

Use of Tree Bark to Monitor Radionuclide Pollution

James D. Brownridge

Department of Physics, Applied Physics and Astronomy, State University of New York at Binghamton, Binghamton, NY 13901

The outer surface bark of many trees is an excellent monitoring source of fallout radionuclides. The accumulation and retention of these pollutants is evident by the presence of ¹⁰⁶Rh, ¹²⁵Sb, ¹⁴⁴Ce and ¹⁵⁵Eu in the outer layer of bark from many trees surveyed during this study. Because of their relative short half-life and no other known source, it is concluded tht these isotopes are the primary result of fallout from the Chinese nuclear weapon test of October 16, 1980. The accumulation and retention of these and other radionuclides suggest that tree bark is an ecosystem monitoring resource that should be exploited for these and possible other environmental pollutants.

The biota most often studied and used to monitor the ecosystem for long-lived nuclear fallout and released materials are lichens (Mattsson 1975; Taylor et al. 1979). However, most lichens are sensitive to other pollutants such as heavy-metals and may become extinct in heavily polluted areas (Koranda and Robison 1978; Folkeson 1984). In these areas and in general, tree bark could offer a viable alternate and second source.

As a monitoring system, tree bark appears to have been neglected. Therefore, the emphasis of this study was a broad survey of the detectability of gamma-ray emitting radionuclides in and on tree bark rather than a narrow quantitative study.

MATERIALS AND METHODS

Samples from 111 trees from ten species were collected in Broome County, New York, in the fall and winter of 1983. The majority of these samples was collected from two sites - one near the southern border of the county west of Interstate Highway 81 and the other east of Interstate Highway 81, 35 km to the north. Trees were selected that did not show any signs of macroscopic lichens on the bark that was to be removed. The outer dead bark was carefully dislodged from the surface of the living trees with a paint scraper or by hand, depending on the physical characteristics of the bark (Kozlowski 1973). When the paint scraper was used, no more than 1 mm of the outer surface was removed from a height of 0.5 to 2.5 m above ground level. The age of these trees ranges from approximately 15 to 125 years.

Live bark samples were collected by felling trees and returning large sections to the laboratory. In these cases, the bark was sectioned into outer and inner bark by carefully separating the epidermis and permiderm from the phloem with some cortex usually in both samples. In general, the bark collected from very young trees (<10 years old) was not sufficient in quantity for the desired sample size of 100 g dry weight and was therefore mixed with bark from adjacent trees of similar age and size.

In an attempt to reduce the minimum detectable activity to the lowest practical level, several large (up to 960 g) samples were ashed at 500°C. As can be seen in Figures 1 and 2, all isotopes identified in the ashed red maple bark sample were easily identified in the oven-dried hawthorn bark. Consequently, it was not necessary to ash or otherwise concentrate large volumes of bark to determine the relative abundance of nuclear fallout products if only the outer bark was used.

All samples not ashed were oven dried at 100°C ground to a powder and placed in a plastic bag for storage. For analysis, an aliquot, or the total sample, was placed in Marenell beakers and analyzed by direct gamma-ray spectrometry using an ultra low background Ge(Li) detector. The system consists of a Ge(Li) detector located in a lead shield 21 cm thick, a 4096 channel multichannel analyzer, an x-y plotter and a printer. The background under the ^{137}Cs 661.6 keV line was 0.030 ± 0.005 counts per minute for a typical 24-hour counting cycle. The minimum observable activity for ^{137}Cs was approximately 0.014 pCi/g of dry bark (Pasternack et al. 1971).

The precision of these measurements was confirmed by redrying, reweighing and recounting many of these samples during the course of this study. Data presented in Table 1 are typical of many samples analyzed and are, therefore, believed to be representative of tree bark in this area.

RESULTS AND DISCUSSION

The following gamma-ray emitting radionuclides from nuclear fission were observed in the outer layer of bark in several species of trees: 2.7y \$^{125}Sb\$; 284d \$^{14}\$^{16}Ce\$; 1.8y \$^{155}Eu\$; 1y \$^{16}Ru\$; 30y \$^{137}Cs\$. Also observed were \$^{7}Be\$ and \$^{40}K\$, along with U and Th and their daughter products. All of the fallout radionuclides observed were superficially deposited on the bark. Although \$^{137}Cs\$ may be taken up by the root system in varying degrees by some trees, it is evident, from this and a previous study, that by far most of the \$^{137}Cs\$ observed in the outer bark was superficially deposited (Brownridge 1984). Data presented in Table 1 are the result of the analysis of outer bark from ten species of trees, one species of lichen and two bald-faced hornet nests. The units are gamma-ray

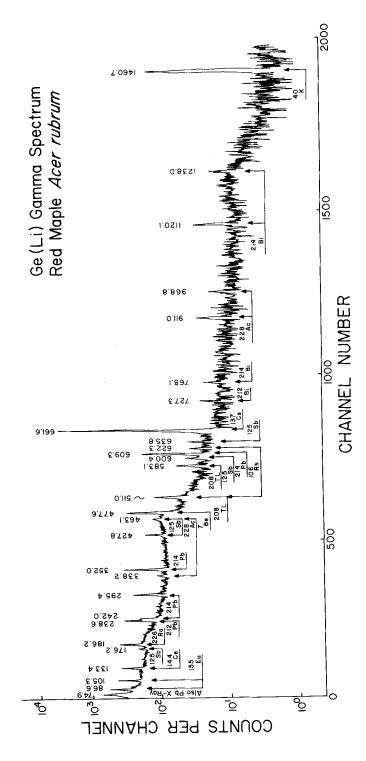


Figure 1. Gamma-ray spectrum of 960 g of oven dried red maple flaky bark collected on 26 July 1983 and ashed in a muffel furnace at $500^{\circ}\mathrm{C}_{\bullet}$. The number above each peak is the gamma-ray energy in keV.

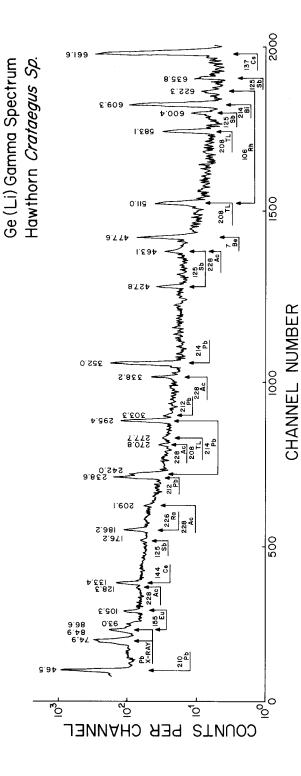


Figure 2. Gamma-ray spectrum of 74 g of oven dried hawthorn scale bark collected on 17 November 1983. The number above each peak is the gamma-ray energy in keV.

photons detected per minute per $100~{\rm g}$ of dry sample at the photon energy listed in Table 1.

With the exception of ¹³⁷Cs, no attempt was made to determine the absolute concentration of the observed radionuclides. However, the relative concentration may be determined from these data. While the surface soil 137Cs concentration measured during this study was relatively uniform from site to approximately 1 pCi/g, the outer tree concentrations varied from approximately 0.2 pCi/g for red pine to 3.0 pCi/g for red maple. Some of this variation is due in part to the inability to remove only the outer "contaminated bark" and none of the inner "clean bark". This is best illustrated in Table 2 where, unlike the samples in Table 1, these trees were felled and whole sections returned to the laboratory where the bark was carefully sectioned into outer and inner bark. The removal of the periderm resulted in the removal of all of the ⁷Be and most of the ¹³⁷Cs in many sam-In old trees with thick trunk bark, such as the white oak, if all of the bark between the cambium and the outer surface of the rhytidome is analyzed as one homogeneous sample, the 137Cs concentration is often only just above the detectable level of 0.014 pCi/q.

Trees that were less than 10 meters apart have been designated as adjacent for the purpose of this study and have been assigned the same sample number in Table 1. The purpose of this designation is to gain some insight into the response of different species to very nearly identical environmental fall-out and deposition conditions. Beryllium-7 produced by cosmic ray and $^{210}{\rm Pb}$, a daughter of $^{222}{\rm Rn}$, are also included for the same purpose. A careful examination of the data presented in Table 1 suggests that there is a positive correlation between the relative high collection and retention of $^{7}{\rm Be}$ and $^{210}{\rm Pb}$ and nuclear fallout radionuclides. These data confirm the suggestion made by Russel, et al. (1981) that $^{7}{\rm Be}$ may be used to describe the behavior of soluble superficial nuclides. It also suggests that $^{210}{\rm Pb}$ is also an equally useful natural tracer.

The response of different species to nearly identical environmental fallout and deposition conditions can be seen in Table 1, samples nos. 9, 10 and 11. Bark from a white oak tree, older than 75 years, is less efficient in accumulating and retaining $^{210}{\rm Pb}$ than red maple. On the other hand, bark from young white oak (<15 years old) has a much higher concentration of $^{210}{\rm Pb}$, $^{7}{\rm Be}$ and $^{144}{\rm Ce}$ than either red maple or the old white oak. The response does appear to be dependent on age or species.

nests. Units are gamma-ray photons detected per minute per 100 g of dry Relative concentration of gamma-ray emitting radionuclides on the surface of tree bark, in lichen and in bald-face hornet sample. Table 1.

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Sample		Age	210_{Pb}	7Be	155 _{Eu}	144 _{Ce}	137_{Cs}	226 _{Ra}	125_{Sb}
Number	Species	(years)	46.5 KeV	477.6 KeV	105.3 KeV	133.4 KeV	661.6 KeV	186.2 KeV	427.8 KeV
	Hop Hornbean(7)	<25	1.61±0.03	0.26 ± 0.01	0.16±0.02	0.15±0.02	2.28±0.02	0.18±0.02	0.08±0.01
2	White $Ash(1)*$	>50	2.59±0.03	0.15±0.01	0.19 ± 0.01	0.18 ± 0.01	2.58±0.02	0.07 ± 0.01	N.D.
2 A	White $Ash(1)*$	>50	2.72 ± 0.08	0.42 ± 0.03	0.20 ± 0.04	0.27 ± 0.04	1.47 ± 0.04	0.25 ± 0.04	N.D.
ന	Black Oak(1)	>75	0.66 ± 0.04	0.11±0.02	0.09 ± 0.02	N.D.	1.69 ± 0.04	0.12 ± 0.02	N.D.
7	Red Pine(7)	>25	0.33 ± 0.02	N.D.	N.D.	0.04 ± 0.01	0.28 ± 0.01	0.06 ± 0.01	N.D.
5	Red Maple(5)	>25	2.18 ± 0.06	0.23±0.02	0.21 ± 0.03	0.17 ± 0.04	2.79 ± 0.05	0.17 ± 0.03	0.13 ± 0.02
9	Apple(1)	>25	0.42 ± 0.05	0.13 ± 0.02	0.16 ± 0.04	0.09 ± 0.03	1.16 ± 0.04	0.27 ± 0.04	N.D.
7	Sugar Maple(1)	>50	1.18 ± 0.04	0.05 ± 0.01	N.D.	0.04±0.01	0.87 ± 0.03	N.D.	N.D.
_∞	White Pine (1)	>50	0.38 ± 0.01	0.03 ± 0.01	0.05 ± 0.01	N.D.	0.40 ± 0.01	0.04 ± 0.04	N.D.
6	Red Maple(1)	>50	0.85 ± 0.03	0.03 ± 0.01	0.08±0.01	0.07±0.01	1.33 ± 0.02	0.08 ± 0.01	N.D.
6	White $0ak(1)$	>75	0.21±0.02	0.02 ± 0.01	0.02 ± 0.01	N.D.	0.31 ± 0.01	0.22 ± 0.01	N.D.
6	White $0ak(3)$	<15	1,69±0,06	0.32 ± 0.03	N.D.	0.37 ± 0.04	0.67 ± 0.03	0.24 ± 0.04	N.D.
10	Hawthorn(5)	>15	2.74±0.07	0.19 ± 0.03	0.41 ± 0.05	0.38 ± 0.04	2.64 ± 0.06	0.24 ± 0.04	N.D.
10	Red Maple(1)	>50	0.79 ± 0.04	0.15 ± 0.03	0.05 ± 0.01	0.06 ± 0.02	0.49 ± 0.02	0.09 ± 0.02	N.D.
11	Red Maple(1)	>25	0.84 ± 0.04	0.13 ± 0.02	N.D.	N.D.	0.65 ± 0.03	0.05 ± 0.02	N.D.
11	White $Oak(1)$	>25	0.22 ± 0.02	0.02 ± 0.01	N.D.	N.D.	0.53 ± 0.01	0.12 ± 0.01	N.D.
12	Lichen(7) +	!	1.04±0.05	0.19±0.02	0.11 ± 0.03	0.07±0.03	2.07±0.05	0.12 ± 0.03	N.D.
12A	Red Maple(7)+	>25	0.84±0.04	0.0490.0	0.12 ± 0.02	0.05 ± 0.02	1.13 ± 0.03	0.12 ± 0.02	0.03±0.01
13	Lichen (2)	1	1.61±0.07	1.09±0.04	0.14 ± 0.04	0.23 ± 0.04	4.51±0.07	0.36 ± 0.04	N.D.
14	Bald-face Hornet	Nest	0.71±0.08	N.D.	N.D.	N.D.	1.46 ± 0.06	N.D.	N.D.
15	Bald-face Hornet	Nest	0.74 ± 0.04	0.55 ± 0.02	N.D.	0.07±0.02	0.55 ± 0.02	N.D.	N.D.

^() Number of trees in the sample \star From the same tree, 2A was 12 meters above ground + Lichen removed from this bark

Denotes not detectable

Table 2 An illustration of the superficial deposition of $^7\mathrm{Be}$ and $^{137}\mathrm{Cs}$ on tree stems. Units are gamma-ray photons detected per minute per 100 g of bark.

Species	Compartment	⁵Be	137 _{Cs}	4 ºK
Red Maple* 1 m above ground	Outer Bark Inner Bark	0.21 ± 0.03 ND	0.25 ± 0.02 ND	$\begin{array}{c} 0.08 & \pm 0.01 \\ 0.08 & \pm 0.01 \end{array}$
Red Maple* 8 m above ground	Outer Bark Inner Bark	0.64 ± 0.03 ND	0.07 ± 0.01 ND	0.21 ± 0.02 0.13 ± 0.01
American Beech	Outer Bark Inner Bark	0.14 ± 0.01 0.02 ± 0.01		$\begin{array}{ccc} 0.11 & \pm 0.01 \\ 0.10 & \pm 0.01 \end{array}$
American Beech	Outer Bark Inner Bark	0.14 ± 0.01 ND	0.32 ± 0.01 ND	$\begin{array}{ccc} 0.11 & \pm 0.01 \\ 0.08 & \pm 0.01 \end{array}$
White Oak	Outer Bark Inner Bark	0.14 ± 0.01 ND	0.66 ± 0.03 0.03 ± 0.01	$\begin{array}{ccc} 0.08 & \pm 0.01 \\ 0.19 & \pm 0.01 \end{array}$
Black Birch+	Outer Bark Inner Bark	10.90 ± 0.40 0.11 ± 0.08	4.20 ± 0.30 0.97 ± 0.08	2.95 ± 0.20 2.56 ± 0.09

^{*} From the same tree + Bark ashed at 500°C

The trees with the overall highest concentration of radionuclides in their outer bark are hornbeam, white ash, red maple and hawthorn.

Since lichens are the biota frequently used to monitor long lived fallout radionuclides in the ecosystem, a comparison between tree bark and lichens in the same environment is desirable. The lichen chosen for this study was parmelia sp. growing on the bark of red maple trees and a building roof. The results are presented in Table 1, samples 12, 12A and 13. Not presented in Table 1 is the relative concentration of $^{40}{\rm K}$ in lichen from trees, the tree bark under the lichen and lichen removed from a building roof. They are 0.24 \pm 0.02, 0.07 \pm 0.01 and 0.61 \pm 0.03 counts per minute per 100 g respectively.

Also included in this survey are two bald-faced hornet nests, samples 14 and 15 in Table 1. Sample 14 was removed from an open field in the fall of 1979 and used as a display item until it was analyzed in March 1984. Sample 15 was constructed in an apple tree in an open field during the summer of 1983. It was removed from the tree and analyzed in May 1984. In addition to the isotopes listed in Table 1, the

ND Denotes not detectable

relative concentrations of 40 K in the nest are 0.33 \pm 0.01 and 0.32 \pm 0.03 counts per minute per 100 g, respectively. The bald-faced hornet chews wood from weathered surfaces to build its coarse paper nest. Therefore, the nest would be expected to contain superficially deposited materials. Notice that while 210 Pb and 40 K concentrations are the same for both nests, the 1979 nest (No. 14) has a higher concentration of 137 Cs than the nest built in 1983. Since the bald-face hornet uses wood fiber from weathered surfaces to build its nest, one would expect environmental erosion to have removed some 137 Cs from exposed nest building materials between 1979 and 1983.

There are very little data in the literature on the environmental half-time for radionuclides deposited on tree bark (Miller & Hoffman 1983). Russel et al. (1981) measured the relative rate of accretion of 144 Ce on needle and twig tissue in the white pine and concluded that a loss constant of zero could be assumed for twig tissue. If the last major release of 284d 144 Ce into the atmosphere was the Chinese nuclear explosion of October 16, 1980, then 144 Ce and the other fission products were indeed held tenaciously as suggested by Russel et al. (1981). Field studies are currently underway to measure the environmental half-time of several fission products on the surface of tree bark.

There have been many studies of plants as monitoring systems for environmental pollutants; however, with few exceptions these have dealt with lichens, grasses, food crops, foliages and the whole tree or shrub (Adriano et al. 1981; Koranda & Robison 1978). Brown and McFarline (1972) suggested that in the absence of man-made collection systems, dormant plants could provide useful information about the release of fallout material. I suggest that tree bark, and in particular the outer surface, may be one of the best and most abundant sources to monitor for environmental pollutants.

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