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Metal Compositions of PM_{10} and $PM_{2.5}$ Aerosols in Taipei during Spring, 2002

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ABSTRACT

Ambient aerosols, collected in Taipei in spring 2002, were measured for concentrations of 15 metals (Al, Ca, Na, Mg, K, Ti, Sr, Ba, Mn, Co, Zn, Pb, Cu, Cd, and Sb). Al was used as a mineral aerosol particle indicator, and based on temporal variations of Al concentration, seven Asian dust storm episodes were identified. The fraction of mineral dust in PM_{10} was estimated to be around 80% during Asian Dust Storm (ADS) episodes and 15% in non-ADS periods. The metals were categorized into three groups based on their source of origin. The first group consisted of metals from crustal sources, Al, Ca, Sr, Ba, Ti, and Co, the second group was from anthropogenic sources, Pb, Zn, Cd, Cu, and Sb; and the third group was of mixed origins; Na and Mg from sea salt and crustal sources, and K and Mn from crustal and anthropogenic sources. The results of this study demonstrated significant variation in concentrations of metals during spring. Sharp increases in concentration were observed during ADS episodes, particularly for crust-derived elements, Al, Ca, Na, Mg, K, Sr, Ti, Ba, and Co. Metals of anthropogenic origin, Pb, Sb, Cd, and Zn, also increased with ADS episodes, which indicates that significant amounts of pollutant were transported with dust to reach Taiwan. Size-distribution analysis revealed that metals derived from crust and seawater sources (Al, Fe, Sr, Ba, Ti, Na, and Mg) tend to reside in coarse particles, and anthropogenic metals (Pb, Zn, and Cd) in fine particles. Air mass backward trajectory analysis suggested that deserts around Mongolia and the Loess Plateau were the dominant source regions of dust aerosols for ADS. Concentrations of metals in dust were found to change and the wet scavenging affect was strongly indicated.

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(Key words: Aerosol, Asian dust storm, Crust-derived element, Anthropogenic metal)

1. INTRODUCTION

Dust storms (DSs) occur annually in arid/semi-arid deserts of central Asia in spring and late winter. Statistics of DSs, for the past 40 years in China, demonstrate a percentage occurrence for March, April and May of about 20%, 58%, and 22%, respectively (Sun et al. 2001). The Gobi Desert in Mongolia and Taklamakan Desert in western China, and the Loess Plateau are main source areas of DSs (Liu 1985). Studies have demonstrated that DSs in China were often associated with frontal systems and the Mongolia cyclonic depression (Sun et al. 2001). Considerable amounts of windblown dust can be entrained into the free troposphere to ~3000 m or higher, by vertical turbulence, and then transported by westerly winds (Zhang et al. 1997). DSs can travel several thousands of kilometers or more from source regions, in the east, to neighboring countries (e.g., Korea and Japan) (Choi et al. 2001) and the North Pacific (Duce et al. 1980; Gao et al. 1992; 1997; Husar et al. 2001). Northeastern winter monsoons can bring Asian dust to Taiwan in the lower atmosphere.

Aerosol particles not only impact on air quality and public health, but also play an important role in radiative forcing and so, climate change (Levitus et al. 2001; Bergin et al. 2001; Kiehl 1999; Yu et al. 2002). For example, Liu et al. (2002) found that increases in anthropogenic aerosols reduced sunshine infiltration in Taiwan. Cheng and Tasi (2000) found that recent increases in aerosols contributed to impaired visibility in central Taiwan. Nutrients in aerosols have also been found to influence marine ecosystems when aeolian particles are deposited and dissolved (Duce et al. 1991; Gao et al. 2001). For example, aeolian Fe fertilizes the ocean, causing biological blooming (Fung et al. 2000). On the other hand, heavy metals such as Zn, Pb, Cu, Sb, and Cd present health hazards due to their toxicity.

Large numbers of ADS investigations have been undertaken in East Asia and the western Pacific (Merrill et al. 1994; Gao et al. 1996; VanCuren and Cahill 2002; Kanayama et al. 2002). However, relatively little research on ADS had been conducted in Taiwan until the so-called "yellow muddy rain" of March 12, 1995 (Young et al. 1997). Liu and Shiu (2001) analyzed air quality monitoring data from EPA monitoring stations in Taiwan, during 1993~2000, and suggested that ADS events contributed to about 10% of the annual loading of PM_{10} around northern Taiwan. Lin (2001) suggested that Asian dust transport from northern China to Taiwan took about two to three days and stayed primarily at an altitude of 500~1500 m. Nevertheless, no systematic experiment on ADSs, to evaluate their impact on air quality of Taiwan, has been undertaken. Rapid economic developments in China over the last few decades have caused severe air pollution. It is conceivable that Asian continental pollutants are transported to Taiwan by winter monsoons and occasionally, DS events.

In spring 2002, the EPA supported a field campaign of ambient aerosols, which devised the first experiment to study DSs, as well as long-range transport of air pollutants. The experiment included measurements of chemical and physical properties of aerosols at six stations around Taiwan and modelling. The findings of the experiment are reported in the companion papers of this special issue.

This study reports measurements of metals in $PM_{2.5}$ and PM_{10} aerosols collected in metropolitan Taipei from February to May 2002. Since however coarse particles appeared to dominate PM_{10} mass concentrations, during ADS episodes, discussion focuses on results of PM_{10}.

2. MATERIALS AND METHODS

2.1 Sampling

The sampling site was located at the Central Weather Bureau (CWB) in metropolitan Taipei. Aerosol samplers were set on the roof of the CWB building, which is approximately 25 m above ground level. The Partisol®-FRM Model 2000 air sampler (Rupprecht & Patashnick Co. Inc.) was employed to collect PM_{10} and $PM_{2.5}$ ambient aerosol particles, simultaneously. An active volumetric flow control system maintained a constant volumetric flow at a rate of 16.7 lpm. PTFE membrane filters (PallGelman, 2.0 μm pore size and 47 mm in diameter with a PE ring) were used for aerosol filtration. The membranes were conditioned for at least 48 hours and then weighed prior to use, by a microbalance (MX 5, Mettler-Toledo Inc.; detection limit 1 μg), in a weighing room, with the relative humidity controlled at 35 ± 5%. Sampling was conducted 2 days before forecast arrival of ADS and generally lasted one week. When there were no ADSs forecasts, sampling was taken in the last week of the month. Samples were collected at 12 hr intervals: from 8:00am to 8:00pm and from 8:00pm to 8:00am the following day. Consequently, samples were categorized by daytime (D) and night-time (N), respectively.

2.2 Analysis

The aerosol samples on PTFE filters were dried, weighed, and then completely dissolved in a mixture of 5 ml HNO_3, 5 ml HF, and 0.5 ml $HClO_4$ (Suprapur grade from Merck) in PTFE beakers.. The dried sample was redissolved in 2 ml HNO_3 and the solution was diluted, with Milli-Q water, to 20 ml (in 2% HNO_3). The digestion method was similar to that used for seawater suspended particles filtered on Nuclepore polycarbonate membranes (Hsu et al. 1998). Note that since PTFE membranes cannot be digested, clean pincers were used to carefully remove residual filters. Digested solutions were stored in a freezer at 4°C for later analysis. All containers used in the study were acid-cleaned prior to use.

Metal determinations were made using a quadrupole-based inductively coupled plasma mass spectrometer (ICP-MS, Elan 6100, Perkin-Elmer™ Instruments, USA). In the study, the detection limit (DL; using units of ng m^{-3}) of each element was assessed by using three times of one standard deviation calculated from ten blanks divided by a 12-hour average flow volume. All DLs obtained were low (Table 1) enough to satisfy requirements of the study. A multi-element standard, prepared from stock (Merck) made up of 2% HNO_3 solution, was used to achieve calibration. Analysis of two types of standard reference materials (SRM): SRM 1648 (urban particulate matter from NIST, National Institute of Standards and Technology, USA) and CJ1 (Chinese Loess, National Research Center for Environment and Measurement, China) were used to validate QA/QC analysis (Nishikawa et al. 2000). Recovered values for all target

elements either fell into this range or within 10% of the certified (or non-certified) values (Table 2). Blanks were used to assess reagents, methods, standards, instruments, and calibrations. Nebulizer gas flow and electrostatic ion lens voltage were adjusted for peak sensitivity. 10 $\mu g\ l^{-1}$ solution, of Mg, Cu, Rh, Cd, In, Ce, Ba, Pb, and U, was used to facilitate mass calibration and instrument optimisation.

Table 1. Detection Limits (DL) for Selected Metals (in a unit of ng m^{-3}).

Element	DL	Element	DL	Element	DL	Element	DL
Al	2	Ca	15	Mn	0.1	Cd	0.01
Na	5	Sr	0.1	Co	0.1	Sb	0.01
Mg	1	Ba	1	Cu	1	Pb	0.2
K	2	Ti	2	Zn	0.5		

Table 2. Comparisons of Analytical and Certified Values for Two Standard Reference Materials, SRM 1648 (urban atmospheric particulates) and CJ1 (Chinese loess) (Nishikawa 2000)

	SRM1648		CJ1	
	Certified values	This work (n=5)	Certified values	This work (n=1)
Al	3.42±0.11%	3.32±0.14%	6.01±0.17%	5.83%
Na	0.425±0.002%	0.42±0.02%	1.33±0.06%	1.37%
Mg	0.8 %*	0.82±0.02%	1.57±0.06%	1.65%
K	1.05±0.01%	0.95±0.04%	1.94±0.10%	1.86%
Ca	---	4.4±0.38%	5.83±0.23%	5.25%
Sr	---	208±15μg/g	277±11μg/g	288%
Ba	737μg/g*	730±37μg/g	504±27μg/g	508μg/g
Ti	0.4%*	0.39±0.02%	0.36%*	0.33%
Mn	786±17μg/g	849±31μg/g	633±44μg/g	684μg/g
Co	18μg/g*	17.8±0.9μg/	12.8μg/g*	14.0μg/g
Cu	609±27μg/g	670±37μg/g	21±3μg/g	22.9μg/g
Zn	0.476±0.014%	0.490±0.024%	59±5μg/g	56.6μg/g
Cd	75±7μg/g	69±3μg/g	---	
Pb	0.655±0.008%	0.635±0.020%	18.4μg/g*	19.3μg/g
Sb	45μg/g*	46±2.3μg/g	---	

*. non-certified values

3. RESULTS AND DISCUSSION

3.1 Temporal Variations in Atmospheric Metal Concentrations

Chester et al. (2000) compiled a list of globally important sources of trace metals in aerosols. These sources included (a) low-temperature crustal weathering (crustal source), (b) a variety of high-temperature anthropogenic processes (anthropogenic source), and (c) sea-salt generation (oceanic source). Chester et al suggested that Al could act as an indicator of mineral dust because Al is one of the most common elements found in crustal sources. Consequently, Al concentrations were used in this study to identify occurrences of ADS.

Temporal variations of Al in PM_{10} indicate that seven episodes (the dates marked gray on the x-axis of Fig. 1) of ADS reached northern Taiwan in winter and spring 2002. They were 0211D, 0306D-0309D, 0318D-0321D, 0323D, 0331D-0401D, 0409D-0413D (0409-0410D and 0412N-0413D episodes were probably a single event), and 0417N-0418N. The timing of dust episodes were very consistent with results of model simulations (Chen et al. 2004).

Chou et al. (this issue) used a different method to identify ADS. This method examines coarse ($PM_{2.5-10}$) to fine ($PM_{2.5}$) particle concentration (C/F) ratios and the results of Chou et al. were reasonably consistent with those found in this study. However, two episodes (0331D-0401D and 0413D) were not identified by the C/F ratio method. In these episodes, Al concentrations were at least two times that of the mean concentration (516 ng m^{-3}) for non-ADS periods, with one exception at 0418D (397 ng m^{-3}). (Local air mass could effect low Al concentrations so this exception presented difficulties for interpretation).

Metal concentrations were separated into two groups, i.e., ADS episodes and non-ADS periods, to facilitate comparison. Table 3 summarizes the statistics (including geometric mean and one standard deviation) of atmospheric concentrations for metals analyzed in PM_{10} and $PM_{2.5}$, from Taipei in spring 2002. The table shows that standard deviations of airborne particulate metal concentrations were very large. This result reveals that variations in concentration were also large. The average Al concentration in PM_{10} was 4589 ng m^{-3} with one standard deviation of 8145 ng m^{-3}, during ADS episodes, and 516 ng m^{-3} with one standard deviation of 800 ng m^{-3}, in non-ADS periods (Table 3). The Al concentration ranged from below the detection limit (2 ng m^{-3}, given in Table 1) to near 30000 ng m^{-3} for PM_{10} (Fig. 1). The Al maxima obtained in this study were comparable with those measured at both Pengchiayu and Taipei sites (Huang 2002; Lin, unpublished data). Apparently, dust from long-range transport and scavenging by precipitation caused large variations in Al concentration. Large temporal variability for mineral-derived elements was consistent with other observations in remote areas downwind and big desert countries, such as northeastern China (Zhang et al. 2001; Liu et al. 2002), Hong Kong (Cheng et al. 2000), and Hawaii (Zieman et al. 1995). In an usual case observed by Kim and Park (2001), a sharp decrease in total PM concentration by one order of magnitude (from approx. 1000 to less than 100 μg m^{-3}) was noted within several hours, during a dust episode in Korea.

Concentrations for Pb in PM_{10} average 34 ng m^{-3} with one standard deviation of 30 ng m^{-3}, during ADS episodes and 15 ng m^{-3} with one standard deviation of 35 ng m^{-3}, during non-ADS periods. On average, comparisons between ADS and non-ADS periods re-

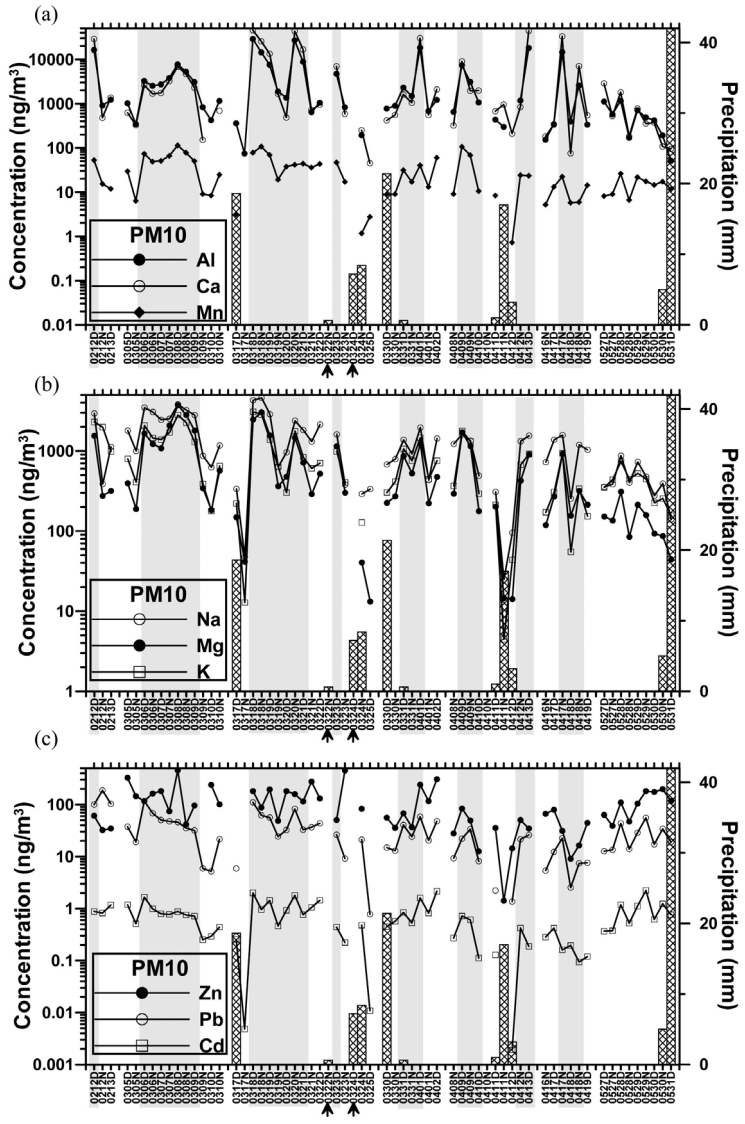

Fig. 1. Temporal variations of selected element concentrations (ng m^{-3}): against sampling date (for example, 0212: February 12) and time (D: daytime or N: night-time). See the upper panel for Al, Ca, and Mn, middle panel for Na, Mg, and K, and lower panel for Zn, Pb, and Cd. Precipitations in corresponding periods, from the Kutin air quality monitoring station of EPA, are also plotted in the bar graph and scaled on the right axis. Samples influenced by dust storm events are marked in grey. Arrows indicate times when no samples were available for analyses because of sampling failure or other causes. Note that the data below detection limits for certain metals were not shown in these plots.

Table 3. Statistical Summary (mean ± standard deviation) of Atmospheric Concentrations (ng m^{-3}) for Selected Metals During ADS Episodes and In Non-ADS Periods for (A) PM_{10} and for (B) $PM_{2.5}$, in spring 2000.

(A)	ADS period (n=26)	non-ADS period (n=35)
PM_{10}	73±31	41±18
Al	4589±8145	516±800
Na	1765±1176	518±526
Mg	997±920	160±211
K	1054±826	295±373
Ca	4820±15420	485±1277
Sr	52±185	3±13
Ba	39±37	8±40
Ti	210±179	37±38
Mn	38±31	11±14
Co	1.2±1.1	0.5±1.2
Zn	74±96	77±106
Pb	34±30	15±35
Cd	0.63±0.52	0.36±0.55
Cu	19±12	13±30
Sb	1.9±1.8	1.9±2.0

(B)	ADS period	non-ADS period
$PM_{2.5}$	29±10	24±15
Al	878±4984	182±855
Na	309±476	77±121
Mg	183±238	34±59
K	462±395	141±416
Ca	592±9118	257±1705
Sr	7±107	2±20
Ba	8±18	3±66
Ti	45±60	16±55
Mn	17±13	6±8
Co	0.6±1.6	0.6±5.2
Zn	84±109	50±114
Pb	21±25	12±49
Cd	0.48±0.64	0.33±0.36
Cu	7±10	4±10
Sb	0.9±1.7	0.8±1.2

vealed that most metals were higher during ADS episodes than in non-ADS periods, except for Zn in PM_{10}. In particular, concentrations of crust-derived elements such as Al, Ca, and Sr were around one order of magnitude higher during ADS episodes than in non-ADS periods.

Figure 1 shows concentrations of nine metal elements in PM_{10} aerosols, including Al, Ca, Mn (crust dominated, Fig. 1a), Na, Mg, K (sea salt dominated, Fig. 1b), Pb, Zn, and Cd (pollution dominated, Fig. 1c). The rainfall was also plotted to evaluate the scavenging effect of wet deposition (Utsunomiya and Wakamatsu 1996). It is worth noting that co-variations of Ca of PM_{10} with Al (Fig. 1a) ($r = 0.97$, $n = 61$; Table 4) correlate well. Concentrations of Na, Mg, and K in PM_{10} also exhibited close correlations with each other (Na vs. Mg, $r = 0.91$ and Na vs. K, $r = 0.89$; Table 4). The general patterns of Zn, Pb, and Cd concentration were similar to one another (Fig. 1c).

Concentrations of Pb, Zn, and Cd correlate poorly with Al and other crustal elements. This result indicates that metal pollutants were not consistently collected by ADS episodes on route to Taiwan. Nonetheless, concentrations of Pb, Zn, and Cd are higher during the ADS periods than the non-ADS periods as shown in Table 3. High concentrations of Pb, Zn, and Cd, during ADS episodes, were probably due (but not always, see Fig. 1) to pick-up of pollutants, when atmospheric conditions were favorable. Assessments of the impact of long-range transport of air pollutants on Taiwan's air quality by Lin et al. (2004) supports this theory.

3.2 Metal Compositions and Aerosol Abundance

Abundances (% for Al, Fe, Na, Mg, K, and Ca, and $\mu g\ g^{-1}$ for others) of each element in PM_{10} and $PM_{2.5}$ aerosol particles, in Taipei for spring 2002, are provided in Table 5. This data was also separated into two groups: ADS episodes and non-ADS periods. On average, Al and Ca in PM_{10} are the predominant metals, during ADS episodes, accounting for 6.3% and 6.6% of the bulk samples, respectively. This finding correlates well with measures in previous studies (Zhang et al. 2001; Ma et al. 2001). Na, Mg, and K constituted 2.4%, 1.4%, and 1.5% of samples, respectively. Compared to non-ADS periods, the weight fractions of major metals were significantly higher. Abundances of Al were five times higher in ADS episodes samples than in non-ADS samples (1.3%). For $PM_{2.5}$, Al, Ca, K, Na, and Mg account for 3.0%, 2.1%, 1.6%, 1.1%, and 0.6% of the total mass during ADS episodes, respectively. These values were also higher than those for non-ADS periods, but less so than those of PM_{10}. This finding is consistent with the fact that coarse particles tend to dominate ADS episodes.

Comparative abundance of heavy metals varied. The abundance of Pb in PM_{10} was 470 $\mu g\ g^{-1}$ in ADS samples, compared to 360 $\mu g\ g^{-1}$ during the non-ADS periods. However, Zn, Cu, and Sb were higher in non-ADS periods. In addition, abundances of Zn, Pb, and Cd in $PM_{2.5}$ were higher than that in PM_{10}, suggesting that fine particles were more enriched with heavy metals of anthropogenic origins compared to coarse particles.

Since the composition of mineral dust components is assumed to be similar to the average crust, of which Al accounts for about 8% by weight (Taylor 1964), Al was used to estimate the weight fraction of mineral dust components. The percentage of dust in PM_{10} averaged at about 80%, during ADS episodes, and only 15% in non-ADS periods. This result is consistent with the fact that sulfate and carbonaceous components make up a major part of urban aerosols on the days without ADS (Lung et al. 2004).

Table 4. Correlation Coefficients (r) Between All Analyzed Metals for (A) PM_{10} and (B) $PM_{2.5}$. (Values > 0.50 are bolded)

(A)	Al	Na	Mg	K	Ca	Sr	Ba	Ti	Mn	Co	Zn	Pb	Cd	Cu	Sb
Al	1.00														
Na	**0.63**	1.00													
Mg	**0.61**	**0.91**	1.00												
K	**0.67**	**0.89**	**0.91**	1.00											
Ca	**0.97**	**0.54**	0.49	**0.56**	1.00										
Sr	**0.97**	**0.49**	0.45	**0.52**	1.00	1.00									
Ba	**0.65**	**0.54**	**0.58**	**0.79**	**0.61**	**0.60**	1.00								
Ti	**0.61**	**0.66**	**0.72**	**0.71**	**0.54**	**0.51**	**0.54**	1.00							
Mn	**0.50**	**0.86**	**0.91**	**0.86**	0.38	0.33	**0.52**	**0.68**	1.00						
Co	0.38	0.38	0.47	0.41	0.31	0.29	0.27	**0.50**	0.45	1.00					
Zn	0.15	0.29	0.31	0.27	0.08	0.07	0.12	0.16	0.40	0.04	1.00				
Pb	0.43	**0.52**	0.42	**0.72**	0.36	0.36	**0.82**	0.35	0.45	0.10	0.19	1.00			
Cd	0.39	0.49	0.37	**0.51**	0.30	0.29	0.35	0.35	**0.51**	0.17	**0.50**	**0.65**	1.00		
Cu	0.22	0.13	0.13	0.19	0.20	0.20	0.20	0.13	0.17	0.14	0.25	0.25	0.42	1.00	
Sb	0.23	0.18	0.09	0.24	0.17	0.18	0.29	0.15	0.24	0.08	0.46	**0.51**	**0.75**	0.40	1.00

(B)	Al	Na	Mg	K	Ca	Sr	Ba	Ti	Mn	Co	Zn	Pb	Cd	Cu	Sb
Al	1.00														
Na	**0.94**	1.00													
Mg	**0.84**	**0.92**	1.00												
K	0.44	**0.59**	**0.63**	1.00											
Ca	**0.98**	**0.92**	**0.79**	0.40	1.00										
Sr	**0.99**	**0.92**	**0.80**	0.42	1.00	1.00									
Ba	0.22	0.21	0.24	**0.71**	0.21	0.25	1.00								
Ti	**0.60**	**0.62**	**0.65**	0.35	**0.58**	**0.58**	0.13	1.00							
Mn	0.46	**0.64**	**0.63**	**0.64**	0.40	0.41	0.09	0.42	1.00						
Co	0.07	0.07	0.11	0.00	0.05	0.05	-0.02	0.48	-0.04	1.00					
Zn	0.04	0.26	0.20	0.36	0.00	0.00	-0.03	0.08	**0.59**	-0.04	1.00				
Pb	0.11	0.19	0.21	**0.84**	0.08	0.12	**0.90**	0.08	0.33	-0.05	0.21	1.00			
Cd	0.25	0.39	0.32	**0.63**	0.21	0.22	0.26	0.21	**0.71**	-0.04	0.44	**0.59**	1.00		
Cu	0.41	0.49	0.36	**0.57**	0.37	0.39	0.48	0.24	**0.53**	-0.04	0.46	**0.57**	**0.62**	1.00	
Sb	0.23	0.28	0.18	0.46	0.22	0.22	0.31	0.24	**0.58**	-0.08	0.24	**0.52**	**0.78**	**0.56**	1.00

Table 5. Statistical Summary (mean ± standard deviation) of Weight Content (% for Al, Fe, Na, Mg, K, and Ca; $\mu g\ g^{-1}$ for others) for Selected Metals During ADS Episodes and In non-ADS Periods for (A) PM_{10} and for (B) $PM_{2.5}$, in spring 2000.

(A)	ADS period (n=26)	non-ADS period (n=35)
Al (%)	6.3	1.3
Na (%)	2.4	1.3
Mg (%)	1.4	0.4
K (%)	1.5	0.7
Ca (%)	6.6	1.2
Sr (μg/g)	696	79
Ba (μg/g)	533	194
Ti (μg/g)	2796	871
Mn (μg/g)	529	264
Co (μg/g)	17	12
Zn (μg/g)	1015	1843
Pb (μg/g)	469	364
Cd (μg/g)	9	9
Cu (μg/g)	261	330
Sb (μg/g)	26	45

(B)	ADS period	non-ADS period
Al (%)	3.0	0.8
Na (%)	1.1	0.3
Mg (%)	0.6	0.1
K (%)	1.6	0.6
Ca (%)	2.1	1.1
Sr (μg/g)	230	91
Ba (μg/g)	266	125
Ti (μg/g)	1563	728
Mn (μg/g)	588	241
Co (μg/g)	20	27
Zn (μg/g)	2955	2168
Pb (μg/g)	756	535
Cd (μg/g)	17	14
Cu (μg/g)	243	178
Sb (μg/g)	31	34

3.3 Size-distributions ($PM_{2.5-10}$ vs. $PM_{2.5}$)

Figure 2 depicts percentages of $PM_{2.5-10}$ (defined as the difference between PM_{10} and $PM_{2.5}$) vs. $PM_{2.5}$, i.e., coarse vs. fine for each element during spring of 2002. Al, Ca Sr, Ba, Ti, Na, and Mg were found to reside predominantly in the coarse mode. This finding is consistent with the fact that they are mainly from windblown dust or sea salt aerosols. Chou et al. (2004) measured the size distribution of total suspended particles, during ADS episodes, and found a mono-mode distribution that peaked at around 2.5 - 5.6 m. This result was quite different from the bi-modal distribution in non-ADS periods.

Figure 2 also illustrates that the coarse mode accounts for ~60% of K, Mn, Co, Cu, and Sb. On the other hand, fractions of Zn, Pb, and Cd in the fine mode were slightly higher than or near equal to those in the coarse mode. In general, these patterns were consistent with earlier studies (Marcazzan et al. 2001; Pakkanen et al. 2001). The results of this study demonstrate that coarse mode aerosols can carry large amounts of polluted species, such as sulfate, nitrate, and heavy metals, to observation sites (Arimoto et al. 1996; Zhou et al. 1996; Kim and Park 2001). Furthermore, during DS transport, compositions of aerosols can be modified by heterogeneous interactions with pollutants, such as SO_2 and NO_X (Chester et al. 2000; Uematsu et al. 2002; Higurashi and Nakajima 2002).

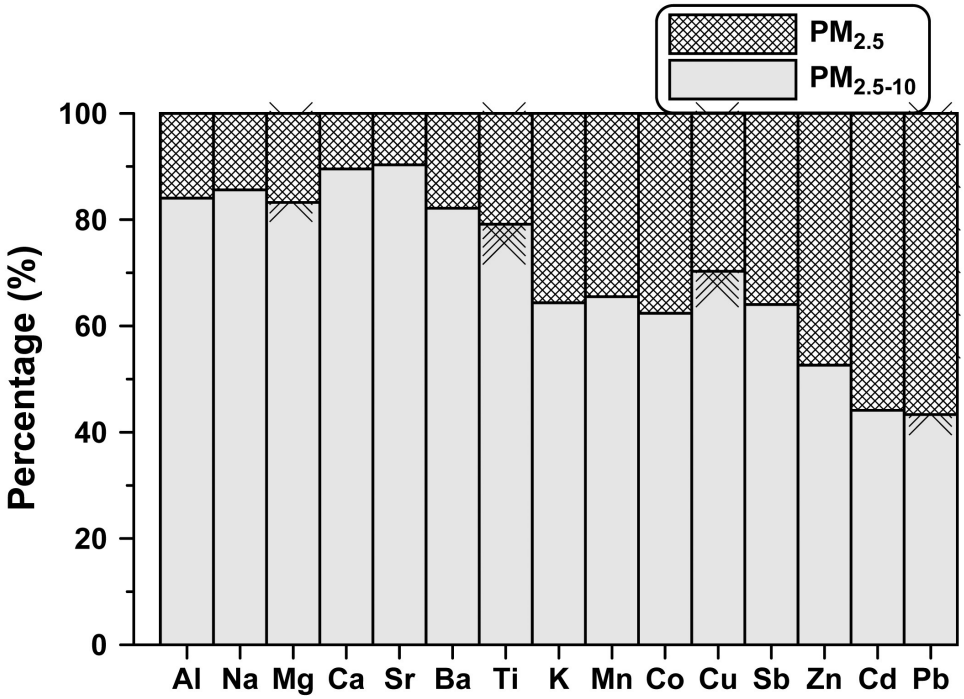

Fig. 2. Size distributions for all selected elements in coarse ($PM_{2.5-10}$) and fine ($PM_{2.5}$) aerosol particles.

3.4 Enrichment Factors and Source Identification for Particulate Metals

Atmospheric concentrations of trace metals can vary significantly. Nonetheless, the normalization of trace metals to crustal concentrations can be used to enable the "chemical character" of aerosol particles to be investigated effectively (Chester et al. 1993). An enrichment factor (EF) can be defined to evaluate anomalies of specific chemical species of ambient aerosols, with respect to representative compositions of a reference material (e.g., average crust and sea water). Al was used as the crustal element indicator, and the EF_{crust} value of element X was calculated according to the following formula:

$$EF_{crust} = (C_{X-aerosol} / C_{Al-aerosol}) / (C_{X-crust} / C_{Al-crust}), \quad (1)$$

in which $C_{X-aerosol}$ and $C_{Al-aerosol}$ are concentrations of element X and Al in aerosols, respectively, and $C_{X-crust}$ and $C_{Al-crust}$ are their concentrations in average crustal material (Taylor 1964). Since, there was a large scattering of data, a conservative EF_{crust} value of about 10 was chosen as the criterion to indicate a significant proportion of a given element, which has a non-crustal source, and these elements with high EF_{crust} values were termed anomalously enriched elements (AEEs).

Figure 3 shows EF_{crust} values of individual metals during ADS episodes and non-ADS periods in PM_{10} (Fig. 3a) and $PM_{2.5}$ (Fig. 3b), respectively. Overall, EF_{crust} patterns for metals in PM_{10} were similar to those in $PM_{2.5}$. The elements were classified into two groups according to their EF_{crust} values. Elements with EF_{crust} values ≤ 10 included Al, Na, Mg, K, Ca Sr, Ba, Ti, Mn, and Co and so indicate elements of crustal sources; whereas those elements with EFcrust values > 10 included Cu, Zn, Pb, Cd, and Sb, and indicate elements of anthropogenic sources. Sequential EF_{crust} values for these particular elements were Cu < Zn < Pb < Cd < Sb.

It is worth noting that EF_{crust} values for each element in PM_{10}, except for Ca and Sr, were lower during ADS episodes than those in non-ADS periods, most notably AEEs (Cu, Zn, Pb, Cd, and Sb), which were lower by a factor of 3 - 10. These results indicate that long-range transport particles were diluted by anthropogenic heavy metals, relative to locally suspended particles. Nonetheless, this does not mean that absolute concentrations of these metals in the air on ADS days were lower than those on non-ADS days. It was also noted that EF_{crust} values of these particular AEEs were slightly higher in $PM_{2.5}$ than in PM_{10}, demonstrating that fine particles contain higher anthropogenic metals compared to coarse particles.

Along with EF_{crust} values, $EF_{seawater}$ values were calculated for Mg, K, Ca, and Sr using equation (1) in which Na was used as an element indicator of average seawater, to evaluate the contribution of sea salt for these elements. Concentrations of sea salt-derived Na were estimated from the total concentration minus crustal-derived Na concentration (i.e., the product of Al concentrations multiplied by the Na/Al ratio of average crust). The mean $EF_{seawater}$ values obtained for Mg, K, Ca, and Sr in PM_{10} aerosols were 1.4, 14, 20, and 11, respectively. This result indicates that other than crustal sources, sea salt was also an important source of Mg, and a minor source of K, Ca, and Sr.

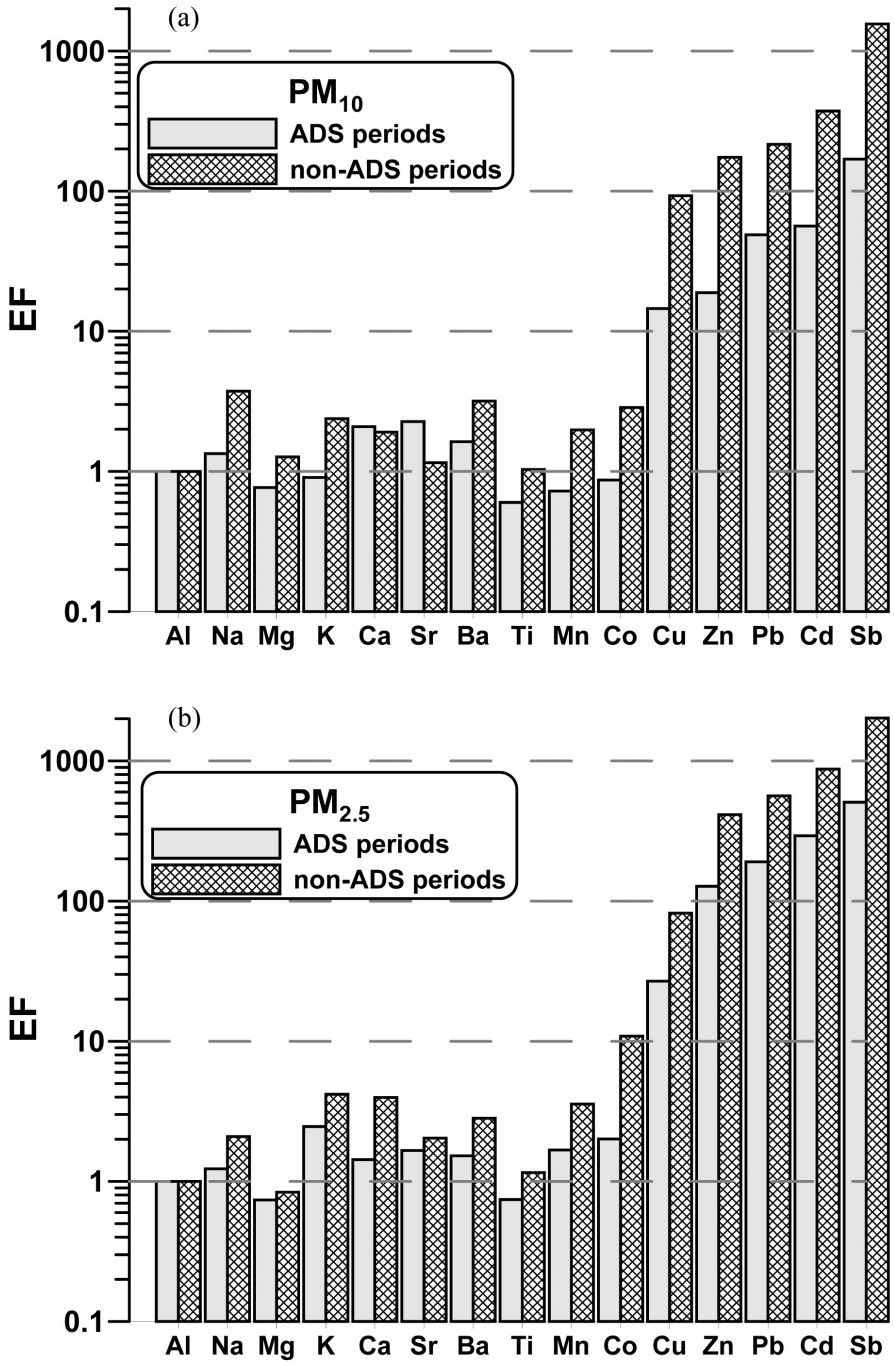

Fig. 3. EF_{crust} values for metals analyzed during ADS episodes and in non-ADS periods for (A) PM_{10} and (B) $PM_{2.5}$.

Correlation of different elements was another method to study elemental sources. Correlation coefficients (r) are given in Table 4. For example, Figs. 4a, b depict correlations Na vs. Mg and K vs. Mn, respectively. The coefficients of Al vs Na, Mg, K, Ca, Sr, Ba, Ti, and Mn in PM_{10} were higher than 0.5 ($P << 0.001$). This result reveals that these elements originate primarily from crustal sources. Furthermore, the correlation coefficient of Na with Mg and K in PM_{10} was extremely high (r~0.9). Obviously, these elements also share a common seawater source. Correlation coefficients between sea salt-dominated metals and crust-dominated aerosols were relatively high (r~0.65). This was to be expected because as ADS is carried by the winter northeasterly monsoon, it passes over a large area of the East China Sea before reaching Taiwan. K demonstrated positive correlations with Mn, Pb, Cd, Cu ($r > 0.5$), and a likely correlation with Sb ($r = 0.46$), indicating that K has an anthropogenic source, as well as crustal and seawater sources. Emissions from biomass burning could have contributed to K because it is abundant in plant tissues (Andreae and Crutzen 1997). K-rich airborne particles were frequently detected in China and countries downwind (e.g., Korea), and these observations have been attributed to coal combustion (Waldman et al. 1991; Wei et al. 1999; Xu et al. 2001; He et al. 2001). Since Mn correlated with K (Fig. 4b) and tended to associate with fine aerosols (Fig. 2) a fraction of Mn might be attributable to anthropogenic origin, such as coal combustion, fossil fuel burning and heavy industrial activity (Winchester and Bi 1984; Lee and Hill 2003).

Antimony was greatly enhanced in PM_{10} and $PM_{2.5}$ relative to average crust compositions ($EF_{crust} >> 100$). Sb pollution has been well documented in China and attributed primarily to coal combustion (Nriagu and Pacyna 1988; Arimoto et al. 1996; Gao et al. 1996) and

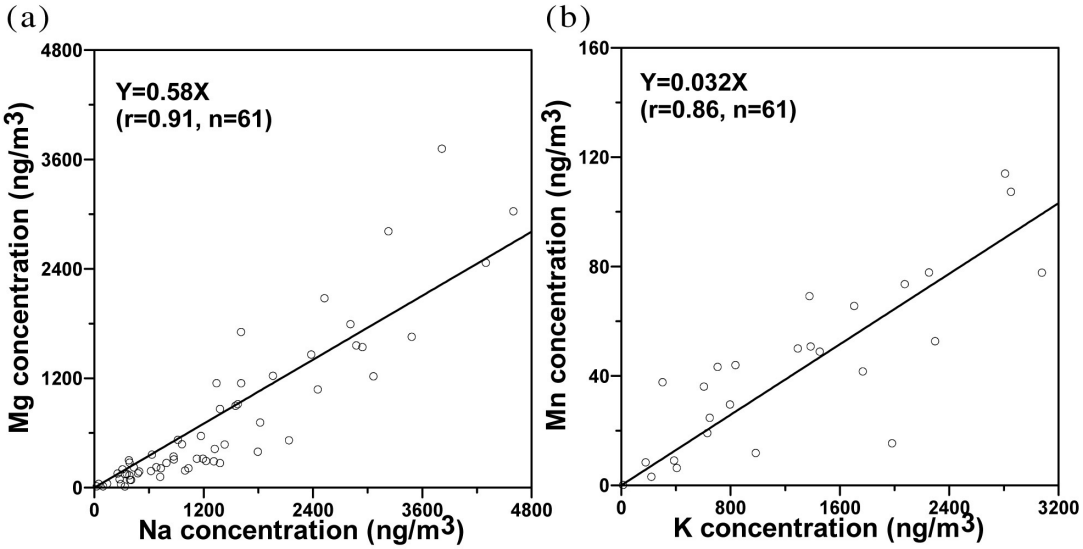

Fig. 4. Regression plots of (a) Mg versus Na concentrations and (b) Mn versus K concentrations.

occasionally to industrial sources (Hashimoto et al. 1994). With EF_{crust} values > 100 in non-ADS periods, Cd was another dominant metal (Fig. 3). Concentration of Cd, during ADS episodes, averaged only slightly above that in non-ADS periods (Table 3), suggesting that contributions of Cd originate from local sources, as well as from long-range transport, during ADS episodes. A major fraction of airborne Cd was emitted from fossil fuel combustion (Nriagu and Pacyna 1988), whereas emissions from refuse incinerators, in areas like Taipei, were the likely source of Cd (Sakata et al. 2000; Hu et al. 2003).

EF_{crust} values of Pb were also considerably higher than 10, and so dominated by anthropogenic sources. Worldwide emissions of Pb primarily come from vehicle exhaust emissions, which account for 65% to 85% of the total emissions of Pb (Nriagu and Pacyna 1988). According to the Environmental Protection Administration (EPA) of Taiwan, leaded gasoline was completely phased out in 2000. Previously, lead content in gasoline was reduced to 0.34 g l^{-1} in 1983 and to 0.026 g l^{-1} in 1997 (http://www.epa.gov.tw). PM measurements indicate that there have been reductions in Pb emissions over the last ten years. Mao and Chen (1996) found a mean concentration of 700 ± 390 ng m^{-3} in PM for Taipei City in 1991, Fang et al. (2002) found several mean concentrations of 20~89 ng m^{-3} in PM_{10} in central Taiwan from mid-1998 to early 2001 and Huang (2002) found a mean concentration of 74 ng m^{-3} in PM for Taipei City from April 2000 to March 2001. A similar trend, in declining Pb concentrations, was also observed in this study. Lead concentrations measured in China have usually been found to be substantially greater than 100 ng m^{-3} (Waldman et al. 1991; Hashimoto et al. 1994; Wei et al. 1999) because leaded gasoline has not been completely phased out. On average, Pb concentrations in Taipei only increased from 15 ng m^{-3} on non-ADS days to 34 ng m^{-3} on ADS days. Note that Pb maxima (~100 ng m^{-3}) occurred in a few ADS samples, e.g., samples 0211D, 0306D, and 0318D.

It is well known that leaded gasoline was primarily responsible for Pb pollution in the ocean, for several decades (Patterson and Settle 1987; Duce et al. 1991). Recently, Lin et al. (2000) investigated seawater Pb concentrations in the southern East China Sea and found that dissolved Pb concentrations were amongst the highest reported for remote oceans. Furthermore, they demonstrated that dissolved Pb concentrations in seawater correlate positively with aeolian Pb influx to this region.

Like Pb, Zn and Cu were also characterized by high EF_{crust} values of > 10. Zn concentrations, during ADS events, were similar to those on non-ADS days, which suggests that local sources were significant contributors. Waste incineration, fossil fuel combustion, and non-ferrous metalliferous industries are the most likely sources of Zn pollution in industrialized countries (Wei et al. 1999). However, this does not explain why atmospheric Zn in Taipei was not enhanced by long-range transport events like other anthropogenic metals, after all, elevated Zn concentrations of several hundreds of ng m^{-3} are common in China (Hashimoto et al. 1994; Wei et al. 1999). EF_{crust} values of Cu were slightly higher than 10, which suggest that Cu was also dominated by anthropogenic sources. In China, Cu pollution has been attributed to combustion of coal and other fuels (Winchester and Bi 1984). These sources may contribute to slightly larger concentrations of Cu, during ADS episodes compared to non-ADS periods.

Ca and Al concentrations in air sharply increased during ADS events and demonstrated a

strong correlation. The mean EF_{crust} value of Ca was about 2 in both PM_{10} and $PM_{2.5}$, which indicate Ca enriched ADS aerosols. Ambient aerosols and soil dust in desert and/or loess areas in China have been characterized by high Ca weight content (Winchester and Bi 1984; Zhang et al. 1993 and 1998; Hashimoto et al. 1994; Zhang and Iwsaka 1999; Nishikawa et al. 2000; He et al. 2001). In addition, Ca-rich aerosols were also frequently observed in the coastal cities of China, Korea, and Japan, as well as North America (Zhou et al. 1996; Choi et al. 2001; Kim and Park 2001; VanCuren and Cahill 2002). Consequently, enhanced Ca concentrations can also be used as an indicator of ADS episodes.

3.5 Case Study of Selected ADS Episodes

Air mass backward trajectory analysis was used in a detailed examination of two ADS episodes, of March 6 and April 9. Temporal variations of Al and Pb concentrations for both episodes are displayed in Fig. 5. The HYSPLIT model of NOAA was used to calculate the isobaric three-days-backward air trajectory at three altitudes of 100, 500, and 1000 m, above the sampling site (CWB). The results for episodes, in March and April, are given in Figs. 6 and 7, respectively.

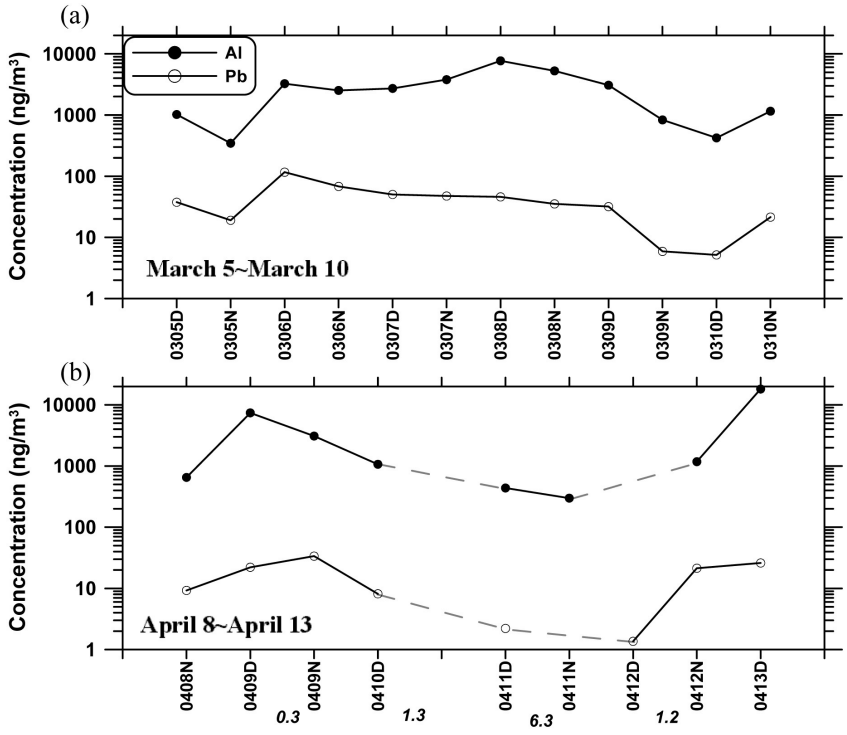

Fig. 5. Temporal variations of Al and Pb concentrations (ng m^{-3}) during two ADS episodes: (a) March 5 to March 10 and (b) April 8 to April 13. The numbers beyond sample IDs denote precipitation (mm).

In the first case, low and middle level trajectories of March 5 came from southern Japan and the upper level trajectory, from the Pacific Ocean (Fig. 6). The concentrations of Al (indicator for mineral dust) and Pb (indicator for pollution) were both low. On March 6, the backward trajectory started at about 3500 m and 1000 m in the inner-Mongolian desert and loess areas for upper and middle levels, respectively, while the low level trajectory started in the Shandon Peninsula and passed through Shanghai, i.e., highly industrialized areas. Both mineral aerosols and pollutants can be expected to be elevated, and in fact both Al and Pb concentrations started to increase (Fig. 5a). On March 7 and 8 backward trajectories at all levels started from around Mongolia. Al concentrations gradually increased and reached a maximum (near 10000 ng m^{-3}) in the daytime of March 8. However, Pb concentrations gradually decreased, despite the fact that the trajectories passed through polluted areas in eastern China. Obviously, trajectories alone did not determine how much pollutant was picked up by the air mass. Vertical mixing probably plays an important role and this notion was examined in detail by Lin et al. (2004). Trajectories at low levels were mostly over the East China Sea on March 9 and 10, and concentrations of both Al and Pb decreased by one order of magnitude.

In the second case, all three levels of backward trajectory for April 8 came from the South China Sea (Fig. 7), and concentrations of both Al and Pb were as low as expected. On April 9, the three-day backward trajectories originated at a similar altitude, of around 3500 m near Mongolia, and after about a day subsided to altitudes of 500 - 1000 m and moved to the East China Sea. Concentrations of Al and Pb initially increased by ten times and three times, respectively (Fig. 5b) but after April 9, concentrations of Al and Pb suddenly decreased until the night of April 12, when both elements recovered. Backward trajectories shifted toward eastern China from April 10 to April 13, but nonetheless provided no obvious clue to explain either concentration decrease or recovery of these metals. However, it rained on Pengchiayu Island, about 100 km north of Taiwan, from April 9 until the night of April 12, and aerosols are known to be susceptible to scavenging by rainfall. It seems reasonable to expect that continuous rainfall (four days) over the ocean removed aerosols from the air mass before it reached Taiwan, although further data is required to verify the scavenging effects of rain.

4. SUMMARY

The major conclusions of this study are summarized as follows:

1. The atmospheric concentrations for most analyzed metals, in particular crustal elements such as Al, Ca, Sr, Ba, and Ti, varied greatly in spring of 2002. Their variability was larger than those of $PM_{2.5}$ and PM_{10} mass concentrations. The dust-derived elements can be sharply increased during ADS passage. Extremely low aerosol and metal concentrations usually follow rainy days, indicating that the wet deposition is an effective removal process.

2. Using Al as an indicator of the mineral dust, seven ADS episodes can be identified in spring of 2002. Furthermore, air mass back-trajectory analyses indicate that all seven

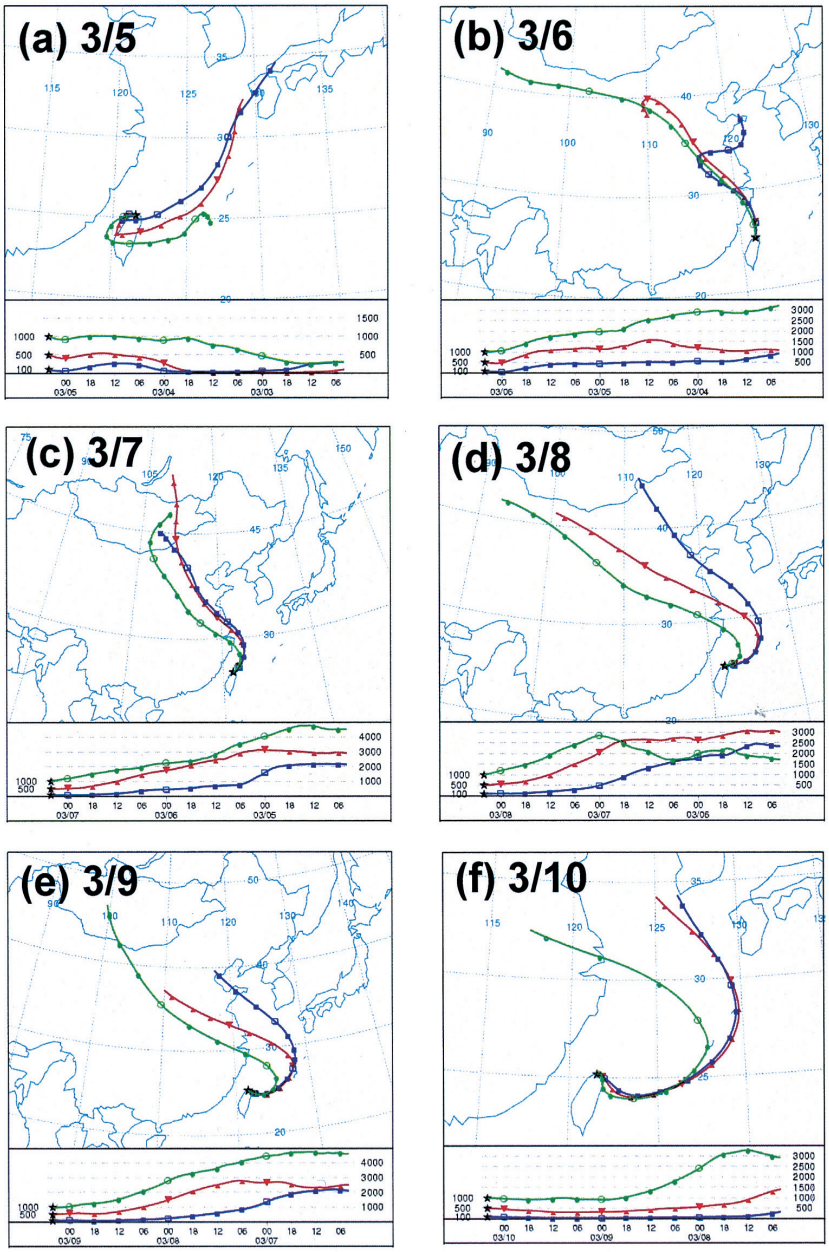

Fig. 6. Three-day air mass back-trajectory analysis obtained from HYSPLIT model for six days from March 5 to March 10. For each day trajectory starts at 0400UTC at three altitudes of 100 m, 500 m, and 1000 m at the CWB site. In each plot the upper and lower panels present the horizontal and vertical motion, respectively.

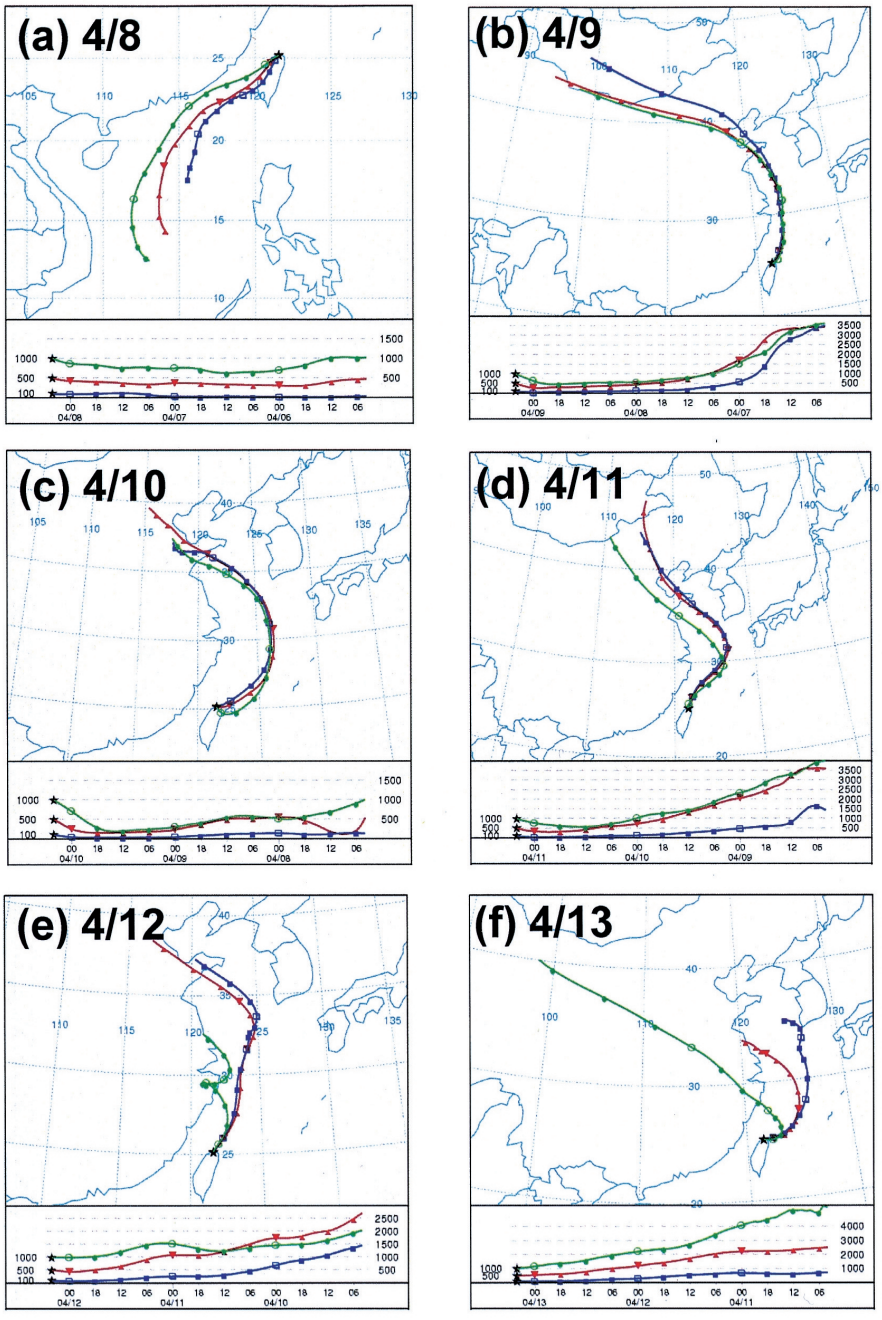

Fig. 7. As in Fig. 5, but for the period from April 8 to April 13.

episodes originated around Mongolia and the Loess Plateau. Additionally, the fraction of dust particles in PM_{10} is estimated to be about 80% during the ADS period, whereas it is only about 15% during the non-ADS period.

3. Metals of anthropogenic origin, namely Pb, Zn, Cd, Cu, and Sb, are characterized by high enrichment factors (EF > 100) relative to the average crust composition, implying that air pollutants are mixed with the ADS. On average, concentrations of anthropogenic metals during ADSs were larger than those of the non-ADS periods by a factor of about two, indicating a significant amount of long-range transport of air pollutants to Taiwan during the ADS. Elevated concentrations of anthropogenic metals often (but not always) occurred concurrently with extremely high Al concentrations during ADS episodes. Furthermore, the correlation of the anthropogenic metals with Al is poor. Obviously, the ADS picks up air pollutants only when the air mass trajectory passes over polluted areas and atmospheric vertical mixing conditions are favorable (2004).
4. Crust-derived elements (such as Al, Ca, Mg, Ti, Sr, and Ba) and sea salt-derived elements (such as Na and Mg) predominantly reside in coarse mode aerosols, while anthropogenic elements (such as Zn, Sb, Pb, and Cd) are nearly equally distributed in coarse and fine mode aerosols.

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REFERENCES

Arimoto, R., R. A. Duce, D. L. Savoie, J. M. Prospero, R. Talbot, J. D. Cullen, U. Tomza, N. F. Lewis, and B. J. Ray, 1996: Relationships among aerosol constituents from Asia and the North Pacific during PEM-West A. *J. Geophys. Res*., **101**, 2011-2023.

Bergin, M. H., G. R. Cass, J. Xu, C. Fang, L. M. Zeng, T. Yu, L. G. Salmon, C .S. Kiang, X. Y. Tang, Y. H. Zhang, and W. L. Chameides, 1999: Aerosol radiative, physical, and chemical properties in Beijing during June 1999. *J. Geophys. Res*., **106**, 17967-17980.

Chen, J. P., Z. Wang, C. Y. Young, F. Tsai, I. C. Tsai, G. J. Wang, W. C. Shieh, H. W. Lin, J. Y. Huang, and M. J. Lu, 2004: Simulations of Asian Yellow Dust Incursion over Taiwan for the spring of 2002 and 2003. *TAO*, **15**, 949-981.

Cheng, M. T., and Y. I. Tsai, 2000: Characterization of visibility and atmospheric aerosols in urban, suburban, and remote areas. *Sci. Total Environ*., **263**, 101-114.

Cheng, Z. L., K. S. Lam, L. Y. Chan, T. Wang, and K. K. Cheng, 2000: Chemical characteristics of aerosols at coastal station in Hong Kong. I. Seasonal variation of major ions, halogens and mineral dusts between 1995 and 1996. *Atmos. Environ*., **34**, 2771-2783.

Chester, R., M. Nimmo, G. R. Fones, S. Keyse, and Z. Zhang, 2000: Trace metal chemistry of particulate aerosols from the UK mainland coastal rim of the NE Irish Sea. *Atmos. Environ*., **34**, 949-958.

Chester, R., M. Nimmo, M. Alarcon, C. Saydam, K. J. T. Murphy, G. S. Sanders, and P. Corcoran, 1993: Defining the chemical character of aerosol from the atmosphere of the Mediterranean Sea and surrounding regions. *Oceanol. Acta*, **16**, 231-246.

Choi, J. C., M. Lee, Y. Chun, J. Kim, and S. Oh, 2001: Chemical composition and source signature of spring aerosol in Seoul, Korea. *J. Geophys. Res*., **106**, 18067-18074.

Chou, C. C. K., C. Y. Lin, T. K. Chen, S. C. Hsu, C. S. C. Lung, S. C. Liu, and C. Y. Young, 2004: Influence of long-range transported dust particles on local air quality: A Case study of the Asian dust episodes in Taipei during the spring of 2002. *TAO*, **15**, 881-889.

Duce, R. A. et al., 1991: The atmospheric input of trace species to the world ocean. *Glob. Biogeochem. Cycle*, **5**, 193-259.

Duce, R. A., C. K. Unni, B. J. Bay, J. M. Prospero, and J. T. Merrill, 1980: Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific: Temporal variability. *Science*, **209**, 1522-1524.

Fang, G. C., C. N. Chang, Y. S. Wu, P. P. C. Fu, C. J. Yang, C. D. Chen, and S. C. Chang, 2002: Ambient suspended particulate matters and related chemical species study in central Taiwan, Taichung during 1998-2001. *Atmos. Environ*., **36**, 1921-1928.

Fung, I. Y., S. K. Meyn, I. Tegen, S. C. Doney, J. G. John, and J. K. B. Bishop, 2000: Iron supply and demand in the upper ocean. *Glob. Biogeochem. Cycle*, **14**, 281-295.

Gao, Y., Y. J. Kaufman, D. Tanre, D. Kolber, and P. G. Falkowski, 2001: Seasonal distributions of aeolian iron fluxes to the global ocean. *Geophys. Res. Lett*., **28**, 29-32.

Gao, Y., R. Arimoto, R. A. Duce, D. S. Lee, and M. Y. Zhou, 1992: Input of atmospheric trace elements and mineral matter to the Yellow Sea during the spring of a low-dust year. *J. Geophys. Res*., **97**, 3767-3777.

Gao, Y., R. Arimoto, R. A. Duce, L. Q. Chen, M. Y. Zhou, and D. Y. Gu, 1996: Atmospheric non-sea-salt sulfate, nitrate and methanesulfonate over the China Sea. *J. Geophys. Res*., **101**, 12601-12611.

Gao, Y., R. Arimoto, R. A. Duce, X. Y. Zhang, G. Y. Zhang, Z. S. An, L. Q. Chen, M. Y. Zhou, and D. Y. Gu, 1997: Temporal and spatial distributions of dust and its deposition to the China Sea. *Tellus B*, **49**, 172-189.

Hashimoto, Y., Y. Sekine, H. K. Kim, Z. L. Chen, and Z .M. Yang, 1994: Atmospheric fingerprints of East Asia, 1986-1991. An urgent record of Aerosol analysis by the JACK network. *Atmos. Environ*., **28**, 1437-1445.

He, K., F. Yang, Y. Ma, Q. Zhang, X. Yao, C. K. Chan, S. Cadle, T. Chan, and P. Mulawa, 2001: The characteristics of $PM_{2.5}$ in Beijing, China. *Atmos. Environ*., **35**, 4959-4970.

Higurashi, A., and T. Nakajima, 2002: Detection of aerosol types over the East China Sea near Japan from four-channel satellite data. *Geophys. Res. Lett.*, **29**, doi: 10.1029/2002GL015337.

Hsu, S. C., F. J. Lin, W. L. Jeng, and T. Y. Tang, 1998: The effect of a cyclonic eddy on the distribution of lithogenic particles in the southern East China Sea. *J. Mar. Res.*, **56**, 813-832.

Hu, C. W., M. R. Chao, K. Y. Wu, G. P. Chang-Chien, W. J. Lee, L. W. Chang, and W. S. Lee, 2003: Characterization of multiple airborne particulate metals in the surroundings of a municipal waste incinerator in Taiwan. *Atmos. Environ.*, **37**, 2845-2852.

Huang, Y. T., 2002: Chemical Characteristics of Total Suspended Aerosols from the Taipei Metropolitan Area. Master's thesis, National Taiwan University, 87pp. (in Chinese)

Husar, R. B. et al., 2001: Asian dust events of April 1998. *J. Geophys. Res.*, **106**, 18317-18330.

Kanayama, S., S. Yabuki, F. Yanagisawa, and R. Motoyama, 2002: The chemical and strontium isotope composition of atmospheric aerosol over Japan: the contribution of long-range-transported Asian dust (Kosa). *Atmos. Environ.*, **36**, 5159-5175.

Kiehl, J. T., 1999: Climate change - Solving the aerosol puzzle. *Science*, **283**, 1273.

Kim, B. G., and S. U. Park, 2001: Transport and evolution of a winter-time Yellow sand observed in Korea. *Atmos. Environ.*, **35**, 3191-3201.

Lee, Y. C., and P. R. Hills, 2003: Cool season pollution episodes in Hong Kong, 1996-2002. *Atmos. Environ.*, **37**, 2927-2939.

Levitus, S., J. I. Antonov, J. L. Wang, T. L. Delworth, K. W. Dixon, and A. J. Broccoli, 2001: Atmospheric warming of Earth's climate system. *Science*, **292**, 267-270.

Lin, C. Y. et al., 2004: Long-range transport of Asian dust and air pollutants to Taiwan. *TAO*, **15**, 759-784.

Lin, F. J., S. C. Hsu, and W. L. Jeng, 2000: Lead in the southern East China Sea. *Mar. Environ. Res.*, **49**, 329-342.

Lin, T. H., 2001: Long-range transport of yellow sand to Taiwan in spring 2000: Observed evidence and simulation. *Atmos. Environ.*, **35**, 5873-5882.

Liu, C. L., J. Zhang, and Z. B. Shen, 2002: Spatial and temporal variability of trace metals in aerosols from the desert region of China and the Yellow Sea. *J. Geophys. Res.*, **102**D, doi: 10.1029/2001JD000635.

Liu, S. C., and C. J. Shiu, 2001: Asian dust storms and their impact on the air quality of Taiwan. *Aerosol Air Qual. Res.*, **1**, 1-8.

Liu, S. C., C. H. Wang, C. J. Shiu, H. W. Chang, C. K. Hsiao, and S. H. Liaw, 2002: Reduction in sunshine duration over Taiwan: Cause and implications. *TAO*, **13**, 523-545.

Liu, T. S., 1985: Loess and the Environment, China Ocean, Beijing., 251p.

Lung, S. C. C., C. H. Liu, S. Y. Huang, T. J. Lin, C. C. K. Chou, and S. C. Liu, 2004: Water-soluble ions of aerosols in Taipei in spring 2002. *TAO*, **15**, 901-923.

Ma, C. J., M. Kasahara, R. Holler, and T. Kamiya, 2001: Characteristics of single particles sampled in Japan during the Asian dust-storm period. *Atmos. Environ.*, **35**, 2707-2714.

Mao, I. F., and M. L. Chen, 1996: Airborne lead pollution in metropolitan Taipei (Republic of China). Water Air Soil Poll., 375-382.

Marcazzan, G. M., S. Vaccaro, G. Valli, and R. Vecchi, 2001: Characterization of PM_{10} and $PM_{2.5}$ particulate matter in the ambient air of Milan (Italy). *Atmos. Environ.*, **35**, 4639-4650.

Merrill, J., E. Arnold, M. Leinen, and C. Weaver, 1994: Mineralogy of aeolian dust reaching the North Pacific Ocean 2. Relationship of mineral assemblages to atmospheric transport patterns. *J. Geophys. Res.*, **99**, 21025-21032.

Nishikawa, M., Q. Hao, and M. Morita, 2000: Preparation and evolution of certified reference materials for Asian mineral dust. *Global Environ. Res.*, **4**, 103-113.

Nriagu, J. O., and J. M. Pacyna, 1988: Quantitative assessment of worldwide contamination of air, water and soils by trace metals. *Nature*, **333**, 134-139.

Pakkanen, T. A., V. M. Kerminen, C. H., Korhonen, R. E. Hillamo, P. Aarnio, T. Koskentalo, and W. Maenhaut, 2001: Use of atmospheric elemental size distributions in estimating aerosol sources in the Helsinki area. *Atmos. Environ.*, **35**, 5537-5551.

Patterson, C. C., and D. M. Settle, 1987: Review of data on eolian fluxes of industrial and natural lead to the land and seas in remote regions on a global scale. *Mar. Chem.*, **22**, 137-162.

Sakata, M., M. Kurata, and N. Tanaka, 2000: Estimating contribution from municipal solid waste incineration to trace metal concentrations in Japanese urban atmosphere using lead as a marker element. *Geochem. J.*, **34**, 23-32.

Sun, J., M. Zhang, and T. Liu, 2001: Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960-1999: Relations to source area and climate. *J. Geophys. Res.*, **106**, 10325-10333.

Taylor, S. R., 1964: Abundance of chemical elements in the continental crust: A new table. *Geochim. Cosmochim. Acta*, **28**, 1273-1285.

Uematsu, M., A. Yoshikawa, H. Muraki, K. Arao, and I. Uno, 2002: Transport of mineral and anthropogenic aerosols during a Kosa event over East Asia. *J. Geophys. Res.*, **107**, D12, doi: 10.1029/2001JD000333.

Utsunomiya, A., and S. Wakamatsu, 1996: Temperature and humidity dependence on aerosol composition in the northern Kyushu, Japan. *Atmos. Environ.*, **30**, 2379-2386.

VanCuren, R. A., and T. A. Cahill, 2002: Asian aerosols in Northern America: Frequency and concentration of fine dust. *J. Geophys. Res.*, **107**, D24, doi: 10.1029/2002JD002204.

Waldman, J. M., P. J. Lioy, M. Zelenka, L. Jing, Y. N. Lin, Q. C. He, Z. M. Qian, R. Chapman, and W. E. Wilson, 1991: Wintertime measurements of aerosol acidity and trace elements in Wuhan, a city in central China. *Atmos. Environ.*, **25**B, 113-120.

Wei, F., E. Teng, G. Wu, W. Hu, W. E. Wilson, R. S. Chapman, J. C. Pau, and J. Zhang, 1999: Ambient concentrations and elemental compositions of PM_{10} and $PM_{2.5}$ in four Chinese cities. *Environ. Sci. Technol.*, **33**, 4188-4193.

Winchester, J. W., and M. T. Bi, 1984: Fine and coarse aerosol composition in an urban setting: a case study in Beijing, China. *Atmos. Environ.*, **18**, 1399-1409.

Xu, L., K. Okada, Y. Iwasaka, K. Hara, Y. Okuhara, Y. Tsutsumi, and G. Shi, 2001: The composition of individual aerosol particle in the troposphere and stratosphere over Xianghe (39.45°N, 117.0°E). *China. Atmos. Environ*., **35**, 3145-3153.

Young, C. Y., K. H. Li, and C. M. Liu, 1997: Comparisons between long-range transported yellow dust and air quality monitoring of local pollutants: Data analysis from March 12 to 16, 1995. *Environ. Prot*., **12**, 1-38. (in Chinese)

Yu, H., S. C. Liu, and R. E. Dickinson, 2002: Radiative effects of aerosols on the evolution of the atmospheric boundary layer. *J. Geophys. Res*., **107**, D12, doi: 10.1029/2001JD000754.

Zhang, D., and Y. Iwasaka, 1999: Nitrate and sulfate in individual Asian dust-storm particles in Beijing, China in spring of 1995 and 1996. *Atmos. Environ*., **33**, 3213-3223.

Zhang, J., Y. Wu, C. L. Liu, Z. B. Shen, Z. G. Yu, and Y. Zhang, 2001: Aerosol characters from the desert region of northwest China and the Yellow Sea in spring and summer: observations at Minqin, Qingdao, and Qianliyan in 1995 - 1996. *Atmos. Environ*., **35**, 5007-5018.

Zhang, X. Y., R. Arimoto, and Z. S. An, 1997: Dust emission from Chinese desert sources linked to variations in atmospheric circulation. *J. Geophys. Res*., **102**, 28041-28047.

Zhang, X. Y., R. Arimoto, G. H. Zhu, T. Chen, and G. Y. Zhang, 1998: Concentration, size-distribution and deposition of mineral aerosol over Chinese desert regions. *Tellus*, **50**B, 317-331.

Zhang, X. Y., R. Arimoto, Z. S. An, T. Chen, G. Zhang, G. Zhu, and X. Wang, 1993: Atmospheric trace elements over source regions for Chinese dust: concentrations, sources and atmospheric deposition on the loess plateau. *Atmos. Environ*., **27**A, 2051-2067.

Zieman, J. J., J. L. Holemes, D. Connor, C. R. Jensen, W. H. Zoller, D. M. Hermann, J. R. Parrington, and G. E. Gordon, 1995: Atmospheric aerosol trace element chemistry at Maunna Loa Observatory 1. 1979-1985. *J. Geophys. Res*., **100**, 25979-25994.

Zhou, M., K. Okada, F. Qian, P. M. Wu, L. Su, B. E. Casareto, and T. Shimohara, 1996: Characteristics of dust-storm particles and their long-range transport from China to Japan-case studies in April 1993. *Atmos. Res*., **40**, 19-31.