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■論 文■

Assessment of Internal Dose by ^{3}H & ^{14}C of Total Diet for Inhabitants near Wolsung Nuclear Power Plants

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Abstract - To assess the internal dose by ^{3}H & ^{14}C in total diet of inhabitants near Wolsung Nuclear Power Plants, TFWT, OBT and ^{14}C concentration in total diet was analyzed for collection region and time. TFWT, OBT and ^{14}C concentrations were in the range of 3.19-42.2 Bq/L , 1.00-39.4 Bq/L, and 0.230-0.856 Bq/gC, respectively. The calculated annual effective dose with TFWT, OBT and ^{14}C is 6.10 $\times 10^{-5}$ mSv/y, 3.71 $\times 10^{-5}$ mSv/y and 7.08 $\times 10^{-3}$ mSv/y, respectively. And then annual internal dose with total diet for inhabitants near Wolsung NPPs is about 7.18 $\times 10^{-3}$ mSv/y, which is about 0.72% of annual effective dose limit 1 mSv/y.
Key words: internal dose, ^{3}H, ^{14}C, Wolsung nuclear power plant, diet, TFWT, OBT

INTRODUCTION

^{3}H and ^{14}C are the biologically important radioisotopes since they are easily absorbed into the human body and their metabolism is the same as that of the stable isotopes, ^{1}H and ^{12}C. They have easy access via food or water to all molecules of human body tissues because they have higher values of max. permissible concentrations than other radionuclides.[1]

Wolsung Nuclear Power Plants(NPPs), which is pressurized heavy water reactor, produces ^{3}H and ^{14}C more than pressurized light water reactor. So it is important that examines the behavior in the environment of ^{3}H and ^{14}C, the concentrations of ^{3}H and ^{14}C in total diet of inhabitants around NPPs and internal exposure dose in human body. Also the behaviors in the environment of ^{3}H and ^{14}C around NPPs and the assessment of dose by ^{3}H and ^{14}C have been studied actively with Japan and Canada as the central figure. The study of radioactive concentration of water, soil, marine creatures and total diet for inhabitants and assessment of internal dose by those is progressed lively.

^{3}H in plant or animal tissue is divided into tissue free water tritium (TFWT) and organically bound tritium (OBT). TFWT or loose tritium is defined as that which can be removed from plant or animal tissue by lyophilization or mild distillation techniques. OBT or bound tritium that which remains after this process and which may be removed by combustion.[2] The biological effectiveness of OBT is greater than that of TFWT for the biological half life of OBT is longer than that of TFWT.[3] ^{14}C is quite mobile, has a long half-life (5730 a) and is readily incorporated

into biological tissues.[4] The radiation dose commitment and the dose rate resulting from the environmental build-up of ^{14}C are considerably larger than those of tritium because of the long half-life.[5]

Therefore we investigated the concentrations of TFWT, OBT and ^{14}C in total diet ingested by inhabitants, the characteristics of the radioactive distribution over area and time and the internal irradiated dose by total diet in order to assess the internal dose by total diet for inhabitants near Wolsung NPPs.

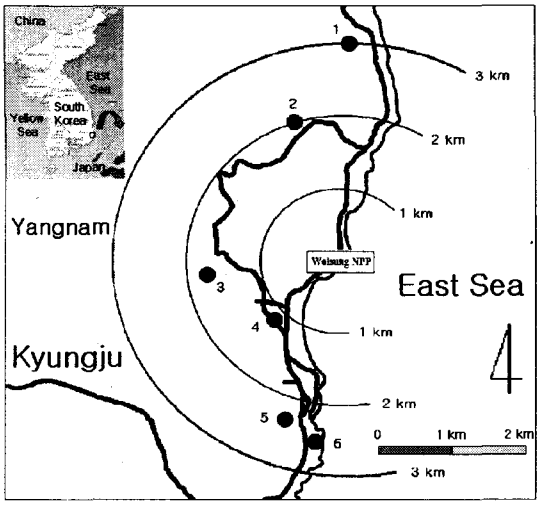

Fig. 1. Sampling locations of total diet near Wolsung NPPs. 1: Bonggil-1ri, 2: Bonggil-2ri, 3: Nasan-ri, 4: Naa-ri, 5: Ubchun-1ri, 6: Ubchun-2ri.

MATERIAL AND METHODS

Wolsung Nuclear Power Plants is located in Narr-ri, Yangnam-myon, Kyungju city in Korea, bordered by the East Sea. Wolsung NPP 1, which began its operation in 1983, is a CANNDU-type reactor with a total gross capacity of 678 MWe. Wolsung NPPs 2, 3 and 4 are the same type of reactors as Wolsung NPP1, and their operation began in 1997, 1998 and 1999, respectively. The total gross capacities of Wolsung NPP 2, 3 and 4 are 700 MWe.

Total diet samples were collected at 6 locations, which are Naa-ri, Nasan-ri, Bonggil-1ri, Bonggil2ri, Ubchun-1ri and Ubchun-2ri within 1km-3km from Wolsung NPPs during the period from July 2000 to January 2001, as shown in Fig. 1. These were meals which an inhabitant of each location ingests one time in a day.

The samples were sealed in polyethylene zip bags and brought back to the laboratory. They were mixed and shattered in small pieces to guarantee their homogeneity since they consisted of various foods and stored in a freezer at 20 ℃. Tissue free water was extracted from the frozen samples about 500 g by lyophilization. The freeze-dried samples were powdered and subjected to the combustion procedure obtaining OBT fraction in water form. The combustion apparatus is consisted of three parts; an oxygen combustion bomb (model 1121, Parr Instrument Co.), an oxygen and nitrogen gas supply system and a freezer, as shown in Fig. 2.

The combustion is performed under high-pressured oxygen atmosphere of 300 psi in a moment. Organically bound tritium (OBT) was obtained from the inner bottom of the combustion bomb in water form. The water as

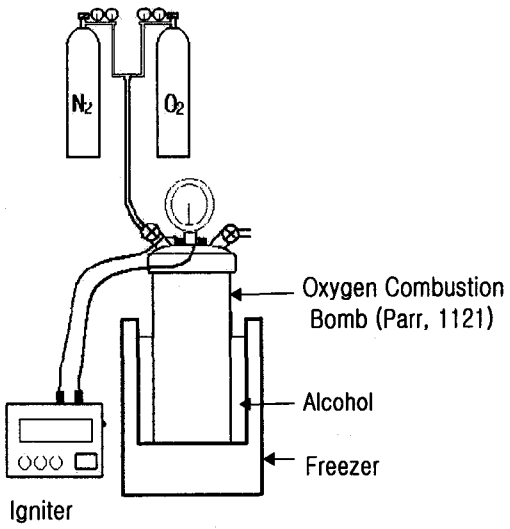

Fig. 2. Combustion apparatus

Table 1. Concentration of TFWT in total diet for inhabitants near Wolsung NPPs.

(Unit: Bq/L)

Location	Month	2000 year			2001 year	Average
		Jul.	Sep.	Nov.	Jan.	
31574.Naa	1	5.85±0.73	12.2±1.0	5.34±0.73	3.19±0.79	9.37±3.74
	2	9.20±0.81	15.1±1.0	10.9±1.0	15.0±1.2	
	3	7.05±0.91	9.05±0.84	8.01±0.91	11.5±1.0	
	Ave.	7.37±1.70	12.1±3.0	8.08±2.78	9.90±6.02	
Nasan	1	38.4±1.6	9.85±0.96	8.70±0.81	10.8±1.0	11.1±9.8
	2	20.7±1.3	7.25±0.93	4.55±0.70	3.92±0.83	
	3	12.3±1.0	5.10±0.69	7.25±.86	4.29±0.71	
	Ave.	23.8±13.3	7.40±2.38	6.83±2.11	6.34±3.87	
Ubchun1	1	14.7±1.2	15.3±1.0	7.90±0.86	7.32±0.93	12.6±6.8
	2	9.60±0.95	7.50±0.92	14.9±1.2	9.55±0.91	
	3	16.2±1.3	31.1±11.6	6.41±0.92	10.3±1.0	
	Ave.	13.5±3.46	18.0±12.0	9.73±4.53	9.06±1.55	
Ubchun2	1	8.14±0.92	8.75±0.92	4.31±0.84	7.05±0.94	7.07±1.33
	2	8.83±0.85	7.76±0.93	6.76±0.92	6.45±0.91	
	3	6.41±0.93	8.24±0.86	6.25±0.96	5.86±0.87	
	Ave.	7.79±1.25	8.25±0.50	5.77±1.29	6.45±0.60	
Bonggil1	1	23.9±1.3	9.25±0.94	8.61±0.91	8.65±0.92	14.5±9.8
	2	15.8±1.1	8.40±0.93	12.7±1.2	11.4±0.98	
	3	42.2±1.6	8.63±0.87	11.6±1.1	13.1±1.1	
	Ave.	27.3±13.5	8.76±0.44	11.0±2.1	11.1±2.2	
Bonggil2	1	40.4±1.6	9.25±0.91	8.42±0.85	12.9±1.2	15.6±12.4
	2	35.2±1.5	8.43±0.89	9.61±0.88	5.75±0.97	
	3	31.3±1.5	8.62±0.90	6.90±0.95	10.3±1.0	
	Ave.	35.6±4.57	8.76±0.43	8.31±1.36	9.65±3.62	
Average		19.2±12.8	10.5±5.7	8.28±2.82	8.74±3.44	11.7±8.46

a TFWT fraction trapped in a cold trap of lyophilization was purified by distillation after the addition of potassium permanganate and sodium peroxide. The water as an OBT fraction obtained from combustion was neutralized with sodium peroxide after the addition of potassium permanganate and purified by distillation. 8 ml of purified water was put into a Teflon-coated plastic vial (model 1200-422, PerkinElmer Co.) and mixed well with 12 ml of a liquid scintillator (Optiphase HiSafe 3, PerkinElmer Co.) to make a total volume of 20 ml.

^{14}C was gotten from CO_2 gas made by combustion using an ammonia solution. The CO_2 gas taken in the ammonia solution was changed for $CaCO_3$ precipitation using $CaCl_2$. The precipitation was dried. The CO_2 in the form of $CaCO_3$ was regenerated by adding extra grade HNO_3 solution and then collected with the 10:10 cocktail solution of carbon absorber (Carbo-Sorb E, PerkinElmer Co.) and the liquid scintillator (Permafluor E+, PerkinElmer Co.) in 20 ml Teflon-coated plastic vial.

The ^{3}H and ^{14}C in the counting samples were measured by a liquid scintillation counter (Quantlus 1220, Wallac,). After the samples stood for about 24 hour in the counter system, ^{3}H and ^{14}C in the samples were measured for

Table 2. Concentration of OBT in totaldiet for inhabitants near Wolsung NPPs.

(Unit: Bq/L)

Location \ Month		2000 year			2001 year	Average
		Jul.	Sep.	Nov.	Jan.	
Naa	1	9.55±0.83	11.6±1.0	11.4±1.0	7.50±0.86	8.00±3.33
	2	5.70±0.84	4.72±0.71	13.5±1.1	10.8±1.0	
	3	3.78±0.83	5.05±0.76	4.25±.82	8.12±0.83	
	Ave.	6.34±2.94	7.12±3.88	9.72±4.85	8.80±1.75	
Nasan	1	29.8±1.5	25.4±1.4	14.2±1.2	12.7±1.3	15.2±8.4
	2	25.8±1.5	15.4±1.2	18.3±1.3	4.23±0.69	
	3	10.7±1.0	7.20±.91	13.8±1.2	4.49±0.79	
	Ave.	22.1±10.1	16.0±9.1	15.4±2.5	7.14±4.82	
Ubchun1	1	18.8±1.2	12.3±1.1	13.4±1.1	29.9±1.5	16.9±8.4
	2	14.0±1.1	1.00±0.46	15.8±1.3	9.55±0.94	
	3	15.4±1.2	19.2±1.3	31.9±1.5	21.2±1.2	
	Ave.	16.1±2.5	10.8±9.2	20.4±10.1	20.2±10.2	
Ubchun2	1	22.0±1.3	9.75±0.92	12.5±1.0	14.0±1.1	15.1±7.8
	2	5.55±0.78	28.3±1.5	26.4±1.4	8.61±0.92	
	3	5.70±0.83	10.1±1.0	17.2±1.3	21.2±1.3	
	Ave.	11.1±9.5	16.1±10.6	18.7±7.1	14.6±6.3	
Bonggil1	1	17.5±1.2	11.6±1.1	10.1±0.98	14.1±1.2	16.1±10.8
	2	7.71±0.92	4.55±0.74	11.3±1.0	34.6±1.6	
	3	34.6±1.6	3.34±.72	15.2±1.2	28.6±1.5	
	Ave.	19.9±13.6	6.50±4.46	12.2±2.7	25.8±10.5	
Bonggil2	1	39.4±1.6	7.35±0.76	8.50±0.91	13.6±1.3	18.1±10.1
	2	27.5±1.5	13.6±1.1	11.7±1.0	21.8±1.5	
	3	30.2±1.5	7.55±0.84	15.2±.2	21.2±1.4	
	Ave.	32.4±6.2	9.50±3.6	11.8±3.4	18.9±4.6	
Average		18.0±11.1	11.0±7.4	14.7±6.2	15.9±9.0	14.9±8.8

300 min (30 min x 10 repeats) and 600 min, (30 min x 20 repeats), respectively.

RESULTS AND DISCUSSION

1. Concentration of TFWT, OBT and ^{14}C in Total Diet

Table 1 and 2 show the TFWT and OBT concentrations in total diet for inhabitants near Wolsung NPP. During the period of July 200 to January 2001, the TFWT concentrations in total samples were in the range of 3.19-42.2 Bq/L and the average was 11.7 Bq/L. The TFWT concentrations in sampling locations Naa-ri, Nasan-ri, Ubchun-1ri, Ubchun-2ri, Bonggil-1ri and Bonggil-2ri were in the range of 3.19-15.1, 3.92-38.4, 6.41-31.1, 4.31-8.83, 8.40-42.2 and 5.75-40.4 Bq/L, respectively.

The OBT concentrations in total samples were in the range of 1.00-39.4 Bq/L and the average was 14.9 Bq/L. The OBT concentrations were in the range of 3.78-13.5, 4.23-29.8, 1.00-31.9, 5.55-28.3, 3.34-34.6 and 7.35-39.4 Bq/L for the sampling locations Naa-ri, Nasan-ri, Ubchun-1ri, Ubchun-2ri, Bonggil-1ri and Bonggil-2ri, respectively.

TFWT and OBT concentrations of total diet studied in this work are lower than the concentrations of the major food sources to Korean such as rice, Chinese cabbage, radish, and green onion.[6] According to the annual report of Ontario Hydro Nuclear, TFWT

concentrations in 1993 terrestrial biota samples from such food as tomatoes, carrots, apples, etc., collected around Pickering NGS, Canada, were in the range of 207-1,009 Bq/L.[7]

Fig. 3 shows the average of TFWT concentrations in total diet at sampling locations. The monthly average of TFWT concentrations at Bonggil is slightly higher than other sampling locations. Fig. 4 shows the monthly average of TFWT concentrations in total diet on sampling months. The monthly average of TFWT concentrations on July 2000 is higher than other months.

Table 3 shows ^{14}C concentrations in total diet for inhabitants near Wolsung NPP. The ^{14}C concentrations in total samples were in the range of 0.230-0.856 Bq/gC and the average was 0.355 Bq/gC. The ^{14}C concentrations in sampling locations Naa-ri, Nasan-ri, Ubchun-1ri, Ubchun-2ri, Bonggil-1ri and Bonggil-2ri were in the range of 0.230-0.500, 0.238-0.835, 0.253-0.469, 0.244-0.856, 0.259-0.671 and 0.251-0.559 Bq/gC, respectively.

^{14}C concentration of total diet studied in this work is slightly higher than the concentration of the major food sources collected at the vicinity of Wolsung site such as rice, Chinese cabbage, radish, and green onion in 1996.[6] This concentration was slightly lower than the range of 0.290-0.485 Bq/gC in 1993 terrestrial biota samples collected vicinity of BNPD facilities.[7]

Fig. 5 shows the average of ^{14}C concentrations in total diet at sampling locations. The average of ^{14}C concentrations at Nasan is slightly higher than other sampling location. Fig. 6 shows the monthly average of ^{14}C concentrations in total diet on sampling months. The monthly average of ^{14}C

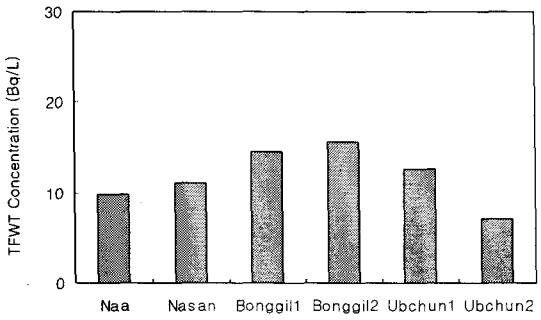

Fig. 3. Concentrations of TFWT in total diet at sampling locations around Wolsung NPPs.

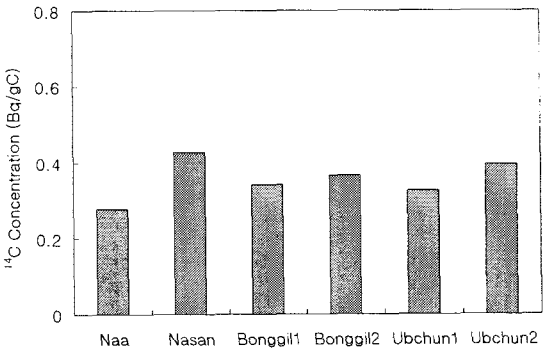

Fig. 5. Concentrations of ^{14}C in total diet with sampling locations around Wolsung NPPs.

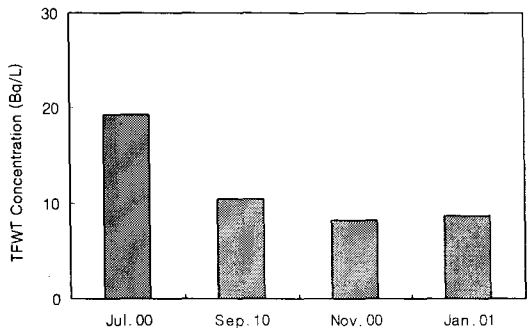

Fig. 4. Concentrations of TFWT in total diet around Wolsung NPPs on sampling month

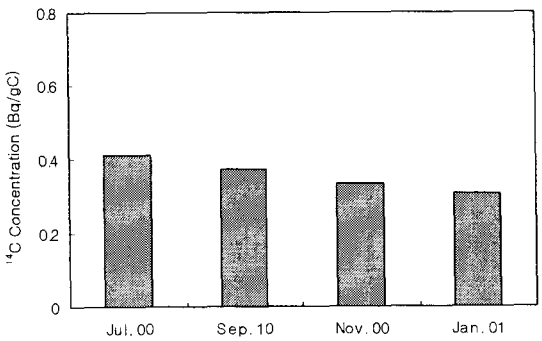

Fig. 6. Concentrations of ^{14}C in total diet around Wolsung NPPs with sampling month.

Table 3. Concentration of ^{14}C in total diet for inhabitants near Wolsung NPPs.

(Unit: Bq/gC)

Location	Month	2000 year			2001 year	Average
		Jul.	Sep.	Nov.	Jan.	
Naa	1	0.232±0.004	0.500±0.005	0.259±0.004	0.251±0.004	0.277±0.071
	2	0.250±0.004	0.230±0.004	0.315±0.004	0.283±0.004	
	3	0.241±0.004	0.231±0.004	0.266±.004	0.265±0.004	
	Ave.	0.241±0.009	0.320±0.155	0.280±0.031	0.267±0.016	
Nasan	1	0.835±0.006	0.393±0.004	0.263±0.004	0.364±0.004	0.427±0.167
	2	0.613±0.005	0.608±0.005	0.276±0.004	0.372±0.004	
	3	0.395±0.004	0.396±0.004	0.238±0.004	0.368±0.004	
	Ave.	0.614±0.220	0.465±0.123	0.259±0.190	0.368±0.004	
Ubchun1	1	0.264±0.004	0.424±0.005	0.389±0.004	0.253±0.004	0.326±0.075
	2	0.374±0.004	0.259±0.004	0.277±0.004	0.258±0.004	
	3	0.392±0.004	0.469±0.005	0.301±0.004	0.253±0.004	
	Ave.	0.343±0.069	0.384±0.110	0.322±0.059	0.255±0.003	
Ubchun2	1	0.760±0.006	0.856±0.006	0.244±0.004	0.266±0.004	0.395±0.202
	2	0.315±0.004	0.316±0.004	0.566±0.005	0.259±0.004	
	3	0.301±0.004	0.323±0.004	0.268±0.004	0.269±0.004	
	Ave.	0.459±0.262	0.498±0.310	0.359±0.179	0.265±0.005	
Bonggil1	1	0.266±0.004	0.344±0.004	0.273±0.004	0.294±0.004	0.342±0.111
	2	0.295±0.004	0.301±0.004	0.671±0.006	0.263±0.004	
	3	0.433±.005	0.259±0.004	0.383±0.004	0.325±0.004	
	Ave.	0.331±0.089	0.301±0.042	0.443±0.205	0.294±0.031	
Bonggil2	1	0.559±0.005	0.266±0.004	0.277±0.004	0.473±0.005	0.365±0.096
	2	0.429±0.005	0.260±0.004	0.336±0.004	0.283±0.004	
	3	0.426±.005	0.251±0.004	0.422±0.004	0.399±0.004	
	Ave.	0.471±0.076	0.259±0.008	0.345±0.073	0.385±0.096	
Average		0.410±0.177	0.371±0.160	0.335±0.117	0.306±0.064	0.355±0.135

concentrations on summer is higher than other season.

2. Annual Internal Dose by TFWT, OBT and ^{14}C in Total Diet

The annual effective dose with tritium and ^{14}C in total diet for inhabitants near Wolsung NPPs is calculated as follows.

$E_{TFWT} = I_{day} \times C_{TFWT} \times R_W \times 365days \times e_{TFWT}$

$E_{OBT} = I_{day} \times C_{OBT} \times R_O \times R_d \times 365days \times e_{OBT}$

$E_C = I_{day} \times C_C \times R_C \times (1 - R_W) \times 1000 \times 365days \times e_C$

Where, E_{TFWT} : annual effective dose of TFWT (Sv/y),

E_{OBT} : annual effective dose of OBT (Sv/y),

E_C : annual effective dose of ^{14}C (Sv/y),

I_{day} : weight of daily ingestion (kg/day),

C_{TFWT} : TFWT concentration (Bq/L),

C_{OBT} : OBT concentration (Bq/L),

C_C : 14C concentration (Bq/gC),

e_{TFWT} : dose conversion factor of

TFWT(Sv/Bq),

e_{OBT} : dose conversion factor of OBT (Sv/Bq),

e_C : dose conversion factor of ^{14}C (Sv/Bq),

R_W : collected TFWT volume (ml)/ fresh sample weight (g),

R_O : collected OBT volume(ml)/dried sample weight(mg),

R_d : dried sample weight(g)/fresh sample weight(g),

R_C : collected C weigh (mg)/dried sample weight (mg).

The dose conversion factors of TFWT, OBT and ^{14}C are used to 1.8×10^{-11}, 4.2×10^{-11} and 5.8×10^{-10} Sv/Bq reported in IAEA Safety Series No. 115. The calculated annual effective doses with TFWT, OBT and ^{14}C are 6.10×10^{-5} mSv/y, 3.71×10^{-5} mSv/y and 7.08×10^{-3} mSv/y, respectively. And then annual internal dose with total diet for inhabitants near Wolsung NPPs is about 7.18×10^{-3} mSv/y, which is about 0.72% of annual effective dose limit 1 mSv/y.

CONCLUSIONS

The TFWT, OBT and ^{14}C concentrations in total diet for inhabitants near Wolsung NPPs during the period of July 200 to January 2001 is investigated. The TFWT concentrations in total samples were in the range of 3.19-42.2 Bq/L and the average was 11.7 Bq/L. The OBT concentrations were in the range of 1.00-39.4 Bq/L and the average was 14.9 Bq/L.

The calculated annual effective dose with TFWT, OBT and ^{14}C is 6.10×10^{-5} mSv/y, 3.71×10^{-5} mSv/y and 7.08×10^{-3} mSv/y, respectively. And then annual internal dose with total diet for inhabitants near Wolsung NPPs is about 7.18×10^{-3} mSv/y, which is about 0.72% of annual effective dose limit 1 mSv/y.

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