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Identification of Sources of Fine Particulate Matter in Kandy, Sri Lanka

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ABSTRACT

Kandy is the second largest city in Sri Lanka and a major tourist destination. It is a fast growing city with continuous construction of buildings, roads and historical places. More than 100 samples of fine particulate matter (PM) were collected using a GENT stacked filter sampler from a fixed site at the regional sampling station of Department of Meteorology situated in Katugastota, Kandy over the period of 2012 to 2014. Black carbon (BC) in these filters were determined by reflectance measurements while their elemental compositions were determined using the X-ray fluorescence spectrometry. Analysis of the elemental data suggests that the PM in Kandy originates largely from re-suspended soil and anthropogenic sources. The fine particulate matter data including BC and major elements (Na, Mg, Al, Si, Cl, Fe, Zn, Ni, Cu, V, S, Br, Pb, Cr, K, Ca and Ti) was analyzed using EPA-PMF version 5.0 (Positive Matrix Factorization) to explore the possible sources of the PM at the study site. Five factors were found and identified as soil, aged sea salt, vehicular emissions, biomass burning, and industrial sources.

Keywords: Particulate matter; Source apportionment; Positive Matrix Factorization; Sri Lanka.

INTRODUCTION

Epidemiological studies have found that air pollution has significant adverse health and visibility effects in the high population cities (Dockery *et al.*, 1993, UNEP, 2002; Katsouyanni, 2005). To protect public health, many countries have instituted air quality standards that set maximum allowable concentrations. For example, the United States Environmental Protection Agency promulgated an airborne particulate matter (PM) in the < 2.5 μm size ($PM_{2.5}$) standard of 12 μg m^{-3} for annual average and 35 μg m^{-3} for 24-hr maximum in 2012. In Australia, the $PM_{2.5}$ goals are 8 μg m^{-3} for annual average and 25 μg m^{-3} for 24-hr maximum. Sri Lanka has a fine $PM_{2.5}$ permissible level of 25 μg m^{-3} for the annual average and 50 μg m^{-3} for a 24 hr maximum. However, for many locations, very few measurements have been made or presented in the scientific literature.

Kandy (Latitude: 7°18′26”N, Longitude: 80°38′55”E) is the second largest city in Sri Lanka and is located in the Central Province (Fig. 1). The surroundings are mountainous and thickly forested. Kandy is a historical city since it was the last capital of the ancient kings' era of Sri Lanka. It was declared a world heritage site by UNESCO in 1988. The city lies in the midst of hills in the Kandy plateau (altitude 473 m) and its condition is similar to a bottom of a basin (Supplemental Fig. S1). Currently its population is more than 1.3 million and approximately 60,000 motor vehicles enter the city of Kandy daily. It is a growing city with the continuous construction of historical places, residences, commercial and official buildings as well as roads. The weather is tropical with average temperatures varying from 19°C to 28°C. The average annual precipitation is 1840 mm. Because of the increased use of vehicles and other human activities driven by a growing population in Kandy area, a long term monitoring study of airborne particulate matter (PM) was initiated in 2012 at the sampling station (Latitude: 7°19'51"N, Longitude: 80°37'36"E) of the Department of Meteorology, Katugastota, Kandy, central province of Sri Lanka. The areal distance from the Kandy city center to the sampling site is 5 km (Fig. 1, right).

The objective of this study was to investigate the airborne particulate matter characteristics including elemental analysis by ED-XRF, and to apply source apportionment methods to the resulting data to determine the sources contributing to the observed concentrations. The source profiles of $PM_{2.5}$ and the relative contribution of each source in different season were identified using positive

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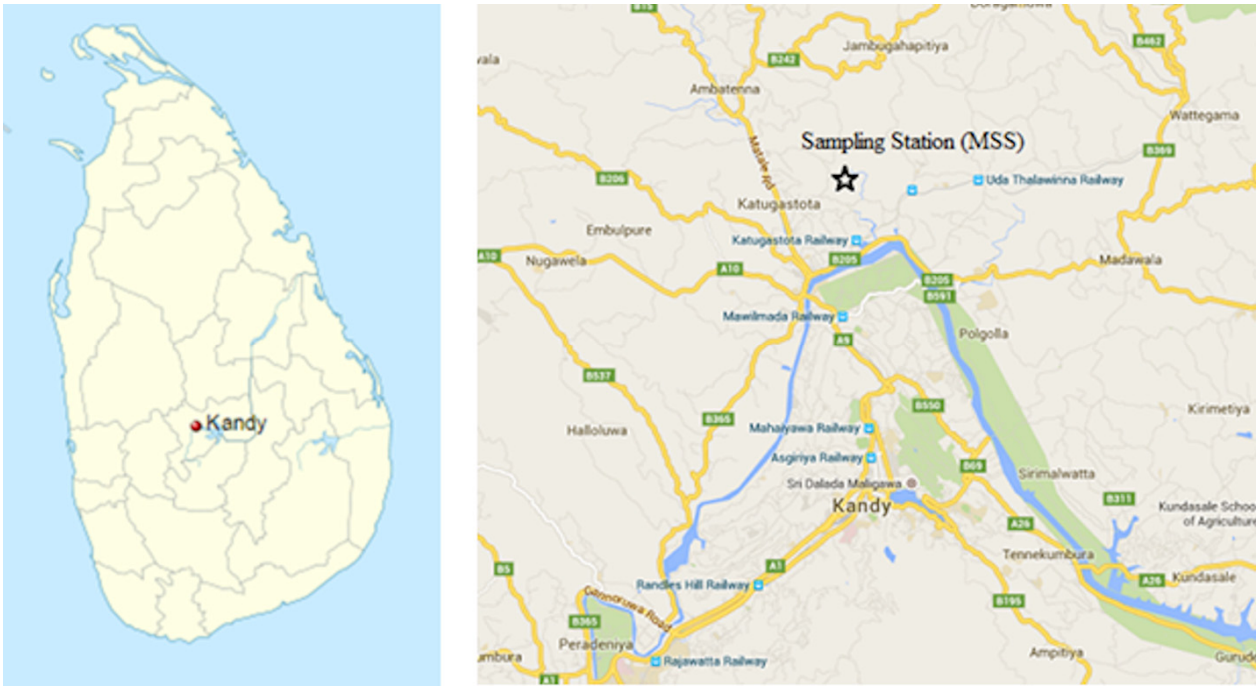

Fig. 1. Maps of Sri Lanka (left) and Kandy showing the sampling site (right).

matrix factorization (PMF) and the locations of the source contributions are explored through conditional probability function (CPF) analyses. A feature of this study is the use of the new error analysis tools that were included in EPA PMF V5. These tools will be introduced and their utility in understanding the PMF results will be shown.

EXPERIMENTAL TECHNIQUES AND METHODS

Sampling was conducted using a "Gent" stacked filter unit particle sampler capable of collecting particulate matter in the size ranges of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ (Hopke *et al.*, 1997). Samples were collected over 24-hour periods from June 23, 2012 to October 20, 2014 using nuclepore filters with 8 µm pore size for the coarse fraction and 0.4 µm pore size filter for the fine fraction. The samples were collected on week days with a flow rate 14 to 16 L min^{-1}. The sampling site is located at the meteorological sampling station (MSS) in downtown Katugastota, Kandy (Fig. 1, right) and the stacked filter unit of the GENT sampler was mounted on the ground with the sampling head approximately 1 m above ground level. The site was in a typical suburban Kandy area with nearby traffic, industry, and biomass burning episodes.

Mass values were determined by weighing the filters on a microbalance (A&D HR202i semi microbalance, sensitivity = 0.1 mg) before and after the sample collection with a 24–hour period of equilibration at 50% relative humidity. Mass was determined for both the coarse and fine particle samples. However, only the fine particle masses were used in these analyses. Energy Dispersive X-ray Fluorescence (EDXRF) spectrometry has been an accepted method for the characterization of particle pollution for many years (Markowicz *et al.*, 1996). This method is well suited for the analysis of filters containing only few hundred micrograms of fine particulate air pollutant. The EDXRF analyses of the fine fraction samples were performed using a Spectro XLAB-2000. Single element MicroMatter standards were used to develop the calibration parameters and samples of NIST SRM 2783 were routinely analyzed with each batch of samples to provide quality assurance of the elemental concentrations. Replicate analyses were made on every 20^{th} filter. XRF analysis of the fine particle filters determined the concentrations of the elements Na, Mg, Al, Si, Ca, Cl, K, S, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br and Pb on fine filters. Analytical errors were estimated using the approach described by Gutknecht *et al.* (2006). The coarse particle filters were not subjected to chemical characterization.

A Smoke Stain Reflectometer (Diffusion Systems Ltd. Model M43D) was used to measure the black carbon (BC) on the fine filters (Edwards *et al.*, 1983) and has been widely used in this region for BC measurements (Hopke *et al.*, 2008). The average fine particle mass absorption coefficient was assumed to be 5.7 m^2 g^{-1} based on prior calibration work for the reflectometer (Biswas *et al.*, 2003).

Advanced data analysis methods such as Positive Matrix Factorization (PMF) and conditional probability analysis (CPF) have been routinely applied to such data sets. The application of PMF has been successful in many similar atmospheric studies (e.g., Chueinta *et al.*, 2000; Begum *et al.*, 2004; Begum *et al.*, 2005; Santoso *et al.*, 2011, Seneviratne *et al.*, 2011) and source apportionment methods have recently been reviewed by Hopke (2016). US EPA PMF (US EPA, 2014) version 5.0 is a program that quantitatively provides both fingerprints and daily time series plots for major source contribution at a given site. These EPA-PMF programs and their applications have been discussed in detail elsewhere (Reff *et al.*, 2007; Norris *et al.*, 2014).

The latest version of the U.S. Environmental Protection

Agency PMF (V5.0.14) incorporates new tools to evaluate the uncertainties in the extracted profiles (Paatero *et al.* 2014). The previously employed bootstrapping method was replaced with a displacement approach (DISP) in which individual values in the profiles were pulled away from the best fit values until the objective function value, Q, rose beyond a specified limit. Also pulling on a value should not cause the factors to change identities ("factor swapping"). DISP can then be combined with bootstrapping, DISP-BS, to examine both rotational ambiguity and measurement uncertainties. There are additional diagnostics provided that allow the identification of bad choices of a base case run and solutions that are not well determined. Thus, these tools permit further exploration of the quality of the derived fit to the data and identify potential problems with the solutions as well as the likely uncertainty bounds on the profile values.

RESULTS AND DISCUSSION

$PM_{2.5}$ Mass Data

A total of 127 filters were collected during 2012 to 2014 at the Kandy, Katugastota MSS site. Fig. S2 shows the daily collected $PM_{2.5}$ mass concentrations for the study period. Twenty-four hour $PM_{2.5}$ concentrations at meteorological (MSS) site ranged from 5 to 60 μg m^{-3} and the mean $PM_{2.5}$ concentrations for MSS site is 16 ± 2 μg m^{-3}, respectively. Sometime small peaks of $PM_{2.5}$ mass of about 30–40 μg m^{-3}, occurred around New Year and Christmas, showing the effects of fireworks and festival celebration on air quality in Kandy.

The annual average ranged from 18 μg m^{-3} in 2012 to 14 μg m^{-3} in 2014 well about the annual US EPA standard (12 μg m^{-3}) and the Sri Lanka permissible level of 25 μg m^{-3} for fine particles. The histogram of daily $PM_{2.5}$ mass values in μg m^{-3} is presented in Fig. S3. The daily $PM_{2.5}$ exceeded 25 μg m^{-3} more than 18% of time during the measurement period.

The south-western monsoon brings rain to the south-west of Sri Lanka between May and September, while the dry season in this region runs from December to March. Because Kandy is situated in the middle of the country in mountainous terrain, it does not have a consistent influence from the seasons, but it is more affected by the solar calendar. Kandy is known for quite mixed weather in both intermonsoon seasons but also adjacent months. The days can be sunny, but if clouds in the lowlands reach the mountains, there can then be persistent rainfall. The monthly distributions of $PM_{2.5}$ mass are shown in Fig. S4. It can be seen that some of the highest concentrations are observed in March and April.

Concentrations Data

The average $PM_{2.5}$ fine particle composition for 2012 to 2014 are summarized in Table 1. Mean values and their standard deviations as well as median and maximum concentrations for each of the measured chemical species are presented. These concentrations are similar to those observed in other major cities across South and Southeastern Asia (Hopke *et al.*, 2008). The variations in constituent concentrations generally follow the variability in mass concentrations. The BC mass concentration distributions are shown in Fig. S5. These distributions have a pattern somewhat different from that of PM mass. There is a minimum in the southwestern monsoon period and a more distinct peak during the winter months when there would be biomass burning for home heating during the cooler months particularly at Kandy's higher elevation. However, some high values were still observed in April and May.

Table 1. Summary statistics for the species measured in $PM_{2.5}$ samples from Kandy, Sri Lanka.

Species	Mean ± SD	Median	Maximum	No. Below Detection Limit
Na (ng m^{-3})	2047 ± 130	374	12171	2
Mg (ng m^{-3})	54 ± 3	35	572	1
Al (ng m^{-3})	203 ± 2	200	820	0
Si(ng m^{-3})	294 ± 2	201	3261	0
S (ng m^{-3})	403 ± 1	348	2175	10
Cl (ng m^{-3})	126 ± 1	89	1912	18
K (ng m^{-3})	218 ± 4	214	782	0
Ca (ng m^{-3})	91 ± 2	66	1201	3
Ti (ng m^{-3})	15 ± 1	12	107	3
V (ng m^{-3})	1.6 ± 0.5	2	4	33
Cr (ng m^{-3})	8.2 ± 0.6	10	17	0
Mn (ng m^{-3})	8.4 ± 0.5	10	25	11
Fe (ng m^{-3})	332 ± 6	368	654	0
Co (ng m^{-3})	3.6 ± 1.9	3	10	1
Ni (ng m^{-3})	49 ± 2	40	100	0
Cu (ng m^{-3})	82 ± 3	99	174	0
Zn (ng m^{-3})	50 ± 2	59	126	34
Br (ng m^{-3})	21 ± 2	20	55	0
Pb (ng m^{-3})	12 ± 1	16	36	54
BC (μg m^{-3})	8.7 ± 0.9	7	25	0
Mass (μg m^{-3})	17.6 ± 11.6	14	75	0

Table 2 show the comparison data of yearly mean compositions in fine particulate matter (PM) reported in selected countries. There were no major differences between these values except that the concentrations of Na in the PM obtained in this study are higher than the values reported in other locations included in Table 2.

Source Apportionment Using EPA Positive Matrix Factorization (EPA - PMF)

EPA PMF version 5.0 (US EPA, 2014) was applied to the $PM_{2.5}$ elemental and BC data to provide a mass apportionment. The Kandy dataset included 127 samples covering the period of 23–June 2012 to 20–October 2014. The fine fraction data set from the MSS site included BC and major elements (Na, Mg, Al, Si, Cl, Fe, Zn, Ni, Cu, V, S, Br, Pb, Cr, K, Ca and Ti). The uncertainties were calculated using the procedure described by Polissar *et al.* (1998) based on the analytical uncertainties and the detection limits for each determination. Organic carbon and ammonium nitrate are often important components of $PM_{2.5}$. However, they were not determined in this study. Therefore, the reconstructed mass was estimated using the procedures of Malm *et al.* (1994) and Begum *et al.* (2006). An additional variable, unmeasured mass (UMM), was calculated as the difference between the measured mass and the reconstructed mass. Similar procedures have been done in prior studies (e.g., Zhao *et al.*, 2007). The uncertainty assigned to UMM was 100%.

The main user input to EPA PMF is the number of factors. The fractional elemental contributions associated with each profile, together with the source contributions to the total fine mass were determined by PMF. The five factor solution was chosen because it provided good fits to the data as observable in the scaled residuals as well as providing physically interpretable source identifications. Four factors resulted in a number of skewed distributions of the scaled residuals and six factors resulted in narrow distributions that clearly represented overfitting of the data.

The profiles and the mass contributions of the five factors for FPM resolved in Kandy are presented in Figs. 2 and 3, respectively. Fig. 2 provides the profile (gray bars), the explained variation (solid circles), the average DISP value (open circles connected by lines to guide the eye) and error bars to indicate DISP minimum and maximum values. The sources were attributed to soil, aged sea salt, automobile traffic, biomass burning, and industrial emissions. The mass values were well fit by the analysis (slope = 1.003 ± 0.006, r^2 = 0.989) and the plot of the predicted versus measured mass concentrations is provided in Fig. S6.

The first factor was identified to be soil containing crustal elements Al, Ca, Fe, Si and Ti (Cohen *et al.*, 2010) and it contributed an average of 3.8% of the mean mass concentration. It can be seen that the bounds on these elements as shown by the error bars are relatively small. There was a substantial value for UMM that likely represents the organic carbon mass associated with suspended soil. Although there is a high value for BC, the error bound includes very low possible values. Similarly, for K, Cr, Mn, Ni, Cu, Zn, and Br, the profile values could include zero. Thus, although the presence of these elements might suggest intermixed sources, the error analysis suggests that only the crustal elements are strongly positioned in this factor. The CPF plot points to southeast that is likely due to the building and road constructions in Kandy and its suburban areas.

The 2nd factor includes Na, Cl, and contributes 3.2% to

Table 2. Comparison of mean compositions of fine particulate matter (elemental concentration in ng m^{-3}, mass concentration in µg m^{-3}).

Species	Australia Brisbane[2]	Belgium Gent[3]	Japan Kashima[4]	Portugal Lisbon[5]	U.K Birmingham[6]	U.S.A California[7]	Thailanad Bangkok[8]	This study Kandy
NO Samples	261	118	64	~132	110	17	101	137
Mass	7.3	20.0	17.7	1.7–75.6	-	48.6	13.4	16.0
Al	29.2	23.2	124	-	-	97.2	211	203.0
Br	16.8	9.0	5.31	-	18.5	48.6	8.0	21.0
Ca	29.2	52	319	269	40.0	97.2	199	91.0
Cl	197	400	761	123	148	486	160	126.0
Fe	51.1	102	124	128	114	194	107	332.0
K	55.5	116	129	146	127	136	120	218.0
Mn	4.02	5.3	1.06	3.55	9.90	27.7	3.9	8.4
Na	-	162	266	442	348	194	135	2047.0
Ti	5.11	3.4	-	5.81	4.70	24.3	25.3	15.0
V	-	11.0	-	-	4.95		4.67	1.6
Zn	26.3	38	62	52.8	297	112	26.5	50.0

[2] Chan *et al.* (1997).
[3] Maenhaut *et al.* (1995).
[4] Okamoto *et al.* (1986).
[5] Freitas *et al.* (1995).
[6] Harrison *et al.* (1997).
[7] Chow *et al.* (1994).
[8] Chueinta *et al*. (2000).

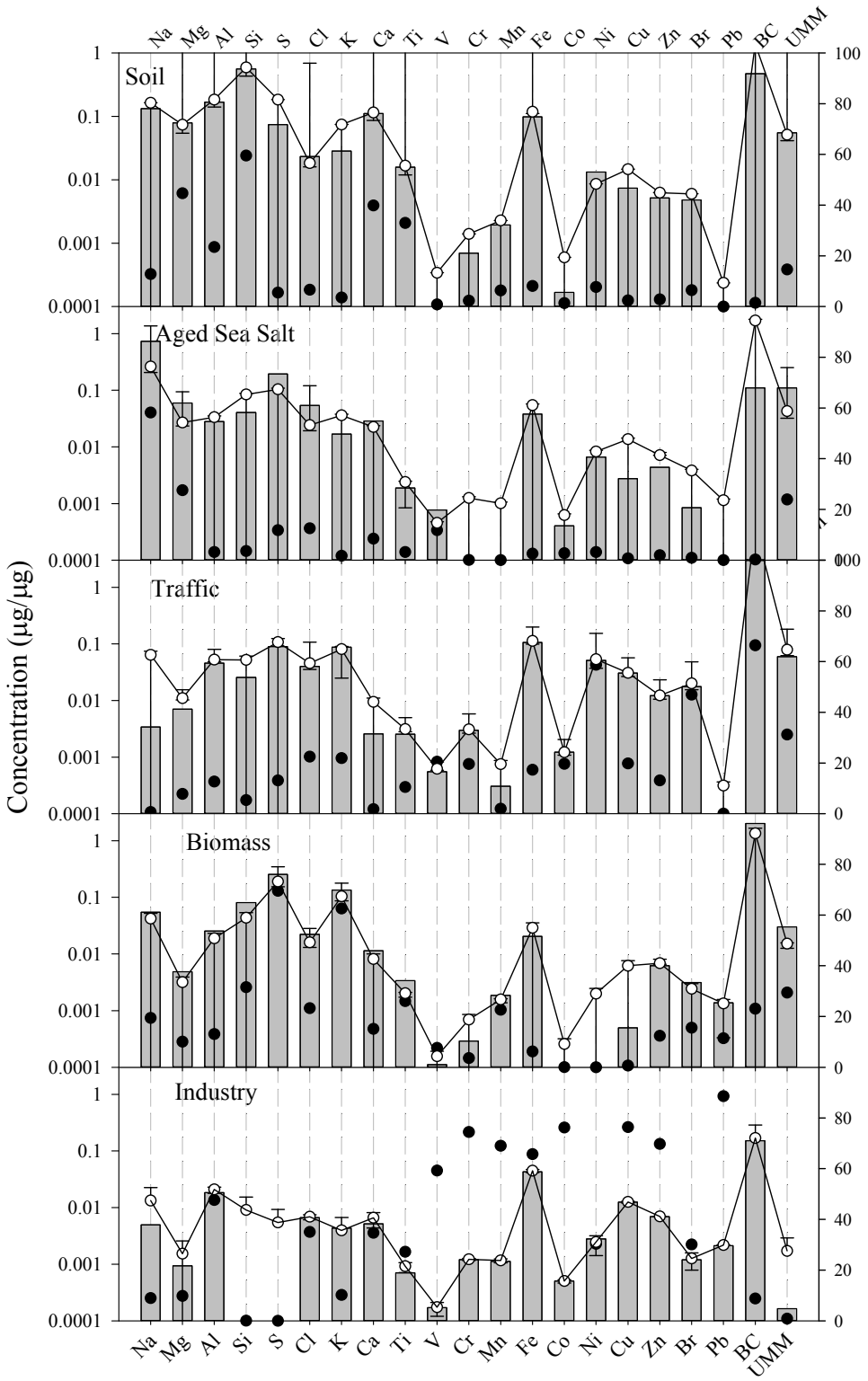

Fig. 2. Source profiles derived from $PM_{2.5}$ data in Kandy, Sri Lanka.

the mass. There is also sulfur and UMM that may represent acid displacement by chlorine by sulfate and nitrate. All of the other species have uncertainty bounds that extend to very small values. It includes a small concentration of V along with the sea salt (Polissar *et al.*, 1998) suggesting the possibility of some input from marine diesel emissions through the combustion of residual fuel oil. The highest sea salt concentrations come during July and August when the monsoon brings the marine aerosol inland to Kandy. The CPF clearly shows this influence of the westerly monsoon winds transporting the marine aerosol from the west coast of Sri Lanka to Kandy.

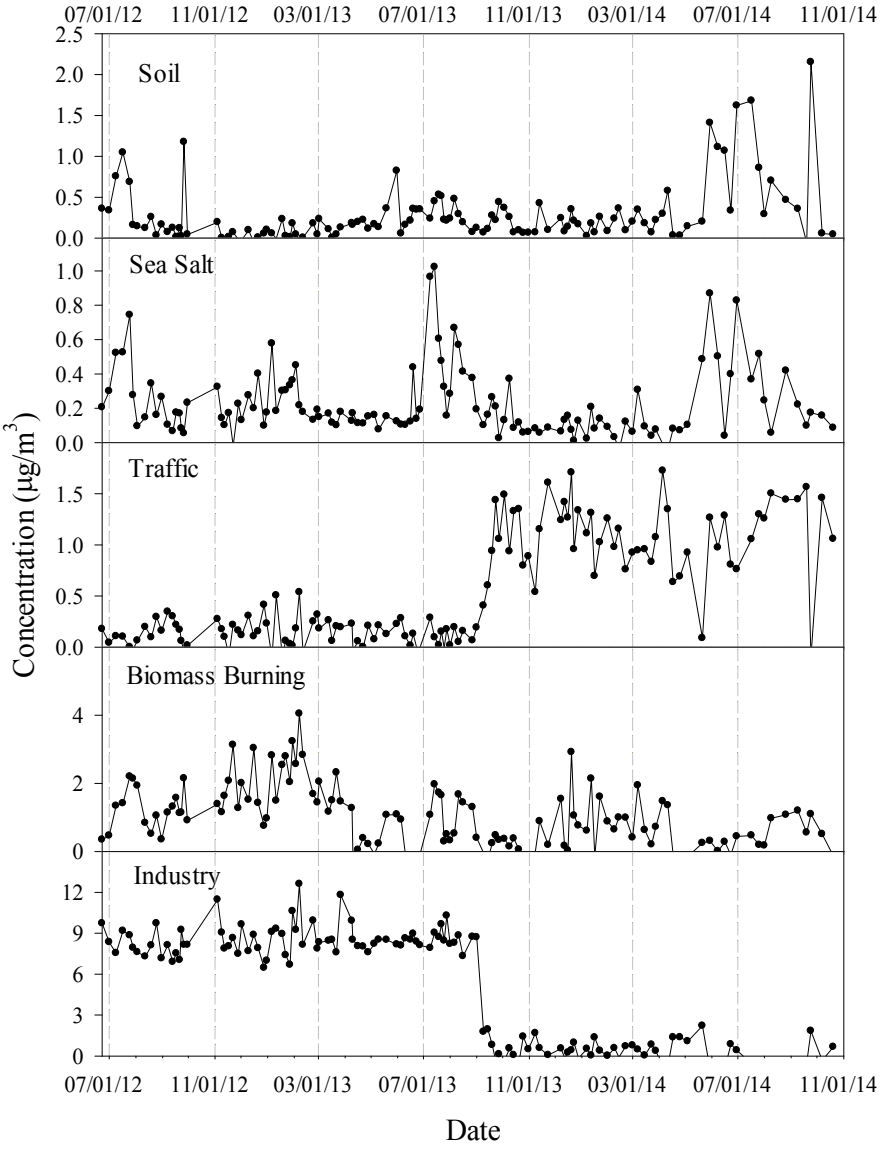

Fig. 3. Time series of mass contributions to the $PM_{2.5}$ measured in Kandy, Sri Lanka.

The 3rd factor includes Automobile Traffic and related to S, Br, Zn, Pb and BC with an average contribution of 7.6% of the average mass concentration. It also includes a large quantity of UMM that is again likely to represent organic carbon. The motor vehicle (diesel and gasoline engine exhaust) source profile characterized by the high BC with tight error bounds and S with broad uncertainty values (Kim *et al.*, 2003, Begum *et al.*, 2004). Diesel fuel in Sri Lanka has a typical sulfur content of 2000 ppm. Combustion in an internal combustion engine results in 5 to 10% of the sulfur emitted as SO_3 that will quickly react with water to form sulfuric acid. Thus, there is primary sulfate emissions from diesel vehicles under these circumstances. This combination of elements suggests a contribution from two–stroke vehicles (Chueinta *et al.*, 2000) due to Zn, BC and Fe still stroke vehicles was stated in 2010 to improve the elements specially Zn. The contribution plot shows that the traffic contribution was high up to January 2013 when it dropped substantially and stayed low for the rest of the sampling period. This change may be the result of changes to traffic regulations in Kandy. The CPF plot points to several main roads around the sampling site within a maximum distance of 5 km (Fig. 1).

The fourth factor profile has high values of K, Si, BC, and UMM with tight bounds except for BC. Again, UMM is likely to represent organic carbon. It is assigned as the emissions from biomass burning that produce high concentrations of carbonaceous particles (Santoso *et al.*, 2008). Burning wood, paper, cardboard and biomaterial including vegetation produce significant emissions in this area since open burning is a common habit. The study by Cohen *et al.* (1997) found that burning was a major source represents 14.1% of fine fraction mass. The biomass burning contributions are highest in the post-monsoon and winter seasons. The increased concentrations could be the result of lower mixed layer and lower wind speeds during this part of the year. There could also be a contribution of long-range transport to the biomass burning concentrations

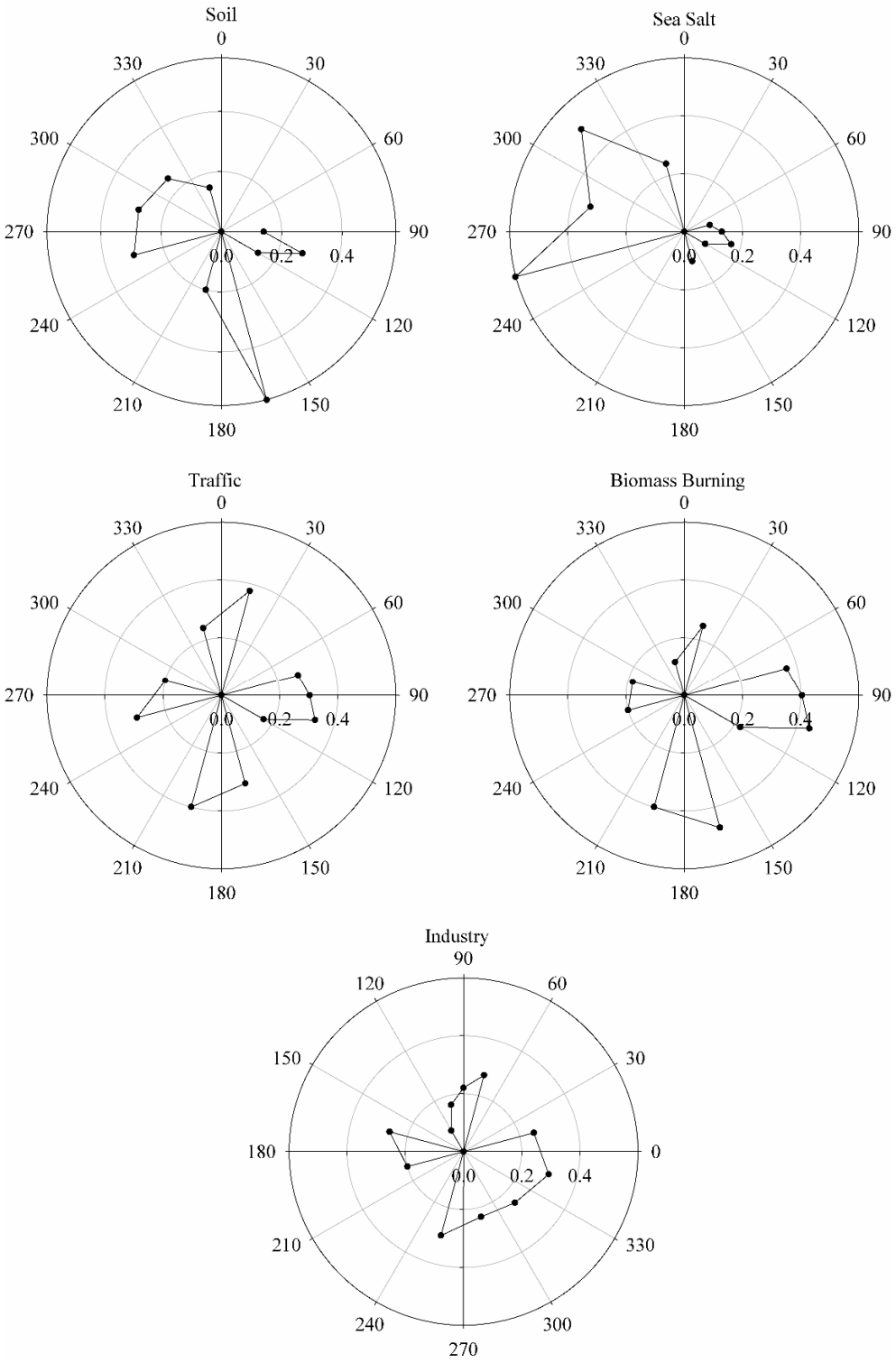

Fig. 4. Conditional probability function plots for the five sources contributing to $PM_{2.5}$ in Kandy, Sri Lanka.

particularly during the late fall. Begum *et al.* (2011) identified transport of biomass burning emissions in India to Colombo. Studies have observed very high concentrations of biomass burning including agricultural burning particulate matter across the Indo-Gangetic Plain area of northern India (Tiwari *et al.*, 2015a, b). The CPF plot points to the south in the direction of the Temple of the Tooth, in which thousands of local and foreign Buddhist/non-Buddhist people light oil lamps and incense-sticks. There are many other temples around the Temple of the Tooth and all the pilgrimages visit and worship to those places also largely through the burning of incense. Thus, it seems likely that local biomass burning is the major source of the estimated contributions.

The 5th factor is identified industrial emissions with Al, Fe and most of the measured transition metals (V, Cr, Mn, Fe, Co, Cu, Zn, and Pb) with high explained variations and tight uncertainty bounds. This factor is largely attributed to metallurgical industries. Kandy is famous for its brass and other metal products. The time series of contributions suggests a substantial reduction of industrial manufacturing in late 2013, and it is not currently known why there was such a sharp decline in emissions. There are several small-scale industries around this area such as brass industry, sawmills, limestone powdering etc. The CPF plot points in the directions of a number of known local small industries.

CONCLUSIONS

The chemical composition data of fine air particulate matter ($PM_{2.5}$) collected from Kandy, historical city was studied using EPA PMF to explore the possible emission sources. The general sources for residential and community areas are founded to be the soil, aged sea salt, traffic, Smoke/biomass burning and industry source. National and anthropogenic sources are clearly distinguished. More sources comprise both crustal and combustion / industry – related species. The air quality in the study site is better than the air quality of other major cities in Sri Lanka.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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