LATEST ^{14}C CONCENTRATIONS OF PLANT LEAVES AT HIGH ALTITUDES IN THE NORTHERN AND SOUTHERN HEMISPHERES: VERTICAL STABILITY OF LOCAL SUESS EFFECT

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ABSTRACT. The radiocarbon concentrations in plant leaves from different altitudes at 3 sampling locations were measured with the new compact accelerator mass spectrometer (AMS) at Yamagata University to investigate air mixing on a global scale. The sampling locations are Yamagata in the mid-latitudes of the Northern Hemisphere (NH), Kenya in the equatorial region (EQ), and Chacaltaya in the Southern Hemisphere (SH). The ^{14}C concentrations of the plant leaves ranged from 102 to 105 pMC. The ^{14}C concentrations at high altitudes and mountain summits showed similar values of 104.2 ± 0.28, 104.3 ± 0.36, and 104.4 ± 0.23 pMC at the Yamagata, Kenya, and Chacaltaya sites, respectively. These results indicate that air from the free troposphere is well mixed on a global scale. The local Suess effect was calculated using the ^{14}C concentrations of leaves at the land surface and mountain summits. The fractions were estimated as 1.25 ± 0.3% and 0.87 ± 0.44% at Yamagata and Nairobi, respectively. This estimation method is more advantageous than the conventional calculation. The life cycle of the leaves sampled is 1 or 2 yr, and hence the leaves allow us to study the ^{14}C concentrations in the ambient atmosphere during a narrow and specific time period.

INTRODUCTION

The latest atmospheric radiocarbon concentrations are studied for evidence of mixing in the atmosphere on a global scale using plant leaves growing in a city area and on a mountain to obtain a vertical distribution of the ^{14}C concentrations. The time profiles from compiled data show that the bomb peak of atmospheric ^{14}C concentrations in the Southern Hemisphere (SH) appears to have a delay of ~3 yr when compared to that of the Northern Hemisphere (NH), but the ^{14}C concentrations are similar in both hemispheres after 1970 (Hua and Barbetti 2004). This indicates that the air is well mixed by atmospheric circulation on a global scale by way of air exchange between the stratosphere and troposphere (Nydal and Lövseth 1983). Also, the ^{14}C concentrations are exponentially decreasing with a time constant of 16 yr, indicating the air-sea exchange of global carbon circulation (Rakowski et al. 2008). Moreover, the ^{14}C concentration in the atmosphere is influenced by CO_2 emissions from the burning of fossil fuels (Suess 1955).

Contemporary ^{14}C concentrations in the atmosphere could be lower than those in the year 2000 due to the continuing air-sea exchange. The increase of CO_2 from fossil fuel combustion additionally depleted the ^{14}C concentrations. In particular, the depletion rates of ^{14}C concentrations in the NH are greater than those in the SH relating to the amount of fossil fuel consumption (Randerson et al.

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2002). One way to check the degree of global mixing of the contemporary ^{14}C concentrations is to investigate these at different altitudes in locations of the NH and SH because the CO_2 from fossil fuels is emitted at the land surface and diluted to the upper atmosphere. In general, it causes local contamination at the ground level and cleaner global conditions in the air. Therefore, the ^{14}C concentrations at higher altitudes could have higher values compared to those at the surface level.

Since it would be difficult to use airplanes and/or balloons to take air samples at different altitudes around the world, an attempt was made to measure ^{14}C concentrations in plant leaves from different altitudes on mountains at 3 locations in the NH, the equatorial region (EQ), and the SH. Generally, the lifespan of plant leaves is <2 yr, so the ^{14}C fixed in the leaves by photosynthesis indicates the ^{14}C concentrations in the ambient atmosphere during the growth period of the leaves, and hence plant leaves are useful for the investigation of ^{14}C concentrations in air at mainly the time of leaf growth (McNeely 1994). In biology, the formative phase of leaves is roughly during the growing season of spring or summer, and hence the structural cellulose of leaves reflects mainly the atmospheric ^{14}C during the growth period in the year and/or the previous year, depending on whether the plants are annuals or perennials (A Tsujimura, personal communication, 2013). Therefore, the choice of timing for sampling the leaves has implications for time resolutions of <1 yr as indicators of local fossil fuels.

SAMPLES AND METHOD

We have chosen 3 sampling locations: Yamagata in Japan, Nairobi in Kenya, and La Paz in Bolivia, taking into account the zones NH2, NH3, and SH categorized by Hua and Barbetti (2004). As shown in Table 1 with their latitudes and longitudes, the sampling sites are located at mid-latitudes in the NH, in the EQ region, and in the SH, respectively.

Table 1 The altitudes and ^{14}C concentrations of leaf samples with the places for the 3 sampling locations.

Sample ID	Place	Altitude (m asl)	Plant	pMC
Yamagata (Japan), 38°15′N, 140°21′E				
YT_s	Yamagata City	165	Sasa leaf	102.7 ± 0.29
YT_p	Yamagata City	165	Pine leaf	103.1 ± 0.28
YM_s	Yamagata City	213	Sasa leaf	103.1 ± 0.30
YM_p	Yamagata City	213	Pine leaf	103.3 ± 0.28
YZ_s	Mt. Zao	1368	Sasa leaf	103.9 ± 0.28
YZ_p	Mt. Zao	1368	Pine leaf	104.4 ± 0.29
Kenya, 0°09′S, 37°20′E				
NI_1	Nairobi City	1637	Leaf	103.3 ± 0.36
NI_2	Nairobi City	1637	Leaf	103.5 ± 0.37
NA_1	Nanyuki	1934	Leaf	104.3 ± 0.36
Chacaltaya (Bolivia), 16°20′S, 68°07′W				
UM_1	La Paz	3424	Leaf	104.6 ± 0.28
UM2_1l	La Paz	3424	Leaf	104.5 ± 0.37
UM2_2l,	La Paz	3424	Leaf	104.3 ± 0.27
UM2_3l	La Paz	3424	Leaf	104.2 ± 0.30
EL_1	El Alto	4132	Leaf (*Buddleja coriacea Remy*)	103.9 ± 0.28
EL_p	El Alto	4132	Pine leaf	104.5 ± 0.29
CM_s	Mt. Chacaltaya	4773	Leaf (*Senecio* sp.)	104.2 ± 0.28
CT_s	Mt. Chacaltaya	5238	Leaf (*Senecio* sp.)	104.5 ± 0.28
CT_r	Mt. Chacaltaya	5238	rice leaf (*Calamagrostis* sp.)	104.4 ± 0.29
CT2_sl1	Mt. Chacaltaya	5238	Leaf (*Senecio* sp.)	104.2 ± 0.26
CT2_sl2	Mt. Chacaltaya	5238	Leaf (*Senecio* sp.)	104.7 ± 0.27

Table 1 also shows the city or mountain name and the altitude at which the leaf sample was collected in each of the 3 sampling locations. Mt. Zao in Yamagata is the summit of a range of mountains with moderate altitude. The 2 sites in Kenya are Nairobi city on the high plateau and Nanyuki, a town at the foot of Mt. Kenya, 250 km north of Nairobi. The wind in this area is most often out of the northeast and east, although in summer there is also a wind from the south. La Paz (~3400 m) and El Alto (~4200 m) in Bolivia are located in a bowl-like valley and above the valley, respectively. Mt. Chacaltaya is the summit of a mountain with a remarkable height of 5200 m. It is located 30 km north of La Paz, and the wind comes from the east during austral summer. A few leaves were carefully sampled from living plants in 2011 and 2012 at the 3 locations. Although the general names (biological names) of several plants are shown in Table 1, other plants are identified as "leaf" only.

The carbon from plant leaves was extracted by burning the samples after they had been chemically cleaned by AAA treatment after rinsing with acetone. The ^{14}C concentrations were measured with an uncertainty of less than 3‰ using the new Compact AMS, which is a 0.5MV Pelletron accelerator (1.5SDH-1) developed by National Electrostatics Corp. at Yamagata University (Tokanai et al. 2011). In addition to measuring $\delta^{13}C$ by AMS for pMC correction, the $\delta^{13}C$ values were independently measured using an IsoPrime™ mass spectrometer for stable isotopes to examine the characteristics of $\delta^{13}C$ of the plant leaves.

For an eucaly leaf sample at La Paz, the ^{14}C concentration in extracted cellulose was checked. It was comparable to that of AAA treatment with 103.68 ± 0.24 and 104.23 ± 0.3 pMC for the cellulose and AAA, respectively, a difference of 0.55 ± 0.38 pMC.

RESULTS AND DISCUSSION

The ^{14}C concentrations of the plant leaves vary from about 102 to 105 pMC with an uncertainty of <0.3 pMC as shown in Figure 1. The squares, diamonds, and circles represent the samples from Yamagata, Kenya, and Chacaltaya, respectively. For Yamagata, the leaves of pine and sasa (dwarf bamboo leaf) are shown with solid squares and open squares containing plus signs (open-plus). The differences between the pine and sasa in 3 pairs were 0.4 ± 0.4, 0.2 ± 0.4, and 0.5 ± 0.4 pMC towards higher altitude, respectively. Although the pine has an offset to the sasa and it might be an indication of the longer life of the pine needles, it is rather difficult to pinpoint a significant systematic offset when taking the uncertainties into account. For Kenya, 2 kinds of leaves at Nairobi and the one at Nanyuki are shown with the symbols open-plus, open-dot, and solid diamonds, respectively. For Chacaltaya, the 4 kinds of circles indicate different kinds of plant leaves, picked at the UMSA campus in La Paz. The solid and right-half-solid circles represent a pine leaf and a leaf at El Alto, respectively. For Mt. Chacaltaya, the 4 upper-half-solid circles represent the same plant leaves, and the open-cross symbols represent a rice leaf.

Figure 2 shows the $\delta^{13}C$ values of leaves measured using the IsoPrime as a function of altitude. The values are scattered between −23‰ and −32‰. The $\delta^{13}C$ values for pines were similar for both Yamagata and El Alto. The $\delta^{13}C$ values for Chacaltaya are roughly segmented to the higher values, except for a few samples similar to those for Yamagata. The high $\delta^{13}C$ values might be characteristic of plants that grow in rocky soils at high altitudes.

Figure 3 shows the ^{14}C concentrations in the plant leaves, as shown in Figure 1, as a function of altitude to examine the relationship between the ^{14}C concentrations and the altitudes. In both figures, the ^{14}C concentrations at Chacaltaya are roughly constant between 104 and 105 pMC with an average and standard deviation of 104.4 ± 0.23 pMC for altitudes from 3400 to 5200 m. However, the ^{14}C concentrations for the altitudes at Yamagata and Kenya were clearly different from the concen-

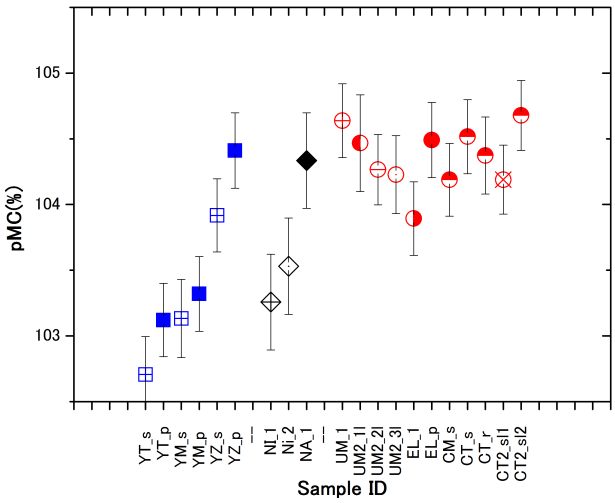

Figure 1 ^{14}C concentrations of the plant leaves at each sampling site. See the text for an explanation of the symbols.

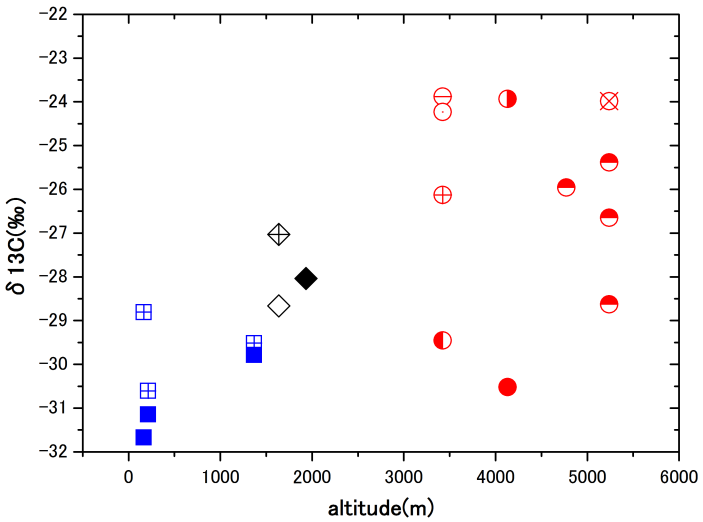

Figure 2 The $\delta^{13}C$ values of plant leaves as a function of altitude. The symbols are the same as in Figure 1.

tration at Chacaltaya. For Yamagata, the increase of ^{14}C concentrations for the pine and sasa leaves was 1.3 ± 0.4 and 1.2 ± 0.4 pMC, respectively, for the ascent of 1200 m. Also, in Kenya, the increase was 0.91 ± 0.5% for the ascent of 300 m. It is an important indication that the ^{14}C concentrations at the high altitudes and the mountain summit show similar values of 104.2 ± 0.28, 104.3 ± 0.36, and 104.4 ± 0.23 pMC at Yamagata, Kenya, and Chacaltaya, respectively.

As the boundary between the free troposphere and the convective mixed layer nearly contacting the land surface is at about 1000 m, the air at the summit of Mt. Zao comes from the free troposphere. Hence, the similar ^{14}C concentrations at the higher altitudes and the mountain summits presumably

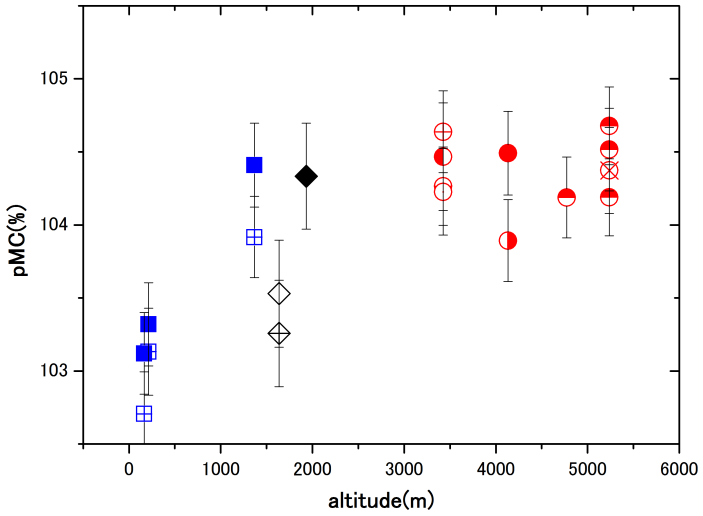

Figure 3 ^{14}C concentrations of the plant leaves as a function of altitude. The symbols are the same as in Figure 1.

indicate that the air of the free troposphere above the convective mixed layer is well mixed on a global scale in the Northern Hemisphere, the equatorial region, and the Southern Hemisphere.

In Figure 4, the average ^{14}C value of 43.7 ± 3‰ (open square) for the high altitudes and the summit at the 3 locations is shown with ^{14}C values of 39.9 ± 4‰ (open circle) and 29.2 ± 3‰ (open triangle) at the land surface in Nairobi and Yamagata, respectively, compared with 2 extrapolated curves derived from the ^{14}C concentrations of tree rings (Hua and Barbetti 2004) and the $^{14}CO_2$ levels for Jungfraujoch (Levin and Kromer 2004). The ^{14}C value at high altitude is between both the extrapolations, indicating that the free tropospheric air is mixed on the global scale. This is caused by the extrapolation from the Jungfraujoch data, at which the altitude is 3450 m asl, and represents the ^{14}C values in the tropospheric air.

Although for the leaves from the 3 same plant species (*Senecio* sp.) picked in 2011 at Chacaltaya at 5200 m, the ^{14}C concentrations were 104.5 ± 0.25 pMC on average. The thin stems of the plant picked in 2011 and 2012 yielded higher ^{14}C values of 106.2 ± 0.28 and 106.9 ± 0.3 pMC, respectively. Using the extrapolated curve from the Jungfraujoch data, these values are equivalent to atmospheric values from 2007. Therefore, although the plant is alive for several years, the ^{14}C concentrations in leaves reflect the ambient atmospheric ^{14}C during the year the leaves were sampled, or the year prior.

The ^{14}C values at the land surface are lower than those extrapolated. The reduction in the ^{14}C values is probably an indication of the influence of fossil fuel consumption. Assuming the CO_2 from fossil fuel combustion at the land surface is well diluted in the free troposphere, we can estimate the fraction of the CO_2 caused by fossil fuel emissions in the local area using the difference in ^{14}C concentrations found between the free troposphere and the land surface. The fractions of CO_2 from fossil fuels were 1.25 ± 0.3% and 0.87 ± 0.44% at Yamagata and Nairobi, respectively. Rakowski et al. (2008) reported the fossil fuel components of atmospheric CO_2 were 5.9 and 6.5 ppmV from tree rings and air CO_2 at Kraków for the period 1983–2003 using the tropospheric $^{14}CO_2$ level at Schauinsland as the ^{14}C concentration of clean air. The fractions at Yamagata and Nairobi are estimated as 4.9 ± 1.5 and 3.4 ± 1.9 ppmV, assuming the CO_2 concentrations at Mauna Loa, respectively.

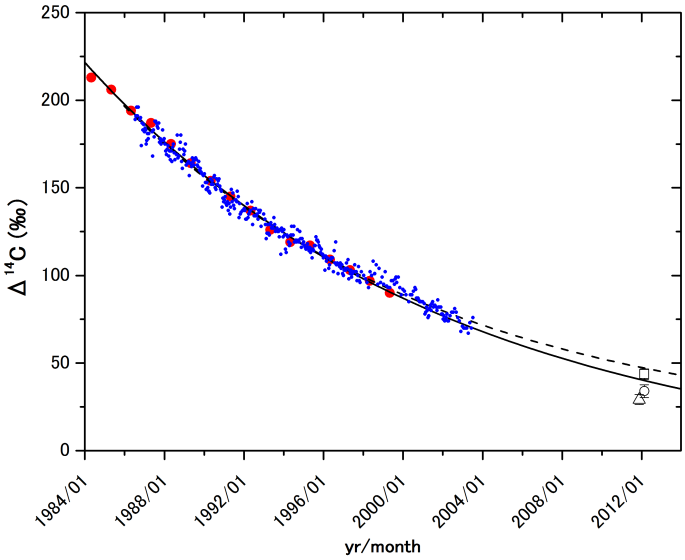

Figure 4 The average ^{14}C at higher altitudes for the 3 locations compared with 2 extrapolated curves (solid line and dashed line) derived from the ^{14}C concentrations of tree rings (Hua and Barbetti 2004) and the tropospheric $^{14}CO_2$ levels for Jungfraujoch (Levin and Kromer 2004), respectively. See the text for an explanation of the symbols.

Although it is necessary to take into account the uncertainties for Kraków, our estimation for Yamagata is comparable to that of Kraków in the NH.

As, in spring, the stratospheric and tropospheric air mutually exchanges at mid-latitudes (Feely et al. 1989), it could affect the fossil fuel fractions with a seasonal cycle for plant leaves, in particular, growing from spring to summer. Observation of cosmogenic nuclide ^{7}Be concentrations in the surface air shows an increase of 40% in spring compared to that of summer (Kikuchi et al. 2009).

These results appear to indicate that ^{14}C in the free troposphere is well mixed on the global scale, and hence the local Suess effect has a vertical limit in the mixing layer that is in contact with the land surface. Moreover, it is efficient to estimate the local Suess effect, normalizing the ^{14}C in leaves at the land surface to the one at the mountain summit. However, for the implementation it is important to compare the data with the ^{14}C in leaves and air samples from several altitudes for multiple locations around the world.

CONCLUSION

The ^{14}C concentrations in plant leaves, picked at different altitudes from 3 sampling locations, were measured with the new compact AMS at Yamagata University to investigate the mixing of air on a global scale. The sampling locations are Yamagata in the mid-latitudes of the Northern Hemisphere, Kenya in the equatorial region, and Chacaltaya in the Southern Hemisphere. The ^{14}C concentrations of the plant leaves were distributed from about 102 to 105 pMC. The ^{14}C concentrations at Chacaltaya were roughly constant with an average of 104.4 ± 0.23 pMC for altitudes from 3400 to 5200 m. However, for Yamagata and Kenya the ^{14}C concentrations increased with increasing altitude, unlike at Chacaltaya. The ^{14}C concentrations at high altitudes and the summits on mountains showed sim-

ilar values of 104.2 ± 0.28, 104.3 ± 0.36, and 104.4 ± 0.23 pMC at the Yamagata, Kenya, and Chacaltaya sites, respectively.

These results indicate that the air in the free troposphere above the convective mixed layer is well mixed on a global scale across the Northern Hemisphere, the equatorial region, and the Southern Hemisphere. Knowing this, we can also estimate the fraction of the CO_2 contributed by fossil fuels at a local site from the difference in ^{14}C concentrations between the free troposphere and the land surface. The fractions of CO_2 from fossil fuels were estimated at 1.25 ± 0.3% and 0.87 ± 0.44% at Yamagata and Nairobi, respectively. This estimation method might have an advantage over the conventional calculation, which uses ^{14}C concentrations in clean air observed at other sites, although it will be necessary to compare the ^{14}C concentrations in leaves and in air samples from several altitudes at various global locations.

Since the ^{14}C concentration of a leaf is clearly lower than that of a stem from the same plant, for the perennial grasses of *Senecio* sp. living at 5200 m that we sampled, the lifespan of the leaves is 1 or 2 yr. Therefore, the leaves are available to observe the ^{14}C concentrations in the ambient atmosphere, comparing them with the tropospheric $^{14}CO_2$ level for Jungfraujoch (3450 m asl). The choice of timing for sampling short-lived leaves, such as those on rice plants, has implications for time resolutions of less than 1 yr as indicators of a local fossil fuel effect.

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