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Transference kinetics of ^{60}Co in an aquatic–terrestrial ecosystem

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Abstract The dynamics of transportation, accumulation, disappearance and distribution of ^{60}Co in a simulated aquatic–terrestrial ecosystem was studied by isotope-tracer technique. In the aquatic system, ^{60}Co was transported and transformed *via* depositing, coupling with ions and adsorption. The absorption resulted in the redistribution and accumulation of ^{60}Co in each compartment of the system. Specific activities of ^{60}Co in water started sharply and gently decreased. The sediment accumulated a large amount of ^{60}Co by adsorption and ion exchange. The hornwort (*Ceralophyllum demersum*) could also adsorb a large amount of ^{60}Co in a short time, because of its large specific surface area. Fish (*Carassius auratus*) and snail (*Bellamya purificata*) had a poor capacity of adsorbing ^{60}Co. The distribution of ^{60}Co in the fish was mainly in the viscera, and the amount of ^{60}Co in the snail flesh was greater than that in the shell. The amount of ^{60}Co in individual compartment in the system was changed with time. The highest specific activity of ^{60}Co in the bean of the terrestrial system remained in the root nodule.

Key words ^{60}Co, Aquatic-terrestrial ecosystem, Compartment model, Transference kinetics

CLC numbers X131.3, X591

1 Introduction

As a necessary trace element, Co behaviors in animals and plants have been well studied. The studies on the adsorption, distribution and accumulation of ^{60}Co in ecosystems are usually focused on identification and description of static behavior. However, there are few reports about quantification and dynamic description of ^{60}Co[1-5]. In this paper, we study the ^{60}Co adsorption, transportation, accumulation and distribution in aquatic-terrestrial ecosystems from dynamic point of view with a compartment model to formulate the ^{60}Co behavior.

2 Materials and methods

2.1 Materials

^{60}Co was supplied by the Academy of Atomic Energy of China. Its specific radioactivity was 3.19×10^7 $Bq\cdot mg^{-1}$ (October 29, 1996), with radiochemical purity of >95%. Prior to use, it was transformed into $^{60}CoCl_2$[6].

Soil samples were collected from an experimental farm in the Huajiachi campus of Zhejiang University. The soil was sieved to remove stones and plant debris. The physico-chemical properties of the soil were described in Ref.[7].

2.2 Methods

An ecological pool of 1m×1m×0.6m was devided into two equal parts by a perforated plastic board. In part 1, as an aquatic ecosystem, the pool was filled with 125 L of water, 50 kg of sediment and aquatic organisms, including hornwort (*Ceralophyllum demersum*), fish (*Carassius auratus*), snail (*Bellamya*

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purificata) etc. In part 2, as a terrestrial ecosystem, the pool was filled with 290 kg of soil on which 15 plants of Glycinemaxes were grown. There was a single introduction of 20.0 mL ^{60}Co-CoCl$_2$ (specific activity 5.53×10^5 Bq·mL^{-1}) into the aquatic ecosystem, with 85.4 Bq·g^{-1} of the initial specific activity.

2.3 Sampling

Samples were collected after 0.25, 1, 3, 7, 15, 22, 29, 37 and 45 days. From the pool, 20 mL water samples were collected at random with disposable plastic cups (*Φ*75mm×110mm) for activity measurements. Two *Carassius auratus*, four *Bellamya purificatas* and a sufficient amount of *Ceralophyllum demersum* were sampled. Skeletal tissues, scales, flesh, gills and viscera were collected and weighed. The fish samples (20 g each) were measured in the *Φ*75mm× 110mm plastic cups.

Two sediment columns were collected from each pool part. They were sectioned into two equal parts, which were smashed and mixed thoroughly. Sediment samples (20 g), in replications of three, were measured in the plastic cups.

Once the experiment for the aquatic ecosystem was completed, the adjacent terrestrial ecosystem was sampled to determine the ^{60}Co specific activity in every part of Glycinemax and the soil[8].

2.4 Measurements

^{60}Co γ rays were measured with a multi-channel γ spectrometer (BH 1224, Beijing Nuclear Instrumentation Factory), with a *Φ*70 mm NaI scintillation detector and a positioning device to fix the sample cup. All the samples were measured on the sampling day. The counting statistics was better than 5%, and the data were normalized with counting efficiency, dead time, disintegration, etc.

The experiments were done at Institute of Nuclear Agricultural Sciences, Zhejiang University.

3 Results and discussion

3.1 Distribution of ^{60}Co in the aquatic ecosystem

When ^{60}Co was introduced into the aquatic system in the form of ^{60}Co-CoCl$_2$, it was transported and transformed *via* deposit and adsorption, and adsorbed by aquatic organisms, leading to the distribution and accumulation in individual part of the system. Over the time, ^{60}Co was desorbed and released, leading to ^{60}Co redistribution in individual parts of the aquatic ecosystem.

3.2 ^{60}Co in the pool water and sediment

The ^{60}Co specific activities in the pool water and the sediment at different days were given in Table 1. The ^{60}Co specific activity in the pool water decreased rapidly on the first hours due to deposit, complexation and adsorption and absorption by sediment and aquatic living organism. In 0.25 d, the ^{60}Co specific activity in pool water reduced to only 7.1% of the initial specific activity (85.4 Bq·mL^{-1}). It decreased gradually, and seven days later, the exchanging of ^{60}Co in water with ions in the ecosystem reached dynamic equilibrium.

Table 1 Changes of ^{60}Co specific activity in water and sediment with time

Time / d	Water / Bq·g^{-1}	Sediment / Bq·g^{-1}
0.25	6.05	96.94
1	4.86	157.13
3	1.21	158.26
7	0.31	108.68
15	0.33	121.80
22	0.25	96.07
29	0.20	102.26
37	0.14	112.90
45	0.09	110.41

As shown in Table 1, due to the deposit of ^{60}Co in water and exchange with the ions in soil, ^{60}Co specific activity in sediment increased rapidly in the first three days to a maximum of 158.26 Bq·g^{-1} on Day 3, and decreased to an average of 110.56 Bq·g^{-1} in the following days. This is because that the ^{60}Co was mainly centralized within the surface sediment and the Co ions that were not combined closely with sediment began desorbing. As time went on, ^{60}Co moved into and combined stably with the deep sediment through the processes of complexation and the adsorption of iron and manganese oxides[8,9].

3.3 ^{60}Co in aquatic organisms

The ^{60}Co specific activity in aquatic organisms on different days was listed in Table 2. On Day 1, the ^{60}Co specific activity increased rapidly in snail. Due to the rapid decrease of ^{60}Co specific activity in water, ^{60}Co in the snail began desorbing and the specific activity was essentially at equilibrium 15 days later. In addition, ^{60}Co specific activity in flesh and shell was determined in the last sample and found to be 23.51 $Bq \cdot g^{-1}$ and 9.93 $Bq \cdot g^{-1}$ respectively; making the activity in the flesh greater than that in shell.

Table 2 ^{60}Co specific activity (in $Bq \cdot g^{-1}$) in aquatic organisms

Time / d	Snail		Hornwort		Fish	
	Fresh weight / g	Specific activity	Fresh weight / g	Specific activity	Fresh weight / g	Specific activity
0	100	0	110	0	500	0
0.25	100	86.3	110	1862.3	500	177.83
1	92	125.1	97	2235.8	460	28.72
3	85	41.7	87	2142.8	425	36.86
7	78	31.9	80	756.6	348	14.98
15	72	17.5	90	438.2	341	8.35
22	66	14.8	95	286.3	306	12.36
29	61	11.7	108	136.9	270	3.99
37	58	13.5	120	74.2	248	4.73
45	55	17.1	130	76.6	218	4.05

The ^{60}Co specific activity in hornwort, which has high ability of absorbing and adsorbing ^{60}Co, increased to 1862 $Bq \cdot g^{-1}$ in 6 hours and to 2235.8 $Bq \cdot g^{-1}$ on Day 1, being much higher than those in snails and fish. This is mainly because hornwort has much larger specific surface to absorb ^{60}Co complex sediment and ^{60}Co-adsorbed suspended particles.

The ^{60}Co specific activities in different parts of the fish in the last sample were given in Table 3. The ^{60}Co specific activity in viscera, 19.81 $Bq \cdot g^{-1}$, is the highest. This is due to the high metabolic activity in the fish organs. The second highest ^{60}Co specific activity is seen in the scales, which exposed directly to the radioactive water, and the gills, through which large amounts of water pass for respiration. The ^{60}Co specific activity in flesh and skeletal tissues are the lowest because ^{60}Co in these two parts are acquired through other organs. In summary, the ^{60}Co specific activities in various parts of the fish descend in the following order: viscera > scales > gills> skeletal tissues > flesh and the total activities descend as: viscera > skeletal tissues > scales > gills > flesh.

Table 3 ^{60}Co specific activity (in $Bq \cdot g^{-1}$) in different parts of fish

Parts of fish	Fresh weight / g	Specific activity	Total activity	Percent
Skeletal tissues	24.52	1.95	47.87	24.39
Scales	2.34	6.20	14.52	7.40
Flesh	12.95	0.92	11.96	6.09
Gills	3.13	4.11	12.85	6.55
Viscera	5.51	19.81	109.09	55.58

3.4 Transportation model of ^{60}Co in simulated aquatic ecosystem

In this experiment, the aquatic ecosystem is composed of water, fish, snails and hornworts. It could be regarded as an open five-compartment system owing to the exchange of water ions between the aquatic ecosystem and terrestrial soil. According to the specific situation of this experiment, the model was established by ignoring some minor processes in Fig.1. The rates of change of ^{60}Co quantity (q_i) in different compartments with time are as follows[10-15]:

$$\frac{dq_1}{dt} = -(k_1 + k_{12} + k_{13} + k_{14} + k_{15}) + k_{21}q_2 + k_{31}q_3 + k_{52}q_5$$

$$\frac{dq_2}{dt} = k_{12}q_1 - (k_{21} + k_{24})q_2 + k_{42}q_4$$

$$\frac{dq_3}{dt} = k_{13}q_1 - k_{31}q_3$$

$$\frac{dq_4}{dt} = k_{14}q_1 + k_{24}q_2 - k_{42}q_4$$

$$\frac{dq_5}{dt} = k_{15}q_1 - k_{51}q_5$$

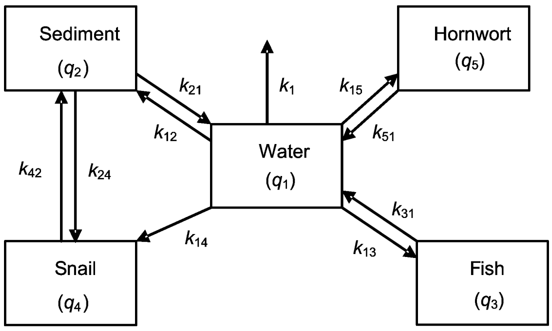

Fig.1 The opened five compartment model of aquatic ecology.

The k_{ij} is transfer rate constant between every two compartments. According to the experimental data and initial conditions, the differential equations were solved by a fitting code and mathematic modes for the dynamic behavior of ^{60}Co in different compartments of the aquatic ecosystem are obtained as follows:

for water,

$C_1=84.20e^{-2.9736t}+0.41e^{-0.803t}+0.16e^{-0.0157t}+0.14e^{-0.1902t}+0.00074e^{-0.4155t}$

r=0.92

for sediment,

$C_2=-111.90e^{-2.9736t}-2.05e^{-0.803t}+117.68e^{-0.0157t}-3.20e^{-0.1902t}+0.49e^{-0.4155t}$

r=0.91

for fish,

$C_3=(-153691e^{-2.9736t}+151827e^{-0.803t}+882e^{-0.0157t}+852e^{-0.1902t}+6.8e^{-0.4155t})/m_{3(t)}$

r=0.69

for snail,

$C_4=(-13105e^{-2.9736t}-391e^{-0.803t}+883e^{-0.0157t}+199e^{-0.1902t}+11796e^{-0.4155t})/m_{4(t)}$

r=0.97

for hornwort

$C_5=(-274022e^{-2.9736t}-6086e^{-0.803t}+7943e^{-0.0157t}+271873e^{-0.1902t}-30e^{-0.4155t})/m_{5(t)}$

r=0.96

where $m_{x(t)}$ is the mass at moment t. Transfer coefficients are shown below with the unit d^{-1}:

k_1=2.1419, k_{12}=0.714, k_{13}=0.0657, k_{14}=0.0028, k_{15}=0.0287, k_{21}=0.021, k_{31}=0.01947, k_{51}=0.021, k_{24}=0.0001, k_{42}=0.4154.

3.5 CF value of ^{60}Co by aquatic organisms

The concentration-factor (CF) values of ^{60}Co by aquatic organisms are shown in Table 4. The ^{60}Co specific activity in aquatic organisms is higher than that in water, and the CF value of ^{60}Co in hornwort was higher than those in fish and snails. The ^{60}Co enriching abilities of the various compartments in the aquatic ecosystem descend as follows: hornwort > snail > fish.

Table 4 CF value of ^{60}Co by aquatic organisms as a function of time

Time / d	0.25	1	3	7	15	22	29	37	45
Hornwort	306	455	1755	2435	1326	1144	685	530	851
Fish	29.39	5.910	30.45	48.33	25.31	49.44	19.93	33.81	88.95
Snail	14.26	25.74	34.44	102.8	53.09	59.17	58.64	96.66	190.3

3.6 ^{60}Co in terrestrial ecosystem

At the end of the experiment (45 d), the average ^{60}Co specific activity in terrestrial soil was 2.8 $Bq \cdot g^{-1}$ and total activity was 812358 Bq, which amount to 7.6% of initial total activity. Compared with the aquatic ecosystem, ^{60}Co specific activity in terrestrial soil was lower than every compartment except water in the last sample.

In the terrestrial ecosystem, the distribution of ^{60}Co in Glycinemax is higher in the root tubercles than in any other part of the plant. This is related to the physiological function of Co in Glycinemax. It is a necessary component of cobamide coenzyme and vitamin B_{12}, and it takes part in nitrogen fixation and the formation of proteins. The total activity of ^{60}Co in Glycinemax is less than 1% of initial total activity.

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