On the Concentrations of Radioactive ^{137}Cs and Stable Cs in the Water of the Isolated Undisturbed Mountain Pond "Yasha-ga-Ike"

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Abstract

The concentrations of radioactive fallout ^{137}Cs and stable Cs in the water of the isolated undisturbed pond "Yasha-ga-Ike", located at an elevation of 1099 m near the ridge of mountain in Fukui Pref., were detrmined. The in-situ collection of ^{137}Cs was done by pumping the water to the packed column of acrylic fibre adsorbent, impregnated with copper hexacyanocobalt ferrate, and the ^{137}Cs collected was determined by gamma spectrometry with Ge detector. The water sample was analyzed by ICP-MS to determine the concentration of stable Cs. The temperature of the nine water samples taken in three years ranged from 12.5 to 25.5 ℃. The concentration ranged from 0.00023 to 0.00085 Bq/l for ^{137}Cs and from 0.005 to 0.018 ppb for stable Cs. Both these concentrations increased with increasing water temperature, and the ratio of ^{137}Cs concentration to stable Cs was regarded to be a constant of about 50 Bq/mg. These results indicate that an equilibrium in water between fallout ^{137}Cs in the sixties and the stable Cs existing for a very long time has been established and hence they behave in the same way in the water.

1. Introduction

A small pond named "Yasha-ga-Ike", with maximum depth of 7.5 m and about 40 are of water surface is located at an elevation of 1099 m near the ridge of mountain in Imajyo, Fukui Prefecture. The pond is supposed to be formed by landslide of the surrounding mountain of chert formation in the latter period of the diluvial epoch. The watershed of the pond is small and none of rivers flow in and out. The pond water is said to have never run dry, and a small Japanese diving beetle "Yasha-Gengoro" specially protected due to its rareness lives. Thus the "Yasha-ga-Ike" is regarded as an isolated undisturbed non-volcanic pond, and it is supposed that all the fallouts from the sky have been accumulated in the pond.

The researches on the pond water and the sedimentary grains have been carried out since 1998, and the properties of the pond water[1)] and the adsorptive properties of the elements onto the grains[2)] were reported. In the present study, the concentration in the pond water of radioactive ^{137}Cs from the global fallout of the weapon testings in the sixties was determined for four water samples, and that of stable Cs was determined for nine samples taken in three

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years. And the results obtained were compared and discussed.

2. Experimental Procedures

Both the in-situ collection of Cs from the pond water and water sampling were carried out for determining the concentrations of ^{137}Cs and stable Cs .

2.1 Method of Collection and Measurement of ^{137}Cs from the Pond Water

The in-situ collections of ^{137}Cs from the pond water were conducted four times on June 9, 1999 (noted as D, water temperature:18 ℃), November 7, 1999 (noted as G, 12.5 ℃), July 17, 2000 (noted as I, 25.0 ℃) and July 23, 2001 (noted as L, 25.5 ℃), as given in Table 1.

Table 1 Outline of in-situ ^{137}Cs Collection at the Pond "Yasha-ga-Ike"

Exp. No.	D		G	I		L	
Date	June 9, '99		Nov. 7, '99	July 17, '00		July 23, '01	
Water Temp (℃)	18.0		12.5	25.0		25.5	
Sampling Site	A	B	A	A	B	A	B
Adsorbent	DAW	DBW	GAW	IAW	IBW	LAW	LBW
No. of Column	2	2	1	1	1	1	1
Water supplied to column (l)	480	480	425	510	510	500	500

The method of collecting ^{137}Cs is schematically shown in Fig. 1. The water taken from two neighboring sites A and B, about 10 m apart from the beach and 25 cm deep, was supplied to two separate column systems. From the bottom of the column, the pond water was introduced by an portable tubing pump, driven by a portable dynamo (Honda EX3000, 110V, 3A), at the rate of 100 liter/hr up to about 500 liters. The ^{137}Cs in the pond water was collected on an acrylic fibre adsorbent impregnated with copper hexacyanocobalt(Ⅰ) ferrrate(Ⅱ), according to a method of Higuchi et al.[3]. The fibre of 130 g was packed in the cylindrical acrylic column with an inner diameter of 10 cm and a height of 30 cm. Two columns connected in series were used for each one site at the first collection trial D, but only one column was adoptd from the second, since the ^{137}Cs was found to adsorb only on the inlet thin layer of the fibre close to the bottom of the column. After the in-situ collection, the columns were brought to the laboratory.

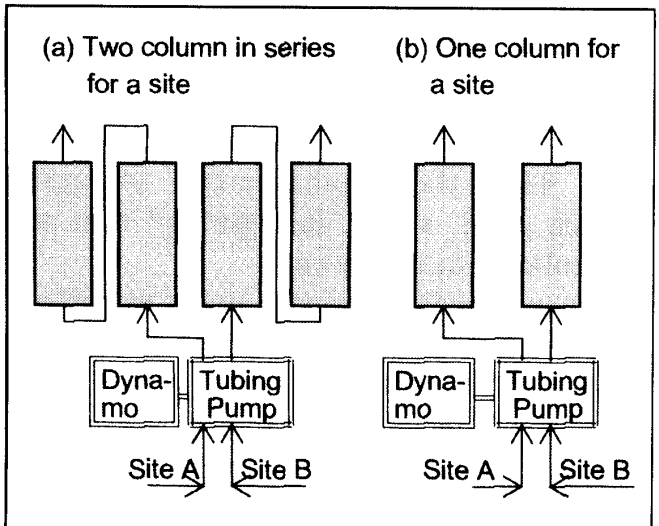

Fig. 1 In-situ ^{137}Cs Collection from Pond Water

For the measurement of ^{137}Cs,

Matsunaga et al.[4] ashed the sample before gamma counting. However, in the present study, the following method shown in Fig. 2 for the case of I sampling was applied. The fibre was taken out from the column IAW, and divided into two portions, lower (IAW1) and upper (IAW2) side, and dried separately. The dried fibre was packed into a stylene vessel with 7.5 cm of inner diameter and 4 cm in height. The radioactivity measurement of the packed fibre was made by gamma-ray spectrometry with Ge(Li) detector (Ortec Co. Ltd). Since the concentration of the adsorbed ^{137}Cs on the fibre was thought to be not uniform throughout the column, measurements were conducted twice for the fibre-packed vessel, as shown by arrows in Fig. 2(a), that is, one for facing the bottom side of the vessel to the detector (IAW1 and IAW2), and the other for the upper lid side (IAW1' and IAW2'). In addition, the spatial distribution of counting efficiency of point source was measured by using an ion exchange bead calibration standard (1 mm in diameter, and gamma emitters of ^{241}Am, ^{57}Co, ^{60}Co and ^{137}Cs are adsorbed, Amersham). Using this point efficiency data, the counting efficiency of the adsorbed fibre portion of the packed vessel was determined, as described later.

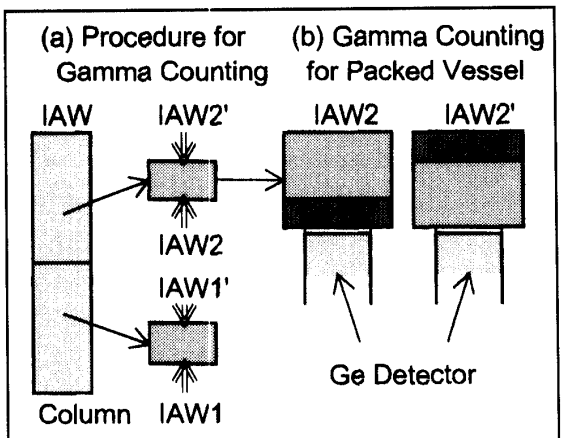

Fig. 2 Gamma Counting of Adsorbent

2.2 Measurement of Stable Cs in the Pond Water

Near the water sampling sites for ^{137}Cs collection, the water samples were taken nine times in three years, where the temperature of water varied from 12.5 to 25.5 ℃. The samples were frozen until measurement. After the melting of the frozen water sample, the water was filtered with 0.42 μm syringe filter, and the concentration of the ^{133}Cs was determined by ICP-MS (Yokogawa HP-4500) using SPEX Multi-Element Plasma Standard XSTC-107 at Research Reactor Institute, Kyoto University.

3. Experimental Results and Discussion

3.1 Concentration of ^{137}Cs in the Pond Water

An example of the measurement of ^{137}Cs radioactivity is shown for the case of I water sample in Fig. 3, where the counting rates from each bottom side of IAW1, IAW2, IBW1 and IBW2 and from each lid of IAW1', IAW2', IBW1' and IBW2' are given for the columns IAW and IBW. Here, the suffix 1 shows the adsorbent fibre of lower half of the column, and 2 that of the upper half. It is seen that the radioactivity was not detected for the upper half, shown as IAW2, IAW2', IBW2 and IBW2', of both columns IAW and IBW and that the counting rate from the bottom side of the vessel packed with the lower half fibre

of the column (IAW1 and IBW1) is higher than that of the rate from the upper lid side (IAW1' and IBW1'). This difference suggests that ^{137}Cs did not adsorbed uniformly whole through the vessel, but adsorbed near the bottom of the vessel. Therefore, the thickness of the uniformly adsorbed layer and the radioactivity of adsorbed ^{137}Cs were estimated in the following way. Assuming that the ^{137}Cs adsorbed uniformly on a certain thickness of the bottom of packed vessel and the countings were held from both the bottom and lid sides, as shown in Fig.2(b), the ratio of both counting rates are compared with the ratio of the counting efficiencies of the uniform layer, which are obtained from efficiency data for the cases of the bottom and the lid side measurements, until both ratios agree. By this procedure the thickness and hence the ^{137}Cs radioactivity can be determined. In this case, the thickness and the radioactivity were determined as 0.4 cm, which corresponds to the height of about 1.5 cm of the column of 30 cm high, and 0.385 Bq for the case of IAW, and 0.6 cm, which corresponds to about 2.3 cm, and 0.413 Bq for the case of IBW. The values of thickness obtained for the other cases were between 0.4 and 0.8 cm, which correspond to 1.5 and 3 cm respectively of the 30 cm column.

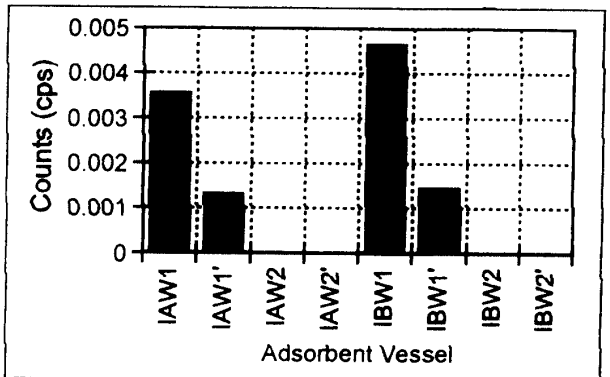

Fig. 3 137Cs Counts of IAW & IBW Adsorbents

The radioactivity measurements for the other cases showed similar results as shown in Fig. 3. In case of the first case D, two columns in series were used, but ^{137}Cs was found only in the lower half of the inlet column. Therefore, the Cs collection after the first case D was carried out by a single column. The results of ^{137}Cs concentrations in the pond water are given in Table 2 for all the cases of D, G, I and L. Since the measurements of ^{137}Cs radioactivity have

Table 2 ^{137}Cs Concentration in the Pond Water

Exp. No.	D		G	I		L	
Date	June 9, '99		Nov. 7, '99	July 17, '00		July 23, '01	
Water Temp (℃)	18.0		12.5	25.0		25.5	
Sampling Site	A	B	A	A	B	A	B
Adsorbent	DAW	BAW	GAW	IAW	IBW	LAW	LBW
^{137}Cs Conc. (Bq/l)	2.64E-4	1.98E-4	2.68E-4	7.55E-4	9.51E-4	8.48E-4	8.34E-4
Ave. Conc. (Bq/l)	0.00023		0.00027	0.00085		0.00084	

been done at different times, the values are corrected to those of the time of the case D. All the cases except G have two experimental values were obtained from the two sampling sites A and B, and so the average value was adopted as an representative of the case. The ^{137}Cs radioactivities ranged from 0.00023 to 0.00085 Bq/l, and they are plotted against the water temperature in Fig. 4. The concentrations of the cases I and L are found to

agree well, although there is one year time lag. The concentration seems to increase with increasing water temperature. These results suggest that the concentration of ^{137}Cs depends on the water temperature.

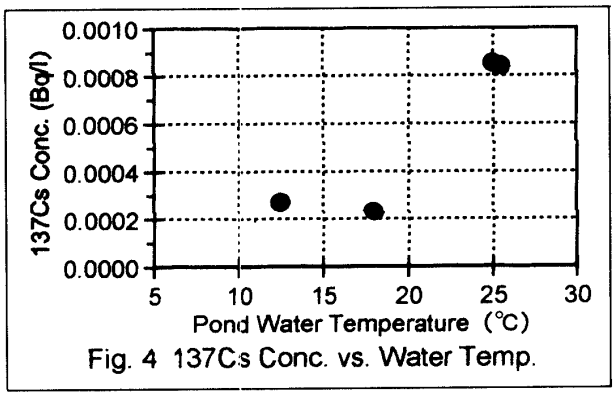

Fig. 4 137Cs Conc. vs. Water Temp.

3.2 Concentration of Stable Cs in the Pond Water

More than four specimens for one water sample was analyzed by ICP-MS, and the average value was taken as the representative. The concentration of stable Cs in the pond water is given in Table 3 for nine water samples including D, G, I, and L. The water samples

Table 3 Stable Cs Concentration in the Pond Water

Exp. No.	D	F	G	H	I
Date	June 9, '99	Oct. 16, '99	Nov. 7, '9	May 23,'00	July 17,'00
Water Sample	DW	FW	GW	HW	IW
Water Temp (℃)	18.0	15.0	12.5	19.0	25.0
Cs Conc. (ppb)	0.005	0.0089	0.0055	0.0122	0.0147
Exp. No.	J	K	L	N	
Date	Oct. 14 '00	May 20 '01	July 23 '01	Oct.13 '01	
Water Sample	JW	KW	LW	NW	
Water Temp (℃)	15.5	20.0	25.5	14.0	
Cs Conc. (ppb)	0.0108	0.011	0.018	0.013	

are named by adding the letter W. The Cs concentrations varied from about 0.005 to 0.018 ppb and the temperature changed from 12.5 to 25.5 ℃. The relationship between the concentration and the water temperature is shown in Fig.5. The concentration seems to increase with increasing the temperature of pond water.

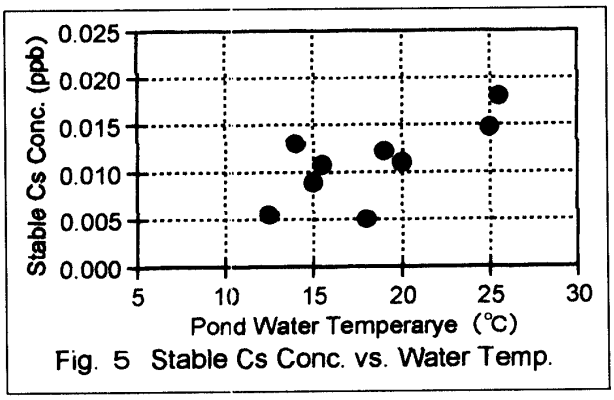

Fig. 5 Stable Cs Conc. vs. Water Temp.

3.3 Relationship Between the Concentrations of ^{137}Cs and Stable Cs

For the data obtained for the cases D, G, I and L, the concentrations of ^{137}Cs are plotted against those of stable Cs in Fig. 6. It is seen that the concentration of ^{137}Cs increases with increasing stable Cs concentration. The ratio of concentration of ^{137}Cs to stable Cs is shown in Fig. 7 for four cases. The ratio seems to be a constant value of about 50 Bq/mg, and does not depend on the time of water sampling nor the water temperature. These results indicate that an equili- brium in water between the ^{137}Cs existing in the pond for these about

40 years and stable Cs existing for a very long time is fully established and hence the ^{137}Cs behaves in the same way as stable Cs in the pond water.

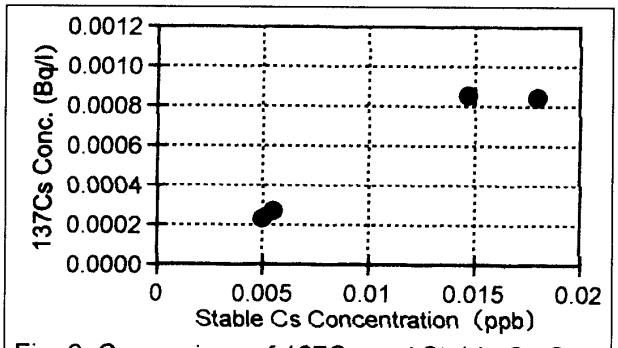

Fig. 6 Comparison of 137Cs and Stable Cs Conc

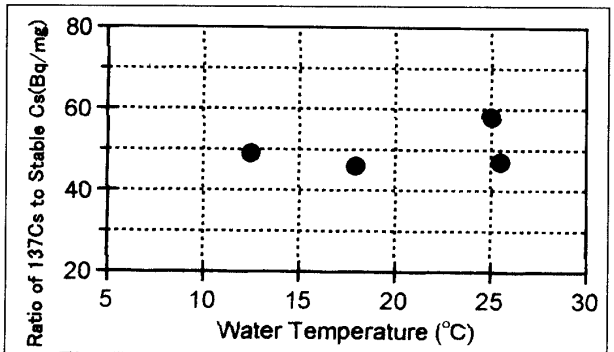

Fig. 7 Ratio of 137Cs to Stable Cs vs. Temp.

4.. Conclusion

The concentrations of radioactive ^{137}Cs and stable Cs in the isolated mountain pond "Yasha-ga-Ike" were determined. The in-situ collection of ^{137}Cs was done by pumping the water to the packed column of acrylic fibre adsorbent, impregnated with copper hexacyanocobalt ferrate, and the radioactivity collected was measured. The concentration of stable Cs in water sample was analysed by ICP-MS. The temperature of the nine water samples taken in three years ranged from 12.5 to 25.5 ℃. The concentration varied from 0.00023 to 0.0085 Bq/l for ^{137}Cs and 0.005 to 0.018 ppb for stable Cs. Both these concentrations increased with increasing the temperature, and the ratio of ^{137}Cs concentration to that of stable Cs was regarded to be a constant value of about 50 Bq^{137}Cs /mgCs. These results indicate that an equilibrium in water between fallout ^{137}Cs in the sixties and the stable Cs existing for a very long time has been established and hence they behave in the same way in the water.

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