



## Characterization and source apportionment of particulate pollution in Colombo, Sri Lanka

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### ABSTRACT

Because of increasing use of vehicles and other human activities in metropolitan Colombo, Sri Lanka, a study of long-term airborne particulate monitoring at two fixed sites was initiated. The specific objectives were to measure the elemental composition of the coarse and fine air particles, to identify the trends in pollution, to identify the main pollutant sources, and to quantify the source contributions. Samples of airborne particulate matter (PM) in the < 2.5 and 2.5–10 μm size ranges (PM<sub>2.5</sub> and PM<sub>10-2.5</sub>) were collected using “Gent” stacked filter samplers at two urban sites in Colombo. The Air Quality Monitoring Station (AQM) of the Central Environmental Authority (CEA) operated during the period of May 2000 to December 2005. The second site at Atomic Energy Authority (AEA) operated from May 2003 to December 2008. Twenty-four hour samples were collected on weekdays. The fine filter samples were analyzed for 18 elements by ED-XRF. The annual averages for PM<sub>10</sub>, PM<sub>2.5</sub>, and black carbon (BC) at the AQM station during 2000–2005 ranged from 50 to 100, 16 to 32, and 8 to 15 μg/m<sup>3</sup>, respectively. The fine fraction data set 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 (Positive Matrix Factorization) to explore the possible sources of the PM at the two study sites. Four factors were found at both sites. The common sources are motor vehicles, road dust, biomass and sea salt. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPILT) back-trajectory model was used to explore possible long range transport of pollution. Smoke and soil dust transboundary events were identified based on fine Si and K in the data base in 2003 and 2004.

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### 1. Introduction

Colombo (Latitude: 6° 49' 35N, Longitude: 79° 51' 45E) shown in Figure 1 is the capital of Sri Lanka. It has an area of 37 km<sup>2</sup> and is located on the west coast of the island. Its population is over 600 000 inhabitants. The city is always congested with a large number of motor vehicles from both private and public transportation systems. The harbor is also located in the city. The weather is tropical with temperatures varying from 25 °C to 30 °C.

Fine particle air pollution (PM<sub>2.5</sub>, particles with diameters ≤ 2.5 μm) has been related to adverse health outcomes (Dockery et al., 1993), poor visibility in Asia (UNEP, 2002; Ramanathan et al., 2005), and long-range transboundary pollution transport (Cohen et al., 2004). Because of the increased use of vehicles and other human activities in Colombo and suburbs, a study of long term airborne particulate monitoring at fixed sites was initiated in 2000 and 2003, respectively.

To protect public health, many countries have instituted air quality standards that set maximum allowable concentrations. For example, the United States Environmental Protection Agency introduced a PM<sub>2.5</sub> standard of 15 μg/m<sup>3</sup> for annual average and 35 μg/m<sup>3</sup> for 24-hr maximum in 2006. In Australia, the PM<sub>2.5</sub> goals are 8 μg/m<sup>3</sup> for annual average and 25 μg/m<sup>3</sup> for 24-hr maximum. Sri Lanka has a fine PM<sub>2.5</sub> permissible level of 25 μg/m<sup>3</sup> for annual average and 50 μg/m<sup>3</sup> for a 24 hr maximum.

This paper presents the results of a multiple year particle characterization study that includes elemental analysis by ED-XRF and the application of data analysis methods for source apportionment. In addition, air parcel back-trajectories were used to identify possible source locations that contribute to the observed concentrations of fine particle pollution at a sampling site in Colombo.

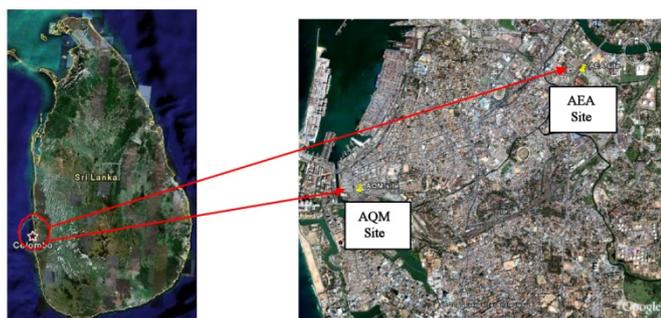
### 2. Techniques and Methods

Sampling was conducted using a “Gent” stacked filter unit particle sampler capable of collecting particulate matter in the range of PM<sub>2.5-10</sub> and PM<sub>2.5</sub> size fraction (Hopke et al., 1997). Samples were collected over 24-hour periods on the weekdays using nuclepore filters with 8 μm pore size for the course fraction and 0.4 μm pore size filter for the fine fraction.

Sampling site 1 is located at the Air Quality Monitoring Station (AQM) in downtown Colombo near the main train station. The samples were collected during the 24-hourly on week days with a flow rate 15 to 18 L/min. Sampling site 2 is located at Atomic Energy Authority (AEA) premises on the northeastern side of Colombo. The sampler was placed on the flat area in the first floor of the AEA building. The AEA building is located near the main road leading to the Colombo metropolitan area.

Mass values were determined by weighing before and after the sample collection with a 24-hour period of equilibration at

50% relative humidity on a microbalance. Mass was determined for both the coarse and fine particle samples.



**Figure 1.** Maps of Sri Lanka (left) and Colombo (right) showing the locations of the two sampling sites.

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 were conducted at Clarkson University (Spectro XLAB-2000) for the fine fraction samples. 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. 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. 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) in the fine filters assuming an average fine particle mass absorption coefficient of  $5.7 \text{ m}^2/\text{g}$ . The elemental analysis methods are mature and have been applied in a large number of prior studies.

Advanced data analysis methods such as Positive Matrix Factorization (PMF) have also been routinely applied to such data sets. US EPA PMF (US EPA, 2010) version 3.0 is a process that quantitatively provides both fingerprints and daily time series plots for major source contribution at a given site. These EPA-PMF processes and their applications have been discussed in detail elsewhere (Reff et al., 2007; Norris et al., 2008).

### 3. Results and Discussion

#### 3.1. PM mass data

About 500 filters were collected during 2000–2008 from two sites. Figure 2 shows the measured  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  mass concentrations for this study period. The  $\text{PM}_{2.5}$  mass concentrations peaked above  $60 \mu\text{g}/\text{m}^3$  in 2002 and 2003. Apart from these increases, there are minor seasonal trends with little excess in the

average annual mass of fine particles and  $\text{PM}_{10}$  over eight years period.

At the AQM station, the annual averages for  $\text{PM}_{2.5}$  showed an increase to 2003 and a decline beginning in 2004. These values are well above the US EPA air quality standards ( $15 \mu\text{g}/\text{m}^3$ ). The annual averages for  $\text{PM}_{10}$  ranged from  $65 \mu\text{g}/\text{m}^3$  in 2000 to  $53 \mu\text{g}/\text{m}^3$  in 2005, again above the value ( $50 \mu\text{g}/\text{m}^3$ ) previously recommended by the USEPA.

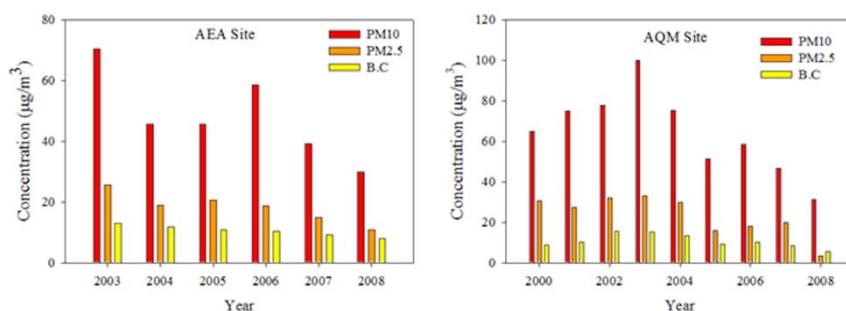
The variation of  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and BC collected at the AEA premises are shown for the period of sampling 2003 to 2008 (Figure 2). The variation of BC is similar to that at the AQM site during the period and annual average was around  $10 \mu\text{g}/\text{m}^3$ . At the AEA station, the annual averages of  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  do not exceed the USEPA recommended values, and showed no consistent pattern in their variation.

#### 3.2. Composition

The average  $\text{PM}_{2.5}$  fine particle composition results over the study period from 2000 to 2008 obtained are shown in Tables 1 and 2. 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 south-eastern Asia (Hopke et al., 2008). The variations in constituent concentrations generally follow the variability in mass concentrations.

**Table 1.** Average elemental concentrations with the standard deviations of  $\text{PM}_{2.5}$  at the AQM site for the period of 2000-2005

Constituent	Mean $\pm$ SD	Median	Maximum
PM ( $\mu\text{g}/\text{m}^3$ )	$29.0 \pm 15.0$	28.7	110.0
BC ( $\mu\text{g}/\text{m}^3$ )	$12.2 \pm 5.9$	11.2	34.6
Na ( $\text{ng}/\text{m}^3$ )	$218.4 \pm 169.5$	163.1	1 018.2
Mg ( $\text{ng}/\text{m}^3$ )	$72.0 \pm 46.1$	58.7	263.1
Al ( $\text{ng}/\text{m}^3$ )	$113.3 \pm 108.0$	85.8	702.2
Si ( $\text{ng}/\text{m}^3$ )	$378.6 \pm 252.1$	319.8	1 683.5
S ( $\text{ng}/\text{m}^3$ )	$709.3 \pm 511.4$	585.8	3 583.7
Cl ( $\text{ng}/\text{m}^3$ )	$145.0 \pm 164.9$	106.3	1 718.7
K ( $\text{ng}/\text{m}^3$ )	$474.0 \pm 360.8$	361.9	2 254.2
Ca ( $\text{ng}/\text{m}^3$ )	$150.4 \pm 165.8$	113.1	1 741.5
Ti ( $\text{ng}/\text{m}^3$ )	$20.4 \pm 16.0$	16.6	129.1
V ( $\text{ng}/\text{m}^3$ )	$6.7 \pm 9.9$	3.5	68.0
Cr ( $\text{ng}/\text{m}^3$ )	$13.7 \pm 11.6$	10.4	90.1
Fe ( $\text{ng}/\text{m}^3$ )	$631.2 \pm 486.9$	523.5	3 247.3
Ni ( $\text{ng}/\text{m}^3$ )	$43.1 \pm 35.0$	35.0	253.9
Cu ( $\text{ng}/\text{m}^3$ )	$130.6 \pm 106.6$	106.4	706.4
Zn ( $\text{ng}/\text{m}^3$ )	$106.4 \pm 91.8$	84.5	656.6
Br ( $\text{ng}/\text{m}^3$ )	$16.0 \pm 9.8$	12.8	79.2
Rb ( $\text{ng}/\text{m}^3$ )	$4.7 \pm 3.5$	3.8	22.6
Pb ( $\text{ng}/\text{m}^3$ )	$37.6 \pm 23.5$	30.3	176.0



**Figure 2.** Annual mean concentrations of  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , and BC at the two sampling sites.

In addition, pseudo-elements (Malm et al., 1994; Begum et al., 2006) were calculated. These variables are combinations of the measured composition values that help to estimate the likely major source types. Soil estimates were obtained by summing the masses of five elements Al, Si, Ti, Ca and Fe when converted to their common oxides. Ammonium sulfate was estimated from the sulfur concentration assuming a fully neutralized aerosol (Malm et al., 1994; Begum et al., 2006).

Non-soil potassium is defined as:

$$K_{non} = (K - 0.6 Fe) \quad (1)$$

Non-soil potassium is a very strong indicator of biomass burning. The average reconstructed mass (RCM) obtained by summing all the analytes (Malm et al., 1994; Begum et al., 2006). This pseudo-element analysis estimates that the average fine particle composition is 48% BC, 9% soil, 11% ammonium sulfate. The average reconstructed mass (RCM), obtained by summing all the analyses and comparing with the gravimetric mass, was  $(70 \pm 30)$  % showing good mass closure for the data (Begum et al., 2006).

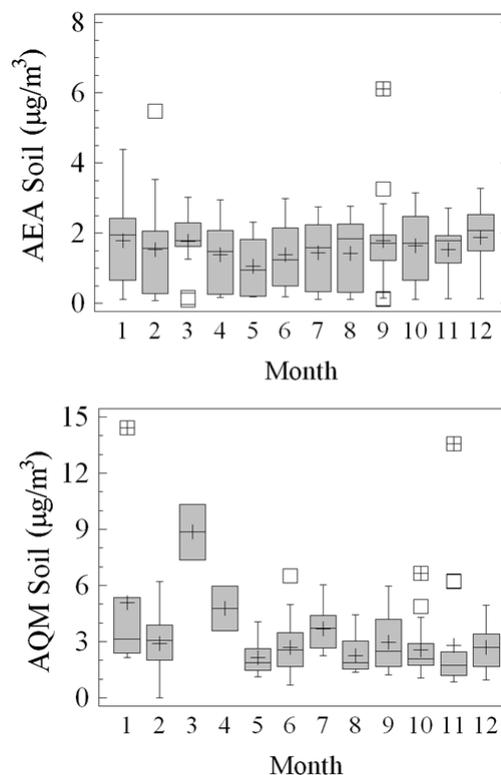
**Table 2.** Average elemental concentrations with the standard deviations of  $PM_{2.5}$  at the AEA site for the period of 2003-2008

Constituent	Mean $\pm$ SD	Median	Maximum
PM ( $\mu\text{g}/\text{m}^3$ )	22.9 $\pm$ 11.9	20.4	72.7
BC ( $\mu\text{g}/\text{m}^3$ )	11.3 $\pm$ 3.6	11.7	23.4
Na ( $\text{ng}/\text{m}^3$ )	109.6 $\pm$ 51.9	105.0	373.2
Mg ( $\text{ng}/\text{m}^3$ )	32.2 $\pm$ 20.8	34.0	132.9
Al ( $\text{ng}/\text{m}^3$ )	113.2 $\pm$ 107.1	71.4	543.1
Si ( $\text{ng}/\text{m}^3$ )	210.9 $\pm$ 141.7	229.0	759.2
S ( $\text{ng}/\text{m}^3$ )	484.7 $\pm$ 304.8	436.4	1990.9
Cl ( $\text{ng}/\text{m}^3$ )	107.5 $\pm$ 87.6	109.1	835.3
K ( $\text{ng}/\text{m}^3$ )	292.2 $\pm$ 152.7	279.7	998.4
Ca ( $\text{ng}/\text{m}^3$ )	113.2 $\pm$ 190.8	92.1	2411.2
Ti ( $\text{ng}/\text{m}^3$ )	12.3 $\pm$ 7.7	12.5	39.8
V ( $\text{ng}/\text{m}^3$ )	5.3 $\pm$ 4.8	4.3	36.5
Cr ( $\text{ng}/\text{m}^3$ )	5.5 $\pm$ 4.9	3.6	32.9
Fe ( $\text{ng}/\text{m}^3$ )	256.3 $\pm$ 215.1	157.4	1355.8
Ni ( $\text{ng}/\text{m}^3$ )	17.0 $\pm$ 15.5	9.3	101.8
Cu ( $\text{ng}/\text{m}^3$ )	53.3 $\pm$ 50.4	26.1	319.6
Zn ( $\text{ng}/\text{m}^3$ )	51.3 $\pm$ 47.2	38.1	329.6
Br ( $\text{ng}/\text{m}^3$ )	9.3 $\pm$ 6.1	9.8	34.5
Rb ( $\text{ng}/\text{m}^3$ )	2.2 $\pm$ 1.5	2.3	11.3
Pb ( $\text{ng}/\text{m}^3$ )	20.8 $\pm$ 12.4	22.5	72.4

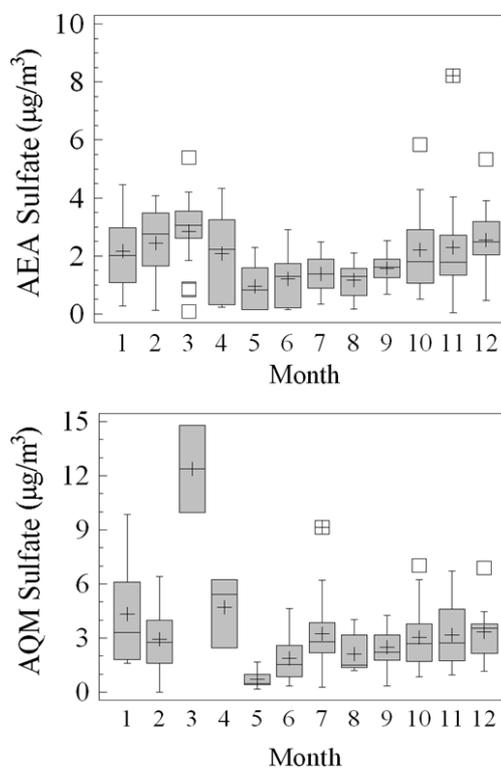
Monthly box and whisker plots for the study period for concentrations of soil and ammonium sulfate are given in Figures 3 and 4, respectively. The (+) sign denotes the monthly mean, the horizontal bar represents the monthly median and the hatched boxes contain 25<sup>th</sup> to 75<sup>th</sup> percentile range of the values for that month. Outliers are shown as dots for the month.

It can be seen that the concentrations at the AQM site are consistently higher than at the AEA site for both soil and sulfate. Although sulfate is normally considered a regional pollutant, there is significant sulfur in diesel fuels in Sri Lanka. The AQM site is adjacent to the major railroad station in Colombo and the trains are diesel powered. There is also substantial light and heavy duty vehicle traffic in this area. There may also be an impact from marine diesel emissions from ships in the harbor (Kim and Hopke, 2008). Thus, the higher source density at the AQM site led to higher concentrations of both soil and sulfate. There were particularly high AQM values observed in samples collected in February. February is often a period when air quality is influenced by long-range transport and this possibility will be discussed later

in this paper. Unfortunately only 10 samples were collected in this month over the whole sampling period.



**Figure 3.** Monthly distributions of soil concentrations as displayed in box and whisker plots.



**Figure 4.** Monthly distributions of  $K_{non}$  (Smoke) concentrations as displayed in box and whisker plots.

If the relative concentrations of soil and ammonium sulfate at the AQM site are considered on a percentage  $PM_{2.5}$  mass basis, they showed distinct seasonal trends. Soil was nearly twice as high as a percentage of total mass in the drier months, while ammonium sulfate showed the opposite trend being higher in the wetter months. In Colombo, the monsoon period is from May to September with heavy rainfall during this interval. Airborne soil is largely resuspended road dust. During the dry season, large quantities of dry dust can be resuspended from unpaved or partially paved roads. Thus, it dominates the concentration relative to sulfate during the dry season. The road emissions are reduced during rainy periods. The rain washes soil from paved roads and reduces emissions from unpaved roads. The sulfate emissions are relatively constant through the year and represent a higher percentage of the measured mass concentrations during the monsoon period.

### 3.3. Source apportionment using EPA positive matrix factorization (EPA – PMF)

EPA PMF Version 3 (US EPA, 2010) was used with  $PM_{2.5}$  elemental data from each site to determine the factors that provided a mass apportionment among the different kinds of sources. The fractional elemental contributions associated with each profile, together with the source contributions to the total fine mass were determined by PMF. A significant choice made by the user is the number of factors. Solutions were chosen that provided good fits to the data as determined by examination of the distributions of the scaled residuals and are physically interpretable sources.

The fine fraction data sets from the two sites included BC and major elements (Na, Mg, Al, Si, Cl, Fe, Zn, Ni, Cu, V, S, Br, Pb, Cr, K, Ca and Ti) The AQM data set included 144 samples covering the period of May 4, 2000 to December 29, 2005 while the AEA data set had 183 samples covering the period of June 18, 2003 to February 25, 2008. They were analyzed separately by PMF to explore the possible sources of the PM. The apportionments of the fine particle mass concentrations for the two sites are presented in Tables 3 and 4. The analysis revealed only four factors for each site. The broad source categories for both sites are motor vehicles, road dust, biomass and sea salt. Plots of the source profiles and corresponding mass contributions for the AQM and AEA sites are presented in Supporting Material (SM) (Figures S1 to S4).

**Table 3.** Source apportionment for the fine particulate matter mass measured at the AQM site

PMF Factor	High loading	Main Sources
Factor 1 48%	BC, S, K, Fe, Pb	Motor Vehicles
Factor 2 27%	Al, Si, Ti, Ca, Pb, Fe, K	Road Dust
Factor 3 21%	BC, K	Biomass Burning
Factor 4 4%	Na, Cl, BC	Sea Salt

**Table 4.** Source apportionment for the fine particulate matter mass measured at the AEA site

PMF Factor	High Concentrations	Main Source
Factor 1 17%	BC, S	Motor Vehicles
Factor 2 9%	Si, Ti, Ca, Pb, Fe, Ni, Cu, Zn, Pb, Br	Road Dust
Factor 3 66%	BC, K, S	Biomass Burning
Factor 4 9%	Na, Cl, BC, Ca	Sea Salt

In both cases, fewer sources were resolved than has often been observed in other locations (e.g., Chueinta et al., 2000; Begum et al., 2004; Santoso et al., 2008). In these two locations, the sites are surrounded by these major areas sources (roads, housing) so that there is little variability in the mixture of sources from day to day. Although there are differences in total emissions from day to day, the relative proportions of many small sources do

not appear to vary sufficiently that they can be resolved in these data sets.

The first factor for the AQM site contributes 48% of average  $PM_{2.5}$  mass concentration and includes BC, S, and minor quantities of metal species. The second factor includes soil elements with BC. This factor with high Si, Na, Mg, Cl, Cr, Fe, Ni, Cu, Pb, Zn, and Br is assigned as the road dust being produced primarily from local traffic, and it contributes 27% of the mass. The third factor is related to K, and BC concentrations in biomass burning which contributes 21% of the average mass. Pb and Br are generally associated with automotive exhaust and are significant in the fine fraction (Seneviratne et al., 1999). Unleaded gasoline was introduced into Sri Lanka in 2003 (ADB-CAIA, 2006). Thus, the nature of automobile emissions has been changing over the time interval of these measurements. The final factor includes Na, Cl, along with BC and contributes 4% to the mass. It also includes the highest concentration of V along with Ni suggesting the impact of ship emissions (Kim and Hopke, 2008).

Among the four factors found for the fine particulate matter in AEA station, the profiles are similar to those at the AQM site, but there are quite different source contributions. The highest contributions were from biomass burning (66%). The facility is surrounded by local housing that uses biomass for cooking. The next highest contributions were from motor vehicle traffic (17%) and road dust (9%). Although there are major roads in the vicinity, there is much less traffic than at the central AQM site. Sea salt contributed 9% of the average mass concentration.

### 3.4. Back-trajectory techniques

An important question is whether the observed concentrations arise entirely from local sources or if there are contributions from distant sources through long-range transport. Recently, Begum et al. (2010) have shown that examination of the highest concentrations indicates that they are often associated with transported particulate matter. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) back-trajectories (Draxler and Rolph, 2010) were used to trace possible medium and long-range transport of pollution to the AQM measurement site.

Soil dust and smoke transboundary events were identified based on the pseudo-elements soil and  $K_{non}$ , an indicator of smoke, in the data base from 2000 to 2006 (Figures 5 and 6). Evidence of these events was identified. There are three major soil events on November 9, 2000, January 14, 2004, and February 21, 2004. Air parcel back-trajectories beginning at noon on these dates are presented in Figures S5–S7 (see the SM). According to the NASA Natural Hazards, there was a dust storm that began on the 15<sup>th</sup> of February 2004 in the Arabian Peninsula (Qatar) (Figure S6 in the SM shows a later storm observed in satellite images on February 21, 2004). Samples were collected on February 21<sup>st</sup> in which soil could be estimated to be  $10 \mu\text{g}/\text{m}^3$  even though the source was 3 500 km from Colombo.

There were 7 large “smoke” episodes with concentrations above  $450 \text{ ng}/\text{m}^3$  observed in Figure 6 on July 12, 2000, February 4, 13, 14, 21, and 23, 2004 and January 1, 2006. The 2000 trajectories (see the SM, Figure S9) extend westward into the area east of Africa and may have collected Saharan and Arabian Peninsula dust leading to high K in ratios relative to Fe that appear to be “smoke” rather than “soil”. In February 2004, there was a more complex situation. The trajectories on days when samples were collected are shown in Figures S7, S10–S13 (see the SM). In general, these trajectories pass over Northern India where smoke from agricultural fires backs up against the Himalaya Mountains. They then pass westward into areas where they might also accumulate soil dust. The high K on January 1, 2006 was from fireworks and thus, trajectories were not calculated.

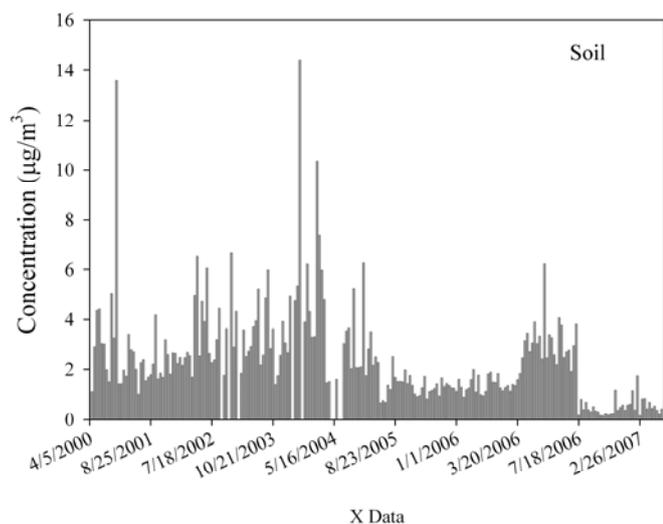


Figure 5. Time series plot of the daily soil concentrations.

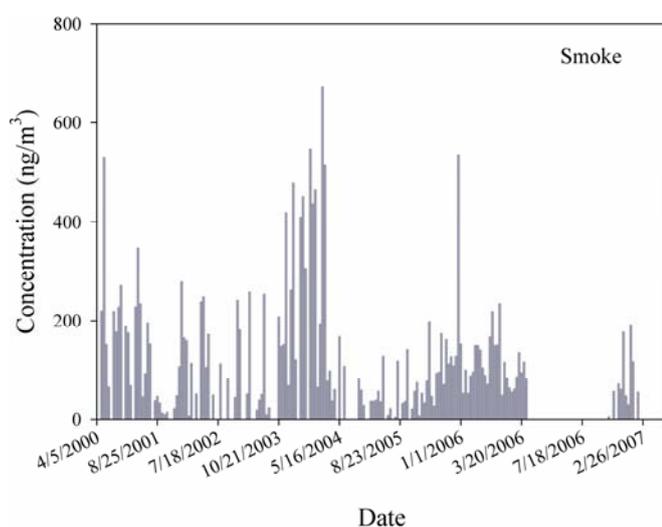


Figure 6. Time series plot of the daily smoke concentrations.

#### 4. Conclusions

The chemical composition data of fine air particulate matter ( $PM_{2.5}$ ) collected from Colombo (AQM) and Orugodawatta (AEA site) were studied using EPA-PMF to explore the possible emissions sources. Factors that have been resolved from two sites show very similar chemical composition profiles. Additionally, few different chemical compositions have been obtained due to local difference. HYSPLIT back-trajectory technique is very handy tool to get an identification of various transboundary events. The influence of a dust storm in Qatar, Iran, on February 15, 2004, could be clearly observed by this technique even though the source region was 3 500 km from Colombo. Similarly, transport of smoke from Northern India in December 2003 could be observed.

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#### Supporting Material Available

Source profiles resolved from the data collected at the AQM site (Figure S1), Source contributions resolved from the data collected at the AQM site (Figure S2), Source profiles resolved from the data collected at the AEA site (Figure S3), Source contributions resolved from the data collected at the AEA site (Figure S4), Back trajectory for November 9, 2000 showing transport from China and Mongolia (Figure S5), Back trajectory for January 14, 2004 (Figure S6), Back trajectory for February 21, 2004 (Figure S7), NASA satellite image of Qatar area on February 21, 2004 (Figure S8), Ensemble of back trajectories for July 12, 2000 (Figure S9), Ensemble of back trajectories starting on February 4, 2004 (Figure S10), Back trajectories starting at different heights on February 13, 2004 (Figure S11), Back trajectories starting at different heights on February 14, 2004 (Figure S12), Back trajectory calculated for February 23, 2004 (Figure S13). This information is available free of charge via the Internet at <http://www.atmospolres.com>.

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