

Temporal and spatial distributions of dust and its deposition to the China Sea

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(Manuscript received 27 March 1995; in final form 28 October 1996)

ABSTRACT

Atmospheric concentrations of aluminum, an indicator of dust substances, have been determined in a set of high-volume aerosol particle samples collected at different locations over continental China and over the China Sea. High concentrations of dust were observed in northern continental China, and at certain locations such as Beijing dust may include an anthropogenic fraction. The mass particle-size distributions of dust varied depending on its distance from source regions, with the mass median diameter for Al of $\sim 1.6\text{--}5.9\text{ }\mu\text{m}$ at Beijing in northern China and $\sim 1.9\text{ }\mu\text{m}$ over off-shore areas of the East China Sea. Model-predicted mean dry deposition velocities of dust particles are from $1.4\text{ to }4.8\text{ cm s}^{-1}$ over northern continental China and from $1.4\text{ to }2.1\text{ cm s}^{-1}$ over the China Sea. Atmospheric deposition models have been applied to estimate the atmospheric fluxes and deposition of dust at different locations. The estimated atmospheric flux of dust at Xi'an of the Loess Plateau is $25\text{ (}4.9\text{ to }44\text{) g m}^{-2}\text{ mo}^{-1}$ which is the highest among the regions we studied. The estimated present-day dust flux is comparable to the late quaternary records of eolian dust accumulation at this site. The total atmospheric deposition of dust to the China Sea is 67 Tg yr^{-1} , accounting for 14% of the total atmospheric deposition of dust to the entire North Pacific. With such a high deposition rate, Asian dust may play an important rôle in biogeochemical cycles of trace substances in the Asia/North Pacific region.

1. Introduction

Dust derived from Asia has drawn increasing attention due to its significant links to the biogeochemical cycles of trace elements. Recent studies have suggested that in certain oceanic regions biological productivity may be limited by the

supply of eolian iron (Duce, 1986; Martin and Gordon, 1988). The mid-latitudes (25° to 40°N) are characterized by high dust concentrations, especially in spring (Uematsu et al., 1983), and this material is a significant source for deep-sea sediments (Duce et al., 1980; Prospero, 1981; Blank et al., 1985; Leinen et al., 1994). Air mass back trajectory analyses verify that the Asian desert dust is carried out over the ocean by the mid-latitude westerlies (Merrill et al., 1989 and 1994).

Investigations of Asian dust are also focusing on the western North Pacific region and Asian

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coastal waters (Okada et al., 1987; Iwasaka et al., 1988; Hayasaka et al., 1990; Chung, 1992; Gao et al., 1992a and 1992b; Hashimoto et al., 1994). High dust concentrations of up to $\sim 1110 \mu\text{g m}^{-3}$ have been observed over the Korean peninsula with dust sources in Northern China and Mongolia (Chung, 1992). The estimated dust flux to the western North Pacific (25° to 50°N , W of 150°E) is about 300 Tg per year (Prospero et al., 1989). The estimated atmospheric deposition of dust to the Yellow Sea is about 9 to $76 \text{ g m}^{-2} \text{ yr}^{-1}$ (4 to 33 Tg yr^{-1}), and this accounts for 20 – 70% of the total input of mineral material to the Yellow Sea (Gao et al., 1992a). These studies suggest that atmospheric dust plays a significant biogeochemical role in the North Pacific oceanic and Asian coastal environments.

Recent studies indicate that dust is 1 of 4 key aerosol species that may cool the planet by absorbing and reflecting solar radiation (Penner et al., 1993). Although compared with other aerosol species the climate forcing by dust particles may not be significant on the global scale due to their relatively short atmospheric residence times and geographical distributions of sources, effects of dust on regional climate are not necessarily negligible. Satellite images from East Asia obtained in spring 1993 indicate that the planetary albedo increased during the dust storms, demonstrating direct connections between dust concentrations and the solar radiation flux to the region (Zhou et al., 1994).

There is relatively little information on Asian dust near its source regions, however. The spatial distribution of dust in the lower troposphere over eastern Asia, especially for the regions close to its source, is not well known due to the limited results of simultaneous ground-based measurements from different locations (Hashimoto et al., 1994). In addition, the atmospheric flux of dust to the entire basin of the Chinese Sea has not been estimated, although one would expect it to be higher than the fluxes to the remote North Pacific. Information regarding the above concerns is essential for a better understanding of the role of dust in the biogeochemical cycles of trace substances, and this information will be helpful to quantitatively interpret the dust-climate interactions in the Asia/western North Pacific region.

From December 1990 to June 1993, high-volume bulk aerosol particle samples were col-

lected at a ground-based station at Beijing. Additional ground-based sampling was conducted in the spring 1992 at Xi'an in the Loess Plateau of the northern China and at two coastal sites in the China Sea, while ship-board measurements were carried in the off-shore region in the East China Sea. In this paper we present results concerning the spatial and temporal distributions of atmospheric dust over east Asia. We also characterize the particle-size distributions of dust based on high-volume cascade impactor samples. We estimate the total deposition and fluxes of atmospheric dust to the China Sea using dry and wet deposition models. Comparisons of our results with those from other studies conducted in the same region are also discussed. We also explored the influence of anthropogenic emissions on the dust concentrations at Beijing, a typical urban area.

2. Sampling and chemical analyses

2.1. Sampling sites

A total of 306 bulk aerosol particle samples, 14 sets of high-volume cascade impactor samples, and 1 set of low-volume cascade impactor sample were collected at 4 ground-based sites and during a cruise (Fig. 1). The first site is at Xi'an in the

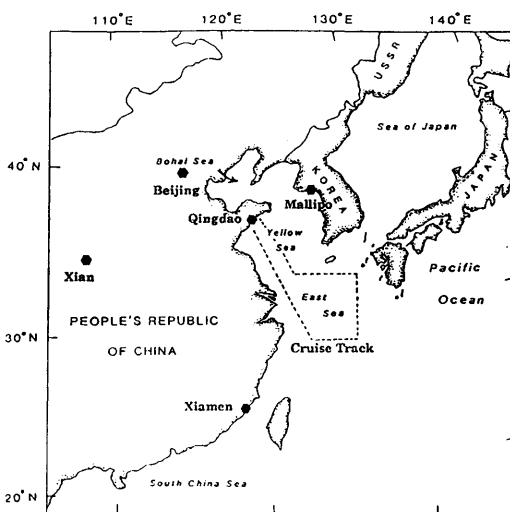


Fig. 1. Map of the sampling locations, including the cruise track.

Loess Plateau, located ~20 km away from the downtown Xi'an; there are no significant industrial activities in the surrounding area. The second site is at Beijing, where the sampler was placed on the roof of a building housing the National Center for Marine Environmental Forecasts in a western suburb. Near this site, there were some construction activities going during sampling and certainly this site is not far from industrial sectors. Thus anthropogenic emissions could possibly affect the atmospheric concentrations of certain trace elements at this site, and the situation at this site is quite different from other sites. The third site was at Qingdao located on the western side of the Yellow Sea. Sampling at this site was conducted at a hydrographic station on Little Wheat Island near the eastern tip of the Qingdao Peninsula. The sampler at this site was ~25 m above sea level. The fourth site was at Xiamen, located on the coast of the South China Sea. Aerosol samples from this site were collected on the roof of a building about 10 m high and about 10 m from the water. The cruise used for sampling was within the East China Sea, primarily the East Sea (SOA Xiangyanghuong Vessel no. 9). The sampling times ranged from ~12 to 24 h at all sites. Detailed sampling information is summarized in Table 1.

2.2. Sampling facilities

High-volume bulk aerosol particle samplers (flow rate of ~1.1 m³ min⁻¹) were used at all sites except during the cruise. Modified Sierra-type high-volume cascade impactors (~1.1 m³ min⁻¹) (Sierra Instruments, Inc., Carmel Valley, CA) were

used at Beijing and Xiamen to separate aerosol particles by size. The high-volume cascade impactors were composed of seven stages (0, 1, 2, 3, 4, 5, B) with corresponding particle cut-off diameters of 17.3 μm, 9.3 μm, 3.6 μm, 2.1 μm, 1.0 μm, 0.5 μm and <0.5 μm (equivalent aerodynamic diameter), respectively. During the cruise, a low-volume bulk aerosol particle sampler (Model KB-120, Laoshan Institute of Electronic Equipment, Qingdao, People's Republic of China) was used, with a flow rate of 0.08 m³ min⁻¹. An Anderson 9-stage low-volume cascade impactor with a flow rate of 0.03 m³ min⁻¹ was also used during the cruise (corresponding aerosol particle-sizes: 11, 7, 4.7, 3.3, 2.1, 1.1, 0.65, 0.43, and <0.43 μm, respectively) (Takadachi Rika Kabushiki Gaisha, Japan). We used Whatman 41® filters (20 × 25 cm) and slotted Whatman 41® cascade impactor filters as the high-volume sampling media. Zefluor® membrane filters (1 μm pore size) were used with both the low-volume bulk aerosol sampler and the low-volume cascade impactor.

2.3. Instrumental neutron activation analyses (INAA)

INAA was used to analyze aerosol samples for the atmospheric concentrations of aluminum (Al), antimony (Sb), scandium (Sc), and selenium (Se) using the 2-MW research reactor and the facilities of the Rhode Island Nuclear Science Center. 2 procedures were used: (a) a short irradiation procedure for Al. For this procedure a 2.2 cm diameter sample filter was irradiated for 60–90 s at a ther-

Table 1. Atmospheric sampling information for different locations

Site	Sampling dates	Location	No. of samples ^a	Environmental feature
Xian	3/17–5/28 1992	38°N, 105°E	18 (H–V)	Loess Plateau,
Beijing	12/7/90– 6/92	40°N, 116°E	249 (H–V) 9 (CI)	Urban, industrial
Qingdao	2/13–6/11 1992	36°N, 120°E	15 (H–V)	Coastal, rural
Xiamen	1/07–6/13 1992	24°N, 118°E	18 (H–V) 5 (CI)	Coastal, rural
East China Sea	4/24–4/29 1992	28°–32°N, 122°–130°E	6 (L–V) 1 (CI)	Off Coast,

^a H–V and L–V: bulk high-volume and low-volume aerosol particle samples, respectively; CI: cascade impactor samples.

mal neutron flux of $4 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$. After cooling for 2–4 min, the sample was counted for 500 s to obtain the gamma-ray spectra for the elements of interest; (b) a long irradiation procedure for Sb, Sc, and Se. For this procedure one quarter of a sample filter was pelleted, sealed, and packaged into a plastic delivery tube and irradiated for 24 h followed by three-week decay and counting for 10–11 h. Counting of all these samples was conducted with the use of Ge(Li) gamma-ray detectors (resolution of 2.5 keV for the 1332-keV gamma ray of ^{60}Co) interfaced to a VAX-workstation 3100 (Model 38). The concentrations of these trace elements were obtained through the analyses of the γ -ray spectra using Genie Gamma Spectroscopy software (Canberra Industries, Inc., Meriden, CT) and proprietary programs. Flux monitors and internal standards were processed at the same time to ensure the precision of the results. The uncertainties associated with the trace element concentrations are ~ 10 –20%, primarily due to uncertainties in the air volume measurements.

3. Results and discussion

In previous dust studies, Al has been generally considered as a representative for mineral aerosol of crustal origin. However, anthropogenic sources may also contribute atmospheric Al at certain locations. Thus Al data at certain sites cannot be simply equated “mineral aerosol” because it possibly contains an anthropogenic fraction. At present, it is difficult to separate mineral aerosol and anthropogenic dust. Based on this consideration, we chose to use “dust” as a general term instead of mineral aerosol in the following discussion. Since most of our discussion is based on samples collected in the spring, we assume that crustal Al is the dominant fraction of atmospheric Al at non-urban areas. This is very likely close to reality in this season, and this will allow us to simplify the discussion and to compare our results with others.

3.1. Spatial distribution of dust

Previous meteorological studies indicate that the major source region for mineral dust over East Asia is the Loess Plateau located in north-central China (Merrill et al., 1989; Hayasaka et al., 1990;

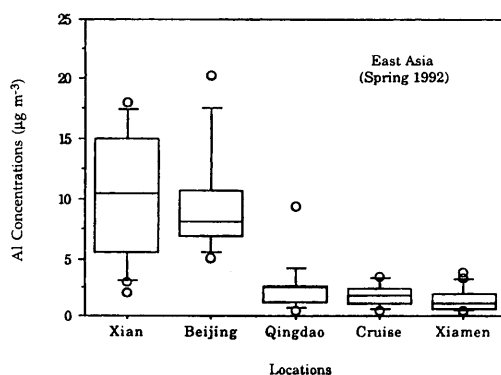


Fig. 2. Geographical distributions of atmospheric Al over Northern China and the China Sea described in a box plot. In this plot, each box is composed of five horizontal lines that display the 10th, 25th, 50th, 75th and 90th percentiles of the Al concentrations at each site. The individual values above the 90th percentile and below the 10th percentile are plotted by open circles.

Gao et al., 1992b). Thus the dust concentrations over the Loess Plateau were expected to be higher than other sampling sites. Fig. 2 summarizes the results of atmospheric Al observed at 5 sites in the spring 1992. The highest mean concentration of atmospheric aluminum during the spring of 1992 was observed at Xi'an (Mean: $10 \pm 5.3 \mu\text{g m}^{-3}$). This mean Al concentration is comparable to the result obtained by Zhang et al. (1993), who found the mean Al concentration was $15 \mu\text{g m}^{-3}$ at Xi'an during the period from December 1990 to May 1991. Since Xi'an is located within the Loess Plateau region, a dust concentration of $140 \pm 80 \mu\text{g m}^{-3}$ may be representative for atmospheric dust in the Loess Plateau region during the spring (Al accounts for $\sim 7\%$ of the total mass of loess, Liu et al., 1985), although dust concentrations are highly variable between dust storm periods and non-dust storm periods. The atmospheric Al concentrations decreased toward the east: $9.6 \pm 4.7 \mu\text{g m}^{-3}$ at Beijing, $2.5 \pm 2.2 \mu\text{g m}^{-3}$ at Qingdao, and $1.9 \pm 1.1 \mu\text{g m}^{-3}$ over the East Sea. The mean atmospheric Al concentration at Beijing is comparable to that at Xi'an although Beijing is several thousand kilometers away from the Loess Plateau. Thus there must be additional sources contributing the atmospheric Al at Beijing, and we will address this question in the following section. Significant differences in the atmospheric Al concentrations exist between the inland regions

Table 2. Results of the Fisher's protected least significant difference for atmospheric Al concentrations between sampling sites

Sites	Mean difference ($\mu\text{g m}^{-3}$)	Critical difference ($\mu\text{g m}^{-3}$) ^a	P-value
Xian, Beijing	0.69	4.5	0.75
Xian, Qingdao	7.5	4.1	0.001
Xian, E. China Sea	8.4	4.6	0.001
Xian, Xiamen	8.9	4.0	0.0001
Qingdao, Xiamen	1.1	1.2	0.08
Qingdao, E. China Sea	0.69	2.0	0.47

^a Significance level: 5%.

and the coastal regions (Table 2). The lower concentrations of dust in the coastal regions are attributed not only to greater distance from the dust source but also to the mixing of continental air with clean marine air by the land-breeze/sea-breeze circulation.

The observed mean concentration of atmospheric Al at Xiamen is $1.5 \pm 1.1 \mu\text{g m}^{-3}$, ~40% lower than that at Qingdao. Most areas of northern China and the East China Sea are on the primary pathway of dust transport. Thus the atmospheric Al concentrations in those areas are strongly affected by the dust activities in the spring, which is the high-dust season. Xiamen, on the other hand, is far from the Loess Plateau and from the dust transport path, and the influence of desert dust on this location is considerably less than at locations in northern China. This geographical difference, i.e., the latitudinal variation in Al concentrations, coincides with results from other western North Pacific regions, where higher concentrations of dust were found in the mid-latitudes rather than in the lower-latitudes (Tsunogai et al., 1985).

The mean Al concentration is $1.9 \pm 1.1 \mu\text{g m}^{-3}$ over the East Sea, which is similar to the concentrations observed over other regional seas of eastern Asia. For example, Tsunogai et al. (1985) found that the mean spring concentrations of Al were $1.3 \mu\text{g m}^{-3}$ at Wajima and $1.7 \mu\text{g m}^{-3}$ at Izumo, which are both located along the west coast of Japan. The annual mean Al concentration observed at Seoul was $1.4 \mu\text{g m}^{-3}$ (Hashimoto et al., 1990).

3.2. Size distributions of dust particles

The concentrations of trace species present in atmospheric aerosols vary strongly with particle-

size. This particle-size dependence varies for different chemical species, reflecting sources, production and removal mechanisms. Soil-derived dust particles are produced through weathering and mechanical wind deflation which primarily generates particles in the coarse mode (Gillette and Walker, 1977; Gillette and Dobrowolski, 1993). This pattern is clearly indicated by results of cascade impactor samples collected at Beijing and Xiamen.

Figs. 3a–c describe the concentration variations of atmospheric Al as a function of particle-size for samples collected at Beijing during the spring and summer of 1992. The concentrations of atmospheric Al in these samples are dominated by aerosol particles between 3.6 and 9.3 μm diameter, which account for 35–63% of the total Al mass in these samples. However, the samples collected from 24 April to 29 April (Fig. 3a) show elevated Al concentrations for the backup filters (<0.5 μm diameter), and this is clearly different from samples collected from April 30 to 8 May (Fig. 3b) and in August (Fig. 3c). The Al concentrations associated with particles in the size range between 3.6 and 9.3 μm diameter for the first four sets of samples account for only 35–45% of the total Al.

One possible reason for these differences could be due to particle bounce in the high-volume cascade impactor with the size distributions being shifted toward smaller particles (Burton et al., 1973; Dzubay et al., 1976; Buat-Menard et al., 1983). Particle bouncing is believed to result from the inability of dry solid particles to adhere to dry impaction surfaces; the extent of particle bouncing depends on the condition of the impactor surface, the nature of aerosol particles, and the ambient relative humidity (Stein et al., 1994; Vasconcelos

et al., 1994). In this case, the elevated concentrations of Al on the backup filters for samples collected during 24–29 April 1992 could be attributed to a low relative humidity (average: 26%) as indicated in Fig. 3a. The sample associated with the highest Al concentration on the backup filter was collected on 24 April, when the mean relative humidity was only 12%. On the other hand during the next sampling period (30 April–8 May), no elevated Al concentrations were found on the backup filters when the mean relative humidity was 48% (Fig. 3b). This is also seen in samples collected in August when the relative humidity was 80% (Fig. 3c).

In addition to particle bounce, aerosol particles in the accumulation mode are produced primarily by high-temperature processes (Brimblecombe, 1986). Beijing is characterized by tremendous coal

combustion due to the rapid industrial development (Kato and Akimoto, 1992). Thus anthropogenic emissions at Beijing could also contribute to the high Al concentrations on the backup filters of the impactors.

Fig. 3d describes the concentration variation of atmospheric Al as a function of particle-size for samples collected at Xiamen in the spring of 1992. The first peak in Al concentration is associated with aerosol particles of $\sim 3.6 \mu\text{m}$ diameter. An elevated Al concentration for particles in the accumulation mode ($\sim 0.5 \mu\text{m}$ diameter) also appeared for two sets of samples. A low relative humidity occurred during the sampling period for these two samples relative to that for the other two samples (Fig. 3d). Since the average relative humidity during the sampling at Xiamen was $\sim 69\%$, the bounce effect is unlikely; high Al concentrations

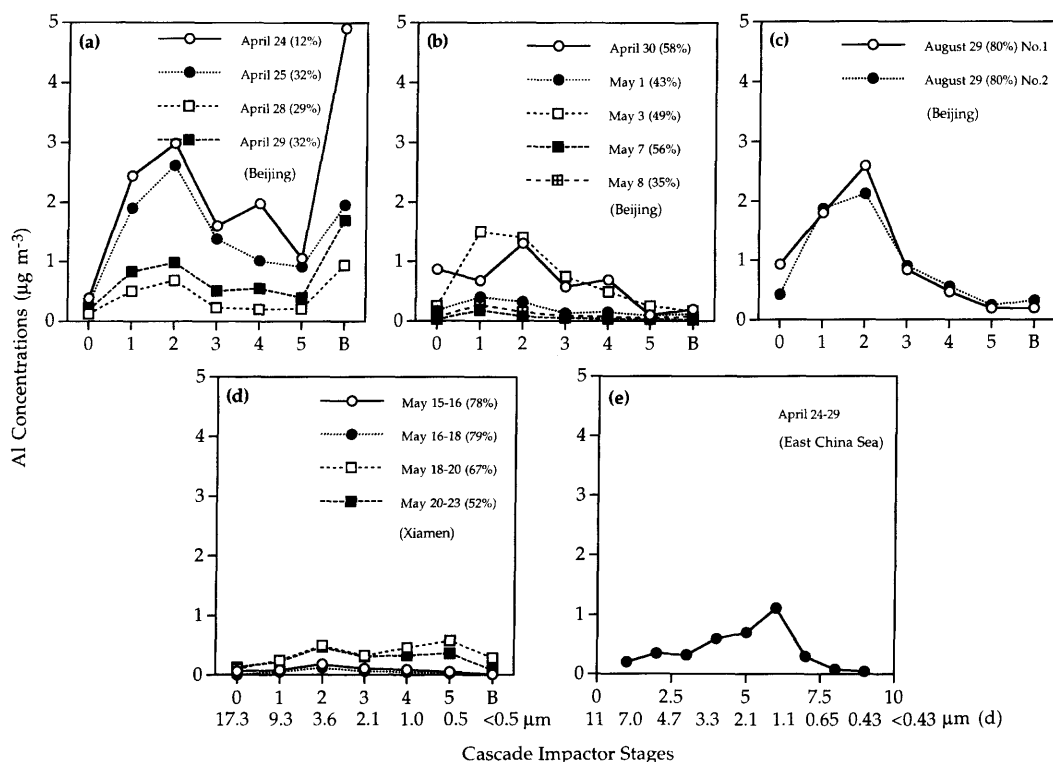


Fig. 3. Concentration variation of atmospheric Al as a function of particle size for high-volume aerosol particle samples collected (a) at Beijing during 24–29 April, 1992; (b) at Beijing during 30 April–8 May, 1992; (c) at Beijing in August, 1992; (d) at Xiamen in May, 1992; (e) over the East China Sea in April, 1992. The % given on the right of the dates are the mean relative humidity during each sampling period.

associated with submicrometer particles may indicate occasional anthropogenic influence, but this needs further investigation.

Fig. 3e shows the results from one cascade impactor sample collected during the East China Sea cruise. This sample should be generally representative of the concentration/particle-size distributions of dust in the marine atmosphere in off-shore regions. The highest Al concentration is associated with aerosol particles $\sim 1.1\text{--}3.3\text{ }\mu\text{m}$ diameter, somewhat smaller than that found in the continental regions. The amount of Al on aerosol particles between 1.1 and $3.3\text{ }\mu\text{m}$ diameter accounts for $\sim 65\%$ of the total Al, indicating that the particle size distributions of dust has shifted toward smaller sizes as the dust particles are transported further away from the continent. This suggests that dust particles of $1.1\text{--}3.3\text{ }\mu\text{m}$ diameter have a longer atmospheric residence time than larger particles, as expected.

3.3. Temporal variation of atmospheric Al at Beijing: additional sources

Unexpectedly high concentrations of Al were associated with both bulk aerosol particles and submicrometer particles at Beijing as indicated in previous sections. It is likely that anthropogenic emissions, especially coal burning, may have played an important role at this site. To address this question, we analyzed 249 bulk aerosol samples collected twice per week at Beijing between December 1990 and June 1993 to investigate the relationships among Al, Sc, Sb, and Se, especially their seasonal patterns. Similar to Al, atmospheric Sc has been considered as a crustal element; atmospheric Sb is almost exclusively from coal burning and atmospheric Se also is mainly dominated also by coal burning.

Fig. 4 shows the temporal variations of atmospheric Al (a), Sc (b), Sb (c), and Se (d) observed at Beijing. Surprisingly, the concentrations of atmospheric Al and Sc, which are commonly considered as crustal elements, co-vary with those of Sb and Se, with their temporal patterns being almost in phase. The maximum concentrations of these four elements occurred in the winter months (November–February) when coal is used most extensively for space heating in addition to its regular consumption for industrial activities. The corresponding concentration residuals for Al, Sc,

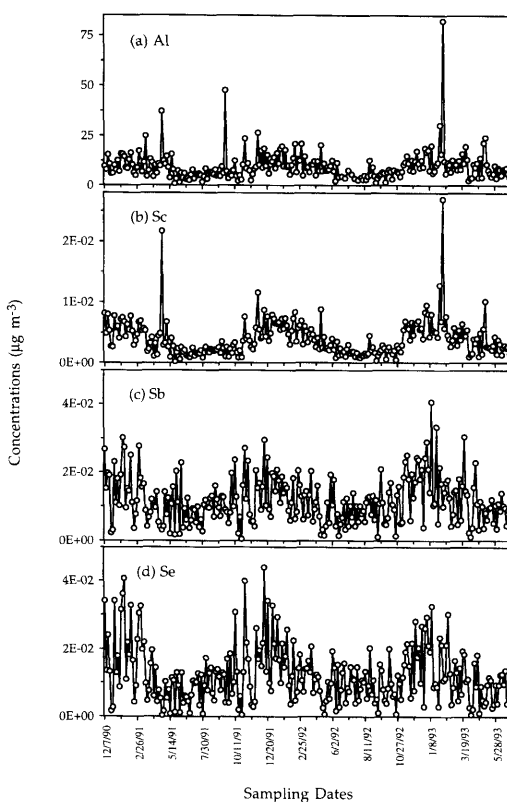


Fig. 4. Temporal patterns of atmospheric Al (a), Sc (b), Sb (c) and Se (d) at Beijing based on high-volume bulk aerosol particle samples collected from December 1990 to June 1993.

Sb, and Sc also show a similar pattern (Fig. 5). This pattern is consistent with recent results by Ando et al. (1996) who found that the maximum concentration of suspended particulate matter at Beijing was observed in the winter season. Thus coal fly ash is a probable source for atmospheric Al at this site, especially in the cold seasons. This source may well generate submicrometer particles which is as what we observed in cascade impactor samples. Although it is difficult at present to quantitatively separate Al of anthropogenic origin from that of desert origin, anthropogenic emissions, especially coal burning, undoubtedly perturb the natural cycle of atmospheric Al at this site.

We should mention that the dust storms are still an important driving force, and they cause springtime Al concentrations to be much higher than the monthly average at Beijing, although the

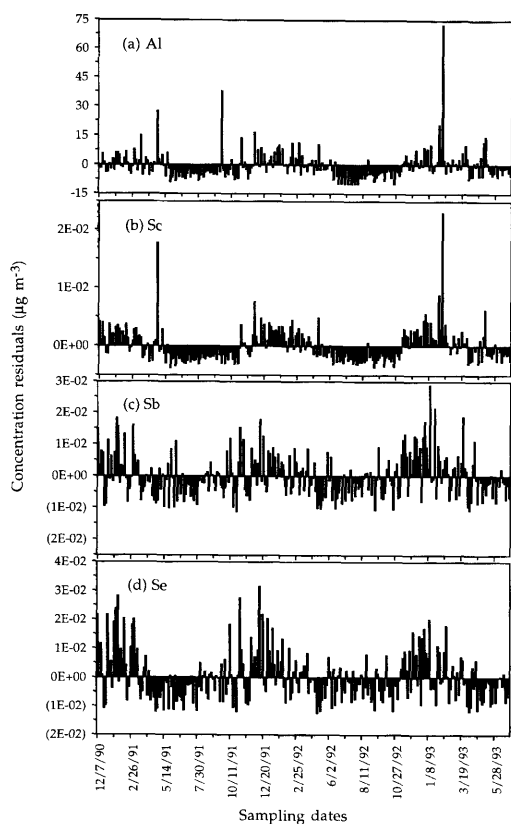


Fig. 5. Temporal patterns of atmospheric concentrations of Al (a), Sc (b), Sb (c), and Se (d) relative to their mean values (concentration residuals) based on samples collected at Beijing from December 1990 to June 1993.

dust storms are sporadic. The atmospheric Al concentration peaked at $37 \mu\text{g m}^{-3}$ on 30 April 1991 (monthly mean: $12.3 \mu\text{g m}^{-3}$), when a heavy dust storm occurred during that day; the corresponding meteorological analyses indicate that a cold frontal system passed Beijing at 0500 on 30 April 1991, accompanied by strong winds and a dust storm. Meteorological charts also indicate that there was a strong high pressure center located to the east of Lake Baikal and a low pressure system was centered over northeast China.

3.4. Modeling atmospheric deposition of dust

Atmospheric aerosols are removed by dry and wet deposition. Although direct measurements of deposition would appear to be the best approach,

problems with sampling, contamination, and the natural variation of the concentration of trace substances in the boundary layer often make direct deposition measurements difficult. So far there are no widely accepted methods to directly measure the total aerosol deposition to a water surface. Removal of aerosol particles by dry and wet processes is often estimated using simple deposition models.

3.4.1. Model for estimating atmospheric dry deposition. The dry deposition of aerosol particles (F_d) is given by the product of the measured air concentration of the substance (C_{air}) and a dry deposition velocity (V_d),

$$F_d = C_{\text{air}} \cdot V_d, \quad (1)$$

where, F_d is the flux of aerosol particles due to dry deposition ($\text{g m}^{-2} \text{s}^{-1}$), C_{air} is the concentration of that substance in the near-surface atmosphere ($\mu\text{g m}^{-3}$), V_d is the dry deposition velocity (cm s^{-1}).

Major uncertainties associated with F_d are primarily attributed to the estimate of the dry deposition velocity (V_d) of particles of different size. We focus on the elemental mass-size distribution of aerosol particles and the modeling of the dry deposition velocity for dust particles in the following discussion.

In the past, the total dry deposition rates were often obtained simply using the dry deposition velocity which corresponds to the mass-median diameter (MMD) of aerosol particles containing the element of interest. The mass-median diameter is defined as the particle-size for which 50% of the mass is associated with smaller particles and 50% of the mass is associated with larger particles. It is now recognized that it is essential to take into account the entire mass-size distribution (MSD) of dust particles to obtain a better estimate of the total dry deposition rates (Arimoto and Duce, 1986; Arimoto et al., 1985; Dulac et al., 1989; McDonald et al., 1982). The MSD of dust particles could not be assessed from direct measurements of aerosol particles in many size intervals owing to limitations in the types of samples available. However we have undertaken a least-squares linear regression method for fitting MSDs to the atmospheric Al data obtained from cascade impactor sampling in this study, as described

below:

$$Y = A + B \log D, \tag{2}$$

where Y is the standard normal deviate of the cumulative % of the total mass of Al on each cascade impactor stage based on tables given by Dixon and Massey (1969), and $\log D$ is the logarithm of the particle diameter. Monomodal lognormal MSDs were assumed for fitting.

For calculating the dry deposition fluxes of dust, we have used a 100-step method developed by Arimoto et al. (1985). This approach takes into account the MSDs of Al and it depends on the fitting of a continuous distribution to the cascade impactor data. The fitted distribution is divided into 100 discrete successive size intervals, with each interval representing 1% of the total Al mass, which has the diameter of the particle at its center ($D_{0.5\%}, D_{1.5\%}, \dots, D_{99.5\%}$). For a total 14 sets of cascade impactor data obtained in this study, the calculated correlation coefficients for log-normal fitting ranged from 0.94 to 0.99.

Thus, we can replace eqn. (1) with

$$F_d = \sum_{i=1}^{100} V_d(D_{(i-0.5)\%}) C_{air}/100, \tag{3}$$

where $V_d D_{(i-0.5)\%}$ is the dry deposition velocity for a particle with the diameter at the center of the size interval i corresponding to i th% of the cumulative mass. An important consequence of

this approach is that the mass of Al is divided into a finite number of size intervals, and these sizes range over the diameters between 0.5% to 99.5% of the cumulative mass. Thus the total dry deposition rate for Al (F_d) was calculated as the sum of the dry deposition for all particle-size intervals based on the above equation. However, uncertainties associated with the model calculations should be expected owing to difficulties in sampling both giant and fine particles. Equally important are the uncertainties in the dry deposition velocities themselves.

Since direct measurements of the dry deposition velocity of aerosol particles are limited (Arimoto et al., 1985; Dulac et al., 1989), dry deposition velocities (V_d) in eq. (3) were derived from a model for dry deposition to a smooth sticky surface (Slinn and Slinn, 1981). It is worth mentioning that large uncertainties still exist in dry deposition models; these have been discussed by Slinn (1983) and Sievering (1984) who concluded that the uncertainties are especially large for submicrometer aerosol fraction.

Table 3 summarizes the calculated mean dry deposition velocities of atmospheric Al and the corresponding MMDs based on cascade impactor samples collected at Xiamen (four sets), Beijing (nine sets) and during the cruise (one set).

The calculated average V_d for samples at Xiamen

Table 3. Mean dry deposition velocities of atmospheric Al (V_d) and the corresponding mass-median diameters (MMDs) (1992)

Location	Date	V_d (cm s ⁻¹)		MMD	MMDs (μm)	
		Arithmetic mean	range		average	range
Beijing	April 24	1.4 (2.8 ^a)	1.4–3.3	1.6	5.0 ^a	1.6–5.9
	25	1.6		2.6		
	28	1.8		1.8		
	29	1.5		1.6		
	30	2.9		5.3		
	May 1	3.0		4.9		
	3	2.1		4.7		
	7	3.0		5.4		
	8	3.0		4.7		
Xiamen	May 15	1.6 (1.4)	1.2–1.6	4.0	3.1	2.1–4.0
	16	1.4		3.5		
	18	1.2		2.1		
	20	1.4		2.9		
E. China Sea	April 24	1.9		3.2		

^a Only samples collected between 30 April and 8 May are considered.

is 1.4 cm s^{-1} , ranging from 1.2 to 1.6 cm s^{-1} with the corresponding MMDs between 2.1 and $4.0 \mu\text{m}$. The calculated average V_d for the Beijing samples is 2.3 cm s^{-1} , with a range of 1.4 to 3.0 cm s^{-1} ; this is higher than that at Xiamen, consistent with larger MMDs of Al at Beijing (1.6 – $5.9 \mu\text{m}$). The calculated average V_d at Beijing is comparable to a mean value of 2.1 cm s^{-1} (1.7 – 2.6 cm s^{-1}) reported by X. Y. Zhang et al. (1993) in their studies conducted at Beijing during the non-dust period in the spring of 1990. X. Y. Zhang et al. (1993) suggested a mean dry deposition velocity of 4.8 cm s^{-1} during dust storms. Since we do not have cascade impactor samples available for dust storm periods, we accept 4.8 cm s^{-1} as a representative mean V_d for the dust storm periods. For the spring, including both dust (4.8 cm s^{-1}) and non-dust periods (2.3 cm s^{-1}), a mean value of 3.6 cm s^{-1} was used for calculating the total dry deposition rates of dust at Beijing.

The calculated mean V_d value over the East China Sea is 1.9 cm s^{-1} , which is comparable to that at Xiamen; this is attributed to similar MMDs: $3.2 \mu\text{m}$ over the East China Sea versus $3.1 \mu\text{m}$ at Xiamen. Since we do not have cascade impactor samples for Qingdao, we assume that the MSDs obtained over the East Sea are applicable to Qingdao. These results are consistent with a V_d of 2 cm s^{-1} which has been widely used to estimate the dry deposition for dust particles for coastal areas (Uematsu et al., 1985; Duce et al., 1991; Gao et al., 1992a). However, the lower mean V_d at Xiamen compared with that over the East China Sea is consistent with the greater distance from the source region and thus smaller particle size.

We should mention that the dry deposition velocity should account for all mechanisms related to the dry deposition process. Each of these mechanisms is a function of a number of factors, including particle size, particle density, wind speed, relative humidity, etc. Therefore it is difficult to predict dry deposition velocities accurately. After careful consideration of deposition process, GESAMP (1989) suggested that the best value for the dry deposition velocity should be associated with an uncertainty of a factor of three. Since most of dust particles are in the size range where gravitational settling is the controlling factor, the mean dry deposition velocities derived from the particle-size spectrum should be relatively reliable.

At Beijing a mean V_d of 2.1 (1.4 – 4.8) cm s^{-1} is appropriate based on a combination of results from this study and the work of X. Y. Zhang et al. (1993). Considering the full range of the dry deposition velocities calculated by our model, uncertainties in sampling and analyses, and limitation of this modeling approach, the mean deposition velocities predicted in this study should include uncertainties at least a factor of 2 to 3.

3.4.2. Model for estimating atmospheric fluxes by wet deposition. When direct precipitation chemistry measurements are not available, an estimate of the atmospheric removal of dust via wet deposition (F_w) can be made using scavenging ratios. Studies have shown that the concentration of a substance in the atmosphere is generally related to the concentration of that substance in rain (Duce et al., 1991). This relationship can be described in terms of a scavenging ratio:

$$S = \rho \cdot C_r / C_a, \quad (4)$$

where S is the scavenging ratio for a substance (dimensionless), C_r is the concentration of that substance in rain (g g^{-1}), C_a is the concentration of that substance in the atmosphere (g m^{-3}), and ρ is the density of air ($\sim 1200 \text{ g m}^{-3}$).

The atmospheric flux via wet deposition (F_w) can be expressed as:

$$F_w = C_r \cdot P, \quad (5)$$

where P is the precipitation rate (m s^{-1}). If C_r is replaced according to (4), eq. (5) can be rewritten as:

$$F_w = P \cdot S \cdot C_a / \rho. \quad (6)$$

Using eq (6), we can estimate the atmospheric fluxes of substances through wet deposition based only on their concentrations in air, the precipitation rate, and the scavenging ratio. Since S is a function of many parameters including particle size, particle shape, the vertical distribution of the atmospheric concentrations, the vertical extent of the rain and the rain cloud, etc., it is difficult to predict its value accurately. GESAMP (1989) reported a range of scavenging ratios for mineral aerosol of 100 to 2000, and suggested that 500 to 2000 are the most appropriate values for coastal seas. The Pacific data of Uematsu et al. (1985) resulting from direct measurements suggest a mean scavenging ratio of 1000 for mineral aerosol with

a range of 500 to 2000; their conclusion also fits observational results obtained from the UK (Cawse, 1977) in spite of large climatological differences between the two regions. Over east Asia, however, few simultaneous measurements of mineral aerosol in air and in precipitation have been made along the Asian coast. Recently the total particle concentrations in precipitation were measured by filtration for a site in the northwest of the Yellow Sea (J. Zhang et al., 1993). If we combine J. Zhang's total particle results in precipitation with our air concentrations of dust particles measured at Qingdao, the resulting mean scavenging ratio for mineral aerosol in the spring would be about 2000. If we assume that the total particles in rain from J. Zhang et al. (1993) were exclusively dust particles, this preliminary calculation indicates that the scavenging ratios of dust for the Asian coast could be toward the upper values of the range reported by GESAMP. However, since these two separate studies did not simultaneously sample precipitation and air, substantial errors in the calculated scavenging ratio could exist. Thus the putative mean scavenging ratio of 2000 for mineral aerosol along the Asian coast needs to be further verified.

For this work, we chose to adopt previous results suggested by GESAMP and Uematsu et al. from the Pacific, and we assume a mean scavenging ratio of 1000 with a range of 500 to 2000 to be the best estimate for mineral aerosol along the Asian coast. It is worth mentioning that substantial uncertainties associated with indirect estimates of wet deposition should always be expected due to difficulties in evaluating the scavenging ratio. In addition, the scavenging ratios for oceanic

environments suggested by GESAMP may not be applicable to continental regions, although they have been used in the past for certain purposes, such as calculation of dust deposition to the Loess Plateau by X. Y. Zhang et al. (1993). The climatological mean precipitation rates for the various sites in this study were obtained from Watts (1969) and listed in Table 4 for calculating wet deposition.

3.5. *Atmospheric deposition and fluxes of dust over inland China and the China Sea*

Atmospheric deposition rates for dust have been estimated for the western North Pacific regions (Tsunogai et al., 1985) and the Yellow Sea of the East China Sea near the Asian continent (Gao et al., 1992a). However, estimates of dust fluxes to most areas of China have not yet been made. Samples collected during this study are the most representative of the springtime atmospheric conditions over the Loess Plateau regions, northern and southern China, as well as the coastal and off-shore regions of the China Sea. Estimations of the atmospheric deposition of dust over continental China and the China Sea based on these data are made below.

3.5.1. Estimated atmospheric fluxes of dust to 5 sites. On the basis of the discussion above, the total atmospheric fluxes of dust in the spring are estimated as the sum of dry plus wet deposition processes at Xi'an, Beijing, Qingdao, Xiamen, and the East China Sea (Table 5). Estimates in Table 5 are based on the concentrations of atmospheric Al observed in the spring (March — May) at

Table 4. *Values used in modeling atmospheric fluxes of mineral aerosol at the different location, Spring 1992*

Locations	Dry deposition velocity (cm s^{-1})		Scavenging ratio		Precipitation rate (mm mo^{-1})	Wind speed (cm s^{-1})
	mean	range	mean	range	mean ^a	mean ^a
Xian	4.9 ^b	3.2–7.1 ^b	1000	500–2000	49.5	—
Beijing	3.6	2.1–4.8	1000	500–2000	25.9	310
Qingdao	2.1	0.9–4.0	1000	500–2000	37.5	570
E. China Sea	1.9	0.9–4.0	1000	500–2000	65.4	510
Xiamen	1.4	0.5–3.0	1000	500–2000	151	290

^a The springtime climatological data of Watts (1969).

^b X. Y. Zhang et al. (1993).

Table 5. Total atmospheric fluxes of mineral aerosol by dry and wet deposition processes in the spring at different locations^a

Locations	Fluxes ($\text{g m}^{-2} \text{mo}^{-1}$)			
	dry deposition F_d	wet deposition F_w	total deposition $F_d + F_w$	% F_w (mean)
Xian ($n=18$)	19 (3.7–33)	6.0 (1.2–11)	25 (4.9–44)	24
Beijing ($n=21$)	15 (5.1–51)	3.3 (1.2–12)	18 (6.3–63)	18
Qingdao ($n=15$)	1.9 (0.33–6.9)	1.1 (0.19–4.2)	3.0 (0.52–11.1)	37
Xiamen ($n=15$)	1.1 (0.33–2.9)	2.5 (0.57–6.9)	3.6 (0.90–9.8)	69
East China Sea ($n=6$)	1.3 (0.36–2.4)	1.4 (0.39–2.7)	2.7 (0.75–5.1)	52

^a Mean total fluxes are reported, with the range in parentheses. n : number of aerosol samples being averaged.

different sites, assuming that aluminum accounts for 7% of loess soils (Liu et al., 1985). For the fluxes by dry and wet deposition processes, all individual flux values at each site are averaged to give the mean flux, with a range indicating the minimum and the maximum individual fluxes as the lower and the upper limits. The total fluxes are calculated as the sum of the mean fluxes from dry and wet deposition processes. The lower limit of the corresponding range for the total mean flux is the sum of the minimum fluxes by dry and wet deposition, and the same approach is applied to the upper limit of the range.

The mean area-weighted total flux of atmospheric dust in the spring is $25 \text{ g m}^{-2} \text{mo}^{-1}$ at Xi'an, with a range of 4.9 to $44 \text{ g m}^{-2} \text{mo}^{-1}$; this is the highest among the regions in this study (Table 5). At Xi'an the total fluxes of atmospheric dust are dominated by dry deposition. Although Xi'an is located in the Loess Plateau, which is the source for mineral dust over much of east Asia, the deposition and resuspension of dust particles, especially coarse particles in the region, is an important process during the dust transport. It is possible that a considerable amount of dust material is recycled between the land surface and the atmosphere, that is, deposited and later re-suspended. Therefore the Loess Plateau can serve as both a source and a sink for mineral dust. The next highest total flux of dust estimated is at Beijing, ranging from 6.3 to $63 \text{ g m}^{-2} \text{mo}^{-1}$. Using

the monthly average concentrations of atmospheric Al at Beijing described in Subsection 3.2. and the above approach of calculating the total deposition, the estimated deposition of dust in the spring (March — May) at Beijing roughly accounts for 34% of the annual deposition (We assume that this is applicable to other sites in northern China in the following calculation of annual deposition).

In the nearshore and off-shore regions of the China Sea, the atmospheric fluxes of dust are much lower compared with those at the continental sites. The total mean atmospheric fluxes of dust are $3.0 \text{ g m}^{-2} \text{mo}^{-1}$ at Qingdao, $2.7 \text{ g m}^{-2} \text{mo}^{-1}$ over the off-shore regions of the East China Sea, and $3.6 \text{ g m}^{-2} \text{mo}^{-1}$ at Xiamen. However, the extent of removal by wet processes are relatively high from ~37% at Qingdao to ~52% over the East China Sea, and ~69% at Xiamen. The percentage of the total fluxes by wet removal is highest at Xiamen, which is due to the high precipitation rate at Xiamen in the spring (151 mm mo^{-1} , see Table 4). This suggests that scavenging by precipitation is particularly important for the removal of dust from the atmosphere in the coastal region of the South China Sea.

3.5.2. *Estimated atmospheric fluxes of dust at some other locations.* In order to obtain information on the atmospheric fluxes of dust at some additional locations where we were unable to

sample, we have applied our deposition models to the results for atmospheric Al obtained from other studies conducted over continental China and the China Sea. These studies include the JACK (Japan-China-Korea) Network (Hashimoto et al., 1994), the Pacific Exploratory Mission (PEM-West) (R. Arimoto, unpublished data, 1994), and the Yellow Sea Project (Gao et al., 1992a). The estimated fluxes of dust at six additional locations are presented in Table 6. In these calculations, we also assume that aluminum accounts for 7% of crustal material. The mean total fluxes of dust are $39 \text{ g m}^{-2} \text{ mo}^{-1}$ at Baotou and $41 \text{ g m}^{-2} \text{ mo}^{-1}$ at Lanzhou higher than at Xian, since these 2 sites are located within the major desert regions. The calculated mean total fluxes of dust in the spring are $3.2 \text{ g m}^{-2} \text{ mo}^{-1}$ at Cheju and $3.7 \text{ g m}^{-2} \text{ mo}^{-1}$ at Mallipo in South Korea, which are comparable to those for Qingdao ($3.0 \text{ g m}^{-2} \text{ mo}^{-1}$) and over the off-shore region of the East China Sea ($2.7 \text{ g m}^{-2} \text{ mo}^{-1}$). The mean total fluxes of dust are $1.4 \text{ g m}^{-2} \text{ mo}^{-1}$ at Kato in Hong Kong and $0.55 \text{ g m}^{-2} \text{ mo}^{-1}$ at Kenting in Taiwan over the South China Sea. The relatively low total flux of dust at Kenting compared with that at Xiamen ($3.6 \text{ g m}^{-2} \text{ mo}^{-1}$) is probably a result of a decreased influence from the mainland.

3.5.3. Present atmospheric fluxes of dust and eolian dust accumulation rates. In the previous section, we estimated the dust fluxes based on

simple atmospheric dry and wet deposition models. One way to test how reliable these estimations are is to compare the atmospheric fluxes with the historic eolian accumulation rates. A recent investigation by Zhang et al. (1994) provides late Quaternary records of the input of eolian dust to the Loess Plateau which make it possible to address this issue. We will focus on this work, comparing our results from Xi'an with their results of late Quaternary records for the Loess Plateau.

The estimated total atmospheric fluxes of dust at Xi'an from this study are 25 (4.9 to 44) $\text{g m}^{-2} \text{ mo}^{-1}$ in the spring. We assume that the deposition of dust in the spring accounts for 34% of the annual deposition at Xi'an, i.e., the same as at Beijing. Thus the annual atmospheric flux of dust at Xi'an would be 220 (43 to 390) $\text{g m}^{-2} \text{ yr}^{-1}$. Based on late Quaternary records (An et al., 1991), during the last glaciation the average dust accumulation rate was equivalent to $210 \text{ g m}^{-2} \text{ yr}^{-1}$ as dust. If we take into account the minimum accumulation rate ($0.43 \text{ g cm}^{-2} 10^{-3} \text{ yr}^{-1}$ for Al) and the maximum accumulation rate ($3.2 \text{ g cm}^{-2} 10^{-3} \text{ yr}^{-1}$ for Al) in the Quaternary records, the corresponding values for dust would be $61 \text{ g m}^{-2} \text{ yr}^{-1}$ and $460 \text{ g m}^{-2} \text{ yr}^{-1}$, respectively. These historical accumulation rates are quite comparable to what is estimated in the present study.

3.5.4. Total atmospheric deposition and fluxes of dust over the China Sea. The China Sea is the

Table 6. Estimated atmospheric fluxes of mineral aerosol by dry and wet deposition processes at additional locations over continental China and the China Sea

Site	Sampling date (mo/yr)	Location	Mineral aerosol ($\mu\text{g m}^{-3}$)	Mean fluxes ($\text{g m}^{-2} \text{ mo}^{-1}$)			
				F_d	F_w	$F_d + F_w$	% F_w
Baotou	10/91	41°N, 110°E	360 ^a	32	6.9	39	18
Lanzhou	10/91	36°E, 104°E	390 ^a	35	5.9	41	14
Kato (Hong Kong)	3–5/92	23°N, 113°E	12 (± 17) ^b ($n=10$)	0.42 (0.061–2.2)	0.96 (0.14–5.0)	1.4 (0.21–7.2)	70
Kenting (Taiwan)	9–10/91	23°N, 120°E	4.8 (± 5.9) ^b ($n=42$)	0.17 (0.005–1.1)	0.38 (0.02–3.8)	0.55 (0.025–4.9)	69
Cheju (S. Korea)	4–5/92	33°N, 127°E	30 (± 27) ^b ($n=8$)	1.5 (0.44–4.3)	1.7 (0.51–4.9)	3.2 (0.95–9.2)	53
Mallipo (S. Korea)	4–5/89	37°N, 128°E	33 (± 19) ^c ($n=14$)	1.6 (0.39–16)	2.1 (0.93–37)	3.7 (1.3–53)	57

^a Derived from the JACK Network (Hashimoto et al. 1994).

^b Derived from the PEM-West (unpublished data, Arimoto, 1994).

^c Derived from the Yellow Sea Project (Gao et al., 1992).

largest regional sea that is adjacent to the Asian continent. Technically, the China Sea includes the East China Sea (1,200,000 km²) and the South China Sea (3,400,000 km²). The East China Sea in turn is composed of the Bohai Sea, the Yellow Sea, and the East Sea. At present information on the dust deposition to the entire basin of the China Sea is limited (Gao et al., 1992; J. Zhang et al., 1993).

Basin-wide estimates of atmospheric deposition of dust are made separately for the East China Sea and the South China Sea (Table 7). This separate estimation is necessary since the mineral dust content of the marine atmosphere is quite different for these two basins. The northern and central parts of the East China Sea are directly under the pathway of dust transport, especially in the spring, and the atmospheric fluxes of dust over the East China Sea are expected to be higher than those over the South China Sea. Since the aerosol samples for this study were collected from both coastal regions and off-shore areas, we have subdivided each basin into two parts: the coastal region and the off-shore region for a more accurate estimation of atmospheric dust deposition. The area of the coastal zone is obtained by multiplying the effective length of the coastline (1,500 km for the East China Sea and 2,500 km for the South China Sea) by 25 km, which is representative of the scale of the land-breeze/sea-breeze circulation for a typical coastal region. The deposition and fluxes of dust to the coastal region are estimated based on the results from samples collected at Qingdao and Xiamen. Results from the cruise in this study and from the work of Arimoto et al. (1996) for the PEM-West Program are used to

estimate the fluxes and deposition of dust for the off-shore areas of the East China Sea. Samples from these two studies were collected in the spring of 1992.

Since there are no samples available in this study from the off-shore areas of the South China Sea, we selected the results from two samples collected during the cruise over the southern region of the East China Sea. This area is closer to the South China Sea and those two samples had the lowest Al concentrations observed (0.51 $\mu\text{g m}^{-3}$ and 1.0 $\mu\text{g m}^{-3}$; the corresponding mean atmospheric flux of dust is 0.40 to 0.81 $\text{g m}^{-2} \text{mo}^{-1}$, respectively). In addition, the atmospheric fluxes of dust at Kato and Kenting in Table 6 were estimated. By averaging the above fluxes, we obtained an flux of 0.48 (0.14 to 1.1) $\text{g m}^{-2} \text{mo}^{-1}$ for the entire basin of the South China Sea.

In terms of annual deposition, we assume that the spring deposition accounts for 34% of the annual deposition at sites in East China Sea as was observed for Beijing. For the South China Sea region, the annual deposition was obtained by integrating the monthly values. The estimated atmospheric fluxes of dust over the coastal regions of the East China Sea range from 4.6 $\text{g m}^{-2} \text{yr}^{-1}$ to 98 $\text{g m}^{-2} \text{yr}^{-1}$ with a value of 27 $\text{g m}^{-2} \text{yr}^{-1}$. The estimated mean atmospheric flux of dust over the coastal region of the South China Sea is 43 (11 to 120) $\text{g m}^{-2} \text{yr}^{-1}$. For the off-shore regions, the estimated atmospheric fluxes of dust are 26 $\text{g m}^{-2} \text{yr}^{-1}$ for the East China Sea and 10 $\text{g m}^{-2} \text{yr}^{-1}$ for the South China Sea. Large uncertainties in total deposition values primarily originate from errors associated with the dry deposition velocities and scavenging ratios as well as the

Table 7. *Atmospheric fluxes and deposition of dust over the China sea*

Name	Surface area ($\times 10^3 \text{ km}^2$)	Total fluxes ($\text{g m}^{-2} \text{yr}^{-1}$) ^a	Total deposition (Tg yr^{-1}) ^a
East China Sea			
coastal	38	27 (4.6–98)	1.0 (0.17–3.7)
off-shore	1160	26 (7.5–63)	30 (8.7–73)
South China Sea			
coastal	63	43 (11–120)	2.7 (0.69–7.6)
off-shore	3300	10 (2.5–52)	33 (8.3–170)
total:	4500		67 (18–260)

^a Estimated annual means with a range in parentheses.

natural fluctuations of the measured concentrations.

To obtain the total amounts of dust deposited, we averaged the atmospheric fluxes of dust for coastal and off-shore regions. For example, the averaged flux for the region of the East China Sea is $27 \text{ g m}^{-2} \text{ yr}^{-1}$ with a range of 6.1 to $81 \text{ g m}^{-2} \text{ yr}^{-1}$. The lower limit in this range is the mean of the minimum fluxes for the coastal and offshore regions, and the same approach is used to obtain the upper limit. This is comparable to the results from a study conducted in the Yellow Sea which is located in the central part of the East China Sea, where the estimated atmospheric fluxes over the Yellow Sea range from 10 to $89 \text{ g m}^{-2} \text{ yr}^{-1}$ (Gao et al., 1992a). The difference between these two studies is that the flux estimates in this study are totally based on individual samples collected at coastal and off-shore regions, whereas the estimates from the earlier study are partially based on aerosol samples and partially based on model estimated concentrations.

The averaged value of dust fluxes to the entire China Sea from our study is $26 \text{ g m}^{-2} \text{ yr}^{-1}$, substantially lower than the average value of $53.7 \text{ g m}^{-2} \text{ yr}^{-1}$ for the China Sea by J. Zhang et al. (1993), who conducted dust measurements at a site to the northwest of the Yellow Sea. We believe that the difference related to the dust flux estimates is due to the difference in methodology regarding the dust measurement. We analyzed our aerosol particle samples for Al concentrations and converted the measured Al concentrations to the dust concentrations by assuming that Al accounts for 7% of the total dust material. However, J. Zhang et al. (1993) considered that all particulate substances which could not pass the $0.45 \mu\text{m}$ filters are dust particles without any further elemental analyses of particulate material. Their estimation of dust fluxes based on the filtration technique could overestimate the dust deposition because the particulate material include not only dust substances but also many other substances, such as organic particulate substances. In addition, their estimate of the dust deposition to the China Sea is based on data from only one site; this may not be representative and applicable to the entire basin of the China Sea, especially since our observations have indicated that the dust concentrations are substantially lower over the South China Sea.

Based on our studies, the estimated total deposition of dust is 31 Tg yr^{-1} for the entire East China Sea. The atmospheric deposition of dust to the South China Sea is estimated to be 36 Tg yr^{-1} . Thus the total deposition of dust to the China Sea is 67 Tg yr^{-1} with a range of 18 to 260 Tg yr^{-1} . This deposition to the China Sea accounts for almost 14% of the total deposition of dust to the entire North Pacific (480 Tg yr^{-1} , Duce et al., 1991). Since the impact of dust materials on the oceanic biosphere largely depends on the quantity of material deposited, such a high deposition rates of dust materials over East Asia must play an important role in biogeochemical cycles of trace substances in that region.

4. Conclusions

Measurements of atmospheric Al at Xi'an, Beijing, Qingdao, Xiamen, and over the East China Sea lead to the following conclusions.

(1) Within China and over coastal waters a trend of decreasing atmospheric concentrations of Al toward east has been observed, with $10 \pm 5.3 \mu\text{g m}^{-3}$ at Xi'an, $9.6 \pm 4.7 \mu\text{g m}^{-3}$ at Beijing, $2.5 \pm 2.2 \mu\text{g m}^{-3}$ at Qingdao, and $1.9 \pm 1.1 \mu\text{g m}^{-3}$ over the East China Sea. This is primarily attributed to the increasing distances from the dust source region, the Loess Plateau. An apparent difference in Al concentrations between Northern China and Southern China was also observed, reflected by low Al concentrations at Xiamen ($1.5 \pm 1.1 \mu\text{g m}^{-3}$).

(2) Results from our 2.5-yr measurements at Beijing reveal that anthropogenic emissions, probably from coal burning, is a significant source for atmospheric Al at this site.

(3) The particle-size distribution of dust varies depending on the distance from aerosol source regions. High concentrations of Al are primarily associated with particles of 3.6 to $9.3 \mu\text{m}$ diameter at Beijing in northern China and 1.1 to $3.3 \mu\text{m}$ diameter over the off-shore areas of the East China Sea.

(4) The model-predicted mean dry deposition velocities of dust particles are 1.4 to 4.8 cm s^{-1} over northern continental China and 1.4 to 2.1 cm s^{-1} over the China Sea, with the corresponding MMDs of 1.6 to $5.9 \mu\text{m}$ and 2.1 to $4.0 \mu\text{m}$, respectively. Based on model estimations, the highest

atmospheric fluxes of dust was obtained at Xi'an of the Loess Plateau: 25 (4.9 to 44) $\text{g m}^{-2} \text{mo}^{-1}$, and this atmospheric deposition rate is consistent with the late Quaternary records of eolian dust accumulation rate. The total atmospheric deposition to the entire China Sea is 67 (18 to 260) Tg yr^{-1} .

To improve deposition estimates of dust, future studies will focus on direct measurements of total deposition of dust at several representative locations and further comparisons between field measurements and deposition modeling. The fractions of atmospheric Al from different sources also should be further evaluated.

5. Acknowledgments

We wish to thank B. Ray, D. Cullen, and T. Urszula for assistance in the laboratory work. Participation of many individuals in field sampling and site operation is highly appreciated. We also thank three reviewers for their critical comments which greatly improved the original manuscript. Comments from M. Uematsu are appreciated. This project was supported by the U. S. NSF ATM-532524 and NASA NAG 11225. Additional support was provided by the Chinese Academy of Sciences, the Chinese Natural Science Foundation (Awards 49291100 and 49373188), and the State Oceanic Administration of the People's Republic of China.

REFERENCES

- An, Z. S., Kukla, G., Porter, S. C. and Xiao, J. L. 1991. Late Quaternary dust flow on the Chinese Loess Plateau. *Catena* **18**, 125–132.
- Ando, M., Katagiri, K., Tamura, K., Yamamoto, S., Li, Y. F., Cao, S. R., Ji, R. D., and Liang, C. K. 1996. Indoor and outdoor air pollution in Tokyo and Beijing supercities. *Atmos. Environ.* **30**, 695–702.
- Arimoto, R., Duce, R. A., Ray, B. J. and Unni, C. K. 1985. Atmospheric trace elements at Enewetak Atoll: 2. Transport to the ocean by wet and dry deposition. *J. Geophys. Res.* **90**, 2391–2408.
- Arimoto, R. and Duce, R. A. 1986. Dry deposition models and the air/sea exchange of trace elements. *J. Geophys. Res.* **91**, 2787–2792.
- Arimoto, R., Y. Gao, M. Y. Zhou, D. S. Lee, L. Q. Chen, D. Y. Gu, and Z. Wang, Atmospheric deposition of trace elements to the western Pacific Basin, In: *Atmospheric deposition of contaminants to the Great Lakes and coastal waters* (ed. J. E. Baker), in press, 1996.
- Blank, M., Leinen, M. and Prospero, J. M. 1985. Major Asian eolian inputs indicated by the mineralogy of aerosols and sediments in the western North Pacific. *Nature* **314**, 84–86.
- Brimblecombe, P. 1986. *Air composition and chemistry*. The Press Syndicate of the University of Cambridge, Cambridge, Great Britain, 224pp.
- Buat-Menard, P., Ezat, U. and Gaudichet, A. 1983. Size distribution and mineralogy of aluminosilicate dust particles, in Tropical Pacific air and rain. In: *Precipitation scavenging, dry deposition, and resuspension* 2 (eds. H. R. Pruppacher, R. G. Semonin, and W. G. N. Slinn). Elsevier, New York, 1259–1269.
- Burton, R. M. 1973. Field evaluation of the high-volume particle fractionating cascade impactor. *J. Air Pollut. Control Assoc.* **23**, 277–281.
- Cawse, P. A. 1977. A survey of atmospheric trace elements in the UK: Results for 1976, AERE-R8869, United Kingdom Atomic Energy Authority, AERE, Harwell, Oxfordshire, England, 22pp.
- Chung, Y. S. 1992. On the observations of yellow sand (dust storms) in Korea. *Atmos. Environ.* **26A**, 2743–2749.
- Dixon, W. J. and Massey, Jr., F. J. 1969. *Introduction to statistical analysis*. MacGraw-Hill, Inc. New York, 462–463.
- Duce, R. A., Unni, C. K., Ray, B. J., Prospero, J. M. and Merrill, J. T. 1980. Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific: Temporal variability. *Science* **209**, 1522–1524.
- Duce, R. A. 1986. The impact of atmospheric nitrogen, phosphorus, and iron species on marine biological productivity. In: *The role of air-sea exchange in geochemical cycling* (ed. P. Buat-Menard). D. Reidel, Norwell, Mass., pp. 497–529.
- Duce, R. A., Liss, P. S., Merrill, J. T., Atlas, E. L., Buat-Menard, P., Hicks, B. B., Miller, J. T., Prospero, J. M., Arimoto, R., Church, T. M., Ellis, W., Galloway, J. N., Hansen, L., Jickells, T. D., Knap, A. H., Reinhardt, K. H., Schneider, B., Soudine, A., Tokos, J. J., Tsunogai, S., Wollast, R. and Zhou, M. 1991. The atmospheric input of trace species to the world ocean. *Global Biogeochem. Cycles* **5**, 193–256.
- Duce, R. A. 1995. Sources, distributions, and fluxes of dusts and their relationships to climate. In: *Aerosol forcing of climate* (eds. R. Charlson and J. Heintzenberg). John Wiley and Sons, London, 43–72.
- Dulac, F., Buat-Menard, P., U. Ezat, Melki, S. and Bergametti, G. 1989. Atmospheric input of trace metals to the western Mediterranean: uncertainties in modeling dry deposition from cascade impactor data. *Tellus* **41B**, 362–378.
- Dzubay, T. G. 1976. Particle bounce errors in cascade impactors. *Atmos. Environ.* **10**, 229–234.
- Gao, Y., Arimoto, R., Duce, R. A., Lee, D. S. and Zhou,

- M. Y. Zhou. 1992a. 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., Arimoto, R., Zhou, M. Y., Merrill, J. T. and Duce, R. A. 1992b. Relationships between the dust concentrations over Eastern Asia and the remote North Pacific. *J. Geophys. Res.* **97**, 9867–9872.
- GESAMP, Group of Experts on Scientific Aspects of Marine Pollution. 1989. The atmospheric Input of Trace Species to the World Ocean. *Reports and Studies no. 38*, 111pp, World Meteorological Organization, Geneva, Switzerland.
- Gillette, D. A. and Walker, T. R. 1977. Characteristics of airborne particles produced by wind erosion of sandy soil, high plains of west Texas. *Soil Sci.* **123**, 97–110.
- Gillette, D. A. and Dobrowolski, J. P. 1993. Soil crust formation by dust deposition at Shaartuz, Tadzhik, U. S. S.R. *Atmos. Environ.* **27A**, 2519–2525.
- Hashimoto, Y., Kim, H. K., Otoshi, T. and Sekine, Y. 1990. Monitoring of atmospheric aerosol components by multi-elemental neutron activation analysis in Seoul, Korea, April 1987–March 1989. *J. Japan Soc. Air Pollut.* **25**, 313–323.
- Hashimoto, Y., Kim, H. K., Otoshi, T. and Sekine, Y. 1991. Air quality monitoring at Seoul, Korea, May 1986–March 1989. *J. Japan Soc. Air Pollut.* **26**, 51–58.
- Hashimoto, Y., Sekine, Y., Kim, H. K., Chen, Z. L. and Yang, Z. M. 1994. Atmospheric fingerprints of east Asia, 1986–1991: An urgent Record of aerosol analysis by the JACK Network. *Atmos. Environ.* **28A**, 1437–1445.
- Hayasaka, T., Nakajima, T. and Yanaka, M. 1990. The coarse particle aerosols in the free troposphere around Japan. *J. Geophys. Res.* **95**, 14,039–14,047.
- Iwasaka, Y., Yamato, M., Imasu, R. and Ono, A. 1988. Transport of Asian dust (KOSA) particles: Importance of weak KOSA events on the geochemical cycle of soil particles. *Tellus* **40B**, 494–503.
- Kato, N. and Akimoto, H. 1992. Anthropogenic emissions of SO₂ and NO_x in Asia: emission inventories. *Atmos. Environ.* **26A**, 2997–3017.
- Leinen, M., Prospero, J. M., Arnold, E. and Blank, M. 1994. Mineralogy of aeolian dust reaching the North Pacific Ocean: 1. Sampling and analysis. *J. Geophys. Res.* **99**, 21,017–21,023.
- Liu, T. S. et al. 1985. *Loess and the environment*. China Ocean Press, Beijing, 215 pp.
- McDonald, R. L., Unni, C. K. and Duce, R. A. 1982. Estimation of atmospheric sea salt dry deposition: wind speed and particle size dependence. *J. Geophys. Res.* **87**, 1246–1250.
- Martin, J. M. and Gordon, R. M. 1988. Northeast Pacific iron distributions in relation to phytoplankton productivity. *Deep-Sea Res.* **35**, 177–196.
- Merrill, J. T., Uematsu, M. and Bleck, R. 1989. Meteorological analysis of long range transport of dust over the North Pacific. *J. Geophys. Res.* **94**, 8584–8598.
- Merrill, J. T., Arnold, E., Leinen, M. and Weaver, C. 1994. Mineralogy of aeolian dust reaching the North Pacific Ocean: 2. Relationship of mineral assemblages to atmospheric transport patterns. *J. Geophys. Res.* **99**, 21, 025–21, 032.
- Okada, K., Kobayashi, A., Iwasaka, Y., Naruse, H., Tanaka, T., and Nemoto, O. 1987. Features of individual Asian dust-storm particles collected at Nagoya, Japan. *J. Meteorol. Soc. Japan* **65**, 515–521.
- Penner, J. E., Charlson, R. J., Hales, J. M., Laulainen, N., Leifer, R., Novakov, T., Ogren, J., Radke, L. F., Schwartz, S. E. and Travis, L. 1993. Quantifying and minimizing uncertainty of climate forcing by anthropogenic aerosols. *Technical Report of U. S. Department of Energy, DOE/NBB-0092T*, 53pp.
- Prospero, J. M. 1981. Eolian transport to the World Ocean. In: *The oceanic lithosphere*, vol. 7: *The sea* (ed. C. Emiliani). John Wiley, New York, 801–874.
- Prospero, J. M., Uematsu, M. and Savoie, D. L. 1989. Dust transport to the Pacific Ocean. In: *Chemical oceanography*, vol. 10 (ed. J. P. Riley, R. Chester and R. A. Duce). Academic, San Diego, Calif., 188–218.
- Sievering, H. 1984. Small-particles dry deposition on natural waters: modeling uncertainty. *J. Geophys. Res.* **89**, 9679–9681.
- Slinn, W. G. N. 1983. Air to sea transfer of particles. In: *Air-sea exchange of gases and particles* (ed. P. S. Liss and W. G. N. Slinn). N. A. T.O. ASI Series. Dordrecht, Holland: Reidel, 299–396.
- Slinn, S. A. and Slinn, W. G. N. 1981. Modeling of atmospheric particulate deposition to natural waters. In: *Atmospheric pollutants in natural waters* (ed. S. J. Eisenreich). Ann Arbor Science, Michigan, 23–53.
- Stein, S. W., Turpin, B. J., Cai, X. P., Huang, P. F., and McMurry, P. H. 1994. Measurements of relative humidity-dependent bounce and density for atmospheric particles using the DMA impactor technique. *Atmos. Environ.* **28**, 1739–1746.
- Tsunogai, S., Suzuki, S., T., Kurata, T. and Uematsu, M. 1985. Seasonal and areal variation of continental aerosol in the surface air over the western North Pacific region. *J. Oceanogr. Soc. Japan* **41**, 427–434.
- Uematsu, M., Duce, R. A., Prospero, J. M., Chen, L., Merrill, J. T. and McDonald, R. L. 1983. Transport of dust from Asia over the North Pacific Ocean. *J. Geophys. Res.* **88**, 5343–5352.
- Uematsu, M., Duce, R. A. and Prospero, J. M. 1985. Deposition of atmospheric mineral particles in the North Pacific Ocean. *J. Atmos. Chem.* **3**, 123–138.
- Vasconcelos, L. A. de P., Macias, E. S., and White, W. H. 1994. Aerosol composition as a function of haze and humidity levels in the southwestern U. S. *Atmos. Environ.* **28**, 3679–3691.
- Watts, I. E. M. 1969. Climates of China and Korea. In: *Climates of northern and eastern Asia*, 8 (ed. H. Arakawa). World Survey of Climatology. Elsevier, Amsterdam, 94pp.
- Zhang, J., Liu, S. M., Lu, X. and Huang, W. W. 1993. Characterizing Asian wind- dust transport to the

- northwest Pacific Ocean: Direct measurements of the dust flux for two years. *Tellus* **45B**, 335–345.
- Zhang, X. Y., An, Z. S., Chen, T., Zhang, G. Y., Arimoto, R. and Ray, B. J. 1994. Late Quaternary records of the atmospheric input of eolian dust to the center of the Chinese Loess Plateau. *Quaternary Research* **41**, 35–43.
- Zhang, X. Y., Arimoto, R., An, Z. S., Chen, T., Zhang, G. Y., Zhu, G. H. and Wang, X. F. 1993. Atmospheric trace elements over source regions for Chinese dust: Concentrations, sources and atmospheric deposition to the Loess Plateau. *Atmos. Environ.* **27A**, 2053–2067.
- Zhou, M. Y., Chen, Z., Huang, R. H., Wang, Q. M., Arimoto, R., Parungo, F., Lenschow, D., Ohada, K. and Wu, P. M. 1994. Effects of two dust storms on solar radiation in the Beijing-Tianjing area. *Geophys. Res. Lett.* **21**, 2697–2700.