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Lead in the southern East China Sea

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Abstract

At present, in most oceans the lead (Pb) biogeochemical cycling has been disturbed by anthropogenic Pb through atmospheric input. The Pb concentrations in the upper water positively correlate with atmospheric input fluxes of Pb. The North Pacific is affected greatly by atmospheric substances via long-range transport from eastern Asia, especially from Mainland China. Mainland China may export considerable amounts of pollutants into the seas via rivers and the atmosphere owing to its recent fast growth in industry and economy. The East China Sea lies in an important geographical position—a transit between Mainland China and the western North Pacific. However, no data are available for seawater concentrations of Pb, a representative element with anthropogenic origin. In this work seawater samples from both 5 and 30–50 m water layers of 15 stations occupied over a cyclonic eddy in the southern East China Sea were analyzed for particulate Pb (PPb) and dissolved Pb (DPb). The Mean concentration of DPb ($\sim$128 ng/l) in the southern East China Sea upper waters ($\leqslant$50 m) is approximately several times higher than those in the Pacific; the high DPb concentrations in the southern East China Sea waters correspond to much higher atmospheric supplies of Pb to the East China Sea. Thus, this study partly fills the 'data gap' of the marginal seas. Also, it indicates that the East China Sea may be considerably contaminated by deposited polluted aerosols. Spatial distributions of DPb in the surface water show a tendency of increasing concentrations with distance offshore, that depends on the magnitudes of atmospheric Pb inputs and on particle scavenging processes. In contrast to DPb, spatial distributions of PPb basically display an 'Ω'-like picture and a tendency of decreasing concentrations with distance offshore. These are related to riverine and scavenging sources and to the drive by the eddy. Additionally, the residence times of DPb in the surface water were estimated to be about 2 years, agreeing well with the reported data.

Keywords: East China Sea; Dissolved lead; Particulate lead; Biogeochemical cycle; Residence time; Scavenging

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1. Introduction

In most oceans the biogeochemical cycling of lead (Pb) has been disturbed by anthropogenic Pb since the beginning of the Industrial Revolution, and the disturbance has become more serious since the inception of consuming alkyl-leaded gasoline. The fact was recorded in sediments and corals (Patterson, 1987; Shen & Boyle, 1989; Veron, Lambert, Isley, Linet & Grousset, 1987). Pb is a particle-reactive element with a strong affinity for certain particles such as clay minerals, Mn oxide, Fe oxyhydroxide and carbonate (Li, 1981; Sturchio et al., 1997; Turekian, 1977; Whitfield & Turner, 1987). Most of the Pb discharged from rivers is removed from overlying water in estuaries or coastal seas (Elbaz-Poulichet, Holliger, Huang & Martin, 1984; Schaule & Patterson, 1981). Atmospheric transport thus becomes the major passage for many anthropogenic constituents and Pb from land to offshore seas. Certainly, the Pb fluxes into the open ocean are dominated by the atmospheric supplies (Nriagu & Pacyna, 1988; Patterson & Settle, 1987).

The western North Pacific receives a large influx of mineral particles and pollutants from eastern Asia, especially from Mainland China through long-range atmospheric transport (Duce, Arimoto, Ray, Unni & Harder, 1983; Gao, Arimoto, Zhou, Merrill & Duce, 1992). The East China Sea (ECS), one of the largest marginal seas in the world, is situated in a transit between Mainland China and the western North Pacific. Massive quantities of terrestrial materials are emitted to the atmosphere in Mainland China (which has a rapidly developing economy and industrial base) and are deposited on the ECS shelf (Gao et al., 1996, 1997). Mainland China has been suffering from serious air pollution and acid rain owing predominantly to coal combustion by nearly two thousand power plants especially in eastern China, and by most households especially in northern China (Qian & Zhang, 1998; Zhao & Sun, 1986). The regional (even global) atmospheric environment could be influenced by these large discharges of SO_2 (around 20 Mt/year at present), greenhouse gases (ranking as third largest generator of the world), and trace constituents (including Pb) (Dod et al., 1986). These pollutants could affect Chinese marginal seas (particularly the ECS) and the Pacific Ocean through the westerlies transport. It can therefore be expected that the ECS Pb system has gradually been affected by eolian inputs of Pb.

Pb concentrations in sediments have been increasing in coastal regions off Mainland China since about 1980, based on the measurements of Pb profiles and the dating of sediment cores (Huh & Chen, 1999). However, the impact on seawater composition corresponding to these deposited eolian constituents is not known. This work attempted to determine the extent of the contamination by eolian inputs from the analysis of seawater Pb. At present, no data are available for dissolved Pb (DPb) concentrations in seawater of the ECS. Flegal and Patterson (1983) have suggested that the DPb concentrations in the surface oceans correspond directly to and correlate positively with magnitudes of atmospheric fluxes of Pb into the oceans. This work could examine that suggestion by taking advantage of the ECS with its unique position, neighboring the western North Pacific.

A total of 30 seawater samples were collected from both 5 and 30–50 m water layers of 15 stations occupied over a cyclonic eddy in the southern ECS off northeastern

Taiwan, and were analyzed for both particulate Pb (PPb) and DPb. The southern ECS is a dynamically energetic marginal sea based upon its geological, physical, chemical and biological features (Chen Lee, 1995; Hsueh, Wang & Chern, 1992; Liu et al., 1992; Wong, Pai, Liu, Liu & Chen, 1991). The Kuroshio Current flows along the eastern coast of Taiwan and collides with the shoaling ECS shelf when it approaches the northeastern tip of Taiwan. As a result, it causes a variety of phenomena, one of which is the development of a cyclonic eddy. Eddies can act as an important vehicle, exchanging seawater constituents between distinct water masses, particularly coastal waters and offshore waters (Hayward & Mantyla, 1990; The Ring Group, 1981). However, the spatial distributions of metals over eddies are poorly understood (Bishop & Fleisher, 1987; Sakamoto-Arnold, Hanson, Huizenga & Kester, 1987). Determining the influences of the cyclonic eddy on the spatial distributions of both PPb and DPb in the upper water is another goal of this work.

2. Materials and analysis

Seawater samples were taken from both 5 and 30–50 m water layers of the southern ECS off northeastern Taiwan during August 1994 on board National Taiwan University's R/V *Ocean Researcher I*. They were taken along three NE–SW parallel transects containing 15 hydrographic stations (Fig. 1) and collected using 20-l acid-pre-cleaned GO-FLO bottles on a CTD rosette. Details of stations and hydrographic parameters are given in Table 1. When the rosette was recovered, seawater was transferred into 20-l PE bottles (which were rinsed twice with the seawater sample before filling) with a filling and venting closure that could prevent contamination from deck air. About 20–40 l of seawater were pressure-filtered through a pre-cleaned and pre-weighed 142-mm 0.4-μm Nuclepore polycarbonate filter using a Sartorius PTFE pressure filter holder and a Masterflex® wriggle pump. One liter filtrate was stored in acid-pre-cleaned 1-l PP bottles and acidified to $pH < 2$ with Suprapur HNO_3 (Merck, Germany) for subsequent analysis of dissolved trace metals in land-based laboratory. Particulate-laden filters were stored in plastic petri dishes. Seawater and particulate samples were kept in a refrigerator and a desiccator, respectively, until analysis.

After drying and weighing, particulate-laden filters were processed by the total digestion method using mixed acids of Suprapur HF, HNO_3 and $HClO_4$ (all from Merck, Germany). The seawater filtrates were concentrated by using Chelex-100 resin (100–200 mesh and ammonium form) columns after adjusting pH to 6.5 in our land-based laboratory. Resin columns were pre-cleaned with 2 N Suprapur HNO_3 and DDW twice before use. The elution also used 2 N Suprapur HNO_3. All treatments for the particulate and seawater samples were carefully processed on class 100 laminar flow bench.

Metals were analyzed using a Hitachi Zeeman graphite furnace atomic absorption spectrophotometer (model Z-8100) equipped with an autosampler SSC-200. Each sample solution (digestion and elution solutions) was analyzed in triplicate at least. The quality of determination was controlled by analysis of BCSS-1 standard

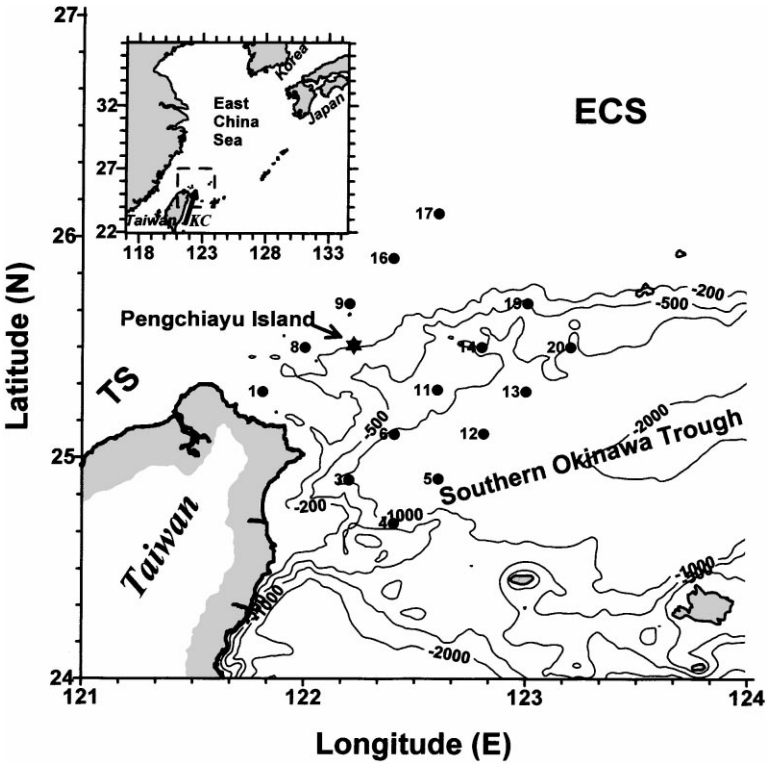

Fig. 1. Sampling locations. Inset is the regional map. The seawater collection is made along three transects: the southern East China Sea (ECS) shelf and slope and the Southern Okinawa Trough. Each transect contains five stations. An offshore island, Pengchiayu Island (shown as a star), is the collection station of marine aerosols, providing the eolian Pb fluxes used in the discussion of the study. Bathmetric map is also shown. TS, Taiwan Strait.

reference samples (National Research Council, Canada). Our Pb data (19.2 ± 0.3 μg/g, $n = 5$) fell at the lower end of certified value (22.7 ± 3.4 μg/g). By spiking a small volume of known amounts of Pb (Pb standard-Merck) into four separated batches of 1 l duplicatedly purified seawater and by subsequent pre-concentration procedures, the recovery and precision of Pb for pre-concentration were obtained to be 77 ± 3 and 4% ($n = 5$), respectively. No corrections were made on the DPb data because the recovery of 77% was good enough for our purposes and the reproducibility was excellent.

To estimate the procedure blank, 20-l seawater samples, which have been purified by passing through a Chelex 100 column twice to eliminate trace metals, were taken on board as 'blank samples'. The blank samples were processed by the same procedures, including shipboard filtration and subsequent pre-concentration for filtrate in land-based laboratory, as 'real samples'. The Pb levels in blank samples detected in this study were much lower than those in real samples by at least one order of magnitude.

Table 1
Station locations, temperature, salinity and both dissolved and particulate Pb (DPb and PPb) as well as particulate Al (PAl) and particulate Mn (PMn) concentrations in the southern East China Sea surface seawater

Station No.	Latitude (°N)	Longitude (°E)	Temperature (°C)	Salinity (psu)	DPb (ng/l)	PPb (ng/l)	PAl[a] (μg/l)	PMn[b] (ng/l)
5 m								
1	25°17.88′	121°48.67′	25.2	34.04	99	15.8	35.1	456
3	24°54.01′	122°12.04′	24.1	34.14	83	12.1	28.2	315
4	24°42.31′	122°24.21′	26.4	33.78	46	10.5	9.5	446
5	24°54.29′	122°36.25′	27.4	34.01	246	3.9	4.3	38
6	25°06.28′	122°24.30′	25.1	34.13	224	12.8	26.9	385
8	25°29.90′	122°00.05′	24.6	34.33	41	4.2	5.1	43
9	25°41.88′	122°12.04′	24.8	34.28	242	5.3	9.1	91
11	25°18.35′	122°35.93′	25.9	34.08	220	23.2	31.5	428
12	25°06.46′	122°48.45′	27.6	34.15	177	9.2	1.0	10
13	25°17.92′	122°59.87′	28.0	34.22	90	1.6	0.2	9
14	25°29.99′	122°48.09′	27.2	34.14	184	9.8	1.1	10
16	25°54.18′	122°23.92′	26.6	34.16	96	5.5	3.3	31
17	26°06.25′	122°36.17′	26.2	34.22	132	4.5	5.1	101
19	25°42.00′	123°00.26′	27.7	33.98	461	3.2	1.5	43
20	25°29.97′	123°11.93′	27.9	34.01	147	2.9	8.4	14
30–50 m								
1	25°17.88′	121°48.67′	23.0	34.04	91	12.7	32.4	345
3	24°54.01′	122°12.04′	20.9	34.44	62	14.6	48.5	429
4	24°42.31′	122°24.21′	23.0	34.57	82	6.6	6.7	48
5	24°54.29′	122°36.25′	25.1	34.58	82	8.1	4.3	32
6	25°06.28′	122°24.30′	22.8	34.37	82	38.7	39.1	504
8	25°29.90′	122°00.05′	21.5	34.60	59	5.1	9.8	116
9	25°41.88′	122°12.04′	22.2	34.66	124	5.5	9.4	90
11	25°18.35′	122°35.93′	24.2	34.30	260	18.7	19.9	339
12	25°06.46′	122°48.45′	25.6	34.49	83	4.4	0.9	9
13	25°17.92′	122°59.87′	26.8	34.38	49	4.8	3.1	83
14	25°29.99′	122°48.09′	25.2	34.64	280	8.0	1.5	16
16	25°54.18′	122°23.92′	23.1	34.84	73	4.1	4.6	46
17	26°06.25′	122°36.17′	24.7	34.61	258	3.3	1.5	19
19	25°42.00′	123°00.26′	25.3	34.57	217	2.9	0.7	13
20	25°29.97′	123°11.93′	26.8	34.35	517	4.6	2.6	13

[a] Taken from Hsu et al. (1998).
[b] Taken from Hsu (1998).

3. Results and discussion

The circulation patterns, particularly the summertime pattern, of the study region are shown in Fig. 2. The generating mechanism of the eddy has been discussed elsewhere (Tang, Hsueh, Yang & Ma, 1999), as have the circulation patterns of the study region (Hsu, Lin, Jeng & Tang, 1998).

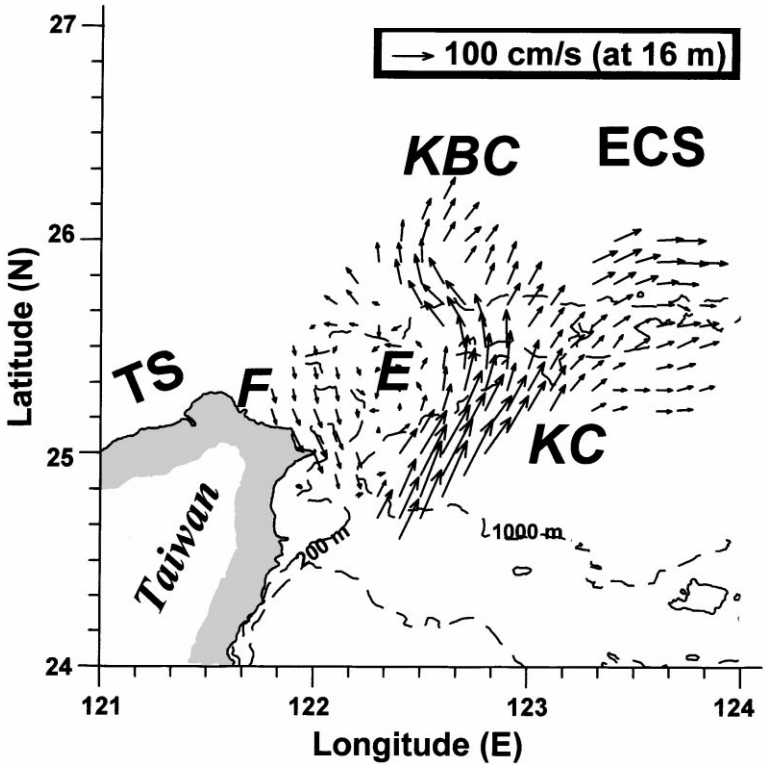

Fig. 2. Circulation pattern (current field) at 16 m depth of the southern East China Sea (ECS) off northern Taiwan, especially for summer season. It is measured by Shipboard Acoustic Doppler Current Profiler (Sb-ADCP) simultaneously on the same cruise as seawater sampling in this study (Tang et al., 1999). KC, Kuroshio Current; KBC, the Kuroshio Branch Current; E, the cyclonic eddy; F, the filament; TS, Taiwan Strait. This map is after Hsu et al. (1998) which gave a detailed review for the entire circulation pattern of the study area.

3.1. Pb concentrations

The concentrations of DPb and PPb in 30 surface seawater samples and the associated hydrographic parameters are summarized in Table 1. DPb and PPb concentrations of all samples ($n = 30$) range from 41 to 517 and 1.6 to 38.7 ng/l, and average 128 ± 117 and 6.8 ± 7.7 ng/l, respectively (Table 2). The PPb accounts for only a small fraction ($< 10\%$ on average) of total Pb (assumed to be the sum of DPb and PPb), agreeing well with earlier studies (Balls, 1985a; Helmers, 1996; Schaule & Patterson, 1981). The DPb concentrations in the southern ECS surface seawaters are comparable with those (range 7–880 ng/l, average 235 ng/l, sampling in May; range 7–800 ng/l, average 301 ng/l, sampling in October) of the coastal seawater of southeastern China reported by Gao and Zou (1998). Their sampling area (25°15′–25°45′N, 119°32′–120°10′E) is approximately 250 km away from our study area. Also, their values and ours fall in the concentration range (0.03–3.26 μg/l with a

Table 2
Means and ranges of dissolved and particulate Pb (DPb and PPb) concentrations in the southern East China Sea surface seawater

		DPb (ng/l)	PPb (ng/l)
5 m ($n=15$)	Range	41–461	1.6–23.2
	Mean	137 ± 107	6.5 ± 5.9
30–50 m ($n=15$)	Range	49–517	2.9–38.7
	Mean	119 ± 129	7.1 ± 9.3
Overall samples ($n=30$)	Range	41–517	1.6–38.7
	Mean	128 ± 117	6.8 ± 7.7

mean of 0.55 μg/l) of the northwestern Pacific Ocean surface seawater off southern Japan (28°30′–36°30′N, 129°–145°E) (Wang, Zou, Lu & Jing, 1990). It has been noted that there were much higher DPb levels (0.9–2.9 μg/l) in the Changjiang River mouth water (Yu, 1995). The DPb concentrations in the southern ECS waters, however, are higher than those of many other oceanic regions. For example, in the Pacific, the surface concentrations of DPb are usually smaller than 20 ng/l (Flegal & Patterson, 1983; Schaule & Patterson, 1981), while in the Atlantic, they fall in a range of 20–75 ng/l, averaging around 30 ng/l (Boyle, Chapnick, Shen & Bacon, 1986; Brugmann, Danielsson, Magnusson & Westerlund, 1985; Helmers, Mart, Schulz-Baldes & Ernst, 1990). Furthermore, the elevated DPb concentrations in surface waters from the study area are higher than those from the Mediterranean Sea but they have declined from 85 ng/l in 1986 to 32 ng/l in 1992 (Nicolas, Ruiz-Pino, Buat-Menard & Bethoux, 1994), and even about two times higher than those from the North Sea with an average concentration of about 60 ng/l (Balls, 1985b; Brugmann et al., 1985). Although surface concentratons of DPb are high, they are still reasonable, based on their relationships with atmospheric fluxes of Pb and the calculated DPb residence times comparable with literature data, as discussed below. Coupled with the above Pb data of the ECS, it can demonstrate that the DPb pool of the entire ECS may be considerably perturbed by anthropogenic Pb.

3.2. Spatial distributions

Spatial distributions of DPb in both 5 and 30–50 m water layers are displayed in Fig. 3; those of PPb, in Fig. 4. The concentrations of DPb tend to increase with distance from the Taiwan coast towards offshore. The distributions of PPb show an opposite pattern to those of DPb, with decreasing concentrations seaward and with an 'Ω'-like shape. The pattern is similar to that of particulate Al (PAl) (Hsu et al., 1998) and particulate Mn (PMn) (Hsu, 1998). PPb concentrations are also strongly correlated with PAl and PMn concentrations: in 5 m water layer, PPb vs. PAl $r=0.82$ and PPb vs. PMn $r=0.81$; and in 30–50 m water layer, PPb vs. PAl $r=0.74$ and PPb vs. PMn $r=0.86$.

The seaward increases of the DPb concentrations suggest that the atmospheric Pb supply, predominantly from Mainland China, either by dry or wet deposition should

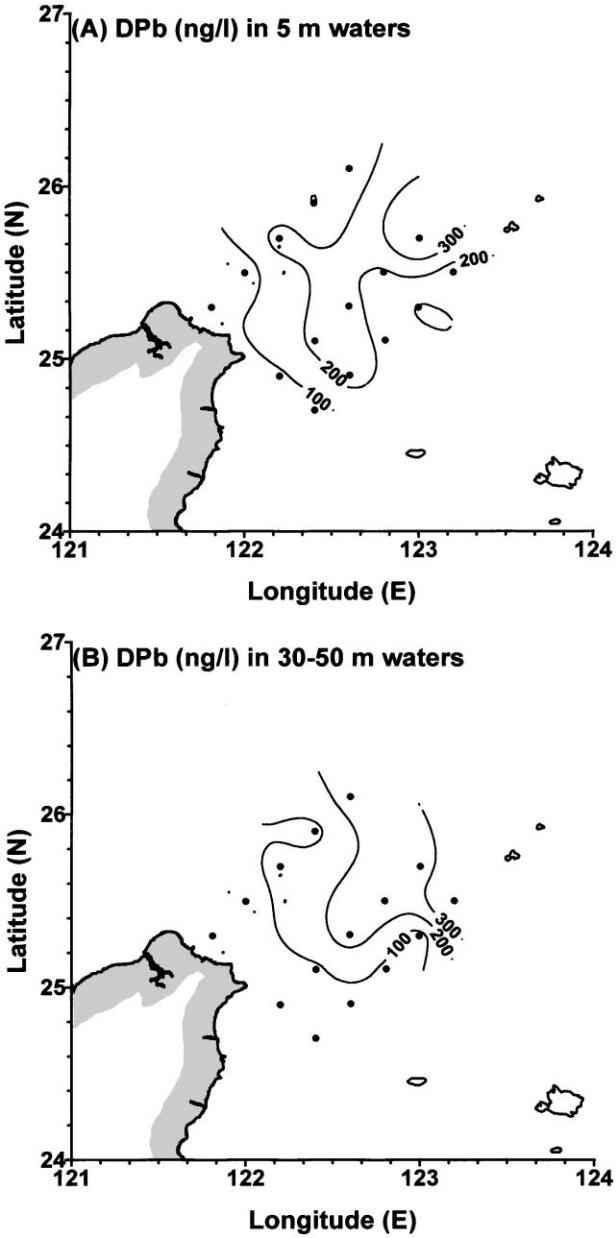

Fig. 3. Spatial distributions of dissolved Pb (DPb) in both (A) 5 m and (B) 30–50 m water layers in the southern East China Sea.

be the major source of the seawater DPb. In fact, the total atmospheric Pb fluxes have been estimated to be 590 ng/cm^2/year (Lin, unpublished data) based on a 2-year aerosol measurement carried out on a small offshore island (Pengchiayu

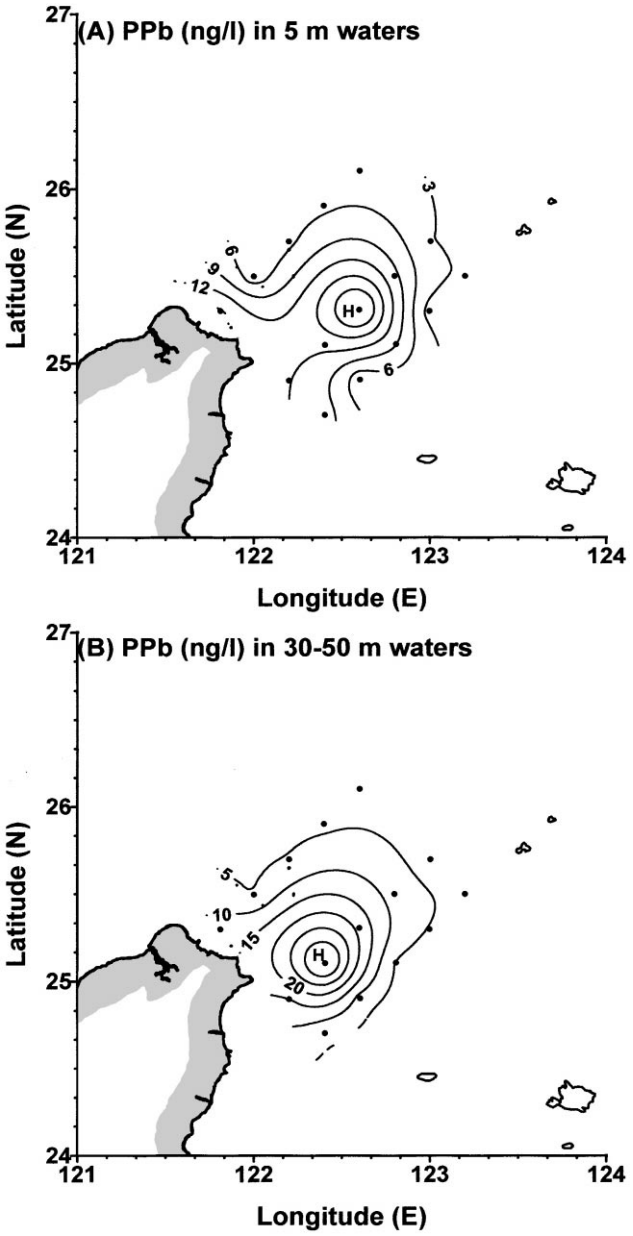

Fig. 4. Spatial distribution of particulate Pb (PPb) in both (A) 5 m and (B) 30–50 m water layers in the southern East China Sea.

Island) located in the study area (Fig. 1). These fluxes are very high compared to many coastal or marginal seas (Injuk, Van Grienek & De Leeuw, 1998; Patterson & Settle, 1987). Effective removal of DPb from overlying waters by particle scavenging

by the high loads of suspended matter in coastal waters is another cause for the DPb variations observed (Fig. 3) (Brugmann et al., 1985; Helmers & Rutgers van der Loeff, 1993). In addition, an examination of the DPb contour patterns shown in Fig. 3 reveals that the isopleth of 200 ng/l curves around the eddy center (Station 11). It is speculated that the cyclonic eddy over the shelf-slope of the southern ECS off northeastern Taiwan affected the DPb distributions by introducing high DPb offshore waters toward coastal area of low DPb waters. The influence of the eddy on the horizontal distributions of PAl (Al was taken as an indicator of lithogenic particles) in the region has also been described (Hsu et al., 1998).

Maximum concentrations of PPb occur at Station 11 in 5 m water layer and at Station 6 in 30–50 m water layer (Fig. 4). The distribution patterns of PPb suggest that riverine input is the major source for crustal elements (like Al and Mn) and likely for anthropogenic elements (like Pb) and that the cyclonic eddy drives their distributions (Hsu et al., 1998). Mean Pb:Al ratios in seawater particulates (1.36×10^{-3}) are high, compared to the mean Pb:Al ratio of shale (0.25×10^{-3}) (Turekian & Wedepohl, 1961). This indicates that Pb is enriched in the ECS surface seawater particulates. Coupled with the landward decrease of DPb concentrations, the results suggest that the PPb concentrations could be enhanced due to DPb transformation via particle scavenging processes. The aluminosilicates (or clay minerals) and manganese oxides (or manganese oxide coating onto various particles) are likely to act as an efficient scavenger to DPb (Li, 1981; Turekian, 1977; Whitfield & Turner, 1987), as indicated by the strong correlation among PPb, PAl and PMn and concurrence of PPb, PAl and PMn maxima in the region. The horizontal distributions of PPb are also affected by the cyclonic eddy, like those for PAl (or lithogenic particles) (Hsu et al., 1998) as indicated by the 'Ω'-like distribution.

3.3. Relationships between DPb concentrations in upper waters and magnitudes of atmospheric Pb input fluxes

Using the analytical results of this work coupled with the earlier results of other researchers, a positive correlation between surface concentrations of DPb and eolian Pb fluxes from other oceanic regions (the North Pacific, the North Atlantic and the North Sea) (Balls, 1985a,b; Boyle & Huested, 1983; Brugmann et al., 1985; Flegal & Patterson, 1983; Helmers et al., 1990; Injuk et al., 1998; Maring & Duce, 1990; Maring, Patterson & Settle, 1989; Patterson & Settle, 1987) can be obtained (Fig. 5). This is further evidence that the sources of Pb pollution are eolian inputs principally from Mainland China. This contrasts to many oceans where the Pb concentrations are declining due to the phase-out of leaded-gasoline recently (Boyle et al., 1986; Helmers et al., 1990; Nriagu, 1996).

3.4. Residence times of DPb in the surface water

In general, the residence time (τ) of an element in seawater can be estimated by the following equation:

$$\tau = I/F$$

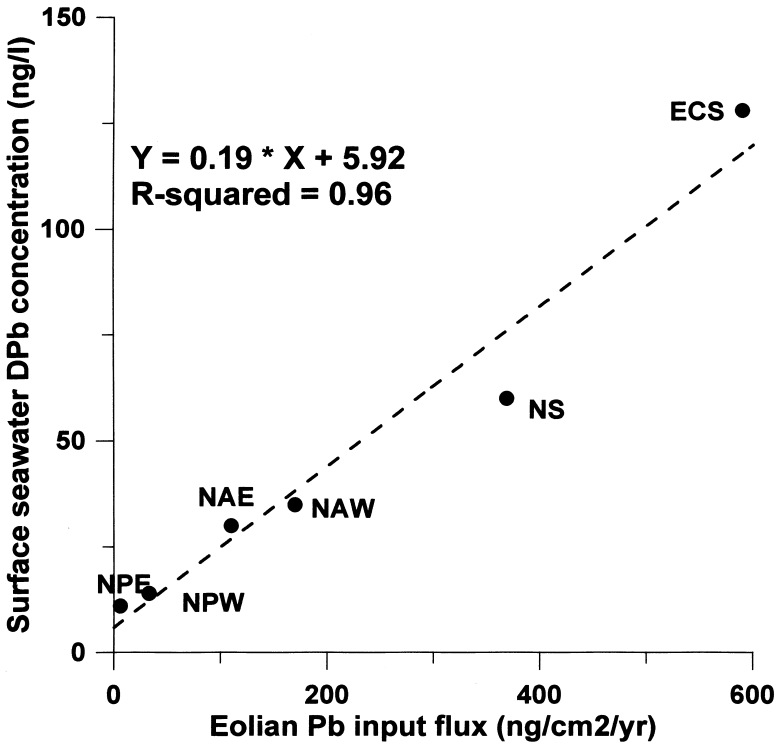

Fig. 5. Relationships between surface seawater dissolved Pb (DPb) concentration and total eolian Pb input flux. Sources: for the North Pacific easterlies (NPE), DPb from Flegal and Patterson (1983), eolian Pb from Flegal and Patterson (1983) and Maring and Duce (1990); for the North Pacific westerlies (NPW), DPb from Flegal and Patterson (1983), eolian Pb from Flegal and Patterson (1983) and Maring et al. (1989); for the North Atlantic easterlies (NAE), DPb from Helmers et al. (1990), eolian Pb from Patterson and Settle (1987); for the North Atlantic westerlies (NAW), DPb from Helmers et al. (1990) and Boyle and Huested (1983), eolian Pb from Flegal and Patterson (1983); for the North Sea (NS), DPb from Balls (1985a,b) and Brugmann et al. (1985), eolian Pb from Injuk et al. (1998); for the East China Sea (ECS), data from this study.

where τ is the residence time of a given element; I, the inventory of a given element in the environmental reservoir of interest; and F, the input or output fluxes of a given element into or from the reservoir. Under steady-state conditions, the rates of input and output are equal.

Firstly, inventories of DPb in the top 50 m at each station were calculated, and then residence times at each station could be estimated if the atmospheric input fluxes of Pb are available. Here we assumed the atmospheric Pb input fluxes to be 295 ng/cm^2/year based on the measurements that the total Pb atmospheric flux is 590 ng/cm^2/year (Lin, unpublished data) and the Pb solubility of aerosols as deposited into seawater is 50% (Duce et al., 1991).

The calculated mean residence time of DPb in the surface water is 2.1 years, very comparable with those estimated from ^{210}Pb (1.8 years) in the same study region

(Chung & Wu, 1995) and also from literature data of common Pb or ^{210}Pb (1.7–5 years) from many seas (Bacon, Spencer & Brewer, 1976; Nozaki, Thomson & Turekian, 1976; Schaule & Patterson, 1981; Veron et al., 1987). It should be mentioned that the temporal variations in surface concentrations of DPb and atmospheric Pb fluxes were not taken into account in the calculations of residence times.

4. Summary

In the southern ECS, DPb concentrations in upper waters are several times higher than those in the North Pacific; the pronounced DPb concentrations correspond to high atmospheric input fluxes of Pb. The results agree well with the suggestion by Flegal and Patterson (1983) that atmospheric inputs and seawater concentrations should be correlated. Also, the results would provide background information of Pb concentrations for monitoring future change in the ECS seawater compositions if Mainland China gradually changes production of pollutants owing to its rapid industrialization. Distributions suggest that DPb is a function of atmospheric supplies of Pb and particle scavenging processes. Whereas for PPb, distributions are controlled by riverine sources with an anthropogenic origin, scavenging and by eddy circulation. The DPb usually accounts for most ($>90\%$) of total Pb; its residence times in the upper water (50 m) are estimated to be about 2 years.

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