

# Characteristics of Atmospheric Metals during Dry Monsoon Season in the Coastal Region of Western Taiwan

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This study investigated the characteristics (elemental concentrations, size distributions, and dry deposition flux) of aerosol particles in the coastal region of western Taiwan during the dry monsoon (fall and winter) season. Factor analysis results show that resuspended soil dust is a significant source of particles. However, factor analysis is unable to define specific emission sources of noncrustal metals, probably because these metals are mixed by long-distance transportation, regardless of their sources. On average, coarse mode particles (with aerodynamic diameters of larger than 1.0  $\mu\text{m}$ ) contribute 66% of the total particle mass. These coarse particles represent 53%, 64%, 62%, and 44% of the mass of elements Al, Ca, Fe, and Ti, respectively. Coarse particles predominate dry deposition fluxes because the average percentages of dry deposition fluxes attributed to fine particles are only 0.2%, 0.2%, 0.18%, 0.22% and 0.19% of the total particle mass, the Al mass, the Ca mass, the Pb mass and the Ni mass, respectively. Both atmospheric concentrations and dry deposition fluxes of Al, Ca, Fe, Mn and Sr are strongly influenced by crustal sources. For Pb, Zn, Ni, Ba, Cu and Cr, the parent aerosol is soil-oriented but the dry deposition is not. Neither the parent aerosol nor the dry deposition of V and Cd is influenced by crustal sources. Enrichment factors of most metal crust elements are considerably lower in dry deposition than in the parent aerosol. Fractionation factor analysis results show that the crustal component of a metal element in a parent aerosol is fractionated from the atmosphere by a dry deposition sequence. Although the noncrustal sources are not important in dry deposition, noncrustal elements can be easily fractionated between particles and dry deposits. Among the elements investigated, Pb had the highest fractionation factor of 12.1.

**Keywords:** metal, size distribution, dry deposition, enrichment factor

## 1. Introduction

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The ever-increasing dispersion of metal elements through the atmosphere, water, and soil is of major concern because such elements endanger human health; initiate possible changes in natural

biochemical processes in all ecosystems, and inevitably accumulate in the food chain (Van Malderen et al., 1996; Fang et al., 2000). The atmosphere is an important environmental medium for the transport and deposition of these substances to terrestrial and aquatic systems (Scudlark et al., 1994; Wu et al., 1994).

Dry deposition - deposition in the absence of precipitation - is a continuous process and may influence the chemical properties of the surface of the earth. Dry deposition is thought to be similar to wet deposition in chemical impact. The former is accepted to be more important than the latter for chemical species associated with large particles (Wu et al., 1994; Golomb et al., 1997). Dry deposition theory suggests that the major aerosol removal mechanisms that occur in the atmosphere are strongly related to particle size, atmospheric turbulence, and atmospheric stability (Quinn and Ondov, 1998).

The metal components of aerosol and the characteristics of their deposition flux have been investigated in many contrasting coastal systems (including for example, the Mediterranean Sea, the North Sea, and the Irish Sea) (Ottley and Harrison, 1999; Chester et al., 2000; Herut et al., 2001). Some studies have estimated the inputs of anthropogenic trace metals into the world's oceans. For industrialized coastal areas, the atmospheric fluxes of some elements exceed that of their river inputs and dominate the total external inputs (Guieu et al., 1997; Guerzoni et al., 1999).

The Taiwan Strait, which separates Mainland China and Taiwan Island, is a longitudinal and narrow open sea. Over recent decades, energy consumption has increased rapidly in China and Taiwan as their economies have grown rapidly. Numerous publications have discussed the natural and anthropogenic sources of air pollution in China, and asserted that anthropogenic source emission strength is rapidly increasing (Akimoto and Narita, 1994; Zhang et al., 2000). Some pollutants, such as

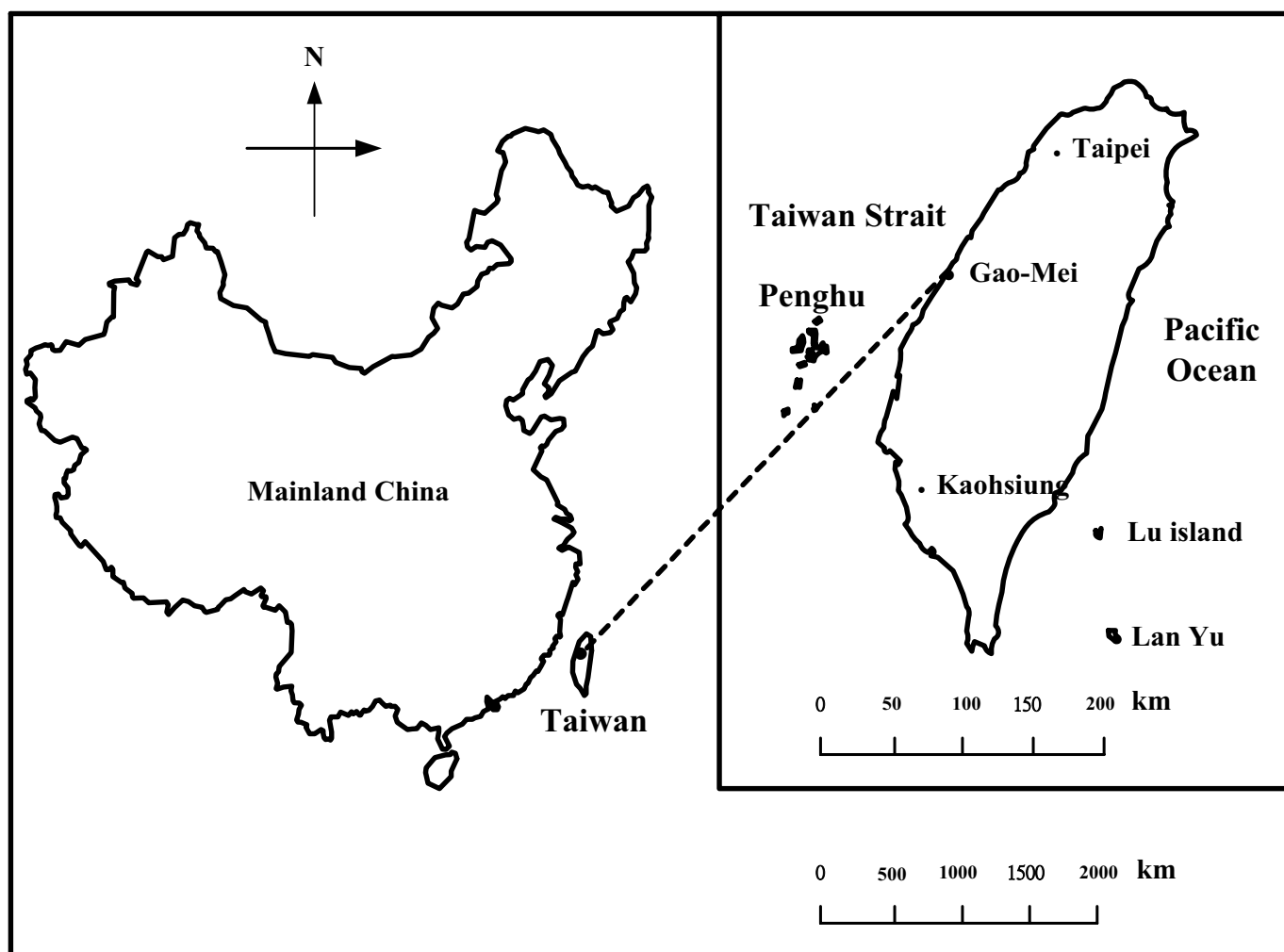
dust particles, are transported eastward by the westerly wind to the western Pacific regions, including Taiwan (Parungo et al., 1994; Zhang et al., 2000). In the past two decades, the population, industrial activity, and motor vehicle usage increased in the Taiwan area by 35%, 300%, and 1200%, respectively. These growths have inevitably exacerbated air pollution in the area. In addition, Taiwan's atmosphere is often influenced by the mixture of air masses from the China and Taiwan's coastal regions. However, aerosol data on the Taiwan Strait are scarce, and, in particular, data on the chemical composition of aerosols are very few. This work first presents the aerosol concentrations, the size distribution, and metal contents in the western Taiwan coastal region. These data were utilized to estimate metal dry deposition fluxes and identify their sources.

## 2. Experiment

### 2.1 Sampling

The prevailing wind in the summer is southerly or southeasterly; in the winter and fall, it is easterly or northeasterly, influenced by monsoons from north China. In the dry monsoon season (fall and winter), sources in both China and local areas contribute atmospheric pollutants in the region. Sampling was performed from September 2000 to March 2001 and all samples were taken over 24 hours. At least ten reliable data were obtained each month. Aerosol samples were collected at Gao-Mei, a coastal town in the western part of central Taiwan, adjacent to the Taiwan Strait (Fig. 1). The samplers were on the roof of a building, about 10 m above ground level.

A high-volume sampler PS-1, dry deposition lates and a MOUDI (Micro-Orifice Uniform Deposition Impactor) were used for sampling. Particulate mass and trace metal concentrations were measured using a high-volume sampler (General Metal Works PS-1), in which were by



**Figure 1.** Location of sampling site.

quartz filters with a 10.2 cm diameter and a 0.8  $\mu\text{m}$  pore size (Whatman International Ltd., 2500QAT-up) could maintain a small drop in pressure over 24-hour sampling period. The quartz filters were weighed before and after sampling to determine the amount of suspended particulate matter collected. The extent of dry deposition was measured by a plate with a smooth surface and a sharp leading edge, mounted on a wind vane (Holsen and Noll, 1992; Lee et al., 1995). The plate used in this study is similar to that used in wind tunnel studies (McCready, 1986). It is made of polyvinyl chloride (PVC) and is 21.5 cm long, 8.0 cm wide and 0.8 cm thick, with a sharp leading edge ( $< 10$ -degree angle) pointing into the wind. Three duplicate plate samples were taken next to each other during each sampling period to minimize system errors during the experiments.

Each plate was covered with Teflon strips (10 cm x 8 cm) coated with silicon grease (NO. 11025 silicon spray, Cling-surface CO., Inc., Angola, NY) to collect impacted aerosols. The strips were weighed before and after sampling to determine the total mass of the particles collected. The strips were then digested and analyzed for metal elements. The size distribution was determined from the data measured with the MOUDI. The MOUDI is an 8-stage cascade impactor with a flow rate of 30 L/min. The particle diameters cut-off by the MOUDI were 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32 and 0.18  $\mu\text{m}$ . Teflon filters were also used in the MOUDI for cases of low metal contamination and pressure-drop problem arose when these two sampling devices were used.

## 2.2 Chemical Analysis

**Table 1.** Blanks of quartz filter

Metals	Wave length (nm)	Content( $\mu\text{g g}^{-1}$ )
Al	396.152	0.228 $\pm$ 0.025
Ca	393.366	0.305 $\pm$ 0.027
Fe	238.204	0.193 $\pm$ 0.012
Ti	323.452	<0.019
Pb	220.353	0.013 $\pm$ 0.001
Zn	213.856	0.016 $\pm$ 0.002
Ni	221.647	0.043 $\pm$ 0.006
V	268.796	<0.018
Ba	455.403	0.009 $\pm$ 0.001
Mn	257.61	<0.001
Cr	205.552	<0.022
Cd	214.438	<0.011
Cu	224.7	0.016 $\pm$ 0.002
Sr	421.552	<0.014

Fourteen metal elements (Al, Ca, Fe, Ti, Pb, Zn, Ni, V, Ba, Mn, Cr, Cu, Cd, and Sr) were analyzed. Samples were first treated with pressurized digestion (Wang et al., 1995). Polytetrafluoroethylene (PTFE) containers were used in the experiments. All chemicals used were supplied by Merck (pro analysis grade) and high-purity water (resistance > 10 M $\Omega$ ) produced by reverse osmosis and demineralization, was used. All stock solutions of the elements of interest (2000  $\mu\text{g/mL}$ ) were prepared from Titrisol concentrates (Merck) by diluting to volume with de-ionized water. The sample was placed in a 25-mL PTFE container and a 5-mL mixture of HNO<sub>3</sub>/HClO<sub>4</sub> (3 + 7 V/V) was added. The sealed container was then transferred into a pressure bomb (supplied by Berghof) and heated on a heating block at 170 °C for 5 hours to facilitate complete dissolution. After the solution was cooled to room temperature, it was transferred into a 25-mL calibrated flask and diluted to volume with distilled water. The digested samples were then analyzed for metal elements by inductively coupled plasma/atomic emission spectrometry (ICP/AES, Jobin-Yvon JY38 Plus).

Unexposed filters were placed in PTFE

containers and the digestion procedures were followed. The background contamination was insignificant for the Teflon filter. Table 1 lists the metal element content in the blank quartz filter. The actual metal concentration was estimated by subtracting the measured metal concentration from the blank concentration. Spectral interferences due to the matrix effect and the background shift were studied qualitatively by examining the standard additions and calibration graphs. The calibration graphs were constructed by measuring multi-element standards prepared in the laboratory. The ranges of recovery efficiencies of the ten samples were between 94% and 105% and the reproducibility results were between 96% and 106% for all metals.

### 3. Results and Discussions

#### 3.1 Identifying Sources of Elements by Factor Analysis

Table 2 presents geometric mean concentrations and associated standard deviations of total suspended particulates (TSP) and metals. TSP concentrations in the air range from 41.9 to 262  $\mu\text{g m}^{-3}$  with a mean  $\pm$  S.D. value of 134  $\pm$  53.6  $\mu\text{g m}^{-3}$ . The level was between those measured in urban (166  $\mu\text{g m}^{-3}$ ) and suburban (71.1  $\mu\text{g m}^{-3}$ ) areas in, Taichung, Taiwan (Fang et al., 2000). Factor analysis of the data sets was performed by the statistical analysis system (SAS, 1997) to categorize these trace metals. The analysis started with the calculation of the eigenvalues of the matrix of correlation coefficients between the variables. The eigenvalues for four factors were larger than 1.0, were retained and the matrix with the loadings was then subjected to VARIMAX rotation.

Table 2 presents the results of factor analysis. The eigenvalues of these factors (before VARIMAX rotation) are 5.7, 2.9 and 1.2, and these factors explain 25.5, 22.5, and 21.0%, respectively,

**Table 2.** Geometric mean concentrations (in  $\text{ng m}^{-3}$ ), standard deviations (S.D.), VARIAMX rotated loadings and the communalities ( $h^2$ ) for the component loadings ( $n = 60$ )

Elements	Mean	S.D.	Rotated Loadings <sup>a</sup>			$h^2$
			Factor1	Factor2	Factor3	
TSP <sup>b</sup>	134	53.6	<b>0.756</b>	-0.153	-0.234	0.68
Al	500	350	<b>0.856</b>	0.041	-0.029	0.23
Fe	883	544	<b>0.830</b>	0.329	-0.203	0.62
Sr	3.3	1.6	<b>0.815</b>	-0.012	-0.211	0.85
Ca	1470	628	<b>0.714</b>	-0.325	-0.113	0.89
Ti	13.9	8.1	<b>0.636</b>	-0.213	-0.280	0.90
Mn	32.0	18.5	<b>0.717</b>	0.052	-0.237	0.90
Zn	156	92	-0.075	0.052	-0.078	0.68
Ni	32.4	25.2	0.263	<b>0.872</b>	-0.119	0.70
Cr	11.5	10.3	0.247	<b>0.768</b>	-0.200	0.76
V	7.8	3.3	0.365	<b>0.700</b>	-0.178	0.63
Cu	18.4	15.3	-0.159	-0.169	<b>0.889</b>	0.81
Pb	75.0	72	-0.331	0.069	<b>0.834</b>	0.74
Ba	15.2	11.1	0.007	0.428	0.421	0.92
Cd	0.50	0.45	0.086	-0.569	-0.231	0.38
Eigenvalue			5.7	2.9	1.2	
$V^c$			3.83	3.37	3.15	
% <sup>d</sup>			25.5	22.5	21.0	

a. Rotated loadings greater than 0.7 are in bold.

b. Unit for TSP is in  $\mu\text{g m}^{-3}$ .

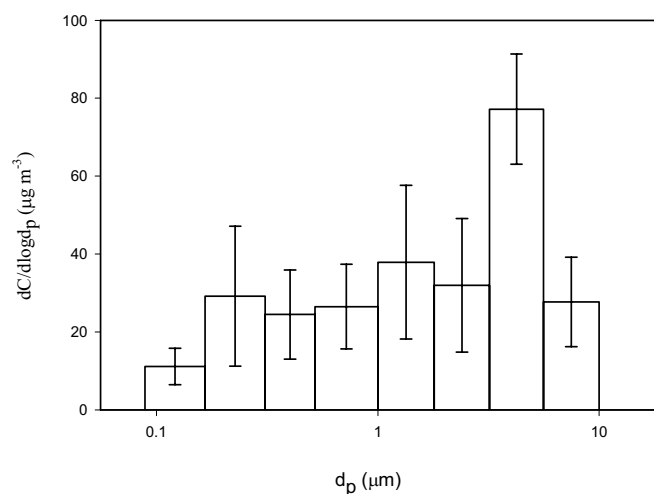
c. Variance explained by each factor.

d. The percentage of variance explained by each factor

of the variance in the data sets. For factor 1, the crustal elements Al, Fe, Sr, Ca, and Ti are highly correlated. Factor 1 can thus be identified as resuspended soil dust. TSP is correlated with factor 1 (loading = 0.756 > 0.7), suggesting that the resuspended soil dust is a significant source of TSP. The second factor has high loadings (larger than 0.7) for Ni, V, and Cr. Ni and V are the indicatory elements of heavy oil combustion (Rahn and Lowenthal, 1984, 1985). Factor 3 has high loadings for Cu and Pb. Inferences about factor 3, sources of Cu and Pb, cannot easily be drawn from current knowledge on industrial emissions.

### 3.2 Distributions of Total Particle Masses and Sizes of Metal Particles

Total particle mass is tri-modally distributed in size with peaks at 0.18-0.32  $\mu\text{m}$ , 1.0-1.8  $\mu\text{m}$  and 1.8-3.2  $\mu\text{m}$  (Fig. 2). The particles are divided into fine (< 1  $\mu\text{m}$ ) and coarse modes (> 1  $\mu\text{m}$ ) in this study. On average, coarse particles constitute 66% of the mass of the samples, indicating that coarse particles dominate the total particle mass. Figure 3 presents the general characteristics of the size spectra for trace metals. Most spectra are bi-modal distributions. For the crustal elements, Al, Ca, Fe, and Ti, significant peaks occur at  $d_p$  larger than 1.0  $\mu\text{m}$ . The fraction of the coarse aerosol mode, important in determining the fluxes for the deposition velocity of the coarse particles, is greater than that of the fine aerosol modes (Quinn, 1994). On average, coarse fraction is 53% for Al, 64% for Ca, 62% for Fe and 44% for Ti. Notably, the crustal



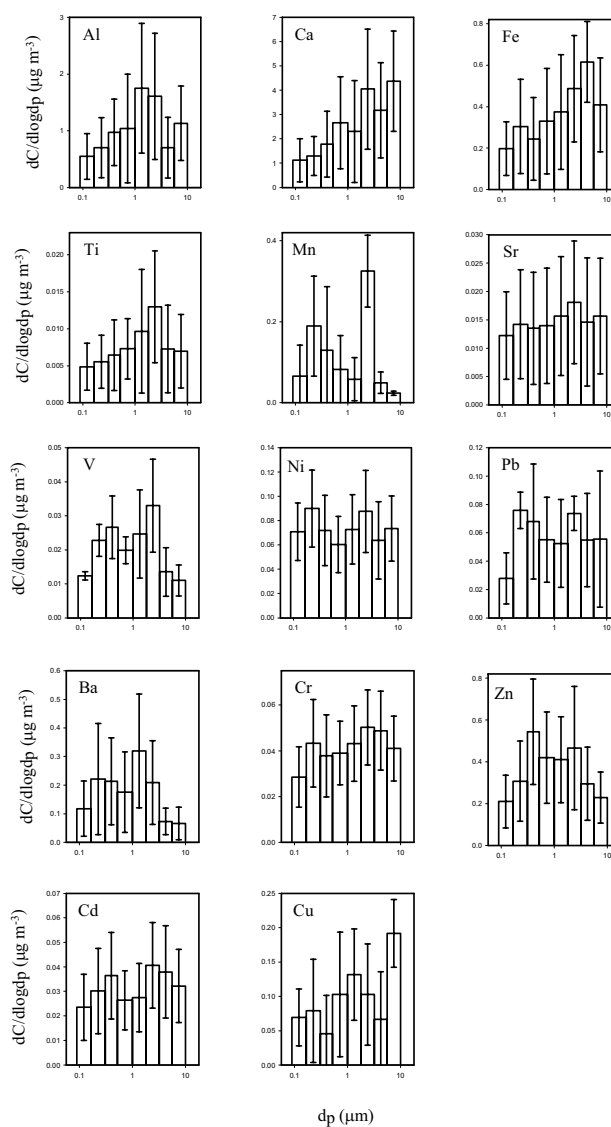
**Figure 2.** Size distribution of particulate matter.

elements Ti and Sr also represent substantial mass fractions in fine mode particles.

In contrast to the elements associated with soil, non-crustal elements are closely associated with fine particles, since their modal diameters in sample size distributions are generally less than 1.0  $\mu\text{m}$ . However, some significant fractions of their masses are associated with the coarse particle mode. For example, the mass fractions associated with mass median diameters (MMDs) of larger than 1.0  $\mu\text{m}$  are Zn, 47%; Ni, 46%; Pb, 55% and V, 54%. Notably, individual spectra of size distributions and mass median diameters of these elements often differ significantly in the same sample, suggesting that emitted particles remain suspended as separate particle populations, and that a homogenizing process, such as cloud processing and coagulation, may be less important in determining their size distribution (Quinn and Ondov, 1998).

### 3.3 Dry Deposition

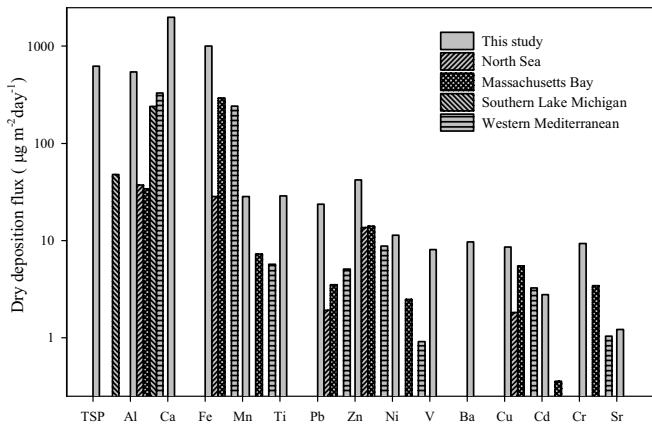
Figure 4 shows the dry deposition fluxes of total particle mass and trace metals. The measured total particle mass dry deposition fluxes vary between 123  $\text{mg m}^{-2}\text{day}^{-1}$  and 2840  $\text{mg m}^{-2}\text{day}^{-1}$  and averages 622  $\text{mg m}^{-2}\text{day}^{-1}$ . The average dry deposition fluxes of the crustal elements are: Al 541  $\mu\text{g m}^{-2}\text{day}^{-1}$ , Ca 1989  $\mu\text{g m}^{-2}\text{day}^{-1}$  and Fe 999  $\mu\text{g m}^{-2}\text{day}^{-1}$ .



**Figure 3.** Average size distribution spectrum of metal elements.

$\text{m}^{-2}\text{day}^{-1}$ . These values are one to two orders of magnitude higher than those of the non-crustal elements, implying that the crustal metals are the major elements of the dry deposition. The dry deposition fluxes measured in this study are higher than those reported for some other regions (North Sea, Massachusetts Bay, Southern Lake Michigan, Western Mediterranean) (Fig. 4).

The dry deposition fluxes of total particle mass and trace metals are calculated by a multistep dry deposition model (Holsen et al., 1993) combined with Sehmel-Hodgson's deposition velocity model (Sehmel and Hodgson, 1978). The multistep dry deposition model divides the particle sizes into intervals, and sums the calculated flux for each

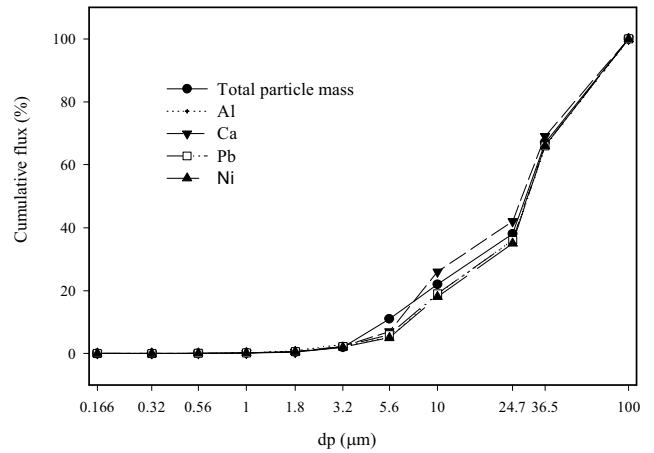


**Figure 4.** The measured dry deposition fluxes of total particle mass and trace metals. Unit of total particle mass is in  $\text{mg m}^{-2}\text{day}^{-1}$ . (Rojas et al., 1993; Golomb et al., 1997; Sofuoglu et al., 1998; Chester et al., 1999)

interval to obtain the modeled flux:

$$\text{Modeled Flux} = \sum_{i=1}^i C_i V_i \quad (1)$$

where  $C_i$  is the concentration for the  $i$ th size interval and  $V_i$  is the dry deposition velocity for the  $i$ th interval.  $V_i$  is calculated using Sehmel-Hodgson's dry deposition velocity model. In this study, a particle density of  $1.0 \text{ g/cm}^3$  and a plate roughness of  $0.001 \text{ cm}$  are assumed. The model also requires wind speed and ambient temperature as inputs. Particles with aerodynamic diameter larger than  $10 \text{ }\mu\text{m}$  were not measured in this study. Although these particles do not dominate the size distribution, their contributions to dry deposition are substantial for their high deposition velocity (Holsen and Noll, 1992; Holsen et al., 1993). Thus, the concentrations of particulate and metal elements larger than  $10 \text{ }\mu\text{m}$  were added according to the method used in Sheu et al.'s study in Taiwan, to provide a more precise explanation of dry deposition results. Figure 5 shows the modeled cumulative flux distributions of total particle mass, two typical crustal metals (Al, Ca) and two anthropogenic metals (Pb, Ni). Fluxes obtained by the multistep flux calculation method are summed



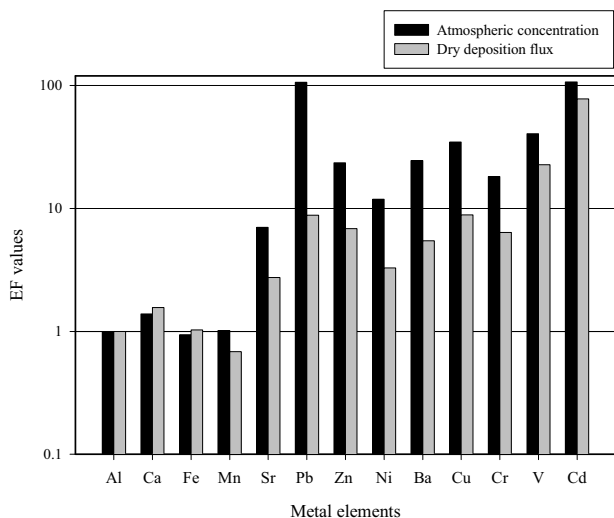
**Figure 5.** Average cumulative fluxes calculated from the multistep model for total particle mass, Al, Ca, Pb and Ni.

in these size ranges and plotted as cumulative percentages. The cumulative flux distributions show that fine particles (diameter  $\leq 1.0 \text{ }\mu\text{m}$ ) contributed only a small fraction of the dry deposition flux. The average percentage of modeled fluxes due to fine particles are only 0.2%, 0.2%, 0.18%, 0.22% and 0.19% for total particle mass, Al, Ca, Pb and Ni, respectively. These results imply that coarse particles dominate dry deposition.

### 3.4 Concentration Distribution of Metals

Concentrations of metals in air may vary over large ranges, but by normalizing them to their crustal concentrations, the concentrations of metal in an aerosol can be compared and assessed (Chester et al., 2000). This study uses an enrichment factor (EF) that incorporates information pertaining to a reference element, indicative of a specific source, to determine the sources of the elements (crustal and non-crustal). Al is normally used as the crustal indicator element. The  $EF_{\text{crust}}$  value is calculated from the following equation:

$$EF_{\text{crust}} = (C_{xp} / C_{Alp}) / (C_{xs} / C_{Als}) \quad (2)$$



**Figure 6.** Average enrichment factor values of trace metals for atmospheric concentration and dry deposition flux.

where  $C_{xp}$  and  $C_{Alp}$  are the concentrations of the subject element x and Al in aerosols, respectively.  $C_{xs}$  and  $C_{Als}$  are their contents in the soil. By convention, an EF value of less than 10 is taken to indicate that a crustal source of a trace metal in aerosol is significant. (The metal originates from the soil.) In contrast, an EF value  $> 10$  is considered to indicate that a significant proportion of an element has a non-crustal source.

Figure 6 presents the average EF values for atmospheric concentration and dry deposition flux. On the basis of these data, the metal elements can be divided into three groups. Group 1 includes Al, Ca, Fe, Mn and Sr. Both atmospheric concentrations and dry deposition fluxes in Group 1 are soil-originated ( $EF_{crust}$  value  $< 10$ ). Group 2 includes Pb, Zn, Ni, Ba, Cu and Cr. The parent aerosol is not soil-originated, whereas the dry deposition is soil-originated. Group 3 includes V and Cd. Neither the parent aerosol nor the dry deposit of these two metals originates from the soil is soil-originated. For most metals, the  $EF_{crust}$  values are lower in the dry deposit than in the parent aerosol. The discrepancy may be associated with fractionation during dry deposition. A crustal source normally generates more coarse particles than fine ones. Coarse particles undergo more effective dry

**Table 3.** Fractionation factors for the dry deposition

Trace metals	Fractionation Factor
Al	1.00
Ca	0.88
Fe	0.91
Mn	1.49
Pb	12.1
Zn	3.43
Ni	3.61
V	1.79
Ba	4.50
Cu	3.91
Cd	1.37
Cr	2.84
Sr	2.56

deposition, resulting in removal of the crustal-originated portion of a metal or of its fractionation from the bulk air mass (parent aerosol). Thus, a dry deposit includes more crustal-originated metal than the parent aerosol, leading to differences in their respective  $EF_{crust}$  values. Dry deposition processes, which remove metals from the air, are a function of particle sizes. A fractionation factor (FF) has been used to measure the extent of fractionation, and is determined by the following equation (Chester et al., 1999):

$$FF = EF_p / EF_d \tag{3}$$

where  $EF_p$  is the enrichment factor of a metal in particle and  $EF_d$  is the enrichment factor in a dry deposit. Element Pb has  $FF = 12.1$ , the highest of all, indicating that Pb is most strongly fractionated between the parent aerosol and dry deposition (Table 3).

#### 4. Conclusions

Aerosol samples collected in the coastal region of western Taiwan during the dry monsoon season were examined. Factor analysis shows that the



crustal elements, Al, Ca, Fe and Ti, are highly correlated and can be identified as resuspended soil dust, explaining 27.3% of the variance. The other factors are difficult to relate to specific emission sources of the samples revealing no discernible patterns. Coarse mode particles (defined by a cut-off point of 1.0  $\mu\text{m}$ ), dominate the total particle mass, accounting for 66% (on average) of it. For Al, Ca, Fe and Ti crustal elements exhibit significant peaks at diameters larger than 1.0  $\mu\text{m}$ . The average mass fractions of coarse particles are 53% for Al, 64% for Ca, 62% for Fe and 44% for Ti. In contrast to the elements associated with soil, non-crustal elements are dominated by fine particles with modal diameters of less than 1.0  $\mu\text{m}$  in average spectra. The average dry deposition fluxes of the crustal elements are Al 541  $\mu\text{g m}^{-2}\text{day}^{-1}$ , Ca 1989  $\mu\text{g m}^{-2}\text{day}^{-1}$  and Fe 999  $\mu\text{g m}^{-2}\text{day}^{-1}$ . These crustal elements are the major components of the dry deposition.

Both atmospheric concentrations and dry deposition fluxes of Al, Ca, Fe, Mn and Sr are soil originated ( $EF_{\text{crust}}$  value < 10). For Pb, Zn, Ni, Ba, Cu and Cr, the parent aerosol does not originate from the soil but the dry deposit does soil. Neither the parent aerosol nor the dry deposition of V and Cd originate from the soil. For most metals investigated, the  $EF_{\text{crust}}$  values are lower in a dry deposit than in a parent aerosol. The crustal parts of a metal are fractionated from the parent aerosol by a dry deposition sequence. The fraction of the mass of an element with a non-crustal source is relatively unimportant in the dry deposition. Among the elements investigated, Pb has the highest fractionation factor (12.1).

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