Silver-110 m in marine organisms, soil and vegetables of Hong Kong

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Abstract. ^{110m}Ag is a fission product inside nuclear reactors and can be released into the environment during accidents. In this study, accumulation and transfer of ^{110m}Ag in simulated marine and terrestrial ecosystems were studied. The kinetic behavior of ^{110m}Ag in the system was summarized. It is noted that most of the ^{110m}Ag accumulated in the sediment and seashore mollusks can absorb more ^{110m}Ag than fish. Accumulation and transfer of ^{110m}Ag in terrestrial ecosystems were studied in pot culture approach where vegetables such as lettuces, spinach, kale, cucumbers, peppers, carrots, egg plants and beans were cultured in soil that contains ^{110m}Ag in 10 Bq g_{soil}^{-1}. Edible vegetable samples were collected and measured at different growth stages. The transfer factors were calculated. The results showed that ^{110m}Ag could be transferred from soil into plants, and spinach may be a critical vegetable. The leaching and migration of ^{110m}Ag in soils were also studied by soil column experiments. Results show that there is no ^{110m}Ag in leaching water and most of it stays in the topsoil of 2 cm deep, and ^{110m}Ag vertical migration is little. A computer model based on ECOSYS-87 to simulate the transport of ^{110m}Ag in the different compartments along the routes will be developed.

1. INTRODUCTION

Silver-110m (β: 0.08MeV, 98.6%; γ: 0.658MeV, 94%; half-life 250 days), as a radionuclide of corrosion product in water-cooled nuclear reactors(PWRs), may be released into the environment and potentially contaminate foodstuffs under both normal operation and accidents(Hirschberg G. et al, 1999; Song H, et al, 1999; H.Van Dam, 1986). The absorption of the radionuclide into marine organisms via seawater and into plants from soil via roots is the critical pathways for dose contribution to human beings. This absorption is affected by various parameters, such as the chemical and physical properties of soil, temperature, sunlight etc. This research is a primary step of a systematic study of biological transport pathways and radiological hazard assessments of ^{110m}Ag.

2. MATERIALS AND METHOD

2.1 Materials

Marine organisms: The fish and mollusks were captured in the HK island seashore, and the sediments were collected from local area. Artificial seawater was made by natural sea salt and water. The normal aquarium could be controlled by artificial condition.

Soil and vegetables: Sandy-loam soil, which is a local representative culture soil, was collected from Hong Kong Tai Lung Experimental Station.(see Tab. 1) The vegetables selected were lettuce, spinach, kale, carrot, peppers, eggplant, beans, and cucumber since they are representative plants in the local vegetable gardens.

Tab.1 Physical and chemical properties of culture soil

Particle-size Distribution(%)			pH	Organic Matter(%)	Total N (%)	Total P (mg/kg)	Total K (mg/kg)	Cation Exchange Capacity (cmol/kg)
Sand	Silt	Clay						
67.50	29.00	3.50	7.47	0.62	0.42	545.47	3473.07	5.17

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2.2 Experimental Design

2.2.1 Marine ecosystem

The marine ecosystem consists of an aquarium containing artificial seawater 60L, sediment 10kg, fish, seashore mollusks and algae, etc. Total 280KBq ^{110m}Ag were added into the aquarium, in which, specific activity of ^{110m}Ag was about 100 Bq/L in water and 20Bq/g in sediment at equilibrium. Samples were collected every 24 to 48 hours up to 705 hours (30d), and then were measured after ashed in a microwave furnace.

2.2.2 Soil column test

Soil in different pH value of 4.24, 4.36, 5.82, 7.06, 7.47, 7.87, 8.15, 8.25 and 8.35 columns (30cm high, 6cm diameter) were made for doing experiments of ^{110m}Ag leaching and vertical migration in soil. 800g of soil was air dried, sieved through 20 mesh and filled into each column uniformly. After four days of leaching with distilled water, 8kBq ^{110m}Ag in 10mL $^{110m}AgNO_3$ solution was added into each of the columns all at once. For the leaching column experiment, distilled water was added continuously and leaching water was sampled in regular intervals. This process was continued for 30 days and then the soil was air dried for one week and sampled in 0.5cm segment for measurement. For the vertical migration column experiment, since no additional water was added, the column was just laid aside for 30 days, then sampled in 0.5cm segment for measurement. Its pH value of the vertical migration test soil is 7.06.

2.2.3 Pot culture test

Pot culture was done in the outdoor controlled area during April, May and June. The temperature was 26~32°C, the relative humidity was 80~95%. The natural culture soil, after crushed, was sieved through 20mesh, then mixed evenly with ^{110m}Ag in 10Bq/g Specific Activity (SA) and placed into plastic pots. The lettuce, spinach, kale, carrot, peppers, eggplant, beans, cucumber were sown and grown in the pot under sprinkling irrigation in outdoors with a shelter. Subsequently, the total edible and inedible parts were sampled separately at different growth stages. The samples were washed under running water, dried naturally and ashed in a microwave furnace (CEM MAS 7000) for measurement.

2.3 Analytical method

For vegetation, soil, marine organisms and liquid samples, measurements were carried out by High Purity Germanium Gamma Spectrometer (Canberra GX6020), with polynomial efficiency of 4.387% for liquid sample and 3.288% for solid sample and FWHM of 2.31keV at 657.75keV channel. The detector was connected to a multichannel analyzer (EG&G ORTEC Digital Gamma Ray Spectrometer) with 4096 channels.

2.4 Calculation of transfer factors

The soil-to-plant transfer factor (TF) is frequently used to describe the transport between the soil and the plant components. It may be defined as the ratio between plant (fresh weight) and soil (dry weight) activities.

$$TF = \frac{^{110m}\text{Ag concentration in vegetables}}{^{110m}\text{Ag concentration in soil}}$$

3. RESULTS AND DISCUSSION

3.1 Simulated marine ecosystem

Based on the measurement, we assume that glass of the aquarium adsorbed 20% of ^{110m}Ag and 80% accumulated in the sediment and dissolved in water. Similar result was concluded in another report (Huang, Z, 1994). Because the seashore mollusks live on the sediment, they absorb and accumulate more radioactivity than other organisms. Figure 1 shows the accumulation of ^{110m}Ag in organisms with time, which shows that fish could absorb little ^{110m}Ag. The interesting fact is that two inedible mollusks in the experiment, *Clibanarius Infraspinatus* and *Chlorostoma Rustica*, absorb more activity than other edible mollusks. However, the data obtained in the experiment is somewhat indefinite, since the number of sample is insufficient.

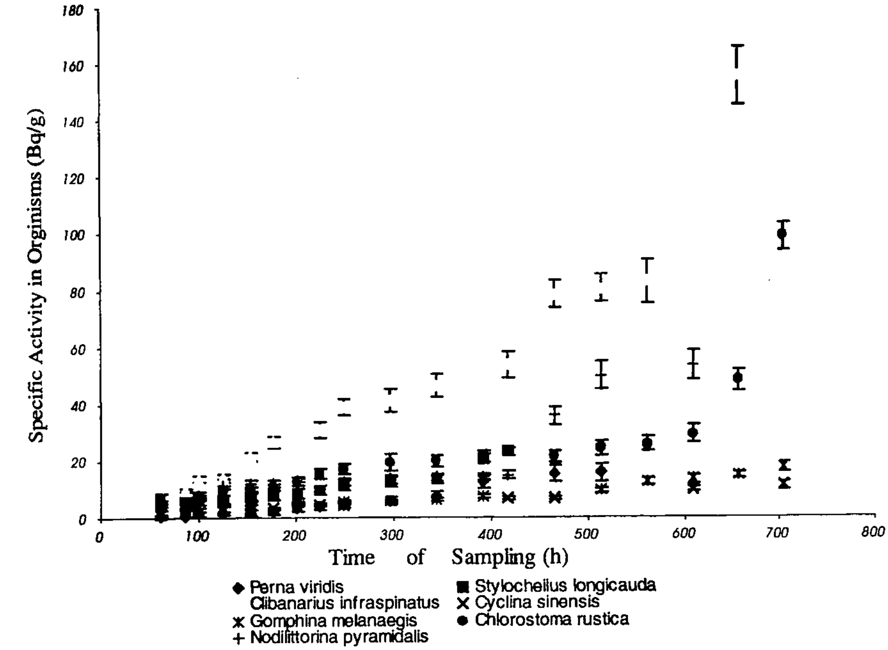

Figure 1 ^{110m}Ag absorption in some marine organisms

3.2 ^{110m}Ag leaching in soil column

The result shows that almost more than 90% of the radionuclide added in the column was retained in the top 1 cm of soil regardless the pH value, and leaching ability of ^{110m}Ag decrease with the pH value increase. The leaching water was also measured, and the results show that there is no ^{110m}Ag within it. That means ^{110m}Ag could be moved down with leaching water but at most up to 1 or 2cm. The result also shows that there is no ^{110m}Ag beyond 6cm. This can be explained by noting that the soil in the column is a cultured soil used for many years to which ploughing, weeding and fertilizer were applied many times, thus has induced plentiful organic matter in the soil. Former studies also showed that most radionuclides were accumulated in the soil layers that have plentiful organic matters and sticky soil particles (Whicker F.W. and Vincent S. 1982; Zhang Z. 1993).

Tab.2 ^{110m}Ag Leaching in Soil Column

Soil layer (cm)	pH4.24	pH4.36	pH5.82	pH7.06	pH7.47	pH7.87	pH8.15	pH8.25	pH8.35
0-0~5	252.28	234.47	377.11	270.13	345.99	328.11	444.86	389.82	375.47
0.5~1	82.19	134.9	96.06	136.53	139.81	120.26	28.85	40.08	37.74
1-1~5	55.42	4.76	3.55	15.27	9.44	25.16	1.3	1.11	0.15
1.5~2	15.48	6.99	1.79	6.13	1.91	0.98	0.14		
2-2~5	6.49	2.96	1.2	5.61					
2.5~3	3.36	1.32	0.97	1.83					
3~4	3.12	1.43	0.36						
4~5	2.1								
5~6	0.23								

3.3 ^{110m}Ag migration in soil column

The result of ^{110m}Ag vertical migration test is reported in Figure 2, which show that ^{110m}Ag stay in 1.5cm depth if no addition water added, and little ^{110m}Ag could migrate into lower and deep soil layers. No ^{110m}Ag was found in the soil samples from 2cm to 24cm column layers. Similar result was observed in a field investigation (Rauret G. et al, 1995). Compared with the leaching test, it seems that ^{110m}Ag itself has no migration in the soil, which means that natural rainfall could push ^{110m}Ag from top into lower soil layers, and subsequently let the process of ^{110m}Ag absorption by plant from soil lasts longer. The former result shows that deep plough has dilution effect for radionuclides in deep soil layers (Sulbu B et al, 1994). In an accident, if ^{110m}Ag had contaminated the topsoil, it would be a feasible approach to deep plough the soil so as to lessen dose contribution from ^{110m}Ag in the topsoil.

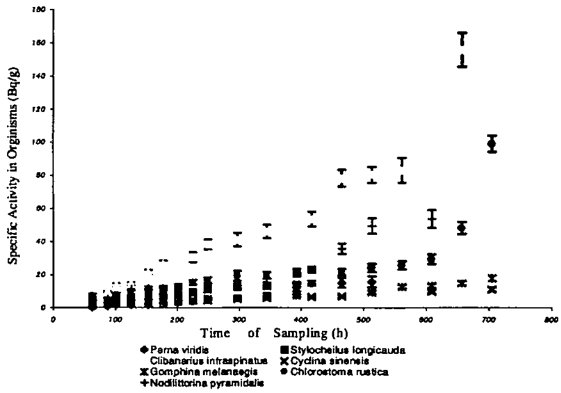

Figure 2 ^{110m}Ag Migration in Soil Column

3.4 Transfer of ^{110m}Ag from soil into vegetables

The result in Table 3 shows that ^{110m}Ag could be transferred from soil into plants via the root absorption. For leafy vegetable, SA and TF of lettuce and spinach are increased by one order of magnitude with growth stage. This can be explained by the increase in root uptake over time, since roots penetrate deeper into the soil where the radionuclide distributed uniformly. As former discussion, ^{110m}Ag leaching from topsoil into lower several centimeters can lead to an increase in transfer values due to the increase of biomass. Compared with leaf and root for the leafy vegetables, since ^{110m}Ag was absorbed via the root, generally, TF of root is higher than leaf. However, the data of root is not so important, since the root is not only inedible, but is not easy to be rinsed off the soil particles using running water after sampling.

Carrot is a root vegetable, which has the biggest TF in the experiment. Therefore, carrot may be as a biomonitor for the radioisotope. For fruit plants, the trend is not clear since the sample number is too few to be analyzed accurately. Further experiments will be done in the near future.

Leafy vegetable is the most important for Hong Kong food ingredient. From the results obtained, spinach behaved as a critical plant among these vegetables since, as a popular leafy vegetable, it could absorb more ^{110m}Ag than others.

Tab. 3 Absorption of ^{110m}Ag from Soil into some vegetables

Vegetables	Growth Stage (d)	Different Organs	Fresh Weight (g)	Ash Weight (g)	Specific Activity (Bq/g fresh sample)	TF
Lettuce	55	Root Leaf	3.65 76.50	0.16 0.83	1.2±0.3 0.003±0.002	0.117 0.0003
	61	Root Leaf	3.60 78.88	0.10 1.02	0.7±0.3 0.005±0.003	0.072 0.001
	68	Root Leaf	6.33 102.09	0.38 1.34	1.2±0.3 0.04±0.02	0.122 0.004
	75	Root Leaf	9.03 71.74	0.54 1.08	4.1±1.4 0.11±0.09	0.405 0.011
	82	Root Leaf	27.60 138.49	1.76 3.87	1.1±0.3 0.5±0.3	0.113 0.048
Spinach	31	Root Leaf	0.52 3.71	0.02 0.14	4.6±2.5 0.07±0.03	0.458 0.007
	38	Root Leaf	0.51 3.45	0.02 0.11	4.9±0.2 0.12±0.06	0.490 0.012
	45	Root Leaf	0.47 1.93	0.03 0.13	6.2±2.9 1.8±0.6	0.619 0.175
	52	Root Leaf	1.72 5.76	0.17 0.39	5.1±1.8 2.1±1.0	0.514 0.212
Kale	52	Root Leaf	0.39 3.91	0.02 0.17	6.0±3.6 0.8±0.5	0.597 0.083
	59	Root Leaf	1.99 20.77	0.09 0.69	1.9±0.9 0.5±0.3	0.189 0.048
	66	Root Leaf	1.02 13.04	0.15 0.45	8.3±2.7 0.48±0.14	0.835 0.048
Carrot	70	Root	10.13	1.79	3.7±1.7	0.369
	77	Root	7.61	2.05	9.1±2.8	0.908
	85	Root	22.55	0.68	4.1±0.8	0.412
Peppers	30	Fruit	4.52	1.16	2.1±1.2	0.209
	36	Fruit	9.52	0.24	0.38±0.21	0.038
	41	Fruit	9.20	0.11	0.87±0.52	0.087
	44	Fruit	15.60	0.36	0.21±0.13	0.021
Egg Plant	85	Fruit	51.95	1.06	0.51±0.11	0.051
Beans	50	Fruit	18.19	0.56	0.35±0.24	0.035
	57	Fruit	10.08	0.13	0.2±0.11	0.020
Cucumber	90	Fruit	14.84	0.80	1.3±0.5	0.135
	97	Fruit	14.56	0.29	0.48±0.16	0.048
	104	Fruit	15.06	0.22	0.33±0.22	0.033

4. CONCLUSION

The conclusion from the experiments could be summarized as follows: most of the ^{110m}Ag in seawater accumulated in the sediment and seashore mollusks can absorb more ^{110m}Ag than other organisms; leafy and fruit vegetables could absorb ^{110m}Ag through root from contaminated soil; spinach may be a critical leafy vegetable and carrot may be a biomonitor for ^{110m}Ag; ^{110m}Ag can leach a couple of centimeter into the lower soil layers from topsoil, but the migration is little. These results suggest that the experimental design is suitable for the study of ^{110m}Ag behavior in marine, soil and soil-plant scenarios. However, the data obtained in marine organisms and vegetables is somewhat indefinite, since the number of sample is insufficient. These preliminary results have been used to design subsequent experiments in order to improve our knowledge of the radionuclide in terrestrial and aquatic ecosystem. Meanwhile, A computer model to simulate the transport of ^{110m}Ag in the different compartments along the routes is being developed.

5. REFERENCE

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