Cosmogenic radionuclide ^{22}Na as an index in evaluating residence time of lake water

Y. Ohtsuka, M. Yamamoto[1], K. Sasaki[2] and K. Komura[1]

Institute for Environmental Sciences, Department of Radioecology, Rokkasho, Aomori 039-3212, Japan
[1] *Low Level Radioactivity Laboratory, Kanazawa University, Tatsunokuchi, Ishikawa 923-1224, Japan*
[2] *Department of Cultural Properties and Heritage, Kanazawa Gakuin University, Kanazawa, Ishikawa 920-1392, Japan*

Abstract. Sodium-22 (^{22}Na; $t_{1/2}$ = 2.6 y, β^+) is naturally occurring radionuclide produced by a spallation between secondary neutron from cosmic ray and argon in the atmosphere and is transported to land and lake surface with rain directly and/or *via* fluvial channels. Because sodium is soluble completely in aqueous systems, ^{22}Na may be useful as an index in evaluating residence time of lake water. The atmospheric deposition rate, fluvial input/output and vertical distribution in lake water of this nuclide were measured in Lake Biwa which is most important source as drinking, industrials for 14,000,000 people in Kinki district, Japan. The mean deposition rate and the fluvial input to the lake surface were estimated to be 0.26 Bq/m^2/y (171 MBq/y in whole lake) and 0.20 Bq/m^2/y (103 MBq/y in whole lake), respectively. There was no substantial gradient of ^{22}Na concentration in the water column through a year. The ^{22}Na concentrations in the lake waters ranged from 0.011 to 0.029 Bq/m^3 and total activity was estimated to be 539 MBq. The mean residence time of ^{22}Na in Lake Biwa was evaluated to be 4 - 6 years, which is comparable to that of lake water (5.3 – 6.2 years).

1. INTRODUCTION

Cosmogenic radionuclides are produced by nuclear reactions between atmospheric constitutions such as nitrogen, oxygen and argon, and secondary neutrons from primary cosmic ray. They have been observed in rain waters and air. These nuclides are transferred to land, lake and ocean surface by precipitations directly and/or *via* fluvial channels in the watershed. Sodium-22 is one of the cosmogenic radionuclides. It is mainly produced constantly by a spallation reaction between argon and secondary neutrons in the stratosphere. Its natural production rate is very small, however, the radiation dosage to men due to ^{22}Na is reported to be higher than that of other naturally occurring radionuclides, such as ^{3}H and ^{7}Be [1]. Since Perkins and Nielsen measured ^{22}Na in rain water [2], this nuclide has been recognized as a useful tracer of air masses on the meteorology similarly to ^{7}Be. For example, $^{22}Na/^{7}Be$ ratio would be used as a sensitive tool for studying the seasonal exchange of stratospheric - tropospheric air [3].

In fresh water systems, ^{22}Na in rivers and lakes was scarcely observed because this nuclide content was very low. However, this isotope may be a useful tracer in the elucidation of geochemical processes of conservative materials because sodium is

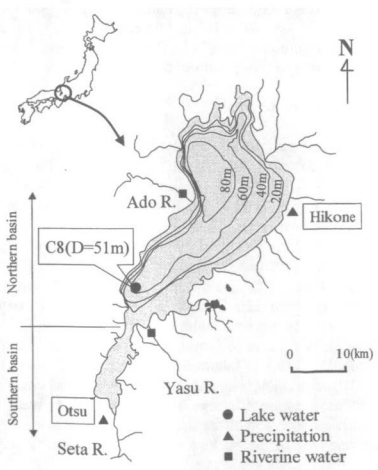

Fig. 1 Study area

Table 1 Physical properties of Lake Biwa [4]

	Unit	North Basin	South Basin	Total
Surface area	km^2	616	58	670.49
Volume	$x10^9 m^3$	27.3	0.2	27.5
Maximum depth	m	103.58	8	103.58
Mean depth	km	43	4	41
Altitude	m	-	-	85.614
Circumference	km	-	-	235.2
Catchment area	km^2	-	-	3174

completely soluble in fresh water systems. For example, the mean residence time of ^{22}Na may become a good indicator of water stagnation. Establishment of ^{22}Na measurement was, therefore, attempted together with geochemical application of it in limnetic systems.

We report here, (1) the technique of low-level ^{22}Na measurement in the natural water and (2) the mass balance calculation of ^{22}Na in a freshwater system, Lake Biwa in Japan.

2. SAMPLING AND MATERIALS

2.1 Study area

Lake Biwa is situated in the center of the Japanese archipelago and is the largest freshwater basin in Shiga Prefecture of Japan, with a total surface area of 670 km^2, volume of 27.5 x 10^9 m^3 and maximum depth of more than 100 m. The physical properties of this lake are given in Table 1. It is also one of the ancient lakes to be compared with the Caspian Sea in central Asia or Lake Baikal in Russia. It has more than 400 input rivers and its outflow is only Seta River which is a tributary of Yodo River flowing down to Osaka Bay. The water of Lake Biwa is utilized for drinking by fourteen million people in the Kinki district (Kyoto-Osaka-Kobe metropolitan areas). The total fluvial inflow and groundwater input to the lake are evaluated to be 3.70 x 10^9 m^3/y and (0.7 - 1.1) x 10^9 m^3/y, respectively [5]. The amount of precipitation and evaporation on/from Lake Biwa and its watershed are reported to be (7.3 - 7.5) x 10^9 and (2.1 - 2.9) x 10^9 m^3/y, respectively. Limnological water residence time is estimated to be 5.3 - 6.2 years [5].

2.2 Sample collections

Lake water samplings were performed at depth of 10, 20, 30 and 40 m in the northern part of the lake (135°58'00" N, 35°10'30" E, Depth = 51 m, Fig. 1) in July, August and November 1999. River water samplings were also carried out at the center of Ado and Yasu Rivers (Fig. 1). The sampling points were situated above about 3 km from each estuary. The sum inflow for this two rivers occupied 23 % of all fluvial water inflows [4].

^{22}Na of the waters in the lake and rivers were preconcentrated by using samplers (see Fig. 2) which had a pump connected with cartridge filter folder (ADVANTEC, Japan, TCW-05-PPS) and ion exchange resin column (ϕ85 mm x 750 mm; ORGANO, Japan; Amberlite: cation exchange resin (IR-120B) and anion exchange resin (IRA-410)). Total volume of collected water was measured by flow meter attached to the end of the resin column. Flow speed was 0.7 - 2.0 L/min and water of about 500 L was passed thought its column for about 4 - 5 hours. After sampling, resins were transferred into plastic cases and brought back to our laboratory (Low Level Radioactivity Laboratory; LLRL).

Precipitation (wet and dry deposition) was collected at Otsu and Hikone from April 1997 to August 1998 at one month interval (Fig. 1). A layer of water at a bucket (0.2 m^2) was constantly maintained to avoid resuspension of dry depositions. In this case, ^{22}Na is undetectable, but ^{7}Be detection is easy because surface area of the buckets at Otsu and Hikone is very small for ^{22}Na detection. The deposition of ^{22}Na

was, therefore, evaluated on the basis of the amount of ^{7}Be depositions in Otsu and Hikone, and annual mean $^{7}Be/^{22}Na$ activity ratio (9.13×10^{3}) in deposition at LLRL where is located at 150 km north away from Lake Biwa,

2.3 Chemical separation and measurements

Sodium in Amberlite resin was removed by washing the resin three times with 3M-HCl. The obtained solution (about 3 liters) was evaporated to dryness. The residue was packed in to a plastic tube (ϕ 18 x 50 mm). ^{22}Na in the tube was measured for 5 - 10 days by using extremely low-background well type HPGe semiconductor detector (Eurisys mesures, France, Relative efficiency 65 %). The detector is special designed for low-level counting set in the tunnel (270 mwe) of former Ogoya copper mine located about 20 km from LLRL [6]. The concentration of ^{22}Na was calculated by using sum peak of 1786 keV (1275 + 511 keV), because that Compton scattring of the 1460 keV gamma-ray from ^{40}K seriously interfaces the detection of 1275 keV emitted directly from ^{22}Na. The recovery was evaluated by comparing the Na concentration between lake (river) water and water after passing through the resin column. Sodium contents in the samples were measured by ICP-AES (VARIAN, Liberty, UK). The recoveries were found to be 98 - 99 % for all samples.

Precipitation was directly passed through the Powdex® (PAO + PCH) resin column [7]. A mixture of PAO and PCH resins effectively collected ionic and particulate radioactive materials from rain water [7, 8, 9]. This column had ability to collect more than 99 % of the sodium ions in rain water [7]. After the resin was dried in an oven at about 100℃, it was packed into a plastic case (ϕ 50 x 50 mm) for determining ^{7}Be by using usual Ge detector.

For the determination of ^{22}Na in precipitation at LLRL, in which the surface area (0.5 m^2) of sampler is 2.5 times larger than that at Otsu and Hikone, precipitation was treated in a similar manner as above and the obtained Powdex resin was ashed at 350℃ in an electric oven overnight. The ash was then packed into a plastic tube (ϕ 18 x 50 mm) and measured for about one week using well type HPGe semiconductor detector.

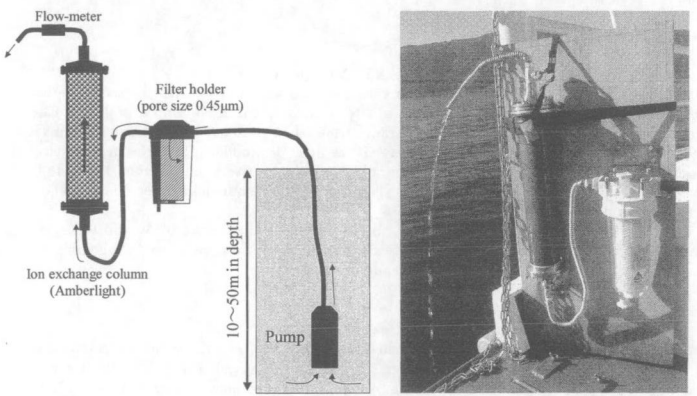

Fig.2. Sampling system for collecting Na in fresh water

Table 2 Results of ^{22}Na concentrations in waters of the lake and rivers and deposition flux to the lake

Lake water				
Date	Depth(m)	Concentration (Bq/m^3)	Volume (10^9x m^3)**	Total activity (MBq)
Jul. 1999	10	0.0205 ± 0.0092	-	
	10	0.0225 ± 0.0101	-	
	40	0.0185 ± 0.0083	-	
	40	0.0160 ± 0.0072	-	
Aug. 1999	10	0.0182 ± 0.0082	-	
	20	0.0293 ± 0.0132	-	
	30	0.0196 ± 0.0088	-	
	40	0.0181 ± 0.0081	-	
Nov. 1999	10	0.0167 ± 0.0036	-	
	20	0.0209 ± 0.0035	-	
	30	0.0237 ± 0.0035	-	
	40	0.0111 ± 0.0033	-	
Mean	-	0.0196 ± 0.0045*	27.5	539
Fluvial water				
Date	Sites	Concentration (Bq/m^3)	Inflow (10^9x m^3/y)**	Input (MBq/y)
Nov. 1999	Ado R.	0.0162 ± 0.0033	0.49	-
	Yasu R.	0.0431 ± 0.0117	0.37	-
Weight mean		0.0277 ± 0.0041	3.70	103
Rain water				
Date	Sites	Deposition flux (Bq/m^2/y)	Inflow (10^9x m^3/y)**	Input (MBq/y)
1997-1998	Otsu	0.249 ± 0.008	-	-
	Hikone	0.261 ± 0.008	-	-
Mean		0.255 ± 0.008	1.14	171

*: Error is one sigma standard deviation. **: Reference [4].

3. RESULTS AND DISCUSSION

3.1 Vertical distribution of ^{22}Na in lake water and ^{22}Na input fluxes

^{22}Na concentrations measured for lake and river waters are shown in Table 2. Also are shown in this table the results of precipitation. The concentrations of ^{22}Na were detected in all samples collected. The ^{22}Na concentrations in lake waters were in a narrow range from 0.0111 to 0.0293 Bq/m^3 with a mean value of 0.0196 Bq/m^3. There were scarcely gradients in vertical distributions of ^{22}Na in water column, independent on the sampling time. The concentrations of ^{22}Na in rivers ranged from 0.0162 to 0.0431 Bq/m^3 with a weighted mean of 0.0277 Bq/m^3. The mean ^{22}Na content in lake water appears to be a little lower than that in river water.

The ^{22}Na deposition fluxes at Otsu and Hikone were estimated to be 0.249 and 0.261 Bq/m^2/y respectively. These deposition fluxes were nearly similar to that in Fukui Prefecture (neighboring to Shiga Prefecture) evaluated previously by Tokuyama *et al.* [10].

3.2 ^{22}Na mass balance and residence time in this system

The total amount of ^{22}Na in the lake was estimated to be 539 MBq from the mean concentration and the total volume of lake water. Total fluvial input (103 MBq/y) was calculated from the average river concentration and total volume of inflow to lake. Atmospheric deposition to lake surface was 171 MBq/y. ^{22}Na output through Seta River was estimated to be 87 MBq/y, by assuming that the concentration of ^{22}Na in Seta River was equal to that of lake surface water (Fig. 3 (a)).

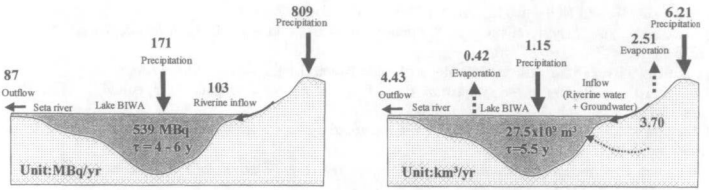

Fig. 3 Mass balance of ^{22}Na (a) and water (b) in Lake Biwa. (Ref. [5])

The mass balances of ^{22}Na in Lake Biwa can be written under a steady-state condition as follows:

$$\frac{\Delta L_{Na}}{\Delta t} = A_{Na} + R_{Na} - L_{Na} \cdot (\lambda + k) \qquad \text{---------- (1)}$$

$$\tau = \frac{1}{k} \qquad \text{---------- (2)}$$

Where, L_{Na}, R_{Na}, λ and k are total amount (539 MBq) of ^{22}Na in the lake water, annual depositon (171 MBq/y) of ^{22}Na to the lake surface, annual fluvial input (103 MBq/y) of ^{22}Na, decay constant (0.266 y^{-1}) and removal rate of ^{22}Na including ^{22}Na outflow from Seta River, respectively. The mean residence time (τ; year) was defined as inverse of k.

The ^{22}Na contribution from the groundwater was assumed to be negligibly small because it might be very low due to the ^{22}Na decay through the underground. By substituting the above values to Eq. (1), the τ value of 4.1 years was obtained. On the other hand, the value of k can be also calculated from the ^{22}Na outflow as shown in the following equation.

$$L_{Na} \cdot k = O_{Na} \qquad \text{-------- (3)}$$

Where, O_{Na} is annual outflow of ^{22}Na from the Seta River. As a result, the τ value was evaluated to be 6.2 years. Difference between two τ values seems to be a rather small and may be due to the errors of parameters such as the volume of fluvial inputs/output and ^{22}Na concentrations of them. Further inventigations are needed for evaluating exactly these parameters. The residence times for ^{22}Na evaluated are comparable to the water residence time (5.5 years) shown in Fig. 3 (b) [5], although the values from 5.3 to 6.2 years as water residence time have been reported by some investigators [5].

Thus, ^{22}Na might be useful as an index in evaluating not only the residence time of lake water itself but also behavior of conservative elements.

Acknowledgments

This work was supported by the Sasakawa research finances (1998). We express thanks to Dr. K. Yokota, Lake Biwa Research Institute of Shiga Prefecture in Japan, for his help in sampling of lake water and precipitation.

References

[1]UNSCEAR, *Sources and effects of ionizing radiation*, UNSCEAR 2000, United Nations, New York (2000) pp. 89.

[2] Perkins R. W. and Neilsen J. M., *Health Phys.* **11** (1965) 1297-1304.
[3] Luyanas V. Yu., Yasyulyonis R. Yu., Shopauskiene D. A., and Styra B. I., *J. Physical Res.* **75** (1970) 3665-3667.
[4] Lake Biwa Research Institute, Data Book of Lake Biwa, (LBRI, Japan, 1988, *in Japanese*).
[5] Fushimi E., in *Report of the symposium on Lake Biwa Research*, Japan, 1990, edited by Lake Biwa Research Institute, pp. 17-37. (*in Japanese*)..
[6] Komura K., in *Proceedings of the 1997 international symposium on environmental radiation, Japan, 1997*, pp. 56 - 75
[7] Matsuura H. Kitagawa T., *Annual report of Fukui Prefectural Institute of Public Health, Fukui-ken* **20** (1983) pp. 55-67 (*in Japanese*).
[8] Tokuyama H. and Igarashi S., *J. Environ. Radioactivity.* **38** (1998) 147-161.
[9] Kimura T. and Hamada T., *Radioisotopes* **27** (1978) 348-349.
[10] Tokuyama H., Oonishi M. and Matsuura H., *J. Environ. Radioactivity.* **21** (1993) 213-218.