C-14 in Rice Scenario

IAEA-EMRAS Tritium/C-14 Working Group

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1. BACKGROUND

Carbon-14 (^{14}C) is one of the major radionuclides released from the nuclear fuel cycle into the environment. It has long been noted that the relatively long half-life of ^{14}C (5,730 years), together with its mobility in the environment and incorporation into man via the food-chain, leads to the long-term homogeneous irradiation of the global population. In addition, ^{14}C discharges from nuclear facilities probably result in the enhancement of ^{14}C levels in biogenic materials in the vicinity and subsequent excess exposure of local population. The development and application of functional mathematical models for predicting short-range ^{14}C transfer in the terrestrial environment is therefore necessary to assess the local radiological impact of the anthropogenic ^{14}C.

The Tokai reprocessing plant (TRP) of Japan Nuclear Cycle Development (JNC) started hot test in September 1977. The total amount of 1,000 tons of spent fuels used at boiling water reactors (BWRs), pressurized water reactors (PWRs) and an advanced thermal reactor (ATR), has been successfully reprocessed as of June 2002. The JNC has conducted careful monitoring for ^{14}C in airborne discharge via 90-m stacks from the TRP, and ^{14}C in atmospheric CO_2 and rice grain (Japanese daily diet) collected around the TRP site. The monitoring data over ten years from 1991 to 2001 would be useful for model-data intercomparison studies on the regional ^{14}C transfer in the environment.

2. SITE DESCRIPTION

Tokai-mura is basically a flat land of agriculture such as rice plant, vegetable and fruit plants. An overview of the Tokai-mura is shown in **Fig. 2.1**. The population of Tokai-mura is about 35,400 as of 2000. The Tokai reprocessing plant of JNC is located in the east end of the Tokai-mura (the longitude 140.6^{o} E, latitude 36.5^{o} N), facing the Pacific Ocean.

3. DISCHARGE MONITORING

3.1. Discharge sources

Carbon-14 has been discharged from three stacks (called “Main stack”, “Sub-1 stack” and “Sub-2 stack”, respectively) of the TRP with gaseous effluent. Dominant sources of ^{14}C discharged from the Main stack are shearing and dissolution of the spent fuel and following solvent extraction processes for separating uranium and plutonium from fission products. Carbon-14 released from the Sub-1 and Sub-2 stacks are mainly from the bituminization of low active level liquid waste and the vitrification of high active level liquid waste, respectively.

The Sub-1 and Sub-2 stacks were located on about 210 m east-northeast and 35 m southwest from the Main stack, respectively. All the stacks had physical height of 90 m, and the release points of them were situated on 96 m above the sea-level. The diameters of outlets (release points) of the stacks were 2.9 m for the Main stack, 2.4 m for the Sub-1 stack and 2.8 m for the Sub-2 stack, respectively. Gaseous effluents including ^{14}C were continuously released from the stacks at exhaust rates listed in **Table 3.1**, but ^{14}C concentrations were less than the authorized detectable limit, 40 Bq cm^{-3}, during non-reprocessing. Almost all (more than 97%) of ^{14}C is proved to be discharged as CO_2 form during reprocessing[(1)].

The effective height of the release is not always the same as the actual physical height of the release point. The upward momentum of released effluents will tend to make the effective height greater than the actual height of the release. The effective plume rise should be simply calculated by applying mean wind speed to a following equation:

$$H = \frac{3 \cdot W \cdot D}{U} \qquad (1)$$

where H is the effective plume rise (m), W the speed of exhaust air at stack (m s^{-1}), D the diameter of the stack outlet (m), and U the mean wind speed at top of the stack (m s^{-1}). The effective height of the release can be therefore derived from the physical stack height + effective plume rise. Air temperatures of stack discharges are also provided in **Tables A.1-A.6** in **APPENDICES**.

3.2. Brief description of monitoring method

The monitoring has been accomplished by sampling airborne effluent from the stack before discharge. A portion of the airborne effluent was introduced into the ^{14}C sampler at a flow rate of 0.4 l min^{-1} for a week. Hydrocarbons and carbon monoxide were catalytically converted into chemical form of CO_2. All carbon was then absorbed as CO_2 in 200 ml of monoethanolamine (MEA; 2-aminoethanol) in a bubbler-type trap after passing through a dehumidifier to remove water vapor in the air. Preliminary experiment demonstrated that the CO_2 absorption efficiency of the trap is almost 100%. At the end of the given sampling period, an aliquot of the MEA was mixed with liquid scintillator and methanol. The ^{14}C activity of the prepared sample was measured using a liquid scintillation counter with 180 min measurement time. The concentration of ^{14}C in the airborne effluent was evaluated using the result of activity measurement and some parameters such as volume of MEA used for sample preparation and the total volume of air collected. The ^{14}C activity discharged in the given period was estimated by multiplying the ^{14}C concentration by air throughput of the stack[(2)]. Weekly monitoring has been accomplished typically by Wednesday-Wednesday sampling.

3.3. Data of ^{14}C discharge rates

Reporting the monitoring data of ^{14}C in airborne effluent from the TRP was officially started in October 1991 for Main and Sub-1 stacks. As for Sub-2 stack, data of ^{14}C in airborne release has been reported since September 1994. The numerical data of weekly monitoring from October 1991 to December 2001 are provided in **Tables B.1-B.6** in **APPENDICES** [(3)].

Fig. 3.1 shows annual discharge rates of ^{14}C from three stacks. It should be noted that the fire and explosion accident at the bituminization demonstration facility on March 1997 stopped the operation of the TRP until July 2000. Variations in monthly atmospheric ^{14}C discharges from three stacks are plotted in **Figs. 3.2-3.4**, respectively. The monthly discharge from the TRP (total of three stacks) is also shown in **Fig. 3.5**. The annual ^{14}C discharge amount was always considerably lower than the authorized discharge limit in the Safety Regulation for the TRP.

4. ENVIRONMENTAL MONITORING

4.1. Monitoring items and locations

Environmental monitoring of ^{14}C has been made mainly for two items: (1) atmospheric ^{14}C in CO_2 form; and (2) ^{14}C in polished rice grain. The atmospheric $^{14}CO_2$ has been sampled every month at five sampling locations. Polished rice grain has been collected at three locations around the TRP in the harvest season of rice. A location map for collecting $^{14}CO_2$ and rice grain samples is presented in **Fig. 4.1**. Detailed information on the sampling locations and available data is summarized in **Table 4.1**. Longitudinal and latitudinal data of discharge sources and sampling locations are also presented in **Table 4.2**. The sampling points of ST-1 and R-1 are located on about 0.6 km west north-west and about 2.1 km west-southwest from the Sub-1 stack, respectively.

4.2. Brief descriptions of monitoring methods

A CO_2 sampler was used to monitor $^{14}CO_2$ concentration in the atmosphere. In the sampler, ^{14}C in CO_2 form was absorbed in NaOH solution with stable CO_2 in a monthly monitoring basis, which made $CaCO_3$ precipitate with NH_4Cl and $CaCl_2$ after sampling. The precipitate was decomposed with H_3PO_4 to re-generate CO_2. The resulting CO_2 was converted to C_2H_2 in a reaction of metallic lithium with water, and then C_6H_6 was synthesized by polymerization of C_2H_2. The C_6H_6 was mixed with liquid scintillator for measuring ^{14}C activity (Bq/gC) by liquid scintillation counting. The error of ^{14}C measurement was normally ± 0.003 Bq/gC. The error presents one standard deviation associated with radiation counting.

Rice grain was completely combusted in a pressurized combustion chamber filled with oxygen, in which CO_2 was produced. The resulting CO_2 was converted to C_6H_6 and analyzed for ^{14}C activity (Bq/gC) by the same manner as $^{14}CO_2$ measurement described above. The error of ^{14}C activity was within ± 0.003 Bq/gC. Carbon-14 activity per unit weight of rice (Bq/kg-raw) also could be calculated by using a factor 0.41 that is carbon content (gC/g-raw) in rice.

4.3. Carbon-14 in polished rice grain in 1991

The ^{14}C activities of rice grain samples collected in 1991 showed no or fewer

enhancements above the background level in Japan, although some amounts of ^{14}C had been discharged into the atmosphere from the TRP. This implies that the accumulation of ^{14}C discharged in the past on the paddy field would be apparently negligible in starting point of the model calculation (1991) in this scenario.

5. METEOROLOGICAL MONITORING

The meteorological monitoring was made at ten minutes interval for following items:

(1) Wind direction and speed (m s^{-1}) at 10 m above the ground;
(2) Wind direction and speed (m s^{-1}) at the top of stack, 90 m above the ground;
(3) Rainfall (mm);
(4) Atmospheric temperature (^{o}C) and relative humidity (%) at 1.5 m above ground;
(5) Solar radiation (kW m^{-2}); and
(6) Atmospheric stabilities.

The item (2) was monitored at the top of a meteorological observation tower built on the JNC site. The height is about 100 m above the sea level, corresponding to the physical height of the stacks above the sea level. Other items were measured near the ST-1 point. The meteorological data averaged over hourly periods are available as input data for model calculations.

6. OTHER INFORMATION

6.1. Background level of ^{14}C in Japan

Fuma et al. (2002) reported environmental background ^{14}C levels in Japan in 1990s[4]. They selected grapes as an indicator of ^{14}C levels in the environment, and determined the specific activities of ^{14}C in ethanol extracted from wine made from grapes cultivated in several prefectures or unknown places in Japan. The specific activities of ^{14}C gradually decreased from 0.260 Bq/gC in 1991 to 0.244 Bq/gC in 2000 (**Table 6.1**).

6.2. Other nuclear facilities in Tokai-mura

It should be noted that there are some potential ^{14}C sources in Tokai-mura such as a

BWR (operated from November 1978) and a gas cooled reactor (GCR, operated from July 1966 to March 1998). These reactors are located in about 3-4 km north-northeast from the TRP site (**Fig.2.1**).

6.3. CO_2 concentration

A thermoelectric power plant near the JNC site shown in **Fig.4.1** has been operated from December 2003. Therefore, CO_2 concentration in the air is to be believed in the normal range as those of rural areas in Japan in the period of 1991-2001. As an example, the CO_2 concentration at 30 cm above the ground in Tokai-mura was estimated to be approximately 380-390 ppmv in 2000[5].

6.4. Management of a paddy field

A schedule for managing paddy field in Tokai-mura in 1999 is presented in **Table 6.2**. Panorama of paddy field is photographically shown in **Figs. 6.1a-6.1d**. For 1991 to 2001, date of transplanting, flowering and harvesting were 10-15 May, 15-20 August, and 18-25 September, respectively, indicating that the difference in timing of each stage is within 7 days.

6.5. Growth curve of a rice plant

Osaki and Tanaka (1979) investigated growth curves of four components in a rice plant in Hokkaido that is northernmost island of Japan[6]. **Fig. 6.2** shows growth curves determined at four growth stages of a rice plant: (1) vegetative stage; (2) flowering stage; (3) milky stage; and (4) harvest stage. The easily respired-substrate and the stored-substrate were rapidly increased in the reproductive phase. Here, the easily respired-substrate means the photosynthesized-substrate released by respiration within a day after CO_2 assimilation. The stored-substrate is temporarily stored in the plant, but will be respired from the plant by harvesting. The dry weight of stems, leaves and roots had a remarkable increase until milky stage. The dry weight of ears showed a different trend of growth rate, which was sharply increased in the ripening phases. At the harvest stage, the dry weights of harvesting organs (ears) and non-harvesting organs (stems, leaves and roots) were almost same (30 g-dry weight per a stock). A stock is a unit bunch of pieces of rice plant. The numerical data is given in **Table 6.3**.

Kondo (2005) investigated more in detail rice growth curves of four components in a rice plant (koshihikari) in Ibaraki in 2004[7]. In this study, four components include (1) leafs; (2) stems; (3) ear stems; and (4) ears. The rice plant were transplanting on 7 May. **Fig.6.3** shows growth curves determined at 6 growing stages of a rice plant. The numerical data is provided in **Table 6.4**.

6.6. Storage and distribution of assimilated-^{14}C in rice plant

Osaki and Tanaka (1979) also investigated storage and distribution of assimilated-^{14}C in rice plant in Hokkaido[6]. Carbon-14 were fed to rice plants at three different growth stages (vegetative, flowering and milky stages), and then ^{14}C retention ratio and its distribution in three components of the rice plant were determined at the harvest stage. The results are presented in **Fig. 6.4**. The total ^{14}C-retention percentages were approximately 50%, 65% and 90% for ^{14}C-assimilation at vegetative, flowering and milky stages, respectively. The ^{14}C-distribution percentages in ears at the harvest stage were about 7%, 28% and 82%, respectively, when ^{14}C was fed at the vegetative, flowering and milky stages. These data suggest that during ripening photosynthates are efficiently translocated to the harvesting organs (ears).

7. CALCULATION END POINTS

Using the information provided above, please calculate

(1) Monthly mean ^{14}C concentrations (Bq/gC) in air collected at two (three) monitoring points ST-1 and ST-2 (and ST-N) from May to October (i.e. the rice growing season) for 1992 to 1997.

(2) Carbon-14 concentrations (Bq/gC) in rice grains collected at two (three) monitoring points R-1 and R-2 (and R-3) for 1992 to 2001.

(3) 95% confidence intervals on all predictions in (1) and (2) above.

REFERENCES

(1) Koarashi, J., Akiyama, K., Asano, T. and Kobayashi, H. (2005). Chemical composition of

^{14}C in airborne release from the Tokai reprocessing plant, Japan. Radiation Protection Dosimetry, in press.

(2) Koarashi, J., Kozawa, T., Yokota, T., Isaka, K., Akiyama, K. and Asano, T. (2004). Methods and their performances for collecting tritium, carbon-14 and radioiodine in airborne effluent from Tokai reprocessing plant, JNC TN8410 2004-003 (in Japanese).

(3) Koarashi, J. and Fujita, H. (2005). Review of monitoring data (1991-2001) for model-data intercomparison studies on ^{14}C transfer in the environment, JNC TN8450 2004-011.

(4) Fuma, S., Inoue, Y., Miyamoto, K., Takeda, H., Iwakura, T., Arai, K., Kashida, Y. and Ichimasa, Y. (2002). Environmental background ^{14}C levels in Japan in 1990s. Radioisotopes, 51, 381-391 (in Japanese).

(5) Koarashi, J., Amano, H., Andoh, M., Iida, T. and Moriizumi, J. (2002). Estimation of $^{14}CO_2$ flux at soil-atmosphere interface and distribution of ^{14}C in forest ecosystem. Journal of Environmental Radioactivity, 60, 249-261.

(6) Osaki, M. and Tanaka, A. (1979). Current photosynthates and storage substances as the respiratory substrates in the rice plant. Nihon Dojou Hiryougaku zasshi (Japanese Journal of Soil Science and Plant Nutrition), 50, 540-546 (in Japanese).

(7) Kondo, M. (2005). Private communication.

APPENDICES

Table 3.1. Representative exhaust rates of three stacks from 1991 to 2001.

Year	Representative exhaust rate (m^3 h^{-1})		
	Main stack	Sub-1 stack	Sub-2 stack
1991	4.02×10^5	1.28×10^5	-
1992	4.17×10^5	1.22×10^5	-
1993	4.17×10^5	1.22×10^5	-
1994	4.17×10^5	1.31×10^5	1.19×10^5
1995	4.17×10^5	1.28×10^5	1.13×10^5
1996	4.17×10^5	1.31×10^5	1.31×10^5
1997	4.17×10^5	8.93×10^4	1.26×10^5
1998	4.17×10^5	1.01×10^5	1.31×10^5
1999	4.17×10^5	8.93×10^4	1.26×10^5
2000	4.17×10^5	1.01×10^5	1.24×10^5
2001	4.02×10^5	8.93×10^4	1.28×10^5

Table 4.1. Information on sampling locations and available data for 1991-2001.

Item	Location		Distance and direction from Main stack	Available data	Notes
Atmospheric $^{14}CO_2$	ST-1	(In JNC site)	0.5 km northwest	Jan.1991-Feb.1994	
	ST-2	(Funaishikawa)	4.2 km northwest	Apr.1993-Mar.1994	
	ST-3	(Nagasuna)	2.8 km southwest	Jan.1991-Jan.1997	
	ST-4	(Koya)	5.2 km west-southwest	May 1991-Dec.1995	Control
	ST-N	(Naka-machi)	14.6 km west-northwest	Jan.1991-June 1996	Control
Polished rice grain	R-1	(Nagasuna)	1.9 km west-southwest	1991-2001	
	R-2	(Terunuma)	1.0 km west	1991-2001	
	R-3	(Nishi-10 km)	11.8 km west	1991-2001	Control

Table 4.2. Longitudinal and latitudinal data of discharge sources and sampling locations.

Item	Location	Longitude	Latitude	Notes
Discharge sources	Main stack	36° 26' 36.1" N	140° 36' 19.0" E	
	Sub-1 stack	36° 26' 38.5" N	140° 36' 26.8" E	
	Sub-2 stack	36° 26' 35.2" N	140° 36' 18.1" E	
Atmospheric $^{14}CO_2$	ST-1	36° 26' 46.1" N	140° 36' 05.6" E	
	ST-2	36° 28' 20.2" N	140° 34' 33.7" E	
	ST-3	36° 25' 24.0" N	140° 35' 10.7" E	
	ST-4	36° 25' 37.7" N	140° 32' 55.3" E	Control
	ST-N	36° 28' 27.2" N	140° 26' 41.0" E	Control
Polished rice grain	R-1	36° 26' 11.1" N	140° 35' 10.3" E	
	R-2	36° 26' 26.7" N	140° 35' 39.9" E	
	R-3	36° 26' 08.1" N	140° 28' 19.5" E	Control

Table 6.1. Environmental background ^{14}C levels in Japan in 1990s.

Year	Specific activity of ^{14}C (Bq/gC)
1991	0.261 ± 0.002[a]
1992	0.264 ± 0.005
1993	0.254 ± 0.003
1994	0.255 ± 0.003
1995	0.254 ± 0.004
1996	0.251 ± 0.002
1997	0.250 ± 0.002
1998	0.252 ± 0.003
1999	0.248 ± 0.004
2000	0.244 ± 0.002

[a]Standard deviation (1σ) of the mean.

Table 6.2. A schedule for managing paddy field in 1999 in Tokai-mura.

Date	Growing stage	Days after transplanting	Depth of water (cm)	Dry/wet ratio of soil weight (%)
12 May	Transplanting	-	0-3	50.9
15 July	Midseason drainage	60	2-5	58.2
4 August	Flowering (Early ripening)	80	3-5	-
20 September	Harvest	130	0	-
28 September	After harvest	140	0	74.1

Table 6.3. Growth of a rice plant observed in Hokkaido.

Days after transplanting	Growing stage	Total weight (g-dry/stock)	Weights of four components (g-dry/stock)			
			Ears	Stems, leaves and roots	Easily respired-substrate	Stored-substrate
50	Vegetative	18.2	0.7	8.0	6.2	3.3
70	Flowering	54.1	6.4	23.1	14.8	9.8
100	Milky	78.5	19.2	29.3	17.0	13.0
120	Harvest	91.6	30.0	30.6	17.9	13.1

Table 6.4. Growth of a rice plant observed in Ibaraki.

Days after transplanting	Total weight (g-dry/m^2)	Weights of four components (g-dry/m^2)			
		Leafs	Stems	Ear stems	Ears
49	90	53	37	-	-
62	346	179	167	-	-
74	629	238	391	-	-
82	849	233	569	-	47
97	1152	209	520	-	423
118	1174	140	409	20	605

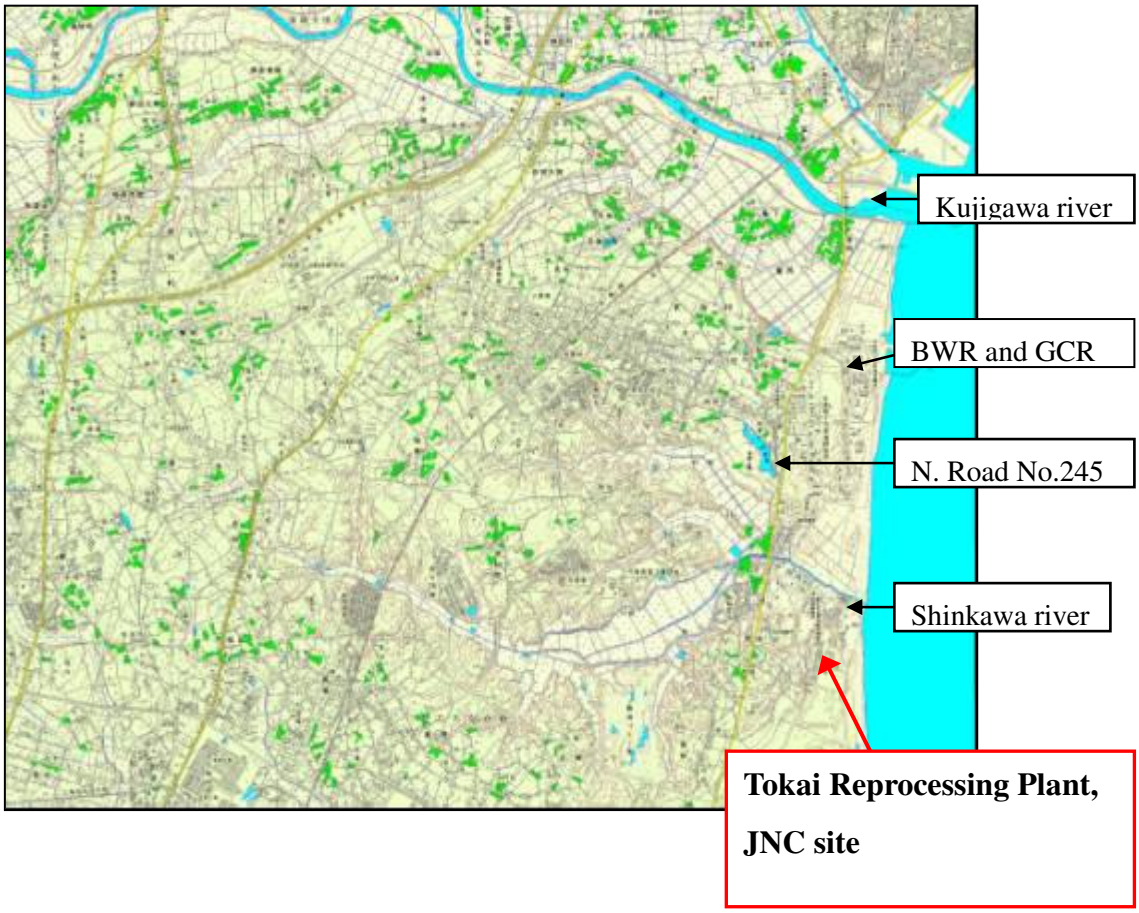

Fig.2.1. An overview of Tokai-mura with the Tokai reprocessing plant.

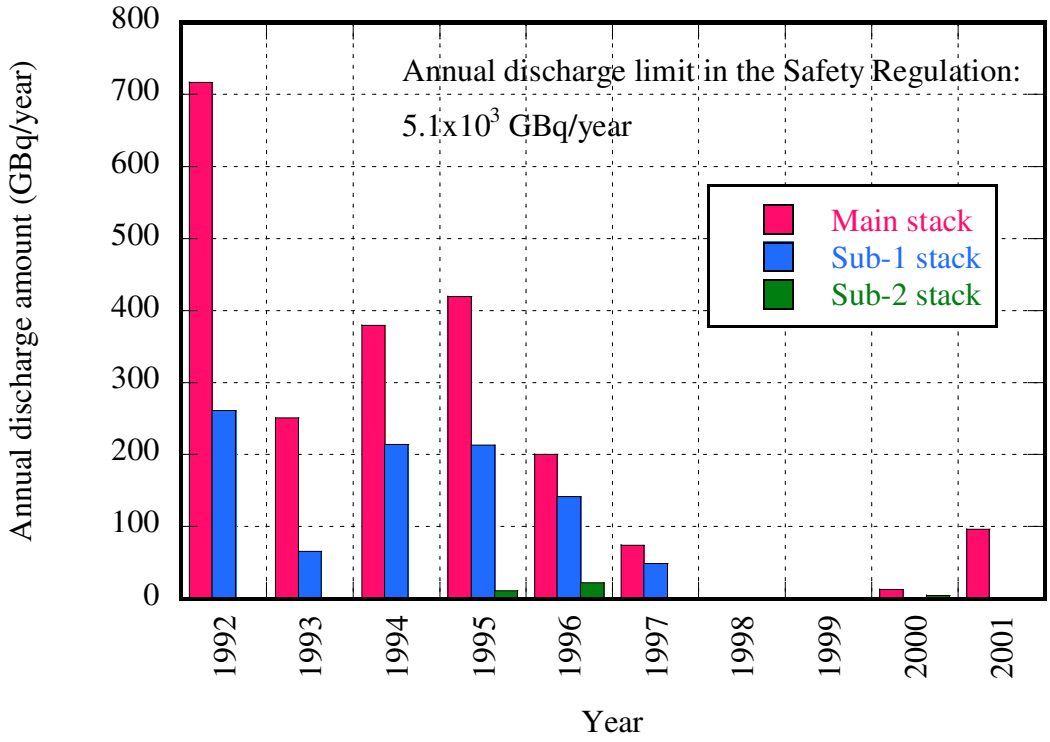

Fig.3.1. Annual discharge rates of ^{14}C from the TRP.

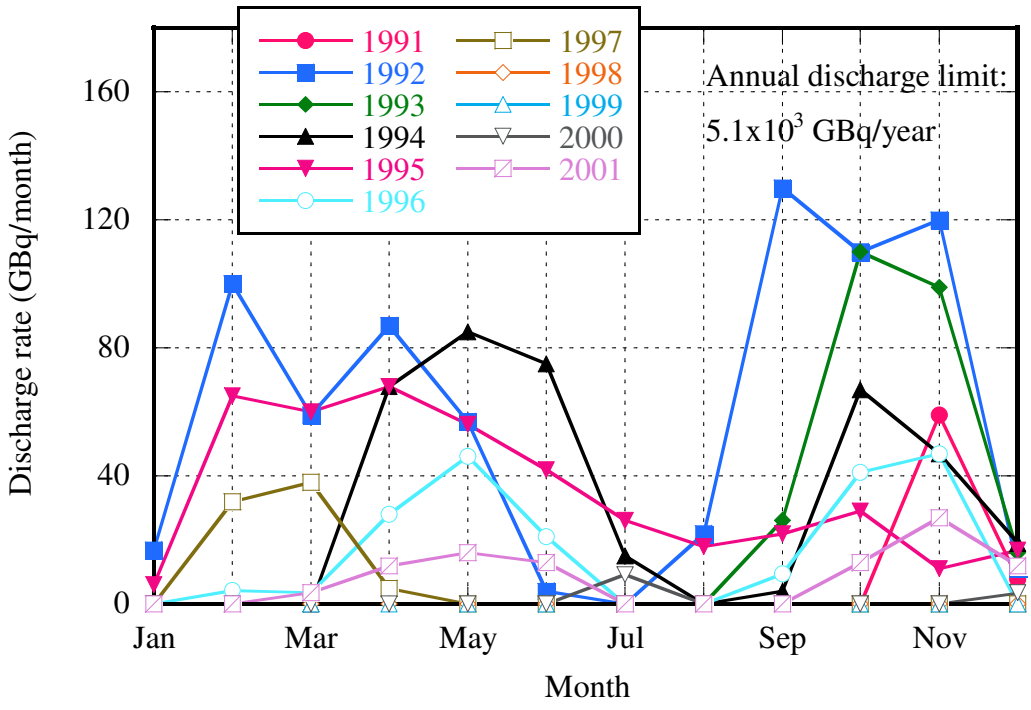

Fig.3.2. Monthly discharge rates of ^{14}C from the Main stack.

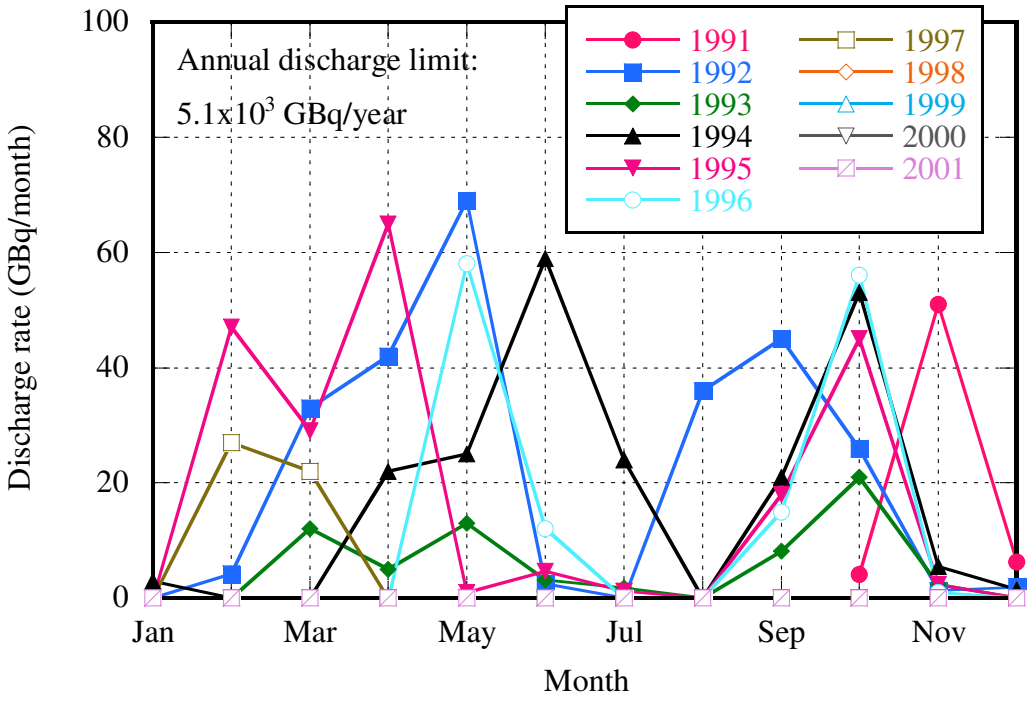

Fig.3.3. Monthly discharge rates of ^{14}C from the Sub-1 stack.

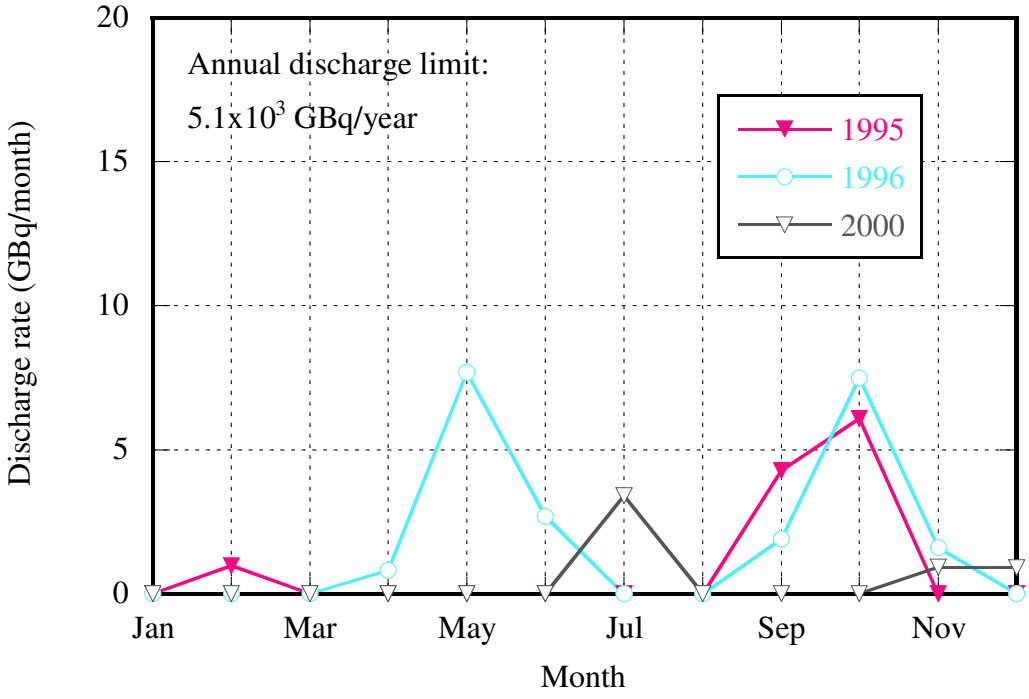

Fig.3.4. Monthly discharge rates of ^{14}C from the Sub-2 stack.

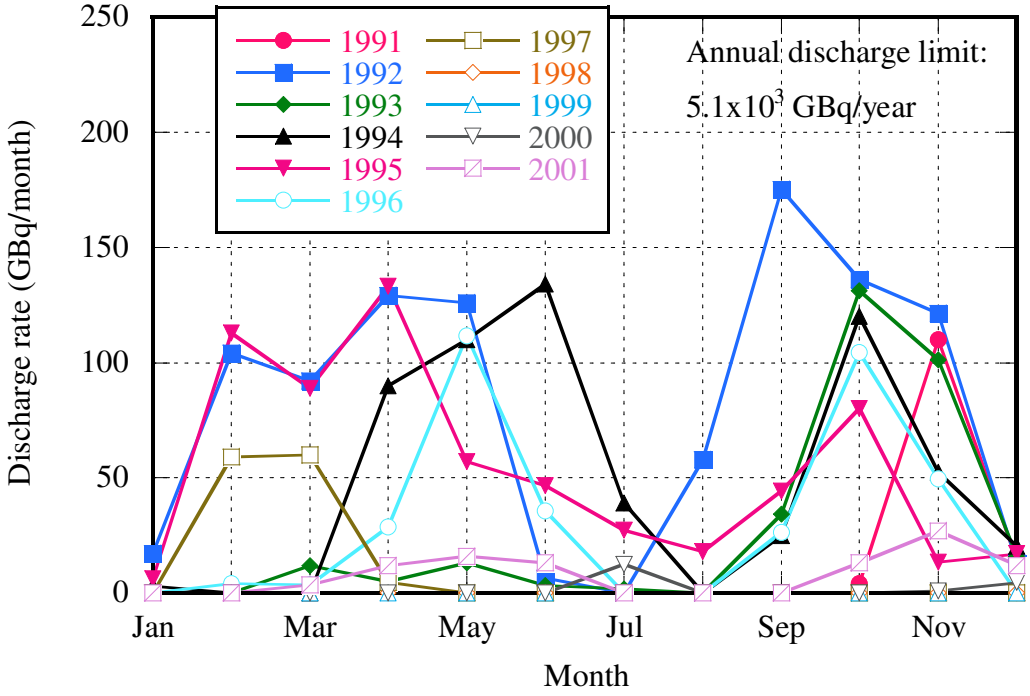

Fig.3.5. Monthly discharge rates of ^{14}C from the TRP.

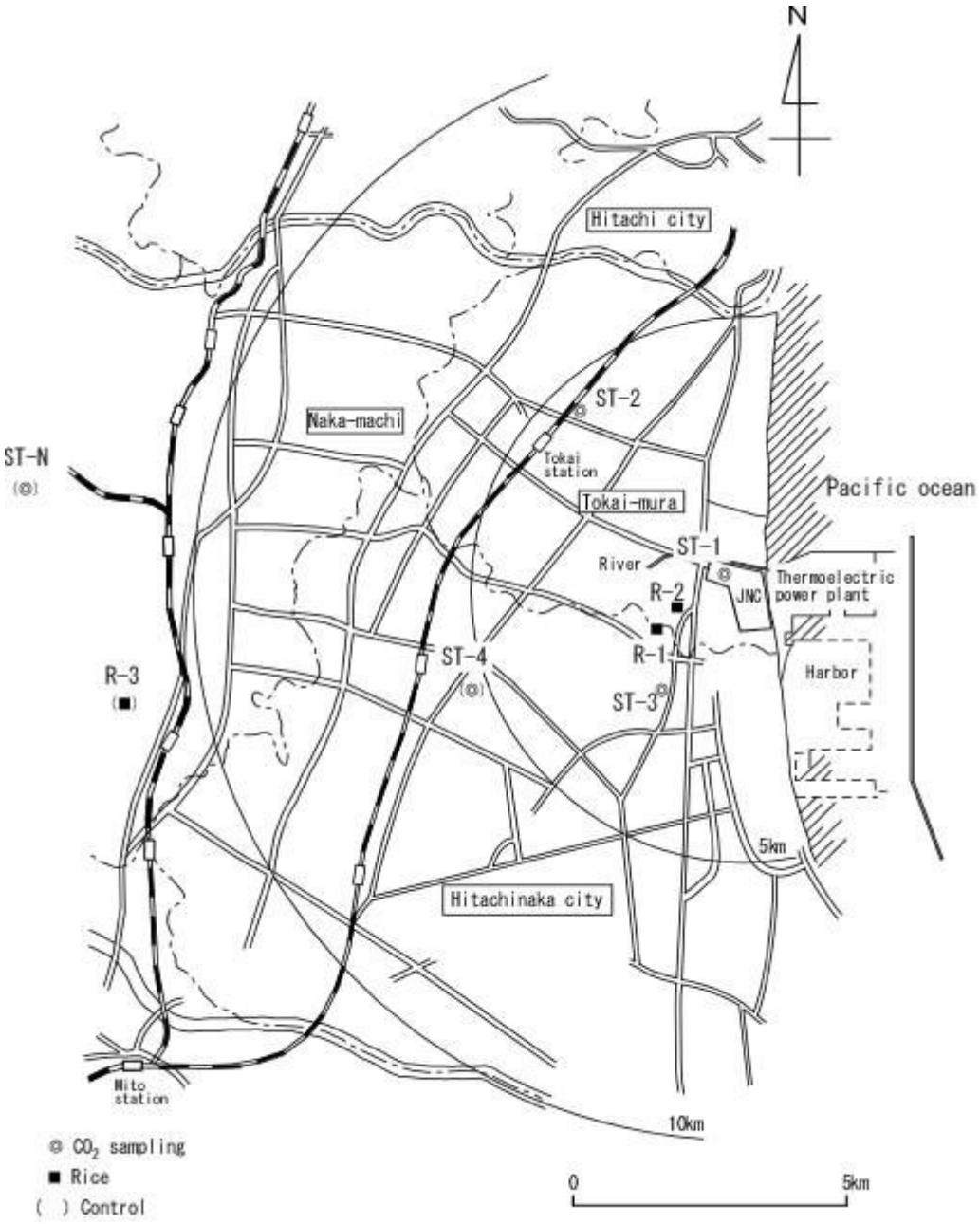

Fig.4.1. Location map for collecting $^{14}CO_2$ and rice grain samples.

Fig. 6.1a. Paddy field before transplanting rice plant (10 May 1999).

Fig. 6.1b. Paddy field after transplanting rice plant (12 May 1999).

Fig.6.1c. Paddy field in flowering stage (3 August 1999).

Fig.6.1d. Paddy field in late ripening phase (13 September 1999).

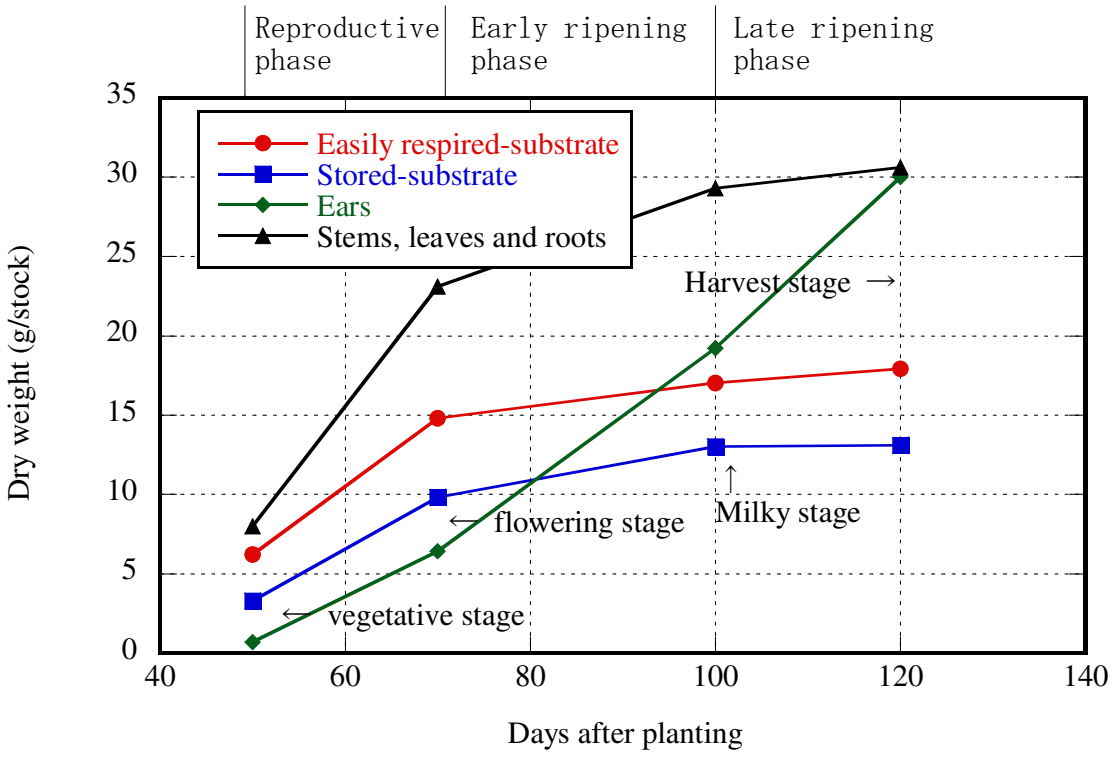

Fig.6.2. Growth curves of four components in rice plant.

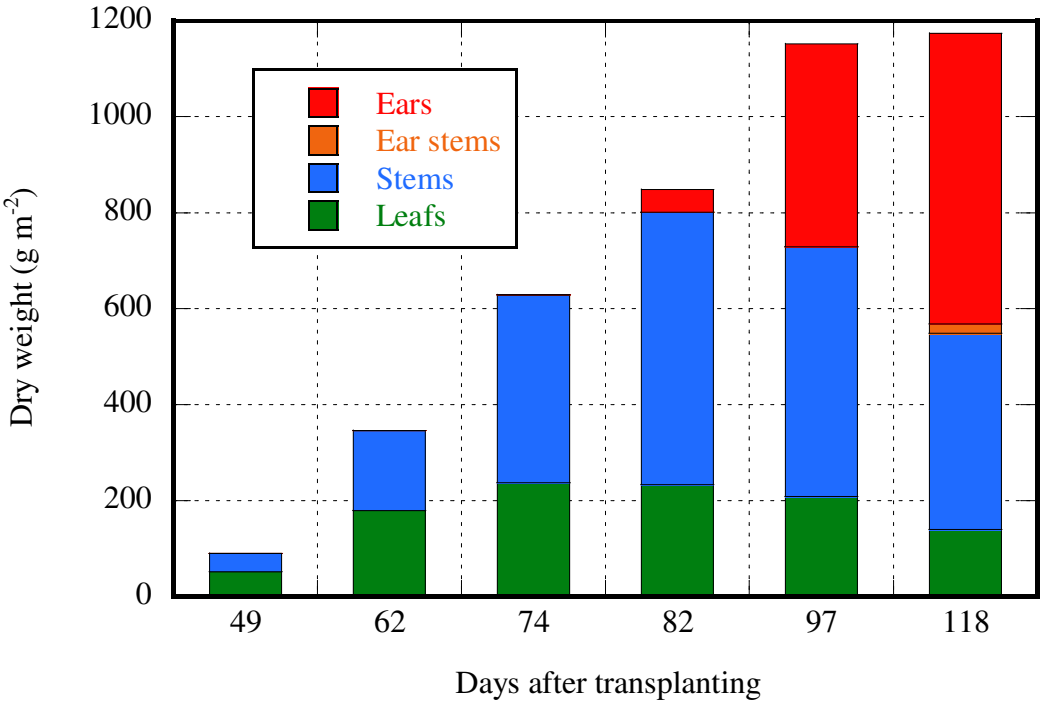

Fig.6.3. Growth of four components in rice plant in Ibaraki.

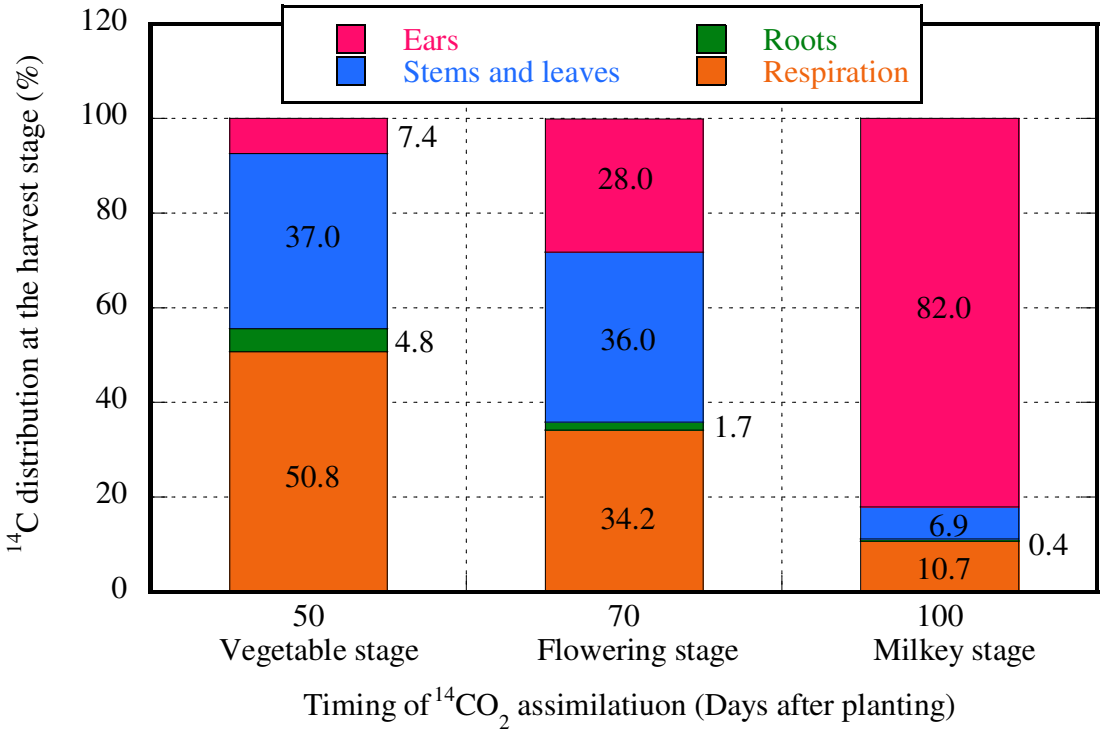

Fig. 6.4. Distribution percentage of assimilated-^{14}C at the harvest stage.