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Distribution and Behavior of ^{239}Pu, ^{137}Cs and ^{60}Co in the Sediment at Urazoko Bay

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Radionuclides/Glycerol/Sediment/Normalization

The concentrations of ^{239}Pu and ^{137}Cs due to fall-out, and ^{60}Co originating from the nuclear power reactor in the sediments sampled at Urazoko Bay, Fukui Pref., Japan, were determined and their correlation with the particle size or the amount of glycerol adsorbed on the sediments as a measure of surface area of the sediment particles were examined. By the size segregation of sediment, the contents of ^{239}Pu, ^{137}Cs and ^{60}Co were found to reach to the highest value in the fraction below 37 μm in diameter. The concentrations of these radionuclides for each size fraction were directly proportional to the amount of glycerol adsorbed on the sediment particles. Based on these facts, the normalization of the concentration of these artificial radionuclides by the amount of adsorbed glycerol was attempted. For the fraction below 0.5 mm in diameter of various sediments collected at around Urazoko Bay during 1975-1977. It was confirmed that the contents of ^{239}Pu and ^{137}Cs per unit amount of adsorbed glycerol were nearly constant, respectively. Furthermore, the pollution characteristics of ^{60}Co in the sediments, such as the distribution and the accumulation, was clearly estimated by using such normalization.

INTRODUCTION

Around Wakasa Bay, Fukui Pref., Japan, nine commercial nuclear power reactors are operating at the coastal area, and releasing waste effluent into the marine environment. Although the sediments from Urazoko Bay situated at the western part of Tsuruga Bay showed some contaminations with ^{60}Co and ^{54}Mn caused by the accumulation of low level waste discharged during the past decade from the Tsuruga Nuclear Power Reactor (BWR : 331MW), any remarkable contamination due to nuclear reactor has not been detected in the other coastal areas[1].

Urazoko Bay is a small shallow bay and has a mouth to the Tsuruga Bay (Fig. 1). Its maximum depth is only 18-20 m at the central zone. The sediments sampled

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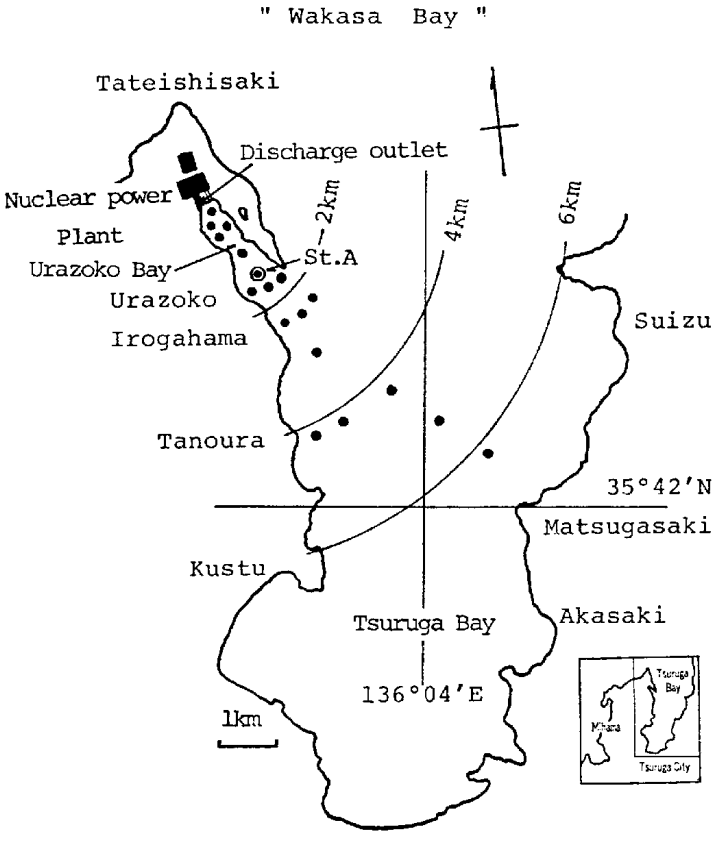

Fig. 1. Position of sediment discussed.

from the sea bottom near the inlet of the cooling water of reactor and the mouth of the bay about 2 km apart from discharge outlet consisted of 40–50% silt and clay, and that sampled at the central part of this bay consisted of 60–80% coarse and fine sand. The amounts of the natural organic components were about 1–3%[2].

Surface sediments, especially in coastal sea or bay, are known to accumulate artificial radionuclides to a large extent by the process of adsorption on the sediment or the sedimentation of suspended particles adsorbing radionuclides[3-7]. Therefore, sediments are one of the most important medium to understand the behavior of radinuclides in the aquatic environment. The uptake of radionuclides by bottom sediment particles is dependent on the nature of particles and the radionuclides in question. To fully understand the distribution and the behavior of radionuclides in the sediment, it is essential to know the difference of the sediment types which causes the wide variation of the concentrations of radionuclides, because the striking change in the composition and physico-chemical properties of sediment exist commonly even among the samples from various sampling points apart from each other

at short distance.

In this paper, the distribution and the behavior of fall-out radionuclides having long half life, such as ^{239}Pu and ^{137}Cs, were examined in comparison with those of ^{60}Co originating from the nuclear power reactor by taking into account of the characteristics of the sediment sampled at Urazoko Bay. Furthermore, to know the characteristics of the sediments, the amounts of glycerol adsorbed on the surface of sediment as a measure of surface area were measured together with particle size of the sediment.

MATERIALS AND METHODS

Sediments were sampled by the Ekman-Bridge bottom sampler at the spots shown in Fig. 1. The samples of 17 spots were sampled during 1975-1977 around Urazoko Bay, and the sample close to the mouth of the bay was sampled intermittently over four years to evaluate the accumulation of ^{60}Co. After rocks, pebbels and large shell were removed, the sediment was air-dried at 110°C for 24-48 hrs., and then pulverized to sift out with standardized sieves (32 mesh). Furthermore, for the study of the adsorption characteristics of the sediment particles, the sample collected from the vicinity of the discharge outlet of the waste from the nuclear power plant in August 1975 were separated into six fractions by passing through several standardized sieves, finest fraction being under 37 μm. In order to estimate the surface area of each sediment sample, Jackson's method[8] was used to measure the amount of glycerol adsorbed on the surface.

To determine the concentration of plutomium, the pluverized samples of 20-30 g for mud or 50-100 g for sand were subjected to radiochemical analysis after adding the known amount of ^{236}Pu as yield tracer. Plutonium was leached out by hot concentrated nitric acid, and the supernatant obtained by filteration was evaporated to dryness. The evaporation residue was dissolved in 8 M HNO_3 and fed on a column of Dowex 1×8 (100-200 mesh, 1 cm×5 cm) preconditionated with 8 M HNO_3. The column was washed with 8 M HNO_3 and then 10 M HCl to remove chemical elements other than plutonium. Then, plutonium was eluted with 10 M HCl/0.1 M HI solution. The eluent was evaporated to dryness and the small amount of organic materials in the eluent was decomposed with perchloric acid. The residue was taken up in 2 M H_2SO_4 with slight warming and the pH of the solution was adjusted to 2-3 with aqueous ammonia or dilute sulfuric acid to perpare the electrolytic solution for the electro-deposition of plutonium on a polished stainless steel disc. After electro-deposition, the stainless disc was rinsed with distilled water and then with acetone, followed by heating it to redness. The amounts and the isotopic composition of plutonium isotopes were determined by alpha spectrometry using Si (Au) surface barrier detector (Ortec : Sensitive area of 450 mm^2) coupled with 400-channel pulse-height analyser (Hitachi : Model PH-403). The chemical yield of plutonium was known by that of ^{236}Pu and found to be 60-80% usually. All values given as the amount of

^{239}Pu in this paper represent the sum of ^{239}Pu and ^{240}Pu, because the spectrometrical discrimination of alpha particle energy of ^{239}Pu and ^{240}Pu is difficult.

The determination of ^{90}Sr in the sediment was carried out by applying the official method[9]. In this method, ^{90}Y in the radioactive equilibrium with ^{90}Sr was extracted with yttrium carrier from radiochemically purified ^{90}Sr. And, the radioactivity measurement of ^{90}Y in the form of yttrium oxalate was made by using a low background gas-flow counter (Fujitsu: Model PBS-1).

The concentrations of ^{137}Cs, ^{60}Co and ^{40}K were analysed by nondestructive gamma-ray spectrometry using Ge (Li) detector (Horiba: Type GLC-153L-S) coupled with 4K-channel pulse-hight analyser (Nothern Scientific, Model NS-720) and minicomputer (Dick: Model PDP-11-05). Furthermore, the concentration of ^{226}Ra was also estimated from the gamma ray peaks of ^{214}Bi in the same spectrum by assuming the radioactive equilibrium.

RESULTS AND DISCUSSION

According to the previous study of marine environmental radioactivities in Urazoko Bay[2], the concentrations of ^{239}Pu and ^{137}Cs were found to be rather high in the sediments consisting largely of fine silt and clay. This fact prompts us to examine the distribution of radionuclides in each size fraction for the sediment collected in August 1975 (Table 1). It is reported by Hetherington *et al.*,[10] that the specific activity of alpha-emitting contaminants increased steadily from sand toward clay in the sediment particles of various size collected at the Newbiggin area near Windscale. The same tendency as Hetherington's observation is recognized for our

Table 1

The concentration of ^{239}Pu, ^{137}Cs and ^{60}Co among each size fraction in the sediment collected from the vicinity of the discharge outlet in August 1975

Size range (mm)	Pu-239	Cs-137	Co-60	Pu-239	Co-60
		(pCi/kg•dry)		Cs-137	Cs-137
2.000−0.500	6.2±0.7	19±7	—	0.33	—
0.500−0.250	8.5±0.8	18±7	34±8	0.47	1.9
0.250−0.105	9.5±0.9	36±6	26±6	0.26	0.7
0.105−0.074	13.0±1.9	23±5	19±7	0.57	0.8
0.074−0.037	14.6±1.7	59±6	56±5	0.25	1.0
<0.037	79.1±7.8	354±7	2527±47	0.22	7.1
*	38.3±3.2	164±4	1048±19		
**	39.1±2.8	185±10	1059±52	0.21	5.8

Errors quoted are those due to counting statistics.

* : The weighted mean concentration of every fractions.

**: The concentration of unsieved sample (<2 mm).

—: Below detection limit in gamma-ray spectrometry.

experimental results as shown in Table 1. The experimental conditions needed for size fractionation, such as the vigorous stirring of the suspension or the lapse of time, did not make any effect to cause the loss of the radionuclides from the samples. Therefore, the radionuclides once adsorbed on the sediments may be hard to be released into the sea water and their desorption may not occur during the movement of suspended sediment.

From our results, it was found that the activity ratios, $^{239}Pu/^{137}Cs$, calculated for each size fraction were in the range of 0.22-0.57, though the values of $^{60}Co/^{137}Cs$ scattered between 0.7-7.1. The $^{60}Co/^{137}Cs$ ratio for the fraction below 37 μm in diameter was found to be much higher than these ratios above 37 μm. Furthermore, on the basis of the concentration of radionuclides and the weight obtained for each size fraction (Table 1), the percentages of the content in each fraction to the total content in the sediment were calculated, and these results, including ^{90}Sr, ^{226}Ra and ^{40}K, are shown in Table 2 together with the percentage of weight. From these data, it was found that the fraction below 37 μm contained above 80% of ^{239}Pu, ^{137}Cs and ^{60}Co in the sediment, while the natural radionuclides, such as ^{226}Ra and ^{40}K, did not so conspicuously in the finest fraction as the artificial radionuclides mentioned above. According to the reports by Nagaya and Nakamura[11], it has been said that the percentage distributions of artificial radionuclides seem to be generally proportional to the percentage of the surface area, though those of natural radionuclides seem to be rather proportional to the percentage of the weight of each size fraction.

Referring to these results, the relationship between the content of radionuclides and the surface area of the sediment particles was examined to elucidate the uptake characteristics of the tracer amounts of elements by the sediment. The adsorption method of glycerol was chosen for this purpose, because of its simplicity in the procedure. The weight of glycerol adsorbed on the particles has been converted generally to the surface area by making use of the conversion factor derived for the montmorillonite. However, since the conversion factor may depend on the surface properties of different mineral particles, the glycerol adsorption amount itself expres-

Table 2

The percentages of the weight and the contents of several radionuclides in each fraction to the total content in the same sediment shown in Table 1.

Size (mm)		Weight (%)	^{239}Pu (%)	^{137}Cs (%)	^{90}Sr (%)	^{60}Co (%)	^{40}K (%)	^{226}Ra (%)
Range	Mean							
2.000−0.500	1.250	11.5	1.9	1.3	2.4	—	13.0	4.3
0.500−0.250	0.375	13.7	3.0	1.5	3.7	0.4	20.1	6.6
0.250−0.105	0.178	14.3	3.5	3.1	9.8	0.4	19.0	11.4
0.105−0.074	0.090	6.2	2.1	0.9	3.2	0.1	6.8	4.9
0.074−0.037	0.056	13.5	5.2	5.3	7.7	0.7	13.9	19.3
<0.037	0.019	40.8	84.3	87.9	73.2	98.4	27.2	53.5

sed by mg(glycerol)/g(soil) was used in this study for discussion. In the glycerol adsorption study, it has been said that the natural organic materials adsorbed on the sediment must be decomposed beforehand and then replaced the surface of sediment with cation in the simple form, because the amount of adsorbed glycerol is affected by the existence of organic materials[12,13] and the polar cationic materials[12,14]. To examine these effect for seven different samples collected from Urazoko Bay in July 1975, the comparative measurments of adsorbed glycerol were carried out for the untreated sediment and for the Ca-form sediment which was prepared by treating the untreated sediment with 1N $CaCl_2$ and 1N $Ca(CH_3COO)_2$ after the decomposition of organic matters with H_2O_2. These results gave only small discrepancy in the every samples, showing that the sediments from Urazoko Bay contain few organic matters and polar cationic ones. Based on this consideration, the untreated sample was commonly used in our work to determine the amount of adsorbed glycerol. The reproducibility of the amount of adsorbed glycerol was checked for the sediments which consisted largely of fine silt and clay or largely of coarse and fine sand. As a result of ten independent determinations, the reproducible values were obtained for both sediments, that is, 21.5±1.2 mg/g for the former sediment and 4.8±0.4 mg/g for the latter one as the mean value and the standard deviation.

The relationship between the concentration of ^{239}Pu and the amount of glycerol

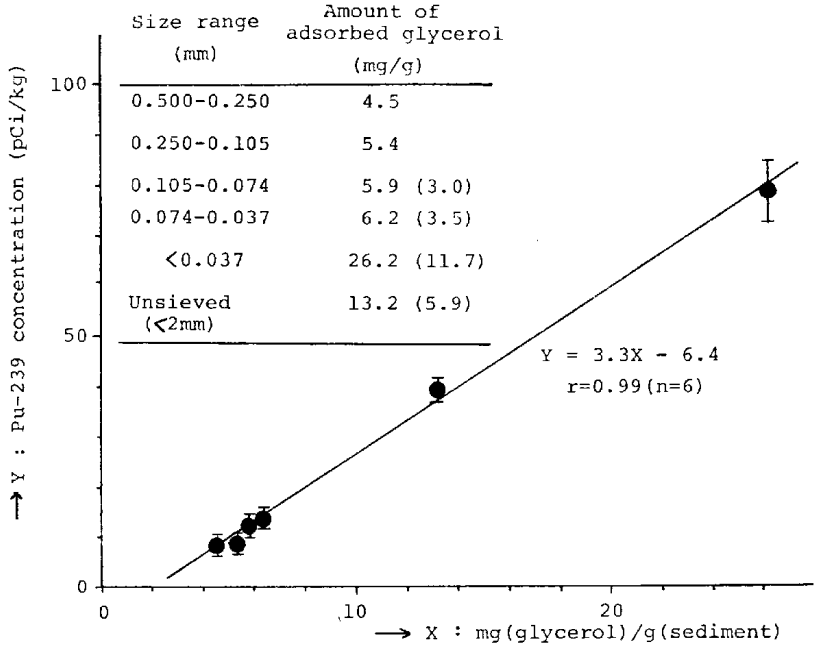

Fig. 2. Correlation of the concentration of ^{239}Pu with the amount of glycerol adsorbed in the each fractions shown in Table 1.

adsorbed on each size fraction was examined for the different size fraction shown in Table 1, and the linear correlation was observed as shown in Fig. 2. The regression line calculated is shown in Fig. 2 together with the simple correlation coefficient (r). Furthermore, the surface area estimated by B. E. T. method utilizing the adsorption of N_2-gas (Sibata: Model P-720) are also shown in the parenthesis. The conversion factor to obtain this surface area from the amount of adsorbed glycerol becomes 0.49 m^2/mg on the average. Similar linear correlations were obtained between the amount of adsorbed glycerol and the concentrations of ^{137}Cs and ^{60}Co, though the reproducibility was poor owing to few samples available.

On the basis of these facts, the normalization of the concentration of these artificial radionuclides by the amount of adsorbed glycerol was attempted. Because, Jones reported that the total activity in the sediment of Irish sea was proportional to the surface area of particles[15]. Futhermore, in the experiments using Dutch Wadden sea and Mediterranean sediment, Duursma and Eisma demonstrated that the total activity in each grain-size fraction showed a constant value per unit surface area of the particles[16]. For the fraction below 32 mesh of various sediments collected at around Urazoko Bay during 1975-1977, Figure 3 shows the relationship

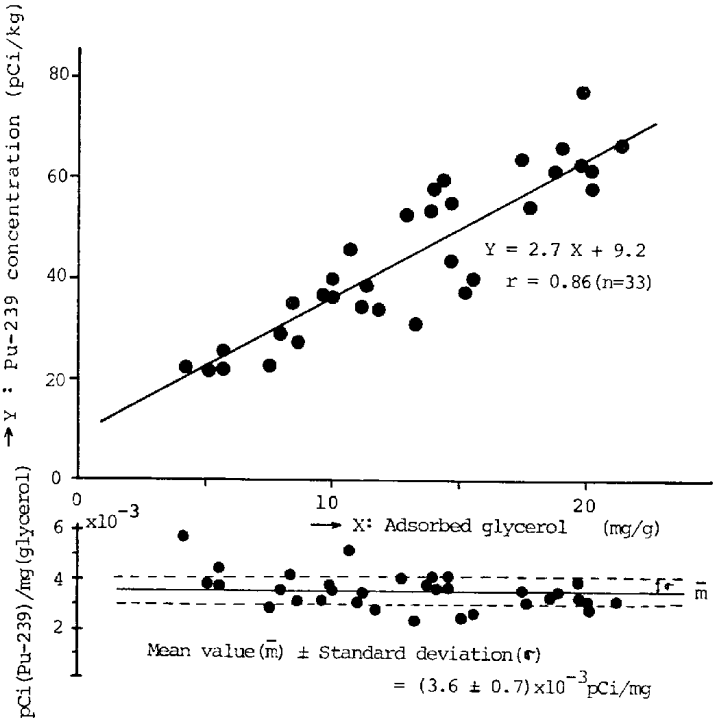

Fig. 3. The relationship between the concentration of ^{239}Pu and that of glycerol adsorbed for 33 samples collected from Urazoko Bay during 1975-1977. The content of ^{239}Pu normalized by the amount of adsorbed glycerol are given in the lower part of this figure.

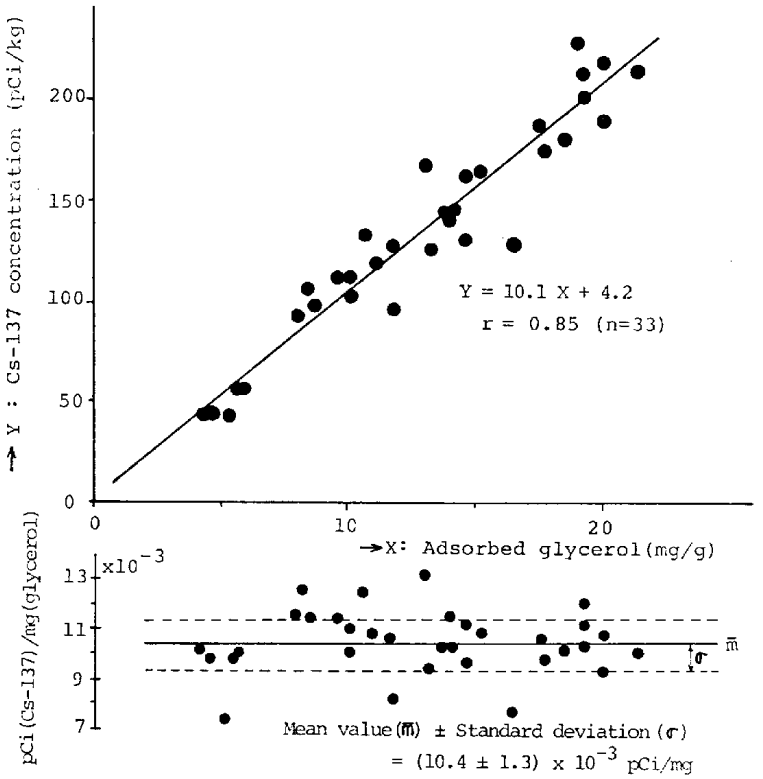

Fig. 4. The relationship between the concentration of ^{137}Cs and that of glycerol adsorbed for 33 samples collected from Urazoko Bay during 1975-1977. The content of ^{137}Cs normalized by the amount of adsorbed glycerol are given in the lower part of this figure.

between the concentration of ^{239}Pu and the amount of adsorbed glycerol. In the lower part of this figure, the contents of ^{239}Pu normalized by the amount of adsorbed glycerol are given. Figure 4 shows the similar examination for ^{137}Cs. The contents of ^{239}Pu or ^{137}Cs per unit amount of adsorbed glycerol were nearly constant, respectively, though the concentrations of ^{239}Pu scattered between 20-80 pCi/kg.dry and those of ^{137}Cs between 40-230 pCi/kg. dry. It's confirmed that the variation of concentrations of ^{239}Pu and ^{137}Cs in the sediments taken at Urazoko Bay becomes small by the normalization mentioned above. The activity ratios of $^{239}Pu/^{137}Cs$ at Urazoko Bay was 0.33 ± 0.08 on the avarage, which was not so different from the value for the sediments in other coastal sea or bay far from nuclear power stations. Therefore, ^{239}Pu and ^{137}Cs in the sediments of Urazoko Bay are thought to be due to the world-wide radioactive fall-out.

In addition, the base-exchage capacity of sediment for calcium was determined for the samples collected in 1977, a part of 33 samples shown in Fig. 3 or Fig. 4[17]. The simple correlation coefficient of 0.95(n=14) was found between the amount of

adsorbed glycerol and the base-exchange capacity. If we refer the work of Cheng and Hamaguchi[18] in which the role of exchangeable potassium in uptake of ^{137}Cs by sediment was noted, it seems reasonable that the concentration of ^{137}Cs is also proportional to the base-exchage capacity of the sediment. In the case of ^{239}Pu, the adsorption may be a dominant factor to concentrate this nuclides owing to its property to form radiocolloid easily and to be adsorbed to fine particles[19-22]. In spite of the rather different chemical properties of ^{239}Pu and ^{137}Cs, similar behavior of both nuclides in the environment has been reported by other researchers for the vertical profile of these nuclides in sediment[23]. Although it's difficult to explain this similar behavior explicitly, a good correlation between the amount of adsorbed glycerol and the base-exchange capacity may be a significant cause of this apparent similar behavior.

In the contrast to the data for ^{239}Pu and ^{137}Cs, the data for ^{60}Co seems to be very interesting from the point of view in regard to the pollution due to the nuclear power reactor. Previously, an investigation concerning the transport and the accumulation of ^{60}Co in the sediment of Urazoko Bay, was reported in detail by Nagaya and Nakamura[11], in which the pollution of bottom sediment with ^{60}Co was studied by using the concentration of ^{137}Cs as an indicator of adsorption capacity of the sediment. The concentrations of ^{60}Co for samples obtained during 1975-1977 were normalized by the amount of adsorbed glycerol and the correlation of these values with the distance from the discharge outlet of the nuclear power station was examined. The results

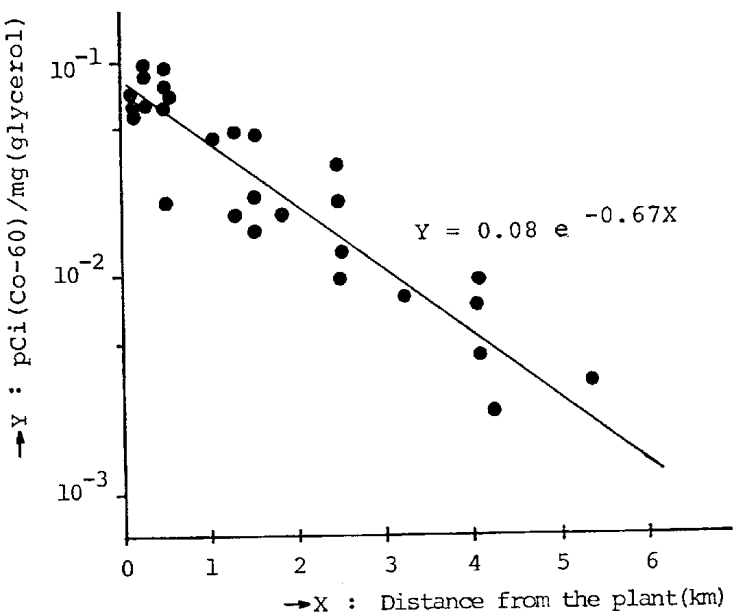

Fig. 5. The concentration of ^{60}Co normalized by the amount of adsorbed glycerol against the distance from the discharge outlet of the nuclear power plant.

are shown in Fig. 5. This relationship fits a simple exponential function, $Y=a\ e^{-bX}$, where Y is pCi (^{60}Co)/mg (glycerol), X is the distance in km, and a and b are constants. The calculated values of a and b for our study were 0.08 and 0.67, respectively. By dividing the value of pCi(^{60}Co)/mg(glycerol) by the that of pCi(^{137}Cs)/mg (glycerol) shown in Fig. 4, Y can be expressed in terms of the ratio of $^{60}Co/^{137}Cs$. In this case, "a" and "b" become 7.7 and 0.67, respectively, and these values agree well with the values of "a"=5.2 and "b"=0.71 reported by Nagaya and Nakamura[11]. The maximum concentration of ^{60}Co in sediment near the discharge outlet reduces to one-half at the point of about 1 km apart from discharge outlet.

Figule 6 shows the variation of ^{60}Co concentration in the sediment near to the mouth of Urazoko Bay where about 2 km distant from discharge outlet (St. A in Fig.

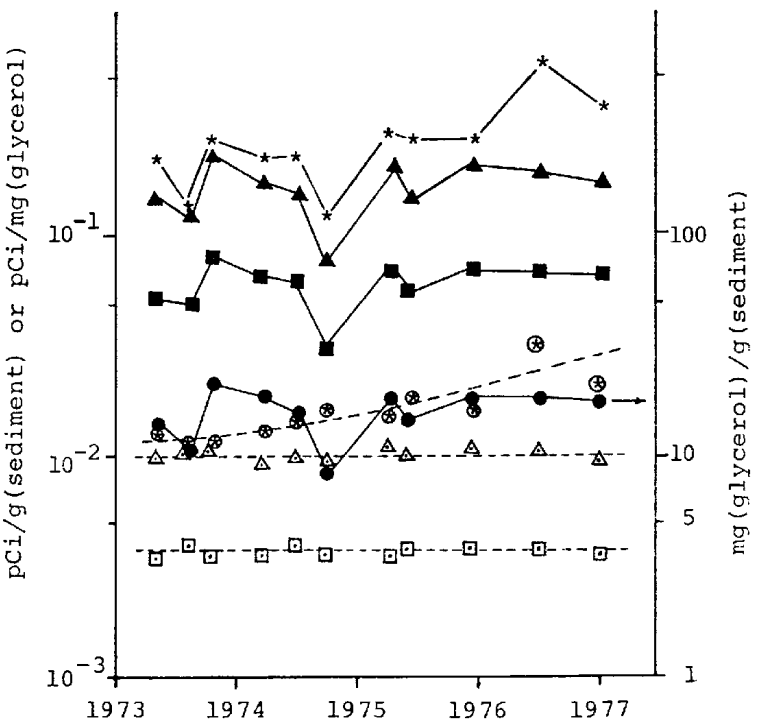

Fig. 6. The concentration of ^{239}Pu, ^{137}Cs and ^{60}Co and their relations to that normalized with the amount of glycerol adsorbed in the sediment at St. A, during 1973-1977.

—★—: Concentration of ^{60}Co(pCi/g).
—▲—: Concentration of ^{137}Cs(pCi/g).
—■—: Concentration of ^{239}Pu(pCi/g).
—●—: Amount of adsorbed glycerol (mg/g).
--⊛--: ^{60}Co
--△--: ^{137}Cs
--⊡--: ^{239}Pu
} Values normalized with the amount of adsorbed glycerol. (pCi/mg-glycerol)

1). It's interesting that even for those time variation data the contents of ^{239}Pu and ^{137}Cs normalized with the amount of adsorbed glycerol show nearly constant values, though the concentrations of both nuclides scattered widely. On the contrary, the normalized content of ^{60}Co is found to be apparently increasing. This fact suggests that the suspended sediment having high concontration of ^{60}Co in vicinity of the discharged outlet is moving more or less to the mouth of the bay. A peak of discharged radioactive nuclides was observed from late 1969 to early 1970 and its levels decreased by almost two or three orders of magnitude after this observation[1]. Furthermore, the regression equation for every year was calculared in a similar manner as the case of Fig. 5, and it was found that the slope of its equation, b, was gradually decreasing with year[24]. The normalization of ^{60}Co concentration by the amount of adsorbed glycerol may be reasonable as compared with the normalization by ^{137}Cs concentration, because this method can be applied even in the environment where the pollution with ^{137}Cs originating from a nuclear power plant or an irradiated fuel reprocessing plant may exist. Such normalization method of the concentration of radioactive nuclides by the surface area of sediment particles may also be effective to examine the depth variation of radionuclides as reported recently by Megumi for ^{210}Pb[25].

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REFERENCES

1. Environmental Radioactivity Survey around Nuclear Power Stations. (1975–1978) *Technical Committee on Environmental Radioactivity Measurement of Fukui Prefecture.* Vol. 7-Vol. 10.
2. M. Yamamoto, H. Hayakawa and T. Kitagawa (1977) Concentration of plutonium in the environmental samples. *Report of Fukui Prefectural Institute of Public Health,* 12: 22.
3. D.P. Kharkar, K.K. Turekian and K.K. Bertine (1968) Stream supply of discharged silver, molybdenum, antimony, selenium, chromium, cobalt, rubidium and cesium to the oceans. *Geochim. Cosmochim. Acta,* 32: 285.
4. C.M. Murray and L. Murray (1973) Adsorption-desorption equilibria of some radionuclides in sediment-fresh-water and sediment-seawater systems. In *Radioactive Contamination of the Marine Environment (IAEA-SM-158)*: 105.
5. R.E. Heft, W.A. Phillips, H.R. Ralson and W.A. Steele (1973) Radionuclide transport studies in the Humbolt Bay marine sediment. *ibid.*: 277.
6. D.E. Robertson, W.B. Silker, J.C. Langford, M.R. Petersen and R.W. Perkins (1973) Transport and depletion of radionuclides in the Columbia River. *ibid.*: 141.
7. K.C. Pillai, N.N. Dey, E. Mathew and B.U. Kothari (1975) Behaviour of discharged radionuclides from fuel reprocessing oparations in the aquatic environment of Bombay Harbour Bay. In *Impacts of Nuclear Releases into the Aquatic Environment (IAEA-SM-198)*: 277.
8. M.L. Jackson (1956) *Soil Chemical Analysis-Advanced Cource., Wiscon.*

9. Science and Technology Agency (1974) *Analytical Method of Radiostrontium.*
10. J.A. Hetherington, D.F. Jefferies and M.B. Lovett (1975) Some investigation into the behaviour of plutonium in the marine environment. In *Impacts of Nuclear Releases into the Aquatic Environment (IAEA-SM-198)* : 193.
11. Y. Nagaya and K. Nakamura (1975) Transport and accumulation of radiocobalt in the marine sediment of Urazoko Bay. *ibid.*: 313.
12. S. Diamond and E.B. Kinter (1958) Surface areas of clay minerals of glycerol retention. *Proc. 5th Nat. Conf. Clays. Clay and Clay Minerals*: 334.
13. J.R. Burford, T.L. Deshapand and D.J. Greenland (1964) Influence of organic materials on the determination of the specific surface area of soils. *J. Soil Sci.*, **15**: 192.
14. B.L. Mcneal (1960) Effect of exchangeable cations on glycerol retention by clay minerals. *Soil Sci.*, **97**: 96.
15. R.F. Jones (1960) The accumulation on nitrosyl ruthenium by fine particles and marine organisms. *Limnol. Oceanogr.*, **5**: 312.
16. E.K. Duursma and D. Eisma (1973) Theoretical, experimental and field studies concerning reaction of radioisotopes with sediments and suspended particles of sea, Part C: Applications to field studies. *Neth. J. Sea Res.*, **6**: 256.
17. S. Ganapathy, K.C. Pillai and A.K. Ganguly (1968) Adsorption of trace elements by near shore sea bed sediment. *BARC-376.*
18. H.G. Cheng and H. Hamaguchi (1968) Studies on the adsorption of radioisotopes on marine sediments, 1. Role of exchangeable cations on the adsorption of cesium and exchange equilibria. *Health Phys.*, **14**: 353.
19. M.L. Templeton and A. Preston (1966) Transport and distribution of radioactive effluents in coastal and esturarine waters into the seas, oceans and surface waters. *Proc. Symp. Vienna, 1966* IAEA, Vienna: 256.
20. Y. Miyake and Y. Sugimura (1968) Plutonium content in the Western North Pacific water. *Pap. Meteoral. Geophys., (Tokyo)* **19**: 481.
21. V.T. Bowen, K.M. Wong and V.E. Noshkin (1971) Plutonium-239 in and over the Atrantic Ocean. *J. Mar. Res.*, **29**: 1.
22. K.C. Pillai and E. Mathew (1976) Plutonium in the aquatic environment. Its behavior, distribution and significance. In *Transuranium Nuclides in the Environment (IAEA-SM-199)* : 25.
23. V.E. Nothkin (1972) Ecological aspects of plutonium dissemination in aquatic environment. *Health Phys.*, **22**: 537.
24. H. Takayama, M. Yoshioka and T. Kitagawa (1979) *Report of Fukui Prefectural Institute of Public Health,* (in preparation).
25. K. Megumi (1978) A problem in ^{210}Pb geochronogies in sediments. *Nature,* **274**: 885.