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Science of the Total Environment 347 (2005) 111–121

Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind

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Variations of Cd/Pb and Zn/Pb ratios in Taipei aerosols reflecting long-range transport or local pollution emissions

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Received 4 August 2004; accepted 17 December 2004
Available online 3 February 2005

Abstract

Along with windblown dust, large quantities of pollutants are annually brought out of continental China by the westerlies in winter and spring; thereafter, they are partly subjected to transport by northeastern monsoon winds to Taiwan. To characterize the heavy metal composition differences between long-range transported and local aerosols and to evaluate metal contributions from long-range transported aerosols during the northeastern monsoon season, both PM_{10} and $PM_{2.5}$ aerosols collected from Taipei, Taiwan from February 2002 to March 2003 were analyzed for three selected heavy metals, namely Pb, Cd and Zn using ICP-MS. Monthly patterns show that Pb concentrations in winter (62 ng/m^3) were over two times higher than those in the other seasons, which is attributed to long-range transport from areas under development in China. Low Cd/Pb (0.017) and Zn/Pb (1.82) ratios were measured in aerosols collected during the Asian dust period, in which the ambient aerosols consisted predominantly of long-range transported pollutants. By contrast, high Cd/Pb (0.030) and Zn/Pb (3.44) ratios were observed during the summer monsoon season, in which aerosols were dominated by local pollutant emissions. Cd/Pb and Zn/Pb ratios appear to be successfully applied to identify the pollutants originating principally from the long-range transport or from local emissions. In addition, by assuming that a significant fraction of heavy metals associated with coarse airborne dust have settled to the sea prior to reaching Taiwan in spring, a mechanism is suggested to explain why higher anthropogenic metal concentrations occurred in winter than those in dust-rich spring.

Keywords: Aerosol; Long-range transport; Local pollution; Taipei

1. Introduction

Asian dust particles are transported over long distances by the westerly winds annually in late winter and spring, which substantially enhance the

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doi:10.1016/j.scitotenv.2004.12.021

ambient aerosol concentration in downwind regions and countries (Duce et al., 1980; Uematsu et al., 1983; Husar, 2001; Mukai and Suzuki, 1996). Along with windblown dust, tremendous amounts of pollutants accompanying these dust-rich atmospheric systems are delivered out of the Chinese continent, primarily because of increased development in industries and common household coal combustion (Kaneyasu et al., 2000; Prospero et al., 2003). Thereafter, part of these natural and polluted substances are subjected to transport southward to Taiwan by cold fronts followed by prevailing northeastern monsoon winds (Liu and Shiu, 2001; Lin, 2001; Hsu et al., 2004; Lin et al., 2004) which dominate almost the whole period of winter and spring. Fig. 1 depicts the dust regions in China and the synoptic transport pathway.

For the areas surrounding China (e.g. Korea, Japan, Hong Kong, etc.), a number of investigations have been conducted on diverse aspects of Asian dust (AD), including chemical, physical and optical properties and transport processes (Merrill et al., 1994; Fang et al., 1999). Extensive studies have also been devoted to characterizing and quantifying the anthropogenic contaminants, particularly sulfur, nitrogen and carbonaceous chemicals (Qi et al., 1995; Zhang and Friedlander, 2000; Streets et al., 2001). It has also been found that coal combustion, industrial emissions, and vehicular exhaust have resulted in more severe air pollution of heavy metals (Zhang et al., 1998; Wei et al., 1999). For example, elevated atmospheric concentrations of Pb almost as high as 1 $\mu g/m^3$ were frequently measured in ambient air in China (Zhang et al., 1998; Zheng et al., 2004), which caused high blood lead content in children (Gao et al., 2001). Nevertheless, until now available data and information on the characterization of airborne heavy metals are still scarce (Wei et al., 1999; Wang et al., 2000; Zhang and Friedlander, 2000). Regarding heavy metals, we are often concerned about Pb and Cd because of their threats to human health. In addition to impacts on public health, they are also significant to ecological environments. Lin et al. (2000) reported that the surface Pb concentration in the southern East China Sea is the highest ever measured over the marginal seas and open oceans of the world. They attributed this result to high levels of eolian Pb depositions predominantly from the Chinese mainland.

Hence, more efforts are merited to qualitatively and quantitatively assess and characterize the contribution and characterization of heavy metals in aerosol particles from long-range transport and local pollution emissions. Atmospheric concentration alone cannot be used to achieve these goals; specific fingerprints of heavy metals may be effective for source apportion-

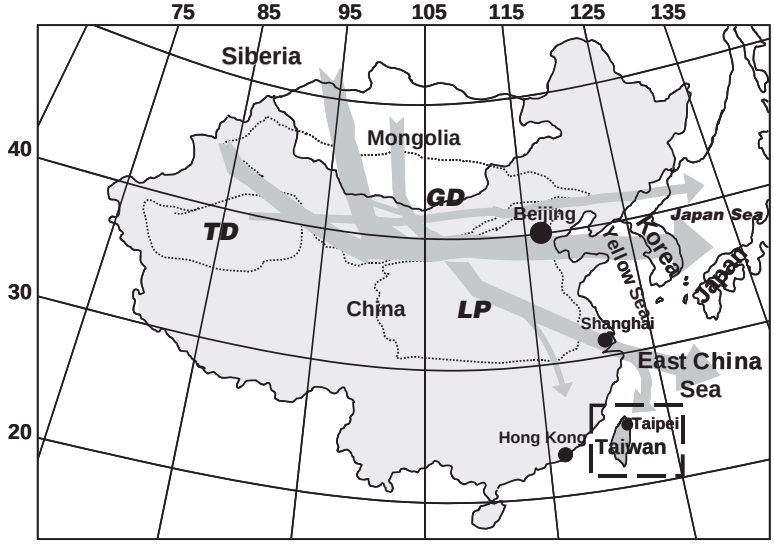

Fig. 1. A regional map for the sampling location (Taipei, Taiwan). Three main dust regions in China are indicated by dotted area, including Takalarmakan Desert (TD), Gobi Desert (GD) and Loess Plateau (LP). The synoptic transport routes are indicated by arrows, which were modified from Zhang et al. (1993), Zhang et al. (1997), Xuan (1999) and Sun et al. (2001).

ment, such as Pb for leaded gasoline (Nriagu and Pacyna, 1988), Sb and Se for coal combustion (Nriagu and Pacyna, 1988; Arimoto et al., 1996), V and Ni for heavy oil (Var et al., 2000), etc. Still, they cannot be used to identify the source regions, i.e. cross-boundary transport vs. local pollutions. Unfortunately, the mixing process of multiple components and/or sources during transport and dispersion will make this identification more difficult. Until now, few characteristic fingerprints were reported by previous studies to differentiate the source regions of anthropogenic metals in air pollutants (Herut et al., 2001), especially for the East Asia region (Mukai et al., 1994). This study is aimed to establish temporal variations of Pb, Cd and Zn concentrations in aerosols on varying time scales and to specify characteristics of these selected metals for differentiating the local pollution emissions from the long-range transport of airborne Pb, Cd and Zn.

2. Materials and methods

The sampling site was located at the Central Weather Bureau (CWB) in metropolitan Taipei, Taiwan (Fig. 1). The Partisol® Model 2300 speciation sampler (Rupprecht & Patashnick, Albany, NY, USA) was employed to simultaneously collect PM_{10} and $PM_{2.5}$ ambient aerosol particles. The samplers were set on the rooftop of the CWB building approximately 25 m above ground level. The medium used for aerosol filtration was polytetrafluoroethylene (PTFE) membrane filters (PallGelman, 2.0 μm pore size and 47 mm in diameter with a PE ring). Sampling was conducted from February 2002 to March 2003. In principle, sampling was carried out in the last week of each month; besides, additional sampling was made when dust events were forecasted by the Environmental Protection Administration (EPA) of Taiwan. Samples were collected at 12-h intervals: from 8:00 am to 20:00 pm and from 20:00 pm to 8:00 am the following day. They were categorized into daytime and nighttime samples, respectively. A total of 186 sets of PM_{10} and $PM_{2.5}$ aerosol samples were collected in the study. Seven strong dust episodes occurred in the springtime of 2002 (as indicated by the grayed area on the *X*-axis in Fig. 2), which has been identified by the presence of crustal-derived elements of high

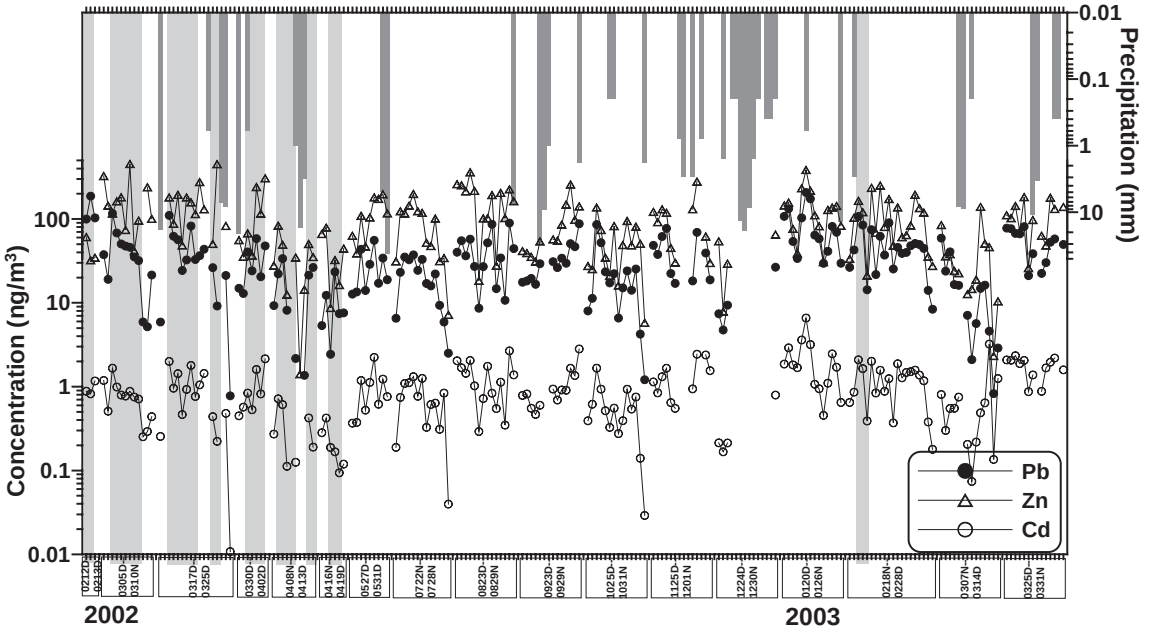

Fig. 2. Temporal variations of Pb, Zn and Cd concentrations in PM_{10} aerosols collected from northern Taiwan from February 2002 to March 2003. Precipitations are also plotted as a bar chart in the upper plot (right axis). The *X*-axis shows the individual observational periods in each month. The periods of Asian dust episodes are indicated by the grayed areas on the *X*-axis.

Al and Ca concentrations as well as air-mass back trajectory analysis (Hsu et al., 2004); only one weak dust episode was measured in 2003.

The aerosol samples on PTFE filters were dried, weighed, and then completely dissolved in a mixture of acids, i.e. 5 ml HNO_3, 5 ml HF and 0.5 ml $HClO_4$ (all Suprapur grade from Merck) in Teflon beakers. The digestion method is described elsewhere (Hsu et al., 2004). All containers used in the study were acid-cleaned prior to use. Metal determinations were made using a quadrupole-based inductively coupled plasma mass spectrophotometer (ICP-MS, Elan 6100, Perkin-Elmer™ Instruments, USA). In the study, the detection limits were 0.2 ng/m^3 for Pb, 0.5 ng/m^3 for Zn and 0.01 ng/m^3 for Cd. Calibration was achieved using a multi-element standard prepared from stock standard (Merck) made up in a 2% HNO_3 solution. The quality assurance and quality control (QA/QC) of analyses were validated by analyzing 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) (Nishikawa et al., 2000). The recovered values for all the target elements fell into the range or within 5% of certified values (except for Cd which was 8% for SRM1648) (Hsu et al., 2004).

3. Results and discussion

3.1. Atmospheric concentrations of Pb, Cd and Zn and their temporal variations

The data of Pb, Zn and Cd concentrations in PM_{10} and $PM_{2.5}$ aerosols collected from northern Taiwan from February 2002 to March 2003 are summarized in Table 1. The annual means of metal concentrations in PM_{10} are 34, 79 and 0.82 ng/m^3 for Pb, Zn and Cd, respectively, and those in $PM_{2.5}$, 26, 60 and 0.66 ng/m^3, respectively. Compared to those concentrations (e.g. 80–2690 ng/m^3 for Pb and 1–70 ng/m^3 for Cd) reported in China (Zhang et al., 1998; Wei et al., 1999), ours are very low. The temporal variations of Pb, Zn and Cd concentrations in PM_{10} aerosols are illustrated in Fig. 2, with the measured rainfall to show the scavenging effect. In general, the three metal patterns are similar peaking at the same time in some cases, for example, 020306D (denoting 2002/03/06 daytime), 020318D, 020829D, 020929D, 021026D, 030120D, 030120N, 030122D–0123D, 030219N and 030223D, of which most (except 020829 and 020929D) were in the NE monsoon season and some in the AD period as indicated in Fig. 2. Fang et al. (1999) also found that atmospheric Pb concentrations can be enhanced by long-range transported AD episodes in Hong Kong. It suggests the three metals originating from common sources (or source regions) in most cases. However, there is some inconsistency as seen in Fig. 2, indicating that other sources and/or transport processes need to be considered for individual metals.

Table 1
Summary of Pb, Zn and Cd concentrations (ng/m^3) in PM_{10} and $PM_{2.5}$ aerosols collected from northern Taiwan from February 2002 to March 2003, in which ranges and annual means (in parentheses) are given

	Pb	Zn	Cd
PM_{10}	ND[a]–207 (34)	ND–454 (79)	ND–6.6 (0.82)
$PM_{2.5}$	ND–285 (26)	ND–577 (60)	ND–11 (0.66)

[a] Non-detectable.

Figs. 3 and 4 respectively illustrate monthly and seasonal variations of atmospheric Pb, Cd and Zn. Geometric means were used for expressing monthly concentrations; seasonal concentrations were arithmetically averaged from monthly concentrations. Winter includes December, January and February, spring includes March, April and May, etc. For Pb, monthly concentrations peak in February of 2002 (124 ng/m^3), which is much higher than those in other months, but only three data are available for this month. In the case of Cd, the highest concentration (1.7 ng/m^3) occurred in January of 2003. The largest monthly average concentration (138 ng/m^3) of Zn is shown in August of 2002, but the differences in Zn concentrations between for all months are less than those of Pb and Cd. Apart from August, other months exceeding the annual concentration for Zn include March (118 ng/m^3), May (100 ng/m^3) and November (98 ng/m^3) of 2002 and January (116 ng/m^3) and February (87 ng/m^3) of 2003; all these months are in the NE monsoon season. Low concentrations (even below detection limits) for all three the metals measured in

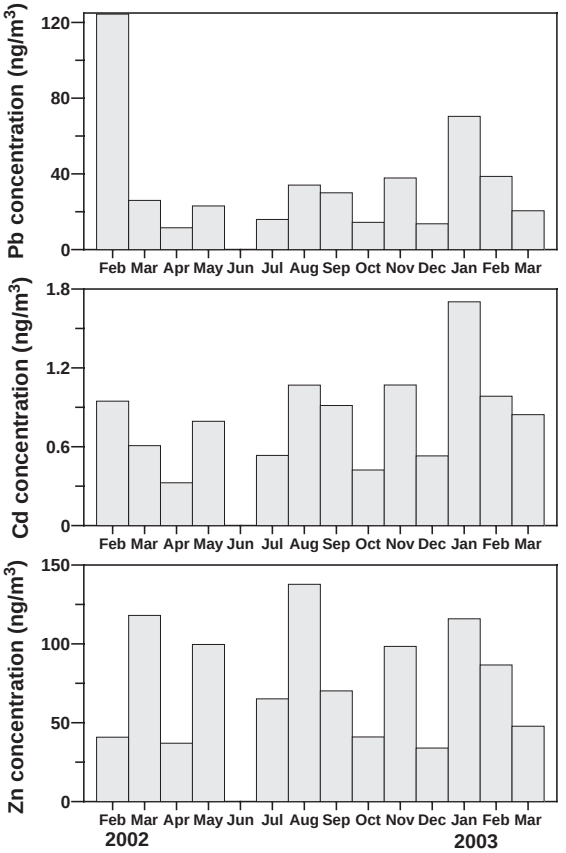

Fig. 3. Monthly variations of Pb, Cd and Zn concentrations in PM_{10} aerosols.

December of 2002 were apparently due to the scavenging effect of frequent rain in that period (Fig. 2) (Hsu et al., 2004). The overall monthly variations coincide with the monthly rainfall patterns, such as high in August and low in December, especially for rainy days (Fig. 4). This demonstrates again the precipitation effect, but the effect is not linearly correlated with rainfall figures. However, concerning wind speeds and directions, no meaningful relationships were found (not shown).

Seasonal patterns show that winter has the highest concentrations of atmospheric Pb and Cd, whereas summer has the highest Zn (Fig. 5). Note that the winter Pb concentration (62 ng/m^3) is over two times higher than the other three seasons (at a 95% confidence level), similar to the observations in Japan (Var et al., 2000), Korea (Kim and Song, 1997) and Hong Kong (Cheng et al., 2000), while the differences in concentrations of Cd and Zn between the four seasons are smaller than that of Pb. The phase-out of leaded gasoline in Taiwan was officially completed in 2000. Reductions in Pb emissions are evident over the last 10 years. For instance, Mao and Chen (1996) determined a mean atmospheric Pb concentration of 700±390 ng/m^3 in Taipei City in 1991; Fang et al. (2002) obtained a concentration range of 20–89 ng/m^3 in central Taiwan from mid-1998 to early 2001. Elevated Pb concentrations of up to several hundreds ng/m^3 were often measured in China (Zhang et al., 1998; Wei et al., 1999) because the utilization of leaded gasoline has not been completely phased out. The present high concentrations of heavy metals in winter seem to be contributed by long-range transport from China since Taiwan is under the influence of the NE monsoon winds in most of winter and spring (Lin et al., 2004). This also caused high winter concentrations of eolian Pb in Korea and Japan (Kim and Song, 1997; Var et al., 2000). As elucidated by the air-mass back trajectory analysis (not shown), the cold air parcels originating from northern China probably mixed with the polluted air when passing through a few major cities like Beijing and Shanghai

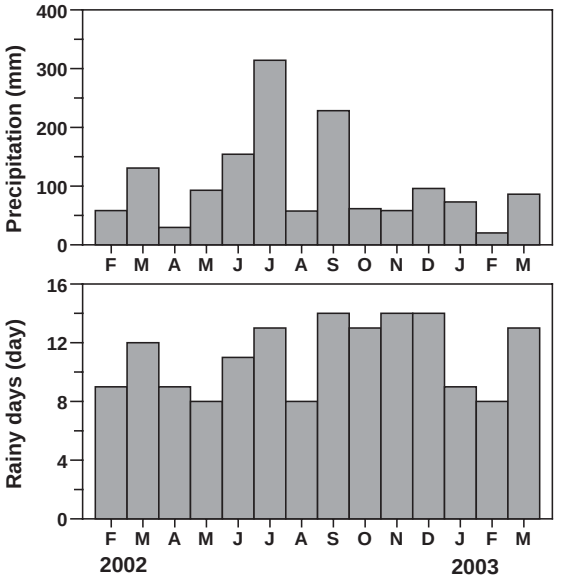

Fig. 4. Monthly patterns of rainfall amount (upper panel) and rainy days (lower panel) in the experimental period (from 2002 February to 2003 March).

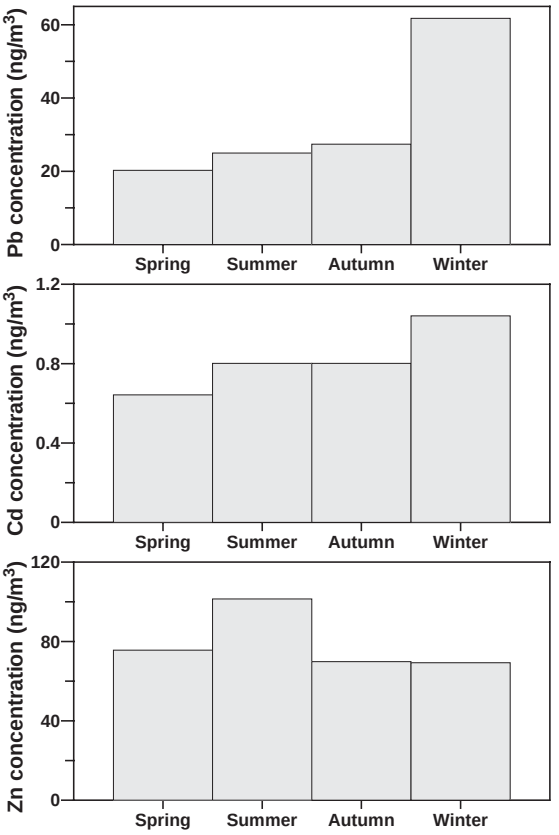

Fig. 5. Seasonal variations of Pb, Cd and Zn concentrations in PM_{10} aerosols.

in the NE monsoon season, and carried pollutants to the neighboring areas. Along with relative contributions from different dominant sources (e.g. long-range transport vs. local pollution emission, or incineration vs. vehicle emission or metalliferous industry), the magnitudes of atmospheric concentrations of certain anthropogenic metals are controlled by other factors (such as removal processes, including dry and wet depositions via which certain chemical species will be susceptible or insusceptible, as well as the behavior of "host" particles of interested metals since emitted metals usually interacted and associated within existing particles such as dust). Of course, the combination of these causes may be possible to make temporal variability of specific metals and the correlation of inter-metals (as described below) more inconsistent.

Additionally, we have also considered the differences between the daytime and nighttime concentrations for studied metals. The results, summarized in Table 2, show statistical differences for Zinc, but not for Pb and Cd. This likely indicates that Zn is contributed primarily by local pollution relevant to more frequent human activities in the daytime than in the nighttime, like heavy traffic for example.

We also raise an important question regarding the occurrence of high concentrations of heavy metals (at least Pb and Cd) in winter as opposed to spring, similar to dust-derived elements (e.g. Al, Ca and Ti, our unpublished data), even though a few pronounced concentrations of Pb, Cd and Zn were observed during the AD episodes (Fig. 2). This probably relates to different source strengths of pollution emissions between winter and spring in China and/or modification and fractionation of aerosol compositions during transport (Herut et al., 2001). It will be discussed in detail in the following sections.

3.2. Size distribution and enrichment factors

Monthly variations of relative proportions of coarse ($PM_{2.5–10}$) and fine ($PM_{2.5}$) Pb, Cd and Zn in aerosols are displayed in Fig. 6, while seasonal variations are shown in Fig. 7. Apparently, these heavy metals tend to reside in fine aerosols. Monthly patterns of the three metals reveal significant changes in relative size proportions with individual months. In general, the fine mode dominates the size distribution for the three metals, especially in winter (January and February), while nearly half (over 40%) of these heavy metals partition within coarse aerosols in the high-dust spring of 2002 (particularly April) (Fig. 6). By contrast, fine modes absolutely dominate the size distribution in winter of both 2002 and 2003 (especially January and February) (Figs. 6 and 7). It is thus inferred that a fraction of heavy metals

Table 2
Daytime and nighttime mean concentration for atmospheric Pb, Cd and Zn

Metal	Daytime mean (ng/m^3)	Nighttime mean (ng/m^3)	p value
Pb	40	36	>0.2[a]
Cd	1.08	0.97	>0.1[a]
Zn	119	88	<0.01[b]

[a] Statistically insignificant.
[b] Statistically significant.

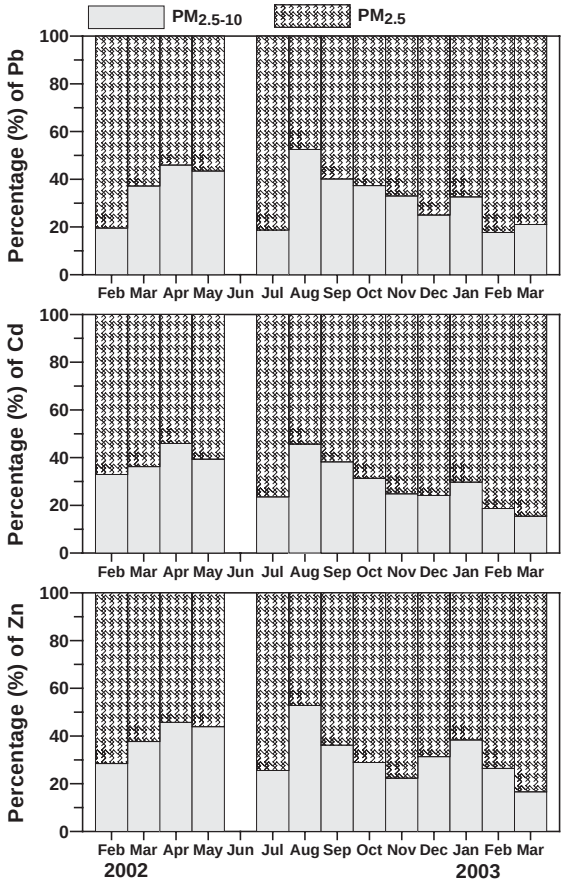

Fig. 6. Monthly variations of relative proportions of coarse ($PM_{2.5–10}$) and fine ($PM_{2.5}$) aerosol particle fractions for Pb, Cd and Zn, which is represented as percentage (%).

associated with coarse dust aerosols have settled with dust particles during transport across the wide-open sea areas in high-dust spring. As a consequence, the amounts of heavy metals delivered to Taiwan are reduced compared to those of the low-dust winter. Such a mechanism may be responsible for the fact that spring concentrations of the three selected metals are not high. It can also explain the observation that the partitions between the two size fractions for the three heavy metals in March of 2003 are more consistent than those in winter (Fig. 6), because no strong AD events occurred in 2003. When considering the seasonality of the two size fractions for the three metals, they obviously show small differences between the four seasons (Fig. 7).

Atmospheric concentrations of trace metals can vary over a relatively large range, but by normalizing them to their crustal concentrations the "chemical character" of aerosol particles can be investigated effectively (Chester et al., 1993). An enrichment factor (EF) can be used to evaluate the anomaly of specific chemical species of ambient aerosols with respect to representative compositions of a reference material (e.g. average crust and sea water). For the crustal source, Al is normally used as the crustal indicator element, and the EF_{crust} value of element X can be calculated according to the following equation:

$$EF_{crust} = (C_{X-aerosol}/C_{Al-aerosol})/(C_{X-crust}/C_{Al-crust})$$

in which $C_{X\text{-aerosol}}$ and $C_{Al\text{-aerosol}}$ are the concentrations of element X and Al in aerosols, respectively,

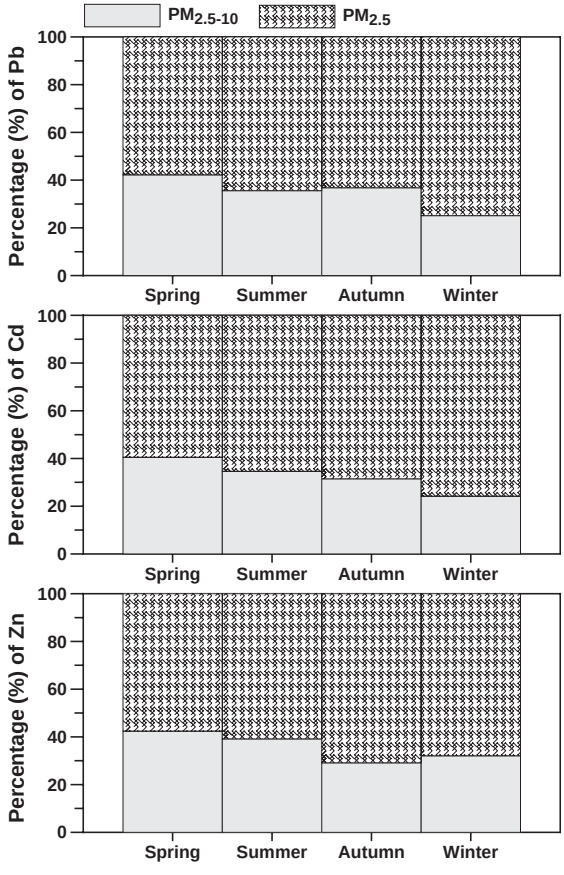

Fig. 7. Seasonal variations of relative proportions of coarse ($PM_{2.5–10}$) and fine ($PM_{2.5}$) aerosol particle fractions for Pb, Cd and Zn, which is represented as percentage (%).

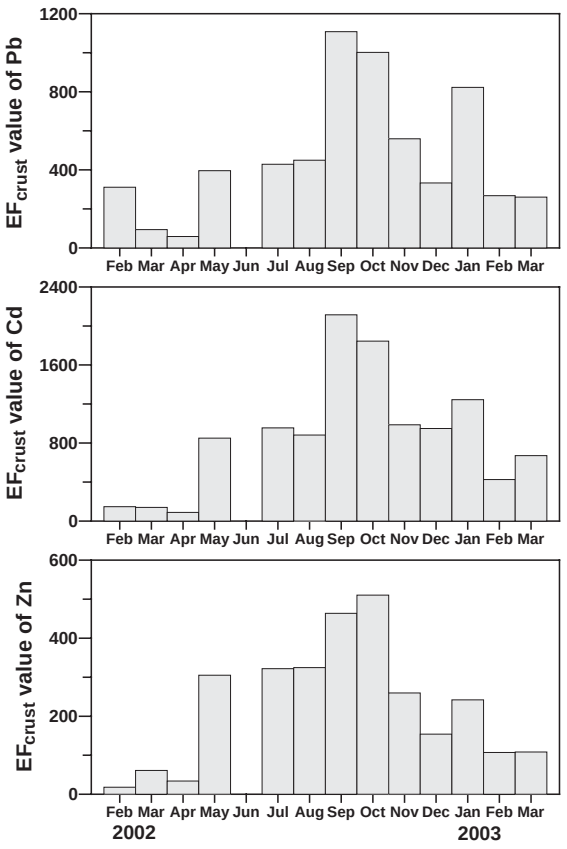

Fig. 8. Monthly variations of enrichment factors relative to average crust compositions (Taylor, 1964) (EF_{crust}) for Pb, Cd and Zn in PM_{10} aerosols.

and $C_{X\text{-crust}}$ and $C_{Al\text{-crust}}$ are their concentrations in average crustal material (Taylor, 1964). The calculated results of enrichment factors for these three anthropogenic metals show several hundred times of enrichment (on average, 250 for Zn, 500 for Pb, and 900 for Cd) relative to natural crust compositions. Fig. 8 depicts monthly variations of EF_{crust} values. Low EF_{crust} values for Pb, Cd and Zn in March and April of 2002 obviously indicate a dilution effect of heavy metals by large amounts of Asian dust (Hsu et al., in press); this is in good agreement with the fact that the proportions of coarse fractions for anthropogenic metals increased in that time (Fig. 6). High EF_{crust} values occur in September and October, which likely results from, to some extent, less dust and high pollutants at that time when local pollution aerosol sources dominate.

3.3. Characteristic inter-metal ratios for differentiating local emissions from long-range transport of pollutants

For comparison, the data on metal concentrations are categorized into three groups, i.e. the AD period (all in the NE monsoon season), the NE monsoon season (from late October to mid-May, excluding the AD period), and the summer monsoon season (from late May to mid-October). The AD case represents the domination of long-range transport, whereas the summer monsoon case, in which the southern to

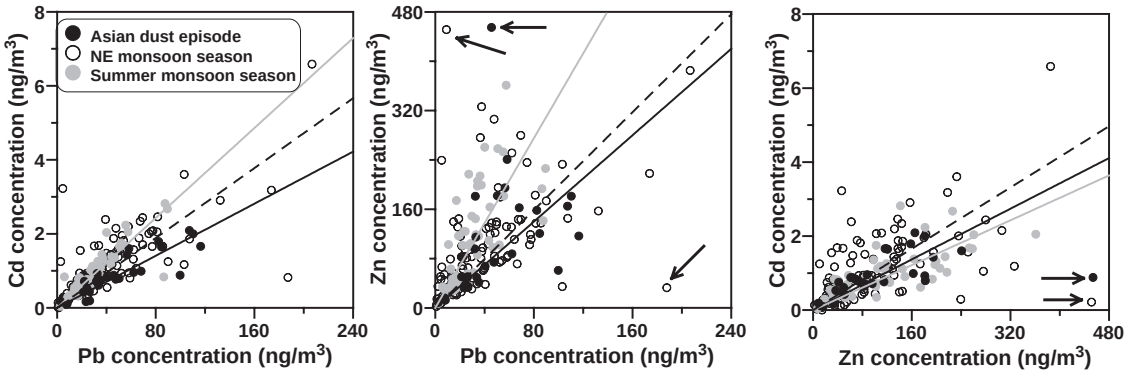

Fig. 9. Linear regression plots of Cd vs. Pb concentrations (left plot), Zn vs. Pb concentrations (middle plot), and Cd vs. Zn concentrations (right plot). Data are grouped into three sets: the Asian dust period, the northeastern monsoon season (excluding the former), and the summer monsoon season (from late May to middle October). Some outliers (indicated by arrows) are excluded for regression.

Table 3
Results of linear regression for Pb vs. Cd, Pb vs. Zn, and Zn vs. Cd

	Linear regression equation	*R*-square	*p* value	Sample number
Pb vs. Cd				
AD period	$Y=0.018X$	0.77	⩽0.001	29
NE monsoon season	$Y=0.024X$	0.52	⩽0.001	108
Summer monsoon season	$Y=0.030X$	0.70	⩽0.001	48
Pb vs. Zn				
AD period	$Y=1.75X$	0.24	<0.01	28
NE monsoon season	$Y=1.98X$	0.34	⩽0.001	103
Summer monsoon season	$Y=3.44X$	0.38	⩽0.001	48
Zn vs. Cd				
AD period	$Y=0.0086X$	0.60	⩽0.001	28
NE monsoon season	$Y=0.0104X$	0.38	⩽0.001	104
Summer monsoon season	$Y=0.0076X$	0.42	⩽0.001	48

southwestern winds prevail, represents the domination of local pollution emissions. Based on this classification, the correlations of Cd vs. Pb, Zn vs. Pb, and Cd vs. Zn are shown in Fig. 9; all correlations are statistically significant at a 95% confidence level (Table 3). The positive correlation may be interpreted as common predominant origins or similar source regions/transport processes for the three metals. High Cd/Pb (0.030) and Zn/Pb (3.44) ratios were obtained for the summer monsoon period, while low Cd/Pb (0.018) and Zn/Pb (1.75) ratios were obtained for the AD period. The NE monsoon period has moderate ratios (0.024 for Cd/Pb and 1.98 for Zn/Pb). The difference in the Cd/Zn ratio between the AD (0.086) and summer monsoon (0.076) periods is insignificant. Nevertheless, a few outliers away from regression lines likely indicate that unidentified local sources of certain anthropogenic metals occasionally occurred. Overall, the mean values of Cd/Pb (0.018) and Zn/Pb (1.75) ratios for the AD period are very consistent with the yields from atmospheric concentrations reported for a few cities of China (Zhang et al., 1998; Liu et al., 2002) and for Hong Kong (Qin et al., 1997; Zheng and Fang, 2000). High atmospheric Pb concentrations perhaps owing to extensive continual use of leaded gasoline and coal combustion in China appear to be responsible for low Cd/Pb and Zn/Pb ratios. Usually, municipal waste incineration is a very important pollution source of heavy metals in many metropolitan cities (Sakata et al., 2000; Hu et al., 2003). Moreover, high Cd/Pb (0.030) and Zn/Pb (3.44) ratios determined in the summer monsoon season are comparable with those measured in the vicinity of incineration sites (Sakata et al., 2000; Hu et al., 2003).

In order to quantify the temporal variability of Cd/Pb and Zn/Pb ratios, we defined the delta Cd/Pb and delta Zn/Pb ratios to be equal to the difference between the measured Cd/Pb and Zn/Pb ratios and the mean ratios for the NE monsoon season (0.024 for Cd/Pb and 2.0 for Zn/Pb) for certain samples:

$$\Delta\mathrm{Cd/Pb} = \mathrm{Cd/Pb}_{\mathrm{measured}} - 0.024$$

$$\Delta\mathrm{Zn/Pb} = \mathrm{Zn/Pb}_{\mathrm{measured}} - 2.0$$

The temporal variability of delta Cd/Pb ratio and delta Zn/Pb ratio is displayed in Fig. 10. It can be clearly seen in these plots that low ratios occur in

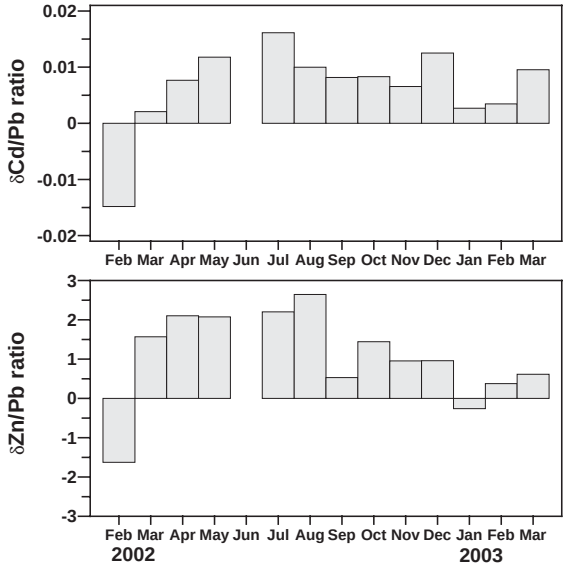

Fig. 10. Monthly variations of delta Cd/Pb ratio and delta Zn/Pb ratio, of which definitions are described in the text.

winter (particularly January and February) and that the values gradually increase with time, reaching maxima in summer (particularly July and August). This demonstrates the change of metal sources (source regions) with seasons, from the domination of long-range transport in the NE monsoon season to that of local pollution emissions in the summer monsoon season. It could be speculated that the growing numbers of vehicles will continually produce large quantities of air pollutants (including eolian Pb), although China has commonly switched to unleaded gasoline since 2000. In other words, lead air pollution will still be a crucial issue in China in future years; low metal to Pb ratios are still characteristic fingerprints to evaluate the impact on the atmospheric Pb from mainland China for the downwind countries.

4. Conclusion

In the study, some conclusions can be drawn as follows:

1. Most of the marked concentrations for aerosol Pb, Zn and Cd were measured in the NE monsoon season (including spring and winter), of which some overlapped within the AD period. Apparently, atmospheric Pb, Cd and Zn can be enhanced by long-range transport from continental China during the northeastern monsoon season.
2. Seasonality of atmospheric Pb and Cd showed a winter maximum higher than the other seasons' concentrations by a factor of 2 to 3, which is due to long-range transported contributions and a spring minimum, while Zn showed a summer maximum that is dominated by local pollution emissions. A mechanism is proposed to explain the spring minima of atmospheric Pb and Cd; they are preferential depositions of coarse airborne particles into the sea during transport before arriving in Taiwan in spring, as a significant proportion of anthropogenic metals possibly associated with high levels of coarse dust aerosols in that time.
3. Low Cd/Pb (averaging 0.017) and Zn/Pb (1.82) ratios were determined in aerosols collected in the Asian dust period and, by contrast, high Cd/Pb (0.030) and Zn/Pb (3.44) ratios were determined in the summertime aerosols. This is in good agreement with other studies conducted in East Asian countries. The chemical characteristics can be applied for differentiating contributions of anthropogenic metals from long-range transport or from local pollution emissions.

Acknowledgments

We are grateful to two anonymous reviewers for their constructive comments. We thank Ms. R.T. Hsu and Ms. Y.W. Chen for their assistance with sample pretreatment and chemical analyses and Messrs. T.K. Chen and C.H. Liu for their help with sampling. Thanks are also extended to Drs. C.C.C. Chou, C.Y. Lin and S.J. Kao for their useful comments and suggestions. This work was supported by the theme research project of Academia Sinica and Environmental Protection Administration grants EPA-91-FA11-03-A062 to S.C.L.

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