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4.29 MEASUREMENT OF C-14 DISTRIBUTION IN FOREST AROUND NUCLEAR FACILITIES

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ABSTRACT

A simple analytical method of C-14 measurement using fast bomb combustion and liquid scintillation counting (LSC) has been developed for measuring C-14 distribution in the terrestrial environment. Specific activities of C-14 in cedar leaves and soils collected from an area around nuclear facilities and control areas were measured using this method. Depth distribution of Cs-137 in soils was also measured at the same sampling sites and compared with the depth distribution of C-14.

C-14 specific activity in cedar leaves examined around nuclear facilities exceeded that in the control areas by 8 to 30 mBq (g carbon)$^{-1}$. The depth distribution of C-14 in forest soil shows that C-14 has peak values in the top 10 cm of the soil profiles ascribed to the highest bomb C-14 level in the 1960's. The data were made available to assess the behavior of fallout C-14 in the surface environment.

KEYWORDS: C-14, Cs-137, Stable isotope ratio, forest

1. INTRODUCTION

Because of the long radioactive half-life of C-14 (5730 yr), fallout C-14 from former nuclear weapon tests remains in soil and C-14 flux from the soil surface to the atmosphere was observed in a forest [1]. Patterns of C-14 enrichment in soil profiles provide important information for estimating carbon turnover and carbon flux from soil. C-14 is also released from nuclear power plants and nuclear reprocessing plants. Recent increase in the peaceful use of nuclear energy has made monitoring of C-14 in the environment more important.

Many studies have been performed to establish the low-level C-14 specific activity in organic materials using liquid scintillation counting (LSC) and accelerator mass spectrometry (AMS). For LSC, the benzene synthesis method is usually adopted because of its high counting efficiency. However, using benzene synthesis-LSC method requires relatively large samples (several grams of carbon) and complex preparation procedures, and a method using AMS is relatively costly. For these reasons, direct CO_2 absorption and the LSC method have an advantage when many samples must be measured. We have developed a simple analytical method of C-14 measurement using fast bomb combustion and LSC. C-14 specific activities in cedar leaves and soils collected from an area around nuclear facilities and control areas were measured using this method. The depth distribution of Cs-137, total carbon content and stable carbon isotope ratio in soils were also measured at the same sampling sites and compared with the depth distribution of C-14.

2. METHODS

2.1 Sampling and pretreatment of cedar leaves and soils

Soils, litters and cedar leaves were collected at Tokai Village and Satomi Village in 2001 and 2002 in Japan [2, 3]. Fig. 1 shows the location of the sampling points. There are nuclear facilities in Tokai Village, including a nuclear power plant and a spent fuel reprocessing plant. Satomi Village is a control site. Litter was divided into 2 layers. After removal of litters, soil samples were collected at depths of 0~2 and 2~5 cm, then collected in increments of 5 cm up to 60 cm from the soil surface. At the sampling points in Satomi Village, newly fallen leaves were collected as the cedar trees were too high to take their leaves directly. At the sampling points in Tokai Village, tips of cedar leaves were collected.

The samples were dried in an oven to attain constant weight. Litters and cedar leaves were ground into very fine powder. Soil samples were ground and sieved through a 2mm mesh after removal of stones, roots and plant debris by hand picking.

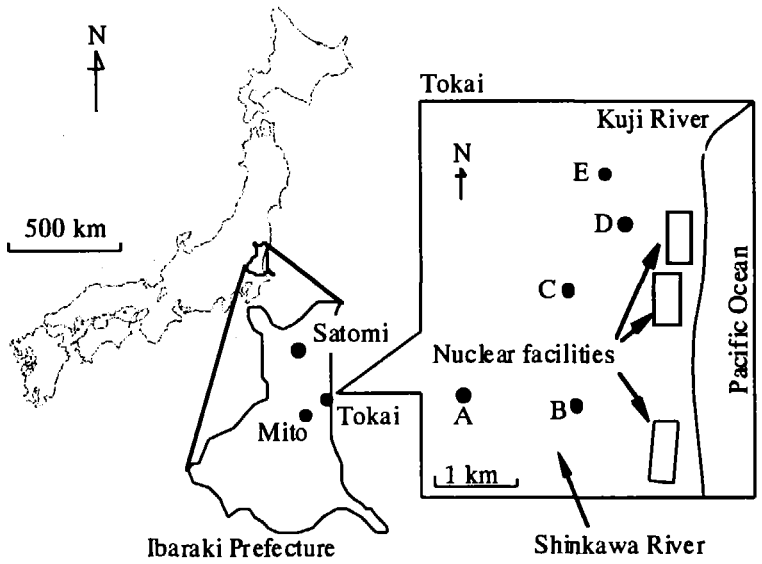

Fig. 1 Location of soil and cedar samples. Points A to E indicate sampling locations in Tokai village.

2.2 Analytical method of C-14 specific activity

C-14 specific activity in plant and soil were measured using fast bomb combustion and LSC [2, 3]. Fig. 2 shows a schematic diagram of the set-up. A sample containing organic material was burned under high oxygen pressure (300 psi) in a fast oxygen combustion bomb apparatus (DELFI SCIENCE INTERNATIONAL, Inc). About 4~56 g samples were burned at a time. A cold trap using liquid nitrogen caught the generated gas mixture, including CO_2. The cold trap was then warmed to room temperature and the trapped gas was passed through a water trap (Drierite, W. A. Hammond Drierite Co. Ltd.) and a CO_2 trap [Molecular Sieve (M.S.) 4A, Union Carbide Co.]. The CO_2 trapped by M.S. 4A was released using a furnace (420 °C) and purged into a CO_2 absorbent material (CARBO-SORB E, Packard Ltd.) mixed with a scintillation cocktail (PERMA FLUORE E^+, Packard Ltd.) with nitrogen gas. After absorption of CO_2, the 20 ml or 100ml mixture of reagents was pipetted into a 20 ml glass vial or 100 ml Teflon vial and weighed. The amount of CO_2 fixed in the absorbent was evaluated by the gravimetric method. The C-14 radioactivity was measured with a low background liquid scintillation counter (LSC-LB III, Aloka) by 500-minute counting. NIST reference material SRM-4990C [oxalic acid with 18.41dpm (g carbon)$^{-1}$] was used to determine the counting efficiency. The average counting efficiency was 60 %. The detection limit in this experimental condition was 20 mBq (g carbon)$^{-1}$, so environmental C-14 levels [about 250 mBq (g carbon)$^{-1}$] could be evaluated by this method.

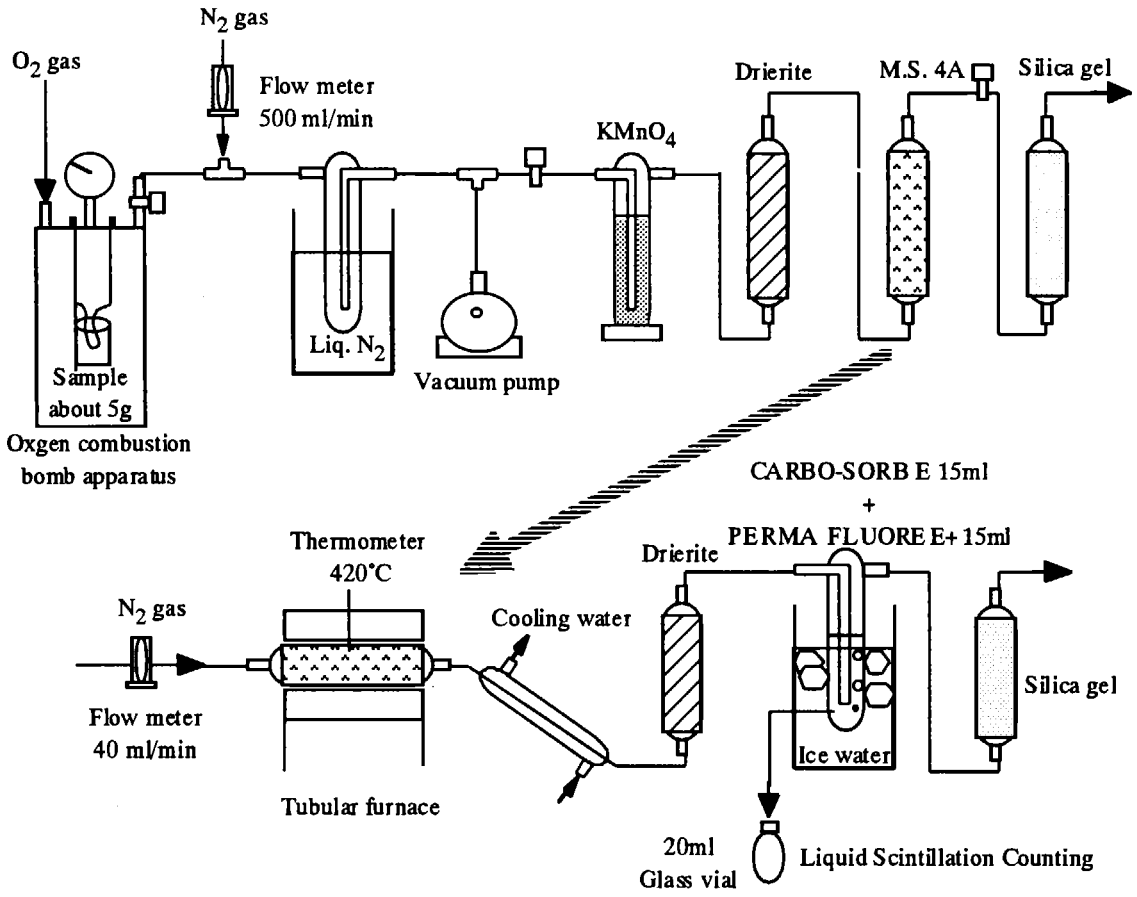

Fig. 2 Schematic diagram of experimental set up [2]

2.3 Carbon stable isotope ratio and carbon content measurement

Less than 10 mg of samples were sealed into tin capsules and combusted with an on-line sample preparation system, i.e. elemental analyzer (NCS-2500, Thermo Quest) linked through a gas purification system to an isotope ratio mass spectrometer (Isoprime-EA, Micromass Ltd). The data are expressed in $\delta^{13}C$ notation by $\delta^{13}C$ (‰) = $(R_{sample}/R_{standard} - 1) \times 1000$, where R_{sample} and $R_{standard}$ are the molar ratios of the heavy to light isotope of the sample and standard (PDB), respectively.

Carbon content in soils was measured using total carbon analyzer (TOC-5000 connected with SSM-5000A, SIMAZU).

2.4 C-137 radioactivity measurement

About 250~500 g of samples were sealed in a plastic container (13 cm in diameter) and counted on a germanium gamma detector coupled to a multichannel analyzer (MCA 7700, Seiko EG & G). Cs -137 radioactivities were determined by its γ emissions at 661.6 keV. Counting geometry was almost the same for all samples. The data obtained are expressed in relative concentration (RC) by RC = CPM of each layer / maximum CPM of each profile.

3. RESULTS AND DISCUSSION

C-14 specific activities in cedar leaves were 249 and 247 mBq (g carbon)$^{-1}$ in Satomi Village and 256~278 mBq (g carbon)$^{-1}$ in Tokai Village, respectively (Table 1). Momoshima et al. reported that

about 20 mBq (g carbon)$^{-1}$ excess was observed around the nuclear power plant in vegetation samples in comparison with the areas located far from nuclear facilities [4]. In this study, C-14 specific activity in cedar leaves around nuclear facilities exceeded that in the control areas by 8~30 mBq (g carbon)$^{-1}$.

Table 1 C-14 specific activity in Japanese cedar leaves [2]

Location	C-14 specific activity (mBq/g carbon)
Satomi Village	
Site 1	249±4[a]
Site 2	247±4
Tokai Village	
A	278±4
B	256±4
C	262±4
D	265±4
E	259±4

[a] Counting error 1σ

The depth profile of carbon content and stable isotope ratio, Cs-137 relative concentration and C-14 specific activity in forest soil were measured for 3 sampling points (Tokai A, Satomi site 1 and Satomi site 2). The results are shown in Fig. 3, Fig. 4 and Fig. 5, respectively. In these figures, the sampling depth in the soil profile is expressed as a negative value and positive 1 cm and 2 cm in soil depth show old and fresh litter on the soil surface. Tokai A is forested mainly with cedar, also with some deciduous trees. Satomi site 1 is forested with cedar and deciduous trees and site 2 is forested only with cedar trees. The annual ring count shows that the cedar trees were planted 30 years ago in Satomi site 2.

The total carbon contents in 3 sampling points show about 50 % in litter, and this decreased dramatically in the upper 10 cm. The carbon isotope ratios ($\delta^{13}C$) show -27 ~ -30 ‰ in the litter. This value is the same as that in the fresh leaves. $\delta^{13}C$ values increased with increasing soil depth, but the tendency shows a difference among 3 sampling points. For Tokai A and Satomi site 2, which are forested mainly with cedar, the $\delta^{13}C$ value increased gradually from litter to 60 cm depth. For Satomi site 1, in which the proportion of deciduous tree is high, the $\delta^{13}C$ value increased steeply between litter and soil, and then the variation of $\delta^{13}C$ in deeper soil was gentler than that for other sampling points. The variation of $\delta^{13}C$ was probably caused by isotopic fractionation in the process of microbial decomposition. Microorganisms preferentially use lighter carbon sources in metabolic reactions associated with litter decomposition and soil organic matter oxidation, and then the residual soil organic matter should become progressively more positive in its $\delta^{13}C$ values. Stable carbon isotope will be a very useful signal for estimating turnover of soil organic matter.

Both Cs-137 relative concentration and C-14 specific activity have a peak in the depth profile. The specific activity of C-14 and Cs-137 in the atmosphere increased dramatically in the 1960's as a result of atmospheric nuclear-weapon tests, and continually decreased to the present. Therefore, the highest peaks of Cs-137 and C-14 in the soil profile are ascribed to fallout from nuclear-weapon tests. For Tokai A and Satomi site 2, the highest concentration of Cs-137 was observed in the horizon of 5~10 cm and 5~7.5 cm, respectively, and the highest concentration of C-14 was observed in the horizon of 2~5 cm for both sites. For both sites, the peak of Cs-137 was in deeper soil than that of C-14. The second peak of C-14 in the horizon of 15~20 cm for Satomi site 2 was probably due to disturbance when the trees were planted. For Satomi site 1, the highest peak of Cs-137 in soil profile was observed in shallower soil (2~5 cm) than that for other sites. This is probably because the difference of the vegetation affected the sedimentation rate. The litter in Satomi site 1, in which the proportion of deciduous trees is high, was decomposed more easily and the soil sedimentation rate was

lower than that in other sites. Though a C-14 peak was observed in the horizon of 5~10 cm in the soil profile for Satomi site 1, the peak concentration was lower and the dispersion of the measured value was big. The reason is not certain yet, but there is a possibility of soil disturbance by earthworms.

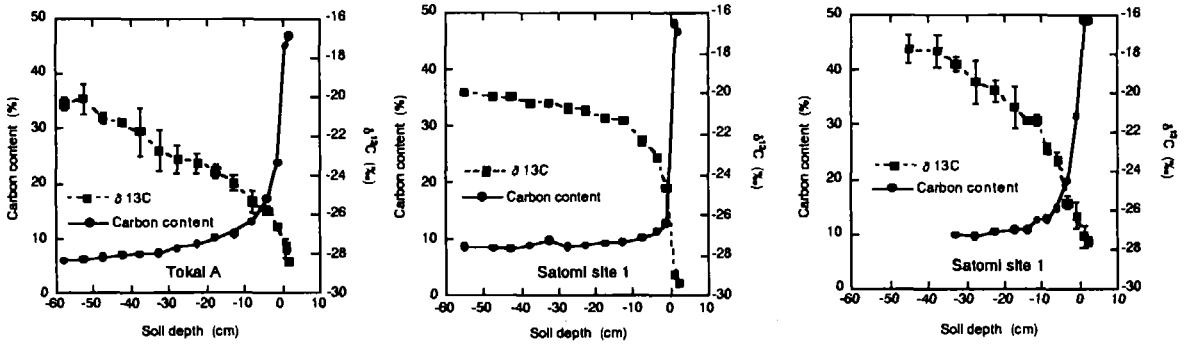

Fig. 3 Depth profiles of carbon content and stable isotope ratio [3]

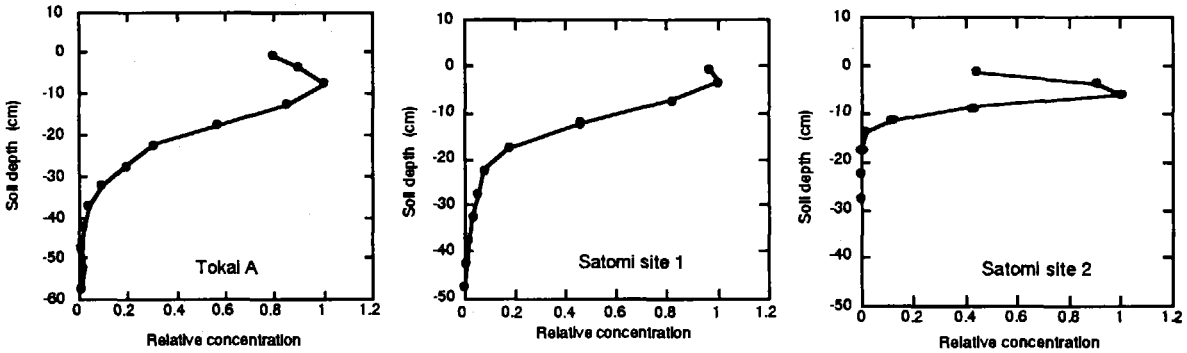

Fig. 4 Depth profiles of Cs-137 relative concentration [3]

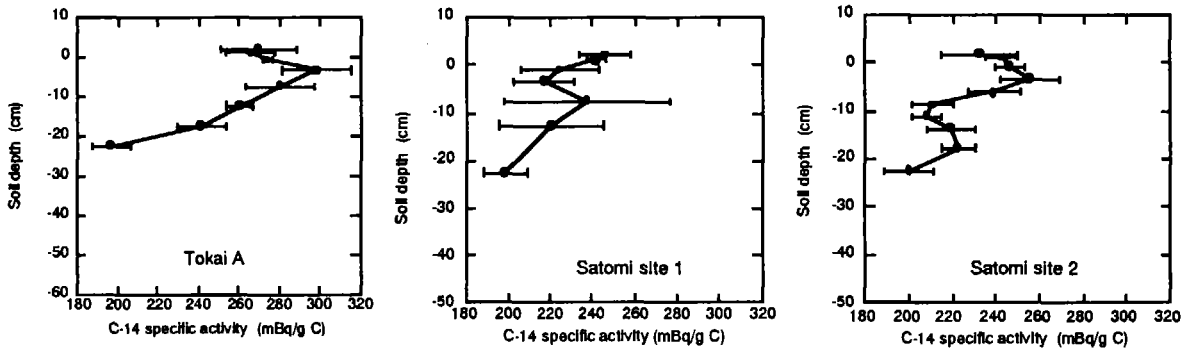

Fig. 5 Depth profiles of C-14 specific activity [3]

4. CONCLUSION

A simple and quick method for C-14 measurement has been developed. C-14 specific activities in cedar leaves and soils collected from an area around nuclear facilities and control areas were measured using this method. Depth distribution of Cs-137, total carbon content and stable carbon isotope ratio in soils were also measured at the same sampling sites and compared with the depth distribution of C-14.

C-14 specific activity in cedar leaves around nuclear facilities exceeded that in the control areas by 8 to 30 mBq (g carbon)$^{-1}$. The total carbon contents in all sampling points show about 50 % in litter

and decreased dramatically in upper 10 cm. The $\delta^{13}C$ values show that the turnover dynamics of soil organic carbon could be described very well by progressive enrichment values of $\delta^{13}C$. The depth distribution of C-14 and Cs-137 in forest soil shows that C-14 and Cs-137 have peak values in the top 10 cm of the soil profiles ascribed to the highest concentration levels in the 1960's as a result of atmospheric nuclear-weapon tests. However, difference of the vegetation and behavior of both radionuclides affected the depth of the peak.

REFERENCES

1. Koarashi, J., Amano, H., Andoh, M., Iida, T. and Moriizumi, J., *Journal of Environmental Radioactivity* **60**, 249 (2002)
2. Guo, J., Atarashi-Andoh, M. and Amano, H., *Journal of Radiation and Nuclear Chemistry* **255**, 223 (2003)
3. Atarashi-Andoh, M. and Amano, H., *Proceedings of 9th International Symposium on Environmental Radiochemical Analysis*, Maidstone, UK, in press
4. Momoshima, N., Kawamura, H., Takashima, Y. and Ezumi, S., *Hokenbutsuri* **30**, 121 (1995), in Japanese