

009 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 the ion exchange resins disposed into the sea water, we have conducted a series of experiments in the 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 the 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 the suspended matters in the sea water containing ^{55}Fe , ^{60}Co and ^{89}Sr will also be discussed.

1. INTRODUCTION.

It is considered to be an important problem to investigate the behaviour and distribution of radioactivities in the ecosystem in the ocean, when the radioactive contamination and its effects due to the radioactive waste disposal into the sea water is studied¹⁻⁶⁾.

If at 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 the sea water

* This paper was presented at the First international congress of the international radiation protection association held in Rome, 1966

and to estimate the behaviour and the distribution of radioactivities released into the sea water, we have conducted a series of experiments in the water bath containing natural sea water using several radioactive nuclides.

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

2. MATERIALS AND APPARATUSES

2.1 Sea water

Natural surface sea water was sampled at about 50 meters apart from the Misaki beach of Osaka-Bay. The total sodium content was estimated at 9.500 ppm, the chlorinity 17.7 0/00 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 gram 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 gram of dry resin.

2.3 Radioactivities

⁵¹Cr (CrCl₃, HCl solution), specific activity 13.3 mCi/mg, 2.8 mCi/ml.

(Commissariat A L'Energie Atomique)

⁵⁵⁻⁵⁹Fe (FeCl₃, HCl solution) carrier free, 26.8 mCi/ml.

(Nuclear Science and Engineering Co.)

⁶⁰Co (CoCl₂, HCl solution) Co 0.44 mg/ml, specific activity 55 mCi/mg, 24.2 mCi/ml

(Oak Ridge National Laboratory)

⁸⁹Sr (SrCl₂, HCl solution) carrier free 1.7 mCi/ml

(The Radiochemical Centre)

Every radioactive chemicals was diluted, and

about 10 or 100 μ Ci 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 also 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 the pH-meter with glass electrode.

2.6 Water baths used for experiments were made of acryl-plastics. The size was 1,000 mm (length) \times 500 mm (width) \times 1,000 mm (height). Cylindrical water baths made of glass and those of poly-ethylene were also used, the sizes of which were 113 mm ϕ \times 145 mm (height) and 300 mm ϕ \times 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 one liter per second. (Plate 1) A glass water bath with a stirrer of a stainless propeller (about 200 r. p. m.) was also used.

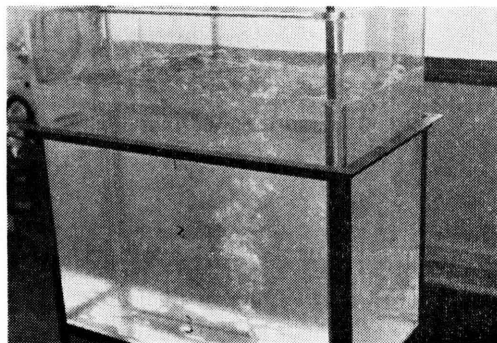


Plate 1. Experimental water bath.

2.8 Suspended matters in the sea water were filtered successively with Toyo-filter paper

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

3. EXPERIMENTAL PROCEDURES

3.1 Weight ratios of ion exchange resin 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 was previously adsorbed on the resins constant.

3.3 Released radioactivity from ion exchange resins.

Ion exchange resins on which radioactive nuclides were adsorbed were placed in the sea water in the water bath for experiments. An aliquot of the water (1 ml) was sampled with definite time interval at each of 5-9 points indicated in Fig. 1 in order to determine the spacial distribution of the radioactivity in the sea water. After equilibrium, the percentage of the released radioactivity

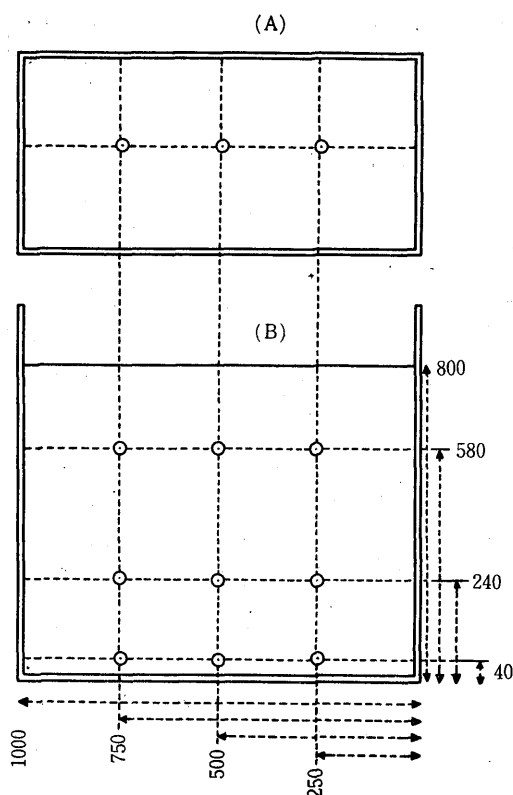


Fig. 1. Experimental water bath (mm) (A) plan, (B) elevation, and sampling position.

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.

With a definite time interval an aliquot of sea water (1.5 l) was sampled several times

Table 1. Properties of filters.

| Filter | Material | Color | Thickness micron | Flow rate* ml/min. cm ² | Mean pore size micron |
|-----------|-------------------|-------|------------------|------------------------------------|-----------------------|
| 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.

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 filtrate was tested with silver nitrate and the ammonium with Nessler's reagent.

3.4.2 Measurement of dry weight of suspended matter in the 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 micro-Kjeldahl 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. Fox⁷⁾.

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

Concentration factor is defined as the ratio of radioactivity (cpm/g) of each fraction of 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 fraction 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 in the above. Residual activities on these filters were subtracted from the above values as backgrounds respectively.

3.6 Autoradiography of accumulation of radionuclides in the suspended matter

The autoradiograms of each fraction of suspended matter in the sea water were taken with 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 the Weight Ratios of Ion Exchange Resin to the Sea Water on the Release of the Radioactivities.

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

As can be seen in this table, the ⁶⁰Co and ⁸⁹Sr were observed to be released nearly 100 % and no marked difference in the release percentages were observed for the weight ratio of 10⁻⁵—10⁻⁷. A similar tendency was observed with the fission product mixture. However, since the relative percentages of the different nuclides included in the fission product mixture may change according to the age, the percentage of release as measured

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.

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.

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 the 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 ⁶⁰Co and ⁸⁹Sr were easily released, while ⁵¹Cr and ⁵⁹Fe were not so easily.

In case of 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. 2.

The photo-peaks corresponding to ¹⁰³Ru, ¹⁰⁶Ru, (¹⁰⁶Rh) and ⁹⁵Zr, ⁹⁵Nb 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 ⁶⁰Co and ⁸⁹Sr.

The release experiment was conducted for the fixed weight ratio of 10⁻³ of the ion exchange resin to the sea water.

The residual activity and the residual amount of the carrier nuclide in the resins at equilibrium state are shown in Fig. 3.

The released percentages of 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. 4.

According to the results of the experiments, the residual percentages of activities in the ion exchange resins were observed to

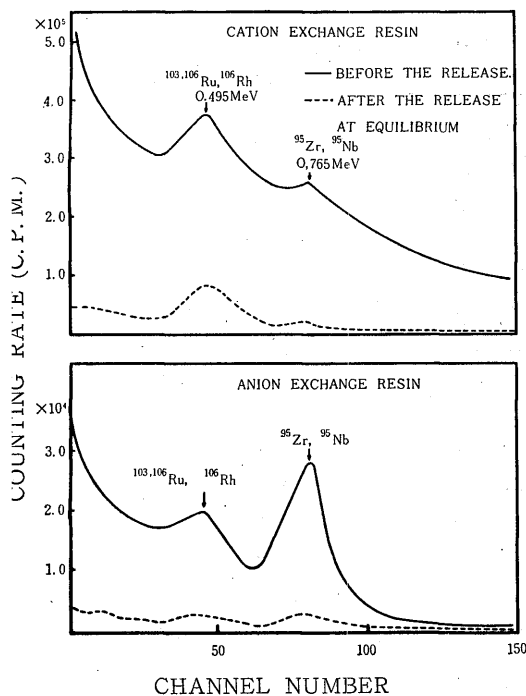


Fig. 2. Gamma-ray spectra of fission products in the resin. Condition of release: Turbulent, with the weight ratio 10^{-3} of the resin to the sea water.

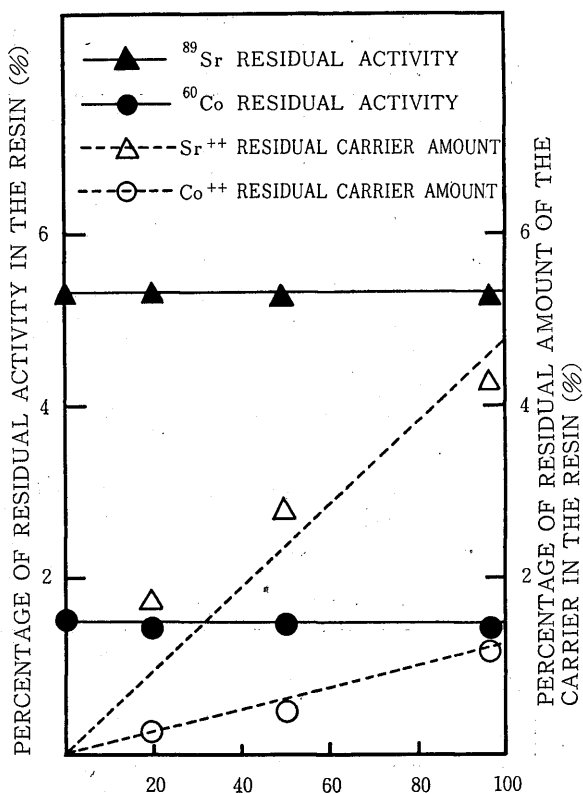


Fig. 3. Effect of the amount of carrier initially adsorbed on the resin on the release of activity. Condition of release: Turbulent, with the weight ratio 10^{-3} of the resin to the sea water.

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.

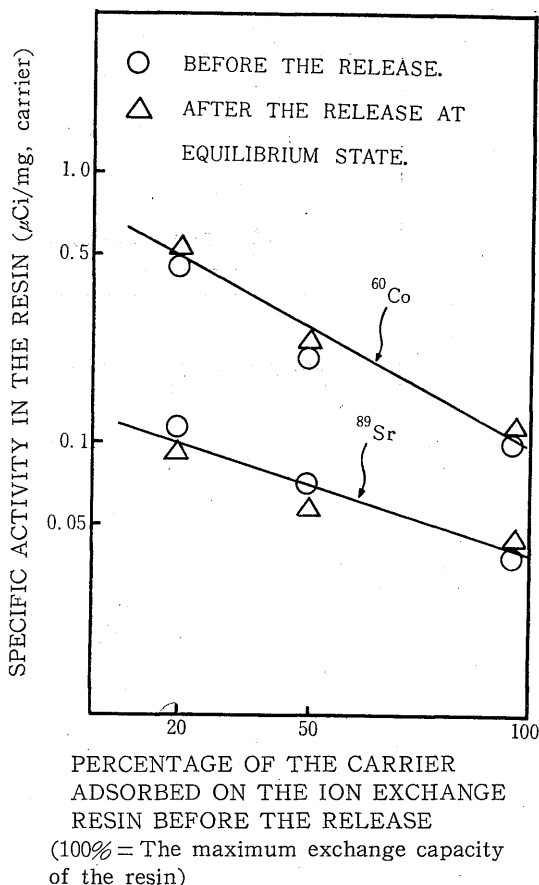


Fig. 4. Specific activity in the cation exchange resin before and after the release. Condition of release: Turbulent, with the weight ratio 10^{-3} of the resin to the sea water.

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 linearly with the increase of the initial amount of carriers as expressed in the percentage against 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 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 the static and turbulent conditions in the experimental bath. The results are shown in Fig. 5-15 as a function of time.

In case of ^{89}Sr (Fig. 5) the distribution was observed to be almost uniform after a few hours. However, in case of ^{51}Cr (Fig. 10) the distribution was observed, to be quite irregular with respect to the time as well as with respect to the 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 the suspended matter in the sea water and accumulation of radioactive nuclides in each fraction.

In Table 5. the analytical data of each fraction of suspended matter in the 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 the Toyo Filter Paper Type No. 5A (mean pore size 18μ), 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. 16.

The accumulation of the radioactive nucli-

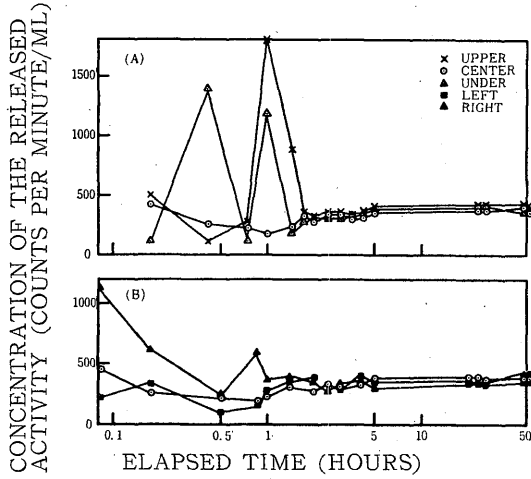


Fig. 5. Distributions of concentration of the released activity of ^{89}Sr adsorbed on the cation exchange resin, under the static condition.
 (A) Vertical distribution
 (B) Horizontal distribution

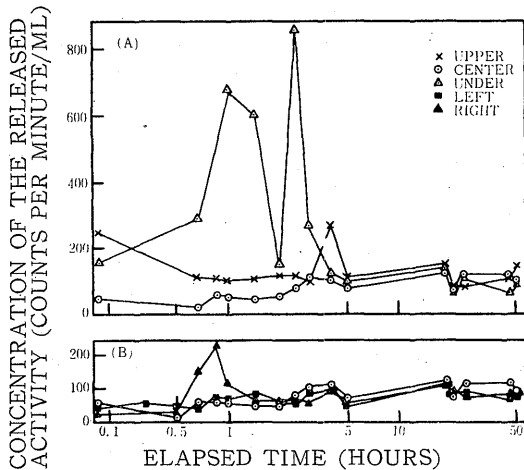


Fig. 6. Distributions of concentrations of the released activity of ^{60}Co adsorbed on the cation exchange resin, under the static condition.
 (A) Vertical distribution.
 (B) Horizontal distribution.

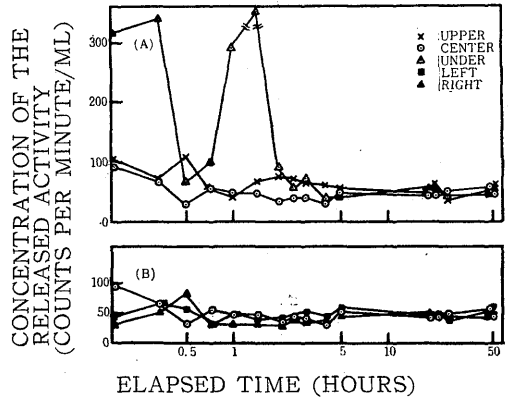


Fig. 7. Distribution of concentration of the released activity of fission products adsorbed on the cation exchange resin, under the static condition.
 (A) Vertical distribution
 (B) Horizontal distribution

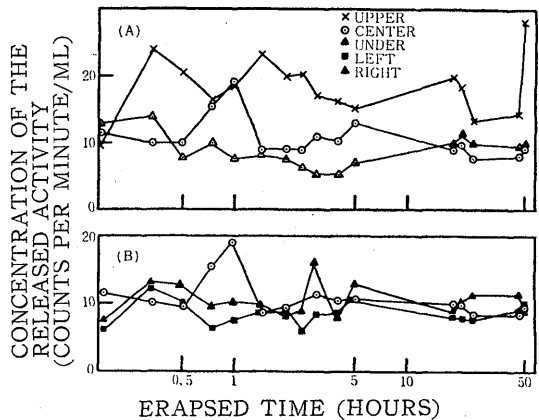


Fig. 8. Distribution of concentration of the released activity of fission products adsorbed on the anion exchange resin under the static condition.
 (A) Vertical distribution
 (B) Horizontal distribution

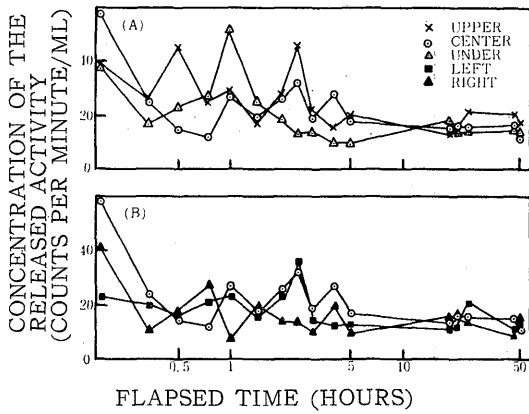


Fig. 9. Distribution of concentration of the released activity of ^{55}Fe adsorbed on the cation exchange resin, under the static condition.

- (A) Vertical distribution
- (B) Horizontal distribution

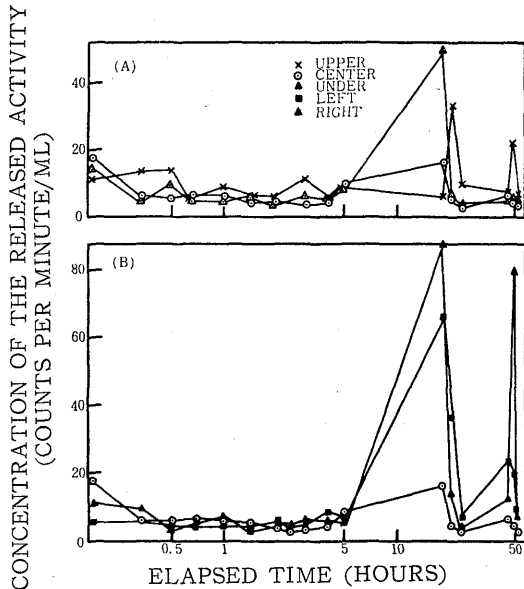


Fig. 10. Distributions of concentrations of the released activity of ^{51}Cr adsorbed on the cation exchange resin, under the static condition.

- (A) Vertical distribution
- (B) Horizontal distribution

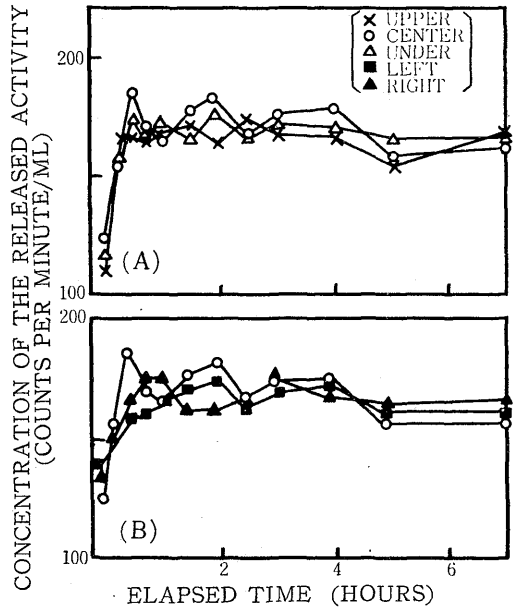


Fig. 11. Distributions of concentrations of the released activity of ^{89}Sr adsorbed on the cation exchange resin, under the turbulent condition.

- (A) Vertical distribution.
- (B) Horizontal distribution.

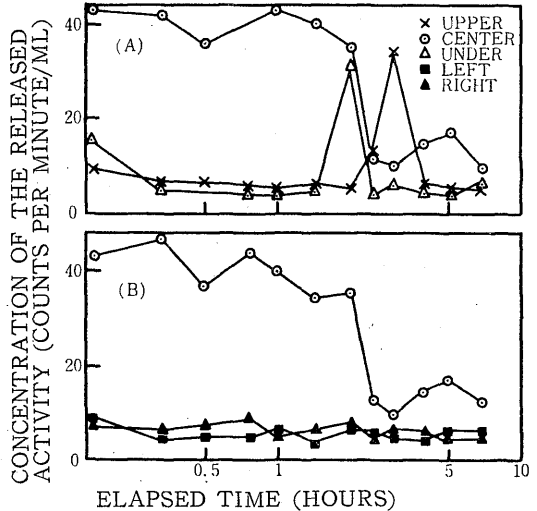


Fig. 12. Distributions of concentrations of the released activity of ^{51}Cr adsorbed on the cation exchange resin, under the turbulent condition.

- (A) Vertical distribution
- (B) Horizontal distribution

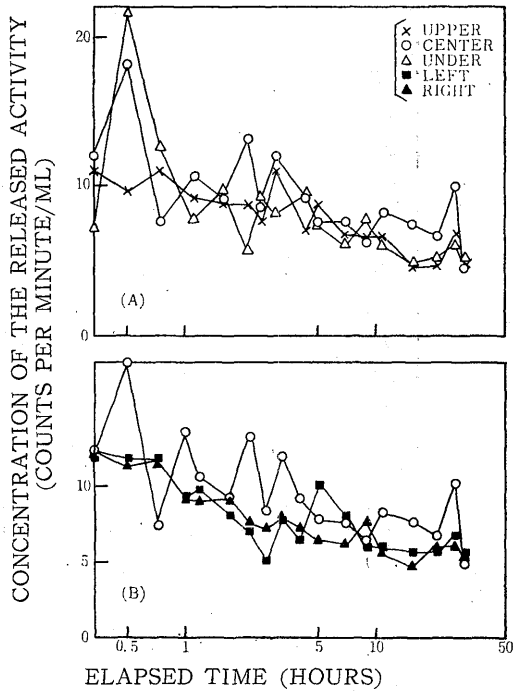


Fig. 13. Distributions of concentrations of the released activity of ⁵⁵Fe adsorbed on the cation exchange resin, under the turbulent condition.
 (A) Vertical distribution
 (B) Horizontal distribution

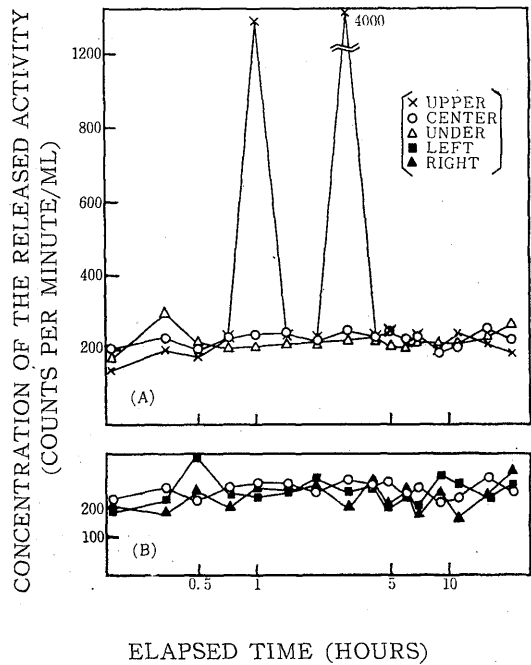


Fig. 14. Distributions of concentrations of the released activity of ⁶⁰Co adsorbed on the cation exchange resin, under the turbulent condition.
 (A) Vertical distribution
 (B) Horizontal distribution.

Table 5. Fractionation of the suspended matter and accumulation of radioactive nuclides. (Initial radioactive concentration in the sea water was about 10⁻⁴ microcurie/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) × 17

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

$$\text{Accumulation fraction} = \frac{\text{Activity of fraction per liter of sea water (cpm/ml)}}{\text{Activity of sea water containing suspended matter (cpm/l)}}$$

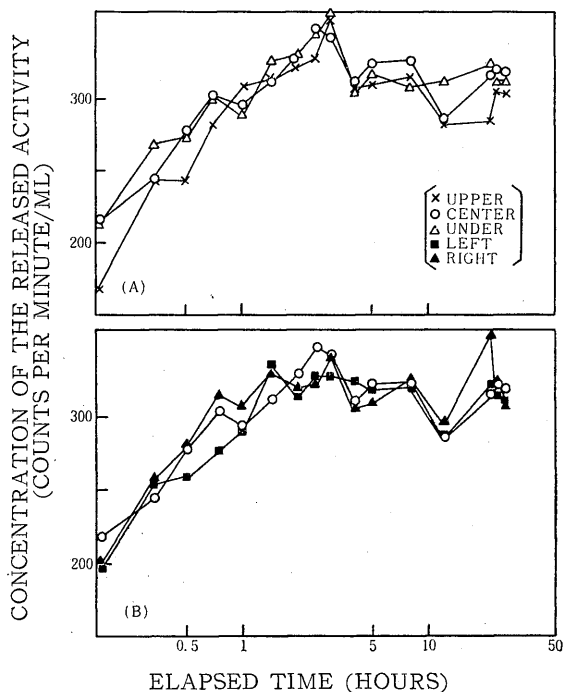


Fig. 15. Distributions of concentrations of the released activity of fission products adsorbed on the cation exchange resin, under the turbulent condition.

- (A) Vertical distribution
- (B) Horizontal distribution.

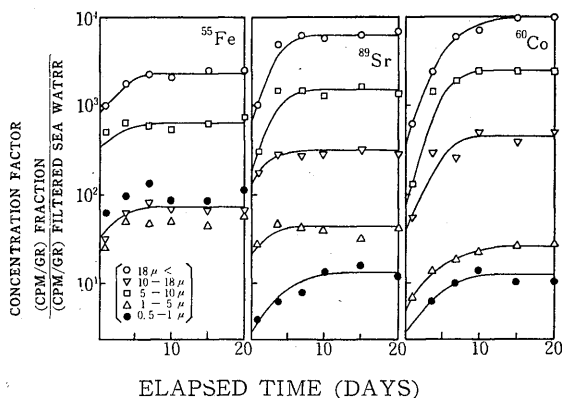


Fig. 16. Uptake of radioactive nuclides in each fraction of suspended matter. (initial activity in sea water = $10^{-4} \mu\text{Ci/ml}$).

des in the suspended matter was observed to reach an equilibrium in about 7-10 days.

The accumulation fraction of radio-activity in each fraction of suspended matter is shown in Fig. 17. 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 that a higher activity of ^{60}Co was observed in the fraction of larger particles, while that of ^{55}Fe in the fraction of smaller particles (Table 5, Fig. 17).

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. 18. A higher activity of ^{55}Fe was also observed in the fraction of smaller particles.

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 the Toyo Filter Paper Type No. 5A are shown in plate 2, 3 and 4 respectively.

It can be seen that the radioactivity was accumulated in relatively large particulates. On the other hand the autoradiograms of radionuclides in the fractions ($5-10 \mu$) recovered by the Millipore Filter Type SM, do not show any large exposed spots as shown in plate 5, 6 and 7.

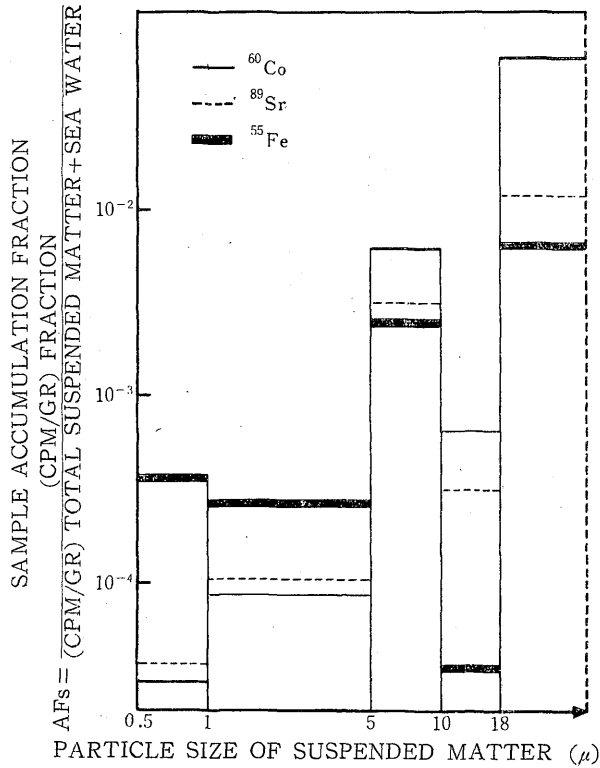


Fig. 17. Sample accumulation fraction of nuclides in each fraction of suspended matter.

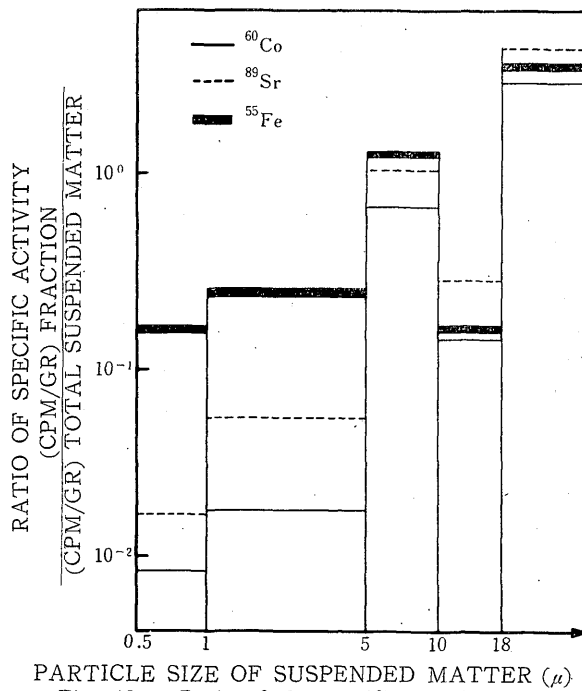


Fig. 18. Ratio of the specific activity in each fraction to the average specific activity of the total suspended matter. (Per dry weight)

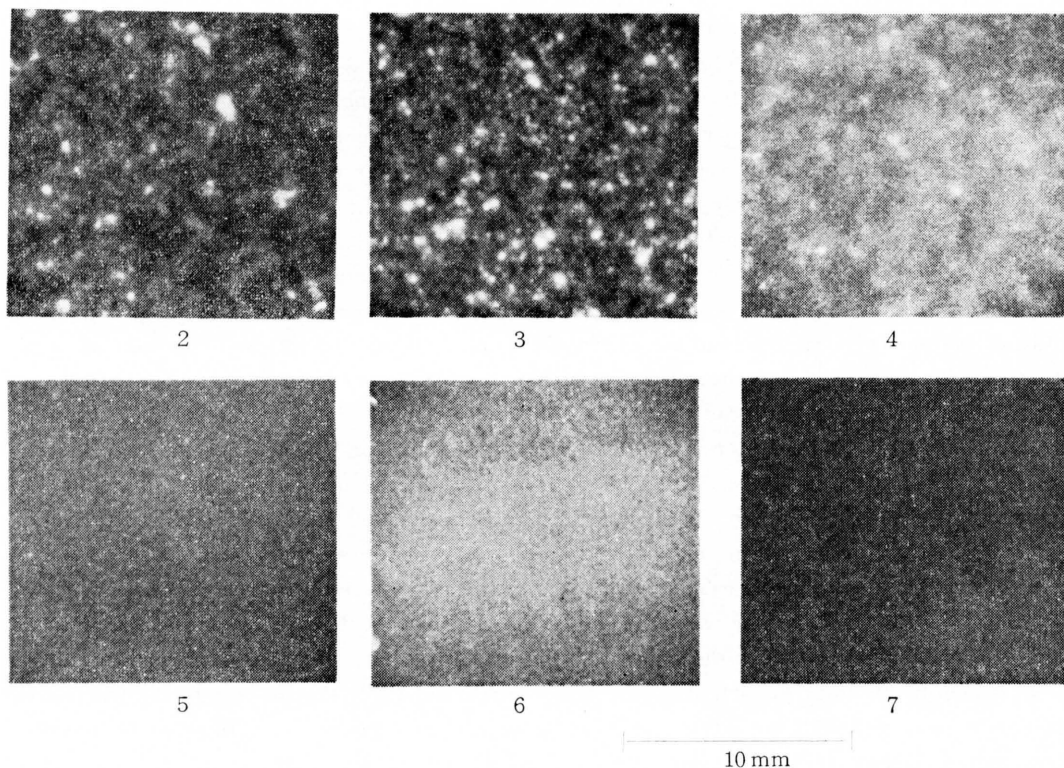


Plate 2-7. The autoradiograms of nuclides in the fraction of suspended matter.

| Plate | Nuclide | Range of Particle Sizes (μ) | Exposure (hr) |
|-------|------------------|-----------------------------------|---------------|
| 2 | ^{59}Fe | > 18 | 48 |
| 3 | ^{60}Co | > 18 | 60 |
| 4 | ^{89}Sr | > 18 | 40 |
| 5 | ^{59}Fe | 5-10 | 48 |
| 6 | ^{60}Co | 5-10 | 60 |
| 7 | ^{89}Sr | 5-10 | 60 |

5. DISCUSSION

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

In the present report, the percentage of the released radioactivities was determined by adding the ion exchange resins containing adsorbed radioactive nuclide into the sea water. This system may be considered an

heterogeneous one consisting of salt solution and particles. It was discussed through what process these adsorbed radioactive nuclides would be released from the resin into the sea water and how they would distribute in the medium.

From these experiments, it was observed that the adsorbed radioactive nuclides were released as soon as the resin particles contacted with the sea water, and that the released nuclides formed a water mass with locally

concentrated radioactivities in the diffusion process and then they diffused gradually through the sea water finally to reach a macroscopically homogeneous equilibrium. In such release experiments, it was found that some nuclides 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 resin to the sea water, and no marked differences in the release were observed for the weight ratio between 10^{-5} — 10^{-7} . The distribution patterns of radioactivity concentration in the sea water after the release seem to depend on the physical-chemical state of nuclides in the sea water. In the experiments with fixed weight ratio of ion exchange resin to the 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 the sea water.

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

Studies on the suspended matter in the sea water have been conducted in the past by many other authors^{6) -15)} from the viewpoint of oceanography and fishery. However, in order to estimate the behaviour and the distribution of radioactive nuclides in the suspended matter in the sea water, the method of the fractional filtration was employed. It was observed that the accumulation of each radionuclides 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 the Toyo Filter Paper

Type No. 5A (mean pore size; 18μ).

It might be as a general rule expected that the smaller the particles, i. e. the larger the surface area per unit weight, would accumulate the larger amount of radioactivity 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 fraction obtained by fractional filtration from natural sea water might have the same composition, and that the organic and inorganic matters 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 that a higher activity of ^{60}Co was observed in the fraction of larger particles, while that of ^{55}Fe in the fraction of small particle sizes. In order to interpret such a tendency on the distributions of radioactive nuclides among the suspended matter in the sea water more adequately, further studies might be necessary.

On assessing the possible effects due to the disposal of radioactive nuclides in environmental hydrosphere, the so-called concentration factor as defined in the beginning of this paper has been used. Oftentimes, the expected average concentration of the activity in the water medium is estimated by simply dividing the total activity of the waste disposal 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 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 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 on evaluating the possible hazard to man at the time of the radioactive waste disposal into the environmental hydrosphere, a much too conservative result may come out for the permissible level of waste disposal, giving the people an erroneous impression. Therefore, in case of the hazard analysis of the waste disposal into hydrosphere, the use of the sample accumulation fraction (AF_s) or the sample concentration factor (CF_s) defined in the below seems to be more reasonable. In the following, some of the relationships and the interrelationships between some of the parameters which may be useful in the analysis of the problems of contamination are introduced.

Notations :

(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 the 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 so-called concentration factor which is the ratio of concentration of 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, at the time of decontaminating 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}

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

(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 the waste disposal into hydrosphere, the expected average initial concentration of the activity (\bar{C}) per unit volume or mass of the water medium is usually estimated by dividing the total activity of the waste disposal 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) in the 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) in the 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) in the above.

From these points of views, 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 also an important factor.

6. SUMMARY

The release of radioactivities from the ion exchange resins into the sea water was studied in the water bath containing natural surface sea water sampled at Osaka Bay using several radionuclides for the different weight ratio of ion exchange resin to the sea water and amount of nuclide initially adsorbed on the ion exchange resins under the static and turbulent conditions. The behaviour and distribution of radionuclides in the suspended matter in the sea water was also studied experimentally by the 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 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. 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 of larger particles recovered by the Toyo Filter Paper Type No. 5 A (mean pore size 18μ), rather than in the fractions of relatively smaller particles.

6. The distribution of radionuclides in each fraction indicated a tendency that a higher activity of ^{60}Co was observed in the fraction of larger particles, while that of ^{55}Fe in the fraction of smaller particles.

7. Finally, after discussing the so-called concentration factor, it is pointed out that it is the ratio of the concentration of activity of the sample to that of the medium, but it 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 activity 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 environmental

hydrosphere.

ACKNOWLEDGEMENT :

Most experimental results in this paper is based on the works conducted under the Atomic Energy Contract No. 1030 (1964) and No. 1142 (1965) between Kinki University Atomic Energy Research Institute and Science and Technology Agency of the Japanese Government on the peaceful uses of the Atomic Energy in Japan. The authors gratefully acknowledge the grant and encouragement by the Atomic Energy Bureau, Science and Technology Agency, Japanese Government.

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