Depth Profiles of Radiocarbon and Carbon Isotopic Compositions of Organic Matter and CO_2 in a Japanese Forest Area

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Abstract

Specific activities of ^{14}C and carbon isotopic compositions ($\Delta^{14}C$) in organic matter and CO_2 in a Japanese forest soil were determined. For investigating the transport of CO_2 in soil, the specific activities of ^{14}C and $\Delta^{14}C$ within the organic layer on the top of soil surface and the atmospheric CO_2 were also determined. The specific activity of ^{14}C and $\Delta^{14}C$ in the organic matter decreased with the increasing soil depth of 0 - 60 cm while that of the soil CO_2 was not significantly variable at the soil depth of 13 - 73 cm and was larger than that in the atmospheric CO_2. Peaks of specific activities of ^{14}C appeared at the depth of 0 - 4 cm and $\Delta^{14}C$ values were positive in the depth range from 0 to 15 cm. These results suggested that the present soil at depth of 0 - 4 cm had been produced since the mid-1950s up until 1963 and the bomb C had reached the depth of 15 cm in the objective soil area. The soil CO_2 at the depth of 53 - 73 cm were assumed to originate only from the decomposition of soil organic matter and the respiration of live roots, the respiration of live roots was estimated to have seasonal variations and be the main source of the soil CO_2 at the deeper horizons in the present forest area.

Key words: soil, soil CO_2, soil organic matter, specific activity of ^{14}C, carbon isotopic composition ($\Delta^{14}C$), depth profiles, sources.

1. Introduction

Radiocarbon is of concern in health physics because of its potential inventory, rapid mobility, and long half-life (5730 y) (Amiro et al., 1991). Two thousands Pg of carbon including radiocarbon is estimated to exist as soil organic matter (SOM) and the SOM exchanges with atmospheric CO_2 at about 60 Pgy^{-1} (IPCC, 2000). Radiocarbon in the SOM enters the food chain by plants through both direct and indirect uptakes from roots and atmosphere, respectively to give a part of the radiation dose to the global populations. The radiocarbon can be indicated by $\Delta^{14}C$, the permil (‰) deviation from the $^{14}C/^{12}C$ ratio of a standard with fixed isotopic composition. As the $\Delta^{14}C$ value is zero for atmospheric CO_2 in the year AD 1950, the positive $\Delta^{14}C$ value indicates the presence of carbon labeled with ^{14}C produced by atmospheric nuclear weapons testing (bomb carbon). Consequently, the $\Delta^{14}C$ in SOM can show the transport of bomb carbon in soil.

Since radiocarbon in SOM ultimately exchanges with atmospheric CO_2 as soil CO_2 (S-CO_2), it is essential to study depth profiles of the specific activity of ^{14}C and $\Delta^{14}C$ in SOM and S-CO_2 simultaneously from the viewpoints of health physics and the study on carbon transport in soil. Some studies have been done on the variation of the specific activity of ^{14}C in SOM (Guo et al., 2003) while few studies have been done on the activity of S-CO_2 due to the difficulties of determining the specific activity of ^{14}C in S-CO_2. Much fewer studies have been concerned on variations of the specific activity of ^{14}C in SOM and S-CO_2 simultaneously.

Forest ecosystems constitute the major reservoir of the global carbon (Houghton et al., 1990; Tan et al., 1990) and have little man-made effect. Forest soil would be a good objective for the study of carbon transport in soil. In the present study, both soil and soil air at different horizons in a Japanese forest soil were collected. Depth profiles of their specific activities of ^{14}C and $\Delta^{14}C$ were obtained. The specific activities of ^{14}C and $\Delta^{14}C$ within the organic layer on soil surface and the atmospheric CO_2 were also determined. The specific activities of ^{14}C in all these samples were measured with an accelerator mass spectrometry (AMS)(Nakamura et al., 2004), which allows the specific activity of ^{14}C measurement on very small sample sizes, about 1000 times smaller than that needed for conventional decay counting methods.

2. Materials and methods

2.1. Site description

The experimental site (35°13' N and 137°34'E) locates in Inabu, Aichi Pref., Japan, as shown in **Fig. 1**, with a mean elevation of 1010 m. The site is forested with about 40-year-old Japanese larch (*LaLix leptolepis*) averaging 23 m in height. The soil belongs to a brown forest one. It is covered by an organic layer with a depth of 7 cm, which consists of a 3-cm leaf-layer (L) and a 4-cm fermentation-layer. The soil is dark at the depth of 0 - 3 cm, yellow-brown at 3 - 13 cm and tan below 13 cm.

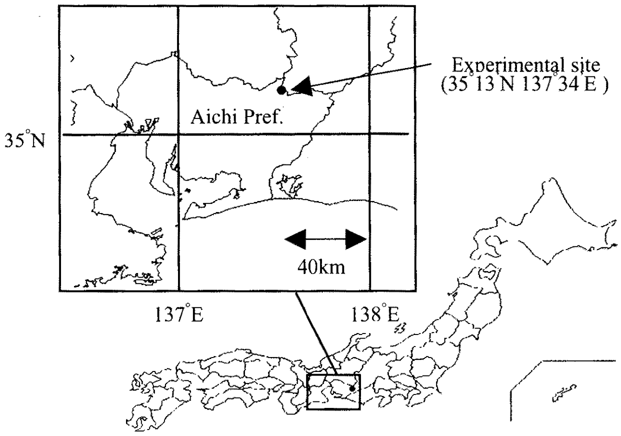

Fig. 1 The location of experimental site.

2.2. Organic carbon measurements

Two sets of soil samples were collected: one set was used to analyze the physical and chemical characteristics of soil such as bulk density, particle density, volumetric water content, air-filled porosity, content of SOM and the value of pH, as shown in **Table 1**. Another was treated with the high temperature combustion pretreatment followed by the AMS (High Voltage Engineering Europe, The Netherlands) measurement to determine its specific activity of ^{14}C and $\Delta^{14}C$. The results were shown in **Figs. 2** and **3**, respectively. Additionally, the specific activity of ^{14}C and $\Delta^{14}C$ in the organic layer were also measured with the same method and were shown in **Figs. 2** and **3**, respectively.

2.3. Air measurements

About 500-mL soil air was collected at different soil depth through the gas-collection probes by a 200-mL syringe. Additionally, a 7000-mL of atmospheric air at either height of 0, 25 and 50 cm above the organic layer was collected by a pump with airflow rare of 500 $mLmin^{-1}$. The CO_2 concentrations in both collected soil air and atmospheric air were firstly measured with a gas chromatograph (GC-14 A, Shimadzu, Japan) with a methanizer and FID. Then, Their specific activities of ^{14}C and $\Delta^{14}C$ were determined with the same method used for the soil samples and were also shown in **Figs. 2** and **3**, respectively.

Table 1 The physical and chemical characteristics of soil samples.

Date	Depth (cm)	Bulk density (ρ_d, g/cm^3)	Particle density (ρ_s, g/cm^3)	Volumetric water content (θ_w, %)	Air-filled porosity[a] (%)	Content of organic matter (wt, %)	pH value*
25 May 2004	0-5	0.63	2.58	29	46.6	17.8	
	5-10	0.72	2.58	49	23.0	11.1	
	15-20	0.69	2.56	61	12.0	15.4	
	35-40	0.85	2.61	63	4.4	11.8	
13 July 2004	0-5	0.46	2.44	37	44.2	16.7	
	5-10	0.58	2.44	34	42.2	15.2	
	10-15	0.82	—	—	—	—	
	15-20	0.85	2.51	61	5.1	13.5	
	20-25	0.82	—	—	—	—	
	25-30	0.73	2.55	57	14.4	12.0	
9 Sept. 2004	0-5	0.59	2.34	49	25.8	20.4	
	5-10	0.48	2.47	31	49.6	16.5	
	15-20	0.88	2.59	56	10.0	13.3	
	25-30	0.77	2.58	55	15.2	13.5	
	40-45	0.96	2.65	59	4.6	9.5	
	45-50	1.12	2.66	58	0.1	7.6	
	55-58	1.28	2.74	46	7.3	7.2	
8 Nov. 2004	0-5	0.50	2.61	43	38.1	20.5	4.32
	5-10	0.67	2.64	34	40.6	14.7	4.80
	15-20	0.78	2.70	57	14.0	13.8	4.82
	25-30	0.65	2.72	60	16.1	14.1	4.84
	35-40	0.86	2.74	63	4.9	12.6	4.90
	45-50	0.83	2.75	62	7.3	12.3	5.22
	50-55	1.02	2.74	62	0.5	29.7	—
	55-60	1.04	2.83	60	3.3	9.6	5.56

[a]: $(1-\rho_d/\rho_s) \times 100\% - \theta_w$; *the mixture of soil and added deionized water (Weight of soil : H_2O=5:10-g)

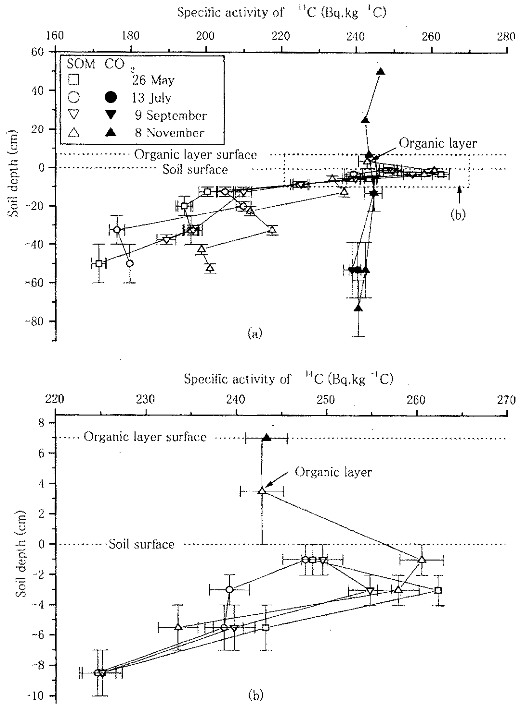

Fig. 2 Specific activity of ^{14}C in the SOM (Soil organic matter), the organic layer, the S-CO_2 (Soil CO_2) and the atmospheric CO_2. The vertical and transversal bars present the depth range of collected sampled and counting error (1σ), respectively. The bottom Figure is the expansion of the b area in upper Figure.

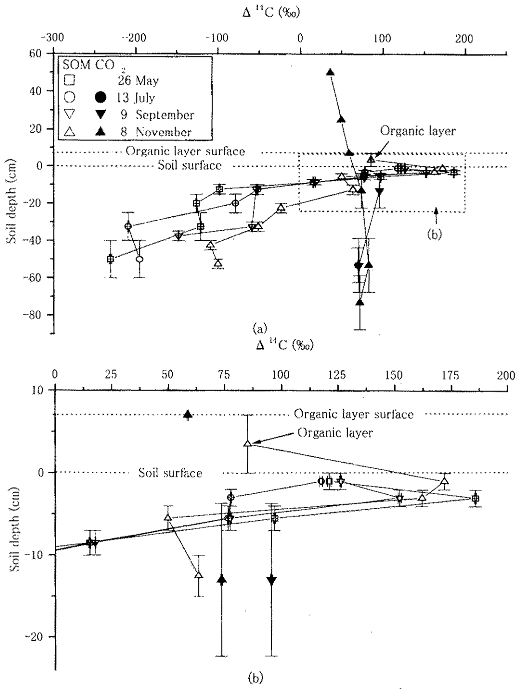

Figure 3 The $\Delta^{14}C$ values within the SOM, the organic layer, the S-CO_2 and the atmospheric CO_2. The vertical and transversal bars present the depth range of collected samples and counting error (1σ), respectively. The bottom Figure is the expansion of the b area in upper Figure.

3. Results and discussions

3.1. Depth profiles of specific activities of ^{14}C and $\Delta^{14}C$ in SOM and S-CO_2

The specific activity of ^{14}C in the SOM (**Fig. 2**) decreased with the increasing depth. It was in the range from 262.3 ± 2.3 to 194.1 ± 2.3 Bq.kg^{-1}C at the depth of 0 - 25 cm, which was in agreement with that published by Guo et al. (2003), who gave 281- 200 Bq.kg^{-1}C in the SOM at the depth of 0 - 22.5 cm in three Japanese soil samples collected in 2001. The specific activities of ^{14}C in SOM were different at different vertical and horizontal positions. They were 236.7±2.2 and 211.6±2.4 Bq.kg^{-1}C at the depth of 10 - 15 and 20 - 25 cm, respectively, for the soil collected on 8 Nov.; however, it was 209.8±1.8 Bq.kg^{-1}C at the depth of 15 - 25 cm for the soil collected on 13 July. At the same depth, such as at the depth of 4 - 7 cm, it was 243.2±2.2, 238.6±2.1, 239.7±3.1 and 233.5±2.2 Bq.kg^{-1}C for the soil sample collected on 26 May, 13 July, 9 Sept. and 8 Nov., respectively. This might be related to soil heterogeneity.

Additionally, peaks of specific activity of ^{14}C in the SOM (**Fig. 2**) appeared at a depth of 2 - 4 cm in the soils samples collected on 26 May and 9 Sept. while they appeared at the depth of 0 - 2 cm on both of 13

July and 8 Nov. It is known that the specific activity of ^{14}C in the atmosphere increased strongly from mid-1950s up until 1963 due to the release of ^{14}C by nuclear weapons testing. Thus, the peaks can be suggested to be the residues of specific activities of ^{14}C in the SOM produced in the mid-1950s until 1963. In other words, the present soils at the depth of 0 - 4 cm have been produced since the mid-1950s up until 1963.

The specific activities of ^{14}C in the S-CO_2 (**Fig. 2**) at the depth of 13 - 73 cm and the organic layer collected on 8 Nov. were in the range from 238.8±2.2 to 244.5±2.3 and 242.8±2.4 Bq.kg^{-1}C, respectively. Since the live roots and the organic layers belonged to the same plant, the specific activity of ^{14}C in the present live roots was considered to be close to 242.8 Bq.kg^{-1}C. Comparing the specific activities of ^{14}C in the SOM of the soil samples collected on 8 Nov. with those in the S-CO_2 collected on the same day, the former ones were less than the latter ones at deeper soil horizons. For example, at the depth of 50 - 55 cm, 200.9±1.9 Bq.kg^{-1}C was observed for SOM while about 242.9±2.4 Bq.kg^{-1}C for S-CO_2. If there is no difference between the specific activity of ^{14}C in SOM and that in the S-CO_2 derived from decomposition of this SOM, the decomposition of SOM at the deeper horizon is not suggested to be the main source of S-CO_2 at the same deeper horizon.

The CO_2 diffuses in soil in the two ways: through the soil air pores and with the soil water (Jassal et al., 2004). Since the diffusion coefficient of CO_2 in air is generally much larger than that in water, for example, it was reported to be approximately ten thousands times that in water at 20°C by Robinson and stokes (1959); Pritchard and Currie (1982), the diffusion of S-CO_2 through the soil air pores are much greater than the transport of S-CO_2 with the soil water. Thus, if there is no depth dependency of the specific activity of ^{14}C in S-CO_2 from the respiration of live roots, the similarity in the specific activities of ^{14}C between the S-CO_2 and live roots at the deeper soil horizon indicates that the respiration of live roots may be the dominant source of S-CO_2 at the deeper soil horizons.

The specific activity of ^{14}C in the atmospheric CO_2 (**Fig. 2**) is in the range from 242.2±2.3 to 246.4±2.3 Bq.kg^{-1}C, which is also approximately same as that in S-CO_2 at the depth of 13 - 73 cm. This may imply that atmospheric CO_2 enters the soil air. The air-filled porosity of the present soil ranged from 10.0 to 49.6% at a depth of 0 - 20 cm and about 4.1% at a depth of 40 - 60 cm, as shown in **Table 1**. This indicates that the vertical exchange of S-CO_2 through the soil pores is stronger at 0 - 20 cm and weaker at 40 - 60 cm. If the volume of collected soil air can be corresponded to a spherical volume of soil, the radius is estimated to be less than 10 cm for 500-mL of soil air at the depth of 13 cm because the air-filled porosity at the same depth is larger than 10%. The 500-mL of soil air sample at the depth of 13 cm may consist of the air at the depth of 3 - 23 cm. The effect of the entering of atmospheric CO_2 had been reported to be important at very shallow depths (less than 10 cm) by (Cerling et al., 1991; Hesterberg and Siegenthaler, 1991), so that the 500-mL soil air at the depth of 13 cm might exist the entering of atmospheric air.

The $\Delta^{14}C$ in the SOM (**Fig. 3**) decreased with the increasing soil depth. Positive $\Delta^{14}C$ in the SOM appeared at the depth of 0 - 10 cm in soil samples collected on 26 May, 13 July and 9 Sept. and 0 - 15 cm in that collected on 8 Nov., respectively. Since positive $\Delta^{14}C$ value is due to incorporation of 'bomb C' (Margarets et al., 2002), the soils at the depth of 0 - 15 cm are considered to dominate by the organic matter fixed in the last 50 years. Therefore, one can be concluded that the bomb C has reached the depth of 15 cm in the present soil area.

The $\Delta^{14}C$ in the S-CO_2 (**Fig. 3**) were not significantly variable at the depth of 13 - 73 cm comparing with the variation of that in SOM with the increasing soil depth. The $\Delta^{14}C$ values in the S-CO_2 were larger than that in the atmospheric CO_2 and approximately same as that in the organic layer. It was much larger than that in the SOM at deeper soil horizon. For example, the S-CO_2 at the depth of 53 - 73 cm on 8 Nov., it was in the range from 72.0±0.3 to 82.6±0.4‰ while that of the SOM was -100.3±0.5‰ at the depth of 50 - 55 cm. Thus, it can also be concluded that the decomposition of SOM at the deeper horizon is not the main source of S-CO_2 at the same deeper soil horizon.

3.3. Quantitative estimation of S-CO_2 originations

The S-CO_2 at the depth of 53 - 73 cm is assumed to originate only from the decomposition of SOM and the respiration of live roots, based on a mass balance approach of $^{14}CO_2$, the following equations can be obtained:

$$R_r + R_s = 1, \quad (1)$$

$$\Delta^{14}C_i = R_r \times \Delta^{14}C_r + R_s \times \Delta^{14}C_s. \quad (2)$$

Combining Eqs. (1) with (2), one can obtain:

$$R_r = \frac{\Delta^{14}C_i - \Delta^{14}C_s}{\Delta^{14}C_r - \Delta^{14}C_s}, \quad (3)$$

where R_r and R_s stand for the fractional contributions of the respiration of live roots and the decomposition of SOM to the total S-CO_2 concentration, respectively; $\Delta^{14}C_i$, $\Delta^{14}C_r$ and $\Delta^{13}C_s$ present the $\Delta^{14}C$ in the S-CO_2, those derived from the respiration of live roots and the decomposition of SOM, respectively. Since the effects of isotopic fractionations on $\Delta^{14}C$ are eliminated with normalization by $\delta^{13}C$ (Stuiver and Polach, 1977), the $\Delta^{14}C$ in the S-CO_2 derived from the respiration of live root can be inferred to be that in the same plant itself. Additionally, little or no significantly seasonal variations occurred in the $\Delta^{14}C$ within the plant tissue and the SOM. The $\Delta^{14}C$ value in the S-CO_2 derived from the live root-respiration can be estimated to be close to 85.1‰, which is the $\Delta^{14}C$ value in the organic layer shown in **Fig. 3**. The $\Delta^{14}C$ value in the S-CO_2 derived from the SOM-decomposition at the depth of 55 - 73 cm can be inferred to be less than -100.3‰ that is the $\Delta^{14}C$ value in the SOM at the depth of 50 - 55 cm on 8 Nov. due to the decrease of $\Delta^{14}C$ in SOM with the increasing soil depth. From **Fig. 3**, the $\Delta^{14}C$ value in the measured S-CO_2 were 69.7, 95.0 - 70.6 and 82.6 - 77.3‰ for the S-CO_2 at the depth of 53 cm on 13 July, 53 - 73 cm on 9 Sept. and 53 - 73 cm on 8 Nov., respectively. Using **Eqs. (1) - (3)**, the R_r at these depths were estimated to be 92%, larger than 92% and larger than 96% on 13 July, 9 Sept. and 8 Nov. 2004, respectively. Therefore, one can be concluded that the fractional contribution of the live root- respiration in the deeper horizon (53 - 73 cm) in the present soil may have a seasonal variations and the live root-respiration is the main source of the S-CO_2 at the deeper soil horizons.

4. Conclusions

The specific activity of ^{14}C and $\Delta^{14}C$ in SOM decreased with the increasing depth from 0 to 60 cm while that in the S-CO_2 was not significantly variable at the depth from 13 to 73 cm and larger than that in the atmospheric CO_2. A peak of ^{14}C specific activity was found at the depth from 0 to 4 cm, which implied that the present soil at the depth of 0 - 4 cm had been produced since mid-1950s up until 1963. Positive $\Delta^{14}C$ values in the depth range from 0 to 15 cm implies that the bomb C have reached the depth of 15 cm in the present forest soil.

The specific activity of ^{14}C and $\Delta^{14}C$ in the S-CO_2 were larger than that in the SOM at the deeper horizons (53 - 73 cm). This implied that decomposition of SOM were not the main source of S-CO_2 at the same deeper horizon. If the S-CO_2 at the deeper horizon was assumed to originate only from the decomposition of SOM and the respiration of live roots, the respiration of live roots was estimated to have an seasonal variations and be the main source of the S-CO_2 at the deeper horizons (53 - 73 cm) in the present forest soil based on the isotope mass balance approach for $^{14}CO_2$.

Acknowledgements

The authors would like to thank Prof. T. Nakamura, Ms T. Ohta and Ms. A. Ikeda, Nagoya University Center for Chronological Research, for their cooperation in mass spectrometry measurements. The authors also wish to thank Prof. E. Shibata, Mr. Y. Imaizumi, Mr. N. Yamaguchi, University Forest, Graduate School of Bioagricultural Sciences, Nagoya University, for their kind assistance and advice during field experiment in the site. This research was carried out as a part of the 21st century COE program on Isotopes for Prosperous Future, Nagoya University.

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3. Domestic presentations

劉衛　森泉　純　山澤　弘実　飯田　孝夫, カラマツ林土壌中の有機物及び二酸化炭素の炭素同位体比. 日本原子力学会春の年会, （平成17年3月）.

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