Receptor Modeling Assessment of Particle Total Exposure Assessment Methodology Data

ELEN YAKOVLEVA AND PHILIP K. HOPKE*

Department of Chemistry, Clarkson University, Box 5810, Potsdam, New York 13699-5810

LANCE WALLACE

U.S. Environmental Protection Agency, Environmental Photographic Interpretation Center, 12201 Sunrise Valley Drive, 555 National Center, Reston, Virginia 22092

Data from the 1991 Particle Total Exposure Assessment Methodology (PTEAM) study in Riverside, CA, were analyzed using a new receptor modeling method. In this study, ambient (outdoor), indoor, and personal particulate matter (PM) concentrations and elemental concentrations of PM_{2.5} and PM₁₀ were measured for a number of participants. These measurements made it possible to relate the pollution to which people were exposed throughout their daily activities with the outdoor air conditions. Personal daytime concentrations of the PM₁₀ and majority of elements were significantly higher than outdoor or indoor concentrations, suggesting that a significant part of personal aerosol exposure is the result of personal daily activities. Possible sources of additional particulate mass include resuspension of particles that penetrate from the outdoors and formation of new particles during cooking, smoking, etc. Positive matrix factorization analysis was performed to describe the sources of personal exposure. To identify relative contribution of different sources, regression of the particulate matter mass against the factor contributions was performed. Major sources of PM_{2.5} were oil combustion, nonferrous metal operations, and motor vehicles. The mass contributions of particles from these sources were similar for outdoor air and personal exposure. Personal exposure to particles from these sources can be controlled by changing outdoor sources. The primary source of PM₁₀ was soil. Concentrations of outdoor, indoor, and resuspended soil particles have different time dependencies. Sea-salt was a significant source only of outdoor coarse particles. A source profile of aerosols formed by personal activities such as cooking and smoking was identified. Good agreement was found between reported daily activities (smoking, cooking, vacuuming) and the related source contribution estimates.

Introduction

The health effects of airborne particles have been studied extensively since 1950s. A strong correlation between airborne

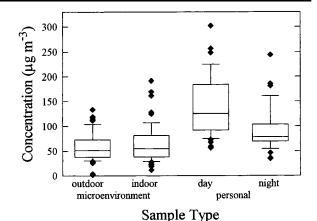


FIGURE 1. Box and whisker plots showing the distributions of outdoor, indoor, daytime, and nighttime personal PM concentrations.

particulate matter (PM) concentration and daily mortality was found during severe pollution episodes, when the estimated PM concentrations reached or exceeded $1000 \, \text{mg/m}^3$ (1930, Meuse Valley, Belgium; 1948, Donora, PA; 1952, London, UK). Subsequently a positive correlation between daily mortality and morbidity and PM was observed for low PM concentrations (1-3).

Recently, ambient (outdoor) air quality standards were promulgated to regulate PM_{10} and $PM_{2.5}$. However, since the average person spends most of their time indoors, ambient concentrations may not accurately represent personal exposure to ambient air pollutants such as airborne particles. Significant efforts will be needed to comply with ambient air quality standards and that compliance may have little or no impact on personal exposure to particles and potential health effects of particulate matter air pollution.

To measure population exposure to PM_{10} , the 1991 Particle Total Exposure Assessment Methodology (PTEAM) study was conducted in Riverside, CA (4, 5). Personal exposures to PM_{10} and indoor and outdoor PM_{10} and $PM_{2.5}$ concentrations were measured for 178 individuals statistically chosen to represent the population of Riverside (139 000). After each observation period, participants were interviewed about their daily activities. The objectives of the study were (i) to estimate the fractions of outdoor and indoor particles that were collected on personal filters, and thus determine the importance of outdoor air conditions for personal PM exposure, and (ii) to model personal exposures using information about the daily activities of participants.

Surprisingly, the population-weighted mean daytime personal PM_{10} concentration was $150~\mu g\ m^{-3},$ more than 50% higher than the mean daytime values measured inside the houses or outdoors (Figure 1). Nighttime personal exposures were similar to indoor or ambient concentrations and were significantly lower than the daytime personal exposures. The correlation between ambient PM concentrations and personal exposures was low. Elemental concentrations showed similar increases in the personal daytime monitors relative to indoor or outdoor monitors, suggesting that the additional particles had compositions similar to indoor or outdoor particles. Electron microscopy observations showed that skin flakes accounted for less than 4% of total mass, and clothes fibers were not present in measurable quantities on the majority of filters and thus were not

^{*} Corresponding author phone: (315) 268-3861; fax: (315) 268-6610; e-mail: hopkepk@clarkson.edu.

TABLE 1. Average Elemental Concentrations of $PM_{2.5}$ and PM_{10} , Percentage Increase in Personal PM_{10} Exposure Relative to Average between Indoor and Outdoor PM_{10} Concentrations

	PΝ	/l _{2.5}	PM ₁₀			
	indoor	outdoor	indoor	outdoor	personal	increase
ΑI	187.9	200.0	1858.3	2590.6	3015.3	35.55%
Si	556.2	577.1	5067.5	6506.3	8044.8	39.02%
Р	28.8	24.4	113.9	71.7	170.8	85.04%
S	1295.5	1513.1	1588.0	1816.0	1669.0	-1.94%
CI	122.6	127.6	374.9	359.0	612.9	67.03%
K	239.4	187.8	896.6	953.8	1347.1	45.60%
Ca	308.4	251.6	1872.0	1958.4	2899.1	51.37%
Ti	28.0	29.9	152.6	178.2	257.6	55.74%
V	4.9	5.6	7.7	9.6	9.0	4.05%
Cr	5.0	2.9	7.9	5.4	9.5	42.86%
Mn	8.9	11.4	30.8	44.9	46.2	22.06%
Fe	280.2	343.7	1448.4	2046.0	2290.2	31.08%
Ni	2.7	2.7	3.8	3.4	7.6	111.11%
Cu	10.6	9.1	19.3	16.4	30.5	70.87%
Zn	38.8	40.2	77.1	61.3	113.2	63.58%
Br	9.0	9.7	12.0	11.9	19.1	59.83%
Sr	4.6	4.9	12.8	15.9	17.7	23.34%
Pb	18.0	21.6	26.8	31.5	33.2	13.89%

responsible for increased PM concentrations (4). Average particle and elemental concentrations as well as increases in concentration due to the person's presence are shown in Table 1. To describe the elevated PM and elemental concentrations in the person's presence, the concept of a "personal cloud" was introduced (6). The particle is considered to be in the "personal cloud" if it would not be in the air without the person's presence and activities.

Possible sources of indoor aerosols include transport of ambient particles by penetration, and aerosols production by various personal activities indoors, such as cooking and smoking. Ozkaynak et al. (5) used stepwise regression to apportion PM concentrations to indoor and outdoor sources. Smoking and cooking were determined to be the major indoor PM sources. Smoking added $27-38~\mu g~m^{-3}$ to the PM_{10} and $PM_{2.5}$ concentrations, while cooking added $12-26~\mu g~m^{-3}$ to PM_{10} . Elements associated with cigarette smoking were potassium, chlorine, and calcium. Elements associated with cooking were aluminum, iron, calcium, and chlorine. Further modeling (5) provided more information about the source profiles, their contributions to personal exposure, and clarified the mechanisms of particle penetration indoors and collection by personal monitors.

Receptor modeling apportions the measured aerosol concentrations to their sources (7). Multivariate receptor models are based on the analysis of the correlation between measured concentrations of chemical species, assuming that highly correlated compounds come from the same source. The commonly used multivariate receptor model is principal components analysis (PCA). PCA extracts the principal components explaining the majority of variance of the data matrix that are then qualitatively interpreted as possible sources. PCA solutions are rarely physically interpretable without further transformation (rotation). The drawbacks of this technique include the inability to handle missing data and data below detection limits, difficulty of finding a satisfactory rotation, difficulty in calculating quantitative source composition, and the assumption of unrealistic weights for the variables in data matrix (8, 9).

A different approach called positive matrix factorization (PMF) has been developed by Paatero (8, 9) to address such problems. In PMF, error estimates for each data value are utilized as point-by-point weights. Such weighting allows the inclusion of uncertain data in the analysis by giving them

low weights. Constraints on the results such as the nonnegativity of the factors are integrated into the computational process. PMF has been successfully applied to the analysis of daily precipitation sample data from Finland (10), for the source identification of bulk wet deposition in Finland (11), of aerosol in Alaska (12), of Arctic aerosols (13), and of CO, PAN, PM, and O₃ at locations across the Midwestern United States (14).

In this report, the set of outdoor (ambient), indoor, and personal PM_{10} and $PM_{2.5}$ aerosol elemental concentrations from PTEAM 1991 California study were analyzed by PMF. On the basis of the calculated chemical composition of factors, the source profiles of personal aerosols were described. The origins of the aerosols formation and deposition on personal filters were derived from the comparison of PMF analysis results of outdoor, indoor, and personal filters.

Sampling and Measurements

The PTEAM study was the first large-scale probability-based study of personal exposure to particles. It was conducted in the Fall of 1991 in Riverside, CA (4, 5). Personal exposure to PM $_{10}$ was measured for 178 participants selected for study on the basis of socioeconomic stratification, after examining their screening interviews. Of 632 permanent residents contacted, 443 (70%) completed the screening interview, 257 of them were asked to participate, and 178 (70%) agreed. They represented 139 $000\pm16~000$ nonsmoking residents of Riverside, California aged 10 and above. Sampling continued during 49 days from September 22 to November 9, 1991.

Each participant wore personal exposure monitors (PEM) for two consecutive 12-h periods. The PEM design is discussed in detail by Ozkaynak et al. (15). PM₁₀ and PM_{2.5} samples were also collected with stationary indoor monitors (SIM) and stationary ambient monitors (SAM) at each home for every observation period. This resulted in 10 samples per household (day and night samples from PEM₁₀, SIM₁₀, SIM_{2.5}, SAM₁₀, and SAM₂₅). A central outdoor site was maintained during the entire period, where PM was measured by two high-volume PM₁₀ samplers (Wedding and Assoc.) and two dichotomous PM₁₀ and PM_{2.5} samplers (Sierra-Andersen), one PEM, and one SAM. Following each of the two 12-h monitoring periods, the participants answered an intervieweradministered questionnaire concerning their activities that might involve the exposure to increased particle levels (nearby smoking, cooking, gardening, etc.) and locations during monitoring time.

All filters were weighed on-site in a van with controlled temperature, humidity, and protection from vibration. Elemental concentrations were determined later by X-ray fluorescence (XRF). Fourteen elements (Al, Si, S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, Sr, Br, Pb) were present in measurable quantities on a majority of the filters. However, many of the elemental concentrations were in the range of $5-15~\text{ng/m}^3$, which can be considered as below detection limit. However, one of the advantages of PMF is its ability to permit the use of below detection limit values because of its ability to individually weight the data point and thus reduce the influence of uncertain points in the analysis (12). Thus, 18 elements could be used in the factor analyses. Mean values for the uncertainties were in a range of 7-15% of the observed concentrations.

Data Analysis

Positive matrix factorization was used for the data analysis. It analyzes the n by m **X** matrix of the measurements of n chemical species in m samples. The objective of multivariate receptor modeling is to determine the number of pollutant sources, p, the chemical composition of each source, and the amount that each of the p sources contributes to each

TABLE 2. Description of the PTEAM Data Sets Including the Number of Samples Analyzed by X-ray Fluorescence and Used in the PMF Analysis

data set no.	measurement type	size fraction	no. of measurements
1	outdoor central site	PM_{10}	195
2	outdoor central site	$PM_{2.5}$	195
3	outdoor backyards	PM_{10}	379
4	outdoor backyards	$PM_{2.5}$	350
5	indoor	PM_{10}	380
6	indoor	$PM_{2.5}$	356
7	personal	PM_{10}	370

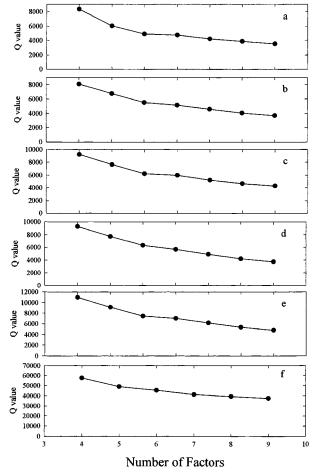


FIGURE 2. Experimental values of the goodness of fit variable, Q_1 as a function of the number of factors for (a) outdoor PM₁₀, (b) outdoor PM_{2.5}, (c) indoor PM₁₀, (d) indoor PM_{2.5}, (e) personal PM₁₀, and (f) three-way data set.

sample. The corresponding two-way factor model can be written as

$$\mathbf{X} = \mathbf{GF} + \mathbf{E} \tag{1}$$

or

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}$$
 (2)

$$i = 1, ..., n; j = 1, ..., m; k = 1, ..., p$$

where **G** is a *n* by *p* matrix of source compositions (source profiles) and **F** is a *p* by *m* matrix of source contributions to the samples. **E** represents the part of the data unmodeled by

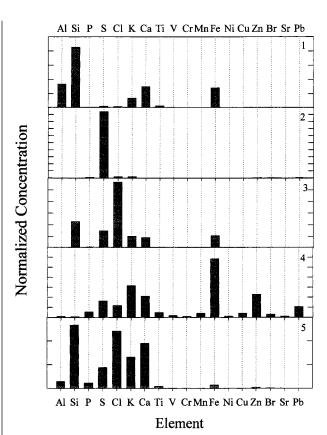


FIGURE 3. Two-way PMF combined analysis results. Factors correspond to (1) soil, (2) secondary sulfate, (3) sea-salt, (4) nonferrous metal smelters and motor vehicles, and (5) personal activities.

the p-factor model. The solution is obtained by minimizing the objective function, $\it Q$, defined as

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \frac{(x_{ij} - g_{ik} f_{kj})^{2}}{\sigma_{ij}^{2}}$$
(3)

where σ_{ij} is a user-defined error estimate for each of the matrix elements of **X**. Nonnegativity constraints are imposed on both the source profiles (**G**) and the source contributions (**F**). All of the species concentrations must be ≥ 0 , and each sample must have each source contribution ≥ 0 . The PMF algorithm then calculates **G** and **F** matrixes that minimize the value of the variable **Q** with $g_{ik} \geq 0$ and $f_{kj} \geq 0$ for k = 1, ..., p. Thus, this is a least-squares problem with the values of **G** and **F** to be determined.

This form of factorization is quite different from PCA. As shown by Paatero (δ), it is not possible to perform factorization by using singular value decomposition on such a point-by-point weighted matrix. PMF uses a unique algorithm in which resulting matrixes are varied simultaneously in each iteration step (δ). It also has the advantage of permitting the inclusion of below detection limit and truly missing data. Polissar et al. (12) by giving such points low weights to reduce the influence of the uncertain points on the analysis.

The two-dimensional factor analytic model may be extended to three dimensions in different ways. PMF three-way model can be defined by

$$\mathbf{X} = \mathbf{A} \otimes \mathbf{B} \otimes \mathbf{C} + \mathbf{E} \tag{4}$$

or

TABLE 3. Source Contribution Calculated by Two-way and Three-way PMF Followed by Multiple Regression of Factor Scores over PM

	PM_{10}			PM _{2.5}						
	personal	indoor	outdoor	indoor	outdoor					
Two-Way PMF Results										
soil	17 ± 13	33 ± 15	49 ± 6	3 ± 1	7 ± 1					
nonferrous metal operations and motor vehicle exhaust	22 ± 7	14 ± 3	19 ± 5	7 ± 1	14 ± 1					
secondary sulfate	21 ± 3	19 ± 2	30 ± 7	14 ± 2	20 ± 3					
sea-salt sea-salt	0	0	4 ± 1	0	4 ±					
personal activities	58 ± 35	14 ± 1	0	19 ± 4	0					
Three-Way PMF Results										
resuspended indoor soil	31 ± 12	0	0	0	0					
ambient soil	0	0	42 ± 6	0	15 ± 1					
indoor soil	12 ± 4	$17 \pm$	0	3 ± 1	0					
nonferrous metal operations and motor vehicle exhaust	10 ± 4	$11 \pm$	8	10 ± 3	11 ± 1					
secondary sulfate	17 ± 2	21 ± 1	22 ± 5	19 ± 3	23 ± 2					
sea-salt sea-salt	0	0	5 ± 1	0	1 ± 0.5					
personal activities	15 ± 5	15 ± 2	0	11 ± 5	0					

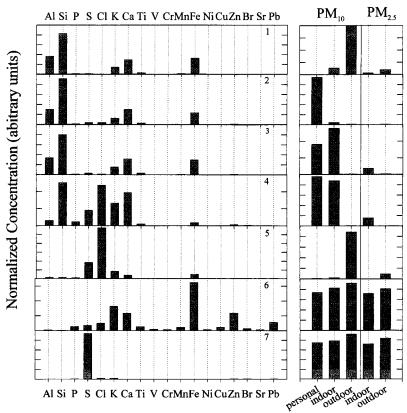


FIGURE 4. Factor profiles (left) and distribution between measurement types (right) of seven-factor three-way PMF model. Factors correspond to (1) ambient soil, (2) resuspended indoor soil, (3) indoor soil, (4) personal activities, (5) sea-salt, (6) motor vehicle, nonferrous metal smelters, and (7) secondary sulfate.

$$x_{ijh} = \sum a_{ik}b_{jk}c_{hk} + e_{ijh}$$

$$i = 1, ..., n; j = 1, ..., m; h = 1, ..., q; k = 1, ..., p$$
(5)

where **X** is a $n \times m \times q$ three-way data array, **A**, **B**, **C** are the resulting two-way factor matrixes for each of the three modes, \otimes indicates a Kronecker product, and **E** is the matrix representing the unmodeled part of **X**. On the basis of extended nonnegativity constraints and minimization criteria, PMF can solve the trilinear model (*9*). This approach has been applied to particulate composition data by Xie et al. (*13*).

Results and Discussion

For the comparison of the aerosol sources in outdoor, indoor, personal fine and coarse samples, seven elemental concen-

trations data sets were constructed and analyzed by PMF (Table 2). The central site SAM measurements were very similar to the backyard SAM measurements and, therefore, produced the same results.

A critical step in PMF analysis is determination of the number of factors. Analysis of the goodness of fit variable, Q, defined in eq 3, can be used to help determine the optimal number of factors. Assuming that reasonable estimates of the individual data point uncertainties are available, then fitting each value should add one to the sum, and the theoretical value of Q should be equal to the number of data points in the data set. However, the resulting solutions also have to make physical sense within the system being studied. Figure 2 show the experimental values of Q. On the basis of this analysis, five factor solutions were chosen for these two-way data sets.

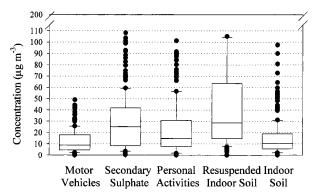


FIGURE 5. Source contributions to personal exposure to PM₁₀ calculated by three-way PMF followed by regression.

Each of the two-way data sets was analyzed separately, and typical results from these analyses will be presented. Two-way PMF identified five sources: "soil", with high concentrations of Al, Si, K, Ca, Ti, Fe; "secondary sulfate" (S); "sea-salt" (Cl), "nonferrous metal operations and motor vehicle exhaust" (Pb, Br, Zn, Fe, Mn), "personal activities" (Cl, Si, K, Ca) (Figure 3). Wienke et al. (16) had also observed the effects of a nonferrous metal source largely associated with zinc in their analysis of the 1987 SCAQS PM2.5 trace element data. The exact nature of this source is uncertain. It was not possible to obtain separate factors for motor vehicles and nonferrous metal operations. This result may be because a large fraction of the trace metal concentrations was below the detection limit. Particles from those sources were found mainly in the fine mode. The "personal activities" source appeared only for indoor and personal data sets. The profile for this factor (chemical composition of particles) cannot be directly related to any outdoor sources. Thus, it was assumed that those particles were generated by personal activities such as cooking and smoking. Contributions of those sources to collected PM were calculated by regression of factor contributions against the individually measured PM mass values (Table 3). Outdoor sources apportionment showed good agreement with the results of the Chow et al. (17) study that was performed in Riverside in 1988. Although two-way PMF analysis provided some understanding of the PM sources in the system, it failed to provide a direct connection between ambient conditions and personal exposure to particles from different sources.

Simultaneous measurements of personal PM_{10} and indoor and outdoor PM_{10} and $PM_{2.5}$ during the Riverside study made it possible to analyze the data as a three-way problem. The three-way array was constructed with the following modes: (1) elemental concentrations (18 variables), (2) samples (303 measurements), (3) sampling type (personal PM_{10} , indoor PM_{10} , outdoor PM_{10} , indoor $PM_{2.5}$, outdoor $PM_{2.5}$). The three-way PMF solution for these data yielded both factor profiles and their contributions and in addition provided the

distribution of each source between personal/indoor/outdoor fine/coarse PM, allowing the analysis of the influence of outdoor sources on personal particulate exposure.

Examination of the Q values (Figure 2f) does not provide a clear indicator of the number of factors to use. After exploring various solutions, it was found that a seven-factor solution provided the best for the three-way model in terms of the quality of the fit and the interpretability of the resulting factors. The seven-factor, three-way PMF source profiles and sample type contributions are shown in Figure 4. Factor contributions were determined by the regressions of PM concentrations against factor contributions. The results are summarized in Figure 5 and Table 3.

Three factors were found to have the same composition and was the same as the soil source as identified by the two-way analysis. In the three-way analysis, several factors can have identical source profiles as long as vectors in samples—factors and sampling type—factors matrixes are different. Equivalent profiles indicate that particles initially were produced by the same source, such as by wind suspending soil particles in the air.

Three soil factors with equivalent profiles but different time dependence and distribution between indoor/outdoor/ personal samples were found by PMF. These factors can be explained by the existence of three independent sources of soil particles. The first factor has high contributions to outdoor coarse PM only. Thus, it can be interpreted as the typical nature of soil particles in the ambient environment. The second factor has high concentrations in personal and indoor samples and thus can be considered to be indoor soil dust (that can penetrate indoors on shoes and clothes). Particles from the third soil source appeared in personal samples only and that led to the assumption that those particles were resuspended by various personal activities inside the house such as moving. From the results of the three-way PMF analysis, comparing source contributions to indoor and personal PM₁₀ resuspended soil particles was the major portion of the "personal cloud".

The time variations for the three soil particle sources showed little correlation (Figure 6), indicating that the outdoor concentration of soil particles was not directly related to indoor or personal exposure to soil particles. Average contributions of "indoor soil" and "resuspended indoor soil" to personal PM were 15% and 30%, respectively.

The "personal activities" particle source appeared in indoor and personal samples. Those particles were generated by everyday indoor personal activities, such as cooking, smoking, and using personal hygiene sprays. The derived composition is characteristic of the mixture of particles from those sources. From the regression analysis, the contribution of those particles to personal PM concentrations was about 15%

The sea-salt source was identified by high chlorine concentrations and its occurrence mainly in the outdoor

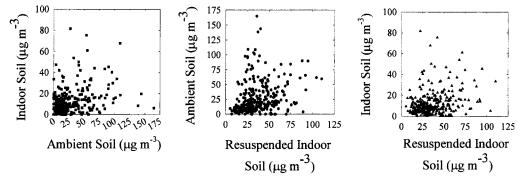


FIGURE 6. Comparison of source contributions of the three soil sources.

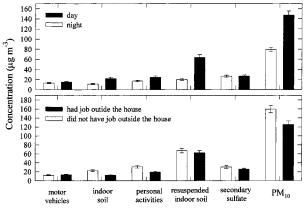


FIGURE 7. Comparison of source contributions to personal PM_{10} (a) for daytime and nighttime monitoring periods and (b) for participants who worked outside the house with those who did not.

coarse particulate mass. Coarse particles do not penetrate indoors effectively and thus do not directly affect personal exposure.

As it was shown earlier, the "indoor soil", "resuspended indoor soil", and "personal activities" contributions to the personal PM_{10} did not depend on the outdoor air conditions. Thus, 60% of personal PM_{10} particle mass was not controlled

by changing outdoor conditions. This conclusion should not be automatically applied to other geographical locations, seasons of the year, weather conditions, etc.

Particles from "secondary sulfate" and "nonferrous metal operations and motor vehicles exhaust" had similar concentrations for all of the sample types in both the fine and coarse mode. In addition, the time variations for those two sources were similar in the personal/indoor/outdoor filters. These two outdoor sources produce mainly fine particles that can more readily penetrate into buildings. Thus, equilibrium can be established between personal/indoor/outdoor environment, resulting in equal amount of particles in all sampling types that are directly related to the ambient concentration of those particles. On average, the "secondary sulfate" source contributed 25% to the personal PM while "nonferrous metal operations and motor vehicles exhaust" contributed 10%.

One of the purposes of the PTEAM study was to relate personal aerosol exposure to participants daily activities. The three-way PMF results were compared with the questionnaire information about activities during each observation period. The results of the comparison are presented in Figures 7–9. Bars show the mean value for each group of participants and the error of the PMF followed by the regression calculations.

A comparison between daytime and nighttime factor contributions is shown in Figure 7 (top). Scores for "personal

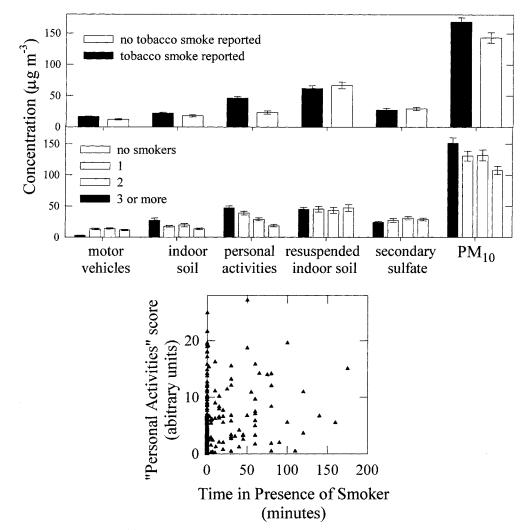


FIGURE 8. Effect of tobacco smoke: (a) comparison of source contributions to personal PM_{10} for participants who were present in an environment with tobacco smoke with those who did not, (b) comparison of source contributions to personal PM_{10} for participants with different number of smokers in the household, and (c) influence of the duration of smoking during the monitoring period on "personal activities" factor score.

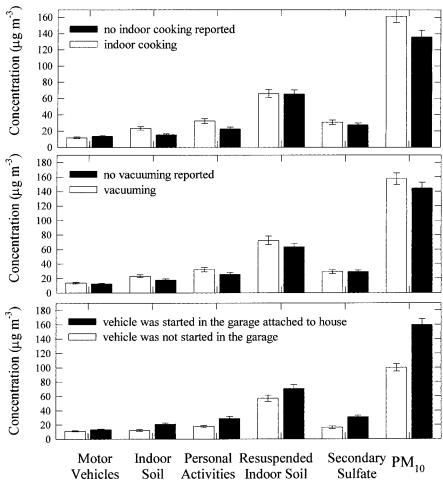


FIGURE 9. Comparison of source contributions to personal PM₁₀ for participants exposed to indoor sources: (a) indoor cooking, (b) vacuuming, and (c) starting vehicle in the garage attached to house.

activities", "indoor soil", and "resuspended indoor soil" showed large differences between daytime and nighttime values. Scores for "secondary sulfate" and "nonferrous metal operations and motor vehicle exhaust" were similar during the day and night. Those results confirm that "personal activities", "indoor soil", and "resuspended indoor soil" sources created the personal cloud and were dependent on daytime personal activities, and "secondary sulfate" and "nonferrous metal operations and motor vehicle exhaust" sources contributions to PM were determined by outdoor conditions and not personal activities.

According to previous results (5), people who stayed in their houses were exposed to more personal cloud pollutants then people who worked. Figure 7 (bottom) demonstrates the increase in personal cloud sources for participants who did not work outside the house. The increase in PM was due to an increase in particles from "indoor soil" and "personal activities" sources.

Many previous studies have identified tobacco smoke as a major indoor pollutant. It was expected that smoking would affect "personal activities" factor contribution and produce high concentrations of Cl, Ca, K. Parts a and b of Figure 8 show the relationship of the source contributions with questionnaire answers about indoor tobacco smoking. The "personal activities" factor contribution showed an excellent correlation with the number of smokers in the house and smoking during the monitoring period. Figure 8 (bottom) shows the comparison of the "personal activities" factor contribution with the minutes of smoking reported by participant during the monitoring period. The poor correlation may be the result of several critical problems: (i)

participants very subjectively estimated the duration of smoking, or (ii) minutes of smoking is not a good indicator of PM from smoking collected by the personal samplers.

Cooking is also a major aerosol source. Figure 9a displays the source contribution in houses where indoor cooking was performed during the monitoring period versus houses where no cooking occurred. "indoor soil" and "personal activities" increased equally, showing that cooking changed the whole indoor environment.

Vacuuming appears to be a major source of resuspended particles in the house. Figure 9b shows that it affected all personal cloud sources, and thus, vacuuming affected the entire indoor environment by resuspensing indoor particles. The "resuspended soil" source was affected by vacuuming in the same manner as "indoor soil" and "personal activities", suggesting that "resuspended soil" may have come primarily from participants clothes and shoes.

Another interesting result came from the comparison of houses where the vehicle was started in the garage attached to house with ones where it was not (Figure 9c). Presumably, personal exposure was increased when participants went to the garage (one of the dustiest parts of a house) a number of times during monitoring period.

Many of questionnaire answers were not related to PM and factor contribution values. Those questions included participants gender, pets, dryer installed in the house, closeness to the road or driveway, household income, etc. A problem in the analysis of the questionnaire data was the small number of participants in a given category (for example outdoor cooking was done only in three houses, and painting was done only in one house).

Concluding Remarks

Major aerosol sources of personal PM_{10} exposure in Southern California in Fall 1991 were the resuspension of the soil particles, generation of the particles by personal activities, and penetration of fine particles from the outdoors, such as emissions from oil combustion or nonferrous metal operations and motor vehicle exhaust. Sources of coarse particles such as sea-salt or ambient soil did not directly influence indoor air conditions and personal exposure to PM_{10} . Sources of fine particles such as secondary sulfate, oil combustion, and nonferrous metal operations and motor vehicle exhaust had similar impacts on outdoor, indoor, and personal PM_{10} . Personal exposure to particles from these sources can be controlled by changing outdoor sources. A major portion of personal exposure to PM_{10} originated indoors and cannot be controlled by changing the outdoor air quality.

Good agreement was obtained between information about daily personal activities and values of factor scores. Smoking, indoor cooking, and vacuuming resulted in elevated "personal activities", "indoor soil", and "resuspended indoor soil" factor scores but did not affect "secondary sulfate" and "nonferrous metal operations and motor vehicle exhaust".

Acknowledgments

The authors would like to thank Dr. Pentti Paatero for his assistance and discussion of the analysis of these data.

Literature Cited

- Dockery, D. W.; Schwartz, J.; Spengler, J. D. Environ. Res. 1992, 59, 362-373.
- (2) Schwartz, J. Public Health Rev. 1992, 19, 39-60.
- (3) Dockery, D. W.; Pope, C. A., III.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G., Jr.; Speizer, F. E. N. Engl. J. Med. 1993, 329, 1753-1759.

- (4) Ozkaynak, H.; Xue, J.; Weker, J.; Koutrakis, P.; Pellizzari E.; Wallace L. The Particle TEAM (PTEAM) study: Analysis of the Data. Final Report. Vol. 1. U.S. EPA: Research Triangle Park, NC. 1994.
- (5) Ozkaynak, H.; Xue, J.; Weker, J.; Koutrakis, P.; Pellizzari E.; Wallace L. The Particle TEAM (PTEAM) study: Analysis of the Data. Final Report. Vol. 3. U.S. EPA, Research Triangle Park, NC. 1994.
- (6) Rodes, C. E.; Kamens, R. M.; Wiener, R. W. Indoor Air 1991, 2, 123–145.
- (7) Hopke, P. K. Receptor Modeling for Air Quality Management, Elsevier: Amsterdam, 1991.
- (8) Paatero, P. Chemom. Intell. Lab. Syst. 1997, 37, 23-35.
- (9) Paatero, P. Chemom. Intell. Lab. Syst. 1997, 38, 223-242.
- (10) Juntto, S.; Patero, P. Environmetrics 1994, 5, 127-144.
- (11) Anttila, P.; Paatero, P.; Tapper U.; Jarvinen O. Atmos. Environ. 1995, 29, 1705–1718.
- (12) Polissar, A. V.; Hopke P. K.; Paatero, P. A.; Malm W. C.; Sisler J. F. J. Geophys. Res. 1998, 103, 19, 045-19, 058.
- (13) Xie, Y. L.; Hopke P. K.; Paatero P.; Barrie L. A.; Li S. M. *J. Atmos. Sci.* **1999**, *56*, 249–260.
- (14) Paterson, K. G.; Sagady, J. L.; Hooper, D. L.; Bertman, S. B.; Carroll, M. A.; Shepson, P. B. *Environ. Sci. Technol.* **1999**, *33*, 635–641.
- (15) Ozkaynak H.; Xue J.; Spengler J.; Wallace L.; Pellizzari E.; Jenkins P. J. Expos. Anal. Environ. Epi. 1996, 6, 57–78.
- (16) Wienke, D.; Gao, N.; Hopke, P. K. Environ. Sci. Technol. 1994, 28, 1023–1030.
- (17) Chow, J. C.; Chung Shing Liu; Cassmassi J.; Watson, J. G.; Zhiqiang Lu; Pritchett, L. C. Atmos. Environ. 1992, 26A, 693–706.

Received for review November 2, 1998. Revised manuscript received May 3, 1999. Accepted June 7, 1999.

ES981122I