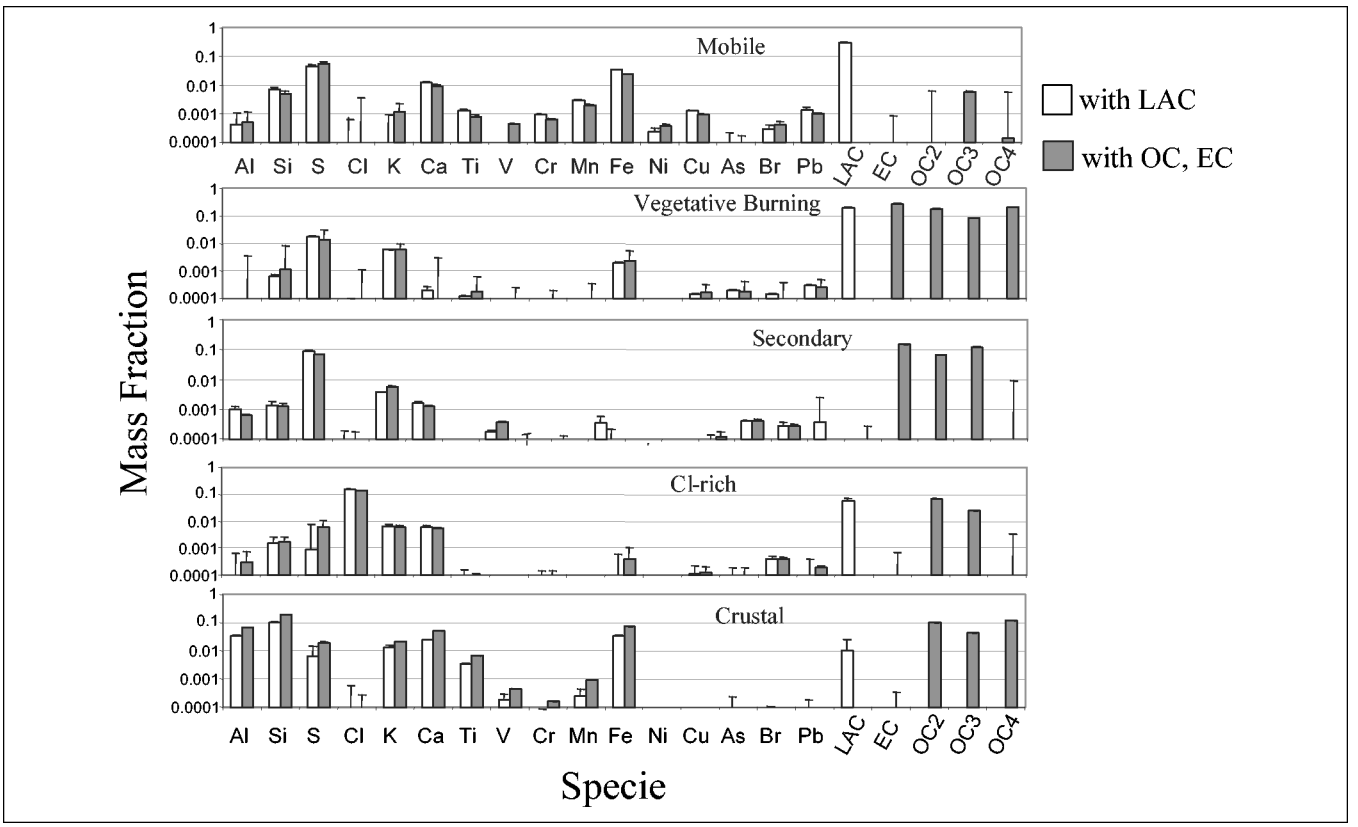

Figure 5. Source profiles for the outdoor samples derived from PMF2 using either LAC (white bars) or temperature-resolved carbon fractions (dark bars). Error bars reflect the 95% confidence intervals.

Exposure Models

Source-specific exposure models were constructed based on the microenvironmental exposure concept.[21] The total personal exposure was modeled as the sum of exposures at home indoors, at home outdoors, and outdoors away from home. Each microenvironmental exposure is a product of the fraction of time spent in that microenvironment times the average PM source estimate in that microenvironment. It is expected that exposures to PM sources originated outdoors can be better predicted than exposures to nonambient sources, such as the crustal sources. In addition, concentrations of personal sources were regressed against reported personal activities using the generalized linear model (GLM) to evaluate activities contributing to the personal sources.

RESULTS AND DISCUSSION

The predicted versus measured $PM_{2.5}$ mass concentrations are shown in Figure 3. PMF-derived source contributions are summarized in Table 2. Vegetative burning contributed the largest amount of mass to the outdoor, indoor, and personal samples (35, 49, and 62% of measured personal, indoor, and outdoor mass as predicted by PMF2), consistent with the fact that most samples were taken during the winter heating season. The average contribution from vegetative burning at the central site was 30% less than those at the home outdoor sites located throughout the city (6.1 $\mu g/m^3$ [SE = 1] compared with 8.5 $\mu g/m^3$ [SE = 1] as estimated by PMF2). This is not surprising because the central site is located near the downtown urban core, whereas the home sites were selected in wood-burning neighborhoods. The average wintertime contribution from vegetative burning at the central site is slightly higher than that previously reported (4 $\mu g/m^3$ via PMF2, SE = 1) for samples collected between 1996 and 1999 at the same location.[22]

PMF2

The source profiles are shown in Figure 4. Six factors were extracted from the outdoor samples and five factors were extracted from the less numerous indoor and personal samples. The vegetative burning, crustal, Cl-rich, mobile, secondary SO_4^{2-}, and fuel oil source profiles were readily identified by comparison with previously reported profiles. Specifically, the vegetative burning profile is similar to those reported elsewhere.[22,35–39] Relatively high levels of LAC and relatively low levels of all trace elements relative to potassium distinguish this profile. In addition, the estimated $PM_{2.5}$ contributions from vegetative burning are correlated with levoglucosan concentrations

Table 3. Correlations between daily source contribution estimates via PMF2 by source and sample type. The first row under each source lists the Pearson's correlation coefficient; values above 0.3 are shown in bold. The second row shows the corresponding *P* value.

	Home Outdoor[a]						Central Site[b]					
	Mobile	Veg Burn	2ndary	Cl-Rich	Crustal	Mobile + Secondary	Mobile	Veg Burn	2ndary	Cl-Rich	Crustal	Mobile + Secondary
						Indoor						
Mobile	**0.76**	0.17	0.03	0.1	0.17		0.03	0.17	−0.12	−0.04	−0.02	
	<0.0001	0.04	0.7	0.23	0.04		0.74	0.03	0.14	0.63	0.79	
Veg burn	0.04	**0.4**	0.02	0.11	−0.28		0.14	**0.36**	−0.02	0.16	**−0.31**	
	0.6	<0.0001	0.76	0.17	0		0.08	<0.0001	0.77	0.05	<0.0001	
2ndary	0.11	0.22	**0.67**	0.14	0.03		0.18	0.22	**0.36**	0.07	0	
	0.19	0.01	<0.0001	0.09	0.69		0.03	0.01	<0.0001	0.37	0.96	
Cl-rich	−0.02	0.06	0.17	0.06	−0.1		0.25	0.12	**0.55**	**0.42**	−0.03	
	0.78	0.5	0.04	0.47	0.24		0	0.13	<0.0001	<.0001	0.73	
Crustal	−0.15	**−0.3**	0.03	−0.07	**0.64**		−0.09	**−0.41**	−0.03	−0.16	**0.4**	
	0.06	0	0.71	0.4	<0.0001		0.24	<0.0001	0.7	0.04	<0.0001	
						Personal						
S, Mn, Fe	**0.31**	−0.03	**0.62**	0	**0.31**	**0.71**	0.01	0.1	0.28	0.05	0.04	0.24
	0.01	0.84	<0.0001	0.97	0.01	<0.0001	0.9	0.36	0.01	0.63	0.7	0.03
Veg burn	**0.32**	**0.45**	0.01	0.14	0.07	0.16	0.03	**0.35**	0.14	0.18	−0.11	0.129
	0.01	0	0.92	0.28	0.61	0.22	0.82	0.001	0.21	0.11	0.31	0.25
Cl-rich	−0.15	**0.34**	−0.1	0.04	−0.21	−0.15	0.13	0.2	**0.4**	0.3	−0.09	**0.39**
	0.25	0.01	0.45	0.73	0.11	0.23	0.25	0.08	0	0.01	0.4	0
Crustal	−0.21	−0.02	0.16	−0.14	**0.31**	0.05	0	**−0.33**	−0.13	−0.22	0.12	−0.108
	0.09	0.9	0.21	0.26	0.01	0.71	0.98	0.001	0.25	0.05	0.32	0.335
Crustal 2	**0.41**	0.14	−0.13	0.19	−0.07	0.07	0.17	−0.05	−0.03	0.04	−0.13	0.035
	0	0.29	0.31	0.14	0.6	0.6	0.13	0.67	0.81	0.72	0.24	0.754

[a]Values based on 148 indoor-home outdoor pairs and 63 personal-home outdoor pairs; [b]Values based on 161 indoor-central site pairs and 82 personal-central site pairs.

measured on a subset of outdoor samples ($r = 0.87$ compared with $-0.2 < r < 0.2$ for all other sources—we do not have levoglucosan measurements on the indoor or personal samples). The Cl-rich profile is presumably dominated by marine aerosol that is present in Seattle,[22] although sodium was not measured in this study. The relatively high abundance of Ni and V as well as SO_4^{2-} (S) and EC (LAC)[22,35–37,39] distinguish the fuel oil profile. The mobile source profile for the outdoor and indoor samples is similar to the diesel profile reported in Phoenix[36,40] and in Seattle,[22] namely high LAC, manganese (Mn), and iron (Fe). This latter profile is similar to the motor vehicle profile reported by Yakovleva et al.,[19] which was derived via PMF2 from the PTEAM data in Riverside. For comparison, Figure 5 shows the source profiles derived from the outdoor samples using LAC versus the outdoor source profiles derived using OC and EC. There is good agreement between the two sets of outdoor profiles, although using the OC/EC data set, a separate fuel oil profile was not able to be distinguished.

The secondary feature or the mobile feature on the personal samples could not clearly be resolved. Instead, as shown in Figure 4, a separate profile rich in S, Mn, and Fe (S, Mn, Fe profile) was found. This personal profile probably represents a composite of the outdoor secondary and mobile profiles, as well as contributions from cooking, as discussed later. The crustal 2 profile is similar to the soil factor reported in PM_{10} personal samples by Yakovleva et al.[19]

Table 3 summarizes the personal versus outdoor and the indoor versus outdoor pair-wise correlations by source category. Notable correlations exist for the mobile, crustal, and secondary sources between indoor and outdoor environments. In addition, as discussed later, there is a notable correlation between the personal S, Mn, Fe source and the outdoor secondary source contributions ($r = 0.62$), with an even stronger correlation observed between the personal S, Mn, Fe source and the sum of the home outdoor mobile and secondary sources ($r = 0.71$). The correlations for vegetative burning are below 0.5 in all cases. In general, the central site source contributions had much lower correlations with home indoor and personal contributions, particularly for the mobile source.

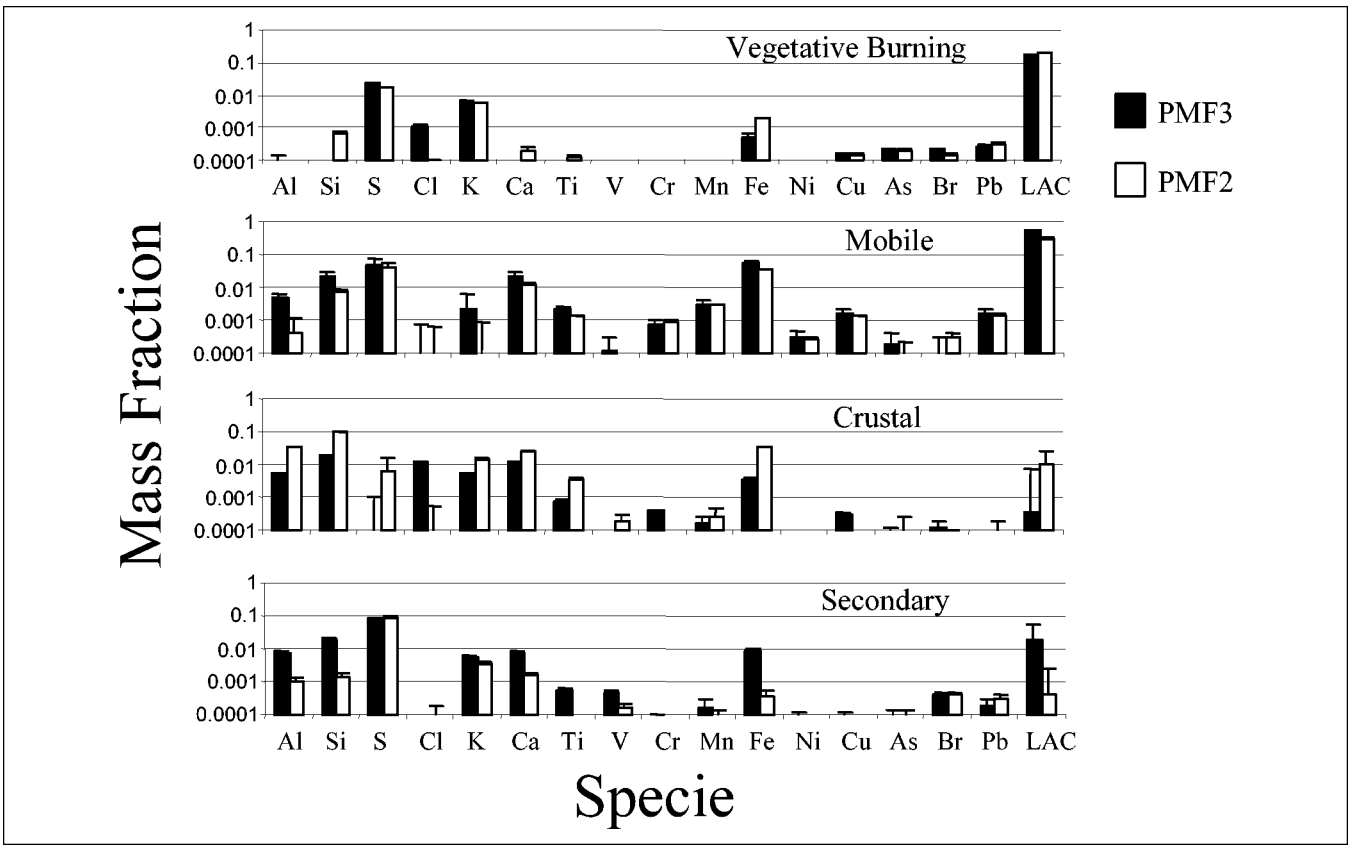

Figure 6. The four source profiles derived from PMF3 (black bars) compared with the corresponding profiles from PMF2 (white bars). Error bars reflect the 95% confidence intervals.

PMF3

Four sources common among the indoor, outdoor, and personal locations were resolved with this model (see Table 2). A five-source solution produced multiple vegetative burning profiles and was not examined further. Hopke et al.[20] used the multilinear engine[41] and resolved SO_4^{2-} (46.3%), unknown (13.6%), soil (2.8%), activity (36.2%), gypsum (0.7%), and personal care (0.4%) sources for the Baltimore apartment and personal samples, with the SO_4^{2-} source contributing 46.3% of the total personal $PM_{2.5}$ mass. Using PMF3, the following contributions to the total personal $PM_{2.5}$ mass were estimated: vegetative burning (41%), mobile (7%), crustal (33%), and secondary (19%). In Figure 6, the four PMF3-derived profiles are compared with the corresponding outdoor profiles derived via PMF2. The PMF3-derived contributions from these four sources are in reasonable agreement with the corresponding PMF2-derived values. The PMF3 results here are also in general agreement with those reported in the PTEAM study[19] in that the major outdoor sources of $PM_{2.5}$ found to also influence indoor samples are motor vehicles and secondary SO_4^{2-} formation. In addition, major impacts were found from wood burning, a source that is not as prevalent in southern California.

Note that PMF3 was better than PMF2 at resolving not only the mobile source on the personal samples but also the secondary source. This is because the secondary source is resolvable in both the indoor and outdoor samples using PMF2 and because PMF3 examines the indoor and outdoor samples together with the personal samples. This result lends support to the hypothesis that the S, Mn, Fe feature found on personal samples via PMF2 is actually most strongly associated with secondary SO_4^{2-} aerosol that is difficult to resolve using only the personal samples. However, PMF3 was unable to resolve either a Cl-rich source or a fuel oil source. The inability of PMF2 or PMF3 to resolve a fuel oil factor (with the exception of outdoor PMF2 results) is probably caused by the high percentage of below-detection-limit values for Ni and V in the indoor and personal samples compared with the outdoor samples (see Table 1). Ni and V are relatively unique tracers of the fuel oil source in Seattle and elsewhere.[22,39]

Source Contribution Ratios

Table 4 summarizes the average indoor to outdoor (I/O) and personal to outdoor (P/O) source contribution ratios as estimated by both PMF2 and PMF3. The PMF3 ratios were computed from the ratios of the c_{hk} values in eq 5 (specifically, c_{2k}/c_{1k} and c_{3k}/c_{1k} for source k, where $h = 1$,

Table 4. Average source contribution ratios predicted by both PMF2 and PMF3.

	I/O	P/O
Via PMF2[a]	n = 148 Pairs	n = 64 Pairs
Veg burn	0.54 (0.03)	0.51 (0.05)
Mobile	0.32 (0.02)	—
Secondary	0.86 (0.03)	—
S,Mn,Fe	—	1.3 (0.1)
Cl-rich	1.3 (0.4)	8 (1.5)
Crustal	2.1 (0.2)	7 (2)[b]
Via PMF3	n = 61 Pairs	n = 61 Pairs
Veg burn	0.62 (0.01)	0.39 (0.01)
Mobile	0.39 (0.02)	0.62 (0.02)
Secondary	0.61 (0.02)	0.7 (0.02)
Crustal	9 (2)	10 (2)

Note: Parentheses denote standard error; [a]Data excluded when outdoor concentration <3 standard deviations above its uncertainty; [b]Includes both crustal and crustal 2.

2, and 3 for outdoor, indoor, and personal sources, respectively). The crustal and Cl-rich features found via PMF2 had much higher average P/O ratios compared with the I/O ratios on the same subset of sample days (for the subset of 61 personal sample days, the average Cl-rich and crustal indoor/outdoor ratios were 2.7 [SE = 1.6] and 2.2 [SE = 0.6], respectively). This result suggests additional sources producing these features are associated with personal activities. However, PMF3 combined the PMF2-derived crustal and Cl-rich features into a single crustal feature that did not show the same difference in I/O versus P/O ratios (see Table 4). Therefore, although it is unclear from these PMF results whether the crustal and Cl-rich features are associated with personal activities or more general indoor sources, it is certainly clear that these two features are not strongly influenced by outdoor sources.

Because the vegetative burning and secondary sources are mostly of outdoor origin, the I/O ratios (see Table 4) provide estimates of the infiltration efficiency for PM generated from these sources. Results from the GLM showed that the PMF2-derived I/O ratios varied significantly by home ($p < 0.001$), indicating significant variation in infiltration efficiencies among homes. This is in agreement with earlier results on infiltration efficiencies using the recursive mass balance model approach.[42] The PMF2 and PMF3 I/O values for vegetative burning are comparable, but the values for the secondary source are different. The average I/O ratio for the PMF3 secondary source (0.61) is identical to the average I/O ratio calculated using the sulfur tracer method.[42]

Effects of Personal Activities

Personal source estimates were evaluated against reported activities on the subjects' TAD. The personal crustal source is related to strenuous personal activities such as sports and exercise and being in a school environment, while the additional crustal 2 source containing Si, Ca, and Fe (see Figure 4) is only associated with being in school indoor environments. For those asthmatic children who spent more hours at school, the personal crustal 2 source exposure increased (see Figure 7a). The vegetative burning source was significantly associated with burning incense and candles, which contributed to an estimated 0.04 $\mu g/m^3$ $PM_{2.5}$/min of such activities ($p = 0.001$), although only two subjects ($N = 10$ subject-days) reported this activity. For the S, Mn, Fe feature, the only significant activity influencing the level of personal exposure is cooking, contributing 0.008 $\mu g/m^3$ $PM_{2.5}$/min of cooking ($p = 0.04$; see Figure 7b). Because this feature is also associated with the secondary and motor vehicle sources, it is not clear which species are most impacted by cooking alone. Interestingly, time spent in transit was not related to any of the identified sources, possibly because of the relatively inaccurate account of in-transit minutes.

Microenvironmental Model Predictions

Personal exposure estimates to various sources based on the microenvironmental model (incorporating home indoor, home outdoor, and central source estimates as well as the fraction of time spent in the corresponding locations) are plotted in Figure 8 against the PMF2-derived personal exposure estimates. Exposures to sources originated outdoors, such as Cl-rich and S, Mn, Fe sources, were more easily predicted than exposures to sources originating in both indoor and outdoor environments. The best agreement was for exposure to the Cl-rich source ($R^2 = 0.66$), followed by the S, Mn, Fe source ($R^2 = 0.53$), the vegetative burning source ($R^2 = 0.35$), and the crustal source ($R^2 = 0.25$). Note that to estimate personal exposure to the S, Mn, Fe source, the sums of the mobile and secondary sources for indoor and outdoor microenvironmental exposures, respectively, were used in the model. By comparison, modeling of total personal $PM_{2.5}$ mass concentrations from the same subjects gave an R^2 of 0.32. By definition, the PMF3-derived indoor, outdoor, and personal source contributions for any given day are constant fractions of each other and, therefore, are not amenable to microenvironmental modeling.

CONCLUSIONS

These results indicate that there are robust features in the outdoor particle data that provide information on outdoor source contributions to indoor concentrations and personal exposures. The ability of both PMF2 and PMF3 to resolve the indoor and personal samples was aided by excluding those indoor and personal samples whose mass concentration was >120% of the corresponding outdoor

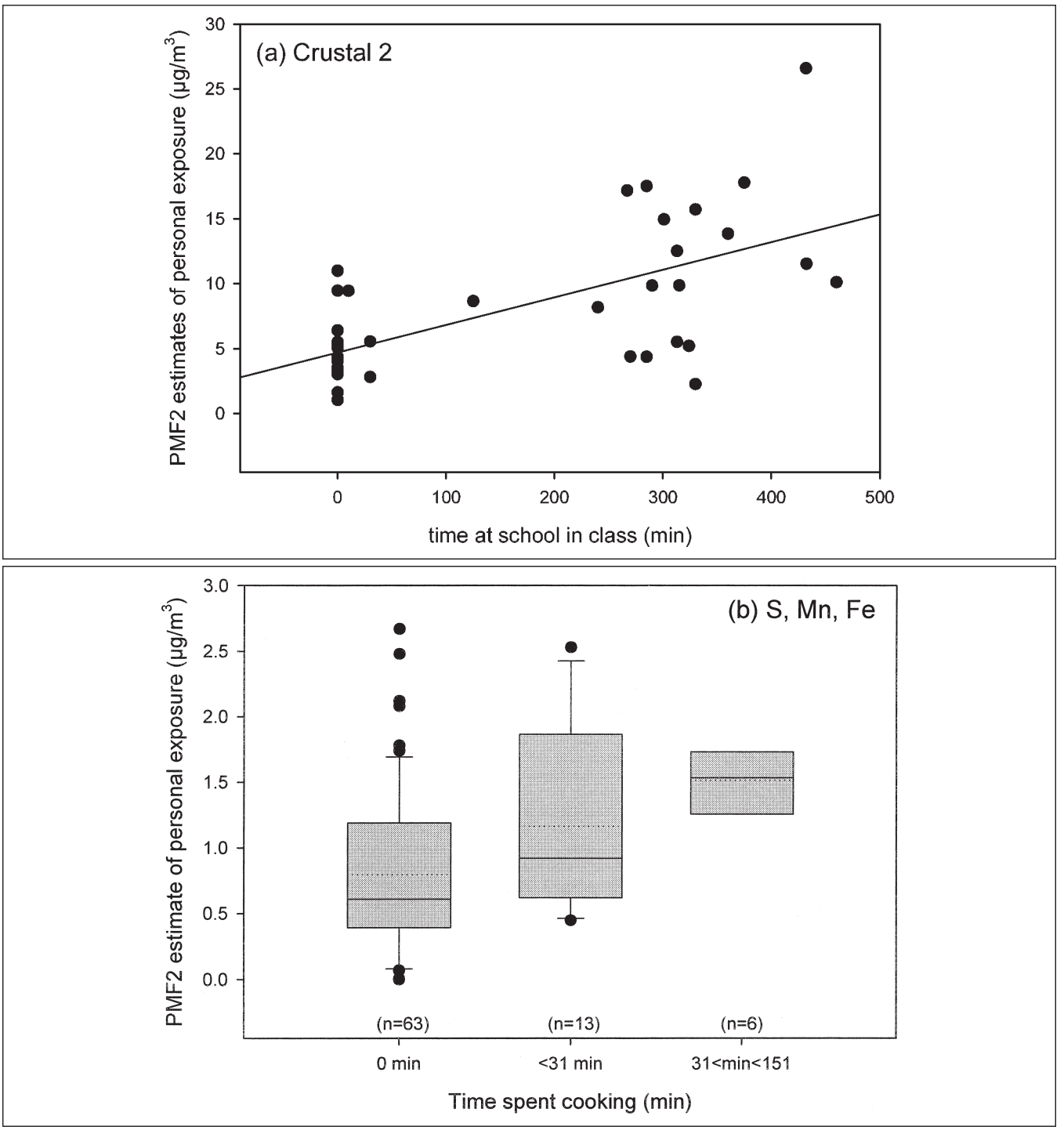

Figure 7. Significant personal activities that influence personal exposures to crustal and S, Mn, Fe sources.

sample. According to both the PMF2 and PMF3 models, vegetative burning contributed more $PM_{2.5}$ mass on average than any other source in all microenvironments. PMF2 is a useful approach in that it derives the source contribution estimates for each microenvironment independently, thereby allowing these estimates to be used in subsequent microenvironmental modeling. However, PMF3 appeared to do a better job of resolving the personal exposures to outdoor secondary and mobile sources than did PMF2. These results are based on a relatively small data set compared with most PMF analyses, especially the PMF3 data set. Further analyses of larger data sets using both PMF2 and PMF3 are warranted.

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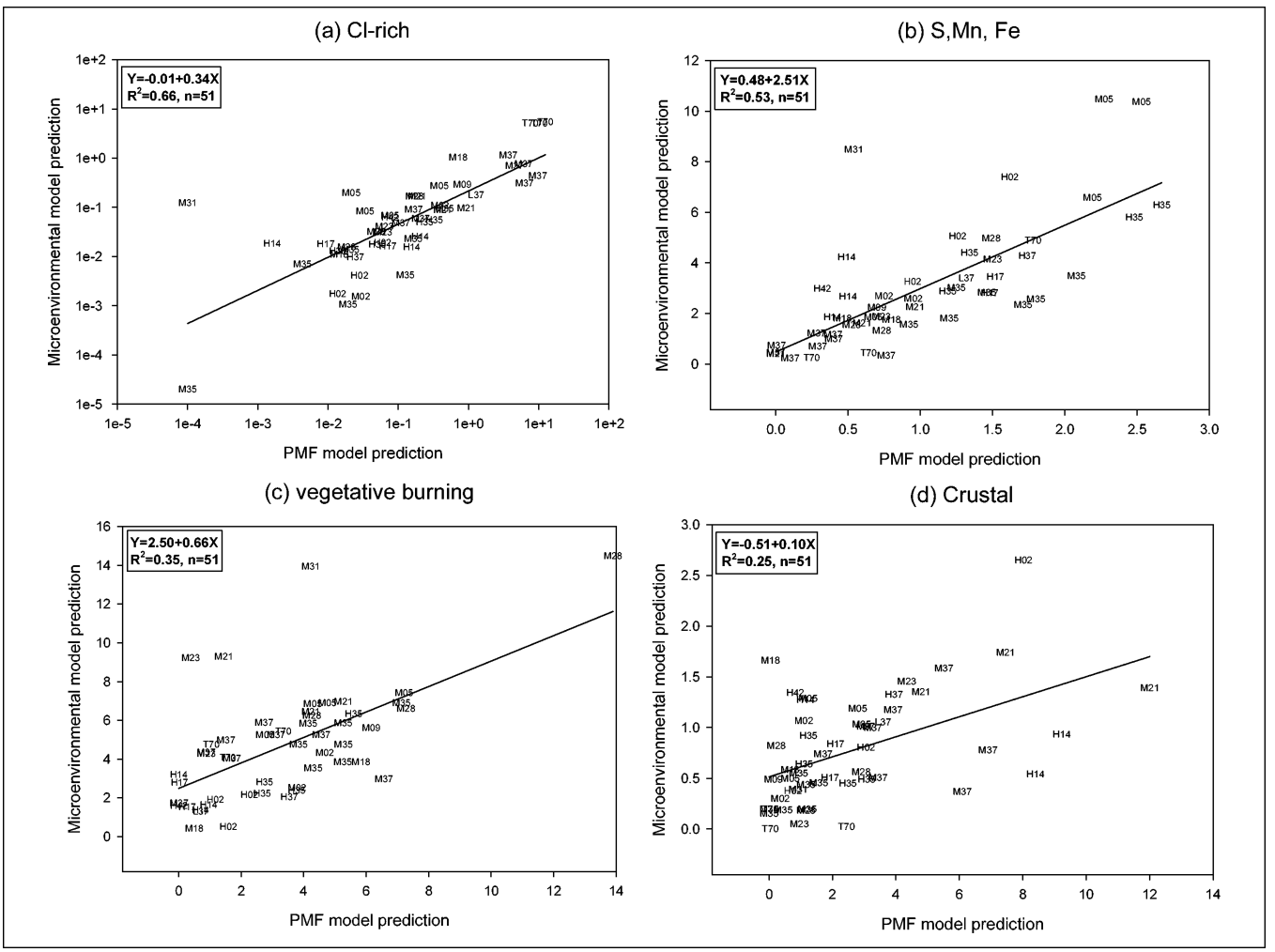

Figure 8. Personal exposures to specific PMF2 sources versus exposure estimates ($\mu g/m^3$) using the 3-microenvironmental model that includes indoor, outdoor, and central source estimates and the TAD information.

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