International Journal of Earth, Energy and Environmental Sciences
ISSN: 2517-942X
Vol:6, No:4, 2012

Chemical Analysis of $PM_{2.5}$ during Dry Deforestation Season in Southeast Asia

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***Abstract*—**In Southeast Asia, during the dry season (August to October) forest fires in Indonesia emit pollutants into the atmosphere. For two years during this period, a total of 67 samples of 2.5 μm particulate matters were collected and analyzed for total mass and elemental composition with ICP - MS after microwave digestion. A study of 60 elements measured during these periods suggest that the concentration of most of elements, even those usually related to crustal source, are extremely high and unpredictable during the haze period. In By contrast, trace element concentration in non - haze months is more stable and covers a lower range. Other unexpected events and their effects on the findings are discussed.

***Keywords*—**Haze, ICP - MS, Particulate Patter, Transboundary Air Pollution.

I. Introduction

IN the past few decades, transboundary air pollution during the dry season in southeast Asia has been attracted much attention due to health problems associated with the particulates. These annual fires are mainly caused by land clearing and "slash and burn" agricultural practices in Indonesia, particularly Sumatra and Kalimantan. This is done in order to clear or rejuvenate the land for cultivation and planting. These agricultural burning activities are common during the traditional dry season between August to October [1-3] that can release massive transboundary air pollution (it has been labeled haze by the general public) at times, and envelope Singapore, Malaysia, Brunei and Indonesia. The direct effect of these fires is the production of large amounts of particulate matters in the region, causing visibility and health problems within Southeast Asia.

Although the phenomenon of fires occurs each year from the late 1990s, relatively few studies have been performed including studies of CO and SO_2 contribution during haze period [1], [2] survey on impact of biomass burning on rainwater acidity and composition [3], investigation on haze transportation [4]–[6] and study of composition and concentration of semi-volatile organic compounds [7], [8].

Particulate matter is categorized as one of the six criteria pollutants, and the most important in terms of complexity for both physical and chemical properties. In order to understand the responsibility of particulate matter in transboundary air pollution and atmospheric chemistry matters, it is important to study their detailed chemical composition.

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Studies on particulate matter in Southeast Asia include reports of total mass, organic carbon/elemental carbon (OC/EC) and water-soluble ionic and organic species over a limited period of time, , but we are not aware of a comprehensive study on elemental analysis of particulate matter during the dry deforestation season in Singapore [9] – [15].

In this work, particulate matter with aerodynamic diameter 2.5 micron ($PM_{2.5}$) or less collected over two years. These fine particles are able to scatter solar radiation, and as a consequence, they often have a large impact on local and regional visibility and contribute to the atmospheric reflection, thus affecting regional and global climate [16] – [18]. In addition, the rates of some chemical reactions responsible for dry and wet acidic precipitation can be increased since these fine particles can be entrained into clouds in the boundary layer [3].

II. Experimental Section

A. Sampling Site

$PM_{2.5}$ sampling was conducted at the roof top of School of Physics and Mathematics (SPMS) around 15 meters above the ground and 67 meters above sea level. This sampling site is situated in the western part of Singapore and is influenced by emissions from major power plants, incinerations, chemical industries and petroleum refineries located in Jurong Island, Tuas and Jurong Industrial Estate and urban vehicular traffic as well.

Singapore is located in Southeast Asia, south of West Malaysia, south of Java in Indonesia and east of Sumatra in Indonesia with latitudes between 11090 N to 11290 N and longitudes between 1031360 E to 1041250 E.

Singapore includes the main island of Singapore and 63 islets within its territorial waters. The main island is about 42 km from west to east and 23 km from north to south.

Because of its geographical location and maritime coverage, its climate is characterized by uniform temperature and pressure, high humidity and abundant rainfall. The climate of Singapore can be divided into two main seasons, the Northeast Monsoon and the Southwest Monsoon season, Fig. 2, separated by two relatively short inter-monsoon periods.

Table I has summarized detailed information about the Singapore climate.

B. Method of Sampling and Analysis

Based on our program, $PM_{2.5}$ sampling started in October 2009 and ended in December 2011 with totally 588 filter samples. For this study, a total 67 samples from August to September for years 2010 and 2011 were analyzed.

TABLE I
SINGAPORE WEATHER INFORMATION

Classification	Duration	Wind Direction	Rainfall
Northeast Monsoon	December - March	from the land toward the sea (Northeast winds)	Continues heavy rain
Southwest Monsoon	June - September	from the sea toward land (Southeast/Southwest Winds)	Isolated to scattered late morning and early afternoon showers
Pre South-West Monsoon	late March - May	Light and variable winds	afternoon and early evening showers often with thunder
Pre North-East Monsoon	October - November	Light and variable winds	Scattered showers with thunder in the late afternoon and early evening.

These sampling periods covered two sever haze periods in September 2010 and October 2011. All samples were collected on 200×250 mm (Paliflex, EW-200010) quartz fiber filters using an Echotech (HiVol – 3000, Australia) high volume air sampler. The sampling period was 24 to 48 hours based on weather conditions with a 67 $m^3 h^{-1}$ flow rate through the filter.

Fig. 1 Singapore Map (A), sampling site location towards main road (B) and Industrial areas (C), Sampling site, SPMS roof top, NTU (D)

Concentration of 60 elements was obtained using an Agilent 7700 series Inductively Coupled Plasma-Mass Spectrometer (Japan) equipped with a 3rd generation He reaction / collision cell (ORS^3) to minimize interferences. Final optimized instrumental operating parameters for this application are summarized in Table II. The developed methods of microwave digestion and ICP - MS analysis were evaluated using 1648a standard reference material (SRM 1648a) of Urban Particulate Matter. Briefly, the average analytical results are good agreement with the related certified values with the difference between the two values being less than 20% for most elements.

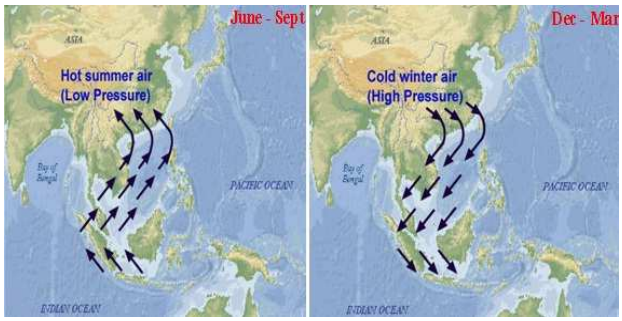


Fig. 2 Northeast Monsoon (Left) and Southwest Monsoon (Right)

C. Reagent

Ultra pure 65% percent HNO_3 was purchased from J. T. Baker, Canada, whereas 30% HCl was purchased from Merck, Germany. ICP-MS single element calibration standards (1000 ppm) were obtained from High Purity Standards (Charleston, SC, USA), Inorganic Venture (Christiansburg, VA, USA) and Environmental Express (Charleston, SC, USA). For all solution preparation and dilutions, high purity water with 18.2, MΩ cm resistivity was used (ELGA, PURLAB Option – Q, UK). HNO_3 – HCl 2% solutions were used as the background matrix of reagent blank, calibrations standards and digested samples to avoid errors related to matrix inconsistencies.

TABLE II
OPERATING CONDITION AND INSTRUMENTAL SETUP FOR ICP-MS

Parameter	ICP-MS
Instrument	Agilent 7700 Series (Japan), MicroMist nebulizer, Quartz spray chamber, 3 channel peristaltic pump
Tuning Solution	Li, Mg, Y, Ce, Tl, Co; 1 µg/L; 2% HNO_3
RF Power	1550 W
Nebulizer gas flow	0.7 L min^{-1}
Auxiliary gas flow	0.35 L min^{-1}
Lens voltage	Extract 1: 0 V, Extract 2: -200 V, Omega Bias: -90 V, Omega Lens: 10.8 V
Octopole Parameter	Octp RF: 200 V, Octp Bias: -18 V
Cone	Ni Sampling cone, Ni Skimmer cone
Collision Cell	3rd generation Octopole Reaction System (ORS^3), He mode
Sample Parameter	Agilent Integrated. Autosampler (I-AS), sample uptake rate 0.3 mL min^{-1}, rinsing time 190 s (Ultra pure water and 2% HNO_3)
Data Acquisiation	3 readings per replicate, 3 replicates, total 123 sec

III. RESULT AND DISCUSSION

During two last years, Singapore experienced two haze periods in 16th to 23rd October 2010 and 8th to 13th of September 2011 with maximum pollution standard index (PSI) values of 96 and 62 respectively.

Usually in August, Southwest Monsoon conditions begin to weaken in the region due to frequent incursions of moist easterly winds blowing from the equatorial Pacific Ocean towards the region. During September, Southwest Monsoon conditions continue to weaken. Towards the last week of the month, the region begins to display signs of gradual transition into Inter-Monsoon conditions, with winds weakening in strength and becoming more variable in direction. Southwest Monsoon conditions continued to prevail in the first half of October and began to weaken around middle of the month as Inter-Monsoon conditions set in over the region. The prevailing southeasterly to southwesterly winds over the ASEAN region also gradually weaken to become more light and variable. The Southeast Asian weather is strongly influenced by the El Nino Southern Oscillation (ENSO) during this period. The southwest monsoon greatly controls the cross equatorial transport of haze from Sumatra and Kalimantan to Peninsular Malaysia and Singapore. Fig. 3 shows a view from SPMS roof top on a clear and hazy day on June 20, 2011 and on September 10, 2011 respectively [19]

The average concentration of all elements from a total 67 $PM_{2.5}$ filters during a 3 month sampling period in years 2010 and 2011 are categorized in clear and hazy days and presented in Table III.

Fig. 3 combination photo shows a view from SPMS roof top on a clear day (top) on June 20, 2011 and shrouded in haze (bottom) on September 10, 2010

As can be seen from Table III, the concentration of most of elements, even those usually related to crustal sources, are extremely high and unpredictable during the haze period. By contrast, trace element concentrations in non haze month are more stable and cover a lower range. Fig. 4 shows time trends for the concentration of all measured elements during the sampling periods.

It is interesting to note that significant increases in the concentration for most of the elements (Li, Be, B, Na, Mg, Al, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Ni, Co, Cu, Ga, Ge, As, Se, Rb, Sr, Zr, Mo, Rh, Pd, Ag, Sn, Te, Cs, Ba, La, Ce, Gd, Hf, Os, Pt, Tl, Pb, Bi, Th, U) are observed during both haze periods (16th to 23rd Oct 2010 and 8th to 13th Sep 2011). Although the average concentration in hazy days is higher compared to clear days, for some of these elements (Ti, V, Ni, Co, Zn, Se, Cu, Ga, Ge, As, Zr, Cd, Mo, Rh, In, Sn, Te, Cs, Ba, Hf, Os and Tl), the slightly higher concentration ranges in clear days suggest that it can be apparently due to variances in their local industrial emissions.

Since measured trace elements during haze period could be affected by background particles, source apportionment is more difficult. These elements may have two different sources: the combustion of vegetation or the remobilisation of particles that were previously deposited on the vegetation.

An interesting feature in this data is a local event, called the Hungry Ghost Festival, that has surprisingly increased concentrations of several elements (B, Na, Mg, Al, K, K, V, Mn, Fe, Ni, Co, Zn, Cu, Ga, Sb, Ba, Hf, W, As, Rb, Zr, Rh, Tl, Pb, Bi) in 24th August 2010 and 14th August 2011.

TABLE III
MESURED TRACE ELEMENT CONCENTRATIONS (NG M^{-3})

	Hazy Days				Clear Days			
Elements	Ave Conc. Range	Std	Median	Geomean	Ave Conc. Range	Std	Median	Geomean
Li	19.9805 8.2508-37.6893	11.8245	16.0051	17.061	6.0311 1.7728-16.9802	3.6109	4.7615	5.2008
Be	0.117 0.0266-0.267	0.0746	0.1032	0.0981	0.0424 0.0172-0.118	0.0244	0.0338	0.0371
B	6235.4271 1456.4189-11999.9731	3792.425	5064.231	5128.932	3412.5945 6.239-46791.6088	5947.141	2173.601	2025.299
Na	20253.6447 7572.6073-38768.6924	12298.2	14565.65	17054.15	9465.6114 161.9391-26013.8209	5660.016	8180.6	7310.855
Mg	2548.1589 914.1165-6877.5636	2257.433	1331.779	1878.834	1324.9365 18.2321-3939.8013	827.2794	1095.084	1010.757
Al	12698.2789 3106.0555-54567.6965	18664.92	4498.426	7196.68	2979.2342 26.9216-10224.3892	2192.82	2641.472	2102.907
K	9046.9679 2874.5809-19377.3105	7245.986	6080.964	6830.997	3049.1087 361.0449-8573.2324	1915.829	2663.004	2474.773
Ca	12267.9515 3364.189-33232.3749	10295.02	7562.698	9502.522	5726.4527 102.1492-16506.777	3459.391	4934.807	4503.695
Sc	1.0631 0.4614-1.9828	0.6739	0.6819	0.8839	0.3615 0.0336-1.4587	0.2611	0.2716	0.2865
Ti	48.4327 12.4474-131.9501	41.7929	28.2742	36.1818	28.4562 1.2344-511.1585	65.3998	18.1431	17.0302
V	45.8442 23.0278-77.5866	24.3747	31.3288	40.5097	22.2372 2.4716-205.8131	27.4858	17.2131	15.7745
Cr	10.4637 3.3998-22.7569	7.5444	6.7607	8.2639	5.097 1.7671-14.9339	2.7544	4.3623	4.5337
Mn	33.094 18.6511-59.206	14.1991	28.6033	30.7825	17.2744 4.7253-51.7692	8.8575	14.8143	15.4004
Fe	368.4022 158.0686-752.2069	213.0715	323.6812	320.7546	223.7371 62.1488-676.6387	131.1478	201.9924	192.5585
Ni	11.7127 5.826-21.9632	5.2776	11.3491	10.7984	9.5802 2.4353-28.5751	4.9336	8.991	8.463
Co	0.3895 0.1538-0.6663	0.2216	0.2986	0.3348	0.3644 0.1249-0.9488	0.1781	0.3118	0.3294
Zn	1839.2445 377.1318-6270.9576	2110.562	904.6238	1130.853	1725.4401 98.034-8499.088	1633.749	1293.603	1103.508
Cu	8.9657 3.3483-21.3342	6.1313	6.6804	7.5661	10.2918 3.3529-35.4614	6.2999	8.2711	8.9025
Ga	128.4386 25.0008-245.5161	90.6852	130.2056	94.826	130.3225 15.568-516.2045	104.7421	110.9063	95.6512
Ge	0.0985 0.0161-0.2207	0.0834	0.0538	0.0673	0.0544 0.0048-0.7153	0.0934	0.0377	0.0369
As	1.5344 0.5538-3.6278	1.1253	0.9159	1.238	1.3407 0.4256-6.881	0.9309	1.1407	1.1592
Se	0.9285 0.4167-1.9307	0.5853	0.5675	0.794	0.5263 0.1197-2.6682	0.3796	0.4525	0.4378
Rb	3.171 1.092-6.1874	2.1126	2.349	2.6179	2.1198 0.5128-4.5137	0.9595	1.9726	1.9114
Sr	98.1222 22.5823-329.8406	108.4363	46.6733	65.1589	61.944 0.4141-177.0685	37.7922	55.3351	45.0738
Zr	3.4523 1.0569-9.1468	2.7518	2.8001	2.707	2.9074 0.2071-34.0478	4.3515	2.0593	2.0234
Mo	0.5195 0.2038-1.1966	0.3421	0.431	0.437	0.3952 0.0582-2.2358	0.3511	0.3241	0.2929
Rh	0.0719 0.027-0.2014	0.0628	0.0418	0.0553	0.045 0.0013-0.2443	0.036	0.0355	0.0342
Pd	0.0281 0.007-0.0845	0.0288	0.0104	0.0183	0.014 0.0008-0.046	0.0093	0.0119	0.0111
Ag	0.0914 0.0285-0.2046	0.0641	0.0827	0.0737	0.0475 0.0138-0.1897	0.0295	0.0378	0.041

TABALE III
CONTINUE

	Hazy Days				Clear Days			
Elements	Ave Conc. Range	Std	Median	Geomean	Ave Conc. Range	Std	Median	Geomean
Cd	0.3368 0.1065-0.7885	0.2436	0.2198	0.2746	0.3714 0.0993-1.3825	0.2313	0.3054	0.3202
In	0.0142 0.006-0.0348	0.0102	0.0095	0.0118	0.018 0.0038-0.1043	0.019	0.011	0.0132
Sn	2.1301 1.2815-3.6169	1.0571	1.4409	1.9279	1.0155 0.1586-5.3134	0.8119	0.891	0.8058
Sb	0.0468 0.0209-0.0987	0.0274	0.0383	0.0412	0.2856 0.0051-14.9105	1.9369	0.0306	0.0304
Te	0.0203 0.0067-0.0491	0.0166	0.0138	0.0156	0.0115 0.0015-0.0596	0.009	0.0093	0.009
Cs	0.1634 0.0583-0.3405	0.1183	0.1289	0.1321	0.1048 0.0346-0.2946	0.0527	0.1011	0.0927
Ba	475.4556 121.3698-1181.1355	379.4028	270.1176	360.0717	506.5501 2.6336-2299.7547	519.7281	285.7684	278.892
La	1.5764 0.3385-3.7773	1.2279	1.1335	1.2157	0.7923 0.0655-2.5052	0.519	0.6606	0.634
Ce	3.4536 0.9891-7.1468	2.3496	3.325	2.755	2.2478 0.0997-6.9809	1.5094	1.9306	1.7543
Gd	0.2783 0.0637-0.6296	0.1921	0.2416	0.2195	0.1285 0.0047-0.3413	0.0789	0.1177	0.1025
Hf	0.0683 0.0248-0.1285	0.0399	0.0632	0.058	0.0672 0.0049-0.917	0.116	0.0464	0.0465
W	0.1431 0.0181-0.2632	0.0974	0.1579	0.1019	0.2479 0.007-5.4607	0.7029	0.1301	0.1134
Os	0.0327 0.0064-0.0749	0.0234	0.0288	0.0252	0.0161 0.0016-0.0976	0.0151	0.0119	0.012
Pt	0.0235 0.0066-0.0677	0.0224	0.0115	0.0166	0.0124 0.001-0.0416	0.0087	0.0105	0.0096
Tl	0.0527 0.0263-0.0865	0.0245	0.046	0.0478	0.0502 0.0213-0.1477	0.0253	0.0434	0.0455
Pb	10.6097 3.1128-22.3145	7.8519	6.4598	8.131	7.7345 2.2862-18.2718	3.5246	7.1562	6.9469
Bi	0.4704 0.0768-1.5855	0.5326	0.2228	0.2994	0.3195 0.0687-0.7902	0.1623	0.2961	0.283
Th	0.5154 0.1575-0.9934	0.2788	0.4603	0.4485	0.2376 0.0084-0.6958	0.1615	0.2003	0.1871
U	0.1772 0.0692-0.359	0.1165	0.1265	0.1478	0.1058 0.005-0.3011	0.0572	0.0933	0.0894

The Hungry Ghost Festival is a traditional Chinese festival and holiday celebrated by the Chinese in seventh month of lunar calendar. The fifteenth day of the seventh month in the lunar calendar is called Ghost Day, and at the beginning of this day people burn bank notes, joss paper and incense all over Singapore. These results are consistent with findings during haze periods that suggest vegetation and papers release same elements.

Another important finding was related to Shell Refinery fires in 28th Sep to 2nd Oct 2011in Pulau Bukom Island, Singapore. The highest concentration in this period was observed for V, Mn, Fe, Ni, Co, Cu, Ag, Rh, Mo, Sr, Ti, Pt and Os.

Other periods with high concentration of elements are listed in Table IV. According to the effected elements, it is likely that these unexpected events are related to local industry events these unexpected events are related to local industry events. However recorded the PSI for these events is not more than 30.

TABLE IV
UNEXPECTED INDUSTRIAL EVENTS

Period	Effected Elements
1 Aug 10	Li, Be, Mg, Ca, Sc, Ti, Cr, Mn, Fe, Ni, Co, Cs,
15 – 17 Sep 10	V, Mn, Fe, Ni, Co, Cu, Mo, Ag, Sn, Te, Tl.
23 – 24 Aug 11	Na, Mg, Al, K, Ca, Sc, Rb, Sr, Rh, Pd, Pt, Tl, Pb, Th, U.
12 – 14 Oct 11	Be, B, Na, K, Sc, Ti, Ge, V, Cr, Co, Zn, Ga, Ge, As, Se, Rb, Sr, Zr, Mo, Rh, Pd, Cd, In, Os,

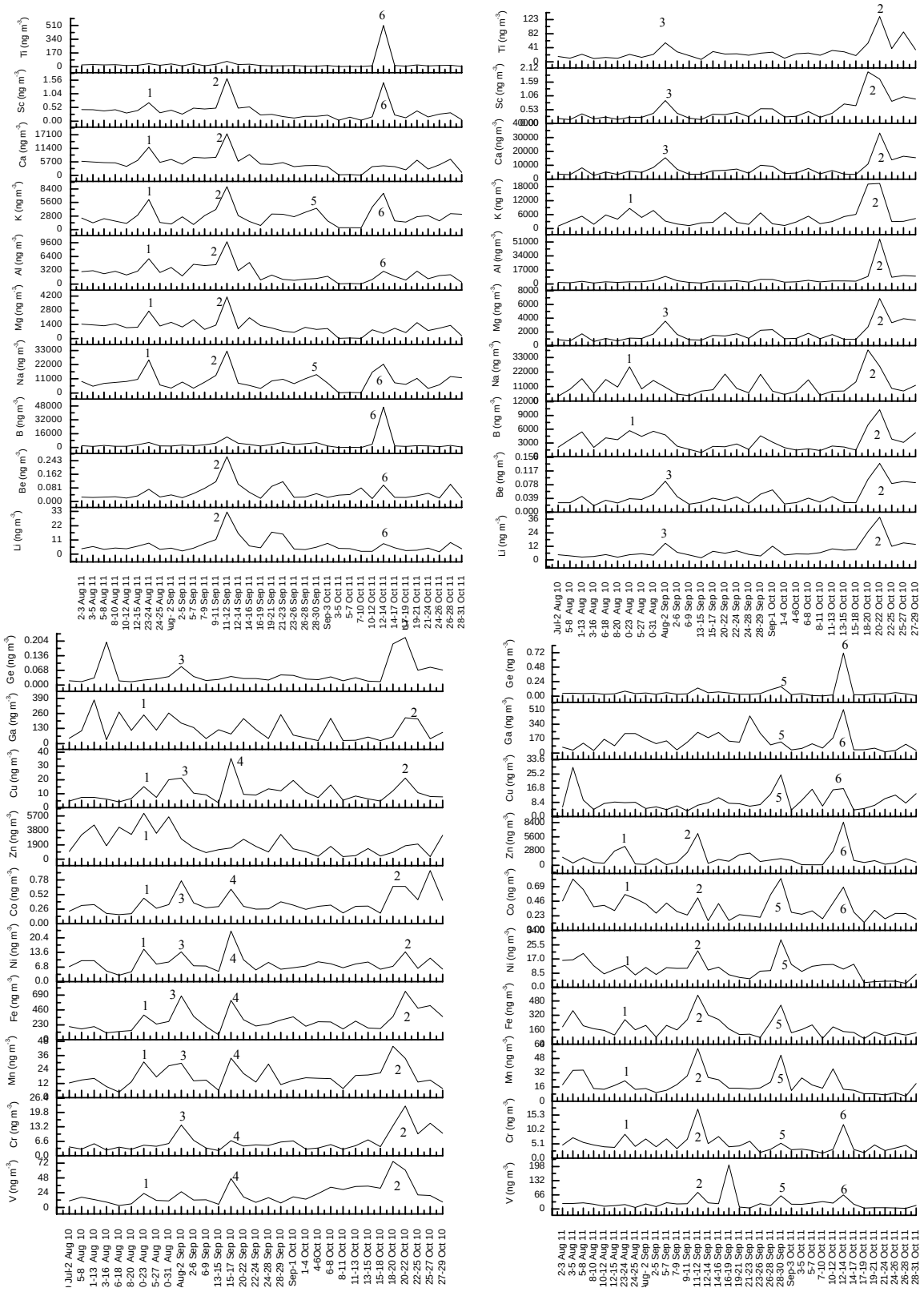
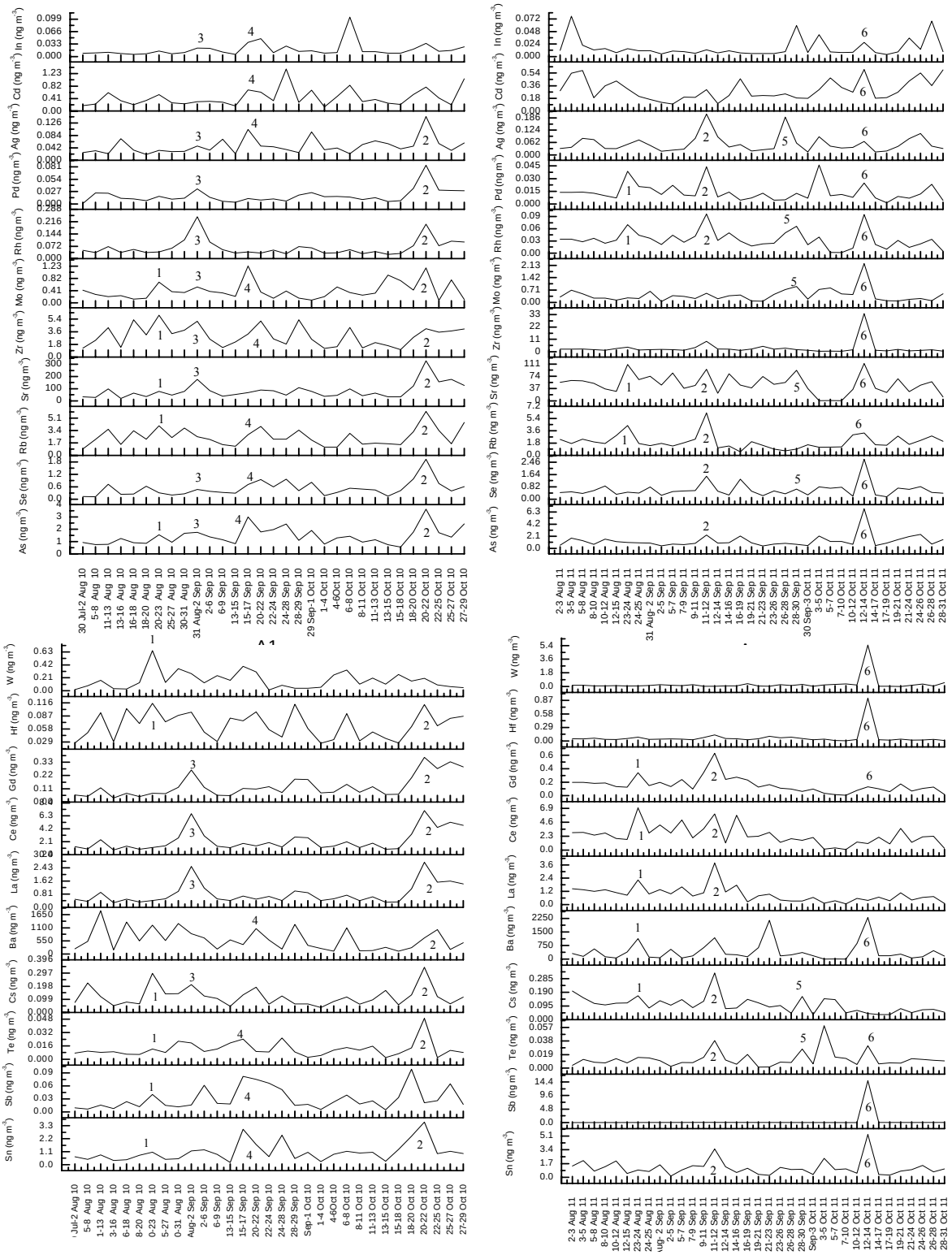


Fig. 4 Time Trends Element Concentration, (1) Ghost Day , (2) Haze Periods, (3), (4) and (6) Unexpected Industrial Events, (5) Shell Oil Refinery Fires.



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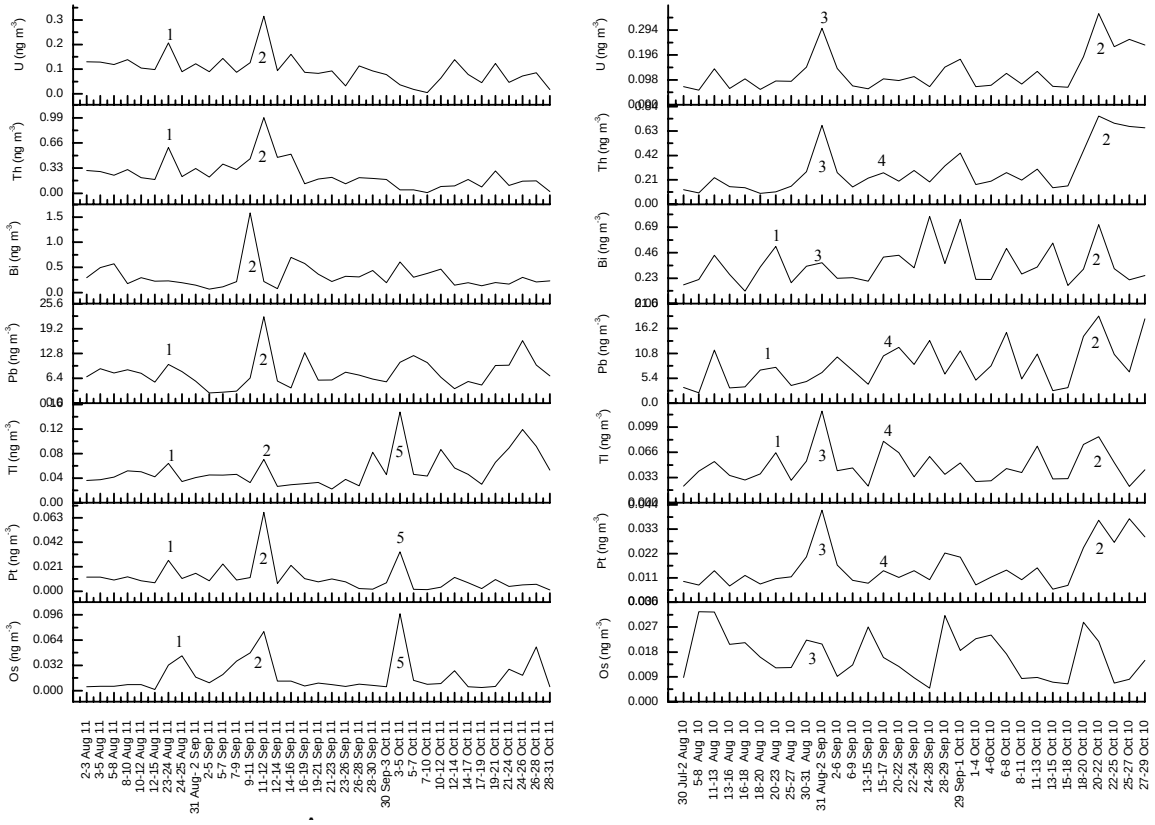

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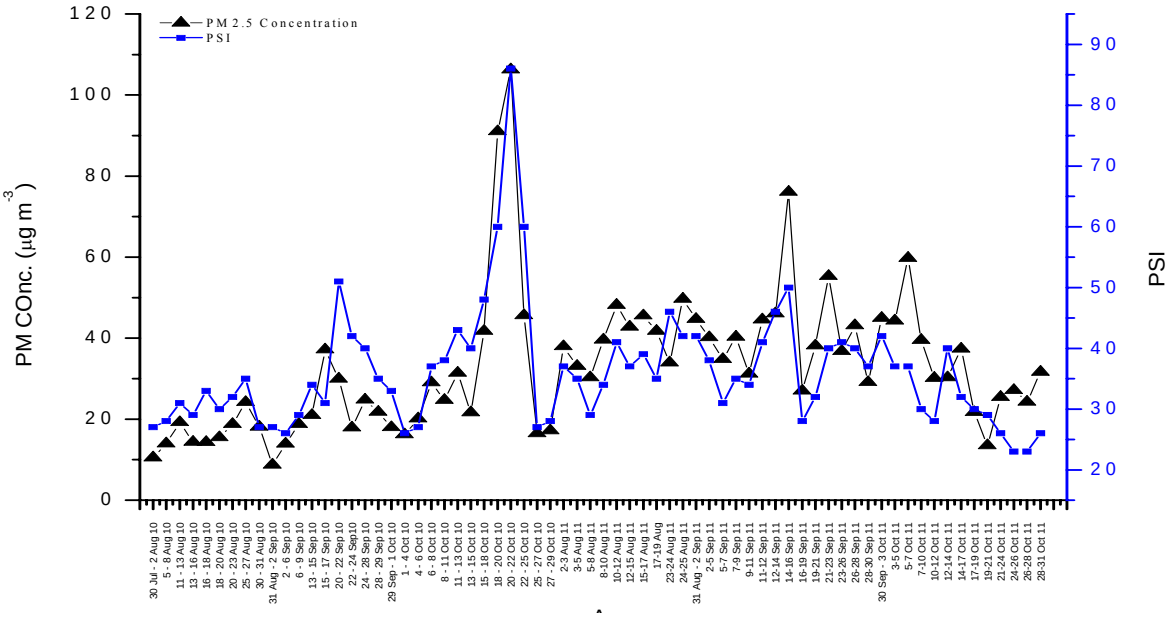

Fig. 5 Temporal Changes in $PM_{2.5}$ concentrations and PSI from August to October 2010 and 2011 (PSI from National Environment Agency of Singapore).

Fig. 5 shows the average concentrations of $PM_{2.5}$ and PSI readings which are indicative of ambient air quality. An index value between 1 to 50 indicates that the air quality is in the good range for that day, 51 to 100 in the moderate range and a value of >100 would mean that the air quality is unhealthy [19]. The pollution index converts the measured concentration of air pollutants (PM_{10}, O_3, CO, SO_2 and NO_2) to a scale of 0 - 500. However, during haze episodes the PSI is always based on PM_{10} concentrations, as these greatly exceed those of other pollutants [12]. Since previous studies suggested that forest fires produce mainly fine-mode particles we expect that PSI and $PM_{2.5}$ concentration data are in good agreement.

The areas of forest fires ranged from several hundred kilometers to around 2000km from Singapore. Day-to-day particle concentrations change significantly in response to spatial and temporal trends of meteorological factors, such as wind conditions (direction and speed) and rainfall (number of rainy days and amount), and of fire activity and Industrial events.

As shown in Fig. 5, both the PSI and the PM concentrations followed the same trend during this sampling period, signifying that the PSI for the day was determined by the prevailing PM concentrations. It could also be seen that there was a periodic increase in the values of both the PSI (maximum 86) and the $PM_{2.5}$ (maximum 91 μgm^{-3}) on a number of occasions during September 2010 and October 2011 which was apparently due to the influence of fires from Indonesia [19].

IV. Microscopic Analysis

Several studies have applied Electron Microscopy techniques to characterize particulate matter over the past thirty years. The SEM uses a focused electron beam across sample to scan it under vacuum. The electron beam interact with sample and produce different effects that can be monitored with suitable detectors.

Fig. 6 shows SEM micrograph of individual particles during haze events. The microscopic observation of the biomass burning PM2.5 sampled confirmed the presence of most major components (microsoot particles, salt condensate, vegetation relicts, soil-dust derived particle) mentioned inother literature [20-21].

Morphology of different type of particles are shown in Fig. 7. The morphological differences between soot aggregates and other particles are mainly related to differences in their formation and size.

V. Conclusion

Among considerable major fires around the world each year, uncontrolled fires in Indonesia during the months of August through October are most notable. These extensive Indonesian forest fires are widespread and caused transboundary air pollution problems in Southeast Asia.

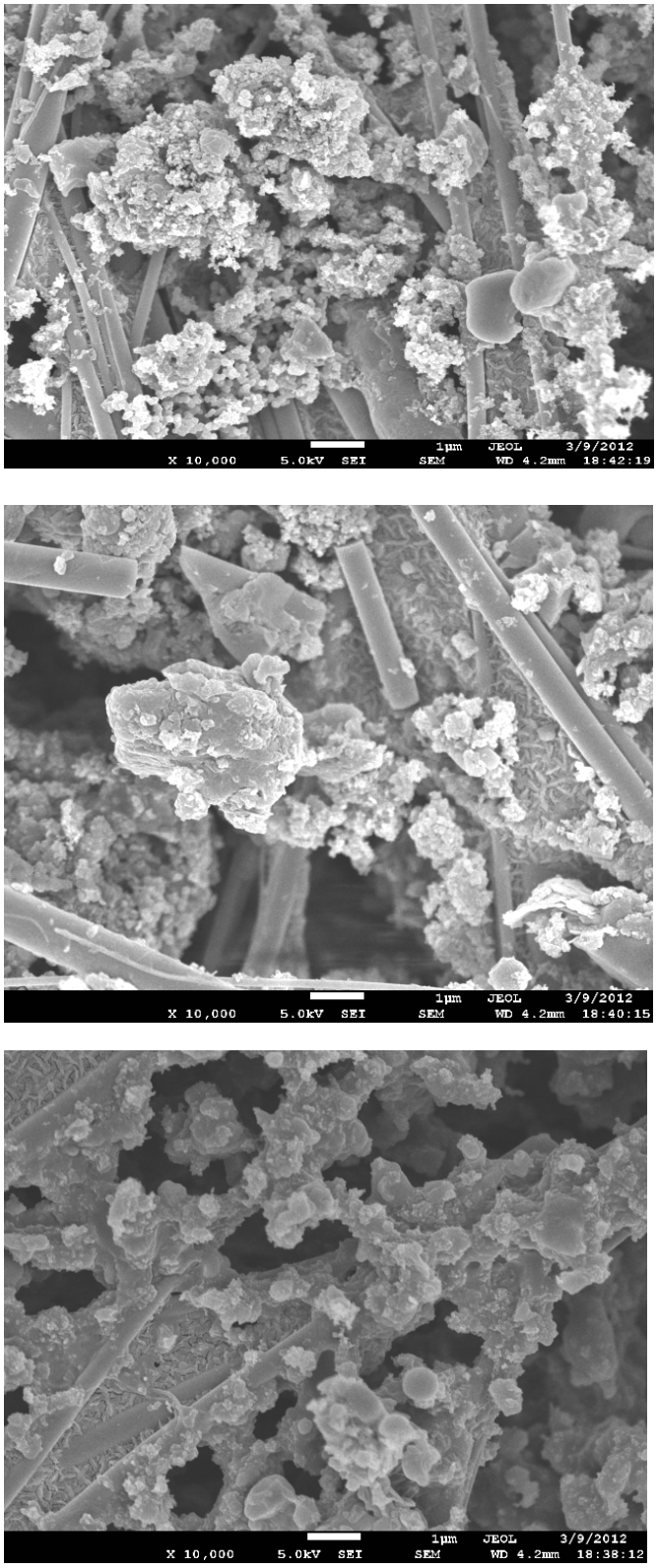

Fig. 6 SEM Micrograph of PM2.5 during Haze Events

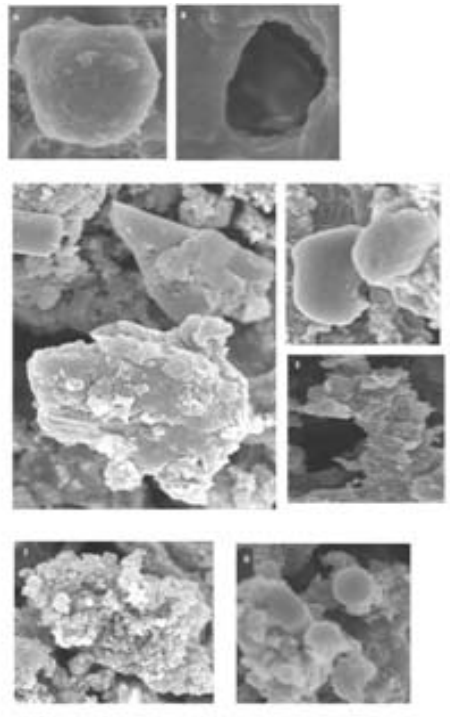

Fig. 7 Morphologies of PM2.5 during Haze period: (A-E) salt condensates and vegetation relicts, (F-G) soot particles

It seems Singapore was very greatly affected by the forest fires in the surrounding region especially those in Sumatra depending on the prevailing wind direction. In this study, we present a detailed chemical characterization of ambient $PM_{2.5}$ during the dry season in 2010 and 2011.

In summary the influence of biomass burning on $PM_{2.5}$ mass was clearly identified and its impact on elemental composition and related concentration was investigated. Accordingly, particulate matter concentration and its source should be monitored in order to prevent adverse health effects. Our results show that huge increases occur in the concentration for most elements during haze period. Because of the background particle composition, source apportionment of data for these periods was complex. Therefore, monitoring of biomass burning emissions should ideally be carried out with the use of several years' data including thee organic and inorganic components to obtain more reliable data for source apportionment.

ACKNOWLEDGMENT

The authors thank "SINGA Scholarship" for financial support.

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