World Journal of Nuclear Science and Technology, 2014, 4, 88-95

http://dx.doi.org/10.4236/wjnst.2014.42014

Transference Kinetics of Radionuclide ^{95}Nb in the Aquatic Ecosystem

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Received 22 December 2013; revised 27 January 2014; accepted 14 February 2014

Abstract

The dynamics of transportation, accumulation, diminishment and distribution of ^{95}Nb in a simulated aquatic ecosystem was studied using the isotope-tracer technique, and a fitting equation was established by application of a closed, five-compartment model. The results showed that when ^{95}Nb was introduced into an aquatic system, it was transported and transformed via deposition in combination with other ions, and adsorption and absorption by aquatic organisms, resulting in redistribution and accumulation in different parts of the organisms. Following addition, the specific activity of ^{95}Nb in water decreased sharply within a short time, and then after reaching a certain value, it decreased more slowly. Sediment accumulated large amounts of ^{95}Nb through the exchange of ions. Hyacinth (*Eichhornia crassipes*) also adsorbed a large amount of ^{95}Nb in a short period of time. Snails (*Bellamya purificata*) and fish (*Carassius auratus*) were found to have a poor adsorption capacity of ^{95}Nb. The amount of ^{95}Nb found in the snail flesh was greater than that in the shell, and the ^{95}Nb found in the fish was mainly distributed in the viscera. The amount of ^{95}Nb in each individual component of the experimental system was affected over time.

Keywords

^{95}Nb, Transference, Enrichment, Distribution, Water, Sediment, Hyacinth, Snail, Fish

1. Introduction

The study of the effects of nuclear fission products on the environment is an active field and is needed for maintaining harmonization between the sustainable development of nuclear power and high environmental quality.

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How to cite this paper: Zhao, X.Y., *et al.* (2014) Transference Kinetics of Radionuclide ^{95}Nb in the Aquatic Ecosystem. *World Journal of Nuclear Science and Technology*, **4**, 88-95. http://dx.doi.org/10.4236/wjnst.2014.42014

^{95}Nb is one of the primary fission products in a nuclear reactor. A primary radionuclide in the radioactive liquid effluents from a pressurized water reactor, its half-life is 35.1 days [1]. In safety assessment of French long-lived nuclear waste disposal, data concerning the mobility and the bioavailability of Nb in soils are needed as well as general trends of its fate in the specific environment around the site of French underground research laboratory [2]. Relevant reports on ^{95}Nb focus on environmental monitoring and evaluation after nuclear accidents, as well as the absorption, distribution and accumulation in natural ecosystems [3]-[7].

In this study, the isotopic tracer technique is used to study the dynamics of ^{95}Nb in a simulated aquatic ecosystem, and its dynamics are described mathematically using a compartment model with a non-linear fitting method.

2. Materials and Methods

2.1. Isotope

$^{95}Nb_2O_5$, in the form of a black powder, was supplied by the Academy of Atomic Energy of China. Its specific radioactivity was 1.14×10^8 Bq·g^{-1} and its radiochemical purity is greater than 95%. Prior to use, it was chemically transformed with HF into $^{95}NbF_5$, at a concentration of 6.17×10^6 Bq·mL^{-1} [8]. Then the $^{95}NbF_5$ solution was diluted to a concentration of 6.17×10^4 Bq·mL^{-1} for this experiment.

2.2. Soil

Sediment was paddy soil on powdery loam from an experimental farm in the Huajiachi campus of Zhejiang University. It was air-dried, pulverized, and sieved to remove stones and plant debris prior to use. The physical-chemical properties of the soil are summarized in **Table 1**.

2.3. Aquatic Ecosystem

The ecosystem was composed of water, sediment, aquatic organisms: hyacinth (*Eichhornia crassipes*), snails (*Bellamya purificata*), and fish (*Carassius auratus*).

2.4. Methods

Three glass tanks with dimensions of 60 cm × 40 cm × 50 cm were constructed. 8 kg of paddy soil on powdery loam as sediment was filled into each pool. The sediment thickness was about 5 cm. The tanks were flooded with 60 L of water, and it was buried 12 plastic sediment sampler with Φ 1 cm × 5 cm. 10 mL of $^{95}NbF_5$ with a specific activity of 4.32×10^5 Bq·mL^{-1} was introduced into the water after one week. The water was stirred gently to get a homogeneous distribution of radionuclide, instantly taking 20 mL water samples. The specific activity of the radionuclide in water was 72 Bq·mL^{-1}. Fifteen water hyacinth, and 30 snails and 15 fish were put into each tank. Each fish weighed between 130 - 220 g. During the period of the experiment, a filling oxygen pump was used to keep the dissolved oxygen in the water constant throughout day and night. Water was added at intervals of 3 days in order to maintain a constant volume of water.

2.5. Sampling

Samples were collected at time intervals of 1, 2, 4, 6, 9, 15, 22, 29 and 34 days, from each pool by randomly taking four 5 mL aliquots of water and depositing them in disposable plastic cups for activity measurements. Each sample from each pool was thus 20 mL. A hyacinth, and 3 snails and a fish were randomly taken from each tank. The hyacinth was divided into root, gourd, leaf, and snails were divided into flesh and shell. The fish was washed in water, and then dried with absorbent paper. The fish was dissected after weighing, and divided into fin, scale, viscera, skin, gill, flesh, skeleton and head. In total, there were eight parts in all (or nine parts

Table 1. Physico-chemical properties of the soil studied.

pH (in water)	Organic matter (%)	CEC (cmol·kg^{-1})	Clay (0.001 mm) (%)
6.0	1.9	0.5	12.5

including the fish eggs). Each part was weighed individually and then cut into smaller pieces. Twenty gram samples from each part were put into the disposable plastic cups for measurements.

In the meantime, three sediment columns were collected from each tank using a sediment sampler. Each sediment column was sectioned into two equal parts, and each part was smashed and mixed thoroughly. Afterwards, 20 g of sediment samples were put into plastic cups for measurement. This was repeated 3 times for each sample.

2.6. Measurements and Statistical Analysis

^{95}Nb emits β and γ particles when it decays. Those were measured with a multi-channel γ spectrometer (model BH-1224, Beijing Nuclear Instrumentation Factory) equipped with a NaI scintillation detector (70 mm in diameter). The spectrometer was installed in a lead-shielded chamber. Disposable plastic cups of 75 mm diameter and 110 mm height were placed on the top of the scintillation detector during measurements. A homemade location device was used for fixing the counting position, to ensure homogeneity in the geometrical position of all samples. The counting error was controlled to be lower than 5%. All samples were measured on the sampling day to avoid errors due to water evaporation. The counts were calibrated with counting efficiency, diminished time, disintegration and other factors [9].

A non-linear regression analysis method was used to describe the variation dynamics of ^{95}Nb in the individual compartment of the system [10].

3. Results and Discussion

3.1. Increase and Decrease of ^{95}Nb in the Pool Water and Sediment

The ^{95}Nb specific activity in the pool water and sediment on different days are shown in **Table 2**. The specific activity of ^{95}Nb in the pool water decreased rapidly in the first few hours due to the deposition, complexation, and adsorption and absorption by the sediment and aquatic living organisms. After 1 day, the specific activity of ^{95}Nb in the pool water reduced to 19.52 Bq·mL^{-1}, only 27.11% of the initial specific activity (72.0 Bq·mL^{-1}). From then on, it decreased gradually, and 29 days later, the exchanging of ^{95}Nb in the pool water with ions in the ecosystem reached dynamic equilibrium.

As shown in **Table 2**, due to the deposition of ^{95}Nb in water and exchange with ions in the soil, the specific activity of ^{95}Nb in the sediment increased rapidly in the first four days to a maximum of 156.55 Bq·g^{-1} on Day 4, and decreased to an average of 118.93 Bq·g^{-1} in the following days. This is because the ^{95}Nb was mainly located within the upper sediment and the Nb ions that were not combined closely with sediment began desorbing. As time went on, ^{95}Nb moved into and combined stably with the deep sediment through the processes of complexation and the adsorption of iron and manganese oxides [11]-[14].

Table 2. Change of ^{95}Nb specific activity in water and sediment with time.

Time (d)	Water/Bq·mL^{-1}	Sediment/Bq·g^{-1}		
		Upper (0 - 2.5 cm)	Lower (2.5 - 5 cm)	Average
1	19.52 ± 1.94	188.70 ± 7.79	1.25 ± 0.10	95.61 ± 7.08
2	12.23 ± 1.13	288.32 ± 12.18	3.04 ± 0.47	147.21 ± 7.19
4	8.22 ± 0.91	304.21 ± 12.68	4.43 ± 0.88	156.55 ± 7.62
6	5.60 ± 0.78	243.05 ± 10.39	7.34 ± 0.93	131.13 ± 6.89
9	4.46 ± 0.63	229.64 ± 9.61	9.60 ± 0.97	122.16 ± 6.79
15	2.35 ± 0.19	218.47 ± 9.12	12.52 ± 1.04	123.46 ± 6.63
22	0.59 ± 0.02	209.05 ± 9.06	13.37 ± 1.13	119.67 ± 6.58
29	0.31 ± 0.00	196.64 ± 8.57	14.23 ± 1.47	111.69 ± 6.17
34	0.23 ± 0.00	185.85 ± 6.97	16.20 ± 1.91	105.45 ± 6.09

3.2. Increase and Decrease of ^{95}Nb in Hyacinth and Snail

The ^{95}Nb specific activity in hyacinth and snail with time were given in **Table 3**. On Day 1, the ^{95}Nb specific activity in hyacinth increased rapidly to a maximum of 247.25 Bq·g^{-1}, and then in the following days, the ^{95}Nb specific activity in hyacinth decreased due to being deabsorbed. Only a little ^{95}Nb was absorbed and steadily combined by hyacinth due to the fact that ^{95}Nb was not an essential element of the organisms. With a water exchange *in vitro* and *in vivo* of the snails, a large amount ^{95}Nb went into the snail body, and the ^{95}Nb specific activity of the snails increased sharply and started to decrease after reaching a maximum. Subsequently, because the specific activity of ^{95}Nb in the water sharply decreased, the ^{95}Nb in the snails began desorbing and the specific activity was essentially at equilibrium 15 days later. The specific activity of ^{95}Nb in the snail flesh was obviously higher than that in the snail shell [15] [16].

3.3. Increase and Decrease of ^{95}Nb in Fish

The dynamic migration and distribution of ^{95}Nb in different parts of the fish in the last sample are given in **Table 4**. The specific activity data of ^{95}Nb in the whole fish is equaled from weighing each part of the body. The

Table 3. Relationship between specific activity and time of ^{95}Nb in aquatic organisms in hyacinth and snail.

Time (d)	Hyacinth of specific activity/Bq·g^{-1}				Snail of specific activity/Bq·g^{-1}		
	Leaf	Gourd	Root	Whole plant average	Flesh	Shell	Whole snail average
1	286.60 ± 12.03	134.13 ± 7.19	319.01 ± 12.98	247.25 ± 10.47	37.54 ± 2.87	17.76 ± 1.40	29.44 ± 2.42
2	259.08 ± 11.08	119.12 ± 6.86	285.39 ± 12.03	221.19 ± 10.87	28.65 ± 2.09	12.98 ± 1.05	17.55 ± 1.59
4	234.23 ± 10.28	112.29 ± 6.65	269.05 ± 11.81	208.53 ± 9.68	17.72 ± 1.91	6.30 ± 0.89	9.75 ± 0.93
6	240.64 ± 10.57	110.63 ± 6.98	265.08 ± 11.33	205.45 ± 9.13	9.87 ± 0.83	4.97 ± 0.59	6.97 ± 0.88
9	163.01 ± 7.58	74.96 ± 4.97	179.60 ± 7.91	139.22 ± 8.03	6.00 ± 0.90	2.96 ± 0.30	3.96 ± 0.75
15	114.28 ± 6.63	52.83 ± 3.56	126.53 ± 7.26	98.12 ± 5.93	6.02 ± 0.87	2.92 ± 0.23	3.93 ± 0.79
22	114.42 ± 6.61	52.61 ± 3.47	126.04 ± 7.01	97.70 ± 5.91	5.98 ± 0.93	2.89 ± 0.27	3.85 ± 0.73
29	107.20 ± 6.91	49.29 ± 3.52	118.09 ± 6.93	91.53 ± 5.63	5.77 ± 0.83	2.73 ± 0.21	3.69 ± 0.64
34	106.81 ± 6.19	49.11 ± 3.13	117.68 ± 6.96	91.21 ± 5.42	5.74 ± 0.88	2.81 ± 0.20	3.64 ± 0.59

Table 4. Relationship between specific activity and time of ^{95}Nb in aquatic organisms in fish/Bq·g^{-1}.

Parts of fish	Sampling time/d								
	1	2	4	6	9	15	22	29	34
Gill	8.36 ± 0.87	7.97 ± 0.83	6.20 ± 0.77	4.93 ± 0.52	3.05 ± 0.37	1.41 ± 0.11	1.93 ± 0.18	0.92 ± 0.08	0.89 ± 0.09
Fin	1.35 ± 0.11	9.60 ± 0.08	1.41 ± 0.10	0.43 ± 0.03	0.62 ± 0.05	0.68 ± 0.06	0.29 ± 0.00	0.19 ± 0.00	0.17 ± 0.00
Viscera	104.11 ± 5.97	140.58 ± 8.16	40.17 ± 2.89	7.99 ± 0.89	9.27 ± 0.90	7.34 ± 0.84	1.83 ± 0.15	1.71 ± 0.13	1.68 ± 0.14
Skin	0.56 ± 0.00	0.99 ± 0.10	0.19 ± 0.00	0.07 ± 0.00	0.04 ± 0.00	0.05 ± 0.00	0.09 ± 0.00	0.03 ± 0.00.	0.01 ± 0.00
Scale	0.22 ± 0.00	0.71 ± 0.00	0.17 ± 0.00	0.28 ± 0.00	0.17 ± 0.00	0.16 ± 0.00	0.11 ± 0.00	0.09 ± 0.00	0.07 ± 0.00
Flesh	0.03 ± 0.00	0.06 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Skeleton	0.17 ± 0.00	0.21 ± 0.00	0.03 ± 0.00	0.00 ± 0.00	0.05 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
Head	0.23 ± 0.00	0.26 ± 0.00	0.15 ± 0.00	0.12 ± 0.00	0.16 ± 0.00	0.09 ± 0.00	0.08 ± 0.00	0.02 ± 0.00	0.03 ± 0.00
Eggs	0.35 ± 0.00	0.00 ± 0.00	0.07 ± 0.00	0.00 ± 0.00	0.03 ± 0.00	0.01 ± 0.00	0.00 ± 0.00	0.04 ± 0.00	0.00 ± 0.00
Whole fish	6.85 ± 0.83	11.52 ± 1.08	3.38 ± 0.47	0.35 ± 0.02	1.41 ± 0.13	0.77 ± 0.08	0.20 ± 0.00	0.14 ± 0.00	0.07 ± 0.00

results showed that the organs taking up and adsorbing ^{95}Nb were mainly the intestines and stomach (viscera). ^{95}Nb was absorbed by the gill and fin through direct contact with water. The specific activity of ^{95}Nb in these three parts reached a maximum in 2 days, after which ^{95}Nb rapidly reduced in water and then gradually decreased tendency with time. 4 days later, the specific activity of ^{95}Nb in these three parts showed a decreased tendency to balance, and the general trend was similar to the increase and decrease found in the snails. The specific activity of ^{95}Nb in flesh, bone, liver and eggs was relatively low, and were only a little higher than the background level. It was indicated that ^{95}Nb remained in the intestines, stomach, gill and fin, and could not readily transport into the inner organs such as the flesh, bone, liver and eggs. The absorption capacity of the fish skin and the scale were lower than that of the fin the maximums respectively being 0.99 Bq·g^{-1} and 0.71 Bq·g^{-1} after 2 days, then gradually reducing as the specific activity of ^{95}Nb in the water reduced. The structure of the head was more complicated, as it comprised bone, flesh, and other organs, as well as the epidermis and mouth organs, which were exposed outside the water surface. ^{95}Nb was mainly found in the external organs which had direct contact with the water, and they were relatively low compared with other parts of the specific activity, their dynamics were similar to the fish skin and scale.

In summary, the specific activities of ^{95}Nb in various parts of the fish descend in the following order: viscera > gill > fin > skin, fish scale > bone, head, eggs > flesh.

3.4. Transportation Model of ^{95}Nb in Simulated Aquatic Ecosystem

In this experiment, the aquatic ecosystem is composed of water, sediment, hyacinths, snails and fish. Due to there being a lack of exchange of water and ions between the system and outside, it could be regarded as a closed five-compartment system. According to the specific situation of this experiment, a model was established by ignoring some minor processes in **Figure 1** [17]-[20].

k_{12}, k_{13}, k_{14}, k_{15} are the transfer rate constants from water to sediment, fish, snails, and hyacinths, respectively. k_{21}, k_{24} are the transfer rate constants from sediment to water and snails, respectively. k_{31} is the transfer rate constant from fish to water. k_{42} is the transfer rate constant from snails to sediment. k_{51} is transfer rate constant from hyacinths to water. The rates of change of quantity (q_1, q_2, q_3, q_4, q_5) of ^{95}Nb in different compartments with time are as follows:

$$\frac{\mathrm{d}q_1}{\mathrm{d}t} = -\left(k_{12}+k_{13}+k_{14}+k_{15}\right)q_1 + k_{21}q_2 + k_{31}q_3 + k_{51}q_5$$

$$\frac{\mathrm{d}q_2}{\mathrm{d}t} = k_{12}q_1 + k_{42}q_4 - \left(k_{21}+k_{24}\right)q_2$$

$$\frac{\mathrm{d}q_3}{\mathrm{d}t} = k_{13}q_1 - k_{31}q_3$$

$$\frac{\mathrm{d}q_4}{\mathrm{d}t} = k_{14}q_1 + k_{24}q_2 - k_{42}q_4$$

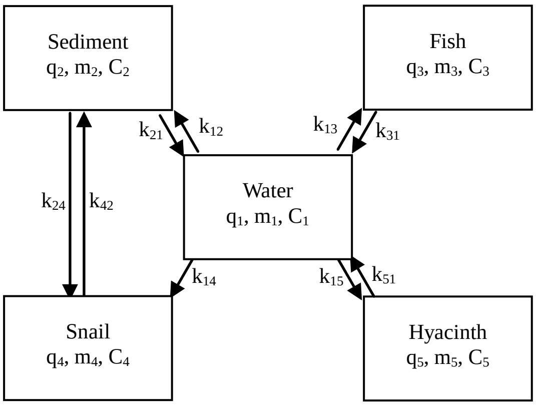

Figure 1. The closed five-compartment model of aquatic ecology.

$$\frac{\mathrm{d}q_5}{\mathrm{d}t} = k_{15}q_1 - k_{51}q_5$$

According to the experimental data and initial conditions, the differential equations were solved by fitting a code and mathematic modes for the dynamic behavior of ^{95}Nb in water (C_1), sediment (C_2), fish (C_3), snails (C_4), hyacinths (C_5) of the aquatic ecosystem are obtained as follows:

$$C_1 = 65.3447e^{-2.2147t} + 0.3285e^{-0.6690t} + 0.1119e^{-0.0129t} + 0.1099e^{-0.1848t} + 0.0006e^{-0.3378t}$$

$$C_2 = 85.7971e^{-2.2147t} + 1.6024e^{-0.6690t} + 91.8122e^{-0.0129t} + 2.9428e^{-0.1848t} + 0.3845e^{-0.3378t}$$

$$C_3 = 118995.7705e^{-2.2147t} + 119516.3543e^{-0.6690t} + 682.5725e^{-0.0129t} + 656.1561e^{-0.1848t} + 5.3418e^{-0.3378t} \Big/ m_{3(t)}$$

$$C_4 = 10198.6263e^{-2.2147t} + 304.2514e^{-0.6690t} + 679.3532e^{-0.0129t} + 155.6352e^{-0.1848t} + 9201.0370e^{-0.3378t} \Big/ m_{4(t)}$$

$$C_5 = 213794.5841e^{-2.2147t} + 4717.3065e^{-0.6690t} + 6210.0565e^{-0.0129t} + 212256.7591e^{-0.1848t} + 23.4203e^{-0.3378t} \Big/ m_{5(t)}$$

The correlation coefficient for water is $r = 0.92$, for sediment $r = 0.91$, for fish $r = 0.69$, for snail $r = 0.97$, for water hyacinth $r = 0.96$. Where $m_{x(t)}$ is the mass at moment t. Transfer coefficients are shown below with the unit·d^{-1}:

$$k_{12} = 0.5765,\ k_{13} = 0.0506,\ k_{14} = 0.0024,\ k_{15} = 0.0246,\ k_{21} = 0.0195,$$
$$k_{31} = 0.0152,\ k_{51} = 0.0169,\ k_{24} = 0.0001,\ k_{42} = 0.3241.$$

3.5. *CF* Value of ^{95}Nb by Aquatic Organisms

Aquatic organisms for radionuclide concentration factor in water is defined as the specific activity of the radionuclide in a certain component of aquatic ecosystem divided by the specific activity of the same radionuclide in the same time and space, and is usually called the concentration-factor (*CF*). The CF values of ^{95}Nb by aquatic organisms are shown in **Table 5**.

The ^{95}Nb enriching abilities of water hyacinth in water are the highest, followed by snail, and then fish. Their CF value increased gradually over time. At the end of the experiment (34 days), the *CF* value of ^{95}Nb for water hyacinth and snail reached 396.57 and 15.83. The CF values of ^{95}Nb for the main part of the fish (fish flesh) remained at almost zero, which shows the ^{95}Nb in water is not enriched by fish. Niobium is not an essential element for use by organisms, it is of universal insolubility and has a very low organism absorption, as show the date for ^{95}Nb.

Table 5. *CF* value of ^{95}Nb by aquatic organisms as a function of time.

Time (d)	*CF*		
	Hyacinth	Snail	Whole fish
1	12.67	1.51	0.35
2	18.09	1.43	0.94
4	25.37	1.19	0.41
6	36.69	1.24	0.06
9	31.22	0.89	0.32
15	41.75	1.67	0.33
22	165.59	6.53	0.34
29	295.26	11.90	0.45
34	396.57	15.83	0.30

4. Conclusions

When ^{95}Nb was introduced into an aquatic system, it was transported and transformed via deposition and absorption, and was adsorbed by aquatic organisms, leading to the distribution and accumulation in individual parts of the system. Over time, ^{95}Nb was desorbed and released, leading to ^{95}Nb redistribution in individual parts of the aquatic ecosystem. After 1 day, the specific activity of ^{95}Nb in water was 27.11% of the initial specific activity.

The ^{95}Nb enriching abilities of the various organisms in the aquatic ecosystem are very different. The ^{95}Nb enriching ability of hyacinth is far greater than for snail and fish, and the ^{95}Nb enriching ability of snail was greater than for fish. The ^{95}Nb in water by fish is not enrichment.

The ^{95}Nb enriching abilities of the different parts of the same organism are very different.

Dynamics of ^{95}Nb concentrations in different components in the aquatic ecosystem can be described by using a tracer kinetic compartment model. Under these study conditions, quantitative expressions reach notable levels.

Acknowledgements

The project was financially supported by the National Natural Science Foundation of China under the approval number 21247003. The critical reviewing of manuscript and comments by anonymous reviewers are greatly acknowledged.

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