

## A COMPARISON OF DISTRIBUTION MAPS OF $\Delta^{14}\text{C}$ IN 2010 AND 2011 IN KOREA

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**ABSTRACT.**  $\Delta^{14}\text{C}$  values of leaves of deciduous trees provide a means to map the regional-scale fossil fuel ratio in the atmosphere. We collected a batch of ginkgo (*Ginkgo biloba* Linnaeus, a deciduous tree) leaf samples from across Korea in the month of July in both 2010 and 2011 to obtain the regional distribution of  $\Delta^{14}\text{C}$ . The  $\Delta^{14}\text{C}$  values of the samples were measured using accelerator mass spectrometry (AMS) at the Korea Institute of Geoscience and Mineral Resources (KIGAM). The average of the  $\Delta^{14}\text{C}$  values from clean air sites in Korea in 2011 measured slightly lower than the average of  $\Delta^{14}\text{C}$  values in 2010. Distribution maps of  $\Delta^{14}\text{C}$  of 2011 and 2010 in Korea were made based on a series of  $\Delta^{14}\text{C}$  values of ginkgo leaf samples from Korea using the Geostatistical and Spatial analyst tools in ESRI's ArcMap software. The distribution maps of  $\Delta^{14}\text{C}$  showed that  $\Delta^{14}\text{C}$  values in the western part of Korea are lower than those in the eastern part of Korea. This is because the western part of Korea is densely populated and contains many industrial complexes, and also because westerly winds from China, containing  $\text{CO}_2$  from fossil fuel use, blow into Korea. We compared the distribution maps of 2010 and 2011 and tried to find traces of the Fukushima power plant accident in Japan.

### INTRODUCTION

Significant fossil fuel emissions from industrial complexes and vehicles have increased atmospheric  $\text{CO}_2$ . Average  $\text{CO}_2$  concentration exceeded 395 ppm in 2011 in Korea and is still increasing (Cho 2012). Furthermore, this process decreases the  $\Delta^{14}\text{C}$  ratio in the atmosphere and subsequently in the biosphere and ocean. These changes are referred to as Suess effect (Suess et al. 1955). In contrast to the Suess effect, nuclear weapons tests and nuclear power plant operations have led to an increase in the atmospheric  $\Delta^{14}\text{C}$ .

$\Delta^{14}\text{C}$  of carbon dioxide ( $\text{CO}_2$ ) in the atmosphere provides a unique tracer for the carbon cycle (Turnbull et al. 2009). Radiocarbon levels in annual plants provide a means to map regional fossil fuel plumes in surface air (Hsueh et al. 2007). A number of studies have taken advantage of this strong effect of fossil fuel  $\text{CO}_2$  emissions on  $\Delta^{14}\text{C}$  to constrain atmospheric mixing ratios of recently added fossil fuel  $\text{CO}_2$  ( $\text{CO}_{2\text{ff}}$ ), demonstrating that  $\Delta^{14}\text{C}$  is likely to be the best method to independently and objectively verify fossil fuel  $\text{CO}_2$  emissions (Turnbull et al. 2009).

In this study, we measured the  $\Delta^{14}\text{C}$  of ginkgo leaves across South Korea in July 2010 and 2011 and made distribution maps of  $\Delta^{14}\text{C}$  in Korea with the Geostatistical and Spatial analyst tools in ESRI's ArcMap software. We are concerned that fossil fuel  $\text{CO}_2$  from industrial complexes and vehicles in cities in Korea and China may have influenced the distribution of  $\Delta^{14}\text{C}$ , and that nuclear power plant operations may also have influenced that distribution.

### SAMPLES AND METHOD

Ginkgo leaves were collected across South Korea from mid-June to early July in 2010 and 2011 at the collection sites presented in Figure 1a. The density of the collection sites in cities is high; however, the average distance between collection sites in rural areas is about 70 km, and the density of collection sites in rural areas is not high.

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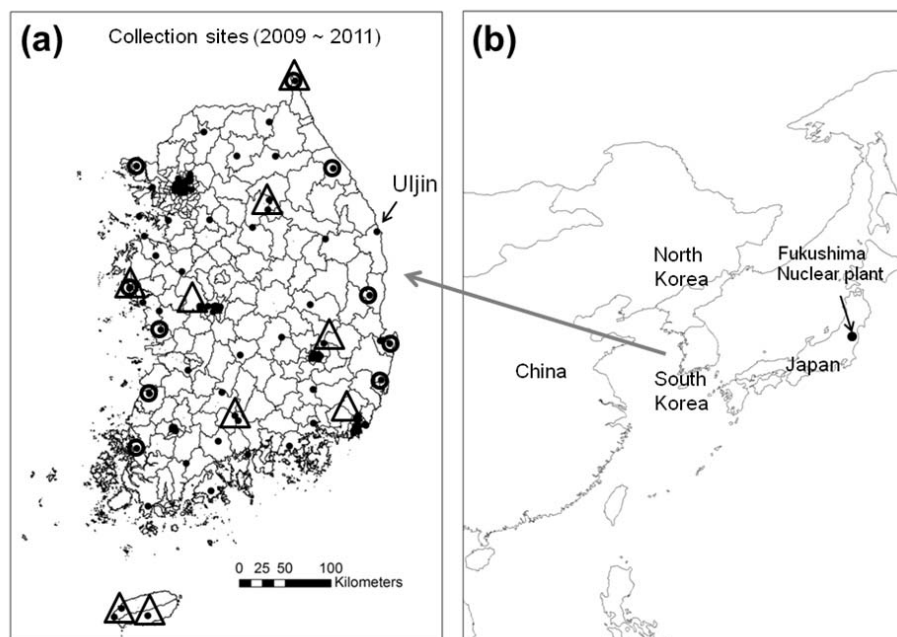


Figure 1 (a) Solid circles represent all of the collection sites. The densities of collection sites in urban areas are high, and densities in rural areas are low, with an average distance between collection sites in rural areas of about 70 km. Solid circles within larger circles are sites selected on the west and east coasts of Korea, which were relatively far from nuclear power plants, large cities, and industrial complexes. Solid circles with large triangles are the main clean air sites, at which samples have been collected from mid-June to early July since 2009. (b) Korea's location between China and Japan

Since different tree species have different growing periods, we have limited our study to 1 species, the ginkgo, in an attempt to obtain an average value during the same growing period. However, as a result of latitude and altitude effects, even the growing periods of the ginkgo can vary by 1 to 2 weeks (Park et al. 2013). Additionally, although isotopic fractionations of the measurement values were corrected during the  $\Delta^{14}\text{C}$  calculation, because large variation of isotopic fractionation may increase uncertainty slightly, the collection sample type was limited to 1 species of ginkgo for more accurate measurement (Park et al. 2013).

The ginkgo is a perennial and deciduous tree found throughout China, Korea, and Japan. The leaves of the ginkgo sprout in April and fall in October, and thus the data from the collected ginkgo leaves provide averages of the period from April to June. The ginkgo is resistant to air pollution, and consequently has been widely planted along many roads in metropolitan areas. Due to its symbolic importance in Buddhism, the ginkgo is also abundantly found near Buddhist temples, which are themselves typically located in mountainous areas that are generally free from fossil-fuel-produced  $\text{CO}_2$  (Park et al. 2013). Therefore, the ginkgo was widely available both in metropolitan and rural areas, and so it was the proper tree species for mapping in both areas.

Large ginkgo leaves of greater than 10 cm were collected for sample pretreatment; they were cleaned using pure water. The ginkgo leaves were treated using the ABA (acid-base-acid) method, combusted in an elementary analyzer, and reduced using an  $\text{H}_2$  and Fe catalyst (Hong et al. 2010a).  $^{14}\text{C}$  activity determination was performed using accelerator mass spectrometry (AMS) at the Korea Institute of Geoscience and Mineral Resources (KIGAM) in Daejeon (Hong et al. 2010b).

## RESULTS AND DISCUSSION

Figure 2 shows distribution maps of  $\Delta^{14}\text{C}$  in 2010 and 2011. The maps were made from all of the  $\Delta^{14}\text{C}$  data except for the  $\Delta^{14}\text{C}$  data from Uljin. The sampling site in Uljin is 2 km from the nuclear power plant that has 6X pressurized water reactors with an electric capacity of 5900 MWe (Wikipedia 2013).  $\Delta^{14}\text{C}$  at Uljin was very high, at 484.4‰ in 2010 and 78.3‰ in 2011, and would impractically distort the  $\Delta^{14}\text{C}$  distribution map; thus, the  $\Delta^{14}\text{C}$  distribution maps were constructed without these values.

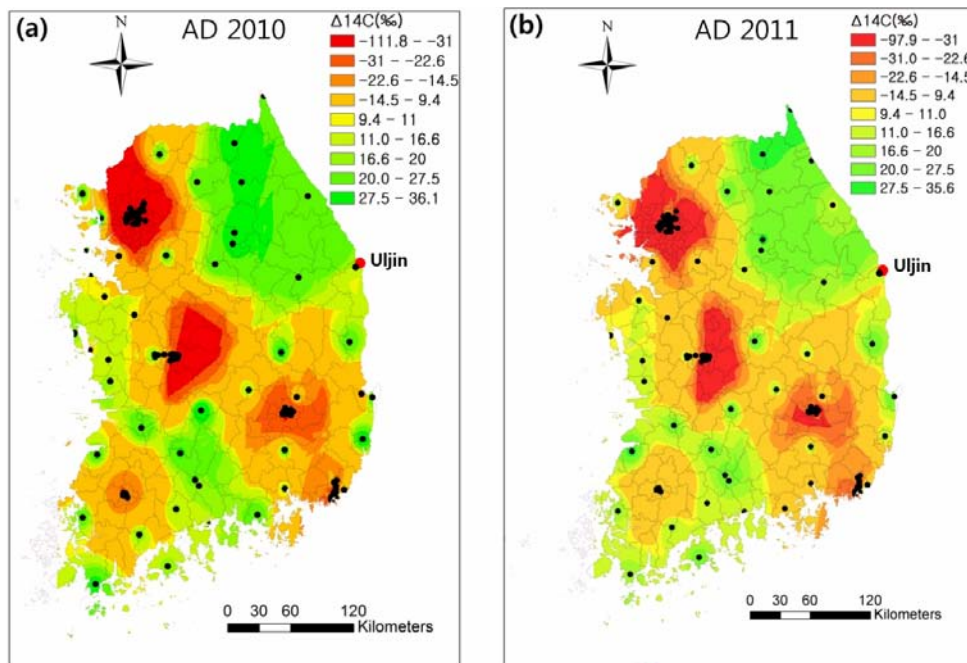


Figure 2 Distribution maps of  $\Delta^{14}\text{C}$  in 2010 (a) and 2011(b). These maps were made from all of the  $\Delta^{14}\text{C}$  data except for the  $\Delta^{14}\text{C}$  data in Uljin (near the nuclear power plant).

Korea is in an area of prevailing westerlies, so the main wind direction is to the west. The Uljin nuclear power plant is located on the east coast of Korea, so the assumption that the air near the nuclear power plant has high  $\Delta^{14}\text{C}$  blowing almost entirely into the East Sea, exerting almost no effect on other sites in Korea. Additionally, these  $\Delta^{14}\text{C}$  data from 2010 and 2011 were from an area near a nuclear power plant under normal operation, but the difference between 2010 and 2011 was very large, and so AMS measurement of Ginkgo leaves near a nuclear power plant may be a very useful tool for monitoring details of the status of nuclear power plant  $\Delta^{14}\text{C}$  distribution. The maps in Figure 2 show that, for 2010 and 2011, the  $\Delta^{14}\text{C}$  values in the NE and SW areas of Korea are larger than those in NW and SE areas of Korea. The densities of industrial complexes and population and urban areas in the NW and SE areas are high, as can be seen in Figure 3, so it is deduced that low values of  $\Delta^{14}\text{C}$  are mainly due to those industrial complexes, the high population density, and the presence of large cities.  $\Delta^{14}\text{C}$  values in the SW area are slightly lower than those in the NE area; this can be ascribed to densities of population and urban areas in the SW area are slightly larger than those in the NE areas and that fossil-fuel-produced  $\text{CO}_2$  from China influences the SW area slightly more because this area is closer to China.

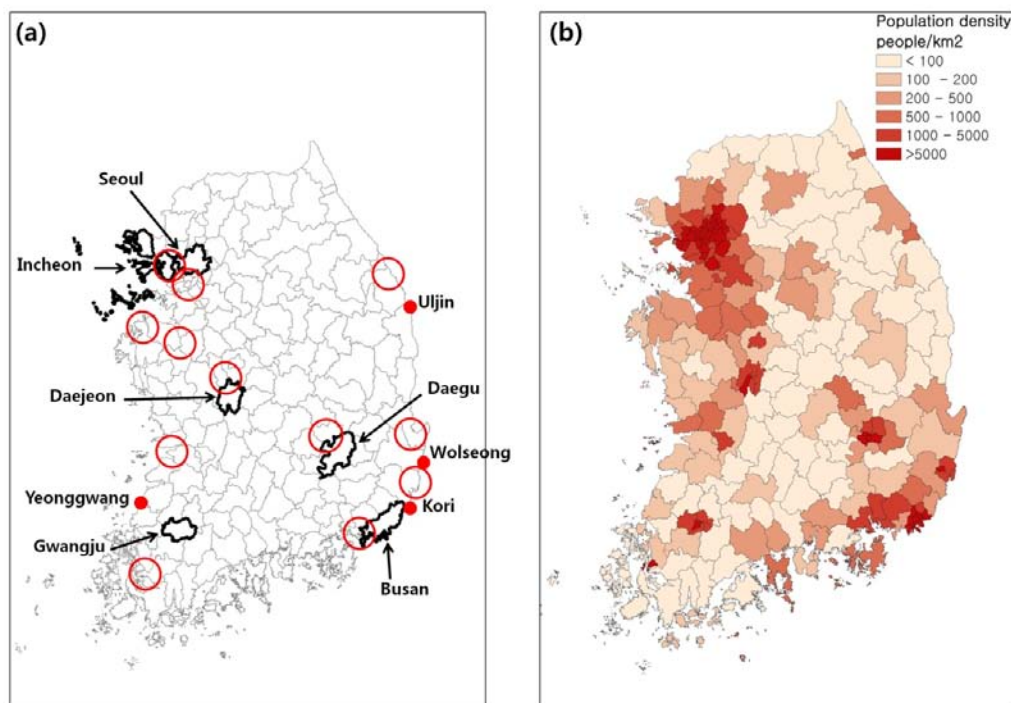


Figure 3 Locations of metropolises are indicated using arrows, the locations of industrial complexes with red open circles, and the locations of nuclear power plants with red closed circles; (b) provides a population density map in 2010 of Korea (Statistics Korea 2013).

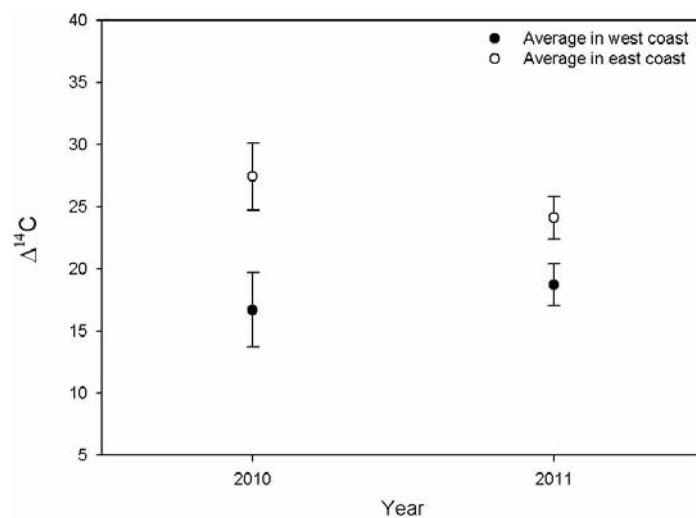


Figure 4 Averages of clean air sites selected on the western and eastern coasts in 2010 and 2011 are compared; the average of the  $\Delta^{14}\text{C}$  data for the east coast sites of Korea is larger than that of the west coast sites.

To clarify this “China effect,” 2 sets of  $\Delta^{14}\text{C}$  data from 2010 and 2011 were averaged. One set is an average of 5  $\Delta^{14}\text{C}$  data at points from sites selected on the west coast of Korea. These sites are

### *Distribution Maps of $\Delta^{14}\text{C}$ in 2010 and 2011 in Korea*

shown at Figure 1a and are relatively far from nuclear power plants, urban areas, and industrial complexes. The other graph shows an average of 5  $\Delta^{14}\text{C}$  data points from sites selected on the east coast of Korea; these sites are shown in Figure 1a and are also relatively far from nuclear power plants, cities, and industrial complexes. The sites selected on the west and east coasts are influenced only indirectly by these sources and are more strongly influenced by other factors. In Figure 4 and Table 1, averages and  $t$  test results for  $\Delta^{14}\text{C}$  data at the sites on both coasts are displayed. The averages of the  $\Delta^{14}\text{C}$  data at the sites selected on the east coast were larger than those selected on the west coast in both 2010 and 2011; the  $t$  test result in 2010 shows that the averages of  $\Delta^{14}\text{C}$  data at the sites selected on the east coast were also larger than selected on the west coast; however, the  $t$  test result in 2011 is different. The main wind direction in Korea is to the west, and so the sites selected on the west and east coasts can both be influenced by Chinese sources, but the west coast of Korea is closer to China (Figure 1b). On the other hand, the sites selected on the east coast can be influenced by fossil fuel  $\text{CO}_2$  from Korea to a greater degree than those sites on the west coast due to the prevailing westerly wind. From the  $t$  test results, the influence of China in 2010 was confirmed but then in 2011 disappeared. We can explain this result as either by less fossil fuel  $\text{CO}_2$  from China in 2011 in the west or more fossil fuel  $\text{CO}_2$  in Korea in the east, respectively.

Table 1 Averages and  $t$  test results of  $\Delta^{14}\text{C}$  data in sites selected on the west and east coasts of Korea. Table S1, online Supplementary files, has the full data set from which these averages derive.

	West coast (‰)	East coast (‰)	One-tailed test ( $t$ test)	Two-tailed test ( $t$ test)	Result
2010	$16.7 \pm 3.0$	$27.4 \pm 2.7$	0.0018 (<0.05)	0.0036 (<0.05)	Different
2011	$18.7 \pm 1.7$	$24.1 \pm 1.7$	0.24 (>0.05)	0.49 (>0.05)	Not different

$\Delta^{14}\text{C}$  in the clean air sites were averaged annually and are shown in Figure 5 and Table 2. Clean air sites are all of the collection sites except for those near the nuclear power plant (Uljin), metropolises, and industrial complexes. The main clean air sites are shown in Figure 1a. Ginkgo leaf samples from the main clean air sites were collected from 2009 (Park et al. 2013). The average  $\Delta^{14}\text{C}$  in the clean air sites was  $22.7 \pm 0.9\text{‰}$  in 2010 and  $21.1 \pm 0.6\text{‰}$  in 2011. The average  $\Delta^{14}\text{C}$  in 2011 decreased 1.6‰ in comparison with the average  $\Delta^{14}\text{C}$  in 2010; considering the  $t$  test result shown in Table 3, average  $\Delta^{14}\text{C}$  in 2010 and that in 2011 are not different, and this amount of annual decrease is small compared to the 5‰ decrease found in other research (Turnbull et al. 2009). Additionally, the amount of decrease in the averages of the  $\Delta^{14}\text{C}$  at the main clean air sites between 2010 and 2011 is slightly smaller than that of between 2009 and 2010. According to the  $t$  test result of Table 3, while the averages of the  $\Delta^{14}\text{C}$  at the main clean air sites between 2009 and 2010 are different, the averages of the  $\Delta^{14}\text{C}$  at the main clean air sites between 2010 and 2011 are not significantly different. This trend may be due to the Fukushima nuclear accident in 2011 in Japan, a reduction of the influence of fossil fuel  $\text{CO}_2$  from China in 2011, and/or a reduction of the production of fossil fuel  $\text{CO}_2$  in 2011 in Korea. The main reason for these overall trends needs to be made clear in future study.

Table 2 Averages of  $\Delta^{14}\text{C}$  data in the main clean air and clean air sites, Korea. Table S2, online Supplementary files, has the full data set from which these averages derive.

Year	Main clean air sites (‰)	Clean air sites (‰)
2009	$34.8 \pm 1.2$	
2010	$24.9 \pm 1.2$	$22.7 \pm 0.9$
2011	$23.1 \pm 1.1$	$21.1 \pm 0.6$

Table 3  $t$  test results of averages of  $\Delta^{14}\text{C}$  data in the main clean air and clean air sites, Korea. Table S3, online Supplementary files, has the full data set from which these averages derive.

Comparison year	$t$ test result		Result
	One-tailed test	Two-tailed test	
2009–2010 (Main clean air sites)	0.003 (<0.05)	0.007 (<0.05)	Different
2010–2011 (Main clean air sites)	0.26 (>0.05)	0.53 (>0.05)	Not different
2010–2011 (Clean air sites)	0.25 (>0.05)	0.49 (>0.05)	Not different

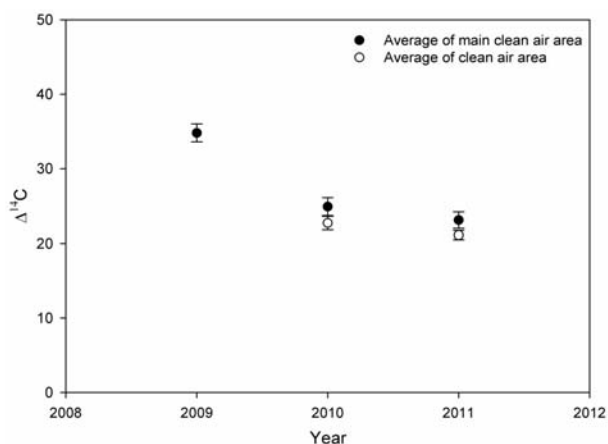


Figure 5  $\Delta^{14}\text{C}$  of the clean air sites and the main clean air site were averaged annually. The difference of the average  $\Delta^{14}\text{C}$  value at the main clean air site between 2009 and 2010 is slightly larger than that of between 2010 and 2011.

## CONCLUSION

Distribution maps of  $\Delta^{14}\text{C}$  in Korea for 2010 and 2011 in Korea were obtained, and local Suess effects in Korea according to metropolises and industrial complexes were confirmed.  $\Delta^{14}\text{C}$  measurement values at sites selected on the west coast are smaller than those on east coast for 2010 and 2011. This may be due to the effect of fossil fuel  $\text{CO}_2$  from China. The amount of decrease in the average  $\Delta^{14}\text{C}$  for the main clean air sites between 2010 and 2011 is smaller than that of between 2009 and 2010. More research is needed to determine what is the main reason for this discrepancy, whether it be the Fukushima nuclear accident in Japan, a reduction of the influence of fossil fuel  $\text{CO}_2$  from China in 2011, and/or a reduction of the production of fossil fuel  $\text{CO}_2$  in 2011 in Korea.

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