Fluxes and inventories of ^{210}Pb and ^{137}Cs in lake sediments from East Asia and their relationships to sedimentary environment

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The activities, fluxes and inventories of ^{210}Pb and ^{137}Cs in sediments collected from freshwater lakes, brackish-water lakes and seabeds in the East Asia are studied to elucidate their geochemical behaviors and characteristics concerning sedimentation environments. The surface concentrations varied widely by location. The average flux of excess ^{210}Pb increased from freshwater lakes to brackish-water lakes to seawater, and the average flux of ^{137}Cs showed an inverse trend. The inventories of excess ^{210}Pb and ^{137}Cs were in good correlation in samples from the same lake or adjacent areas. The inventory ratio ($^{137}\text{Cs}/\text{excess } ^{210}\text{Pb}$) was in the order of samples from seawater, increasing in brackish-water, further increasing in freshwater, and samples from Chinese lakes showed higher values. It is inferred that the inventory ratio may become a kind of index of sedimentation environment, geological, and/or radiochemical events.

Key words: ^{210}Pb , ^{137}Cs , lake sediment, East Asia, inventory, flux

Introduction

The sedimentation rate is one of the most important pieces of information for the analysis of sedimentary process and environment. ^{210}Pb and ^{137}Cs have been used to date recent lake sediments or sea bottom sediments (for example; Appleby and Oldfield, 1978; Cochran et al., 1998; Goldberg and Koide, 1962; Koide et al., 1972; 1973; Krishnaswami et al., 1971). The former nuclide belongs to the uranium series nuclides and is produced from ^{222}Rn gas through short half-life nuclides. Its half-life is 22.3 y, which is appropriate for dating within about 100 years. On the other hand, the latter nuclide has been produced mainly from the nuclear weapon tests and the nuclear power industries. Its detection in sediments started in 1954, and its release became maximum in 1963 (Peirson, 1971). Its half-life is 30.3 y, and the variation of its activity is also used for marking the date (1963 or

1954).

A simple ^{210}Pb chronology is based on the following assumptions.

- (1) The produced ^{210}Pb (unsupported ^{210}Pb ; excess ^{210}Pb) is adsorbed on the aerosol, removed from the atmosphere within a week (Poet et al., 1972), and deposited with suspended matter in the water column.
- (2) The residence time of excess ^{210}Pb in water is by far shorter than the sedimentation time, and excess ^{210}Pb is successively incorporated into the sediment.
- (3) Excess ^{210}Pb in the sediment does not move after sedimentation. The sediment column is a closed system for excess ^{210}Pb .
- (4) ^{210}Pb in the sediment grain (supported ^{210}Pb) is in equilibrium with ^{226}Ra in the sediment grain.
- (5) The excess ^{210}Pb concentration in the surface bottom sediment is always constant (CIC model), or the flux (F) of excess ^{210}Pb is constant (CRS model; Appleby and Oldfield, 1978).

The activity (A_i) of excess ^{210}Pb at depth z is expressed as follows in a simple model.

$$A_i = A_0 \cdot \exp(-\lambda \cdot z/w) \quad \text{eq. [1],}$$

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where A_0 , λ , and w are the activity at the surface sediment, the decay constant of ^{210}Pb , and the sedimentation rate, respectively. The flux (F) is the annual input of radioactivity into the sediment:

$$F = A_0 \cdot w \quad \text{eq. [2].}$$

The inventory (I) is the depth-integrated radionuclide activity through the sediment column and is calculated as follows;

$$I = \sum \rho_s \cdot (1 - \phi) \cdot A_i \cdot \delta_z \quad \text{eq. [3],}$$

where ρ_s , ϕ , and δ_z are the solid density (assumed as 2.45 in this study), the porosity, and the depth increment in sediment column, respectively. There is a relationship between F and I (Kanai, 2000);

$$F = \lambda \cdot I \quad \text{eq. [4].}$$

The chronology by use of ^{137}Cs is also based on the resembled assumptions mentioned above, except (4) and (5). The radionuclides are assumed to associate with the aerosol or suspended materials, deposit with suspended matters in water column, and successively incorporated into the sediment.

We have studied the sedimentation rates in some lakes in Japan, China, and Nepal (Kanai et al., 1995; 1997; 1998a; 1998b; 2000; 2002) and in seas (Kanai and Ikehara, 1995). In this study, validation of the model is checked by using these data. Especially much attention was paid to the fluxes and inventories of the radionuclides to distinguish the characteristics of the sediments such as fresh/brackish environment.

Samples and experimental

The lake sediments were taken from Japan, China, and Nepal. In Japan, Lake Suwa, Lake Shinji, and Lake Nakaumi were selected. Lake Suwa is located in the center of Nagano Prefecture, in central Japan. It is a freshwater lake, and 11 cores with 50–90 cm lengths (about 72 cm on average) were taken from the bottom (Kanai et al., 1995; 1997). Lake Shinji and Lake Nakaumi are located in Shimane and Tottori Prefectures, western Japan. They are brackish-water lakes. Although the salinities in these lakes vary with location, water depth, and season, those reported are 2.6–3.1‰, 1.8‰ (surface water), 3.3–4.9‰, 1.8–9‰, >7‰ (bottom water) for Lake Shinji, and 11–14‰, 16.0‰, 5–10‰ (surface water), 18–25‰, 25–30‰, 20–25‰ (bottom water) for Lake Nakaumi (Nakamura, 1993; Takayasu and Tokuoka, 1993; Tokuoka et al., 1990). Fifteen and thirteen cores ranging from 31 to 96

Fig. 1 Sampling locations of studied areas.

The map is created by using “pscoast” (URL: woodshole.er.usgs.gov/mapit/index.html).

cm were taken systematically in about 2 km intervals in the two lakes, respectively (Kanai et al., 1998b; 2002). Three cores were taken from off Niigata, the Sea of Japan (Kanai and Ikehara, 1995). Two Chinese lake cores with 60–85 cm lengths were taken from Lake Daihai and Lake Heiquan (Kanai et al., 1998a). Two cores (40 and 60 cm long) in Nepal lakes were taken from Lake Begnas and Lake Rupacot (Kanai et al., 2000). The detailed locations are shown in Fig. 1.

Radioactivity measurement was conducted as follows. Each core sample was cut at 1–5 cm intervals, and the divided samples were dried and ground prior to analysis. About 1–5 g of powdered sample was taken into the Nalgene centrifuge tube with a cap and sealed for more than 1 month to maintain radioactive equilibrium between ^{226}Ra and ^{214}Pb . Then the activities of ^{210}Pb (peak energy: 46.5 keV), ^{214}Pb (352 keV), ^{137}Cs (661.6 keV), and ^{40}K (1461 keV) were measured by gamma-ray spectrometry, using well-type Ge semiconductor detectors (ORTEC GWL-140230-S and GWL-120230-S). The peak intensities were corrected by the sample configuration. The activity of excess ^{210}Pb is calculated by subtracting that of ^{214}Pb from that of ^{210}Pb , assuming the above assumption (4). The additional information is shown in the references.

The adsorption experiment was conducted as follows. The bentonite powder, kaolinite powder (Wako Pure Chemical Industries, Ltd.), and iron oxyhydroxide (FeOOH ; Nacalai Tesque, Inc.) were taken in test tubes with 10 ppm Cs solutions, respectively (solid: solution ratio is 1: 100). An aliquot of the reaction solution was taken after one month, and the Cs concentration was determined by an atomic emission spectrometer (Japan Jarrel Ash Ltd., AA-8500).

Results and discussions

1. Activities of surface sediments and fluxes

Most of excess ^{210}Pb , after disintegrations from ^{222}Rn in the atmosphere, is deposited in the sediments with aerosol particles or suspended materials. Its concentration or flux (F) depends on the ^{222}Rn concentration in the air, and ^{222}Rn concentration depends on the amount of ^{226}Ra and ^{238}U . Although the temporal ^{222}Rn concentration varies with time and weather conditions, showing high in the morning and low in the evening (Eisenbud and Gesell, 1997; Iida et al., 2000), it can be assumed to be almost

constant on average for the relatively long term under concern.

Table 1 shows the activity of the surface sediment with the data on sedimentation rate, flux, inventories of excess ^{210}Pb and ^{137}Cs in the studied areas. Figure 2a depicts the range and the average of surface activities of excess ^{210}Pb . The data obtained from the same lake vary widely. For example, those from Lake Shinji range from 0.06 to 0.7 Bq/g. Those from Lake Suwa and Chinese lakes (average = 0.1 Bq/g) appear to be lower than those from others. Because there is a relationship (eq. [2]) among flux (F ; Bq/cm²/y), surface concentration (A_0 ; Bq/g) and sedi-

Fig. 2 (a): Range (straight line) and average ($\square$) of excess ^{210}Pb activity at surface sediment. (b): Range (straight line) and average ($\square$) of ^{137}Cs activity at surface sediment.

Table 1 Activity of the surface sediment, sedimentation rate, flux, and inventory of excess ^{210}Pb and ^{137}Cs in the studied areas.

	activity at surface sediment				sedimentation rate		Flux	Inventory		Inventory ratio
	depth	excess ²¹⁰ Pb	¹³⁷ Cs	²¹⁴ Pb(²²⁶ Ra)	²¹⁰ Pb method		excess ²¹⁰ Pb	excess ²¹⁰ Pb	¹³⁷ Cs	¹³⁷ Cs
	cm	Bq/g	Bq/g	Bq/g	cm/y	g/cm ² /y	Bq/cm ² /y	Bq/cm ²	Bq/cm ²	excess ²¹⁰ Pb
● Lake Suwa (Kanai et al., 1995; 1997)										
S-15	1-5	0.19	0.008	0.017	1.30	0.45	0.071	1.71	0.27	0.16
S-18	0-1	0.20	0.016	0.003	0.67	0.20	0.032	1.04	0.23	0.22
S-22	3-4	0.16	0.010	0.013	1.39	0.47	0.060	1.69	0.22	0.13
S-10B	1-2	0.09	0.009	0.006	0.96	0.39	0.022	0.58	0.30	0.52
S-24A	1-2	0.07	0.007	0.019	1.09	0.35	0.025	0.75	0.21	0.28
S-33	0-1	0.05	0.008	0.016	2.08	1.07	0.044	0.37	0.06	0.16
S-46	0-1	0.09	0.007	0.016	0.40	0.11	0.014	0.42	0.04	0.09
ST-4	0-1	0.12	0.008	0.018	0.73	—	—	—	—	—
ST-31	0-1	0.09	0.009	0.017	0.65	—	—	—	—	—
ST-130	0-1	0.07	0.008	0.020	1.76	—	—	—	—	—
ST-149	0-1	0.05	0.005	0.013	0.84	—	—	—	—	—
● Lake Shinji (Kanai et al., 1998a)										
11	0-1	0.45	0.057	0.032	0.06	0.03	0.014	1.23	0.09	0.08
13	0-1	0.69	0.049	0.041	0.28	0.10	0.072	2.44	0.30	0.12
15	0-1	0.56	0.048	0.037	0.24	0.12	0.054	1.86	0.39	0.21
17	0-1	0.40	0.048	0.042	0.14	0.07	0.032	0.99	0.12	0.12
19	0-1	0.23	0.022	0.040	0.12	0.09	0.026	0.92	0.19	0.21
40	0-1	0.34	0.027	0.037	0.50	0.26	0.094	2.69	0.55	0.20
42	0-1	0.55	0.037	0.035	—	—	—	2.16	0.68	0.31
44	0-1	0.64	0.045	0.030	0.08	0.02	0.018	1.07	0.06	0.05
46	0-1	0.17	0.022	0.040	0.05	0.02	0.005	0.48	0.02	0.04
48	0-1	0.43	0.040	0.024	0.08	0.04	0.021	0.91	0.12	0.13
50	0-1	0.43	0.028	0.031	0.23	0.11	0.053	2.25	0.20	0.09
52	0-1	0.06	0.007	0.032	0.04	0.03	0.003	0.23	0.01	0.05
69	0-1	0.38	0.024	0.035	0.51	0.28	0.092	2.63	0.67	0.25
71	0-1	0.47	0.039	0.038	0.44	0.20	0.115	3.40	0.45	0.13
73	0-1	0.49	0.031	0.037	0.14	0.06	0.032	1.81	0.09	0.05
● Lake Nakaumi (Kanai et al., 2002)										
3	0-1	0.25	0.004	0.017	0.32	0.12	0.032	1.08	0.02	0.02
5	0-1	0.09	0.001	0.017	0.24	0.17	0.056	1.87	0.09	0.05
10	0-1	0.75	0.016	0.010	0.97	0.23	0.172	5.42	0.22	0.04
12	0-1	0.48	0.014	0.025	0.23	0.09	0.048	1.90	0.03	0.02
14	0-1	0.35	0.041	0.023	0.35	0.12	0.057	2.33	0.07	0.03
16	0-1	0.58	0.015	0.023	0.46	0.16	0.102	3.17	0.15	0.05
32	0-1	0.34	0.022	0.019	1.06	0.44	0.186	5.81	0.58	0.10
34	0-1	0.72	0.023	0.020	0.47	0.17	0.103	3.21	0.12	0.04
36	0-1	0.62	0.024	0.025	0.21	0.07	0.069	2.04	0.09	0.05
38	0-1	0.47	0.015	0.047	0.99	0.38	0.269	4.74	0.49	0.10
40	0-1	0.39	0.024	0.030	0.17	0.08	0.035	0.98	0.02	0.02
51	0-1	0.51	0.020	0.030	0.10	0.04	0.028	0.76	0.03	0.04
53	0-1	0.80	0.017	0.019	0.28	0.09	0.067	2.60	0.15	0.06
● Nepal lakes (Kanai et al., 2000)										
B-1	0-1	0.60	0.049	0.103	0.33	0.15	0.063	1.96	0.29	0.15
					* 0.23	0.09	0.039	1.25	0.23	0.18
R-2	0-1	0.53	0.037	0.133	—	—	—	2.35	0.45	0.19
					* 0.53	0.13	0.053	1.58	0.29	0.18
● Chinese lakes (Kanai et al., 1998)										
DH-1	0-1	0.11	0.024	0.035	0.34	0.36	0.038	1.07	0.72	0.67
KS-2	0-1	0.13	0.025	0.033	0.09	0.07	0.013	0.37	0.19	0.52
● The Sea of Japan (Kanai and Ikehara, 1995)										
B-91	0-2	0.48	0.008	0.022	1.90	0.17	0.091	6.62	0.13	0.02
B-126	0-2	0.62	0.017	0.023	4.00	0.27	0.250	7.43	0.22	0.03
B-135	0-2	0.39	0.005	0.022	4.00	0.34	0.170	5.02	0.12	0.02

*: excluding flood layers

mentation rate (w ; $\text{g}/\text{cm}^2/\text{y}$) in an ideal sedimentation model, the surface concentration may vary with the flux and the sedimentation rate. Flux, sedimentation rate and surface concentration are calculated from the depth profile of excess ^{210}Pb activities using the regression line by least square method. A nearly linear correlation is observed between the sample surface concentration and the calculated surface concentration, though the sedimentary condition is not always ideal and there are some discrepancies between sample and calculated surface concentrations probably because of bioturbation or mixing of the sediments that would make the sample activity lower.

Figure 3a depicts the range and the average of excess ^{210}Pb fluxes. Although the fluxes from Lake Nakaumi and the Sea of Japan are scattered, the average has an increasing trend in the order of Chinese lakes ($0.026 \text{ Bq}/\text{cm}^2/\text{y}$), Lake Suwa ($0.038 \text{ Bq}/\text{cm}^2/\text{y}$), Nepal lakes ($0.046 \text{ Bq}/\text{cm}^2/\text{y}$), $\sim$ Lake Shinji ($0.045 \text{ Bq}/\text{cm}^2/\text{y}$), $<$ Lake Nakaumi ($0.094 \text{ Bq}/\text{cm}^2/\text{y}$), and $<$ the Sea of Japan ($0.170 \text{ Bq}/\text{cm}^2/\text{y}$). The ^{210}Pb flux of $0.011 \text{ Bq}/\text{cm}^2/\text{y}$ in Lake Michigan is reported (Robbins and Edgington, 1975). In central and eastern America, the average ^{210}Pb flux on the soil is reported as 0.017 and $0.026 \text{ Bq}/\text{cm}^2/\text{y}$ (Nozaki et al., 1978). Similar depositional flux data in New Haven

Fig. 3 (a): Range (straight line) and average ($\square$) of flux of excess ^{210}Pb activity at surface sediment. (b): Range (straight line) and average ($\square$) of flux of ^{137}Cs activity at surface sediment.

(0.02 Bq/cm²/y), Bermuda (0.0115 Bq/cm²/y), and south-eastern Michigan (0.0235 Bq/cm²/y) are reported (Turekian et al., 1983; McNeary and Baskaran, 2003). In Japan, the depositional fluxes are reported to be 0.026–0.047 Bq/cm²/y in Hokkaido (Tsunogai and Shinagawa, 1977; Fukuda and Tsunogai, 1975; Kurata and Tsunogai, 1986), 0.015 Bq/cm²/y in Tokai (Ueno et al., 2003), 0.018–0.023 Bq/cm²/y in Tokyo (Tsunogai and Shinagawa, 1977; Kurata and Tsunogai, 1986), 0.025 Bq/cm²/y in Shiga (Kurata and Tsunogai, 1986), and 0.023 Bq/cm²/y in Nagasaki (Honda et al., 2002). Those in the studied lakes (0.026–0.094 Bq/cm²/y) are slightly higher than other data reported. This suggests the existence of additional ²¹⁰Pb accumulation mechanisms. On the other hand, the ²¹⁰Pb flux in the Sea of Japan (0.170 Bq/cm²/y) is comparable to those on the continental slopes and marginal basins in the Gulf of Papua, which range from 0.02 to 0.213 Bq/cm²/y (Muhammad et al., 2008). This infers that the seawater has some effect on the higher flux of ²¹⁰Pb_{ex}.

Meanwhile, ¹³⁷Cs is produced by the nuclear power industries and the past nuclear weapon tests. During the 1950s and 1960s, there were many atmospheric tests of nuclear weapons. After the nuclear moratorium in 1959 and the test ban treaty in 1963, the intense release of ¹³⁷Cs into the environment decreased drastically. Because the amount of ¹³⁷Cs released into the atmosphere is very small nowadays, those detectable activities in the atmosphere are considered to originate by a resuspension of the past fallout deposit (Fujiwara et al., 2007; Igarashi et al., 2001; 2003). The surface activity of ¹³⁷Cs in the bottom sediment is depicted in Fig. 2b, showing a large variation. The apparent surface fluxes of ¹³⁷Cs can be calculated using the surface activity and sedimentation rate (eq. [2]). They are rough estimates and are shown in Fig. 3b. Compared with Fig. 2b, the average fluxes in various locations appear to be nearly equal (0.003–0.005 Bq/cm²/y), though there is a large scatter. It is well known that ¹³⁷Cs deposition density deduced by an atmospheric model has a latitudinal variation (UNSCEAR, 2000). The sampling locations in this study are within a relatively narrow range of 28.2–40.4°N. Therefore this infers that the global air movement mixes the recent fallout homogeneously in the East Asia studied. However, the critical inspection shows that it has an increasing trend of the Sea of Japan, < Lake Nakaumi, < Lake Shinji, < Lake Suwa, < Nepal lakes, < Chinese lakes. This trend of ¹³⁷Cs flux is nearly an inverse relation to that of ²¹⁰Pb flux, and some systematic sedimentation mechanisms, in addition to the mixing by the global air movement, are suggested. One of

the reasons is thought to be the catchment area of the lake. In Japan, the catchment area is in the order of Lake Suwa (531 km²), < Lake Nakaumi (595 km²), and < Lake Shinji (1288 km²), different from that expected. Considering that Lake Nakaumi is adjacent to Lake Shinji and the river water flows from Lake Shinji to Lake Nakaumi, the effective catchment area of Lake Nakaumi should be more than 595 km², probably including that of Lake Shinji.

Since the amount of ¹³⁷Cs released into the air is minor nowadays, a resuspension of the past fallout deposit may be important. The annual atmospheric depositions in Sapporo, Akita, Sendai, Tokyo, Osaka and Fukuoka in 1982 (Katsuragi, 1983; Katsuragi and Aoyama, 1986) are less than 0.7% of those in 1963. The current deposition in Tsukuba decreased further to the 10⁻²–10⁻³% level of those in the early 1960s. Recent direct atmospheric deposition flux of ¹³⁷Cs is reported to be 0.014–0.035 mBq/cm²/y in Tsukuba during 1993–1999 (Igarashi et al., 2003) and 0.053 mBq/cm²/y in Tokai during 1993–2001 (Ueno et al., 2003). Igarashi et al. (2003) attributed the source of ¹³⁷Cs onto contemporary sediment to the resuspension of soil dust, Asian dust (Kosa). However, the direct atmospheric deposition of ¹³⁷Cs is only about 1% of the sediment flux in this study. In addition, the activity of surface sediment is 18–86% of the maximum activity of the sediment cores. This implies that the atmospheric deposition is insufficient to explain the ¹³⁷Cs activity in the surface bottom sediment and that there should be other larger sources of ¹³⁷Cs.

2. Sources of excess ²¹⁰Pb and ¹³⁷Cs in sediments

The main sources of excess ²¹⁰Pb in sediment are considered to be wet and dry deposition from the atmosphere, the aeolian transport of soil dust particles from Asian continent, the in-situ production from the soluble ²²⁶Ra in water column, and the inflow through the river from catchment. Excess ²¹⁰Pb in the atmosphere deposit with aerosol particles or suspended materials after disintegrations from ²²²Rn (half-life is 3.8 days). Rainfall is one of the important factors controlling excess ²¹⁰Pb flux (McNeary and Baskaran, 2003; Yeager et al., 2005). An additional amount of excess ²¹⁰Pb caused by the Asian dust would be less important because the amount and its seasonal fluctuation in Yamagawa Prefecture was relatively small (Kanai and Yanagisawa, 2002). Although the temporal excess ²¹⁰Pb flux varies with space, time, and weather condition, the atmospheric depositional flux of excess ²¹⁰Pb in a small area can be assumed to be almost constant during the relatively long term under concern.

Another source of excess ²¹⁰Pb is ²²⁶Ra dissolved in

Fig. 4 Relationship between excess ^{210}Pb activity and ^{137}Cs activity at the surface.

water. The concentration of ^{226}Ra in seawater shows the increasing profile with depth and is concentrated on the seafloor. ^{210}Pb derived from ^{226}Ra dissolved in seawater would be rapidly scavenged from water by adsorption on suspended particles and deposit on the seafloor. The concentration of ^{226}Ra in seawater is by far higher than that in fresh water (Craig et al., 1973). Therefore the sediment under seawater would accumulate more excess ^{210}Pb .

The flux of inflow via rivers, in other words, from the catchment, implies the transport of particles by erosion. The flux may vary by the climate and topography in the area concerned. It should correlate with the sum of the present and the past depositions.

In this connection, the present flux of ^{137}Cs is owed to this source because its exhaustion into the atmosphere is little nowadays. The dominant flux is supposed to be the inflow by erosion of catchment soil and the stirred soil dust. The trend of ^{137}Cs flux in Japanese lakes also suggests the importance of the catchment area.

It is well known that fine particles such as clay contain more ^{137}Cs and excess ^{210}Pb , because the surface area is larger and both nuclides can be adsorbed on the grain. He and Walling (1996) showed a linear relationship between the specific surface area and the concentrations of ^{137}Cs and ^{210}Pb and a linear correlation between the two nuclides by laboratory experiment. Figure 4 shows the relationship between ^{137}Cs and excess ^{210}Pb in surface sediment samples studied in this study. Although the sources of the two nuclides are different, a general linear correlation is observed among the samples belonging to

the same lake or area, with a different slope by the group. If each flux should behave differently, *i.e.*, a constant atmospheric deposition and a various inflow from catchment should make up the total flux of excess ^{210}Pb in sediments, the ^{137}Cs activity of the surface sediment would not vary in proportion to excess ^{210}Pb . This linear correlation suggests nearly constant apparent flux ratios within a group and the strong adsorption and fixation of both excess ^{210}Pb and ^{137}Cs on suspended particles (Kanai, 2009). This may also suggest that the fluxes of direct atmospheric deposition and inflow from catchment are mixed together within the water column in the area considered, or that the flux of inflow is superior to the flux of atmospheric deposition than ever considered. The wide range of scatter and the different slopes are attributed to the diversities of catchment area, sedimentary environment, grain size, composition of particles such as inorganic/organic and minerals of surface sediments taken from various locations.

3. Inventories of excess ^{210}Pb and ^{137}Cs

Inventory is the total amount of deposited material from the past, the time integration of flux. The inventory of excess ^{210}Pb can be calculated by several methods; an integration of ideal activity-depth profile (Flux/λ ; eq. [4]) or a summation of real activity profile. In this study, the latter method is adopted because the excess ^{210}Pb activity-depth profile is not always ideal and the inventory of ^{137}Cs needs to be also calculated in the same way (Table 1).

The inventory of excess ^{210}Pb ranges from 0.2 to 7.4 Bq/cm^2 (Table 1). A larger inventory is observed in the

Fig. 5 Correlation between the inventories of excess ^{210}Pb and ^{137}Cs .

seafloor sediment. Thomson et al. (1993) reported the excess ^{210}Pb inventory of $0.18\text{--}0.79\text{ Bq/cm}^2$ in the north-east Atlantic Ocean. That in the Gulf of Papua was $0.48\text{--}6.88\text{ Bq/cm}^2$ (Muhammad et al., 2008). A relatively larger inventory for the marine environments may be attributed to the fact that the seawater is a big reservoir of excess ^{210}Pb , because the inventories in seawater column (not sediment) of $7.7\text{--}67\text{ Bq/cm}^2$ in the Malacca Straits and 313 Bq/cm^2 in the southern South China Sea are reported (Mohamed et al., 2006), while they depend on water depth. They are by far high compared with those in sediments.

On the other hand, the inventory of ^{137}Cs in the sediment ranges from 0.01 to 0.72 Bq/cm^2 (Table 1). Yeager et al. (2005) observed the ^{137}Cs inventories in the flood plain cores in the United States and reported 0.52 Bq/cm^2 on average. The ^{137}Cs inventories in undisturbed soils in Vietnam were in the range of $0.01\text{--}0.33\text{ Bq/cm}^2$, 0.06 Bq/cm^2 on average (Hien et al., 2002). Andrello and Apoloni (2004) reported those in undisturbed soils in Brazil to be $0.03\text{--}0.04\text{ Bq/cm}^2$, one order smaller than in the Northern Hemisphere ($0.16\text{--}0.25\text{ Bq/cm}^2$ in USA, Canada, UK). Not only excess ^{210}Pb , but also ^{137}Cs inventory in seawater (not sediment) is much more complicated because of inflow, transport, and subduction mechanisms of water mass, and $0.05\text{--}0.41\text{ Bq/cm}^2$ in the Japan Sea (Ito et al., 2003), $0.19\text{--}0.32\text{ Bq/cm}^2$ in South China Sea, and Sula Sea (Yamada et al., 2006) are reported. Although the inventory of excess ^{210}Pb in the seawater column is two orders of magnitude larger than that of

sediment, the inventory of ^{137}Cs in the seawater column is comparable to that of sediment. Therefore the reservoir function of seawater for ^{137}Cs is supposed to be smaller than excess ^{210}Pb .

The ^{137}Cs and the excess ^{210}Pb are both supposed to deposit with suspended matter in water. Figure 5 shows the correlation between the inventories of excess ^{210}Pb and ^{137}Cs with the additional data on the coastal marine sediments by Suzuki (1993). Although each inventory has a wide range of variation, some linear correlation appears to exist among the samples from the same lake or area. The trend, the slope being in the order of the Sea of Japan, < Lake Nakaumi, < Lake Shinji, < Lake Suwa, Nepal lakes, < Chinese lakes, resembles that observed in Fig. 4.

As is explained above, the sedimentary condition in seawater is preferable to accumulate ^{210}Pb . Although both ^{210}Pb and ^{137}Cs in the temporal sediment are strongly fixed in the particle lattice (Kanai, 2009), those in the past sediment, especially ^{137}Cs , are thought to have been adsorbed on the surface of particle in water and then strongly fixed. Therefore the salinity effect during past adsorption must be taken into consideration. Salinity is a main factor controlling the water chemistry. As the clay minerals in the sediment and the oxyhydroxides such as iron or aluminum oxyhydroxides, sometimes play important roles in the adsorption and coprecipitation (Tessier et al., 1985) as well as organic materials, an adsorption experiment was conducted using clay minerals (kaolinite and bentonite) and iron oxyhydroxide to elucidate this

Fig. 6 Partition coefficient (K_d) of Cs for bentonite, kaolinite, and iron oxyhydroxide with varying pH and salinity.

Table 2 Ratios of inventories of ^{137}Cs to those of excess ^{210}Pb .

location	slope of regeression line	R*	ratio of both inventories at each location	salinity(‰)
Lake Suwa (n=7)	0.18	0.26	0.22 ± 0.14	estimated < 1
Lake Shinji (n=15)	0.17	0.77	0.14 ± 0.08	1-9
Lake Nakaumi (n=13)	0.07	0.82	0.05 ± 0.03	7-31
Chinese lakes (n=2)	0.65	0.99	0.60 ± 0.10	estimated < 1
Nepal lakes (n=4)	0.18	0.92	0.18 ± 0.02	estimated < 1
Sea of Japan (n=15)	0.02	0.95	0.02 ± 0.01	35

*: coefficient of correlation

process as a model. The partition coefficient (K_d ; ml/g), which is the concentration ratio of solid phase to liquid phase, is shown in Fig. 6. The bentonite, whose main mineral is smectite, has a large K_d value in a wide range of pH. The K_d of kaolinite increased with increasing pH, while that of iron oxyhydroxide was nearly unchanged. With the increase of the NaCl concentration up to 3%, which is almost equal to that of seawater, all K_d s examined in this experiment decreased. This model experiment shows that the salinity has a strong negative influence on K_d s. Organic carbons also adsorb the metal ions, and their K_d s would have the same trend as in this model experiment. Therefore the scavenging ability of depositional particles should be influenced by salinity.

Table 2 shows the slope of each group, supposing that a linear regression line might pass the origin. The average and standard deviations of the ratio of both inventories at each location are also shown. The slope is in the order of the Sea of Japan, < Lake Nakaumi, < Lake Shinji, < Lake Suwa, Nepal lakes, < Chinese lakes. The Sea of Japan is sea; Lake Nakaumi is a brackish-water; Lake Shinji is a slightly brackish-water; and others are freshwater lakes. Therefore this order seems to correspond to the salinity of water. Another characteristic revealed from

Fig. 5 is that the slope in Chinese lake samples is rather higher than expected from other freshwater lakes. Hirose et al. (1987) reported that the annual deposition of radioactive debris resulting from Chinese nuclear weapon tests was observed during 1968-1981, following 1963. So the amount of local fallout within China might be much higher than the average of the global fallout mixed well and diluted by the air movement.

Figure 7 shows the relationships between the salinity and inventory ratio of ^{137}Cs to excess ^{210}Pb . The salinity of brackish-water lakes has a wide variation, and its variation depends on the mixing ratio of freshwater with seawater in this figure. The inventory ratios (the slope in Table 2 is shown as a dotted line, and the average ratio as a straight line in Fig. 7) changed in almost one order range, in accordance with that of K_d s in the adsorption model experiment (Fig. 6). Therefore the differences of slope among samples from freshwater lakes, brackish-water lakes, and coastal sea are partly caused by the flux from ^{226}Ra and the salinity (influence of seawater in the past), i.e., the difference of K_d s under different aquatic conditions in the past. Because the variation of excess ^{210}Pb is in inverse direction of that of ^{137}Cs , the ratio of both inventories makes the environmental difference more

Fig. 7 Relationship between inventory ratio of ^{137}Cs to excess ^{210}Pb and salinity.

apparent.

The correlation between the inventories shown in Figs. 5 and 7 may discriminate the sedimentary environment of the lakes in the area, if many other inventory data are accumulated. The data of sediment core samples from the freshwater lake will fall in the group of the freshwater lakes. Similarly, those from brackish-water lakes will fall in the group of brackish-water lakes. If the plot of the inventories deviates from the group, it will mean that the site might have suffered from some geological event, such as turbidity or radiochemical accident, which would have changed the flux.

4. Inventories of brackish-water lakes

As mentioned above, the ratio of ^{137}Cs inventory to excess ^{210}Pb inventory tends to vary by the influence of seawater. However, the degree of influence by seawater, *i.e.*, salinity, in brackish-water lakes, varies widely by place and season. Therefore the ratio of ^{137}Cs inventory to excess ^{210}Pb inventory in Lake Shinji and Lake Nakaumi was studied further.

Although there are more than 20 rivers that run into Lake Shinji, the Hii River is the largest, which pours into the western part of the lake. The three shaded circles in Fig. 8 are the data at the locations near the mouth of the Hii River. Although the slope for the whole data was 0.17, which is near to that of fresh water lake (0.18), the linear regression line had a slope of 0.12 if the three shaded circles were excluded. It is shown as “line A” in the Fig. 8. The correlation coefficient was improved from 0.77 to

0.84. Therefore it is inferred that the three locations near the mouth of the Hii River are out of the group, less brackish, and much influenced by the terrestrial particles.

In the same way, Lake Nakaumi has two large inflows from the Ohashi River and the Iinashi River. The two shaded triangles in Fig. 8 are the data taken at the mouths of both rivers. If they were excluded, the calculated slope became 0.05 from 0.07. The linear regression line is shown as “line B” in Fig. 8. The correlation coefficient became 0.91, from 0.82. Therefore these locations near the mouths of rivers are less brackish and much more influenced by the terrestrial particles than other locations in the same lake. The data near the Ohashi River that pours from Lake Shinji is plotted near the “line A”. This suggests that the sedimentary environment at this location might be similar to that in Lake Shinji.

In this way, the inventory ratio of ^{137}Cs to excess ^{210}Pb , or the relationships between the inventories of both ^{137}Cs and excess ^{210}Pb among the groups, is supposed to become an important index of the sedimentary conditions.

Conclusion

^{210}Pb and ^{137}Cs , which are often used for geochronology, in the sediments from freshwater lakes, brackish-water lakes, and coastal sea in East Asia, are discussed from the point of view of geological setting. The inventories of excess ^{210}Pb and ^{137}Cs were in good correlation in the samples from the same lake or adjacent area. The

Fig. 8 Correlation between the inventories of excess ^{210}Pb and ^{137}Cs in Lake Shinji and Lake Nakaumi.

inventory ratio ($^{137}\text{Cs}/\text{excess } ^{210}\text{Pb}$) was in the order of samples from seawater, < brackish-water, < freshwater sediments, and that from Chinese lakes showed higher values. It is inferred that the inventory ratio may become a kind of index of sedimentary environment, geological event, and/or radiochemical events in the past. For the critical discrimination, many inventory data should be accumulated and evaluated statistically.

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東アジア地域の湖底堆積物中の ^{210}Pb と ^{137}Cs のフラックス及びインベントリーと堆積環境

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東アジア地域の淡水湖, 汽水湖, 海底等から採取した堆積物中の鉛-210, セシウム-137 の測定結果から, 放射能強度, フラックス, インベントリーについて, 堆積環境に関する特徴を抽出するために検討を行った. 表層濃度は場所によってかなりの変動を示した. 鉛-210 の平均フラックスは, 淡水湖<汽水湖<海底の順に増加する傾向を示し, 一方, セシウム-137 はその逆であった. 鉛-210 とセシウム-137 のインベントリーは, 同じ湖もしくは隣接する地域で良い相関を示した. そのインベントリーの比 ($^{137}\text{Cs}/^{210}\text{Pbex}$) は海底堆積物<汽水湖<淡水湖の順であり, 中国の湖における比は高めであった. このように, インベントリーの比は堆積環境, 地質学的なイベント, 放射化学的な出来事を示唆する指標になりうる事が明らかとなった.