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Investigation of sources of atmospheric aerosol at urban and suburban residential areas in Thailand by positive matrix factorization

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Abstract

Samples of fine and coarse fractions of airborne particulate matter were collected in an urban residential area of metropolitan Bangkok from June 1995 to May 1996 and in a suburban residential area in Pathumthani, Bangkok's boundary province, from September 1993 to August 1994. The samples were analyzed for elemental concentrations by instrumental neutron activation analysis. The data sets were then analyzed by positive matrix factorization followed by rotation to identify the possible sources of atmospheric aerosols in both areas. The best solutions were found to be six factors for elemental compositions of each of the fine and coarse particulate matter fractions at the urban site and five factors each for both fine and coarse fractions at the suburban location. Soil was the major source of airborne particulate matter identified for all data sets. The motor vehicle factor showed much higher concentration for Br in urban than in suburban area. A motorcycle factor with high concentrations of Zn and Mn were found at the urban site. The factor containing highest concentrations of Na and Cl was attributed to sea-salt and was clearly seen in the urban atmosphere. The site was located 35 km to the north from the Gulf of Thailand and was influenced by wind from the south and southwest for most of the year. Charcoal/wood burning and incineration factors were likely to be the local sources. A factor with high concentration of Ca was attributed to a construction near the urban residential site and from two plaster manufacturing factories close to the suburban residential site. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Positive matrix facteorization; Receptor modeling; Airborne particulate matter; Thailand

1. Introduction

Bangkok (latitude 13°45′N, longitude 100°29′E) is the capital and the most populated city in Thailand. It has a population of over 5.6 million inhabitants in an area of approximate 1500 km^2. The city is congested with a large number of motor vehicles, including both public and private transportation. More than 3.9 million vehicles circulate within the city. Moreover, construction of high ways and buildings takes place continuously throughout the city. Many small-scale factories are also located in the area. With these sources, Bangkok has become highly polluted. Several organizations have studied the amount and composition of urban particulate matter (PM) for a number of years. They have found that air pollution is very severe in the areas of heavy traffic flow in the Bangkok metropolis and the most important problem is the particle emissions. However, the utilization of the PM data for the optimal determination of possible sources has remained limited. The importance and the need for local source identification in Bangkok area has been recognized as important in order to obtain a better understanding of the ambient air quality within the regional background conditions.

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In order to identify sources, multivariate receptor modeling can be applied to the observed PM composition data. Multivariate approaches are based on the idea that the time dependence of a chemical species at the receptor site will be the same for species from the same source. Chemical species are measured in a large number of samples gathered at a single receptor site over time. Species of similar variability are grouped together in a minimum number of factors that explain the variability of the data set. It is assumed that each factor is associated with a source or source type. Among the multivariate receptor modeling used for aerosol source identification, positive matrix factorization (PMF) is a relatively new technique developed by Paatero and Tapper (1993, 1994) and Paatero (1997). PMF has special features of the use of realistic error estimates to weight the data values and the imposition of non-negativity constraints in the factor computational process. The application of PMF have been successful in several atmospheric studies (Juntto and Paatero, 1994; Anttila et al., 1995; Polissar et al., 1996, 1998; Xie et al., 1999; Paterson et al., 1999).

The main objective of this study was the source apportionment of the atmospheric aerosol in Bangkok. However, since the urbanization has rapidly expanded from metropolitan area to its neighboring provinces, it is useful to get information of the air quality of both urban and suburban areas of the city. The work focused on PM with aerodynamic diameter of less than 2.2 μm (fine particle-FPM) and between 2.2 and 10 μm (coarse particle-CPM), which together constitute the 'inhalable particle' size range. Size-fractionated PM was sequentially collected at two sites representing urban and suburban residential area. The elemental composition of the samples was determined by Instrumental Neutron Activation Analysis (INAA). The data sets obtained were then analyzed with PMF.

2. Sampling and elemental concentration determination

Fig. 1 shows locations of the sampling sites in Bangkok and its surrounding area. The urban residential site was located at Bangkhen, approximately 10 km from the city center. The population within 10 km of the site is nearly 1.2 million and the residential population within 1 km is about 6500. In comparison, the suburban site at Klongha, Pathumthani, which is 40 km to the north of Bangkok, has a residential population of 85,000 within 10 km and only around 800 people within 1 km of the site.

The 'Gent' stacked filter unit (SFU) (Hopke et al., 1997) was operated at 16 l min^{-1} for 24 h sampling periods. FPM and CPM were collected separately on Nuclepore filters of 0.4 and 8 μm pore size, respectively. The aerosol sampling at Bangkhen was performed from June 1995 to May 1996. A total of 101 pairs of FPM and CPM samples consisted of 74 samples collected on weekdays and 27 samples collected on weekends. The sampling at Klongha had been done previously from September 1993 to August 1994. However, only 42 samples of FPM and CPM were collected and mostly on weekdays.

The filter samples were analyzed for elemental concentrations by INAA. By making two different irradiations and four gamma-ray counts after appropriate decay times, up to 25 elements could be determined (Chueinta, 1997). Since some elements had a large number of missing data and/or values below detection limits, data for only 17 measured elements were used in the data analysis.

3. Data analysis

In this study, PMF was used with the INAA data. PMF is a new variant factor analysis method and is described in detail by Paatero (1997). Only a brief description of the technique is given here.

3.1. PMF and data handling

PMF uses a weighted least-squares fit with the known error estimates of the elements of the data matrix used to derive the weights. The factor model (PMF2) can be written as

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

where X is the known $n \times m$ matrix of the m measured chemical species in n samples. G is an $n \times p$ matrix of source contributions to the samples (time variations). F is a $p \times m$ matrix of source compositions (source profiles). Both G and F are factor matrices to be determined. E is defined as a residual matrix, i.e., the difference between the measurement X and the model Y as a function of factors G and F.

$$e_{ij} = x_{ij} - y_{ij} = x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}$$

$$(i = 1, \ldots, n; j = 1, \ldots, m; k = 1, \ldots, p). \tag{2}$$

The objective of PMF is to minimize the sum of the squares of the residuals weighted inversely with error estimates of the data points. Furthermore, PMF constrains all of the elements of G and F to be non-negative; meaning that sources cannot have negative species concentration ($f_{kj} \geqslant 0$) and sample cannot have a negative source contribution ($g_{ik} \geqslant 0$). The task of PMF analysis can thus be described as to minimize Q, which is defined as

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} (e_{ij}/s_{ij})^2 \tag{3}$$

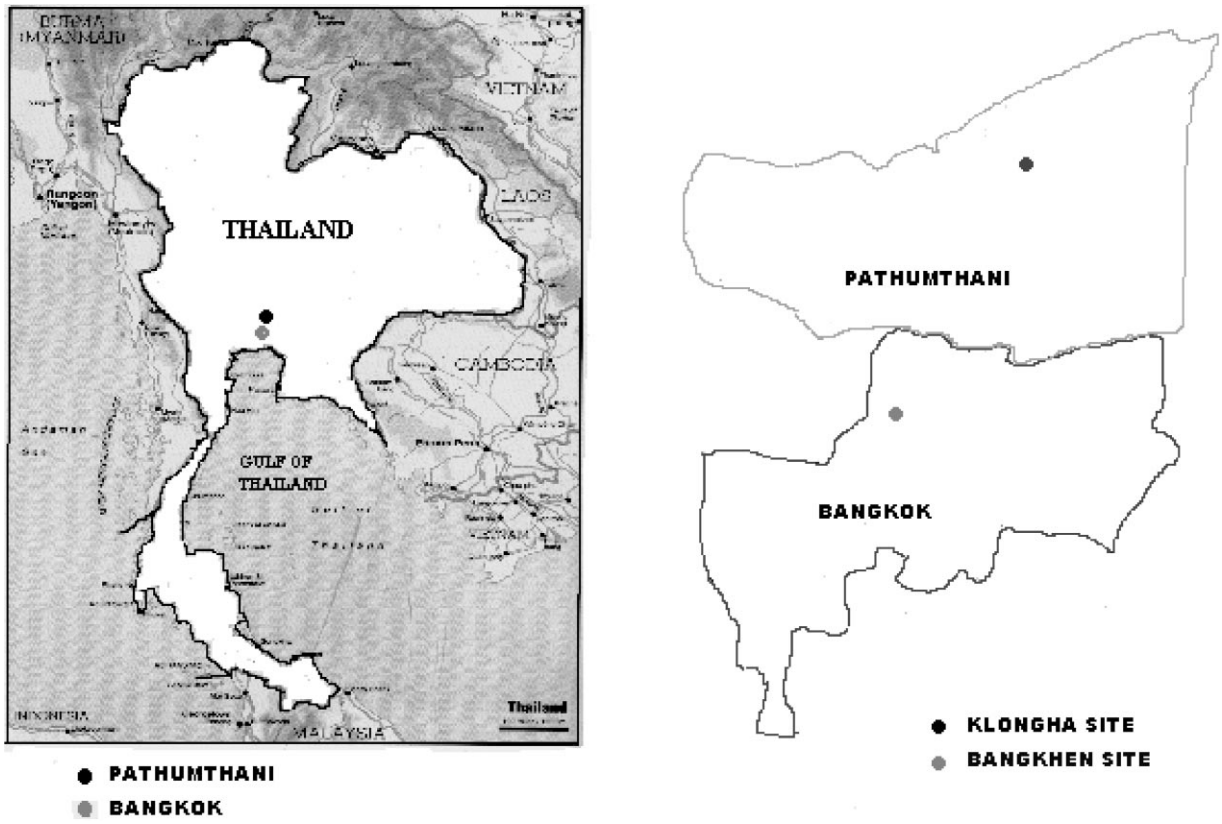

Fig. 1. Location of sites in Thailand.

with $f_{kj} \geqslant 0$; $g_{ik} \geqslant 0$ and s_{ij} is the error estimate for x_{ij}. The solution of Eq. (3) is obtained by a unique algorithm PMF2 in which both matrices, G and F, are adjusted in each iteration step. The process continues until convergence (Paatero, 1997).

A special advantage of PMF2 is the ability of the model to handle the incomplete data such as missing data, below detection limit data and the negative values after blank correction by giving low weights to such data points. In this work, any missing data were replaced by the arithmetic mean of corresponding elements. Half of the detection limit was used for any value below detection limit and its uncertainty. PMF2 can compute heuristic error estimates, s_{ij}, for x_{ij} based on the data point and its original error estimate, σ_{ij}. According to the error model used in this work, the equation is

$$s_{ij} = C_1 + C_3|x_{ij}|, \tag{4}$$

where C_1 is the observed error estimate, σ_{ij}, and C_3 the dimensionless values between 0.1 and 0.2 were chosen so that the relative uncertainty of each data set was reasonable.

Beside the ad hoc computation of error estimates, the program can take outliers into account. It must be noted that 'outlier' is not the same as 'bad data'. It is any data that significantly deviates from the distribution of the other data. The robust mode was selected to handle outlier values in the data matrix. The robust factorization based on the Huber influence function (Huber, 1981) is a technique of iterative reweighing of the individual data values. The least-squares formulation, thus, becomes to

$$Q = \sum_{i=1}^{m} \sum_{j=1}^{n} (e_{ij}/h_{ij}s_{ij})^2, \tag{5}$$

where

$$h_{ij}^2 = \begin{cases} 1 & \text{if } |e_{ij}/s_{ij}| \leqslant \alpha, \\ |e_{ij}/s_{ij}|/\alpha & \text{otherwise,} \end{cases}$$

where α is the outlier threshold distance and the value of $\alpha = 4.0$ was chosen.

The other important feature for this analysis was the desired rotation by using FPEAK to control rotations in PMF2. By setting a positive value of FPEAK, between 1.5 and 2, the routine is forced to subtract the F factors

from each other yielding more physically realistic solutions. The method requires a complicated mathematical process which is not described here (see Paatero, 1997).

4. Result and discussion

The distributions of sample masses of FPM and CPM collected at the two sites are plotted as the "box and whisker" plots shown in Fig. 2. A line in the box represents the median of the particle mass concentration. The range of the box depicts the bounds of the 25th and 75th percentile of the data. The whiskers extending from the box represent the bounds of the 10th and 90th percentile of the data. Outliers in each data set are also shown. For about half of samples collected at Klongha site, no mass concentration data are available and the exact sampling date cannot be specified. The data from Klongha site, therefore, is not separated while the data from Bangkhen site can be separated into samples of weekday and weekend. It can be seen that the particulate masses of each size-fraction PM at both sites are in the same range. The arithmetic means of FPM at Bangkhen and Klongha are 13.4 and 18.1 μg m^{-3} and the arithmetic means of CPM are 36.5 and 28.1 μg m^{-3} at Bangkhen and Klongha, respectively. Bar graphs showing average elemental concentrations with standard deviations of FPM and CPM from both sites are plotted together in Fig. 3. Tables 1 and 2 show the comparison data of yearly mean compositions in fine and coarse particulate matter reported in some countries. There are no significant differences between these values except that the concentrations of Al in the PM obtained from this study are, apparently, higher than the values reported in other countries included in Tables 1 and 2.

The importance of the elemental data is, however, to be utilized in the PMF analysis for the source apportionment that is the main objective of this study. For the determination of number of factors in PMF, the primary consideration is basically to obtain a good fit of the model to the original data. The theoretical Q-value should be approximately equal to the number of degree of freedom, or approximately equal to the number of entries of data array, provided that correct values of s_{ij} have been used in equation (3). Often there is no reliable information on s_{ij} and the presence of outliers (even in the robust mode) complicates the situation. Then it may be impossible to determine whether the observed value of Q is normal or too large. It is helpful to examine the distributions of scaled residuals (e_{ij}/s_{ij}). In a well-fit model, the residuals e_{ij} and the error estimates s_{ij} should be about equal and the ratios e_{ij}/s_{ij} should fluctuate between ± 2 (Juntto and Paatero, 1994). In this work, the calculated Q-values obtained from trials with five or six factors did not show that either was much better than the other. In the end, the factors were inspected to find

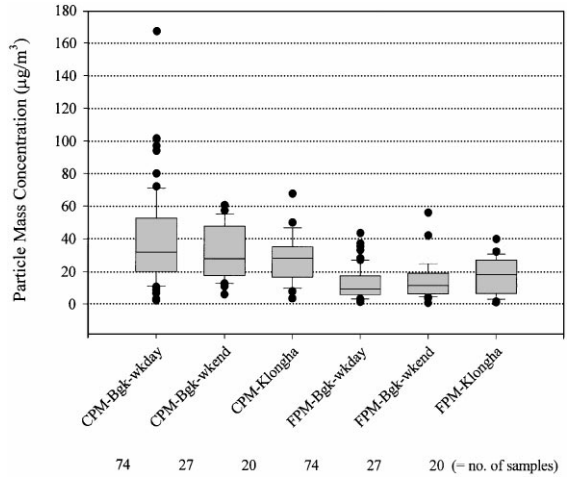

Fig. 2. Particle mass concentrations.

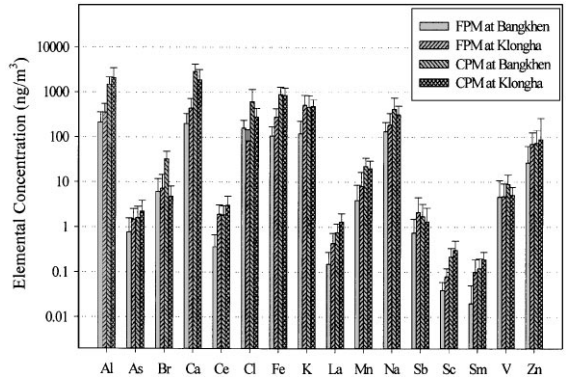

Fig. 3. Elemental concentration means and standard deviations of PM.

the most interpretable factor patterns. The analysis, with the application of the robust mode and with rotation, resulted in six factors for FPM and CPM at the Bangkhen site and five factors for FPM and CPM at the Klongha site. The number of factors, the identified source types, the theoretical and the calculated Q-values are summarized for each data set in Table 3. The calculated Q-values can be expected to be higher for the robust mode and applied rotations. However, if the values are not too large, it is reasonable to conclude a good fit to the original data even if the scaled residuals show a small number of outliers.

In this study, the time variation factors were normalized to a unit mean. Since only neutron activation elemental data are available and thus major elemental species like S, C, and Si are not measured, a normalization by using the measured mass concentration cannot

Table 1
Comparison of mean compositions of fine particulate matter (elemental concentration in ng m^{-3}, mass concentration in μg m^{-3})

Species	Australia Brisbane[a]	Belgium Gent[b]	Japan Kashima[c]	Portugal Lisbon[d]	U.K. Birmingham[e]	U.S.A. California[f]	This study	
							Bangkhen	Klongha
No. sample	261	118	64	~ 132	110	17	101	41
Mass	7.3	20	17.7	1.7–75.6	—	48.6	13.4	18.1
Al	29.2	23.2	124	—	—	97.2	211	356
As	—	—	—	0.79	4.20	—	0.77	1.5
Br	16.8	9.0	5.31	—	18.5	48.6	8.0	7.26
Ca	29.2	52	319	269	40.0	97.2	199	439
Ce	—	—	—	0.36	—	—	0.37	1.96
Cl	197	400	761	123	148	486	160	82.1
Fe	51.1	102	124	128	114	194	107	279
K	55.5	116	129	146	127	136	120	520
La	—	—	—	0.28	—	—	0.15	0.44
Mn	4.02	5.3	1.06	3.55	9.90	27.7	3.9	8.07
Na	—	162	266	442	348	194	135	187
Sb	—	—	—	1.71	—	—	0.74	2.14
Sc	—	—	—	—	4.60	—	0.04	0.08
Sm	—	—	—	0.012	—	—	0.02	0.1
Ti	5.11	3.4	—	5.81	4.70	24.3	25.3	—
V	—	11.0	—	—	4.95	—	4.67	4.78
Zn	26.3	38	62.0	52.8	297	112	26.5	70.2

[a]Chan et al. (1997).
[b]Maenhaut et al. (1995).
[c]Okamoto et al. (1986).
[d]Freitas et al. (1995).
[e]Harrison et al. (1997).
[f]Chow et al. (1994).

be performed. Hence, all plots of time variation of source contributions and source compositions are in arbitrary unit. The plots of source compositions are shown in Figs. 4, 6, 8 and 10. The time variations plots are shown in Figs. 5, 7, 9 and 11 for CPM and FPM collected at Bangkhen and Klongha, respectively.

For PM at Bangkhen site, the CPM had 6 possible source contributors (see Figs. 4, 5 and 12). The first factor shows high values of Na and Cl that clearly indicates sea salt. The Gulf of Thailand is located approximately 35 km to the south of the site. The polar plot of the relationship of this factor with wind direction shows the dependence on southerly wind direction as can be seen in Fig. 12a. The second factor contains high concentration of Zn and Mn. It suggests the contribution from motor vehicle, especially those of two-stroke engines such as motorcycle/motor scooter. In 1996, the number of motorcycles registered in metropolitan Bangkok was approximately 1.5 million and there were about 1 million cars in the city. The high number of motorcycles allows us to distinguish their impact from motor vehicles interpreted in the third factor which has the highest loading of Br. Because the analyzes were performed by INAA, lead values are not available. It should be noted that Al, Ca and Fe also appear in both factor two and three possibly suggesting contaminated road dust. These elemental compositions are commonly found correlated with a motor vehicle factor (Huang et al., 1994; Chow, 1995). The vehicle factor is also observed to be influenced by wind mostly from the south (see Fig. 12b). The toll-way and the junction of the main street are to the south and southwest of the sampling site. The time series plot of vehicle factor shows lower contribution during the rainy season (July–October).

The next factor has K as its only major constituent, which is then identified as charcoal/wood burning. The time series plot of this factor shows the highest peak in November and a few other high peaks in January. It was determined that there was considerable cutting and burning of tree branches in the area of the site during that winter. The fifth factor contains Al, Mn and Ti and indicates a crustal or soil dust source. High loadings of Ca and Fe appeared in factor six were probably from the building construction near the sampling site and is interpreted as cement since the cement was mixed at the construction site. The construction site was about 500 meters to the northwest of the sampling site and appears strongly in this wind direction polar plot for this

Table 2
Comparison of yearly mean composition of coarse particulate matter (elemental concentration in ng m^{-3}, mass concentration in μg m^{-3})

Species	Australia Brisbane[a]	Belgium Gent[b]	Japan Kashima[c]	Portugal Lisbon[d]	U.K. Birmingham[e]	U.S.A. California[f]	This study	
							Bangkhen	Klongha
No. sample	261	118	64	~ 132	110	17	101	41
Mass	10.4	11.8	17.5	1.7–75.6	—	22.5	36.5	28.1
Al	187	114	805	—	—	742	1476	2060
As	—	—	—	0.50	1.45	—	1.61	2.24
Br	4.16	2.7	3.5	—	12.2	0.00	32.6	4.86
Ca	239	290	962	2430	168	788	2865	1878
Ce	—	—	—	0.90	—	—	1.9	3.04
Cl	1082	470	2398	1240	682	990	618	279
Fe	198	240	595	470	187	1058	891	849
K	78	97	—	376	71.5	284	471	489
La	—	—	—	1.07	—	—	0.76	1.31
Mn	3.85	8.1	28	7.06	6.40	20.5	22.5	20.2
Na	—	440	1330	1520	698	1148	422	319
Sb	—	—	—	1.24	—	—	1.73	1.32
Sc	—	—	—	—	8.05	—	0.22	o.31
Sm	—	—	—	0.05	—	—	0.12	0.19
Ti	32.2	11.1	52.5	49.5	11.9	119	113	—
V	—	3.8	—	—	2.40	—	8.94	5.08
Zn	8.32	20.1	17.5	80.0	55.6	758	73.6	87.7

[a]Chan et al. (1997).
[b]Maenhaut et al. (1995).
[c]Okamoto et al. (1986).
[d]Freitas et al. (1995).
[e]Harrison et al. (1997).
[f]Chow et al. (1994).

Table 3
Possible source types, total number of factors and *Q*-values by PMF2 for 4 elemental data sets

Source type	Bangkhen		Klongha	
	CPM	FPM	CPM	FPM
Cement	—		—	
Soil dust	—	—	—	—
Sea-salt	—	—	—	
Charcoal/Wood burning	—	—	—	—
Refuse incineration		—	—	—
Vehicle	—	—		—
Motorcycle	—	—		
Oil combusion				—
Theoretical *Q*-value	1717	1717	656	656
Calculated *Q*-value	2485	2278	678	811
Number of factors	6	6	5	5

—shows the presentation of the factor contribution to the particular data set.

factor (Fig. 12c). Both soil and cement contributions seem to have a similar pattern with somewhat higher abundance during the winter (November–February).

The six factors for FPM at Bangkhen are described in Figs. 6, 7 and 13. They are attributed to charcoal/wood burning, motorcycle, vehicle, soil, sea salt and refuse incineration based on marker species K, Zn, Br, crustal elements, Na and Cl, and Sb, Zn and As, respectively. It can be noted that the wood-burning factor has a peak in January showing some correspondence to the same source for coarse fraction. The motorcycle and motor vehicle patterns are also similar to those seen in the CPM and show an even stronger influence of southerly wind (see Figs. 13a and b). There is no significant seasonal variation in sea-salt source contributions, but it can be seen from the polar plot in Fig. 13c that this factor depended on the wind from the south and the southwest. Refuse incineration and wood combustion seem to be local sources. Trash/wood/grass burning is common in many places. Furthermore, many residents still use wood and charcoal for cooking and heating.

The PMF analysis resulted in five factors for both CPM and FPM collected at Klongha. Figs. 8 and 9 provide the results for the CPM. The first factor consists of soil components such as Al, Ca, Fe, La and Mn. However, it also contains high value of K. Therefore, this factor could represent local soil strongly contaminated

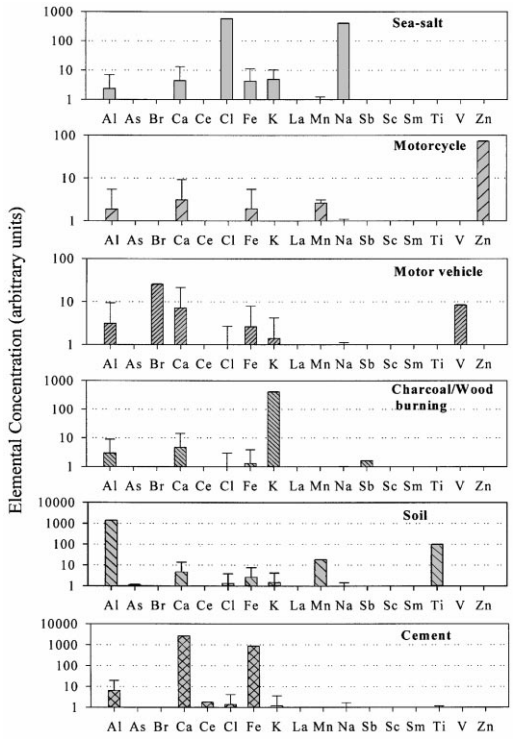

Fig. 4. Source compositions for CPM at Bangkhen.

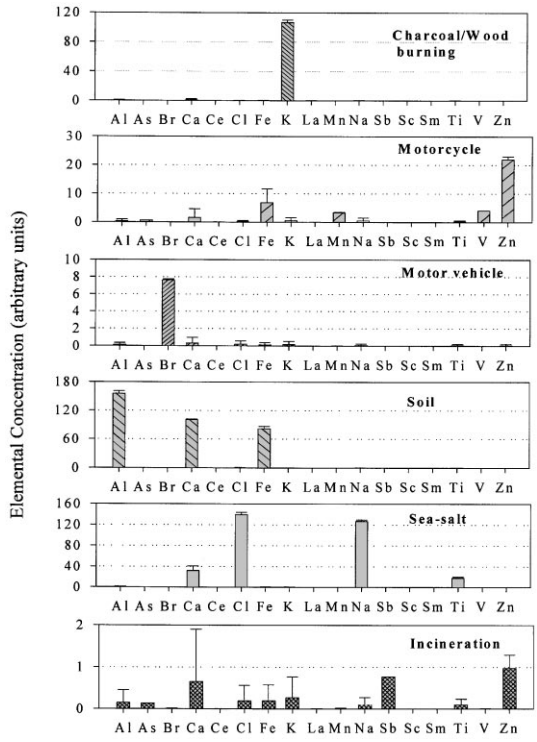

Fig. 6. Source compositions for FPM at Bangkhen.

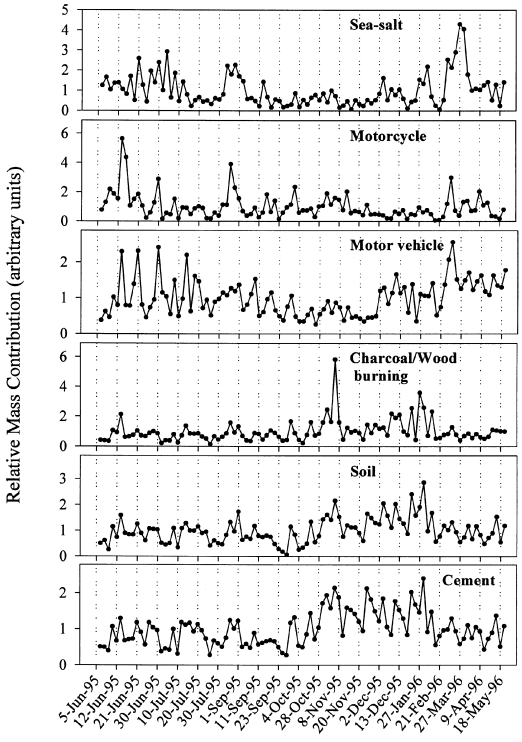

Fig. 5. Time variations for CPM at Bangkhen.

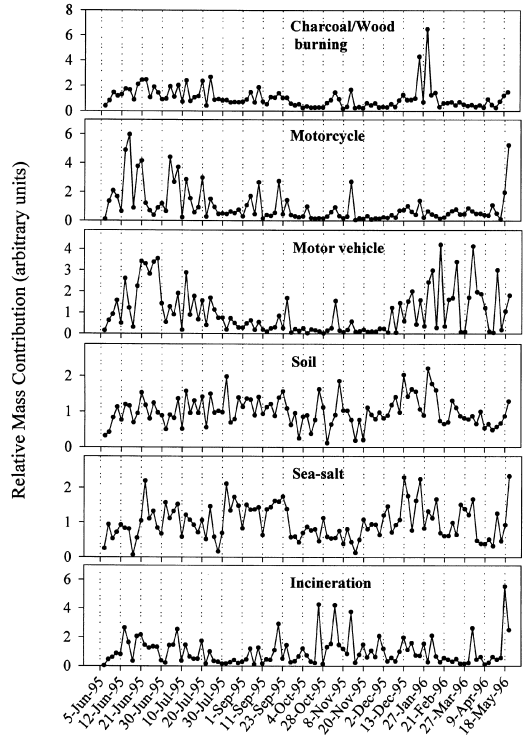

Fig. 7. Time variations for FPM at Bangkhen.

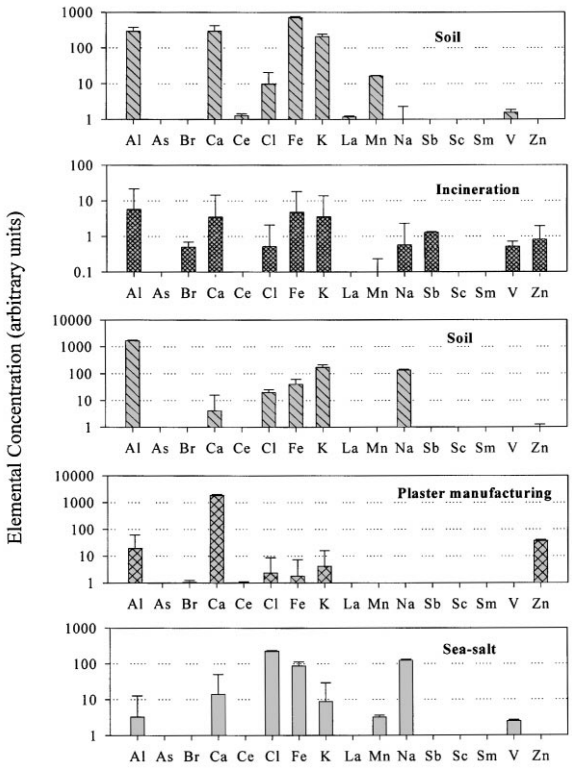

Fig. 8. Source compositions for CPM at Klongha.

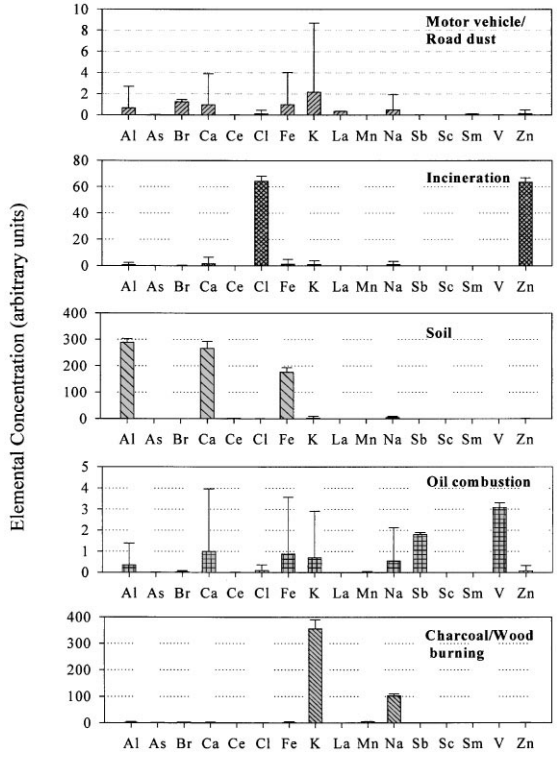

Fig. 10. Source compositions for FPM at Klongha.

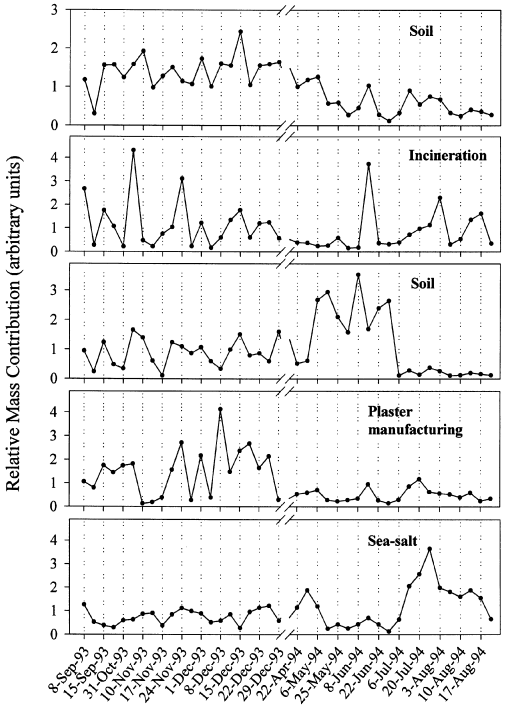

Fig. 9. Time variations for CPM at Klongha.

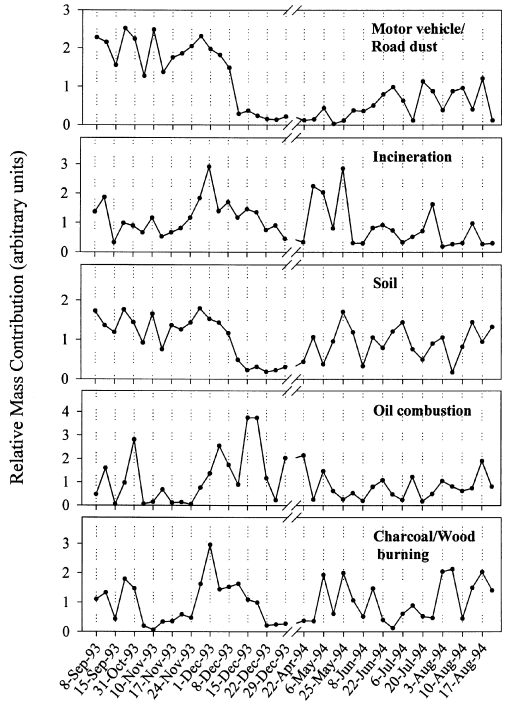

Fig. 11. Time variations for FPM at Klongha.

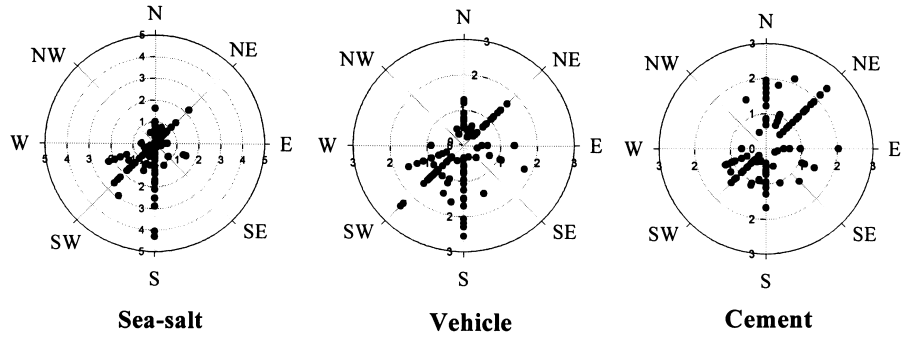

Fig. 12. The relationship of source contributions with wind direction for CPM at Bangkhen: (a) sea salt, (b) motor vehicle, and (c) cement.

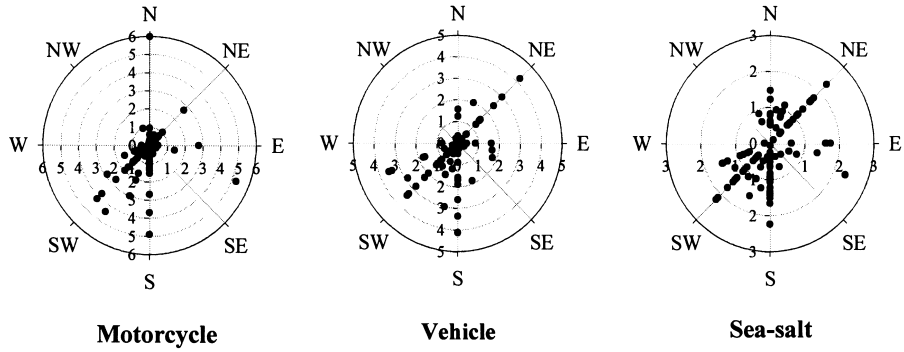

Fig. 13. The relationship of source contributions with wind direction for FPM at Bangkhen: (a) motorcycle, (b) motor vehicle, and (c) sea salt.

by biological materials or wood ash. The second factor was attributed to refuse incineration based on the high loadings of Sb with Zn. Minor amounts of other crustal elements were also observed. Al dominates the third factor and is, thus, considered to be the second soil dust source. The fourth factor, on the other hand, has very high Ca. This source is assumed to be the two plaster manufacturing factories located approximately 1–2 km from the sampling site. The last factor characterized by Na and Cl indicates the sea-salt source. From the time series plots, it can be observed that soil and cement contributions are, apparently, diminished during rainy season but the sea-salt source is enhanced as might be expected from higher winds in the monsoon season. However, with the limited number of samples analyzed and the lack of meteorological data, it is difficult to extend our analysis in greater detail.

The results obtained for FPM at Klongha were not as readily interpreted as the other data sets. Nevertheless, five factors provided the explanation for the data (see Figs. 10–13). The first factor contains equivalent loading for Br, Al, Ca, Fe, including K. However, the elements other than Br have much higher uncertainties and are not as significant as Br. The factor might be accounted for the mixing of sources among vehicles, road dust and emissions from vegetation or wood smoke. Zn and Cl dominate the second factor. Since it is in fine fraction, Zn was more likely attributed to refuse incineration. Cl in this factor does not show a correlation with Na. This lack of relationship implies that Cl may not come from sea spray but possibly from polyvinylchloride plastic in the burning trash. The third factor is crustal material. The fourth factor has significant Sb and V and reflects possible emission from incineration and/or oil combustion (Greenberg et al., 1978; Church et al., 1988; Xie et al., 1999). Duce et al. (1975) reported that anthropogenic V in aerosol was a result of combustion of heavy fuel oil containing V-porphyrin complex. The heating process in plaster manufacturing factories might be the source of this factor. On the other hand, there is a small electric generation plant with a capacity of 237.5 MW using fuel oil, located in southern Pathumthani (north of Bangkok). There is also a privately installed generator (approximately 60 MW) for local use by Pathumthani province using fuel oil and diesel. The final factor is described as charcoal/wood burning because of the high abundance of K. The time-series plots of fine fraction results do not provide much information.

5. Conclusion

By collecting size-fractionated PM and determining elemental composition by INAA, the major particle sources can be identified by applying PMF2 at an urban and a suburban residential site in Bangkok, Thailand. PMF analysis reveals six sources of size-fractionated PM at Bangkhen, the urban area, and five factors for those samples from the Klongha site, the suburban area. The general sources for both residential areas are found to be soil, vehicle, charcoal/wood combustion, incineration, sea salt, and cement.

Soil is the major source of PM and was identified in all data sets. The vehicle factor shows much higher loading of Br in Bangkhen than in Klongha. The seasonal patterns of both soil and vehicle factors show the high peaks during winter season and lower values during the monsoon period. Motorcycle emissions can be identified at Bangkhen site. The sea-salt factor is clearly seen at Bangkhen. Sea-salt factor contributing to the coarse fraction of aerosol at Klongha was identified with the maximum contributions during the monsoon period. Charcoal/wood burning and incineration factors are likely to be local sources. Cement factors with a high concentration of Ca was contributed by construction near by the Bangkhen site and from two plaster factories close to the site at Klongha.

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