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Original Article

Radioactive iodine analysis in environmental samples around nuclear facilities and sewage treatment plants

Ukjae Lee, Min Ji Kim, Hee Reyoung Kim*

Ulsan National Institute of Science and Technology, Ulsan, 689-798, Republic of Korea

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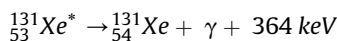
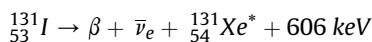
ABSTRACT

Many radionuclides exist in normal environment and artificial radionuclides also can be detected. The radionuclides (^{131}I) are widely used for labeling compounds and radiation therapy. In Korea, the radionuclide (^{131}I) is produced at the Radioisotope Production Facility (RIPF) at the Korea Atomic Energy Research Institute in Daejeon. The residents around the RIPF assume that ^{131}I detected in environmental samples is produced from RIPF. To ensure the safety of the residents, the radioactive concentration of ^{131}I near the RIPF was investigated by monitoring environmental samples along the Gap River. The selected geographical places are near the nuclear installation, another possible location for ^{131}I detection, and downstream of the Gap River. The first selected places are the “front gate of KAERI”, and the “Donghwa bridge”. The second selected place is the sewage treatment plant. Therefore, the Wonchon bridge is selected for the upstream of the plant and the sewage treatment plant is selected for the downstream of the plant. The last selected places are the downstream where the two paths converged, which is Yongshin bridge (in front of the cogeneration plant). In these places, environmental samples, including sediment, fish, surface water, and aquatic plants, were collected. In this study, the radioactive iodine (^{131}I) detection along the Gap River will be investigated.

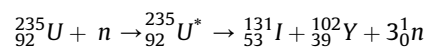
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1. Introduction

The radioactive iodine (^{131}I) has a short half-life of 8.05 days with beta minus and gamma emissions (Fig. 1). The transformation into stable ^{131}Xe is in two steps:



It is widely used as a general-purpose nuclide for labelling compounds, including for the purpose of clinical examination [1]. Furthermore, this radionuclide can be used in scintigraphy, metabolism, and the treatment of hyperthyroidism and thyroid cancer [2]. Radioactive iodine is also used as a tracer and is generated as a nuclear fission reaction product in nuclear reactors [3,4]:



An artificial nuclear species of ^{131}I has been detected in the general environment around hospitals, because of its use in the medical field. In addition, as the use of ^{131}I for medical diagnosis and treatment has increased, it is released into river systems through sewage treatment plants and contaminates the environment [5–7]. Radioactive iodine is highly volatile and mostly enters the body through respiration. It accumulates in the thyroid gland with generating beta and gamma rays. The gland cells that are affected by radiation die or mutate and develop cancer. Therefore, periodic monitoring of ^{131}I in the environment is essential to protect human health.

There are regulations for nuclear facilities in terms of environmental radiation sampling and environmental impact assessment [8]. Gamma isotopes, such as ^{131}I , are monitored by many ways in terms of the quality and safety of air, drinking water, groundwater, rainwater, surface water, sediments, agricultural products, and organisms. Details of the environmental radiation monitoring program around nuclear facilities are shown in Table 1, which is informed by the Korean radiation safety act. According to this act, if radioactivity concentration is detected by the operation of a nuclear

Abbreviations: KAERI, Korea Atomic Energy Research Institute; RIPF, Radioisotope Production Facility.

* Corresponding author.

E-mail address: kimhr@unist.ac.kr (H.R. Kim).

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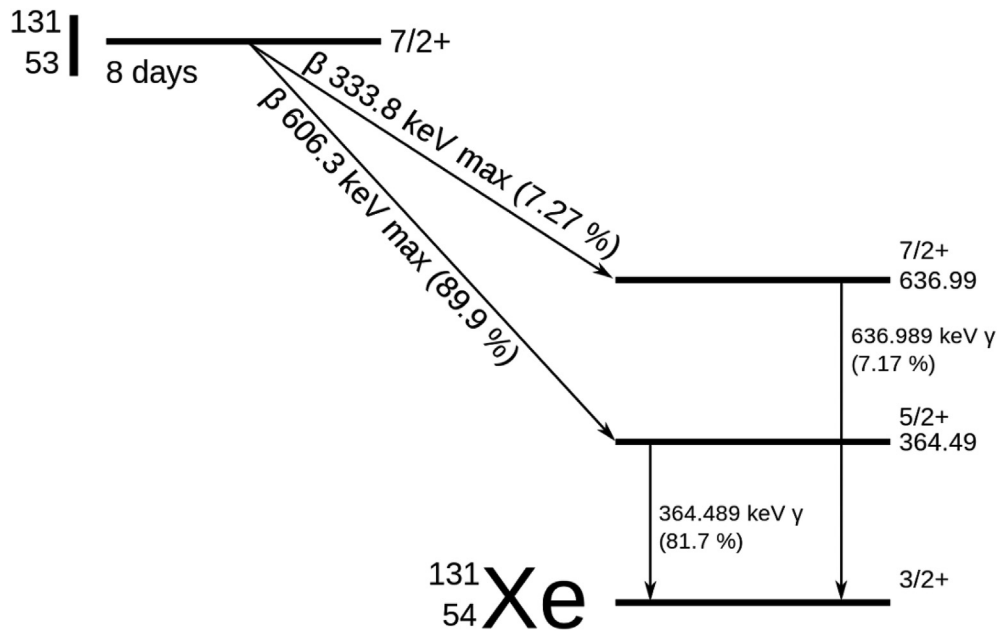


Fig. 1. Decay scheme for iodine.

facility, the concentration value shall be more than five times the range of fluctuation over a period of three years, or an artificial radionuclide such as ^{131}I should be reported to the Nuclear Safety and Security Commission [9].

The Korea Atomic Energy Research Institute (KAERI) in Daejeon (Fig. 2) houses the Radioisotope Production Facility (RIPF), which handles about 1000 Ci of ^{131}I per year [10]. To investigate the effect of the RIPF on the surrounding environment, samples were collected from sediments, fish, surface water, and aquatic plants in the nearby Gap River, because these constitute the common elements of the concerned ecosystem. The samples were analyzed to determine whether the radioactive iodine (^{131}I) detected in the general environment originated from the RIPF [11].

2. Materials and methods

2.1. Collection of environmental samples

Gapcheon is the largest river going through Daejeon. The main

purpose of this paper is to clarify the detection of ^{131}I in the environment. To prove this, the first selected geographical places are near the nuclear installation, which is the “front gate of KAERI” and “Donghwa bridge”. The second selected place is another possible place for ^{131}I detection, which is the sewage treatment plant. Therefore, the Wonchon bridge is selected for the upstream of the plant and the sewage treatment plant is selected for the downstream of the plant. The last selected place is the downstream location where the two paths converged, which is Yongshin bridge.

Sediment, surface water, and aquatic plant samples were collected from the KAERI's front gate, Donghwa bridge, the drain of the Daejeon sewage treatment plant and front of the cogeneration plant, and Yongshin bridge. These sampling points are in the Donghwawool river system, which flows into the Gap River from Deokjin-soryu. The comparative samples were collected from the Wonchon bridge upstream of the sewage treatment plant. The sampling points are shown in Fig. 2. Considering the distance and geographical factors in Gapcheon, the Gongju

Table 1
Monitoring items and cycles on radiation environment survey and radiation environmental impact assessment around nuclear facilities.

Category	Monitoring Item		Monitoring Cycle	
	Contents	Monitoring nuclide	Sampling frequency	Analysis frequency
Radiation	Environmental radiation	Spatial gamma dose rate Total integrated dose(TLD)	Continuous monitoring	Once a month Once a quarter
Ground sample	Air	Gross beta, ^{14}C , ^{131}I , U, Gamma isotopes	Continuous sampling	Once a month
	Moisture in air	^3H		2 times per month
	Drinking water/groundwater	^3H , U, Gamma isotopes	Once a quarter	Once a quarter
	Rain/surface water	Gross beta, ^3H , U, Gamma isotopes	Once a month	Once a month
	Superficial layer soil	^{90}Sr , Pu, U, Gamma isotopes	2 times per year	2 times per year
	Sediment	U, Gamma isotopes	Once a quarter	Once a quarter
	Agricultural product	^3H , ^{14}C , ^{90}Sr , U, Gamma isotopes	Harvest	2 times per year
	Meat	^{14}C , Gamma isotopes	2 times per year	2 times per year
	Milk	^3H , ^{90}Sr , ^{129}I , ^{131}I , Gamma isotopes	Once a month	Once a month
		^{14}C	Once a quarter	Once a quarter
Sea sample	Indicator organism	^{90}Sr , Gamma isotopes	2 times per year	2 times per year
	Sea water	Gross beta, ^3H	Once a week	Once a month
		^{90}Sr , Pu, Gamma isotopes		Once a quarter
	Sub-marine sediment	^{90}Sr , Pu, Gamma isotopes	2 times per year	2 times per year
	Fish and sells	^{90}Sr , Pu, Gamma isotopes	2 times per year	2 times per year
Seaweed	^{90}Sr , ^{99}Tc , ^{129}I , ^{131}I , Gamma isotopes	2 times per year	2 times per year	

Table 2

Calibration result of gamma spectrometer.

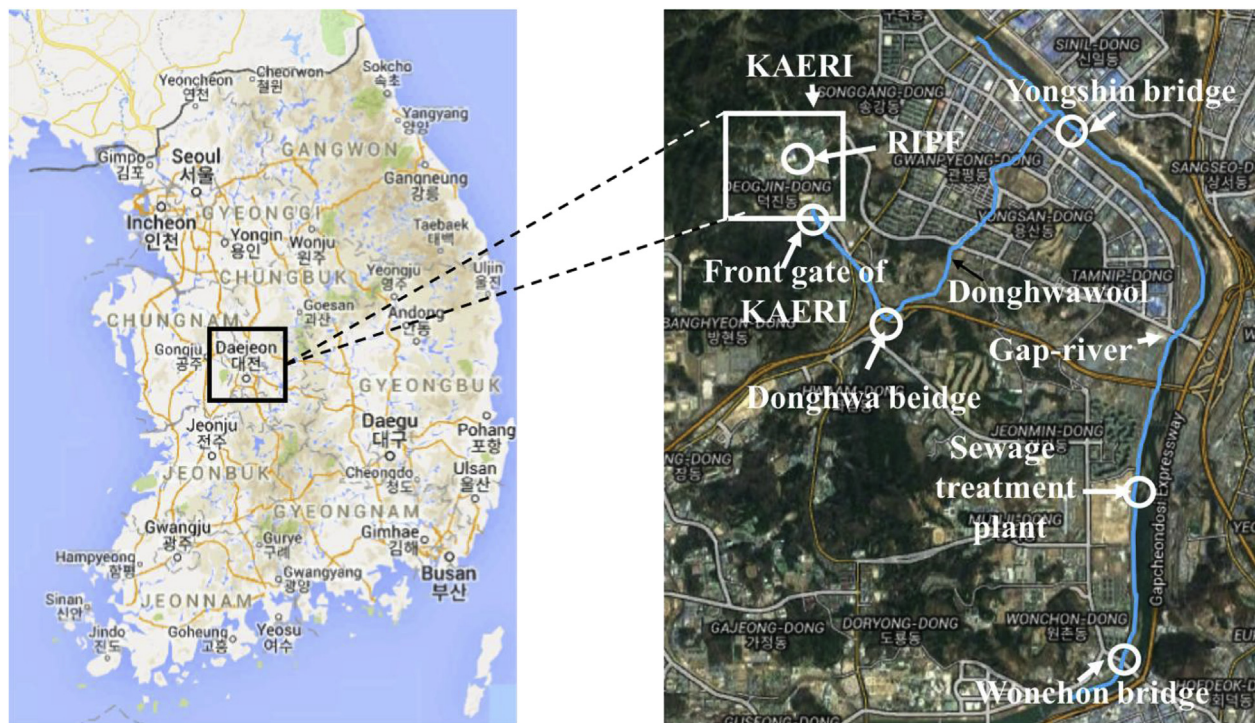
	Calibration Source (QCY48 Mixed Gamma Source)		Energy Calibration		Efficiency Calibration	
			KeV	Channel	KeV	Efficiency
			MCB#1 (EG&G Ortec MCA)	3.25	1L Marinelli Beaker	88 661 1332
	10.26	1L Marinelli Beaker	88 661 1332	446.9 3360.6 6768.2	88 661 1332	0.0457 0.0110 0.0060

MCB#1 System:

Detector Type: HPGe (GMX) -Relative Eff.: 25%.

Resolution: 1.81 keV at 1.33 MeV -Peak-to-Compton Ratio 60Co: 48.

Crystal Diameter: 53.6 mm -Crystal Length: 71.2 mm.

**Fig. 2.** Sampling points in the study area around Gab River in Daejeon, Korea.

reservoir (Fig. 3), which was not exposed to radioactive contamination, was selected as the control site in similar environmental conditions. Fig. 4 shows each monitoring point. Sediment samples were collected from more than 5 times per one sampling at a uniform azimuth and at about a 5 m radius from each sampling site. The depth of sampling was 5 cm, and more than 500 g samples were collected with plastic bag at each point. The sediment samples were placed in plastic bags to inhibit evaporation and transferred to the laboratory. Surface water samples were taken from the middle of the water flow at each sampling point, and samples of 20 L were collected.

Fish samples were caught by fishing at both the sewage treatment plant and in the Gongju reservoir. Two cornet fish (185 g–210 g), seven crucian carp (355 g–495 g), and two carp (1855 g–2355 g) were caught downstream of the sewage treatment plant (Fig. 5). Three crucian carp (255 g–395 g), one carp (1025 g), and two leather carp (720 g–975 g) were caught at Gongju reservoir (Fig. 5).

2.2. Pretreatment of samples

The equipment for the pretreatment process is shown in Fig. 6 [12]. In the laboratory, the sediment samples were weighed, after which they were placed in a stainless-steel tray and dried in a drying oven at 100 °C for at least 24 h. Subsequently, the dry weight was measured to calculate the percentage of water contained in the soil. The sample was pulverized using a 1 mm sieve (Fig. 6 (a) and (e)). To determine the fineness of the sample, almost gravel was crushed with a mortar, and the sample was partially sampled and placed into U-8 vial (Fig. 6(b)), which was used for gamma isotope analysis. The aquatic plants were thoroughly dried in a dryer and pulverized with a grinder [13]. The surface water samples were evaporated and concentrated to 1 L. The measuring vessel was a Marinelli beaker (Fig. 6(b)) [14].

Each fish was pulverized using a grinder (Fig. 6(c)). After weighing the samples, foreign substances were removed by washing with distilled water the samples three times. Thereafter,



Fig. 3. Distance between Gongju reservoir and the radio-isotope production facility in the sampling area.

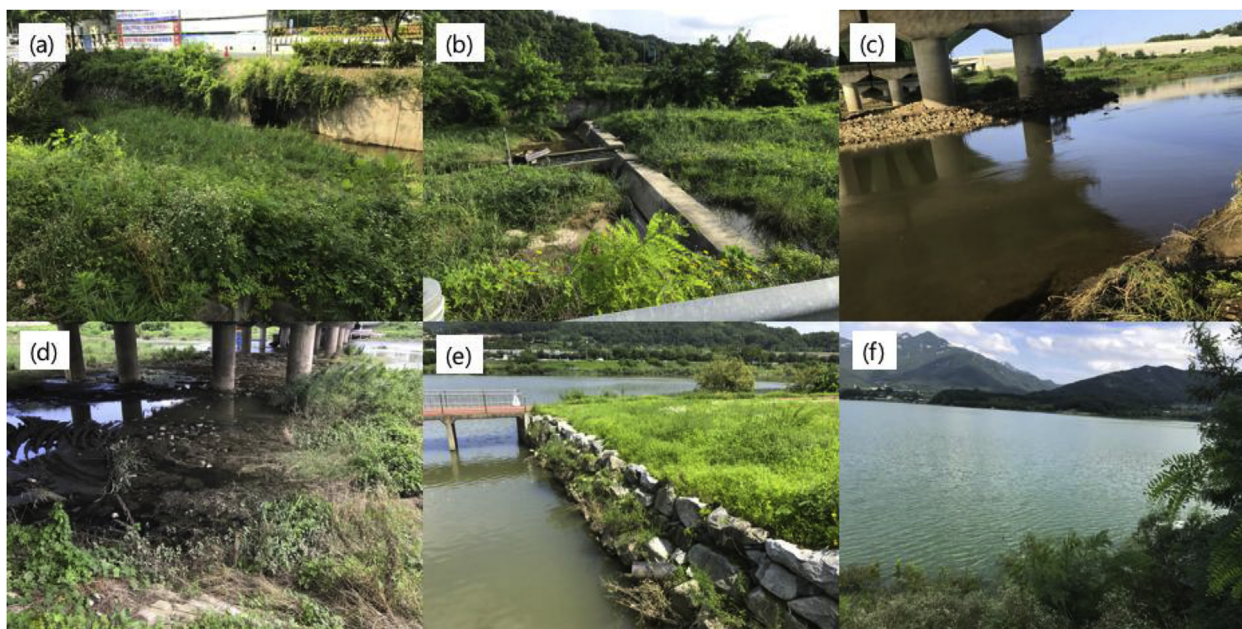


Fig. 4. Sampling sites of this study: (a) front of main gate of the Korea Atomic Energy Research Institute (KAERI), (b) Donghwa bridge, (c) Yongshin bridge, (d) Wonchon Bridge, (e) sewage treatment plant drain, and (f) Gongju reservoir.

the samples were dried at 100 °C for at least 24 h. Incineration was subsequently carried out in the following steps: drying (at 200 °C for 10 h); carbonization (at 300 °C for 48 h); and incineration (at 450 °C for 24 h) was carried out to prevent ^{137}Cs loss. Fig. 7 shows the temperature history of the pretreatment of each fish sample. After combustion, the mass of the incinerated samples was obtained, and the rate of incineration can be calculated. Thereafter, the whole amount of the sample was placed into U-8 vial, which was used for gamma isotope analysis while uniformly mixing the sample.

3. Results and discussion

The results of ^{131}I concentrations for sediment, surface water, and aquatic plant samples by applying the pretreatment procedure are described in this section. Table 3 shows the radioactive iodine (^{131}I) concentration of sediment, aquatic plant, and surface water.

3.1. Radioactive iodine (^{131}I) concentration in sediment

Radioactive iodine (^{131}I) was not detected in the sediments collected at the KAERI's front gate and Donghwa bridge during the

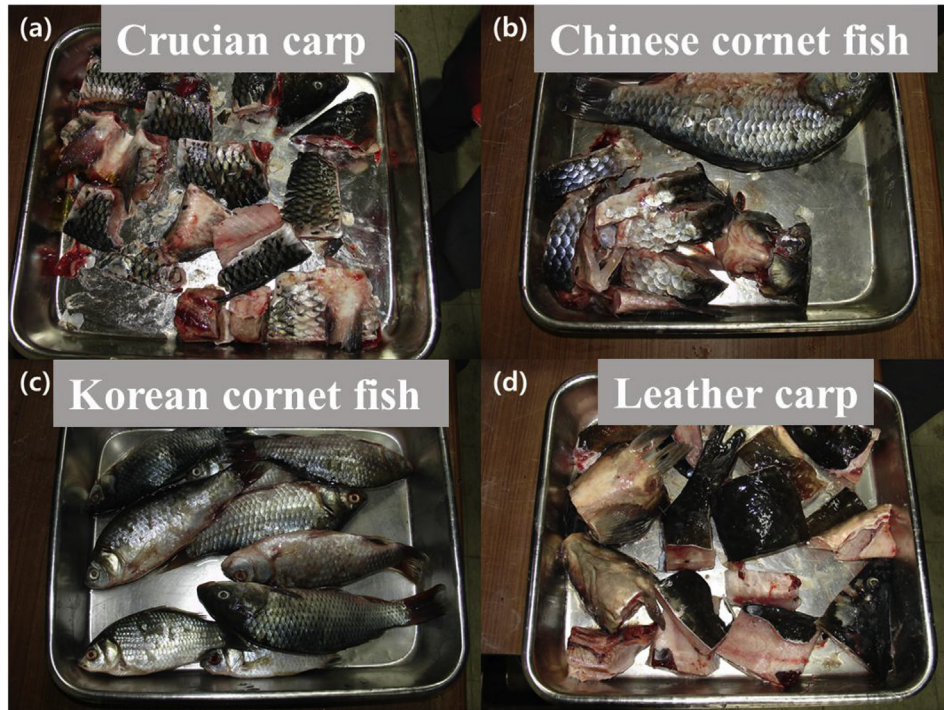


Fig. 5. Samples of fish collected from the sewage treatment plant drain and Gongju reservoir.

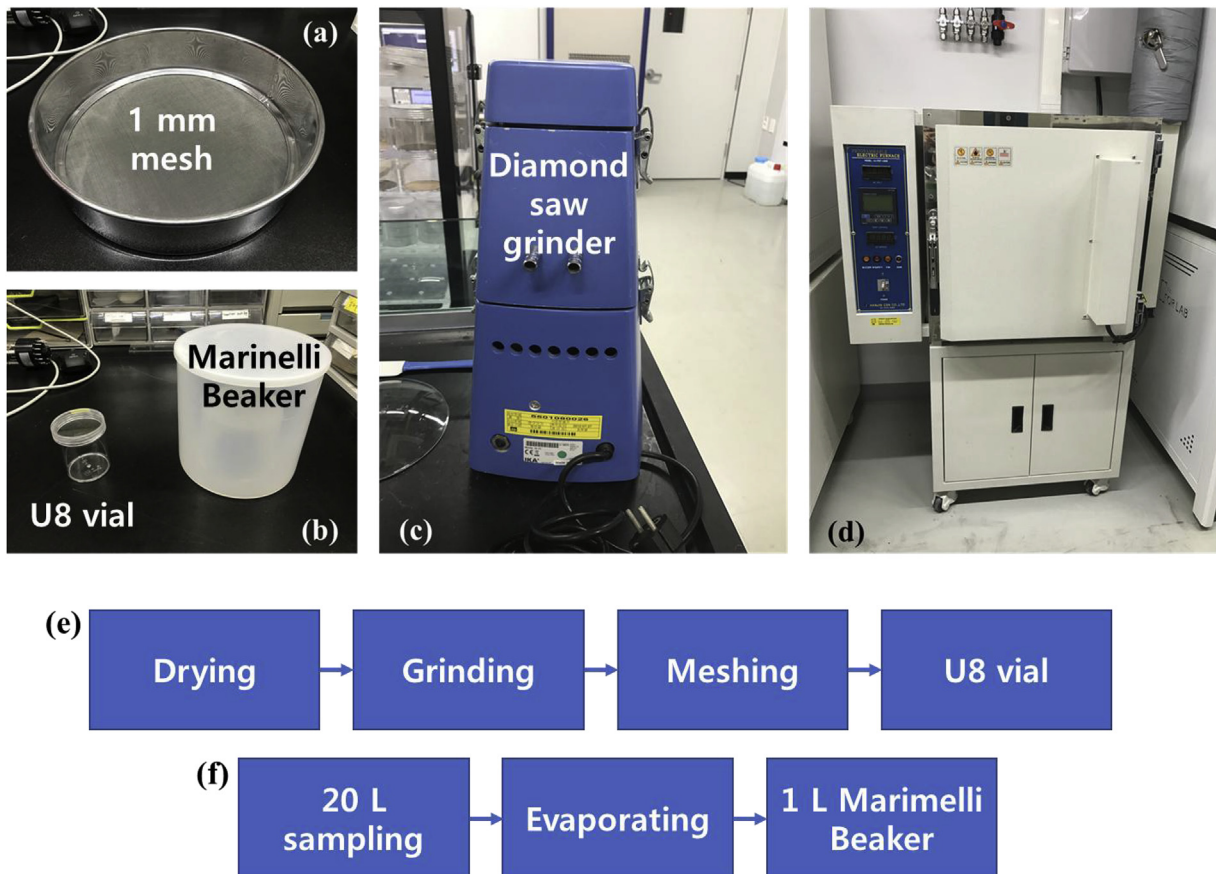


Fig. 6. Equipment for pretreatment process included (a) a sieve, (b) a U-8 vial and Marinelli beaker, (c) a grinder, and (d) electrical furnace; and pretreatment processes of (e) sediment and aquatic plant samples and (f) surface water samples.

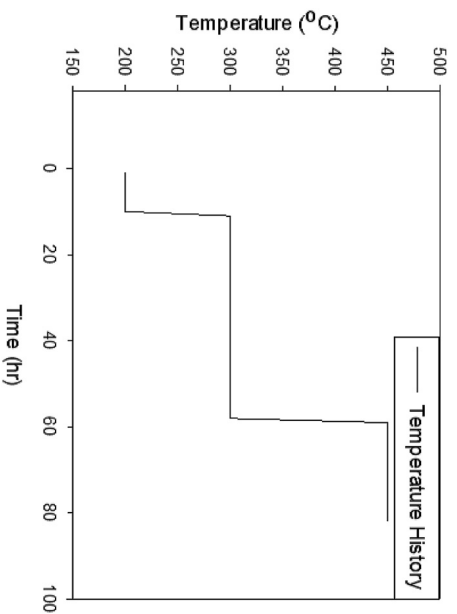


Fig. 7. Temperature history for pretreatment of samples.



Fig. 8. Gamma spectrometer with HPGe detector and Model 747 lead shield.

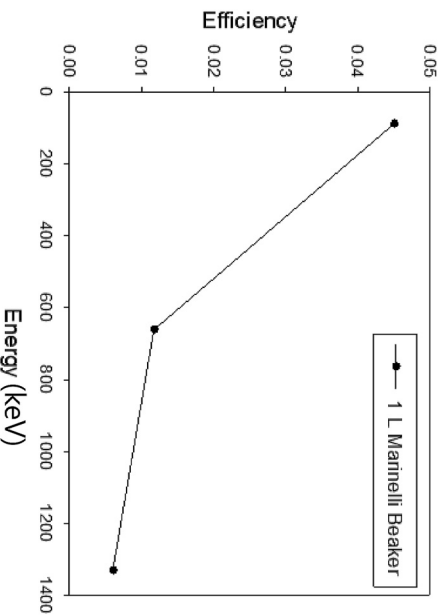


Fig. 9. Coefficient curve for gamma spectrometer.

Table 3

¹³¹I concentration of sediment, aquatic plant, and surface water at specific months.

Sample	Point	Sample number									
		August	October	November	December	February	March	April	May	June	July
Sediment (Bq/kg)	Front of main gate	<0.142	<0.155	<0.133	<0.151	<0.140	<0.179	<0.153	<0.131	<0.118	<0.0825
	Donghwa bridge	<0.114	–	–	–	–	–	–	–	–	–
	Wonchon bridge	<0.124	<0.270	<0.207	<0.207	<0.421	3.00 ± 0.108	<0.220	<0.225	<0.108	1.53 ± 0.0843
Aquatic plant (Bq/kg)	Drain of sewage treatment plant	7.39 ± 0.26	8.93 ± 0.199	4.49 ± 0.125	7.64 ± 0.195	2.30 ± 0.104	0.428 ± 0.0921	6.88 ± 0.17	5.47 ± 0.124	7.24 ± 0.174	10.1 ± 0.218
	Front of cogeneration plant	<0.125	0.766 ± 0.0747	0.843 ± 0.169	1.67 ± 0.0828	0.969 ± 0.131	2.02 ± 0.157	<0.273	3.27 ± 0.157	3.02 ± 0.156	4.70 ± 0.214
	Front of main gate	<0.0768	0.165 ± 0.0532	4.01 ± 0.367	1.61 ± 0.451	0.584 ± 0.0613	29.8 ± 0.719	5.58 ± 0.170	<0.291	<0.198	0.565 ± 0.129
Surface water (Bq/L)	Donghwa bridge	<0.0727	–	–	–	–	–	–	–	–	–
	Wonchon bridge	2.42 ± 0.13	1.92 ± 0.117	0.507 ± 0.156	1.09 ± 0.101	4.81 ± 0.192	1.77 ± 0.0998	3.66 ± 0.333	14.4 ± 0.314	5.36 ± 0.203	3.32 ± 0.126
	Drain of sewage treatment plant	60.5 ± 1.22	89.8 ± 1.80	627.6 ± 12.1	214.7 ± 4.06	123.7 ± 2.416	27.9 ± 0.179	2.73 ± 0.189	274.0 ± 5.23	386.6 ± 7.45	510.6 ± 9.83
	Front of cogeneration plant	12.4 ± 0.29	39.2 ± 1.39	1.64 ± 0.145	69.9 ± 1.40	47.2 ± 0.979	154.6 ± 3.02	378.1 ± 7.21	23.8 ± 0.510	24.1 ± 0.547	5.25 ± 0.144
Surface water (Bq/L)	Front of main gate	–	<0.00394	<0.00242	<0.00214	<0.00268	<0.00273	<0.00401	<0.00304	0.0138 ± 0.00187	<0.00295
	Drain of sewage treatment plant	–	0.713	0.343	3.21	<0.00534	0.839	0.834	0.661 ± 0.0130	1.51 ± 0.0298	0.482 ± 0.00966
	Front of cogeneration plant	–	±0.0151	±0.00816	±0.0607	±0.0173	±0.0169	0.322 ± 0.00698	1.28 ± 0.0257	0.125 ± 0.00298	
			0.334	0.324	1.07	0.757	0.896	0.0694			
			±0.00804	±0.00750	±0.0208	±0.0152	±0.0180	±0.00284			

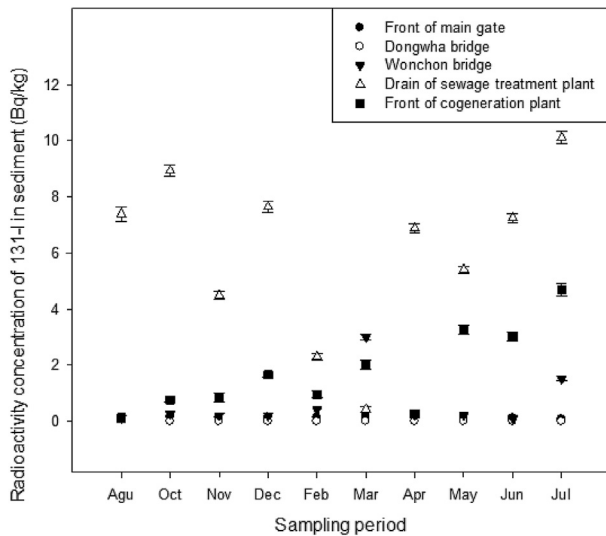


Fig. 10. Radioactivity concentration of sediment over the sampling period.

entire sampling period. A small amount of ^{131}I was detected in March and July at the Wonchon bridge. In the sewage treatment plant drain, ^{131}I was detected throughout the entire sampling period, and the highest value was measured in July. At the front of the cogeneration plant, radioactivity was detected for most of the sampling period, but it was lower than at the sewage treatment plant drain. Fig. 10 shows the trend of radioactivity concentration in sediment samples. The data from the sewage treatment plant shows a higher value than the other sampling points. The maximum radioactivity concentration at this sampling point was 10.1 Bq/kg, whereas that at the Yongshin bridge was 4.7 Bq/kg and that at Wonchon bridge (upstream of the sewage treatment plant) was much lower (1.53 Bq/kg and 3.00 Bq/kg).

3.2. Radioactive iodine (^{131}I) concentration in aquatic plants

Fig. 11 shows the radioactive iodine (^{131}I) concentration of aquatic plant samples. Radioactivity was detected at the Wonchon bridge, sewage treatment plant drains, and front of the cogeneration plant. The sewage treatment plant drain had the highest ^{131}I concentration overall, except for March and April, after which it

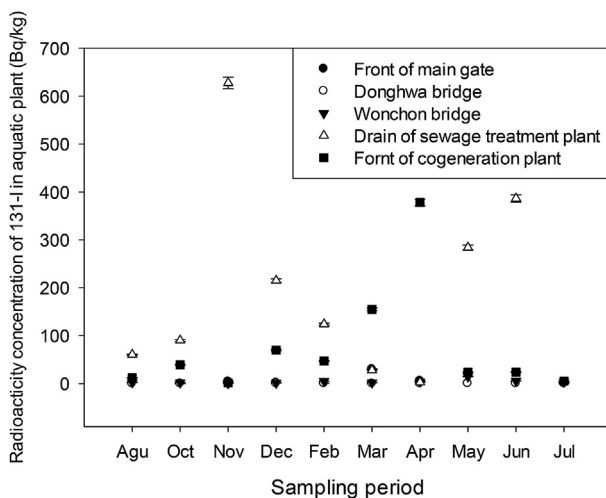


Fig. 11. Radioactivity concentration of aquatic plants over the sampling period.

increased again (Fig. 11). On the other hand, the radioactive iodine (^{131}I) concentration at the front of the cogeneration plant increased until April and decreased after April. The radioactive iodine (^{131}I) concentration in the aquatic plants collected at the sewage treatment plant drain had a minimum value of 27.9 Bq/kg in April and a maximum value of 627.6 Bq/kg in November. These samples also had a minimum value of 1.6 Bq/kg and a maximum value of 69.9 Bq/kg at Yongshin bridge. Radioactive iodine (^{131}I) concentration was detected seven times at the KAERI's front gate and Donghwa bridge with averages of 2.2 Bq/kg and 4.6 Bq/kg, respectively. At the Wonchon bridge, radioactivity was detected throughout the sampling period (with an average of 3.9 Bq/kg).

3.3. Radioactive iodine (^{131}I) concentration in surface water

Fig. 12 shows the radioactive iodine (^{131}I) concentration in surface water throughout the sampling period. The radioactive iodine (^{131}I) was not detected at the KAERI's front gate or Donghwa bridge, but it was detected at the sewage treatment plant drain and the front of the cogeneration plant. The highest ^{131}I concentration at the former was in December (3.21 Bq/kg), and the latter had the highest value in June (<0.00534 Bq/kg), which indicates MDA. Furthermore, the radioactive iodine (^{131}I) concentration at the sewage treatment plant drain and cogeneration plant increased in June and decreased in July. Table 4 shows the range and average ^{131}I concentration of detected samples. The maximum ^{131}I concentrations at the sewage treatment plant drain and Yongshin bridge were 3.21 Bq/L and 1.28 Bq/L, respectively. No ^{131}I was detected at the Wonchon bridge.

3.4. Radioactive iodine (^{131}I) concentration in fish

Radioactive iodine (^{131}I) was detected in all fish caught at the sewage treatment plant drain (Table 5). The maximum concentration was 11.35 Bq/kg in the 2355 g carp, and the mean value was 3.93 Bq/kg in crucian carp. On the other hand, ^{131}I was not detected in the fish captured at the Gongju reservoir (Table 6). This indicated that ^{131}I flowed into Gap River.

3.5. Radioactivity analysis of ^{131}I

For radioactivity analysis, the background counting factor was calculated under the same conditions as in the sample analysis and

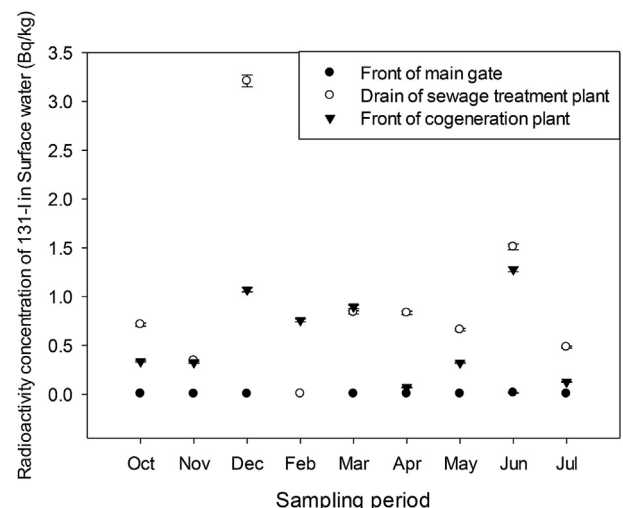


Fig. 12. Radioactivity concentration of surface water over the sampling period.

Table 4
The range and average ^{131}I concentration of sediment, aquatic plant and surface water samples in the sampling area.

	^{131}I Concentration (Bq/kg-dry, Bq/L)		
	Sediment(Average)	Aquatic plant(average)	Surface water(average)
Front of main gate	<MDA	<MDA ~ 5.6(2.2)	<MDA
Donghwa bridge	<MDA	<MDA ~ 18.7(4.6)	<MDA
Wonchon bridge	<MDA ~ 3.0(2.27)	1.1 ~ 14.4(3.9)	<MDA
Drain of sewage treatment plant	0.43 ~ 10.1(6.09)	27.9 ~ 627.6(282)	0.34 ~ 3.21(1.07)
Yongshin bridge	<MDA ~ 4.7(2.16)	1.6 ~ 69.9(26.2)	0.07 ~ 1.28(0.52)

Table 5
 ^{131}I concentration of fish samples caught at the sewage treatment plant drain in the Gap River.

	Mass	Radioactivity concentration of ^{131}I (Bq/kg)
Cornet fish 1	185 g	0.861 ± 0.0955
Cornet fish 2	420 g	1.19 ± 0.0734
Crucian carp 1	365 g	5.08 ± 0.156
Crucian carp 2	495 g	2.38 ± 0.127
Crucian carp 3	440 g	3.57 ± 0.106
Crucian carp 4	475 g	3.63 ± 0.109
Crucian carp 5	445 g	3.19 ± 0.109
Crucian carp 6	355 g	4.06 ± 0.140
Crucian carp 7	575 g	5.61 ± 0.184
Flesh of carp	875 g	0.699 ± 0.0626
Bone of carp	1040 g	2.39 ± 0.0945
Internal organs of carp	440 g	8.26 ± 0.227

Table 6
 ^{131}I concentration of fish samples captured in the Gongju reservoir.

	Mass	Radioactivity concentration of ^{131}I (Bq/kg)
Crucian carp 1	675 g	<0.0549
Crucian carp 2	395 g	<0.0972
Crucian carp 3	390 g	<0.134
Carp	1025 g	<0.0323
Leather carp 1	720 g	<0.0869
Leather carp 2	975 g	<0.0484

subtracted from the sample counting rate to calculate the net counting rate of the sample itself. The same empty container that was used for the samples (U-8 vial for sediment and fish samples and Marinelli beakers for the surface water samples) was measured for a sufficient time under the same conditions as the sample measurement. There were no radiation sources (natural or artificial radionuclides) in the measurement room that could increase the background. The radioactivities (^{131}I) of all pretreated samples were measured with a gamma spectrometer (HPGe detector with MCA, ORTEC Co.) [15] in a standardized container, as shown in Fig. 8. The gamma spectrometer was calibrated according to the following procedure: the standard mixed source (^{109}Cd , ^{54}Mn , ^{57}Co , ^{60}Co , ^{137}Cs , ^{88}Y , etc.) was diluted to a certain amount (0.1 Bq/g–10 Bq/g) and weighed. The detection efficiency was calculated using the HPGe detector for each source of energy and nuclear species for the standard source of each sample measured (Table 2) (Fig. 9) [16–20].

Based on the background and blank counts, the minimum detectable activity (MDA) for this geometry was derived using Curie's method [21]:

$$\text{MDA} = \frac{\sigma\sqrt{B}}{\epsilon PTW} \text{ (Bq/kg)}$$

where σ is the statistical coverage factor ($= 1.645$) ($P \leq 0.05$), B is the background radiation of the radionuclide of interest, ϵ is the counting efficiency of the detector, P is the absolute transition probability of γ -decay, W is the dried sample weight (g), and T is

the counting time (s). In this study, the counting time for each sample was 80,000 s.

3.6. Analysis of outbreak location and contamination of ^{131}I

The radioactive iodine (^{131}I) was mostly detected at the sewage treatment plant drain, but it was not detected at Wonchon bridge located upstream of the plant. At the sewage treatment plant, wastewater from Daejeon city is collected, including that from hospitals with patients diagnosed by means of radioactive iodine. The radioactive iodine (^{131}I) was detected in all fish captured in the sampling points downstream of the sewage treatment plant, but it was not detected in the fish captured in the Gongju reservoir. This indicates that fish were affected by ^{131}I -contaminated river water. The radioactive iodine (^{131}I) was detected throughout the sampling period at the sewage treatment plant drain and sediment and at the Yongshin bridge, but it was not detected at the KAERI's front gate and Donghwa bridge. This suggests that the presence of ^{131}I in the Gap River is independent of the RIPF. In the case of aquatic plants, traces of ^{131}I were detected at the KAERI's front gate, Donghwa-wool, and Wonchon bridge. This is probably due to the direct release of excrement to the environment by patients who have been diagnosed by means of radioactive iodine, and it seems to be due to the accumulation of ^{131}I in aquatic plants [22].

According to the "The Table of Food Supply and Demand" of the Korea Rural Economic Institute, the annual supply of fish per capita was 24.5 kg in 2015 [23–26]. The internal exposure dose assessment was carried out according to the scenario of consuming fish with the highest radioactivity concentration for one year. According to the Annex of the International Commission on Radiological Protection Publication 119, effective dose coefficient for the ingestion of ^{131}I radionuclides for adult members of the public up to 70 years of age is $2.2\text{E}-8$ Sv/Bq [27]. The aquatic plants that were sampled at the sewage treatment plant drain in November had the highest value of radioactivity concentration as 8.26 Bq/kg. Therefore, internal exposure due to the ingestion of fish can be calculated as the product of annual intake and radioactivity concentration and dose coefficients (0.0045 mSv/year). This value is lower than 1 mSv/year, which is the legal limit for the public [28–30]. The legal limit of the radioactive iodine concentration for food is 100 Bq/kg, and all the fish samples showed the value 1/12 lower than this standard.

4. Conclusions

Radioactive iodine (^{131}I) is a radionuclide that has been detected in river water and aquatic organisms. This radionuclide enters rivers in large cities through sewage treatment plants due to an increase in medical use. This study indicated that ^{131}I was detected in environmental samples from the Gap River, but the radioactivity concentration by location showed that it did not enter the river at the RIPF. In addition, the wastewater from the RIPF is collected in the storage tank, and after analyzing the nuclide, it is confirmed that the nuclide is not detected and transferred to the wastewater

treatment facility. The radioactive iodine (^{131}I) was not detected in surface water at the RIPF, but it was detected in the drain of a sewage treatment plant located downstream of the RIPF. Therefore, it is concluded that hazardous ^{131}I detected in environmental samples could have entered the environment through the wastewater of facilities using radioactive isotopes, such as hospitals. Through this paper, we hope that the public perception of ^{131}I being emitted from the RIPF to the environment will be improved.

Declaration of interest

None.

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