MEASUREMENTS OF ^{137}Cs AND ^{40}K CONCENTRATIONS FROM FISH IN JEJU SEA

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Abstract

A variety of artificial and natural radioactive isotopes were distributed to the earth atmosphere due to the nuclear weapon tests, nuclear power plant accident, the cosmic rays and the lithosphere rays. With the aim of assessing their effects to the public health, the concentrations and the resulting effective doses of ^{137}Cs and ^{40}K from fish in Jeju Sea were measured and derived: ^{137}Cs and ^{40}K as the representatives of the artificial and natural radioactive isotopes, respectively.

Hairtail, mackerel, filefish and squid were chosen as the samples, as they are consumed much by the public in Jeju. 5kg of each species were sampled and sorted by flesh, bone and intestines.

After drying, carbonization and ashery processes, the concentrations of ^{137}Cs and ^{40}K from the samples were measured by Gamma-ray Spectrum Analysis using a high purity germanium detector and multi-channel analyser.

The concentrations of ^{137}Cs showed the distribution from MDA (Minimum Detectable Activity) to 133mBq/kg·flesh weight and the distribution of ^{40}K concentrations was from MDA to 147Bq/kg·flesh weight. While ^{137}Cs was mainly detected in all eating parts except squid, ^{40}K was detected in all parts. The concentration of ^{137}Cs detected in eating part of the hairtail was most high(133 ± 15.9·flesh weight) and the case of ^{40}K showed the highest concentration in eating part of a filefish (147 ± 6.20 Bq/kg·flesh weight).

These results were within the average range of concentrations of ^{137}Cs (61.3 mBq/kg·flesh weight~262.5mBq/kg·flesh weight) by the KINS(Korean Institute of Nuclear Safety) study in 2002. Also the ^{40}K concentrations in the mackerel and hairtail come under the measurement range of KINS.

The effective dose per year through the intake of fish was about 1.6067×10^{-3} mSv, which was very small compared to the total effective dose (2.4mSv per year) by the natural radiations reported from UNSCEAR 2000. When compared with 0.29mSv, the effective dose per year through the intake of foods, this figure is about 1/180.

Introduction

Various artificial radioactive isotopes were emitted to the atmosphere all over the world due to Chernovyl accident and nuclear weapon tests. Especially, a lot of ^{137}Cs (Half life: 30.2 yr), an γ emitter, had been dispersed to the outer space as floating form of fall-out. Since γ ray from ^{137}Cs has strong energy enough to cause harmful effects on all the livings than any other isotopes, it is very important to monitor the environmental radioactive contamination by measuring the concentration of ^{137}Cs for human beings.

The ^{137}Cs existing in the atmosphere falls by weather change and then is absorbed into the soil or seawater. Thus, human can be exposed to the hazard of radioactive contamination by direct inhalation of ^{137}Cs or by ingesting polluted plants or seafood. ^{137}Cs accumulated in the human body is known to affect the digestive system or muscle and can cause the cancers, in particular.

This study is focused on internal exposure effect in case of taking in seafood especially fish, which could provide with fundamental data for radioactivity environment of seawater regarding fish.

Backgrounds

1. Radioactive isotopes

Natural and artificial radioactive isotopes exist in the environment follow as.

1) Natural radioactive isotopes

There are about 14 natural radioactive isotopes that do not accomplish the disintegration family. Among them, ^{40}K, ^{87}Rb, ^{50}V, ^{115}In, ^{138}La, ^{147}Sm, ^{176}Lu are easily detected around us.

Seeing the ^{40}K, this is one major aiming isotope in this study and its atomic number and mass number are each 19 and 40. It is decays by through β-ray emission and electron capture and its half life is 12.5 hundred million years. ^{40}K is radioactive isotope that has being remained in the earth's surface with the beginning of the earth.

2) Artificial radioactive isotopes

Most artificial radioactive isotopes are created by fission reactions and the number is about 200. Among them, radioactive isotopes which have mass number from 72 to 161 are detected. Seeing the ^{137}Cs, this is the other major aiming isotope in this study. Its half life is 30.2 yr and is converted to ^{137m}Ba. During this process, the ^{137}Cs emits β-rays which have each maximum energy of 1.176 MeV(6.5%), 0.514 MeV(93.5%) and then is changed to ^{137m}Ba(Half life: 2.55 min) emitting γ ray of 0.662 MeV(=662 KeV). Finally it is converted to the stable ^{137}Ba by isomeric transition.

2. Behavior of radioactive materials

Fig.1 shows that the route of radioactive isotopes absorbed into human body through sea living things from the atmosphere.

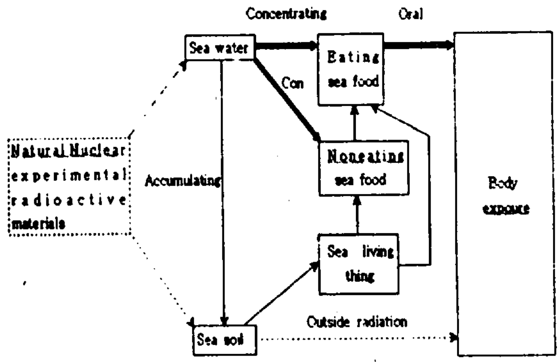

Fig 1. The route of radioactive material through sea.

3. Assessment of effective dose

Assessment of effective dose is carried out in order to calculate internal exposures which can be caused by intake of food from radioactive isotopes. In case of several intestines which were exposed to equivalent dose, the relevant symptoms and hazards differ according to the exposed intestines. Decrease of residual radioactivity in the human body complies with exponential attenuation rule as radioactive decay and half life is controlled by effective half-life that is composition effect by two of exclusion by physical decay and metabolism in this time. The following many factors including a accumulating factor, sensibility of human intestines, effective half life were applied in order to calculate effective doses.

$$R_{apj} = 1.12\times10^{9}\ q_i\ R_i\ B_{ip}\ D_{aipj}\ \exp(-\lambda_i t_p)$$

R_{apj}: exposed dose by intake of seafood (μSv/yr), age (a), exposed path (p), target intestines (j)

M_p: mixing coefficient at dose calculation target position

U_{ap}: intake of seafood (kg-amount of live weight/yr)

q_r: emissivity of liquid phase radioactivity effluence (ft^3/sec)

q_i: emissivity of radioactive materials (TBq/yr)

R_i: reaccumulating factor

B_{ip}: living body accumulating factor (λ/kg-eating part; liveweight standard)

D_{aipj}: dose conversion factor of intake (Sv/Bq-intake)

λ_i: decay constant of radioactive materials (hr^{-1})

t_p: Time that takes though liquid phase radioactivity effluence moves to dose calculation target position (hr)

1.12×10^9: unit conversion factor

Measurement Methods and Procedures

1. Samples preparing and pretreatment

Hairtail, mackerel, filefish and squid were chosen as representative fish consumed much. From 5 kg of each species, samples were prepared by eating part, bone and intestines and then washed clearly under running water and each liveweight was calculated.

Samples were treated by dryness for 2 hr, carbonization for 5 hr and ash stage, finally.

2. Standard samples manufacturing and efficiency calibration methods

1) Standard sources (calibration sources)

The subject for gamma radioactivity analysis in the environmental samples is from 50 keV to 3 MeV of γ rays energy. Thus the isotopes releasing γ rays which have from 100 keV to 2 MeV are used as standard samples in general.

In this study, standard sources used for gamma calibration are the mixed sources (QCY48) which consist of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, ^{60}Co.

Table 1 indicates energy calibration and peak efficiency calibration by using standard sources.

Table 1. Standard sources (QCY48, Amersham) use for analysis of HPGe detector.

Nuclear	Energy (keV)	Yield	Half-life (day)	Counting rates	Reference radioactivity(Bq)*	Counting activity (Bq)**
^{241}Am	59.54	0.36	1577745.3	4.62915	169.5085	169.4519
^{109}Cd	88.03	0.0363	462.6	4.16982	96.9264	86.4941
^{57}Co	122.06	0.8560	271.79	4.04515	87.1586	71.8016
^{139}Ce	165.86	0.7987	137.64	3.68822	106.9947	72.9700
^{203}Hg	279.2	0.8148	46.595	3.30629	286.2710	92.4218
^{113}Sn	391.7	0.6489	115.09	5.3038	316.6262	200.3369
^{85}Sr	514.01	0.9840	64.849	5.50235	585.7660	259.9747
^{137}Cs	661.66	0.8510	11012.7	6.37072	369.8231	368.0582
^{88}Y	898.04	0.9400	106.63	7.23235	944.4688	576.2753
^{60}Co	1173.24	0.9986	1924.2	4.83494	501.9134	488.3590
^{60}Co	1332.5	0.9998	1924.2	4.40792	502.2140	488.6514
^{88}Y	1836.06	0.9936	106.63	4.25621	998.4170	609.1922

*Reference date : 2004. 12. 01.

**Counting date : 2005. 02. 15.

2) Standard samples manufacturing

Standard sources mixed with gamma releasing isotopes were diluted fittingly by 1M HCl solution (Fig. 2) and put into samples chamber and then calculated their radioactive concentrations. Because the height of environmental samples is difficult to be packed changelessly, 11 of standard sources which have a difference of 5 mm for efficiency and energy calibration samples were manufactured. Each weight of the samples was quantified exactly in order to calibrate the density by self-absorption effect (Fig. 3).

Fig. 2. 1M HCl solution Fig. 3. standard sources.

3) Efficiency calibration

The manufactured standard samples were detected by Gamma-ray Spectrum Analysis using a high purity germanium detector and multi-channel analyzer for 80,000 sec. Table 2 presents properties of HPGe(High-Purity Germanium) detector.

Table 2. Properties of HPGe detector.

Properties	
Resolution(FWHM) at 1.33MeV, ^{60}Co	1.85 keV
Peak-to-Compton ratio, ^{60}Co	58/1
Relative efficiency at 1.33MeV, ^{60}Co	30 %
High voltage	3200 V
VCrystal diameter	57.7mm
Crystal length	75.3mm
End cap to crystal	3 mm

To reflect the detecting efficiency as well as the calibration and resolution of energy on the measured spectrum, the calibrated spectrum was made by applying Aptec. program and establishing the interpolated correlation about those factors at intervals of 1 mm from 2.5 mm to 50 mm. The detecting efficiency of standard sources decreases due to summing effect of ^{60}Co, ^{88}Y and the density of samples causes it to decrease due to self-absorption effect through interacting materials of samples with γ rays. Thus, this study calibrated the summing effect and self-absorption effect of samples.

Measurement Results and Analysis

1. Results of samples pretreatment

Table 3 shows an ash quantity and an ash rate after pretreatment processes were finished.

Table 3. Live-weights an d ash rate of samples.

Samples	Region	Purchasing liveweight (kg)	Ash quantity (g)	Ash rate (%)	Detecting liveweight (kg)	Average height of samples (mm)
File fish	Eating part	1.60	54.76	3.42	1.60	47.2
	Bone	2.10	108	5.13	0.76	49.2
	Intestine	1.10	19.0	1.73	1.10	13.4
Hair tail	Eating part	2.58	82.1	3.18	1.70	48.5
	Bone	1.93	124	6.43	1.21	49.0
Mackerel	Eating part	2.45	50.8	2.75	2.45	45.5
	Bone	1.70	94.2	5.54	1.18	49.2
	Intestine	0.40	15.3	3.83	0.40	11.4
Squid	Eating part	3.93	135	3.44	1.56	48.5
	Intestine	0.86	15.8	1.84	0.86	13.5

The purchasing live-weight presents the real weight of each part when were purchased and the detecting live-weight presents real weights used for detection.

2. Spectrum of background and standard sources

Fig. 4 and Fig. 5 present general forms of background and standard's spectrum. ^{40}K and Th or U family isotopes easily detected around us were detected in the background spectrum files.

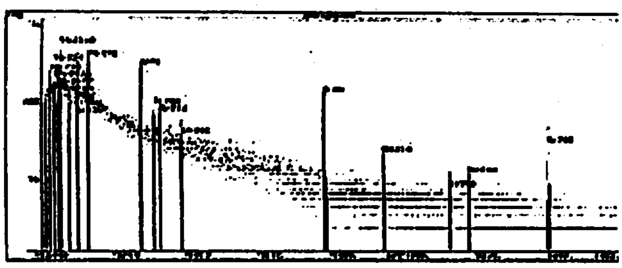

Fig. 4. Spectrum of background.

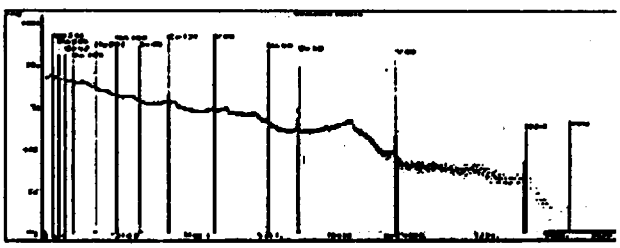

Fig. 5. Spectrum of standard sources.

Table 4 presents the results of detecting efficiency depending on the height of standard sources. As the height of sources rises, the detecting efficiency decreased due to effects by samples' density. As energy rises, the detecting efficiency decreased except below 122.06 keV.

3. Results of analyzing samples' spectrum

Among results which were analyzed about the samples' spectrum of ^{137}Cs and ^{40}K, Fig. 6 and Fig. 7 were much higher than any other samples.

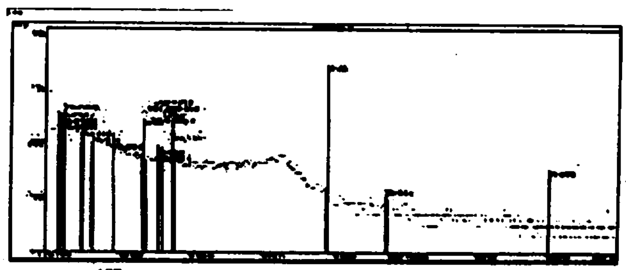

Fig. 6. ^{137}Cs spectrum of hairtail's eating part.

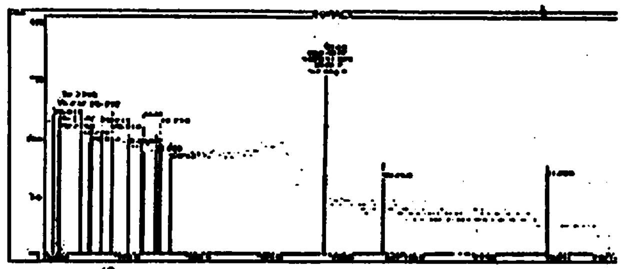

Fig. 7. ^{40}K spectrum of file fish's eating part.

4. The concentrations of ^{137}Cs and ^{40}K among samples

The concentrations of ^{137}Cs and ^{40}K among samples were showed in the Table 5.

The measured average value and standard

Table 4. Detecting efficiency following height of stadard sources.

	^{241}Am	^{109}Cd	^{57}Co	^{139}Ce	^{203}Hg	^{113}Sn	^{85}Sr	^{137}Cs	^{88}Y	^{60}Co	^{60}Co	^{88}Y
	59.54	88.03	122.0	165.86	279.2	391.7	514.01	661.66	898.04	1173.2	1332.5	1836
1.8	7.47	13.7	16.1	14.0	9.85	7.15	5.65	4.51	3.04	2.36	2.10	1.61
5.3	6.70	12.1	14.2	12.4	8.76	6.33	5.05	4.03	2.74	2.15	1.90	1.47
11.1	5.94	10.6	12.3	10.8	7.57	5.53	4.38	3.53	2.42	1.90	1.70	1.31
15.4	5.15	9.93	10.8	9.54	6.71	4.90	3.87	3.15	2.18	1.72	1.53	1.19
20.2	4.65	8.54	9.73	8.49	6.04	4.39	3.52	2.82	1.99	1.57	1.40	1.08
25.1	4.23	7.79	8.83	7.80	5.49	4.04	3.21	2.60	1.85	1.46	1.32	1.01
30.4	3.81	6.95	7.85	6.96	4.97	3.63	2.90	2.34	1.67	1.31	1.19	0.92
35.5	3.41	6.27	7.12	6.34	4.46	3.30	2.63	2.13	1.54	1.22	1.10	0.85
40.4	3.17	5.81	6.47	5.84	4.18	3.06	2.47	2.00	1.44	1.14	1.03	0.79
45.3	2.91	5.35	6.11	5.42	3.85	2.85	2.30	1.86	1.35	1.07	0.96	0.75
50.0	2.73	4.96	5.63	5.05	3.58	2.65	2.12	1.73	1.26	0.99	0.90	0.70

Table 5. Concentrations of ^{137}Cs and ^{40}K among samples.

Samples	Region	^{137}Cs (mBq/kg·flesh weight)		^{40}K (Bq/kg·flesh weight)	
		Concentrations	MDA	Concentrations	MDA
File fish	Eating part	69.4 ± 19.7*	63.1	147 ± 6.20	0.542
	Bone	58.3 ± 31.7	104	94.7 ± 4.15	0.914
	Intestine	20.0 ± 11.3	37.0	29.0 ± 0.58	0.392
Hair tail	Eating part	133 ± 15.9	45.6	94.1 ± 4.00	0.474
	Bone	113 ± 18.7	55.7	73.9 ± 3.20	0.560
Mackerel	Eating part	105 ± 12.0	35.9	114 ± 1.43	0.345
	Bone	20.1 ± 22.0	73.2	80.0 ± 3.45	0.662
	Intestine	103 ± 23.4	72.8	81.3 ± 1.36	0.807
Squid	Eating part	1.12 ± 11.2	59.9	115 ± 4.88	0.499
	Intestine	11.1 ± 16.3	54.2	93.6 ± 1.38	0.473
Total**		307.4		470.1	

*Detect errors, **Calculation of only eating part's concentrations

deviation of ^{137}Cs concentrations are 104.7±17.94 mBq/kg·flesh weight.

The range of ^{137}Cs concentrations was from MDA (Minimum Detectable Activity) to 133 mBq/kg·flesh weight and ^{40}K was from MDA to 147 Bq/kg·flesh weight.

^{137}Cs concentrations were detected in all eating parts except squid and also were detected in hair tail's bone and mackerel's intestine.

^{40}K was detected in all parts. The highest concentrations of ^{137}Cs and ^{40}K were detected in hair tail's eating part and file fish's eating part.

5. Assessment of effective dose per year

In this study, the effective dose per year was calculated by using following relations which consist of concentrations of fish, intake of fish per year consumed by jeju's people, conversion coefficient excluding accumulating factor, sensibility of human body.

Table 6 presentstotal effective dose and effective dose per year by ^{137}Cs and ^{40}K.

$$H(mSv/y) = A(mSv/Bq) \times B(kg/y) \times C(Bq/kg \cdot flesh)$$

Table 6. Total effective dose and effective dose per year by ^{137}Cs, ^{40}K.

Samples	Intake (kg)	Effective dose by ^{137}Cs (mSv)	Effective dose by ^{40}K (mSv)	Total Effective dose* (mSv)
Total all eating parts	5.44	2.17×10^{-5}	1.585×10^{-3}	1.6067×10^{-3}

*^{137}Cs + ^{40}K

A : conversion coefficient according to oral intake(^{137}Cs: 1.3×10^{-5}, ^{40}K: 6.2×10^{-6})

B : intake of fish per year

C : radioactivity concentrations in fish

Conclusion

The radioactivity concentrations of ^{137}Cs showed the distribution from MDA (Minimum Detectable Activity) to 133mBq/kg·flesh weight and the radioactivity concentrations of ^{40}K showed the distribution from MDA to all eating parts except squid. ^{40}K was detected by all parts.147Bq/kg·flesh weight. ^{137}Cs was detected in all eating parts except squid. ^{40}K was detected by all parts,

The radioactivity concentrations of ^{137}Cs were the highest in eating part of a hairtail(133 ± 15.9 mBq/kg·flesh weight) and the concentrations of ^{40}K was the highest in eating part of a filefish(147 ± 6.20 Bq/kg·flesh weight). These results were within the average range of concentrations of ^{137}Cs (61.3 mBq/kg·flesh weight ~262.5 mBq/kg·flesh weight) by the KINS (Korean Institute of Nuclear Safety) study in 2002. Also the ^{40}K concentrations in the mackerel and hairtail come under the measurement range of KINS. The effective dose per year through the intake of fish was about 1.6067×10^{-3} mSv, which was very small compared to the total effective dose (2.4 mSv per year) by the natural radiations reported from UNSCEAR 2000.

Thus it is concluded that the effective doses caused by the intake of fish show the negligible effects to the public health. Also this study provides fundamental radioactivity concentration data in fish regarding the artificial and natural radioactivity concentration distributions covering eating and non-eating parts.

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Key words : natural and artificial radioactive isotope, ^{137}Cs and ^{40}K concentration of fish, effective dose