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Transport of atmospheric trace elements and dust-soil to the Western Pacific Ocean

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Abstract—— Atmospheric aerosol and rainwater samples collected in the different Western Pacific area were analyzed by instrumental neutron activation and proton induced X-ray emission to (1) determine the atmospheric concentrations of trace elements over the Western Pacific and (2) to estimate the atmospheric deposition of trace elements and dust-soil material to this region.

High abundance of pollutant and crustal elements relative to oceanic sources was observed. Some characteristics of marine atmosphere relating to long-range transport of crustal and anthropogenic elements from continent to the remote ocean are discussed. The total dust-soil particle mass is estimated to be 0.066—1.2 $\mu g/m^3$ over the Western Pacific Ocean areas. Atmospheric inputs of dust-soil particles control the marine particle concentrations of crustal elements.

In addition, the characteristics of marine rain water are discussed.

Keywords: dust-soil; marine rain-water; atmospheric trace elements.

1 Introduction

Atmospheric aerosol can be transported for long-range by the atmospheric motion, Chinese geographical position is located in the westerlies, so aerosol over China can be transported eastwards to the Pacific Ocean. Therefore, it is one of important causes for marine environment pollution and marine sedimentation.

The role of atmospheric transport and deposition in the biogeochemical cycling of trace elements has been recognized only in the past 20—30 years. Dust-soil aerosol particle studies are important from a chemical standpoint because certain trace elements associated with dust-soil particles, especially iron, and several enriched elements i.e. copper and manganese, are important as catalysts for atmospheric oxidation-reduction reactions. Dust-soil particles react with acids, and they often exist as internal mixtures with atmospheric seasalts, sulfaterich particles, or the other types of aerosols.

The objectives of studying the chemical composition of aerosols over the oceans are to asses the effect of long-range transport of natural and anthropogenic trace substances on the composition of marine aerosols, and to estimate the deposition of crustal material and air pol-

lutants onto the ocean surface and their contribution to marine sediments, and to offer the background concentrations of trace elements in aerosol particles over remote marine regions.

2 Sampling methods and analytical techniques

2.1 Sampling

Aerosol samples were collected on a platform on the Chinese research vessel at an elevation of about 20m above the sea surface. A KB-120 air sampler and anderson cascade impactors were used, the KB-120 sampler was used to measure the total amount of aerosols, the anderson cascade impactor collected samples in different size ranges. The sampling times and collection speeds were 8 hours and 90 L/min for KB-120 sampler; 48h and 2 L/min for the cascade impactor.

Sample changing in the field was conducted with great care to minimize contamination. A clean area was prepared in advance for loading and unloading the filters. When one sampling interval was completed, the holder with the filter was put in a clean plastic bag and brought back to a clean area for unloading. Sample filters were sealed in small clean polyethylene bags and stored in a freezer until analysis.

2.2 Analytical techniques

Instrumental neutron activation analysis (INAA) (Yang, 1985) and proton induced X-ray emission (PIXE) (Hu, 1987) techniques were used to determine the concentrations of 46 elements in the atmospheric aerosol particle samples. Some elements analyzed by INAA and PIXE overlap each other and the two methods are mutually complementary.

Quality assurance is routinely carried out in our laboratory by concurrent analysis of suitable standard reference materials, namely SRM-1648 (airborne particulate matter), SRM-1632a (trace elements in coal), and SRM-1633a (trace elements in coal fly ash).

3 Results and discussion

3.1 Characteristics of enrichment factor for western pacific aerosol

The principal nature-sources for most marine atmospheric trace elements are the oceans and earth's crust. The crustal and marine enrichment factors for selected elements are given in Table 1.

Table 1 Enrichment factors calculated relative to crust and seawater for aerosol samples

	Locality											
	119.7°E	13.5°N	134.2°E	3°N	154.5°E	2°S	155°E	2°S	155.5°E	2.1°S	156°E	2°S
	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}
Al	1	35000	1	30000	1	220000	1	200000	1	130000	1	120000
As	810	420	—	—	—	—	—	—	450	1200	280	480
Br	58000	0.98	75000	1.1	12000	1.3	12000	1.2	23000	1.4	17000	0.95
Ca	24	0.94	15	0.51	2.3	0.56	1.7	0.40	8.3	1.2	5.8	0.80

Table 1 (continued)

Cl	310000	0.93	360000	0.91	53000	0.98	54000	0.92	88000	0.93	210000	0.89
Co	4.3	1800	—	—	4.4	11000	3.5	7000	3.8	6000	5.0	7200
Cu	430	170000	260	87000	1400	340000	110	250000	370	530000	330	440000
Fe	3.7	41000	2.2	20000	2.3	150000	8.1	500000	3.7	150000	10	370000
I	—	—	—	—	37000	850	29000	600	44000	570	28000	340
K	24	0.73	20	0.51	6.8	1.2	22	3.5	18	1.8	14	1.4
La	6.2	27000	—	—	0.97	26000	0.89	21000	1.4	21000	—	—
Mg	140	0.90	240	1.3	43	1.8	44	1.6	70	1.5	61	1.3
Mn	6.7	46000	—	—	1.2	54000	4.6	180000	2.0	49000	1.3	89000
Na	890	1	1000	1	140	1	160	1	250	1	270	1
Pb	1400	800000	350	170000	—	—	1000	340000	630	130000	470	88000
S	19000	2.4	17000	1.8	4700	3.6	9900	7.0	16000	7.0	20000	8.5
Sb	2000	210	790	180	580	950	1300	2000	940	880	1000	890
Se	15000	8000	16000	7500	8100	27000	15000	47000	18000	35000	14000	26000
Si	3.6	140	7.9	260	1.6	390	3.3	740	8.4	1200	8.0	1000
Zn	420	4200	370	3100	540	33000	1300	75000	310	11000	160	5300

Enrichment factors have shown that many trace elements exhibit atmospheric concentration ratios similar to those in average crustal material or in bulk seawater. Aluminum and sodium are used as reference element for calculating crustal and seawater enrichment factors, respectively. The enrichment factors computed in our studies indicate that natural processes may dominate the atmospheric concentrations of many elements. Seasalt elements (Na, Cl, Mg, Ca, Br, and K) exhibit relative proportions in marine aerosol particles and rainwater that generally are similar to those in bulk seawater. The seasalt aerosols do not necessarily have the same relative trace element composition as bulk seawater. Iodine may be affected by fractionation, that occurs during the seawater bubbles burst process, it was enriched about 1000-fold on aerosols. The second group of trace elements (Al, Co, Fe, Mn, Si and rare earth) often display relative proportions in remote aerosol particles similar to those in average crustal material. The concentrations of these elements may be controlled by the long-range transport of dust-soil aerosol particles from the continents, because the concentrations of crustal elements in aerosols were high over the ocean area close to the Asian continent and decreased very rapidly with increasing distance from land, but the crustal enrichment factors do not obviously demonstrate that their ultimate source is either crustal or natural. The enrichment factors computed for the third group of elements (As, Cu, Pb, Sb, Se, and Zn) have shown that these elements are enriched relative to both crustal and seawater sources. It is suggested that the enrichment elements over the Western Pacific were influenced mainly by long-range transport of anthropogenic pollutants from continent. The long-range transport of atmospheric substances from Asia not only transports dust-soil but also carries pollutants,

nutrients, and a variety of other atmospheric substances to the remote atmosphere and oceans.

3.2 Characteristics of marine rainwater

During this cruise from December 1992 to March 1993, we have collected 15 rainwater samples at the Western Pacific area. The pH value of the rain samples varied very little, from 5.3 to 5.8, exhibit acidity. This imply that the acidic materials such as SO_2, NO_x etc., may affect the remote oceanic environment, those acidic materials are mainly come from continents.

In Table 2, enrichment factors showed that the concentrations of Fe, Mn, Co, La, and Si in rainwater were dominated by a crustal source. The As, Pb, S, Sb, Se, Zn, Na, Cl, K and Br are enriched relative to the crustal material, the enrichment of Na, Cl, Br, Ca, and K are due to seasalt. However, enrichment of other elements are due to anthropogenic sources. The enrichment factors for element Na relative to Al in rain were higher than those in aerosols, it is easy to understand that the enrichment factor of Na in rain samples was higher than in aerosols, because seasalt is a good cloud condensation nucleus and Na could been enriched in cloud in the process of cloud formation.

Table 2 Enrichment factors calculated relative to crust and seawater for rain samples

	Locality											
	156°E	2°S	155.6°E	2°S	145°E	2.1°S	155°E	2.2°S	154.8°E	2.1°S	154.7°E	2°S
	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}	EF_{crust}	EF_{sea}
Al	1	27000	1	69000	1	200000	1	50000	1	60000	1	49000
As	680	270	930	940	210	600	—	—	190	160	370	270
Br	10000	1.3	33000	1.1	8400	0.80	40000	0.95	33000	0.96	45000	1.0
Ca	61	1.8	36	2.7	13	2.9	24	1.4	24	1.6	150	8.2
Cl	500000	1.1	160000	0.92	58000	0.93	250000	1.0	200000	0.98	260000	1.0
Co	32	8900	19	13000	6.6	13000	1.0	2300	1.0	600	1.2	21000
Fe	1.8	15000	1.4	30000	1.5	16000	0.33	1500	0.27	1800	1.3	19000
K	96	2.0	21	1.2	22	3.4	17	0.65	16	0.78	10	0.90
La	—	—	—	—	—	—	1.2	7500	0.74	5400	3.0	41000
Mg	290	1.4	—	—	30	1.1	110	1.0	83	0.92	120	1.1
Mn	2.1	10000	—	—	1.9	73000	3.9	38000	2.3	28000	4.6	44000
Na	1200	1	460	1	760	1	630	1	520	1	650	1
Pb	3300	1400000	—	—	810	2500000	—	—	—	—	—	—
S	16000	1.5	4600	1.1	1300	0.93	4600	0.81	4200	0.90	3000	0.85
Sb	11000	2200	7200	3700	1600	2300	240	90	250	110	710	260
Se	13000	5500	19000	20000	3300	10000	2300	1800	1700	1600	3300	2400
Si	3.8	110	27	2000	1.0	230	0.85	47	1.1	75	1.0	52
Zn	2200	17000	3400	67000	1500	82000	560	7900	530	9000	1100	15000

3.3 Deposition of atmospheric trace elements and dust-soil particles in the Western Pacific Ocean

The atmospheric input of certain trace substances exceeds those from more commonly investigated sources such as rivers, sewage outfalls, dumping, etc. (Duce, 1991). However, there is a little consideration of the transport and deposition of chemical elements from continent to the adjacent waters.

The atmospheric deposition of trace elements can be estimated through the use of a total deposition velocity, V_d(cm/s). That is $F_d = C_a \cdot V_d$. Where F_d is the total deposition flux ($\mu g/(cm^2 s)$). C_a is the measured concentration of aerosol particles in air. According to Gao (Gao, 1992) calculated results, for calculation of the total deposition of trace elements, it is probably accurate to use $V_d = 4.5$ cm/s for seasalt elements, 3 cm/s for crustal elements, and 0.55 cm/s for enriched elements. Using these total deposition velocities, we calculated the total fluxes of atmospheric trace element to the Western Pacific Ocean (Table 3).

Table 3 Total fluxes of atmospheric trace elements to the Western Pacific area

Element	$\mu g/(m^2 \cdot d)$	Element	$\mu g/(m^2 \cdot d)$
Al	14—260	La	0.0034—0.60
As	0.034—0.86	Mg	0.43—14
Br	1.4—55	Mn	0.26—20
Ca	30—2700	Na	1100—120000
Cl	2400—190000	Pb	0.095—11
Co	0.0029—0.41	S	190—2900
Cu	0.52—14	Sb	0.0048—0.24
Fe	3.1—600	Se	0.043—0.43
I	0.90—2.9	Si	370—3100
K	45—3000	Zn	0.95—15

The value of the atmospheric dust-soil aerosol is estimated by analysis of the continental components of these marine aerosol particles. We assumed that the concentrations of Al in the aerosol particles reflected the dust-soil loading in the atmosphere, the crustal material is 8.13% Al by weight. During this cruise from December 1992 to March 1993, the total dust-soil particle mass is estimated to be 0.066—1.2 $\mu g/m^3$ over the Western Pacific Ocean areas. This data similar to those at Enewetak 0.05—0.68 $\mu g/m^3$ reported by Uematsu *et al.* (Uematsu, 1983).

Our studies have shown that large quantities of soil materials are transported by winds out of Asia and over the Western Pacific Ocean.

Atmospheric inputs of dust-soil particles control the marine particle concentrations of crustal elements, when the amount of dust-soil in the atmospheric over the Western Pacific changed, the amount of crustal elements in aerosol particles changed correspondingly. The dust-soil concentrations decreased with increasing distance from land over the ocean area

close to Asian land and fluctuate around its average value over the remote ocean regions. The amounts and frequencies of precipitation also varied significantly from season to season and from site to site over the Western Pacific.

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