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Implications of excess ²¹⁰Pb and ¹³⁷Cs in sediment cores from Mikawa Bay, Japan

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Abstract

Four sediment cores were collected from Mikawa Bay, Japan, and excess ²¹⁰Pb and ¹³⁷Cs were measured by gamma spectrometry. Sedimentation rates for the four cores were determined by ²¹⁰Pb method. The sedimentation rate range is 0.10–0.70 g/(cm²·year). The bio-mixing depth for each core is less than 7.0 cm, and was determined by the excess ²¹⁰Pb profiles as well. Therefore, the bioturbation is slight. The ²¹⁰Pb-derived dates coincided with the results from ¹³⁷Cs geochronology. Acceleration in sedimentation rate due to environmental alteration has been found in cores A2.5 and 05AS8, representing two depocenters due to their topography. Evidence of the Tokai Flood in 2000 was found in core 05AS8 according to the profiles of both radioisotopes and trace metals.

Key words: ²¹⁰Pb; ¹³⁷Cs; sedimentation rate; Mikawa Bay **DOI**: 10.1016/S1001-0742(08)62328-1

Introduction

The ²¹⁰Pb with a half-life of 22.3 years, as a member of the naturally occurring ²³⁸U decay series, is one of the most widely-used radioisotopes to determine the sedimentation rates in coastal seas on a timescale of around 100 years (Koide *et al.*, 1972; Robbins *et al.*, 2000; Lu and Matsumoto, 2005c). Meanwhile, ¹³⁷Cs, an artificial radioisotope, has often been used as a tracer to provide supplementary information on sedimentation rates to compare with the result derived from ²¹⁰Pb method (Ritchie and McHenry, 1990; Smith, 2001; Lu, 2004; Lu and Matsumoto, 2005c; Palinkas and Nittrouer, 2007).

Mikawa Bay facing a highly urbanized and industrialized region is one of the largest semi-enclosed seas in Japan. Due to the coastal urbanization and industrialization, sedimentation rates in the coastal seas have been significantly affected by changes in river sediment discharge (Owen and Lee, 2004; Siakeu *et al.*, 2004). The understanding of sedimentation in coastal seas is of great importance to coastal zone management, for example, pollution, harbor maintenance, fisheries, coast protection, conservation or recreational activities (Coltorti, 1997; Hansom, 2001). Therefore, this study focused on the determination of sedimentation rates in Mikawa Bay over the last century using the ²¹⁰Pb method and insights into its implications.

Change in natural system (e.g., climate change) can also cause change in sedimenation rate. Environmental change

events are often recorded in sediment cores. There was a heavy rain in the Mikawa Bay area and the adjacent Ise Bay area (Ushiyama and Takara, 2002). A flood caused by the heavy rain has been called the Tokai Flood. It was expected to find sedimentation evidence in the Ise Bay or Mikawa Bay sediments. However, the Tokai layer could not be observed in Ise Bay (Lu and Matsumoto, 2005c). Therefore, another aim of this study was to check if there was a record of the Tokai Flood in Mikawa Bay.

1 Sampling and methods

1.1 Study area

Mikawa Bay, located on the Pacific side of Honshu Island, Japan, is a semi-enclosed embayment separated from Ise Bay by the Chita Peninsula (Fig. 1). It has a surface area of 602 km^2 and a volume of about 5.5 km³. The catchment area of Mikawa Bay is 3752 km^2 , with a population of about 2.4 million. The annual discharges of the Yahagi River and the Toyo River are 50 and $32 \text{ m}^3/\text{s}$, respectively. Tidal currents through the Moroyoki and Nakayama channels bring oceanic water into the bay; once inside, the currents branch to North and East (Saijo and Unoki, 1977; Bodergat *et al.*, 1997). Due to the relatively low river discharge and the semi-enclosed nature of the Mikawa Bay, bay water exchange is slow (Saijo, 2002).

The bay is close to the Chubu industrial district of Japan (Nagoya, Toyota, Toyohashi etc.). Due to an increasing load of nutrients from rapid industrial development since

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the mid-1950s, as well as from livestock farms and municipalities, the bays have become highly eutrophicated and hence have induced intense red tides almost every year during summer (Bodergat *et al.*, 1997). To satisfy the intensified need of land to develop industries after World War II, reclamation works in coastal seas of Japan increased significantly after 1945, particularly during the 1960s–1980s (Wakabayashi, 2000). The reclamation in Mikawa Bay, with an area of more than 30 km² up to 1996, was carried out in Mikawa Harbor (near Toyohashi) and Kinuura Harbor (north to the Yahagi River estuary) (Saijo, 2002).

Sediments in Mikawa Bay are predominantly silt- and clay-sized mud. Coarse-grained sediments, for example sand- and gravel-grained sediments, are deposited near the bay mouth.

1.2 Sampling

Using a modified gravity corer consisting of an acrylic pipe with a 5.5-cm inner diameter, four sediment cores were collected onboard the T/S Seisui-maru, Mie University during two cruises in July 2003 and September 2005. The sampling sites are shown in Fig. 1 and Table 1. Lowering the corer as slowly as possible, sediment cores with undisturbed sedimentary strata were obtained. Immediately after collection, the sediment cores were sectioned at 2–5 cm intervals and then sealed in plastic containers (Lu and Matsumoto, 2005a, 2005b). The sediments were later homogenized and dried for radionuclide analyses including ²¹⁰Pb, ²¹⁴Pb, and ¹³⁷Cs.

1.3 Analytical methods

The analytical details have been described in Lu and Matsumoto (2005c). The homogenized sediment samples were dried at 110°C and ground to fine powder in a quartz mortar. An EG&G ORTEC model GWL-120230 (USA) well-shaped germanium detector equipped with a SEIKO EG&G model 7800 MCA (Japan) was used for the low-energy gamma-ray spectrum analyses. The dried and homogenized samples weighing 2.5 g each in calibrated geometries were counted for 48 to 72 h. Uranium-Thorium ore DL-1a from the Canadian Center for Mineral and Energy Technology was used as a reference standard. The activities of ²¹⁰Pb, ²¹⁴Pb, and ²¹⁴Bi were measured based on their various characteristic gamma peaks (46.5 keV for ²¹⁰Pb, 295.2 keV and 351.9 keV for ²¹⁴Pb, 609.3 keV for ²¹⁴Bi). The radioactivity of ²²⁶Ra (supported ²¹⁰Pb) and its error are calculated using a weighted average of activities

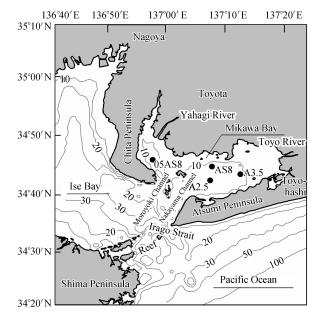


Fig. 1 Sampling sites (solid circle) in Mikawa Bay, Japan.

of ²¹⁴Pb and ²¹⁴Bi. Excess ²¹⁰Pb profiles were analyzed using a program named LEADAT (Lu and Matsumoto, 2006). In addition, the activities of ¹³⁷Cs were measured by counting its 661.63 keV gamma radiation.

2 Results

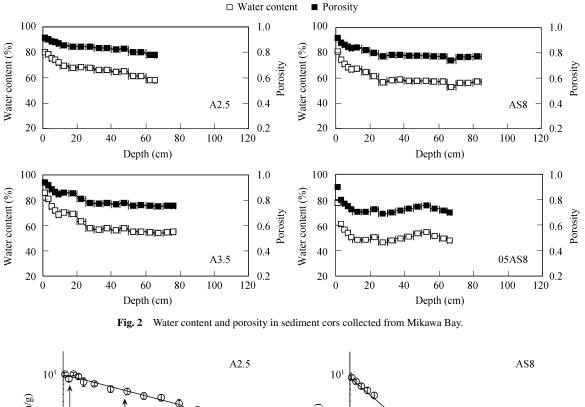
The collected sediments are generally grayish homogeneous clay or silty clay. However, sediments for core 05AS8 are characterized as greenish gray homogenous silt with fine sand. Fine sedimentary structures such as lamina were not found in any cores.

To remove the compaction effect, mass depth (g/cm^2) instead of depth (cm) has been used (Lu and Matsumoto, 2005a, 2005c; Lu, 2007). Mass depth was calculated using the data of water content and porosity shown in Fig. 2. Density of solid phase in sediment is assumed to be 2.55 g/cm³ (Robbins, 1978). Sedimentation rates were determined by the slope of excess ²¹⁰Pb profiles using least squares regression (i.e., the CIC model, Appleby and Oldfield, 1992). Mixed layers were determined by the flatten activities of excess ²¹⁰Pb in upper parts of the cores (Fig. 3). The mass depth vs. date curves derived from the excess ²¹⁰Pb profiles have been given in Fig. 4 to compare with the ¹³⁷Cs penetration mass depth.

Table 1 Locations, water depth, and the chararastics of ²¹⁰Pb and ¹³⁷Cs

Core	Latitude (N)	Longitude (E)	Water depth (m)	Sampling date	Supported ²¹⁰ Pb	Excess ²¹⁰ Pb		¹³⁷ Cs
						Inventory (dpm/cm ²)	Flux (dpm/ (cm ² ·year)	Inventory (dpm/cm ²)
A2.5	34°42.58′	137°07.49′	17	July 17, 2003	1.15 ± 0.17	111.58 ± 3.13	3.47 ± 0.10	7.61 ± 0.39
A3.5	34°43.71′	137°12.45′	12	July 17, 2003	1.04 ± 0.17	35.25 ± 1.74	1.10 ± 0.05	2.89 ± 0.20
AS8	34°45.05′	137°07.59′	12	July 17, 2003	0.98 ± 0.07	35.55 ± 1.86	1.11 ± 0.06	1.79 ± 0.35
05AS8	34°46.19′	136°57.91′	15	Sep 17, 2005	0.94 ± 0.14	276.66 ± 4.59	8.62 ± 0.14	11.57 ± 0.76
				-		$(70.94 \pm 4.87*)$	$(2.21 \pm 0.15^*)$	$(2.94 \pm 0.80^*)$

* Values denotes that the contribution from the Tokai Flood in 2000 has been removed. dpm: decay per minute.



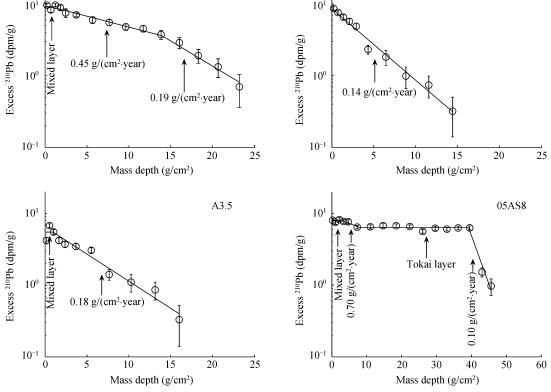


Fig. 3 Profiles of excess ²¹⁰Pb (1 dpm/g = 60 Bq/g). Sedimentation rates have been calculated from the least squares lines.

2.1 Mixing mass depth and sedimentation rates derived from ²¹⁰Pb profiles

As shown in Fig. 3, for core A2.5, the mass depth for its mixed layer is 0.60 g/cm² (2.61 cm). Sedimentation rates for sections in 0.60–13.95 g/cm² and below 13.95 g/cm² are 0.45 and 0.19 g/(cm²·year), respectively. The shift in sedimentation rate occurred in ca. 1970 (A.D.).

For core A3.5, the mixed layer is down to the mass depth, 0.92 g/cm^2 (4.41 cm). Sedimentation rate below the

mixed layer is 0.18 g/(cm²·year). For core AS8, sedimentation rate is 0.14 g/(cm²·year), and there is no clear mixed layer.

The excess ²¹⁰Pb profile for core 05AS8 is complex. The mixed layer is down to 3.11 g/ cm² (6.63 cm). Sedimentation rate for section 3.11–8.05 g/cm² is 0.70 g/(cm²·year); excess ²¹⁰Pb at mass depth from 8.05 to 39.17 g/cm² (ca. 10–55 cm) shows almost no change; sedimentation rate below 39.17 g/cm² is 0.10 g/(cm²-year).

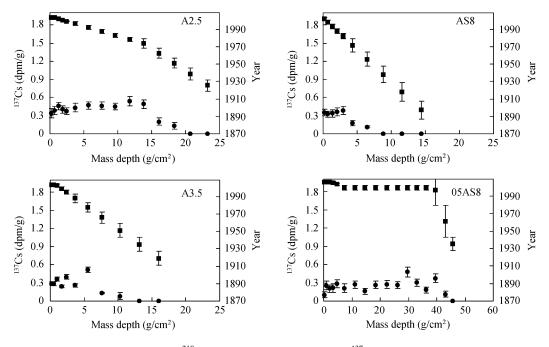


Fig. 4 Mass depth vs. ²¹⁰Pb-derived dates (solid squares) and ¹³⁷Cs profiles (solid circles).

2.2 ¹³⁷Cs profiles

In Japan, ¹³⁷Cs was first introduced into the environment in the early 1950s and showed a fallout peak in 1963 (e.g., Matsumoto, 1987). In sedimentary environments, the 1963 peak is often flattened or absent due to one or more factors, such as the mixing effect, the additional catchment-derived input, averaging effects within residence time (system time averaging) and sectioning thickness (Kim et al., 1997; Robbins et al., 2000; San Miguel et al., 2004; Lu and Matsumoto, 2005a). As shown in Fig. 4, the 1963 peak is not obvious for the profiles of ¹³⁷Cs for the four cores in Mikawa Bay. Therefore, the maximum penetration mass depth of ¹³⁷Cs, represented as the early 1950s, was used to compare with the ²¹⁰Pb-derived date vs. mass depth curve. The maximum penetration mass depths of 137 Cs are 18.29 ± 1.11 , 10.27 ± 1.42 , 6.46 ± 1.12 , and 43.05 ± 1.83 g/cm² for cores A2.5, A3.5, AS8 and 05AS8, respectively. The corresponding ²¹⁰Pb-derived dates at the above maximum mass depths are 1951 ± 6 , 1951 ± 8 , 1956 ± 8 , and $1961 \pm$ 18 (A.D.), showing in good consistency with 137 Cs dating within dating errors.

2.3 Inventories and fluxes for excess ²¹⁰Pb and ¹³⁷Cs

Excess ²¹⁰Pb and ¹³⁷Cs inventories were calculated as the sum of excess ²¹⁰Pb and ¹³⁷Cs through a sediment core, respectively. The mean input flux of excess ²¹⁰Pb was estimated using that inventory times the decay constant of ²¹⁰Pb. The results are listed in Table 1.

3 Discussion

3.1 Implications of the excess ²¹⁰Pb profiles

Due to urbanization and industrialization in the coastal areas, it is well known that metal concentrations in coastal

marine sediments generally show an increasing trend from the bottom to the surface (e.g., Matsumoto 1988; Dai et al., 2007). Owen and Lee (2004) also recognized an increasing trend in sedimentation due to coastal reclamation and dredging. Indeed, a generally increasing trend in sedimentation rates was previously found in Ise Bay due to environmental alteration of coast and hinterland over the last century (Lu and Matsumoto, 2005c). This might be used to explain the sedimentation rate change in ca. 1970 for core A2.5, which might be a local depocenter in the eastern part of Mikawa Bay according to its topography. There are, however, no obvious changes in sedimentation rate for cores A3.5 and AS8. Similarly, according to the bathymetric topography (Fig. 1), core 05AS8, where water depth is deep, might be the other depocenter in the western part of Mikawa Bay. The abrupt shift in sedimentation rate indicates that the local sedimentary environment has changed greatly.

For core 05AS8, the uniform excess ²¹⁰Pb for section 8.05–39.17 g/cm² (ca. 10–55 cm) might be related to the Tokai Flood in the Yahagi River catchment in September 2000 (Ushiyama and Takara, 2002). This estimation is supported by the dating results shown in Fig. 4. The Tokai layer was dated as 2000 ± 3 (A.D.) by ²¹⁰Pb geochronology. As reported by Ohno (2007), the concentrations of Cr, Co, and Zn in a duplicate core of core 05AS8 also appeared to have no significant change in section 3.11-8.05 g/cm² (10–55 cm), which is similar to excess ²¹⁰Pb profile. The profiles of metals reflect that the section 10–55 cm might be deposited during one sedimentation event, i.e., the Tokai Flood.

The lack of the Tokai layer in the adjacent Ise Bay may be attributed to the much weaker circulation and slower water exchange in Mikawa Bay than those in Ise Bay (Saijo, 2002). Namely, the flood discharge in Ise Bay might disperse more broadly than that in Mikawa Bay. Moreover, the maximum rainfall was observed in the upstream of the Yahagi River during the Tokai flood (Ushiyama and Takara, 2002).

In addition, core 05AS8 has a mixing depth of 0– 6.63 cm and shows evidence that eutrophication and weak exchange of bay water caused oxygen depletion in the deep layer (Saijo, 2002). Likely, bioturbation would therefore be limited due to the oxygen depletion.

3.2 Inventories and fluxes for excess ²¹⁰Pb and ¹³⁷Cs

The ²¹⁰Pb flux from atmosphere to land has been reported 1.5-2.8 dpm/(cm²·year) at Hakodate (Fukuda and Tsunogai, 1975) and 0.8-1.2 dpm/(cm²·year) in Osaka and Tokyo (Yamamoto et al., 2006). The higher fluxes for cores A2.5 and 05AS8 indicate a supplementary riverine input of excess ²¹⁰Pb. In particular, the Tokai Flood in 2000 has a significant contribution for core 05AS8. If subtracting the excess ²¹⁰Pb within the Tokai layer, the excess ²¹⁰Pb inventory and flux for core 05AS8 is 70.94 dpm/cm² and 2.21 dpm/(cm²·year), respectively, which can be interpreted by the direct atmospheric flux. Furthermore, the range of the excess ²¹⁰Pb fluxes (1.10–2.21 dpm/(cm²·year)) for the four cores in Mikawa Bay coincides the range in the nearby Ise Bay (0.51–4.31 dpm/(cm²·year)) as reported by Lu and Matsumoto (2005c). In addition, the lower excess ²¹⁰Pb fluxes for cores A3.5 and AS8 might be related to the relatively low scavenging efficiency of excess ²¹⁰Pb from the water column due to the low sedimentation rates.

The observed values of 137 Cs inventory range from 1.79 to 11.57 dpm/cm², which coincide with 1.14–34.92 dpm/cm² at Lake Nakaumi (Kanai *et al.*, 2002) and 2.54–29.50 dpm/cm² at Ise Bay (Lu and Matsumoto, 2005c). Taking radioactive decay into account, the total fallout of 137 Cs in Tokyo until 2003 is 17.99 dpm/cm² (Matsumoto, 1987; Lu, 2004; Meteorological Research Institute of Japan, 2005). The 137 Cs inventories for all cores are lower than this. This may be caused by the inefficient scavenging and the mobility of 137 Cs (e.g., Simpson *et al.*, 1976; Sholkovitz and Mann, 1984; Cooper *et al.*, 1995; San Miguel *et al.*, 2004).

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References

- Appleby P G, Oldfield F, 1992. Application of lead-210 to sedimentation studies, In: Uranium-series Disequilibrium: Application to Earth, Marine, and Environmental Sciences (Ivanovich M, Harmon R S, eds.). 731–778.
- Bodergat A M, Rio M, Ikeya N, 1997. Tide versus eutrophication:

impact on ostracods populations structure of Mikawa Bay (Japan). *Revue de Micropaléontologie*, 40: 3–13.

- Coltorti M, 1997. Human impact in the Holocene fluvial and coastal evolution of the Marche region, Central Italy. *Catena*, 30: 311–335.
- Cooper L W, Grebmeier J M, Larsen I L, Solis C, Olsen C R, 1995. Evidence for re-distribution of ¹³⁷Cs in Alaskan tundra, lake, and marine sediments. *The Science of the Total Environment*, 160/161: 295–306.
- Dai J C, Song J M, Li X J, Yuan H M, Li N, Zheng G X, 2007. Environmental changes reflected by sedimentary geochemistry in recent hundred years of Jiaozhou Bay, North China. *Environmental Pollution*, 145: 656–667.
- Fukuda K, Tsunogai S, 1975. Pb-210 in precipitation in Japan and its implication for the transport of continental aerosols across the ocean. *Tellus*, 27: 514–521.
- Hansom J D, 2001. Coastal sensitivity to environmental change: a view from the beach. *Catena*, 42: 291–305.
- Meteorological Research Institute of Japan, 2005. Artificial Radionuclides in the Environment 2005. http://www.mrijma.go.jp/Dep/ge/reportsJP.htm.
- Kanai Y, Yamamuro M, Inouchi Y, Tokuoka T, 2002. Sedimentation rate and environment of Lake Nakaumi, Shimane and Tottori Prefectures. *Geochemistry*, 36: 161–178.
- Kim G, Hussain N, Church T M, Carey W L, 1997. The fallout isotope ²⁰⁷Bi in a Delaware salt marsh, a comparison with ²¹⁰Pb and ¹³⁷Cs as a geochronological tool. *The Science of Total Environment*, 196: 31–41.
- Koide M, Soutar A, Goldberg E D, 1972. Marine geochronology with ²¹⁰Pb. *Earth and Planetary Science Letters*, 14: 442– 446.
- Lu X Q, 2004. Application of the Weibull extrapolation to ¹³⁷Cs geochronology in Tokyo Bay and Ise Bay, Japan. *Journal of Environmental Radioactivity*, 73: 169–181.
- Lu X Q, Matsumoto E, 2005a. How to cut a sediment core for ²¹⁰Pb geochronology. *Environmental Geology*, 47: 804– 810.
- Lu X Q, Matsumoto E, 2005b. How to cut a sediment core for ²¹⁰Pb geochronology: A supplement. *Environmental Geology*, 49: 251–252.
- Lu X Q, Matsumoto E, 2005c. Recent sedimentation rates derived from ²¹⁰Pb and ¹³⁷Cs methods in Ise Bay, Japan. *Estuarine, Coastal and Shelf Science*, 65: 83–93.
- Lu X Q, Matsumoto E, 2006. LEADAT: a MATLAB-based program for lead-210 data analysis of sediment cores. *Acta Oceanologica Sinica*, 25(6): 128–137.
- Lu X Q, 2007. A note on removal of the compaction effect for the ²¹⁰Pb method. *Applied Radiation and Isotopes*, 65: 142– 146.
- Matsumoto E, 1987. Pb-210 geochronology of sediments. *Natural Environment*, 3: 187–194.
- Matsumoto E, 1988. Residence times of trace metals and nutrients in Tokyo Bay water. In: Oceanic Processes in Marine Pollution Volume 5: Urban Wastes in Coastal Marine Environments, (Wolfe D A, O'Connor T P, eds.). Florida: Robert E. Krieger Publishing Company. 211–218.
- Ohno H, 2007. Composition of organic solvent extractable metals as an indication of coastal pollution. Master thesis, Nagoya University, Japan.
- Owen R B, Lee R, 2004. Human impacts on organic matter sedimentation in a proximal shelf setting, Hong Kong. *Continental Shelf Research*, 24: 583–602.
- Palinkas C M, Nittrouer C A, 2007. Modern sediment accumulation on the Po shelf, Adriatic Sea. *Continental Shelf*

Research, 27: 489-505.

- Ritchie J C, McHenry J R, 1990. Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: a review. *Journal of Environmental Quality*, 19: 215–233.
- Robbins J A, 1978. Geochemical and geophysical applications of radioactive lead. In: Biogeochemistry of Lead in the Environment, (Nriagu J O, ed.). Amsterdam: Elsevier Scientific. 285–393.
- Robbins J A, Holmes C, Halley R, Bothner M, Shinn E, Graney J, Keeler G, tenBrink M, Orlandini K A, Rudnick D, 2000. Time-averaged fluxes of lead and fallout radionuclides to sediments in Florida Bay. *Journal of Geophysical Research*, 105: 28805–28821.
- Saijyo Y, 2002. Natural History of Closed Embayment: to Rejuvenation of Mikawa Bay. Nagoya: Arumu Co., Ltd. 76.
- Saijo Y, Unoki S, 1977. Oceanographic characteristics and primary production of Ise Bay and Mikawa Bay. *Bulletin on Coastal Oceanography*, 14: 10–18.
- San Miguel E G, Bolivar J P, Garcia-Tenorio R, 2004. Vertical distribution of Th-isotope ratios, ²¹⁰Pb, ²²⁶Ra and ¹³⁷Cs in sediment cores from an estuary affected by anthropogenic releases. *The Science of the Total Environment*, 318: 143– 157.

- Sholkovitz E R, Mann D R, 1984. The porewater chemistry of ^{239,240}Pu and ¹³⁷Cs in sediments of Buzzards Bay, Massachusetts. *Geochimica et Cosmochimica Acta*, 48: 1107–1114.
- Siakeu J, Oguchi T, Tatsuto A, Esaki Y, Jarvie H P, 2004. Changes in riverine suspended sediment concentration in central Japan in response to late 20th century human activities. *Catena*, 55: 231–254.
- Simpson H J, Olsen C R, Trier R M, Williams S C, 1976. Manmade radonuclides and sedimentation in the Hudson River estuary. *Science*, 194: 179–183.
- Smith J N, 2001. Why should we believe ²¹⁰Pb sediment geochronologies? *Journal of Environmental Radioactivity*, 55: 121–123.
- Ushiyama M, Takara K, 2002. Characteristics of heavy rainfall in Tokai District in September 2000, based on precipitation data since 1901. *Journal of Natural Disaster Science*. 21(2): 145–159.
- Wakabayashi K, 2000. History of Environmental Problems in Tokyo Bay. Tokyo: Yohikaku Publishing Co., Ltd. 408.
- Yamamoto M, Sakaguchi A, Sasaki K, Hirose K, Igarashi Y, Kim C K, 2006. ²¹⁰Pb and ⁷Be deposition: features of the Japan Sea side of Japan, *Journal of Environmental Radioactivity*, 86: 110–131.

