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Identifying the PCA-Resolved Components by Source tracer Elements

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ABSTRACT

An indoor and outdoor source apportionment study was conducted to quantify fine particulate matter PP 205 sources. The data consist of mass and elemental composition of PM2.5 samples collected inside 70 smokers, 70 non-smokers homes, and at an outdoor site in Tehran, Isfahan and Gilan provinces, two communities participating in a large health effects study. The data were divided into winter and summer seasons and analyzed separately. The apportionment results were used to reconstruct the contribution of various sources to the measured PM2.5 mass concentrations. Tehran results show that sulfur-related, auto-related and soil sources contribute to PM2.5 levels indoors. In Isfahan, an outdoor composite source and soil impacts were identified during winter months. In summer, the resolved outdoor composite, sulfur-related and auto-related sources showed significant contributions to indoor PM2.5 mass. Additionally, a wood-smoke source was resolved during winter months in both the places. The PCA analyses of smokers homes show that the resolved tobacco smoke is the major contributing PM2.5 source.

Keywords: PM2.5 source, Indoor outdoor composite, Sulfur-related, Auto-related, Aerosols.

INTRODUCTION

People are exposed to a variety of indoor and outdoor pollutants. The pollulant concentrations and the relative contributions of various sources may also vary across seasons. Atmospheric dispersion chemical reaction rates, and permeability of indoor enclosures are some of the factors that contribute such seasonal variations. In order to quantify the relative contribute of fine particulate mat matter sources, principal component analysis (PCA) was applied to indoor and outdoor samples collected to two communities. The communities, Isfahan, are contrasted by the level of urbanization and industrialization: the former being industrial while the latter is primarily agricultural. In this work, the sources of indoor and outdoor fine particulate matter PM2.5 have been identified and apportioned using PCA. The analysis was carried out separately for winter and summer seasons. PCA was applied to indoor data separated on the basis of reported smoking habit (yes/no) of its inhabitants. The details of indoor analyses and outdoor analyses have been presented elsewhere [1,2].

MATERIALS AND METHODS

The indoor and outdoor samples were collected for a one year period during 2008-09 at Emam Hosein and Shush. These two communities are participants in Municipality Air Pollution station [2]. The samples were divided into winter [December, 2008 to March, 2009] and summer [June-September 2009] seasons. Indoor monitoring consisted of week-long samples collected at 70 smokers and 70 non-smokers homes once during winter and summer, respectively. Twenty-four hour PM2.5 samples were collected at an outdoor receptor site in each place. The samples were analyzed for their elemental content using x-ray fluorescence [3]. Details of QA/QC procedures applied to the data se are presented elsewhere [4].

PCA was applied to identify and apportion the possible source. PCA is a multivariate statistical technique commonly used to reduce the dimensionality of a set of inter-correlated variable into a smaller number of principal components. In this study, the elemental correlations about their means were analyzed using varimax rotate PCA. The method resolved the observed variance into possible components attributable to source. The mass and elemental contributions of each resolved source were also estimated using regression of absolute principal component scores [5,6].

RESULTS

The PCA resolved components were identified using dominant source tracer elements that were associated with each of the resolved component. Si and Al have found to be suitable tracers of soil emission [7] Many studies [8,9] have reported that Pb, Br and Zn are found in automobile exhaust, and tire dust emissions resulting from transportation-related activities. Elemental sulfur was attributed to possible impacts from sulfur-related sources. In Tehran ambient iron and steel production emission have been identified using Fe as a tracer element [10]. The sources lised so far are predominantly outdoor origin. Therefore, published source emission compositions could be directly used for the identification of the resolved component (s).

In the PCA analyses, the attribution of principal component (s) to indoor sources required additional investigation. For this purpose, the concentration ratios between the dominant element (s) in the resolved component and an outdoor source element were computed. The ratios, presented in table 1, were calculated with reference to S and Pb which are predominantly of outdoor origin. If we assume that there are no indigenous indoor sources for lead and sulfur, then the computed rations should show higher values for elements of indoor origin.

Indoor PCAs resolved two components aligned with potassium and calcium respectively. Elemental potassium has been associated with wood smoke emissions from fire places and wood-stoves [11,12]. However, in Tehran and Isfahan the indoor PM2.5 and K concentrations were significant higher in smokers' homes than non-smokers' homes. We also observed that the K/S, and K/Pb ratios were higher for smokers' homes than non-smokers' homes by a factor of two (Table 2). The major distinction between these two home groups is the smoking habit of the residents. These finding suggest that tobacco smoke source may be partially contribute to PM2.5, and K levels measured in smokers' homes. During summer, with the absence of wood-burning activities, the resolved K-related principal component for the smokers' homes was attributed to tobacco smoke source. The sources of indoor calcium are intriguing. Table 1 shows that Ca/S, and Ca/Pb ratios were higher indoors (both for smokers' and non-smokers' homes) than outdoors. Also the ratios were found to be similar for smokers' and non-smokers' homes. Calcium is a

crustal element associated with soil emissions. If the indoor elemental Ca were due to soil impacts, Al and Si would also display alignment with the resolved principal component. But this was not the case in our indoor PCAs. These evidences suggest that the resolved Ca-related component may be indoor oriente.

Table 1. Selected elemental ratios for winter and summer months

Ratio	Isfahan Province			Tehran Province		
	Smokers' homes	Non-smokers' home	Outdoor site	Smokers' homes	Non-smokers' home	Outdoor site
Season: Winter						
K/S	0.288	0.146	0.077	0.524	0.208	0.052
Ca/S	0.057	0.047	0.040	0.110	0.147	0.021
Br/S	0.006	0.005	0.004	0.005	0.0026	0.0024
Fe/S	0.109	0.109	0.113	0.057	0.053	0.025
Si/S	0.111	0.116	0.114	0.139	0.174	0.072
Zn/S	0.036	0.035	0.029	0.022	0.024	0.011
S/Pb	30.7	25.8	37.6	65.5	61.9	102.
K/Pb	9.21	3.91	2.91	35.2	12.6	5.33
Ca/Pb	1.76	1.30	1.40	7.43	10.7	2.17
Br/Pb	0.18	0.13	0.16	0.35	0.15	0.25
Fe/Pb	3.42	2.84	0.11	3.83	3.22	2.60
Si/Pb	2.89	3.02	3.74	9.55	11.2	7.22
Zn/Pb	1.11	0.93	1.09	1.43	1.44	1.09
Season: Summer						
K/S	0.108	0.054	0.047	0.141	0.052	0.042
Ca/S	0.023	0.022	0.014	0.042	0.052	0.024
Br/S	0.002	0.001	0.002	0.002	0.001	0.0016
Fe/S	0.069	0.065	0.057	0.038	0.037	0.031
Si/S	0.067	0.069	0.066	0.118	0.136	0.116
Zn/S	0.019	0.017	0.017	0.009	0.009	0.006
S/Pb	91.5	89.2	58.9	165.	161.	143.
K/Pb	10.9	5.41	3.36	19.2	8.40	5.81
Ca/Pb	2.13	2.14	1.01	6.49	8.72	3.39
Br/Pb	0.19	0.11	0.12	0.31	0.18	0.24
Fe/Pb	6.16	5.85	4.01	6.38	6.06	5.02
Si/Pb	6.14	6.22	4.59	19.6	22.5	18.3
Zn/Pb	1.67	1.65	1.16	1.38	1.46	0.87

Table 2. Reconstructed source contributions for Isfahan province

Source	*Smokers' homes*	*Non-smokers' homes*	*Outdoor site*
	Winter		
Soil	7.9 (3.45)	17.6 (3.45)	9.6 (1.79)
Wood smoke	9.5 (4.15)	21.2 (4.15)	23.0 (4.31)
O.C.-I	10.3 (4.47)	22.9 (4.47)	24.8 (4.65)
Tobacco smoke	45.6 (19.9)	n/a	n/a
Unexplained	26.7 (11.6)	38.3 (7.47)	42.6 (7.95)
Total measured	100 (43.57)	100 (19.54)	100 (18.7)
	Summer		
Sulfur-related	17.8 (8.90)	33.3 (8.23)	52.5 (15.5)
Auto-related	7.3 (3.65)	14.8 (3.65)	5.3 (1.55)
O.C.-II	8.8 (4.40)	16.5 (4.07)	26.0 (7.67)
Tobacco smoke	53.7 (26.8)	n/a	n/a
Indoor dust	7.4 (3.70)	15.0 (3.70)	n/a
Unexplained	5.0 (2.4)	20.4 (5.05)	16.2 (4.78)
Total measured	100 (49.85)	100 (24.7)	100 (29.5)

Additionally, calcium concentration at the outdoor site was found to be lower than that indoors (I/O>1). Therefore we tentatively term this component indoor dust possibly due to general house he dust. Application of PCA separately for smokers' and non-smokers' homes resolved principal component attributable to sources of indoor and outdoor origin accounting for more than 80% of the observed variance. To estimate the source contributions, the observed PM2.5 mass concentrations were regressed against the absolute principal component scores. But in smokers' homes, the resolved labacco smoke-related source was found to be the only significant contributor to the mean measured PM2.5 concentration. The workers [13] reported larger variance in the measured respirable particular mass associated with to bacco smoking. The between home variation in PM2.5 concentration was also found to be large for homes with possible tobacco smoke source. Thus in the regression analysis, PM2.5 contribution of the lesser sources could not be estimated for the smokers' homes. To overcome this limitation we assumed that smoker and non-smoker homes behaved similarly with respect to the penetration of outdoor pollutants and the generation of other non-tobacco pollutants. Therefore the resolve outdoor sources from non-smokers' homes PCA were used to reconstruct the source contributions for smokers' homes. The concentration ratios for elements of outdoor origin (Al, Si, Pb, Si and Zn) were found to be similar for smokers' and non-smokers homes (Table 1). These results may support our assumption outlined above. The reconstructed source contributions for Tehran and Isfahan are presented in table 2 and 3 respectively.

Table 3. Reconstructed source contribution for Tehran Province

Source	*Smokers' homes*	*Non-smokers' homes*	*Outdoor site*
	Winter		
Sulfur-related	13.2 (4.56)	30.7 (4.56)	39.2 (4.04)
Auto-related	5.1 (1.78)	12.0 (1.78)	17.3 (1.78)
Soil	3.8 (1.31)	8.8 (1.31)	13.4 (1.38)
Tobacco smoke	71.0 (24.6)	n/a	n/a
Wood smoke	2.7 (0.94)	6.3 (9.4)	13.0 (1.34)
Unexplained	4.2 (1.38)	42.2 (6.23)	17.1 (1.80)
Total measured	100 (34.6)	100 (14.8)	100 (10.3)
	Summer		
Sulfur-related	23.3 (5.80)	38.1 (5.30)	45.8 (6.23)
Auto-related	18.1 (4.50)	29.6 (4.12)	35.6 (4.84)
Soil	7.5 (1.86)	13.4 (1.86)	16.5 (2.25)
Tobacco smoke	40.1 (9.99)	n/a	n/a
Unexplained	11.0 (2.75)	18.9 (2.62)	2.10 (0.28)
Total measured	100 (24.9)	100 (13.9)	100 (13.6)

All figures in % (μgm^{-3}); O.C.-I: Iron & steel, and auto-related sources; O.C.-II: Iron & steel, and soil sources

DISCUSSION AND CONCLUSION

From indoor PM2.5 measurements in these two places with very different ambient sources, we found that the mean PM2.5 concentrations were higher for smokers' homes than non-smokers' homes by a factor of two. The smokers' home PM2.5 concentrations were also higher than similar outdoor concentrations. The application of PCA to elemental composition data attributed much of this excess PM2.5 mass to the resolved tobacco smoke source. In analyses stratified by season, we found that tobacco smoke contributed between 40 and 70% of the observed PM2.5 levels inside smokers' homes in Isfahan and Tehran. It is clear that targest indoor source is associated with tobacco smoke.

Surfur-related sources contribute a major fraction of the PM2.5 mass in non-smokers' homes. In Isfahan the estimated concentration of this source was 33.3% during summer months. In Tehran, sulfur-related sources contribute 30.7 and 38.1% of the measured PM2.5 mass concentrations in winter and summer, respectively. The resolved wood smoke source showed higher contributions for Isfahan than Tehran homes ($4.15\mu gm^{-3}$ Vs $0.94\mu gm^{-3}$). In Tehran we observed that soil related emissions contributed approximately equal amounts to the observed PM2.5 concentrations during winter and summer months ($1.31\mu gm^{-3}$ Vs $1.86\mu gm^{-3}$). In Isfahan data sets, the ambient source tracers were found to be strongly correlated with each other. Since the PCA is based on correlation matrix, the resolution of the outdoor impacts into separte source components was only partially successful. This resulted in clustering of possible iron and steel emissions with other outdoor sources as seen in the outdoor composite source terms. The clustering was observed in both smokers' and non-smokers' homes data such that reconstruction of sources also resulted in the same resolved outdoor composite source. PCA resolution of PM2.5 mass into source contributions was not always perfect. We found that the mass unexplained by the resolved sources was over 35% in two indoor analysis cases (non-smokers' homes in winter for Isfahan, and Tehran). But the resolved sources explained more than 80% of the elemental variance in these data sets.

Tobacco smoke source alone contributes half the observed PM2.5 concentrations inside smokers' homes. Ambient sulfate-related source impact the indoor environment, equally across smokers' and non-smokers' homes, with higher contribution in summer months. Outdoor sources are significant contributors to aerosol in homes characterized by non-smoke home class.

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