STUDIES ON THE RELEASE OF RADIOACTIVITIES FROM THE ION EXCHANGE RESINS INTO THE SEA WATER

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Abstract—For the purpose of estimating the release of radioactivities from ion exchange resins disposed into sea water, we have conducted a series of experiments in a water bath containing natural sea water, using such radioactive nuclides as ^{51}Cr, ^{55}Fe, ^{60}Co, ^{89}Sr and fission products by changing the weight ratio of ion exchange resin to the sea water and the amount of nuclide initially adsorbed on the ion exchange resin under static and turbulent conditions. The results are summarized as follows:

1. The radioactivities were rapidly released from the ion exchange resin particles (Dowex 50 and Dowex 1, 50–100 mesh) on contact with the sea water. However, some differences were observed in the degree of release depending on the different nuclides. Such nuclides as ^{60}Co and ^{89}Sr were easily released, while ^{51}Cr and ^{55}Fe not so easily.

2. The released activity was observed to increase with the decreasing weight ratio of the ion exchange resin to the sea water, but with the weight ratio below 10^{-4} of the ion exchange resin to the sea water no remarkable difference was observed.

3. The results of fractionation of suspended matter in sea water containing ^{55}Fe, ^{60}Co and ^{89}Sr will be discussed also.

1. INTRODUCTION

It is considered to be an important problem to investigate the behaviour and distribution of radioactivities in the eco-system in the ocean, when radioactive contamination and its effects due to radioactive waste disposal into sea water are studied.[1-6] If by any chance high-level activities of spent ion exchange resins were disposed into the sea, the possible effect on man who might consume the marine products contaminated with radioactivity should be adequately assessed. To consider the effects, it is necessary as a first step to investigate the percentage of the release of radioactivities adsorbed on the ion exchange resin and to investigate the behaviour and distribution of released radioactivities in the sea water. In order to estimate the release of radioactivities from the ion exchange resins disposed into sea water and to estimate the behaviour and distribution of radioactivities released into sea water, we have conducted a series of experiments in a water bath containing natural sea water, using several radioactive nuclides.

Further, the behaviour and distribution of radioactive nuclides in suspended matter in sea water were also studied experimentally by fractional filtration.

2. MATERIALS AND APPARATUSES

2.1. Sea Water

Natural surface sea water was sampled at about 50 m distance from the Misaki beach of Osaka-Bay. The total sodium content was estimated at 9500 ppm, the chlorinity 17.7 $^{\circ}/_{\circ\circ}$ and pH 8.0–8.2.

2.2. Ion exchange resins

H-types of cation exchange resin, Dowex 50-X8 (50–100 mesh), were used. Their exchange capacity was estimated to be 4.44 meq

for Na^+ per 1 g of dry resin, 3.37 meq for Sr^{++} and 3.49 meq for Co^{++}. Cl-types of anion exchange resin, Dowex 1-X8 (50–100 mesh), were also used. Their exchange capacity was estimated to be 1.91 meq for Cl^- per 1 g of dry resin.

2.3. *Radioactivities*

^{51}Cr ($CrCl_3$, HCl solution), specific activity 13.3 mCi/mg, 2.8 mCi/ml.
  (Commissariat à l'Energie Atomique)
^{55}, ^{59}Fe ($FeCl_3$, HCl solution) carrier free, 26.8 mCi/ml.
  (Nuclear Science and Engineering Co.)
^{60}Co ($CoCl_2$, HCl solution) Co 0.44 mg/ml, specific activity 55 mCi/mg, 24.2 mCi/ml.
  (Oak Ridge National Laboratory)
^{89}Sr ($SrCl_2$, HCl solution) carrier free 1.7 mCi/ml.
  (The Radiochemical Centre)

Each radioactive chemical was diluted, and about 10 or 100 μCi was adsorbed on ion exchange resin for one experiment.

2.4. For measuring β-radioactivities of samples a G-M counting system and a low-background gas-flow type counting system were used. A scintillation counter was used for counting γ-radioactivities.

2.5. A spectro-photometer was used to determine the quantities of cobalt and strontium carriers. Sodium content of sea water was measured with a flame-photometer, pH of sea water was measured with a pH-meter with glass electrodes.

2.6. Water baths used for experiments were made of acryl-plastics. The size was 1000 mm (length) × 500 mm (width) × 1000 mm (height). Cylindrical water baths made of glass and those of polyethylene were also used, the sizes of which were 113 mmϕ × 145 mm (height) and 300 mmϕ × 430 mm (height), respectively.

2.7. Sea water in the acryl plastic water bath was stirred with compressed air which was led into the center of the water bath through a vinyl chloride pipe (inner diameter 13 mm) connected to a blower-pump with a flow-rate of 1 l./sec (Fig. 1). A glass water bath and a stirrer with a stainless steel propeller (about 200 r.p.m.) were also used.
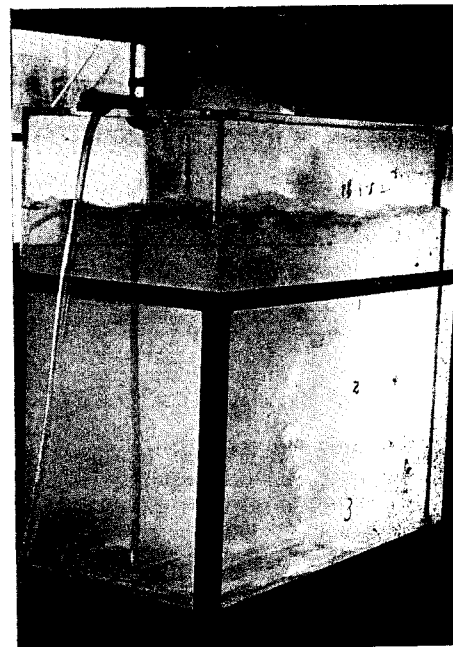
2.8. Suspended matter in the sea water was

FIG. 1. Experimental water bath.

filtered successively with Toyo-filter paper No. 5A (mean pore size 18 μ), milli-pore filter OS type (mean pore size 10 μ), SM type (mean pore size 5 μ), RA type (mean pore size 1 μ), and HA type (mean pore size 0.5 μ).

3. EXPERIMENTAL PROCEDURES

3.1. *Weight ratios of ion exchange resins to sea water*

Keeping the volume of sea water in the water bath constant, the weight of ion exchange resins mixed with the sea water was changed for the range of weight ratio from 10^{-3} to 10^{-7}.

3.2. *Initial carrier quantities in ion exchange resins*

The quantities of stable carriers adsorbed on the cation exchange resins were changed, keeping the radioactivity previously adsorbed on the resins constant.

3.3. *Released radioactivity from ion exchange resins*

In performing these experiments, ion exchange resins on which radioactive nuclides were adsorbed were placed in the sea water

contained in the water bath. An aliquot of the water (1 ml) was sampled with definite time interval at each of the 5–9 points indicated in Fig. 2 in order to determine the spacial distribution of the radioactivity in the sea water.
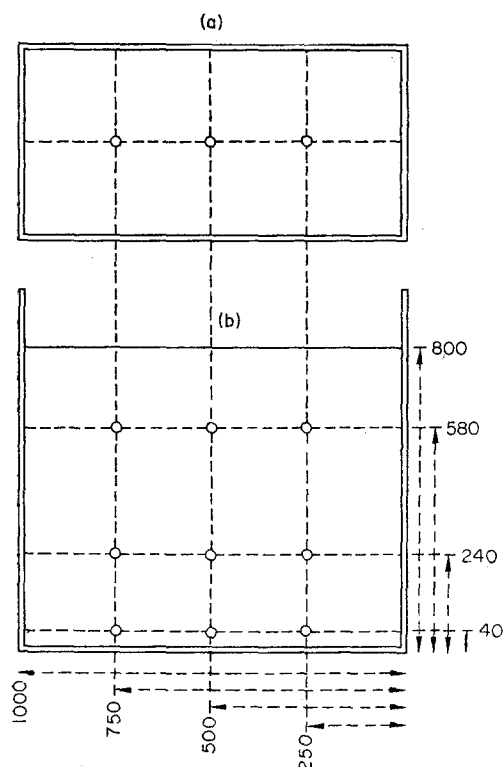

FIG. 2. Experimental water bath and sampling position.

After equilibrium, the percentage of released radioactivity was estimated from the residual radioactivity in the resins.

3.4. *Measurement of suspended matter in the sea water*

3.4. 1. *Fractionation of suspended matter*. At definite times an aliquot of sea water (1.5 l.) was sampled and filtered successively through five different types of filters, as listed in Table 1, in the order of pore sizes, to fractionate suspended matter in the sea water by suction with rotary vacuum pump. Each fraction was rinsed with distilled water. The chloride in the filtrate was tested with silver nitrate and the ammonium with Nessler's reagent.

3.4. 2. *Measurement of dry weight of suspended matter in sea water*. Fractionated suspended matter was dried in a desiccator until the weight became constant, and weighed.

3.4. 3. *Measurement of organic suspended matter*. Total nitrogen of dry samples was determined by the microkjeldahl method. The quantities of nitrogen were used as index of organic suspended matter. The amount of total organic matter was estimated by multiplying the nitrogen quantities by 17 according to D. L. Fox et al. [7]

3.5. *Concentration factor and accumulation fraction of radioactive nuclides in suspended matter in sea water*

Concentration factor is defined as the ratio of radioactivity (cpm/g) of each fraction of

Table 1. *Properties of Filters*

Filter	Material	Color	Thickness microns	Flow rate* ml/min. cm^2	Mean pore size microns
TR-No. 5A	Cotton	White	200	—	18
MF-OS	Cellulose plastic	White	280	500	10
MF-SM	Cellulose plastic	White	170	560	5
MF-RA	Cellulose plastic	White	150	300	1
MF-HA	Cellulose plastic	White	150	65	0.5

TR: Toyo Roshi (Toyo Filter Paper), Japan.
MF: Millipore filter, U.S.A.
* Flow rate of water at 25°C with a pressure 70 cm Hg.

suspended matter in the sea water per dry weight to the radioactivity concentration (cpm/ml) of final filtered sea water (sea water filtered with HA type filter).

Concentration factor

$$= \frac{\text{Radioactivity of each fraction per dry weight (cpm/g)}}{\text{Radioactivity concentration of final filtered sea water (cpm/ml)}}$$

At equilibrium state, the accumulation fractions of radioactive nuclides in each fraction were calculated by the following formula.

Accumulation fraction

$$= \frac{\text{Accumulated radioactivity in each fraction of suspended matter per liter of sea water containing suspended matter (cpm/l.)}}{\text{Total radioactivity per liter of sea water containing suspended matter (cpm/l.)}}$$

This final filtered sea water was again filtered through five other new filters of the same types used above. In each case residual activities on these filters were subtracted from the above values as backgrounds.

3.6. Autoradiography of accumulation of radionuclides in suspended matter

The autoradiograms of each fraction of suspended matter in the sea water were taken by the contact method. Fuji medical X-ray films (PX type) were used.

3.7. Temperature

All experiments were conducted at room temperature.

4. EXPERIMENTAL RESULTS

4.1. Release of Radioactivities from the Ion Exchange Resins into the Sea Water

4.1. 1. *Effects of weight ratios of ion exchange resin to sea water on release of radioactivities.* The percentages of activities of ^{60}Co, ^{89}Sr and fission products released from the cation exchange resins into the sea water were measured under static and turbulent conditions for different weight ratios (about 10^{-4}, 10^{-5}, 10^{-6}, 10^{-7}) of ion exchange resin to sea water. The final percentages of released activities after the equilibrium state was reached are shown in Table 2.

As can be seen in this table, the ^{60}Co and ^{89}Sr were observed to be released nearly 100% and no marked differences in the release percentages were observed for the weight ratio 10^{-5} to 10^{-7}. A similar tendency was observed with the fission product mixture. However, since the relative percentages of the different

Table 2. Effects of the Weight Ratio of Cation Exchange Resin to Sea Water on the Release of the Radioactivities, under the Static and Turbulent Conditions

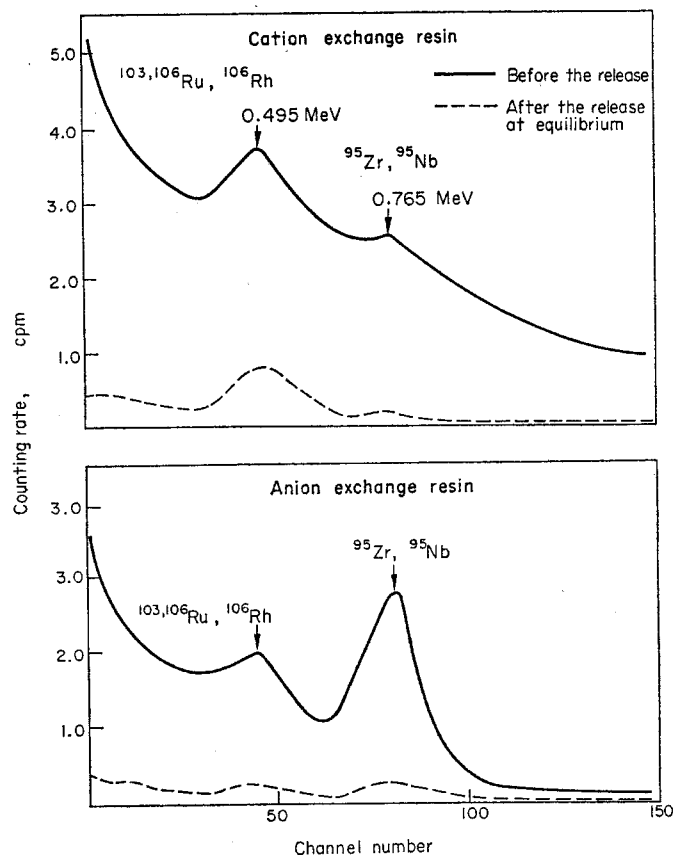| Weight ratio resin : sea water	Release of activity (%)			Condition of release
	^{60}Co	Fission products	^{89}Sr	
10^{-4}	99.25	89.85*	98.80	Turbulent
10^{-4}	—	—	98.50	Static
10^{-5}	99.92	96.26*	99.95	Turbulent
10^{-5}	—	—	99.94	Static
10^{-6}	99.97	98.30*	99.99	Turbulent
10^{-6}	—	—	99.96	Static
10^{-7}	—	—	99.97	Static

* The gross beta activity.

FIG. 3. Gamma-ray spectra of fission products before and after the release. (The weight ratio of resin: sea water $= 10^{-3}$).

Table 3. *Final Percentages of the Released Activities of Various Nuclides, after the Equilibrium State or about* 50 *hours elapsed, under the Static and Turbulent Conditions (the weight ratio of resin: sea water $= 10^{-6}$)*

Radioactive nuclides	Ion exchange resin	Release of activity (%)	
		Static condition	Turbulent condition
[89]Sr	Cation exchange resin	99.96	99.99
[51]Cr	Cation exchange resin	44.0	47.0
[55]Fe	Cation exchange resin	19.0	36.6
[60]Co	Cation exchange resin	99.79	99.97
F.P.	Cation exchange resin	75.0*	98.3*
F.P.	Anion exchange resin	80.0*	97.4*

* The gross beta activity.

nuclides included in the fission product mixture may change according to age, the percentage of release as measured with the gross activity may also change.

4.1. 2. *The percentages of the release of activities of different nuclides.* The percentages of the release of activities of different nuclides were measured under static and turbulent conditions for the fixed weight ratio 10^{-6} of the ion exchange resin to the sea water. The results are shown in Table 3.

As can be seen in this table, such nuclides as ^{60}Co and ^{89}Sr were easily released, while ^{51}Cr and ^{55}Fe not so easily.

In the case of the fission product mixture, the release experiment was conducted not only with the cation exchange resins but also with the anion exchange resins. The gamma ray spectrum of the fission product mixture on the ion exchange resins was measured before and after the release.

The results are shown in Fig. 3.

The photo-peaks corresponding to ^{103}Ru, ^{106}Ru (^{106}Rh) and ^{95}Zr, ^{95}Nb were observed. Because of the marked lowering of the corresponding peaks after the release, it may be estimated that the corresponding nuclides might have been released.

4.1. 3. *The effects of the amounts of nuclide initially adsorbed on the ion exchange resins on the release of activities.* Prior to the release experiment, different amounts of non-radioactive carriers of Co and Sr (varying from nearly zero to about maximum exchange capacity of the resins) were adsorbed on the ion exchange resins together with fixed activities of ^{60}Co and ^{89}Sr.

The release experiment was conducted for the fixed weight ratio 10^{-3} of the ion exchange resins to the sea water.

The residual activity and the residual amount of carrier nuclide in the resins at equilibrium state are shown in Fig. 4.

The percentages of released activities are given in Table 4.

The specific activities in the ion exchange resins before and after the release of the activities are shown in Fig. 5.

According to the results of the experiments, the residual percentages of activities in the ion

exchange resins were observed to remain the same in spite of the large differences in the initial amount of carriers. In other words, the residual percentages of the carriers in the ion exchange resins were observed to increase
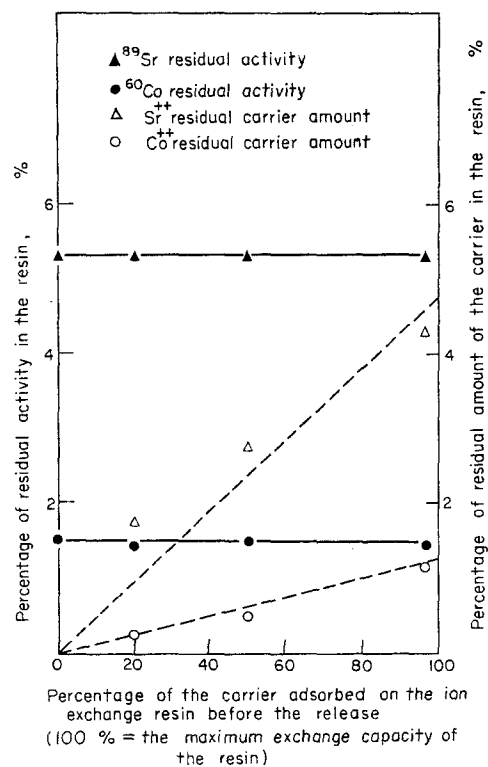

Percentage of the carrier adsorbed on the ion exchange resin before the release
(100 % = the maximum exchange capacity of the resin)

Fig. 4. Effects of the amount of carrier initially adsorbed on the release of activity.

Table 4. *Effects of the Initial Amount of Carrier in the Resin on the Released Activity, under the Turbulent Condition (the weight ratio of resin : sea water = 10^{-4})*

Amount of carrier in the resin before the release* %	Release of activity	
	^{60}Co %	^{89}Sr %
20	98.55	95.75
50	98.50	94.82
100	98.52	94.48

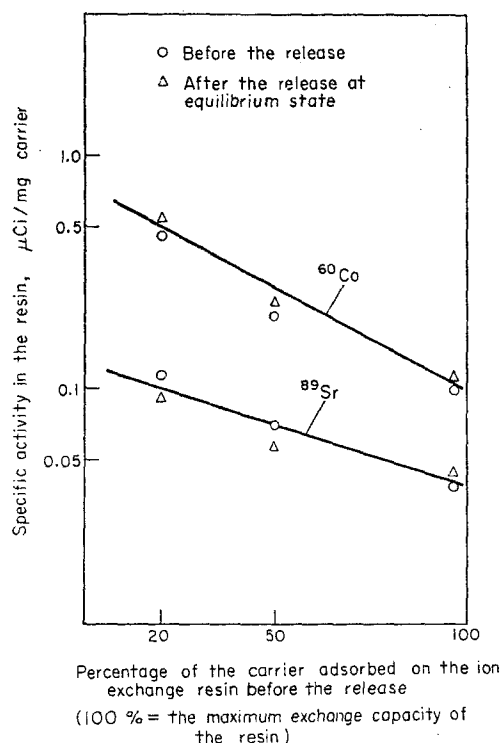* 100% = The maximum exchange capacity of the resin.

FIG. 5. Specific activity in the cation exchange resins before and after the release.

linearly with the increase of the initial amount of the carriers expressed as percentage of the saturation capacity of the ion exchange resins. From these results it was confirmed that the mass action law holds in the exchange reactions under the condition of the experiments.

The specific activities in the ion exchange resins before and after the release remained unchanged in each experiment within the limit of error in these experiments. The behaviour of the non-radioactive carrier nuclides was the same as that of the radioactive and therefore the use of the carrier was justified in this case.

4.1. 4. *The distribution of concentration of the released activities.* Vertical and horizontal distributions of the activities released from the ion exchange resins into the sea water have been measured under static and turbulent conditions in the experimental bath. The results are shown in Figs. 6–16 as a function of time.

In the case of ^{89}Sr (Fig. 6), the distribution was observed to be almost uniform after a few hours. However, in the case of ^{51}Cr (Fig. 11) the distribution was observed to be quite irregular with respect to time as well as with respect to location. Such a difference in the distribution pattern might be ascribed to the difference in the physico-chemical state of the nuclide in the sea water.

4.2. *Fractionation of suspended matter in sea water and accumulation of radioactive nuclides in each fraction*

In Table 5 the analytical data of each fraction of suspended matter in sea water and the concentration factors of radionuclides per dry weight of suspended matter are given. It can be seen that the quantity of organic matter was relatively less than that of inorganic matter and that the greatest concentration factors and accumulation fractions of radionuclides were obtained in the fraction of large particle sizes recovered by Toyo Filter Paper Type No. 5A (mean pore size 18 μ), rather than in the fractions of relatively small particle sizes.

The concentration factors of radionuclides in each fraction of suspended matter as a function of elapsed time are shown in Fig. 17.

The accumulation of radioactive nuclides in suspended matter was observed to reach an equilibrium in about 7–10 days. The accumulation fraction of radioactivity in each fraction of suspended matter is shown in Fig. 18. It can be seen that the majority of radioactivity was not retained on any of the filters used in this experiment.

4.3. *The distribution of radioactive nuclides in each fraction of suspended matter*

The distribution of radionuclides in each fraction indicated the tendency for a higher activity of ^{60}Co to be present in the fraction containing the larger particles, while the activity of ^{55}Fe was greater in the fraction containing the smaller particles (Table 5, Fig. 18).

The ratio of the specific activity per dry weight of suspended matter in each fraction to the average activity per dry weight of the total suspended matter is shown in Fig. 19. A higher activity for ^{55}Fe was also observed in the fraction containing the smaller particles.
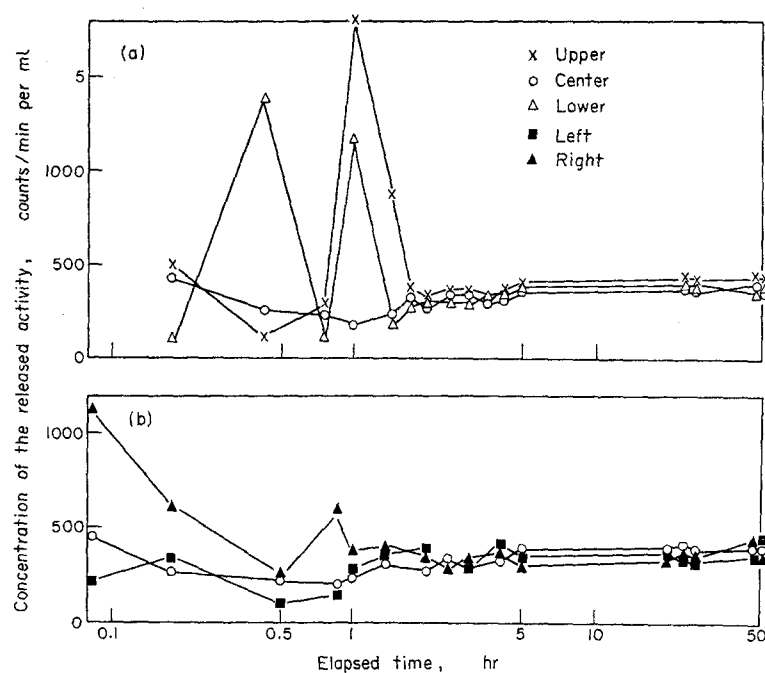
FIG. 6. Distributions of concentrations of the released activity of ^{89}Sr adsorbed on cation exchange resin under the static condition.
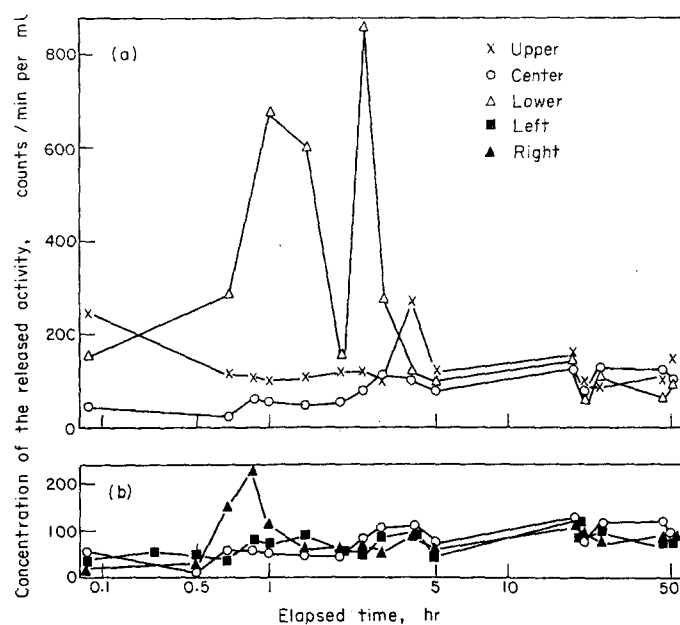

FIG. 7. Distributions of concentrations of the released activity of ^{60}Co adsorbed on cation exchange resin, under the static condition.
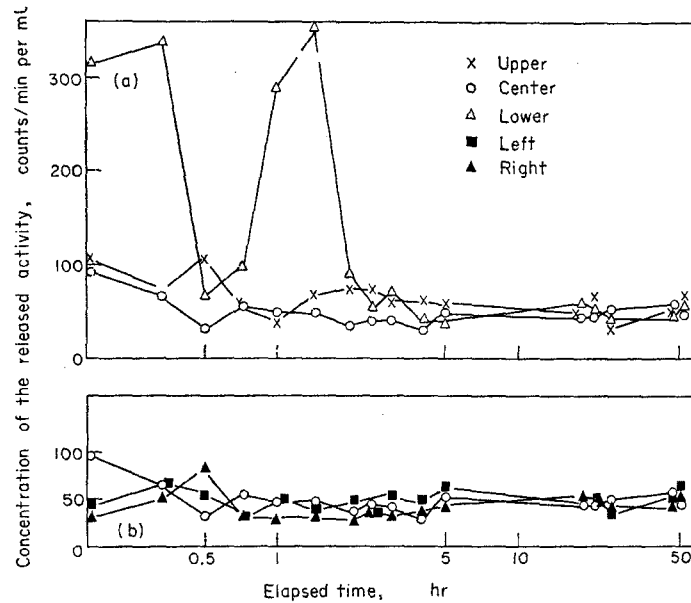
FIG. 8. Distributions of concentrations of the released activity of fission products adsorbed on cation exchange resin, under the static condition.
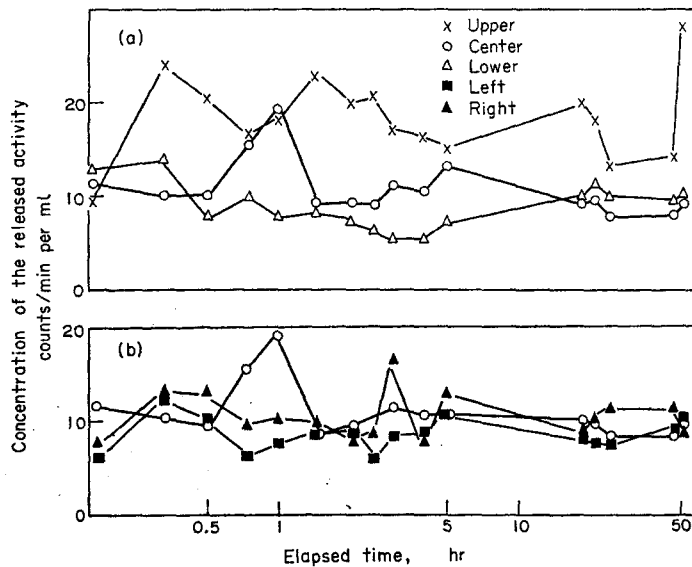

FIG. 9. Distributions of concentrations of the released activity of fission products adsorbed on anion exchange resin, under the static condition.
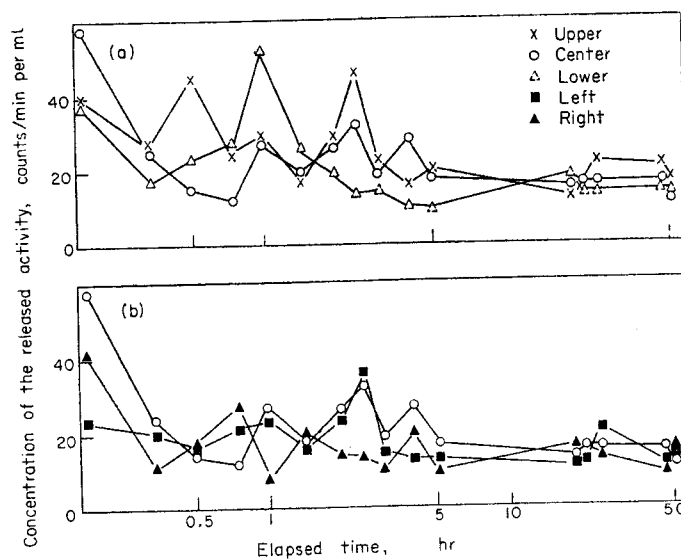
Fig. 10. Distributions of concentrations of the released activity of ^{55}Fe adsorbed on cation exchange resin, under the static condition.
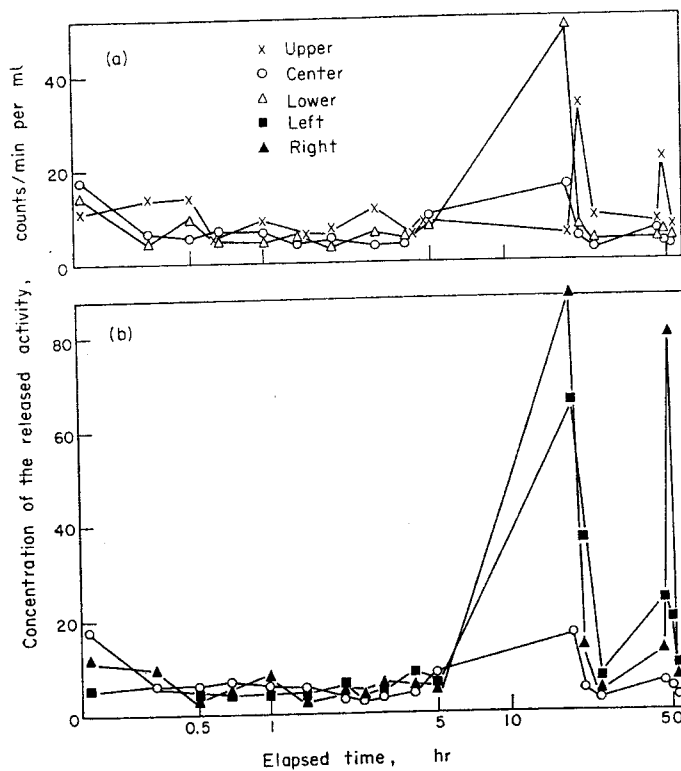

Fig. 11. Distributions of concentrations of the released activity of ^{51}Cr adsorbed on cation exchange resin, under the static condition.
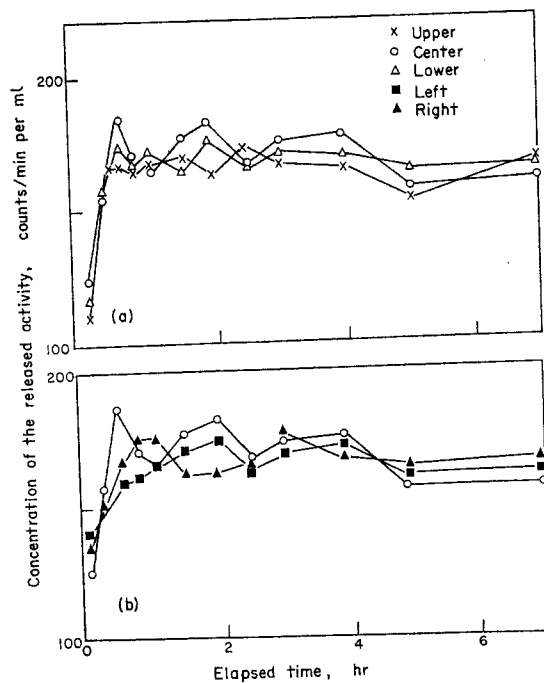
FIG. 12. Distributions of concentrations of the released activity of ^{89}Sr adsorbed on cation exchange resin, under the turbulent condition.

FIG. 13. Distributions of concentrations of the released activity of ^{51}Cr adsorbed on cation exchange resin, under the turbulent condition.
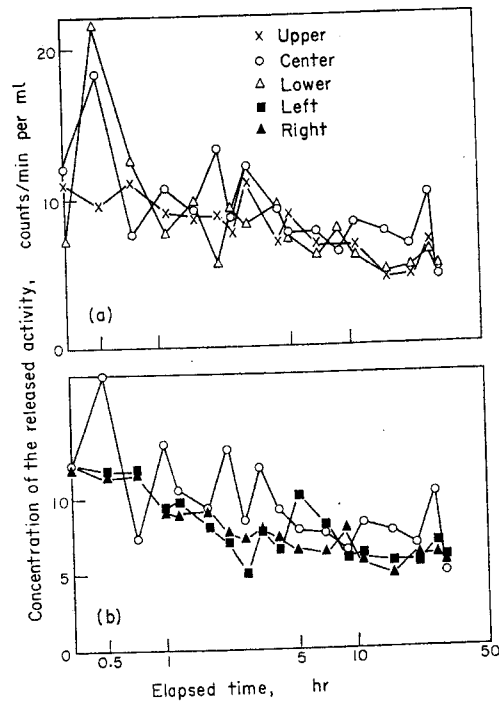
FIG. 14. Distributions of concentrations of the released activity of ^{55}Fe adsorbed on cation exchange resin, under the turbulent condition.
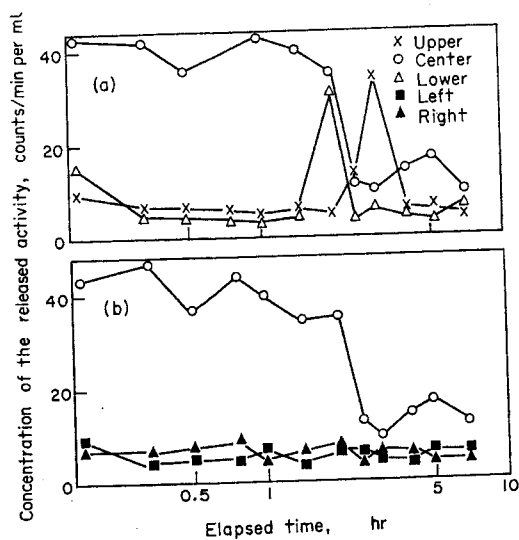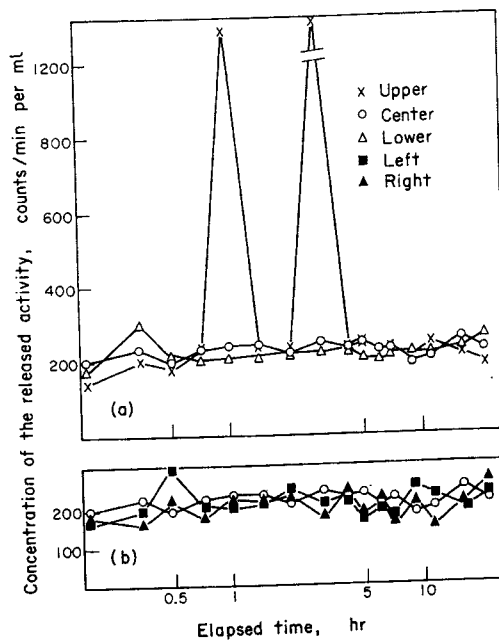
FIG. 15. Distributions of concentrations of the released activity of ^{60}Co adsorbed on cation exchange resin, under the turbulent condition.
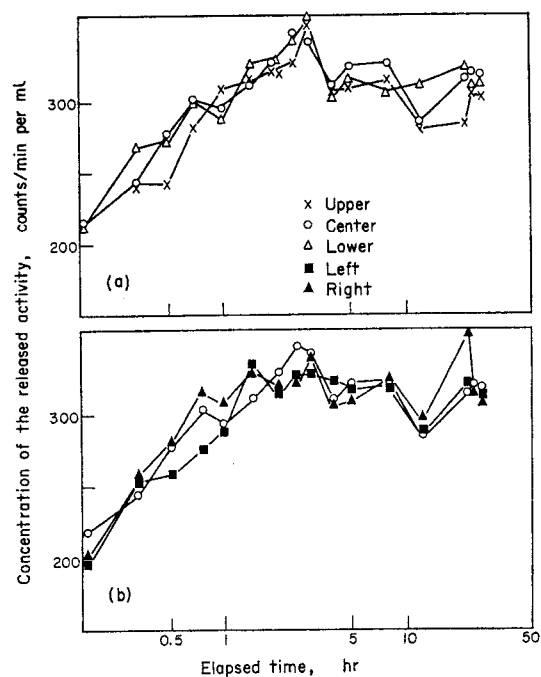
FIG. 16. Distributions of concentrations of the released activity of fission products adsorbed on cation exchange resin, under the turbulent condition.
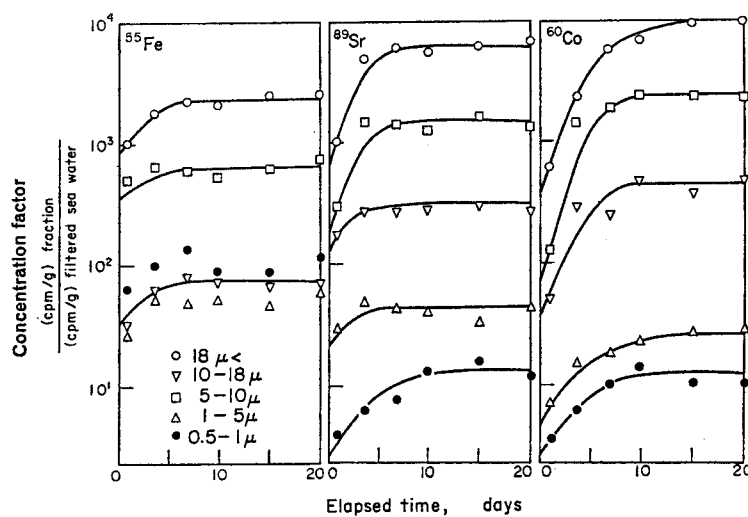

FIG. 17. Uptake of radioactivity in each fraction of suspended matter (per dry weight).

Table 5. Fractionation of the Suspended Matter and Accumulation of Radioactive Nuclides (initial radioactive concentration in the sea water was about 10^{-4} μCi/ml)

Size range of suspended matter micron	Dry weight mg/ml	Organic N mg/ml	Organic matter* mg/ml	Inorganic matter by difference mg/ml	Concentration factor per dry weight			Accumulation fraction		
					^{55}Fe	^{60}Co	^{89}Sr	^{55}Fe	^{60}Co	^{89}Sr
					—	—	—	—	—	—
18	4.19	0.06	1.02	3.17	23.0×10^2	93.0×10^2	52.0×10^2	5.2×10^{-2}	56.1×10^{-2}	2.0×10^{-4}
10–18	0.94	0.02	0.34	0.60	0.6×10^2	4.2×10^2	2.6×10^2	0.8×10^{-2}	7.8×10^{-2}	0.4×10^{-4}
5–10	2.74	0.01	0.17	2.61	8.0×10^2	24.0×10^2	14.0×10^2	2.9×10^{-2}	10.2×10^{-2}	0.8×10^{-4}
1–5	4.05	0.03	0.51	3.54	0.6×10^2	0.27×10^2	0.4×10^2	0.7×10^{-2}	1.3×10^{-2}	0.5×10^{-4}
0.5–1	3.90	0.04	0.68	3.22	0.6×10^2	0.11×10^2	0.4×10^2	0.5×10^{-2}	0.7×10^{-2}	0.4×10^{-4}

* Organic matter (mg/ml) = Organic nitrogen (mg/ml) $\times$ 17.

Concentration factor $= \dfrac{\text{Activity of fraction (cpm/g)}}{\text{Activity of filtered sea water (cpm/ml)}}$

Accumulation fraction $= \dfrac{\text{Activity of fraction per liter of sea water (cpm/ml)}}{\text{Activity of sea water containing suspended matter (cpm/l)}}$
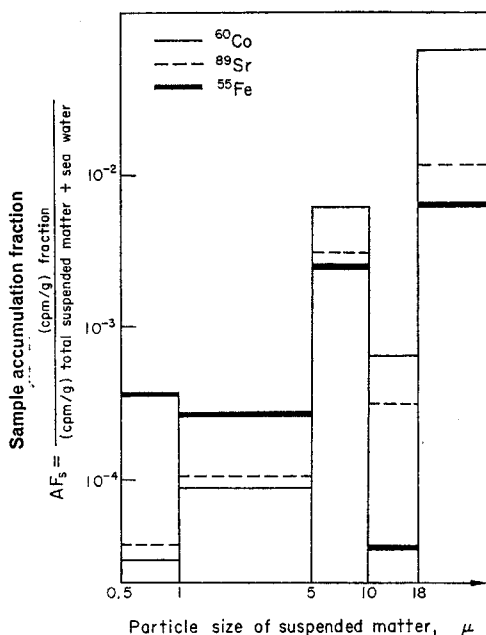
FIG. 18. Sample accumulation fraction of nuclides in each fraction of suspended matter.
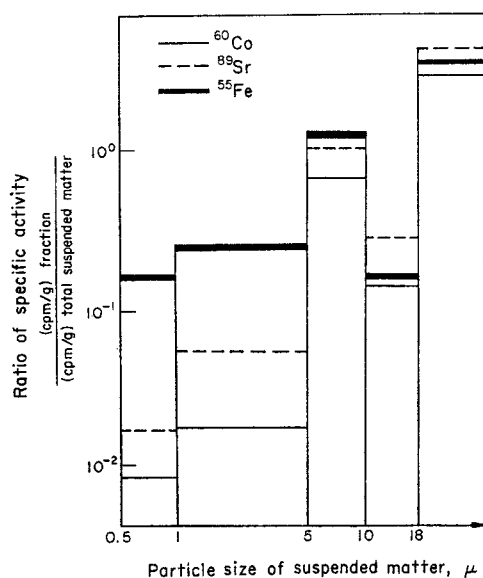
FIG. 19. Ratio of the specific activity in each fraction to the average specific activity of the total suspended matter (per dry weight).

4.4. *Autoradiography of radioactive nuclides in the suspended matter*

The autoradiograms of ^{59}Fe, ^{60}Co and ^{89}Sr in the fraction ($> 18\,\mu$) recovered by Toyo Filter Paper Type No. 5A are shown in Fig. 20. It can be seen that the radioactivity was accumulated in relatively large particulates. On the other hand, the autoradiograms of radionuclides in the fractions (5–10 μ) recovered by Millipore Filter Type SM, do not show any large exposed spots as shown in Fig. 20.

5. DISCUSSION

The exchange reactions of ion exchange resins have been investigated by many authors not only from the basic viewpoint but also from the viewpoint of applications.

In the present report, the percentage of released radioactivities was determined by adding the ion exchange resins containing adsorbed radioactive nuclides to the sea water. This system may be considered a heterogeneous one consisting of salt solution and particles. The processes by which these adsorbed radionuclides would be released from the resin into the sea water and how they would distribute in the medium have been discussed.

From these experiments, it was observed that the adsorbed radioactive nuclides were released as soon as the resin particles came in contact with the sea water, and that the released nuclides formed a water mass with locally concentrated radioactivities by the diffusion process and then diffused gradually through the sea water finally to reach a macroscopically homogeneous equilibrium. In such release experiments, it was found that some nuclides such as ^{60}Co and ^{89}Sr were easily released, while others as ^{51}Cr and ^{55}Fe not so easily. The percentage of the release of activities increases with the decrease of weight ratios of ion exchange resins to the sea water, and no marked differences in the release were observed for the weight ratio between 10^{-5} to 10^{-7}. The distribution patterns of radioactivity concentration in the sea water after the release seem to depend on the physical-chemical state of the nuclides in the sea water. In the experiments with a fixed weight ratio of ion exchange resins to sea water, it was observed as a tendency that the more the initial amounts of adsorbed activities on the

resins, the less, although not so remarkably, the activities released from resins into sea water.

From the abovementioned results the mechanisms of releasing adsorbed activities from resins into sea water were considered to depend exclusively on the ion exchange reaction, although some other reactions might also occur.

Studies of suspended matter in sea water have been conducted in the past by many other

method of fractional filtration was employed. It was observed that the accumulation of each radionuclide reached equilibrium in 7–10 days and that the maximum concentration factor, expressed as a ratio of the activity in the suspended matter per dry weight to that in the sea water per milli-liter, of each radionuclide was found in the fraction of larger particles recovered by Toyo Filter Paper Type No. 5A (mean pore size 18 μ).
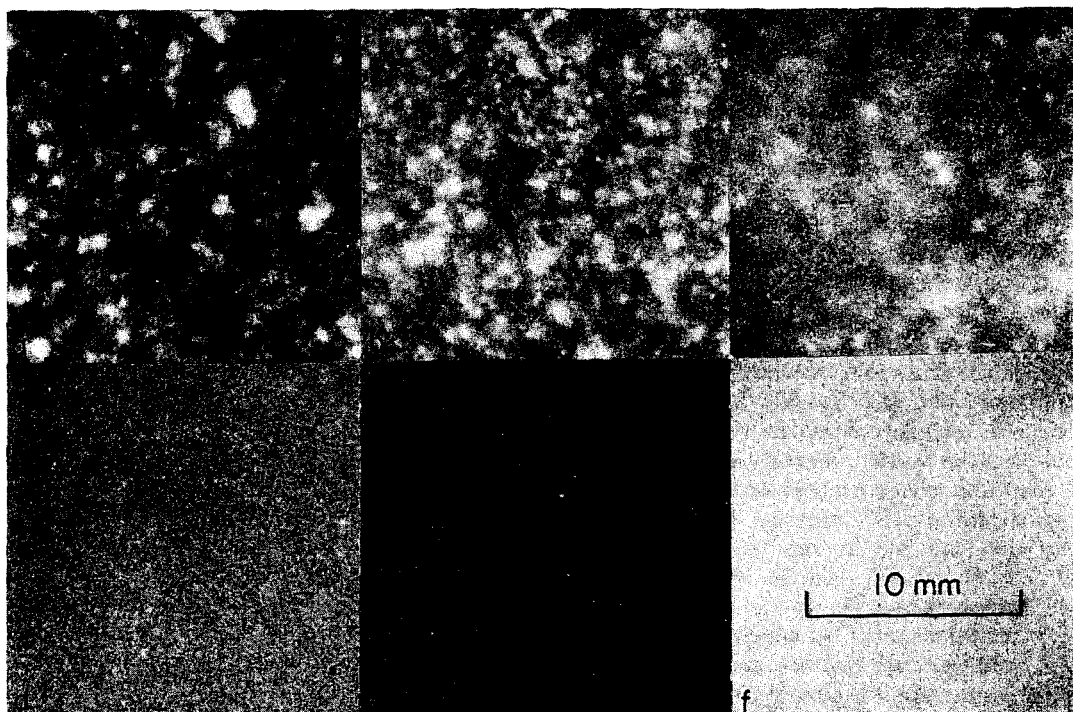

FIG. 20. *The autoradiograms of nuclides in the fraction of suspended matter.*

Plate	Nuclide	Range of particle sizes (μ)	Exposure (hr)
a	^{59}Fe	> 18	43
b	^{60}Co	> 18	60
c	^{89}Sr	> 18	40
d	^{59}Fe	5–10	43
e	^{60}Co	5–10	60
f	^{89}Sr	5–10	60

authors [6-15] from the viewpoint of oceanography and fishery. However, in order to estimate the behaviour and distribution of radioactive nuclides in suspended matter in sea water, the

It might be expected as a general rule that the smaller the particles, i.e. the larger the surface area per unit weight, the larger the amount of radioactivity which could be

accumulated per unit weight. In these experiments, on the contrary, the accumulation of radioactivity in the fraction with larger particles was greater. Judging from these results, it may be inferred that not all particulate fractions obtained by fractional filtration from natural sea water have the same composition, and that the organic and inorganic matter of particulates accumulating nuclides in each fraction would be different in quality and quantity. The distribution of radioactive nuclides in each fraction indicated the tendency for a higher activity of ^{60}Co to be present in the fraction containing the larger particles, while the activity of ^{55}Fe was greater in the fraction containing the small particle sizes. In order to interpret such a tendency more adequately, further studies might be necessary.

In assessing the possible effects due to the disposal of radioactive nuclides in the environmental hydrosphere, the so-called concentration factor as defined in the beginning of this paper has been used. Oftentimes, the expected average concentration of activity in a water medium is estimated by simply dividing the total activity of the waste present by the estimated total volume of the environmental water medium, and then the concentration of activity in the biological sample is estimated by just multiplying the expected average concentration of activity in the water medium by the so-called concentration factor. If the concentration factor is really the factor by which the biological sample may concentrate the activity from the environmental water medium, or if the so-called concentration factor is the one obtained under the equilibrium condition which could simulate the actual situation of the waste disposal, and if the sample fraction or the concentration of sample in the medium is sufficiently small, the estimation of the final concentration of activity in the biological sample by the above procedure may be approximately justifiable.

However, if a highly contaminated fish at the highly contaminated part of the sea water medium swims out into the surrounding low contaminated water, and if the so-called concentration factor is estimated as the ratio of the concentration of activity of the highly contaminated fish sample to that of the low contaminated water medium where the fish was caught, an

unusually high concentration factor may result. In this case, the concentration factor is merely a ratio of the concentration of the sample to that of the medium at the place where the sample was caught, but it does not represent the factor by which the sample may concentrate the activity from the environmental water medium. If such a high concentration factor were used evaluating the possible hazard to man at the time of the radioactive waste disposal into the environmental hydrosphere, a much too conservative result may be indicated for the permissible level of waste disposal, giving the people an erroneous impression. Therefore, in the case of the hazard analysis of the waste disposal into hydrosphere, the use of the sample accumulation fraction (AFs) or the sample concentration factor (CF_S), as defined below, seems to be more reasonable. In the following, some of the relationships and interrelationships between some of the parameters which may be useful in the analysis of problems of contamination are introduced.

Notation:

(Medium + Sample) = MS, Medium = M, Sample = S

$A_{(M)}$ = activity of medium

$A_{(S)}$ = activity of sample

$A_{(MS)} = A_{(M)} + A_{(S)}$

A/kg = concentration of activity per kg of material concerned

$A_{(S)}/kg_{(S)}$ = concentration of activity per kg of sample

$A_{(M)}/kg_{(M)}$ = concentration of activity per kg of medium

$A_{(S)}/kg_{(MS)}$ = activity of the sample included in kg of medium plus sample

$A_{(M)}/kg_{(MS)}$ = activity of the medium included in kg of medium plus sample

In the following relations, the activity per unit mass is used instead of the activity per unit volume because strictly speaking the volume may be dependent on the temperature. However, even if we replace the activity per unit mass with the activity per unit volume, it is obvious that similar relations should hold.

(1) *Sample to Medium Concentration Ratio:* $CR_{(S-M)}$

$$CR_{(S-M)} = \frac{A_{(S)}/kg_{(S)}}{A_{(M)}/kg_{(M)}}$$

This is the so-called concentration factor which is the ratio of the concentration of the activity of the sample to that of the medium.

(2) *Medium to Sample Concentration Ratio:* $CR_{(M-S)}$

$$CR_{(M-S)} = \frac{A_{(M)}/kg_{(M)}}{A_{(S)}/kg_{(S)}} = \frac{1}{CR_{(S-M)}}$$

This relation may be important when the medium is more useful than the sample itself, for instance, when decontaminating a water medium by the biological sample in it.

(3) *Sample Accumulation Fraction:* AF_S

$$AF_S = \frac{A_{(S)}/kg_{(MS)}}{A_{(MS)}/kg_{(MS)}} = \frac{A_{(S)}}{A_{(MS)}} \leq 1$$

(4) *Medium Accumulation Fraction:* AF_M

$$AF_M = \frac{A_{(M)}/kg_{(MS)}}{A_{(MS)}/kg_{(MS)}} = \frac{A_{(M)}}{A_{(MS)}} \leq 1$$

(5) *Sample Concentration Factor:* CF_S

$$CF_S = \frac{A_{(S)}/kg_{(S)}}{A_{(MS)}/kg_{(MS)}}$$

(6) *Medium Concentration Factor:* CF_M

$$CF_M = \frac{A_{(M)}/kg_{(M)}}{A_{(MS)}/kg_{(MS)}}$$

(7) *Sample to Medium Ratio:* $R_{(S-M)}$

$$R_{(S-M)} = \frac{kg_{(S)}}{kg_{(MS)}}$$

(8) *Medium to Sample Ratio:* $R_{(M-S)}$

$$R_{(M-S)} = \frac{kg_{(M)}}{kg_{(S)}} = \frac{1}{R_{(S-M)}}$$

(9) *Sample Fraction:* F_S

$$F_S = \frac{kg_{(S)}}{kg_{(MS)}}$$

Fraction of the sample per $kg_{(MS)}$.

(10) *Medium Fraction:* F_M

$$F_M = \frac{kg_{(M)}}{kg_{(MS)}}$$

Fraction of the medium per $kg_{(MS)}$.

(11) *Some of the Important Interrelationships.*

$$(CF_S)(F_S) = (AF_S) \leq 1$$
$$(CF_M)(F_M) = (AF_M) \leq 1$$

From these relationships and interrelationships, it is clear, for instance, that if the sample concentration factor (CF_S) is 10^5, the sample fraction (F_S) or the concentration of sample in the aqueous medium must be smaller than 10^{-5}. In the case of hazard analysis of waste disposal into the hydrosphere, the expected average initial concentration of activity $(\bar{C})$ per unit volume or mass of the water medium is usually estimated by dividing the total activity of the waste present by the estimated total volume or mass of the environmental water medium which may include the sample in question, regardless of the sample fraction in the medium. In this case, the final concentration of the activity in the biological sample $(A_{(S)}/kg_{(S)})$ may be estimated by using the sample concentration factor (CF_S) defined in (5) above from the following relation:

$$A_{(S)}/kg_{(S)} = (\bar{C})(CF_S)$$

where $\bar{C}$ is the average concentration of the activity per $kg_{(MS)}$ and may be considered to be equivalent to the overall average concentration $A_{(MS)}/kg_{(MS)}$ in the above notation.

Similarly, the final total activity transferred to and accumulated in the sample per kg of medium plus sample $(A_{(S)}/kg_{(MS)})$ may be estimated by using the sample accumulation fraction (AF_S) defined in (3) above from the following relation:

$$A_{(S)}/kg_{(MS)} = (\bar{C})(AF_S)$$

If the concentration of the sample in the medium, the sample fraction (F_S) or the medium fraction (F_M) is known, the factors involved in the above estimation could be checked, whether they are grossly in error or not, by using the interrelation indicated in (11) above.

From these points of view, the effective volume or mass of the medium associated with the sample or the average volume or mass of the medium that could be effectively utilized by a biological sample in question seems to be an important factor also.

6. SUMMARY

The release of radioactivities from ion exchange resins into sea water was studied in a water bath containing natural surface sea water sampled at Osaka Bay, using several radionuclides for the different weight ratios of ion exchange resins to sea water and the amount of nuclide initially adsorbed on the ion exchange resins under the static and turbulent conditions. The behaviour and distribution of radionuclides in suspended matter in sea water was also studied experimentally by fractional filtration using several types of filters with different pore sizes. The results are summarized as follows:

1. The radioactivities were rapidly released from the ion exchange resin particles on contact with the sea water. However, some differences were observed in the degree of release depending on the different nuclides. Such nuclides as ^{60}Co and ^{89}Sr were easily released, while ^{51}Cr and ^{55}Fe not so easily.

2. The released activity was observed to increase with the decreasing weight ratio of ion exchange resins to sea water, but with the weight ratio below 10^{-4} of ion exchange resin to sea water no remarkable difference was observed.

3. With the weight ratio below 10^{-3} of the ion exchange resin to the sea water, the initial adsorbed amount on the ion exchange resin gave no remarkable effects on the release of radioactivities.

4. The accumulation of the radioactive nuclides in suspended matter was observed to reach an equilibrium in about 7–10 days.

5. The greatest accumulation and concentration of radioactivity per dry weight of suspended matter in the sea water were observed in the fraction containing the larger particles recovered by Toyo Filter Paper Type No. 5A (mean pore size $18\,\mu$), rather than in the fractions containing relatively smaller particles.

6. The distribution of radionuclides in each fraction indicated a tendency for a higher activity of ^{60}Co to be present in the fraction containing the larger particles, while the activity of ^{55}Fe was greater in the fraction containing the smaller particles.

7. Finally, after discussing the so-called concentration factor, it is pointed out that the ratio of the concentration of activity of the sample to that of the medium may not necessarily represent the real factor by which the biological sample may concentrate the activity from the environmental water medium. The use of the sample concentration factor, which is the ratio of the concentration of activity of the sample to the average concentration of medium plus sample, is proposed for the estimation of the final concentration of the biological sample for the purpose of hazard analysis due to radioactive waste disposal into the environmental hydrosphere.

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