Lead-210 in the Japan Sea*

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Abstract: This is the first detailed study on the distribution of lead-210 in the Japan Sea water. The content of lead-210 ranged from 9.3±2.1 dph/l in the surface water to 3.4±0.8 dph/l in the deep water—a quite low content as compared to that in the deep water of the North Pacific. Vertical profiles show that the content of lead-210 abruptly decreases below the seasonal thermocline (10–20 m in depth) and nearly uniform in the deep water. It is suggested that a significant amount of air-borne lead-210 deposited over the Japan Sea is transported along with the Tsushima Current to the open ocean. The budget of lead-210 is calculated by using a simple box-model and the mean residence time of lead-210 in the Japan Sea is estimated to be 15 yr.

1. Introduction

The hydrography of the Japan Sea has been studied by many workers (e.g. UDA, 1934; HATA, 1962 and FUKUOKA, 1965) and considered to be relatively simple: The main water mass, known as the Japan Sea Proper Water, is semi-closed, because the Japan Sea opens to outer only through the four narrow straits shallower than 200 m. The Tsushima Warm Current, runs in the southern sector from southwest to northeast along the Japanese Islands (Figure 1). In winter, cooling in the northern part of the Japan Sea induces a vigorous convective mixing which carries down the surface water to the deep and results in the high level of dissolved oxygen concentration more than 4 ml/l throughout the entire water body. While, in summer, a well-developed seasonal thermocline (10–20 m in depth) is formed in the whole area of the Japan Sea.

The present study is concerned with the distribution and behavior of naturally occurring lead-210 (half life, 21.4 yr) in the Japan Sea. Lead-210 in the surface water is mainly derived from air-borne lead-210 which is scavenged by meteoric precipitation after its production from radon-222 in the atmosphere. In the deep water, the content of lead-210 is balanced by supplies from the surface and the decay of its progenitor, and by removal as settling particulate. The distribution of lead-210 in the deep water may visualize the general behavior of

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Fig. 1. Sampling locations and the schematic movement of the currents (HATA, 1962) in the Japan Sea. Triangles, open circles, solid circles and crosses indicate the stations of Tansei-Maru (KT-67-6), Seifu-Maru (1968), Seifu-Maru (1969) and Hakuko-Maru (KH-70-4), respectively. The numbers in the figure show the stations where the deep samples were collected.
heavy metals in the sea.

The first determination of lead-210 in seawater was carried out by RAMA, KOIDE and GOLDBERG (1961), who estimated the short residence time of lead less than 2 yr in the surface mixed layer down to 100 m in depth. TSUNOGAI and NOZAKI (1971) found the pronounced latitudinal variation in concentrations of lead-210 in the surface water of the Pacific, which showed the highest value in the northern mid-latitudes around 30°N, and concluded that the concentration of lead-210 in the surface water reflects its deposition rate from the atmosphere. Furthermore, the observed surface concentrations in the North Pacific did not vary longitudinally and revealed the rapid transport of lead-210 from the continent to the ocean through the atmosphere (NOZAKI and TSUNOGAI, 1973). CRAIG, KRISHNASWAMI and SOMAYAJULU (1973), and NOZAKI and TSUNOGAI (1973) found the depletion of lead-210 relative to radium-226 in the deep water of the Pacific, owing to the rapid removal of lead by settling as particles even in the deep water.

We intended to clarify the distribution of lead-210 in the Japan Sea with special reference to the hydrographic condition and behavior of lead in the deep water.

2. Samples and analytical procedure

Samples were collected at the 4 cruises from 1967 to 1970 (Table 1 and Figure 1). Most of samples were collected in summer.

The analytical procedure for lead-210 in seawater has been described (NOZAKI and TSUNOGAI, 1973). For each determination, a 30-501 portion of seawater was used. Lead-210 was determined by counting α-activity of polonium-210, a granddaughter of lead-210, after storage of several months. The counting error was less than ±8%.

3. Results and discussion

Distribution of lead-210 in the Japan Sea

Horizontal distribution of lead-210 in the surface water (0 m in depth) is shown in Figure 2. The average content of lead-210 was 9.3±2.1 dph/l, which is nearly comparable with, or somewhat lower than, those observed in the western North Pacific. The relatively higher contents were observed in the warm water region of the Tsushima Current. The contents lower than 8 dph/l observed in the coastal water near the Japanese Islands may be due to the active scavenging effect of larger particulate materials derived from the land.

Vertical profiles of lead-210 down to 500 m in depth (Figure 3) show a different feature compared to those observed in the North Pacific (NOZAKI and TSUNOGAI, 1973). The content of lead-210 abruptly decreases at a depth below the seasonal thermocline layer.
Fig. 3. Vertical distribution of lead-210 and salinity down to 500 m in depth. Open circles and crosses represent lead-210 and salinity, respectively.

Fig. 4. Distribution of lead-210 in the deep water. The data at KT-4, KT-5, ST-19, ST-21, St-26, St-41, St-50, KH-2, KH-5 and KH-8 are plotted.

and 120 m. The content of lead-210 of the Tsushima Warm Water, is very low, about 4 dph/l, inspite of its long way through the region receiving the heavier input of lead-210 from the atmosphere. A possible explanation is that lead-210 in the original water forming the Tsushima Current has been effectively scavenged by detrital materials whirling from the bottom or derived from the land, in passing through the shallow continental shelf and the Tsushima Strait.

The content of lead-210 in the deep water is $3.4 \pm 0.8$ dph/l, as an average, and nearly constant below a depth of 500 m (Figure 4). This content is quite low compared to those in the deep water of the North Pacific where the observed content is always higher than 10 dph/l and increases with increasing depth, up to 17 dph/l at 3,500 m in depth (NOZAKI and TSUNOGAI, 1973). Since the content of radium-226 is uniformly about 6 dph/l in the surface water of the oceans and increases with increasing depth owing to the dissolution of radium-226 from the bottom sediment (e.g. BROECKER, Li and CROMWELL, 1967; SZABO, 1970, etc.), it is expected that the content of radium-226 is 6 dph/l or more in the deep water of the Japan Sea. Even if we use 6 dph/l as a mean content of radium-226 in the deep water, the content of lead-210 is only a half of its radiochemical equilibrium concentration with radium-226 in the deep water. This fact clearly reveals that lead-210 is removed from the deep water
of the Japan Sea by its settling.

**Horizontal transport of lead-210 in the surface water**

The surface water above the seasonal thermocline which appears in summer and vanishes in winter is characterized by the low salinity of 33.0–33.8 % and the high temperature of 20°–30°C. Judging from its low salinity, the surface water is formed by mixing the sub-surface water below the seasonal thermocline with fresh waters, such as meteoric precipitation and river water.

The content of lead-210 in the surface water is higher by about 5 dph/l than that in the sub-surface and deep water, owing to the input of air-borne lead-210. The deposition rate of lead-210 from the atmosphere was estimated to be 2.0 dpm/cm²/yr at Hakodate, Japan, by FUKUDA and TSUNOGAI (1972). Therefore, the increase rate of lead-210 becomes 5 dph/l/month in the surface water of 20 m thickness. By using the increase rate and the concentration difference between in the surface water and in the deeper water (5 dph/l), the mean life of the surface water in the Japan Sea is found to be more than 1 month. The lower limit, one month is the case when the transfer of lead-210 from the surface to the deep is neglected.

Although the uncertainty of the calculated value may be large, the mean life of the surface water is shorter than the period of stratification of water in the Japan Sea—probably several months. This requires that the surface water is renewed and transported from the Japan Sea to the open ocean along with the Tsushima Current. Thus, it is suggested that only a small portion of lead-210 deposited from the atmosphere penetrates to the deeper water in the Japan Sea. This is, partly, the reason why the content of lead-210 is low in the deep water of the Japan Sea.

Based on the above consideration, a transport model of lead-210 in the Japan Sea in summer (Figure 5) is constructed by referring the distribution of temperature and salinity and the model proposed by SUGIURA and YAMAMOTO (1968). Because air-borne lead-210 is continuously supplied to the sea-surface, the flow of the surface water illustrated in Figure 5 may result in the distribution of the higher content of lead-210 in the warm water region of the Tsushima Current (Figure 2).

**Budgets of lead-210 in the Japan Sea**

As the Japan Sea is semi-closed, the budget of lead-210 (Figure 6) is estimated by using a simple box-model. At a steady state condition, the balance of lead-210 is given by the following equation.

\[ I + P = (\lambda + \mu)N \]

where, \( I \) (dpm/cm²/yr) is the input rate of air-borne lead-210 from the atmosphere, \( P \) (dpm/cm²/yr) is the production rate of lead-210 from radium-226 in seawater, \( \lambda \) (0.032 yr⁻¹) is the radioactive decay constant of lead-210 and \( \mu \)
(yr⁻¹) is the removal rate constant assuming that the removal rate is proportional to the amount of lead-210 (N, dpm/cm²). The mean residence time of lead-210 in the Japan Sea (τₑ, yr) is defined as 1/μ.

Taking 2,000 m as a mean depth of the Japan Sea, the amount of lead-210 in seawater (N) is 12 dpm/cm² (Figure 4). Assuming a mean radium-226 concentration of 0.1 dpm/l in the Japan Sea, Ρ is estimated to be 0.7 dpm/cm²/yr. The deposition rate of lead-210 from the atmosphere is 2.0 dpm/cm²/yr (Fukuda and Tsunogai, 1972). However, as discussed earlier, a significant amount of the airborne lead-210 is transported from the Japan Sea to the open ocean by the Tsushima Current. If the penetration of the airborne lead-210 into the deeper water takes place only in colder season of about half a year, in the northern cold water region, which occupies about a half of the whole area of the Japan Sea, only a quarter of the airborne lead-210 is virtually supplied to the deeper water of the Japan Sea. Therefore, the actual input rate from the atmosphere (I) becomes 0.5 dpm/cm²/yr.

Then, the sedimentation rate of lead-210 (μN) from the overlying water is estimated to be 0.8 dpm/cm²/yr. The sedimentation rate corresponds to the existence of 24 dpm excess lead-210 (relative to radium-226), on an average, in the surface layer of the Japan Sea sediments. This excess lead-210 may be used as a geochronological tool of the recent sediment in the Japan Sea (Koide, Soutar and Goldberg, 1972).

The residence time of lead-210 in the Japan Sea is calculated as follows,

\[
\tau_{ₑ} = \frac{12}{0.5 + 0.7 - 0.4} = 15 \text{ yr}
\]

The residence time obtained here agrees well with 230 days in the surface mixed layer of the western North Pacific down to 100 m in depth obtained by Nozaki and Tsunogai (1973), when we take each depth of the model into account. Although the value of \(\tau_{ₑ}\) varies from 5 yr (in the extreme case of \(I=2.0 \text{ dpm/cm}²/\text{yr}\)) to 40 yr (\(I=0 \text{ dpm/cm}²/\text{yr}\)), depending on the estimated deposition rate of lead-210 from the atmosphere, this agreement supports the validity of the discussion on the horizontal movement of lead-210 in the Japan Sea described in previous and present sections.

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References
日本海における Pb-210

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要旨: 日本海における Pb-210 の分布を初めて詳細に測定した。Pb-210 含量は表面水における 9.3±2.1 dph/l から深層水における 3.4±0.8 dph/l の範囲にあった。深層水の値は、太平洋深層水の濃度に比べて極めて低い。鉱直分布は夏季のサーキュラライン (10-20 m 深) 以深で Pb-210 含量が急激に減少し、深層水中ではほぼ一様であることを示している。日本海に大気圏から降下してきた Pb-210 のかなりの部分が対馬暖流とともに外洋に流れ出していることが推定される。現象を用いて日本海の Pb-210 の収支を計算し、表面を除いた日本海水中の Pb-210 の平均滞留時間を 15 年と見積もった。