

STUDIES ON RADIOECOLOGICAL CONCENTRATION PROCESSES IN THE AQUATIC ENVIRONMENTS
-UPTAKE AND RETENTION OF Am-241 BY FISH-

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1. INTRODUCTION

Much attention has been paid for the environmental contamination with transuranics, especially plutonium having long half-lives and high radio-toxicity, from nuclear installations or weapon testing. On the other hand, recently, the environmental concern for americium has increased from the radiological protection point of view (1). However, only limited knowledge has been obtained on the behavior of americium in the aquatic environments (2-6). To elucidate radioecological concentration processes of americium by aquatic organisms, the uptake and retention of Am-241 by fingerlings of rainbow trout were investigated under laboratory conditions using natural river water with aeration. The results were compared with those of some other radionuclides such as Co-60, Ru-106, I-131, Cs-137 and Ce-144, in relation to their physical states.

2. MATERIALS AND METHODS

2.1. Radionuclides

Am-241 in $\text{Am}(\text{NO}_3)_3$, Co-60 in CoCl_2 , I-131 in NaI, Cs-137 in CsCl, Ce-144 in CeCl_3 , Ru-106 in chloro and RuNO complexes of RuNO-nitro, -nitrate and -binuclear which were prepared according to the method described by Fletcher et al. (7) and were used in the present experiments.

2.2. Fish and their rearing method

The fingerlings of rainbow trout, *Salmo gairdneri irideus*, were about 5 months old after hatching. The average body length and weight of the fish were 5 cm and 1 g, respectively. The fish were reared with aeration in plastic vessels which held 20 liters of a river water of pH 7.2 contaminated with each radionuclide at the radioactive concentration of about 1-10 $\mu\text{Ci/l}$. The temperature of rearing water was kept at $12 \pm 1^\circ \text{C}$. The fish were transferred to the vessels in a group of about 200 fish when the radioactive concentration of the rearing water reached at an approximate constant level. During retention experiments, the rearing water was frequently changed for the fresh one without radioactivity to prevent a buildup of contaminants. Throughout the experimental periods, the fish were reared without specific feeding.

2.3. Fractionation of radionuclide in rearing water

To elucidate physical states of radionuclides in rearing waters, the fractionation experiments of the radionuclides were carried out by filtrating 5 ml of rearing water with Millipore HAWP filters (0.45 μm of pore size).

The filters which retained Am-241 were also observed by the alpha-track autoradiography using cellulose nitrate film detectors.

2.4. Measurement of radioactivity

The individual fresh sample made up in triplicate was measured for its radioactivity by using automatic gamma counting system equipped with well type NaI(Tl) detector (Auto-well). The Am-241 concentration in samples was also measured by using low energy photon spectrometer (LEPS). Furthermore, the alpha-track autoradiograms were taken with frozen sections of fish.

3. RESULTS AND DISCUSSION

3.1. Fractionation of Am-241 in rearing water

The changes of Am-241 concentration in rearing water are shown in Fig. 1 with the alpha-track autoradiograms of filters retaining Am-241. The results obtained by both Auto-well and LEPS counting showed quite similar trend with each other. The radioactivity of Am-241 retained on filters increased with time after transfer of fish into the water and reached to an apparent equilibrium state in about 5 days. The fractionation of the radionuclides in rearing waters showed that at apparent equilibrium states, about 90% of Am-241 were retained on the filters in the same way as Ce-144, while only about 20% was retained for RuNO-nitrato and less than 10% for the other radionuclides. These results indicate that more than 50% of Am-241 or Ce-144 and some of Ru-106 in fresh water are of particulate form, while most of Co-60, I-131 and Cs-137 are of quite soluble form as previously reported (8). Am-241 should behave in a similar way to the lanthanide elements in the marine environment in many respects (4). In the present studies, Am-241 also behaved in a similar way to cerium in the fresh water environment. As can be seen from the plate in Fig.1, the alpha-track autoradiograms of the filters retaining Am-241 showed homogeneous tracks before transfer of fish into the water, while aggregations observed after transfer of fish seemed to indicate adsorption of Am-241 on suspended matter. Murray and Fukai (9) reported that most of Am(III) in the filtered Var River water of pH 8.1 was in particle form and the particle formation of Am(III) in both river water and sea water seemed to be influenced much more than that of Pu(IV) by the occurrence of unfiltered substances.

3.2. Uptake and retention of Am-241 by fish

The uptake and accumulation of Am-241 in the whole-body of fingerlings are shown in Fig. 2 with those by their tissues of viscera and gills. The curves were drawn on the basis of assumed exponential model (10). The pattern of whole-body uptake of Am-241 was similar to that of Ce-144 and radioactivities in the fish reached apparent equilibrium in about 10 days, whereas only several days were required for the other radionuclides (11). Table 1 summarizes the rate of uptake (u) and the turnover rate (β) calculated from the uptake curves on the basis of exponential model (10). The concentration factors are also listed in this table, though the concentration factor of Am-241 for lake trout in the Lake Michigan was not explicitly reported by Wahlgren et al. (5). As can be seen in this table, the estimated concentration factors from the ratio u/β are quite similar to those observed.

It is noticeable that the highest uptake of Am-241 were found in the viscera including digestive tracts and was followed by that in gills. The alpha-track autoradiograms of frozen section of fish also demonstrated the occurrences of aggregated Am-241 in the digestive tracts.

These results suggest that the fish swallowed suspended matters, on which Am-241 was adsorbed. On the other hand, the highest uptakes among various tissues were found in viscera for Ru-106, Ce-144 and Co-60, in gills for I-131 and in muscle for Cs-137 (11-12).

The retention patterns of Am-241 by whole-body, viscera and gills are shown in Fig. 3.

The whole-body retention of Am-241 was about 30% in 10 days and thereafter gradually decreased. The elimination of Am-241 from the tissues was highest in gills. Comparing the retention in 20 days, only about 9% of whole-body was found for gills, whereas about 63% for viscera including digestive tracts. This higher retention of Am-241 in viscera was similar to the previously reported value for Ce-144 (11).

REFERENCES

- (1) ELLETT, W.H., et al., Allowed health risk for plutonium and americium standards as compared with standards for penetrating radiation, In "Transuranium Nuclides in the Environment", 587 (IAEA-SM-199/113), IAEA, Vienna (1975)
- (2) EMERY, R.M., et al., Ecological behavior of plutonium and americium in a fresh water ecosystem, Phase I. Limnological characterization and isotopic distribution, BNWL-1867 (1974)
- (3) EMERY, R.M., et al., Ecological behavior of plutonium and americium in a fresh water ecosystem, Phase II. Implications of differences in transuranic isotopic ratios, BNWL-1879 (1974)
- (4) NEVISSI, A., et al., Distribution of plutonium and americium in Bikini Atoll Lagoon, Health Physics 28 5 (1975) 539
- (5) WAHLGREN, M.A., et al., Study of the behavior of transuranics and possible chemical homologues in Lake Michigan water and biota, In "Transuranium Nuclides in the Environment", 9 (IAEA-SM-199/44), IAEA, Vienna (1975)
- (6) EDGINGTON, D.N., et al., Plutonium and americium in Lake Michigan sediments, In "Transuranium Nuclides in the Environment", 493 (IAEA-SM-199/44), IAEA, Vienna (1975)
- (7) FLETCHER, J.M., et al., Nitrate and nitro complexes of nitrosylruthenium, J. Inorg. Nucl. Chem., 1 6 (1955) 378
- (8) KIMURA, Y., et al., Uptake and elimination of some radionuclides by eggs and fry of rainbow trout(I), Contributed to J. Radiat. Res. (1976)
- (9) MURRAY, C.N., et al., Adsorption-desorption characteristics of plutonium and americium with sediment particles in the estuarine environment, In "Impacts of Nuclear Releases into the Aquatic Environment", 179 (IAEA-SM-198/25), IAEA, Vienna (1975)
- (10) HIYAMA, Y., et al., Uptake of radioactive nuclides by aquatic organisms: The application of the exponential model, In "Environmental Contamination by Radioactive Materials", 463 (IAEA-SM-117/17), IAEA, Vienna (1969)
- (11) KIMURA, Y., et al., Uptake and elimination of some radionuclides by eggs and fry of rainbow trout(II), Contributed to J. Radiat. Res. (1976)
- (12) ICHIKAWA, R. "Accumulation of radionuclides by fish" Ch. 7, Radioactivity and Fish (EGAMI, N. Ed) In Japanese, Koseisha-Koseikaku, Tokyo (1973)

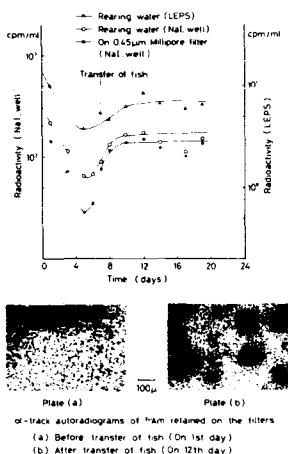


Fig.1 Radioactivity of ^{241}Am in the rearing water

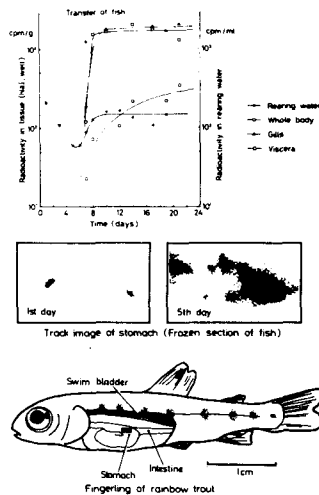


Fig.2 Uptake of ^{241}Am by fingerling

Radionuclide	Rate of uptake (u)	Turnover rate (β)	Concentration factor	
			Estimated (U/β)	Observed (average)
^{241}Am	0.19	0.06	3.2	1.8
^{60}Co	1.82	0.32	5.7	6.5
^{131}I	0.27	0.35	0.8	1.9
^{137}Cs	0.24	0.15	1.6	1.3
^{144}Ce	0.97	0.41	2.4	1.8
^{106}Ru -chloro	0.60	0.89	0.7	0.8
$^{106}\text{RuNO}$ -nitro	—	—	—	1.7*
$^{106}\text{RuNO}$ -nitrate	10.73	3.99	2.7	3.5
$^{106}\text{RuNO}$ -binuclear	0.07	0.07	1.0	0.9

— : Could not be estimated due to death of the fishes

* : On 10th day

Table 1 Rate of uptake, turnover rate and concentration factor for radionuclide in whole-body of fingerling

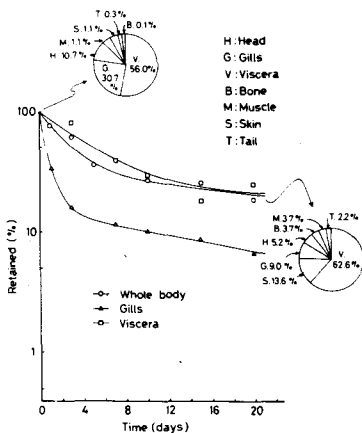


Fig.3 Retention of ^{241}Am by fingerling