

**Atmospheric transport and deposition of anthropogenic substances
from the Asia to the East China Sea**

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Abstract

Experiments conducted aboard the R/V *Hakuho Maru* (Cruise KH-02-3) in the autumn of 2002 were designed to study the pathways of atmospheric aerosol transport, selected trace elements and major ions composition of particles and quantify their dry deposition over the East China Sea. We determined the concentrations of trace elements Al, Mn, Pb, Se, and Zn and major ions Na^+ , SO_4^{2-} and NO_3^- in aerosol particles. Aerosols were continuously collected in fine and coarse size fractions with a cut off at 2.5 μm . Particle number densities (volumes) were determined in four size ranges between 0.1 and $<0.5 \mu\text{m}$. High Al concentrations in the coarse mode fractions suggest continental origin of these aerosols. Weather charts revealed conducive meteorological conditions for northeastward air mass transports from the Asian continent. Back trajectory analysis results corroborated with the meteorological observations. Nss-SO_4^{2-} , Mn, Pb, Se and Zn were more abundant in the fine than coarse mode fraction, but NO_3^- was associated with the latter. About three-fold higher Mn/Al ratio in fine mode than in coarse aerosols (0.042 vs. 0.015, respectively) and a strong correlation between non-crustal Mn and nss-SO_4^{2-} suggest that fine Mn originates from industrial sources. Similarly, non-crustal Pb and Zn in the fine fraction were again significantly correlated with nss-SO_4^{2-} indicating major contributions from anthropogenic activities. Our results highlight the fact that atmospheric pollutants from Asia, including heavy metals and major ions, are carried to the East China Sea even during non-dusty periods. It is necessary to obtain the atmospheric deposition fluxes, wet and dry, through out the year both by periodic and continuous atmospheric measurements in order to find the biogeochemical significance of Asian aerosols to the East China Sea and North Pacific Ocean.

Keywords: Atmospheric transport, Aerosols, Anthropogenic substances, Trace metals, Major ion species, East China Sea

1. Introduction

Anthropogenic substances are carried by winds from heavily polluted industrial regions in Asia to the western Pacific and beyond (Zhou et al., 1990; Uematsu et al., 1992). These atmosphere contaminants emanate from fuel combustion, industrial processes, biomass/biofuel burning, etc, and their releases are on a continuous rise. For example, in the 1990s, anthropogenic nitrogen oxide emissions from Asia surpassed those from North America and Europe, which is expected to continue for decades to come (Akimoto, 2003). The anthropogenic substances are atmospherically transported in forms of gases and particles that include secondary particles formed *via* gas-to-particle conversions of sulfur (SO₂), nitrogen (NH₃ and NO_x), and organic compounds. The transport and deposition of anthropogenic substances is important not only for biological processes in coastal and offshore regions but also for trace element geochemical processes in the Chinese marginal seas (Lin et al., 2000) and the North Pacific Ocean (Maring and Duce, 1990).

Surface waters of the East China Sea, a large marginal sea (area $\sim 1250 \times 10^3$ km²), are susceptible to the air/sea transfer and deposition of materials due to its proximity to intense natural and anthropogenic activities in Asia. Earlier studies over the Yellow Sea (Hong et al., 1998) and East China Sea (Gao et al., 1996; 1997) have reported concentrations of substances in bulk aerosol samples but little or no information is provided on their distribution in different size-groups of particles. Zhou et al. (1990) used a cascade impactor to collect size separated aerosols over the western Pacific with a focus only on trace elements. In the present attempt, we studied both trace elements (Al, Mn, Pb, Se and Zn) and major ions (Na⁺, SO₄²⁻ and NO₃⁻) in size-fractionated aerosols along with the chemical associations between these two groups. The results were used to evaluate the transport of anthropogenic substances to the East China Sea and their deposition during September and early October when dust concentrations are normally low.

2. Sampling and analytical methods

2.1. Study area and sample collection

The area lying within 124°E -132°E and 26°N-34°N in the East China Sea was studied from 26 September to 9 October 2002. The experiments were conducted on board the vessel R/V *Hakuho Maru* during its leg 2 of cruise KH-02-3 (Fig. 1). The cruise originated in Kagoshima and ended at Hakata Kyushu Island in Japan during which a total of 27 aerosol filter samples was collected. The atmospheric particles were continuously collected using a high-volume virtual dichotomous air sampler (Kimoto Electric, Co., Ltd., Model AS-9) that was operated at a mean flow rate of 13 m³ h⁻¹. The sampler was mounted on the upper deck of the ship, 13 m above the sea surface. The sampling duration was about 12 h that amounted to a mean filtered air volume of ~140 m³. The virtual impactor separated coarse (diameter, d > 2.5 μm) and fine (d < 2.5 μm) particles, wherein both the fractions were collected on a single 90 mm diameter Teflon™ (Advantec, PF040 PTFE). The collection efficiency was 95% for 0.3 μm sized particles.

We also determined the number densities of particles in four size (diameter) ranges: 0.1 μm < d < 0.15 μm, 0.15 μm < d < 0.2 μm, 0.2 μm < d < 0.3 μm, and 0.3 μm < d < 0.5 μm). The measurements were made for every 5 min using an optical particle counter (OPC, Rion Co., Ltd., Model KC18).

2.2. Chemical methods

The aerosol samples were digested using a mixture of strong acids (HNO₃ + HF + H₂O₂) following the procedure of Minakawa and Uematsu (2001). The concentrations of trace metals Al, Mn, Pb, and Zn in aerosols were determined using an inductively coupled plasma atomic emission spectrometer (ICP-AES of Jarell Ash Co., Ltd., Model IRIS Advantage ICAP). The detection limits were found to be: 54 ng mL⁻¹ for Al, 0.2 ng mL⁻¹ for Mn, 4 ng mL⁻¹ for Pb, and 8 ng mL⁻¹ for Zn. The analytical errors were within 10%. The Se concentrations were determined by the DAN-fluorometric technique (Nakaguchi et al., 1985) in combination with high-performance liquid chromatography (Hattori et al., 2002). The Se detection limit was 0.1 fg mL⁻¹. The errors associated with Se analysis are within ~10%.

Water-soluble ions (Na⁺, SO₄²⁻ and NO₃⁻) in aerosol particles were extracted into high-purity water (Milli-Q) and their concentrations immediately determined by ion chromatography (Sasakawa et al., 2003) through the use of DX-120 system (Dionex

Corp., Sunnyvale, CA). The detection limits were $0.01 \mu\text{g mL}^{-1}$ for Na^+ , $0.02 \mu\text{g mL}^{-1}$ for NO_3^- , and $0.03 \mu\text{g mL}^{-1}$ for SO_4^{2-} . The amount of non-sea-salt SO_4^{2-} (nss- SO_4^{2-}) in aerosols, the sulfate fraction in excess over that expected from sea salt, was calculated by using the equation:

$$[\text{nss-SO}_4^{2-}] = [\text{total SO}_4^{2-}]_m - ([\text{SO}_4^{2-}]/[\text{Na}^+]_{\text{seawater}}) \cdot [\text{Na}^+]_m$$

Here, a mass weight ratio (w/w) of 0.251 was used for $([\text{SO}_4^{2-}]/[\text{Na}^+]_{\text{seawater}})$ and the subscript m denotes the quantities measured in the aerosol samples.

The non-crustal (nc) concentrations of the trace elements were calculated as follows:

$$[\text{TE}]_{\text{nc}} = [\text{TE}]_m - (\text{TE}/\text{Al})_{\text{crust}} [\text{Al}]_m$$

where $[\text{TE}]_{\text{nc}}$ is the non-crustal trace element (TE) concentration, $[\text{TE}]_m$ and $[\text{Al}]_m$ are measured TE and Al concentrations, and $(\text{TE}/\text{Al})_{\text{crust}}$ is the ratio in the crust (Taylor and McLennan, 1985).

2.3. Air mass trajectories

Isentropic trajectories extending 4-days backwards in time were calculated using the hybrid single-particle Lagrangian integrated trajectory (HYSPLIT 4) model (NOAA Air Resources Laboratory, <http://www.arl.noaa.gov/ready/hysplit4.html>). At the three different altitudes, 500, 1000, and 1500 m, trajectories were obtained to identify the origin and flow of typical air masses.

3. Results and Discussion

3.1. Particle numbers and weather conditions

High aerosol loadings at certain locations during the cruise were found in the size range 0.1 to $< 0.5 \mu\text{m}$ (aerodynamic equivalent diameter, Fig. 2). Primary aerosol particles formed from terrestrial or seawater sources typically are larger than secondary

aerosols formed *via* gas-to-particle conversion. Therefore, during the events of higher aerosol loading larger volumes of smaller-sized particles (Fig. 2) provide evidence for the prevalence of secondary aerosols than in the first few days of the cruise.

The pressure fields on weather charts (Fig. 3) suggest that winds favoring the aerosol transport from Asia to the North Pacific Ocean occurred to the north of a cold front and the northwestern parts of the present cruise tracks. Back trajectory analyses suggest that on the days of high particle number densities (volumes) the air masses had passed over the continent within the past 24 hrs. Furthermore, the calculated air-mass back trajectories agreed well with the airflows expected from the pressure fields. Figures 2 and 3 thus suggest that we sampled continental air containing large volumes of small sized secondary particles of likely anthropogenic origin. The results of chemical analyses of collected aerosols, specifically those of the events labeled (a) to (d) in Figs. 1-3, are studied in detail (Fig. 4-7) to understand processes governing the composition and origins of these particles, with particular reference to anthropogenic perturbations.

3.2. Variations in Al and crustal components

Aluminum is commonly used as a chemical tracer for mineral dust. High concentration of Al in coarse ($>2.5 \mu\text{m}$) particles (Fig. 4(b)) indicates that dust from soil/crustal sources was one of the aerosol components that contributed to the high aerosol loading events. The air over the East China Sea is thus strongly influenced by air transported from dust-producing areas of the continent even in autumn when dust storms are infrequent. The fact that the Al concentrations in fine particles were $\sim 100 \text{ ng m}^{-3}$ during the events shows that considerable quantities of these aerosols also originated from the land. The arithmetic mean of total Al concentration over the East China Sea during the cruise, off-dust season, was 820 ng m^{-3} . The highest Al concentration found was $1,840 \text{ ng m}^{-3}$, which is considerably lower than that observed over the Yellow Sea in spring (Hong et al., 1998) and the East China Sea (Gao et al., 1997) but much higher than that observed over islands in the remote North Pacific ($< 100 \text{ ng m}^{-3}$) at the same time of year (Uematsu et al., 1983). Coarse fraction Al contributed 75–82% to the total Al, which is higher than that of 59–69% found over the Kagoshima-Okinawa region or the 30–60% over the Central North Pacific (Uematsu et

al., 2002). Hence, the clay mineral fraction transported from Asia appears to be significant in aerosols over the East China Sea.

3.3. Variation in non-crustal trace elements

Fig. 4 shows that concentrations of Pb, Se and Zn, the three TEs with strong continental, but not crustal, sources peaked during the high aerosol loading events coinciding with nss-SO_4^{2-} and NO_3^- (see Table 1). In contrast to that for Al, the fine mode aerosols accounted for more than half (i.e., 65-76%) of the total Pb, Se and Zn mass concentrations (Fig. 4). On the other hand, Mn distribution was somewhat different because ~40% of its mass was in fine particles, which is intermediate in behavior to Al and the other three non-crustal TEs (Pb, Se and Zn).

The variations in Pb, Se and Zn observed during the cruise suggest a spatial difference in the strength of the continental influence on aerosol composition over the East China Sea. Aerosols over the northwestern region generally exhibited higher abundances of TEs. The highest concentrations were observed in samples of air masses that passed over the industrial area near Shanghai (e.g., October 4, see Fig. 3 (c)); which is one of the largest cities in China, with a population of over 15 million. Anthropogenic aerosols are mainly released, in this region, from motor vehicles and industries (Shu et al., 2001; Yao et al., 2002; Ye et al., 2003). This trend is consistent with the trajectories shown in Fig. 3. In contrast, aerosol samples from the southeastern region of the study area contained the lowest concentrations of Pb, Zn and Se, especially the first two, in both the size fractions. During the sampling in this region, near Kagoshima and areas around Okinawa, the collected aerosols were from air masses that were transported from the western North Pacific Ocean by easterly winds (Fig. 3).

Higher levels of TEs in the fine mode aerosol could result either from direct emissions in this mode or transformations (adsorption/condensation) of precursor substances during their transport through the atmosphere. Emissions of Pb, Se and Zn in fine mode occur mainly from anthropogenic activities, some involving processes at high temperatures (Arimoto et al., 1989).

3.4. Relationship between trace elements and major anions

Although both nss-SO_4^{2-} and NO_3^- in the atmosphere are primarily derived from industrial activities their partitioning is different in the two size groups of aerosols. Nss-SO_4^{2-} was dominant in the fine fraction while NO_3^- is more in the coarse mode (Fig. 4). The nss-SO_4^{2-} in fine mode particles is formed from the oxidation of SO_2 released by human activities or from conversion of biogenically emitted reduced sulfur gases (e.g., dimethylsulfide). The abundance pattern of nss-SO_4^{2-} in small sized aerosols is similar to the trends observed for pollutant metals, mainly Pb, Se and Zn (Fig. 4) suggesting the significant anthropogenic influence on the aerosol composition over the East China Sea. It is interesting to note that the production mechanisms of nss-SO_4^{2-} , which forms from pollutant gas-to-particle conversion, and the trace metals, which are expected to be released as particulates during the high temperature combustion processes, are different. Calculated correlations (Table 2) provided statistical support for an anthropogenic origin for these substances in aerosols, even if sources are not the same.

On the other hand, the anthropogenic NO_3^- is present in higher concentrations in coarse than fine particles facilitated by its reactions with Na^+ and Ca^{2+} while substituting for Cl^- and CO_3^{2-} , respectively. This reasoning is supported by the positive linear correlation observed between Ca^{2+} and NO_3^- and implying that HNO_3 reacted with some compounds in the coarse mode dust particles (Nakamura et al., 2005). In addition, Cl^- deficiencies in coarse aerosols have been reported in a parallel study carried out during the same cruise (Nakamura et al., 2005).

3.5. Behavior of the non-crustal trace element fraction

To evaluate the extent of trace element pollution over the East China Sea we calculated mass ratios between TE and Al. A TE enrichment in aerosol particles relative to its crustal abundance is arguably an indicator of the level of anthropogenic perturbation. While many samples with high TE concentrations expectedly did have high TE/Al ratios, it is interesting to note high ratios in some samples, from the southeastern legs of the cruise, with low TE concentrations (Table 3). Large TE/Al ratios in the latter samples mainly resulted from very low Al (dust) concentrations in the air transported from the open ocean. Commonly, enrichments are quantified relative to a reference element of the crust. We calculated enrichment factors ($\text{EF} = [\text{TE}/\text{Al}]_{\text{m}} / [\text{TE}/\text{Al}]_{\text{reference}}$) using the Al and TE data for average crustal rock (Taylor and McLennan,

1985) and presently measured concentrations in aerosols. In the fine fraction, the EFs were $\sim 1,200$ for Pb, $\sim 33,000$ for Se, and ~ 400 for Zn. The EFs for the coarse fractions of these TEs were much lower, ~ 40 (Pb), $\sim 1,000$ (Se), and ~ 30 (Zn). The evaluated EFs reveal that these metals in both the coarse and fine fractions were of non-crustal origin.

The behavior of Mn is different from the other TEs as it was less concentrated than Pb or Zn, but more than Al (Table 3, Fig. 5). The slopes obtained for Mn/Al in two size fractions were clearly different, with the ratio three times higher for the fine mode particles (slope = 0.042) than for the coarse mode (slope = 0.015). This we believe as evidence for Mn enrichment in fine particles resulting from anthropogenic emissions; most likely from ore processing and other metallurgical activities (Mielke et al., 2002).

The spatial distributions of the non-crustal fractions of metals (Fig. 6 shown for Pb and Zn as examples) are in concurrence with the transportation pathways of atmospheric pollutants in the northwest. Furthermore, fine fraction Pb_{nc} , Se_{nc} and Zn_{nc} components showed significant correlations with $nss-SO_4^{2-}$, with r^2 values of 0.76, 0.63 and 0.85, respectively (Fig. 7), suggesting once again that these substances have similar anthropogenic origins. Manganese also showed significant correlation with $nss-SO_4^{2-}$ implying that the excess Mn, i.e., the Mn over that expected from mineral dust, may originate from sources that also emit SO_2 .

3.6. Dry deposition fluxes of particulate trace elements

We estimated the dry deposition flux of particulate trace elements in order to understand the effect of the atmospheric trace metal supply to the East China Sea (Table 4). The atmospheric dry deposition flux of TE (F) is defined as:

$$F = V_d C_a$$

where V_d is the dry deposition velocity and C_a is the atmospheric aerosol concentration. Based on experimental and model studies of Duce et al. (1991) we estimated 0.1 cm s^{-1} to be the dry deposition velocity for fine mode aerosols and 2 cm s^{-1} as that for coarse mode particle deposition (Yeatman et al., 2001). The contribution of coarse mode aerosol dry deposition was more than 80% of total deposition for TEs and NO_3^- , and

70% of that for nss-SO₄²⁻ (Table 4). Coarse mode Pb and nss-SO₄²⁻ contents represented relatively small fractions (less than 19%) but their percent deposition fluxes were much higher than that of the fine mode. This comparison suggests that measurement of coarse mode aerosol is quite important in the estimation of atmospheric deposition flux. Longer residence time of TEs in fine mode aerosols facilitates their further transport to the open Pacific (Duce et al., 1983) and or beyond. Direct measurements of the wet deposition fluxes are essential to obtain the total deposition fluxes. We recommend a long-term continuous sampling at fixed sites because of episodic removal of aerosols by precipitation. This scavenging mechanism makes wet deposition more significant than the dry deposition flux (Uematsu et al., 1985). Recent model estimates suggested that precipitation scavenging of dust particles accounts for ~60% of the total deposition over the open ocean (Gao et al., 2003).

4. Conclusions

We studied trace elements Al, Mn, Pb, Se, and Zn and major ions Na⁺, SO₄²⁻ and NO₃⁻ in the atmosphere of the East China Sea through continuous collection of aerosols, in fine and coarse size fractions together with particle number densities in the autumn of 2002. Higher Al concentrations in coarse fraction suggest land origin of these aerosols carried by air masses to the ocean. Nss-SO₄²⁻, Mn, Pb, Se and Zn occurred in higher concentrations in the fine fraction while NO₃⁻ was largely in coarse particles. A high Mn/Al ratio in fine aerosols over that in coarse particles and the non-crustal-Mn relation with nss-SO₄²⁻ suggest an industrial origin for Mn in fine mode aerosols. The non-crustal components of Pb, Se and Zn in fine fraction are also significantly correlated with nss-SO₄²⁻ revealing their origin due to anthropogenic activities. Atmospheric pollutants including trace metals from the Asian continent could be carried to the East China Sea even during non-dust periods, in which wet deposition could be significant.

Atmospheric deposition of these aerosol substances, after coagulation or otherwise, could have significant influence on biogeochemical cycles as dissolution of bio-essential substances such as NO₃⁻, Se (Vandermeulen and Foda, 1988) and Zn (Bruland et al., 1991) from aerosols can occur in surface waters and alter productivity patterns. Especially during the dusty periods, the deposited dust particles may act as

ballast facilitating the faster scavenging of organic materials from the surface to deep ocean (Armstrong et al., 2002) and carbon draw down in the East China Sea.

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Figure Captions

Fig. 1. Ship track followed during the cruise KH-02-3 of R/V *Hakuho Maru* in the East China Sea. Locations identified by closed circles were starting points of aerosol collections, with a sampling time of 12 hours for each filter. Positions of open circles for (a) Sept. 28, (b) Sept. 30, (c) Oct. 4, and (d) Oct. 8) were starting points for back air trajectories computations.

Fig. 2. Variations in particle number densities (by volume), along the cruise track for aerosols in four size groups, plotted against sampled dates. Bars (a) to (d) correspond to starting dates for air mass back trajectories (see Fig. 1).

Fig. 3. Computed air mass back trajectories (panels on the far left) and relevant weather charts (panels on the right) for four days identified in Figs. 1 and 2. The low pressure systems from China moved to the Northwest Pacific Ocean via the northern parts of Japan.

Fig. 4. Variations in Al, Pb, Se, Zn, nss-SO_4^{2-} , and NO_3^- concentrations in fine and coarse mode aerosols over the East China Sea. Bars are as detailed in Fig. 2.

Fig. 5. Relations between Mn and Al in fine and coarse mode aerosols collected over the East China sea. Regression equations and lines indicate Mn enrichment in fine mode particles than in coarser fraction..

Fig. 6. Distribution of non-crustal fractions of Pb and Zn in fine mode aerosols over the East China Sea.

Fig. 7. The relations between nss-SO_4^{2-} and non-crustal components of Pb, Mn, Se and Zn in the fine fraction of aerosols collected over the East China Sea.

Table 1
Average concentrations of elements and major anions over the Pacific Ocean and East China Sea during the cruise of the autumn 2002 (unit: ng m⁻³)

	Pacific Ocean* (n=11)			East China Sea* (n=16)			Overall (n=27)		
	fine (<2.5 µm)	coarse (>2.5 µm)	Total	fine (<2.5 µm)	coarse (>2.5 µm)	Total	fine (<2.5 µm)	coarse (>2.5 µm)	
Al	15 (N.D. - 44)	54 (N.D. - 270)	70 (8 - 310)	140 (13 - 380)	730 (62 - 1,560)	820 (74 - 1,840)	87 (N.D. - 380)	420 (N.D. - 1,560)	500 (8 - 1,840)
Mn	0.7 (N.D. - 2.4)	1.4 (0.1 - 4.3)	2.1 (0.1 - 6.4)	7 (0.8 - 15)	12 (1.2 - 27)	19 (2.4 - 42)	4.4 (N.D. - 15)	7.6 (0.1 - 27)	12 (0.1 - 42)
Pb	1.8 (N.D. - 5.4)	0.8 (N.D. - 2.8)	2.6 (0.3 - 8.2)	33 (4.5 - 120)	7.7 (0.9 - 16)	41 (5.4 - 130)	20 (N.D. - 120)	4.9 (N.D. - 16)	25 (0.3 - 130)
Se	0.29 (0.06 - 0.72)	0.08 (0.02 - 0.20)	0.37 (0.11 - 0.92)	1.5 (0.24 - 3.7)	0.45 (0.08 - 0.85)	1.9 (0.49 - 4.5)	0.99 (0.06 - 3.7)	0.30 (0.02 - 0.85)	1.3 (0.11 - 4.5)
Zn	4 (N.D. - 17)	3 (N.D. - 14)	7 (0 - 18)	40 (4 - 110)	17 (2 - 44)	57 (6 - 160)	26 (N.D. - 110)	11 (N.D. - 44)	37 (0 - 160)
nss-SO ₄ ²⁻	2,780 (860 - 4,900)	310 (92 - 620)	3,090 (960 - 5,400)	11,300 (2,150 - 21,700)	1,340 (290 - 2,420)	12,600 (2,590 - 23,800)	7,820 (860 - 21,700)	920 (92 - 2,420)	8,740 (960 - 23,800)
NO ₃ ⁻	140 (44 - 350)	730 (105 - 1,230)	870 (150 - 1,580)	310 (120 - 490)	2,070 (260 - 4,300)	2,380 (420 - 4,670)	240 (44 - 490)	1,530 (105 - 4,320)	1,770 (148 - 4,670)

Table 2

Correlation (r^2) matrix for elements and major anions in fine mode aerosols for all data

Element	Al	Mn	Pb	Se	Zn	nss-SO ₄ ²⁻	NO ₃ ⁻
Al	1.00						
Mn	0.68	1.00					
Pb	0.28	0.61	1.00				
Se	0.35	0.46	0.64	1.00			
Zn	0.33	0.74	0.94	0.65	1.00		
nss-SO ₄ ²⁻	0.43	0.76	0.76	0.65	0.85	1.00	
NO ₃ ⁻	0.47	0.54	0.34	0.42	0.44	0.41	1.00

Table 3

Average metal/Al ratios in fine mode aerosols over the Pacific Ocean and East China Sea

(x10 ⁻³)	Pacific Ocean	East China Sea	Overall	Earth crustal ratio
Mn/Al	50±28 (n=8)	55±25 (n=16)	54±26 (n=24)	7.5
Pb/Al	295±204 (n=9)	287±288 (n=16)	290±261 (n=25)	0.25
Se/Al	28±17 (n=10)	14±11 (n=16)	20±15 (n=26)	0.0006
Zn/Al	429±264 (n=10)	350±294 (n=16)	380±285 (n=26)	0.88

Earth crustal ratio based on Taylor and McLennan (1985).

Table 4

Dry deposition flux of TEs calculated for fine and coarse fractions of aerosols over East China Sea

($\mu\text{g m}^{-2} \text{ day}^{-1}$)	Fine	Coarse	Total
Al	12	1250	1260
Mn	0.6	20	21
Pb	2.8	13	16
Se	0.1	0.8	0.9
Zn	3.5	29	33
nss-SO ₄ ²⁻	980	2300	3300
NO ₃ ⁻	27	3600	3630

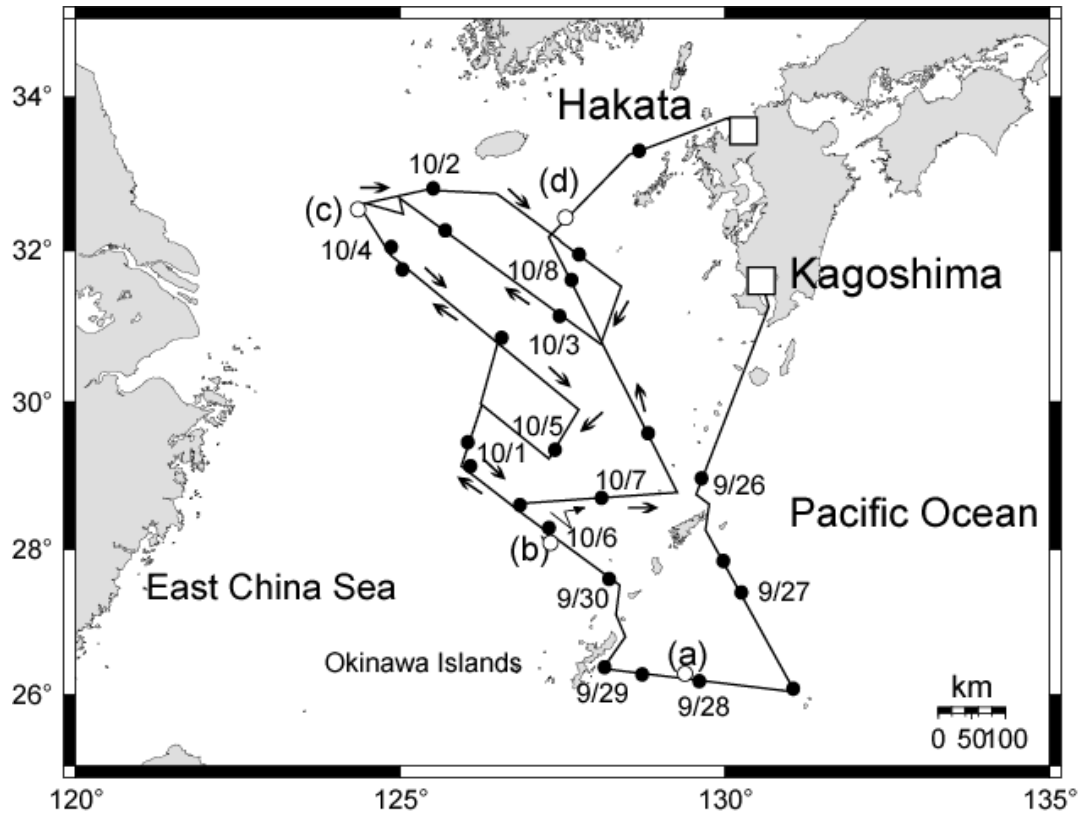


Fig. 1

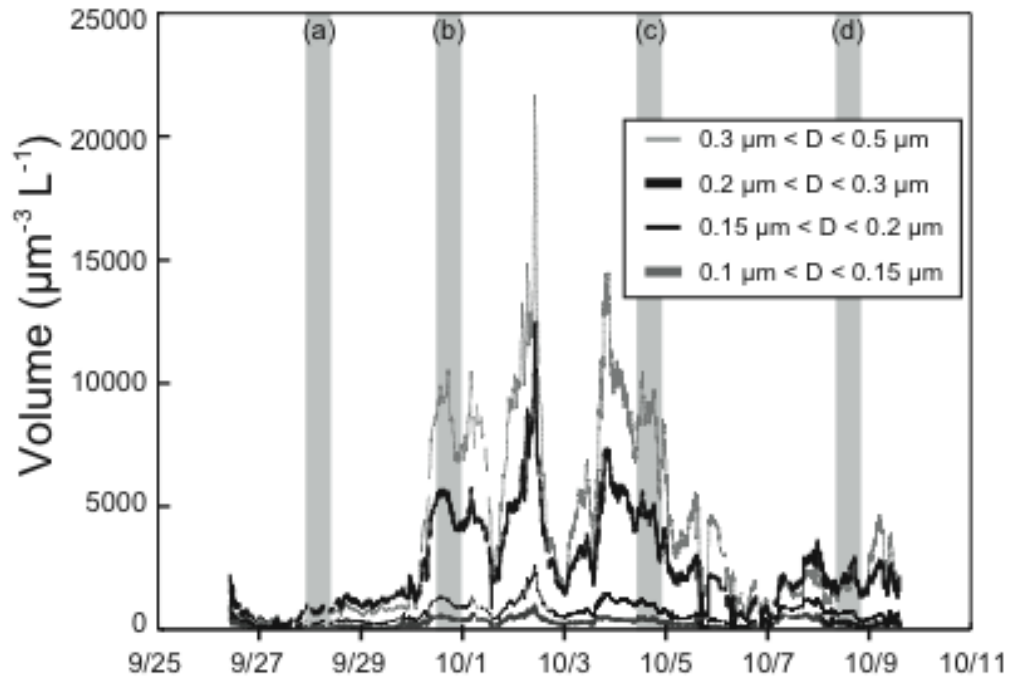


Fig. 2

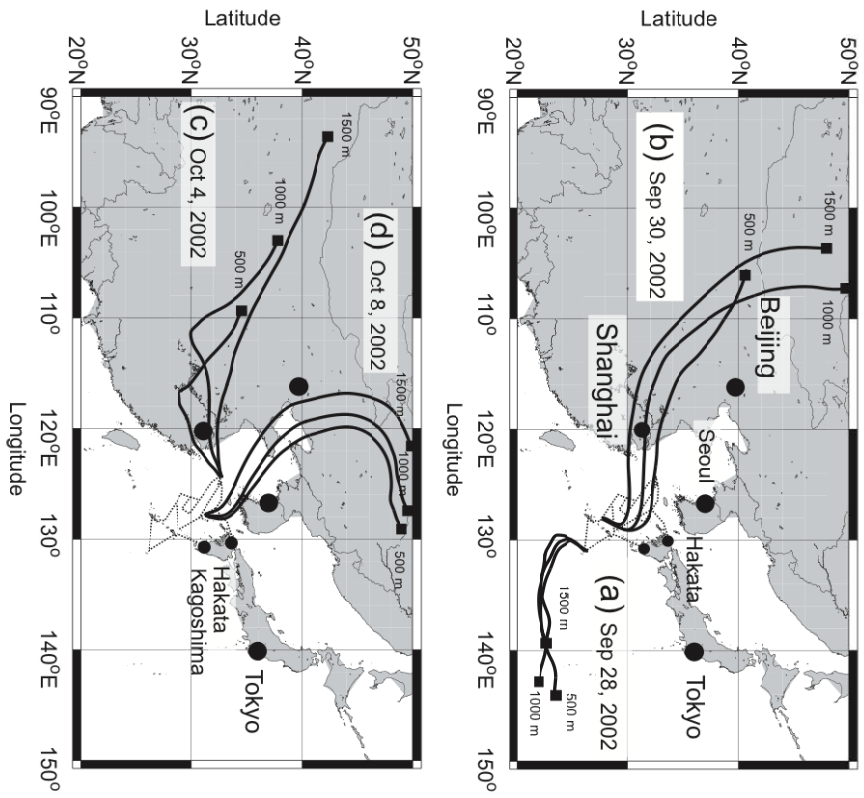
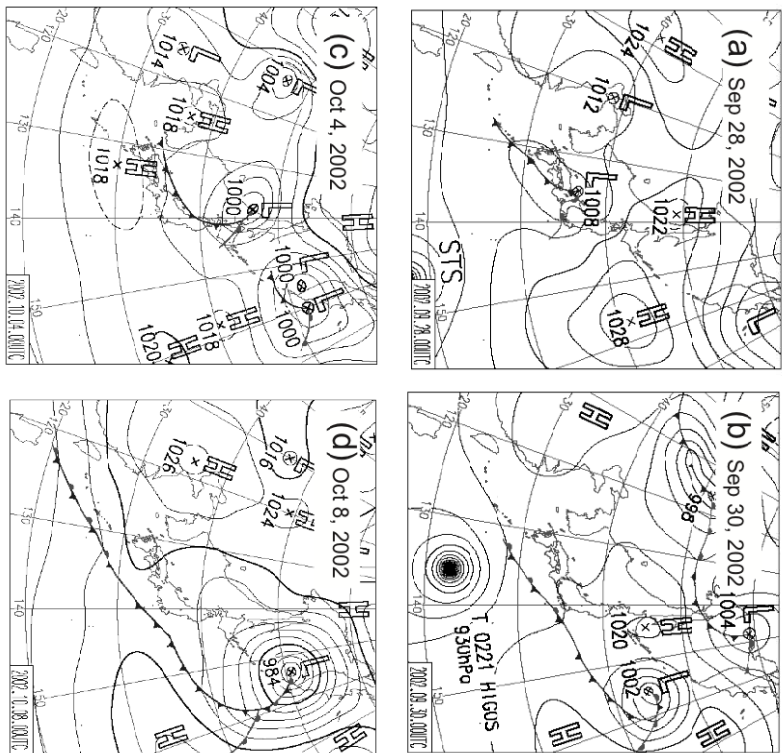


Fig. 3



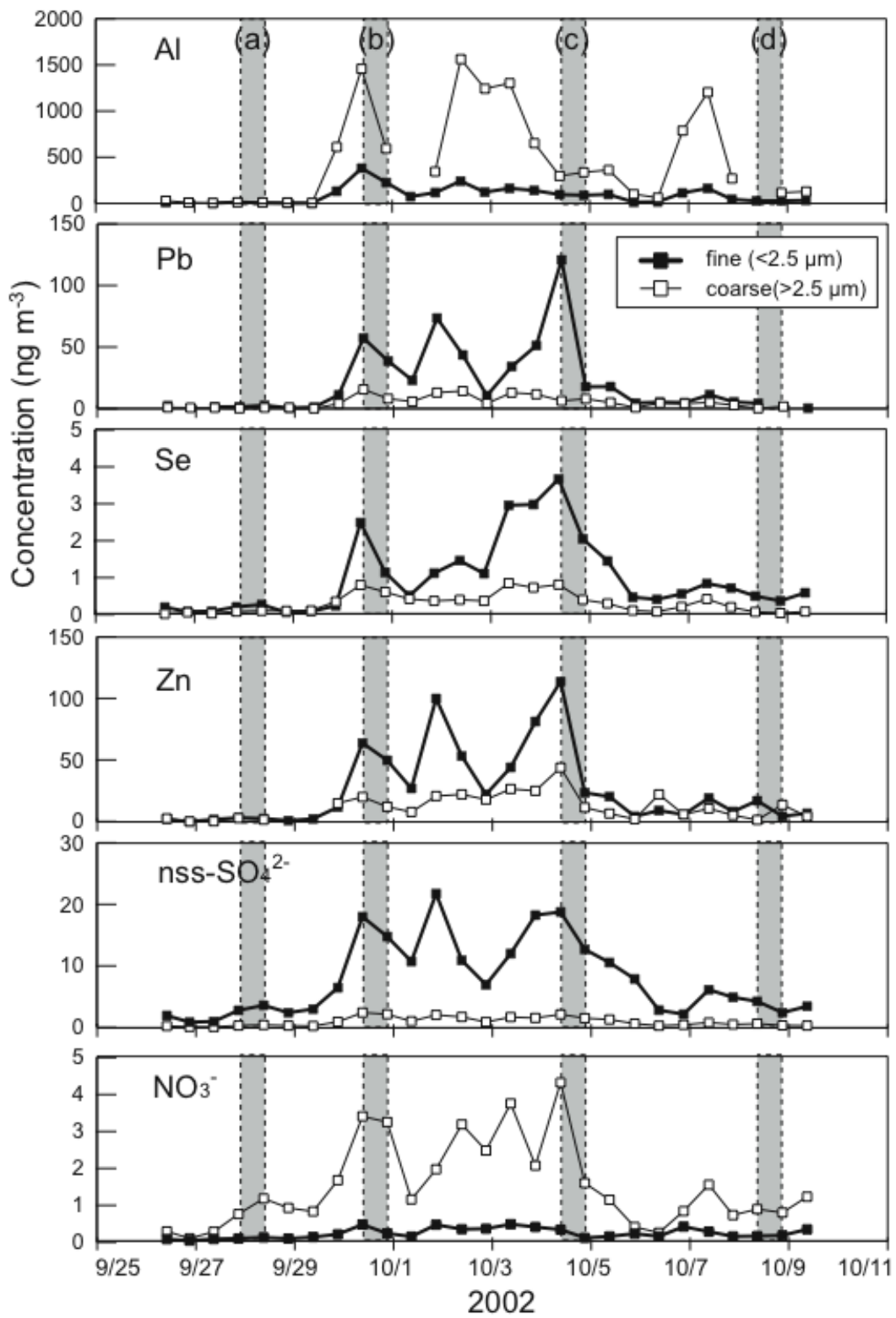


Fig. 4

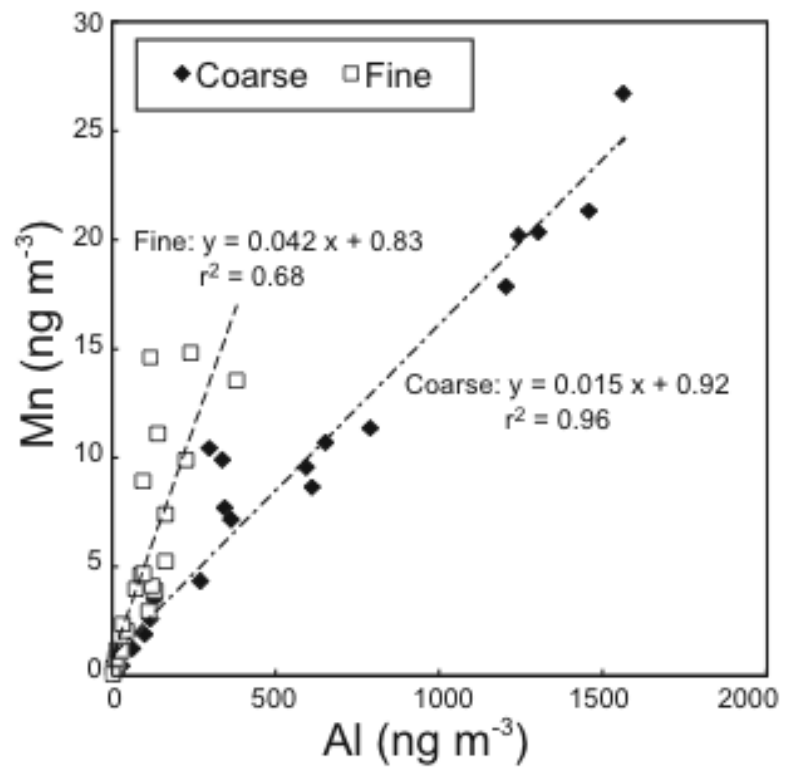


Fig. 5

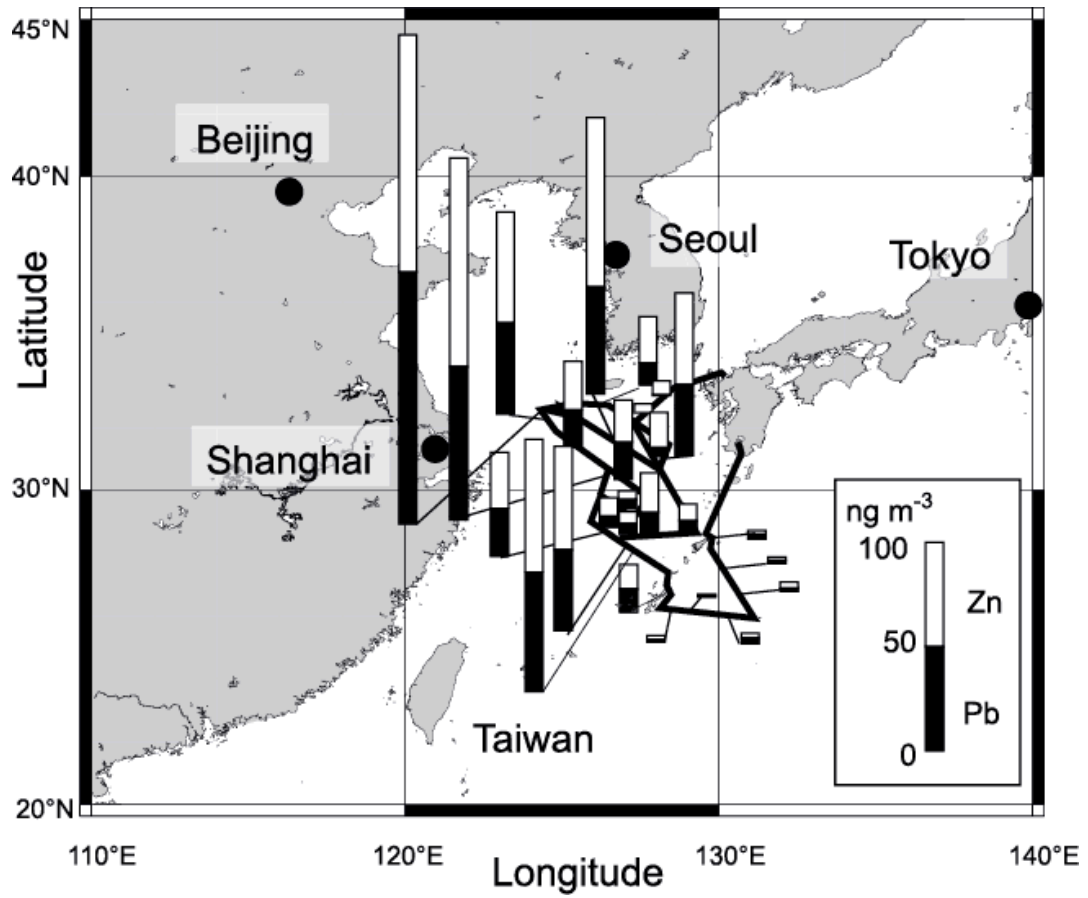


Fig. 6

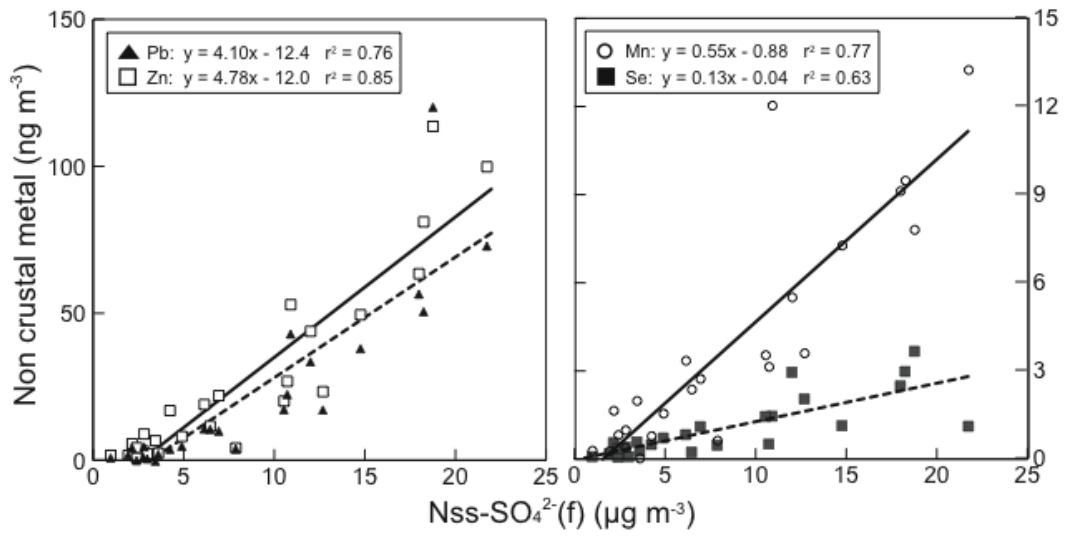


Fig. 7