

# Occurrence of Cesium-137 and Other Radionuclides in the Surface Layers of Soil in Ohio and Antarctica<sup>1</sup>

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**ABSTRACT.** The testing of nuclear weapons in the atmosphere in the 1960s and the accidental explosion of a nuclear reactor in 1986 near Chernobyl, Ukraine, caused large amounts of fission-product radionuclides to be deposited over most of the surface of the Earth. The present study was undertaken in order to compare the decay rates of <sup>137</sup>Cs in the surface layer of soil in central Ohio at 40°N and in Antarctica at 77°S latitude. The measurements were made by means of gamma-ray spectrometry on 10 gram samples of bulk soil collected from the surface. The results indicate that the decay rate of <sup>137</sup>Cs in the Ohio soil sample is  $(2.2 \pm 0.8) \times 10^{-1}$  pico Curies/g, whereas the rate of decay of this radionuclide in the antarctic soils is below the limit of detection ( $<1 \times 10^{-2}$  pCi/g). In addition, roots in the Ohio soil do not contain detectable <sup>137</sup>Cs, confirming that this radionuclide is not absorbed by plant roots because of its strong sorption on the surfaces of clay minerals. Soils in Ohio and Antarctica do contain long-lived unstable <sup>40</sup>K, as well as short-lived unstable daughters of <sup>238</sup>U (<sup>214</sup>Pb and <sup>214</sup>Bi) and of <sup>232</sup>Th (<sup>228</sup>Ac, <sup>212</sup>Pb, <sup>208</sup>Tl, and <sup>228</sup>Th). The apparent absence of <sup>137</sup>Cs in the surface layer of antarctic soils is most likely the result of nondeposition of radioactive fallout at the extreme southern latitudes of Antarctica.

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## INTRODUCTION

The accidental explosion of a nuclear reactor near Chernobyl, Ukraine, on 26 April 1986, released about  $10 \times 10^{16}$  Becquerels (Bq) of <sup>137</sup>Cs which is a radioactive fission product of <sup>235</sup>U with a half-life of 30.1 years (1.0 Becquerel = 1 disintegration per second). The plume of radioactive dust arrived over the USA about 370 hours (15.4 days) after the explosion and deposited  $2.8 \times 10^{14}$  Bq of <sup>137</sup>Cs, or about 0.3% of the total amount of <sup>137</sup>Cs that was emitted. Much larger amounts of <sup>137</sup>Cs were deposited on countries adjacent to the Ukraine, including Poland ( $9.2 \times 10^{15}$  Bq), Romania ( $6.7 \times 10^{15}$  Bq), and Sweden ( $3.4 \times 10^{15}$  Bq) (Anspaugh and others 1988; Hakanson 1999; Hinton and others 1999).

Prior to this latest episode, <sup>137</sup>Cs and other products of nuclear fission of <sup>235</sup>U were released into the atmosphere as a result of testing nuclear weapons starting in 1944 and ending in 1980 (Leifer and others 1984). As a result, the deposition rate of <sup>137</sup>Cs in the Chicago area peaked in 1963 at about  $3 \times 10^2$  milli Curies/km<sup>2</sup> (1 Curie =  $3.7 \times 10^{10}$  Bq) and declined to less than  $2 \times 10^1$  milli Curies/km<sup>2</sup> after 1967 (Gustafson and others 1970).

The purpose of the present study was to measure the rate of decay of <sup>137</sup>Cs in the surface layers of soils in Ohio and Antarctica in order to determine whether this and other products of nuclear fission of <sup>235</sup>U are transported through the atmosphere from point sources in the northern hemisphere to the extreme southern latitudes of Antarctica. Previous work by Machta and others (1956) was inconclusive because of the absence of relevant data for the southern hemisphere. Information compiled by UNSCEAR (1969) and reviewed by Eisenbud (1987) indicates only that the cumulative accumulation of radioactive <sup>90</sup>Sr in soils of Antarctica is less than 10%

of that present at about 40°N latitude in the Pacific Ocean and in North America. The global north to south distribution of <sup>137</sup>Cs and other fallout radionuclides is a proxy for the distribution of other kinds of industrial contaminants that are released predominantly in the northern hemisphere. The present study is based on <sup>137</sup>Cs rather than on <sup>90</sup>Sr because <sup>137</sup>Cs emits a gamma ray with an energy of 661.7 Kev (kilo-electron volts) as it decays to stable <sup>137</sup>Ba, whereas <sup>90</sup>Sr does not emit a gamma ray and therefore cannot be detected by gamma-ray spectrometry.

## MATERIALS AND METHODS

A sample of soil weighing 5292 g (dry) was collected during the spring of 1997 from the surface of the prairie on the Marion Campus of the Ohio State University. The sample was taken from an area of 324 square inches (0.21 m<sup>2</sup>) to a depth of 2 inches (5 centimeters). The sample was dried in air, homogenized by mixing, and the enclosed plant roots were removed. The prairie site was chosen because the soil has not been disturbed by cultivation since before the reactor accident at Chernobyl.

In order to characterize the Ohio soil sample used in this study, its grain-size distribution was determined by sieving and its major minerals were identified by X-ray diffraction using Cu K-alpha X-radiation (Department of Geological Sciences, The Ohio State University, Columbus). In addition, its chemical composition was determined by X-ray fluorescence spectrometry (X-Ray Assay Laboratories, Toronto, Ontario). The results of the chemical analysis are listed in Table 1. The grain-size distributions and chemical compositions of the Antarctic soil samples will be presented elsewhere.

Ten gram aliquots of the less than 125 micrometer (μm) fractions of soil and a similar amount of plant roots in the Ohio soil were analyzed by gamma-ray spectrometry (Teledyne Brown Engineering Environmental Services of Westwood, NJ). The observed counting rates

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TABLE 1

Chemical composition of surface soil collected on the prairie, Marion Campus, The Ohio State University.

Oxide	Concentration %	Oxide	Concentration %
SiO <sub>2</sub>	75.3	Fe <sub>2</sub> O <sub>3</sub> <sup>‡</sup>	3.17
Al <sub>2</sub> O <sub>3</sub>	9.44	MnO	0.04
CaO	1.1	TiO <sub>2</sub>	0.808
MgO	0.69	P <sub>2</sub> O <sub>5</sub>	0.12
Na <sub>2</sub> O	1.18	Cr <sub>2</sub> O <sub>3</sub>	0.04
K <sub>2</sub> O	1.76	LOI <sup>†</sup>	6.6

<sup>‡</sup> Total iron

<sup>†</sup> Loss on ignition: Water of hydration and carbon dioxide (Analyst: X-Ray Assay Labs, Toronto, Ontario).

were converted to decay rates by reference to radiochemical standards. The results in Table 2 are expressed in terms of pico Curies per gram of sample (p Ci/g). The prefix "pico" (p) represents a factor of 10<sup>-12</sup>. Therefore, a decay rate of 1.0 pCi is equal to 3.70 × 10<sup>-2</sup> Bq.

## RESULTS

### Mineralogy and Chemical Composition

The grain-size distribution of the Ohio soil in Figure 1 indicates that it consists of a mixture of sand (55.8%) and silt (42.8%). Clay-sized particles make up only 1.3% by weight. The soil collected on the prairie at Marion is therefore classified as a sandy loam. The soils in the ice-

TABLE 2

Decay rates of <sup>137</sup>Cs and naturally occurring radionuclides in the less than 125 micrometer (μm) size fraction of the surface layer of soils in Ohio and Antarctica in picoCuries/gram (pCi/g).

Nuclide	Ohio		Antarctica		
	Soil	Roots	Beacon	Wright	Taylor
<sup>137</sup> Cs	2.2 × 10 <sup>-1</sup>	< 1 × 10 <sup>-2</sup>	< 8 × 10 <sup>-2</sup>	< 8 × 10 <sup>-2</sup>	< 8 × 10 <sup>-2</sup>
<sup>40</sup> K	11	32.5	9.7	11	15
<sup>208</sup> Tl	2.7 × 10 <sup>-1</sup>	n.d. <sup>†</sup>	1.2 × 10 <sup>-1</sup>	1.4 × 10 <sup>-1</sup>	2.0 × 10 <sup>-1</sup>
<sup>212</sup> Pb	7.2 × 10 <sup>-1</sup>	n.d.	5.4 × 10 <sup>-1</sup>	5.9 × 10 <sup>-1</sup>	6.9 × 10 <sup>-1</sup>
<sup>214</sup> Pb	7.2 × 10 <sup>-1</sup>	n.d.	2.8 × 10 <sup>-1</sup>	2.7 × 10 <sup>-1</sup>	3.8 × 10 <sup>-1</sup>
<sup>214</sup> Bi	5.5 × 10 <sup>-1</sup>	n.d.	2.6 × 10 <sup>-1</sup>	n.d.	2.5 × 10 <sup>-1</sup>
<sup>228</sup> Ac	6.3 × 10 <sup>-1</sup>	n.d.	n.d.	6.8 × 10 <sup>-1</sup>	6.9 × 10 <sup>-1</sup>
<sup>228</sup> Th	n.d.	0.73 × 10 <sup>-1</sup>	n.d.	n.d.	n.d.

<sup>†</sup>n.d. = not detected

Locations:	Prairie, Marion, OH:	40°33'N, 83°05'W
	Beacon Valley, Antarctica:	77°49'S, 160°39'E
	Wright Valley, Antarctica:	77°31'S, 161°50'E
	Taylor Valley, Antarctica:	77°37'S, 163°00'E

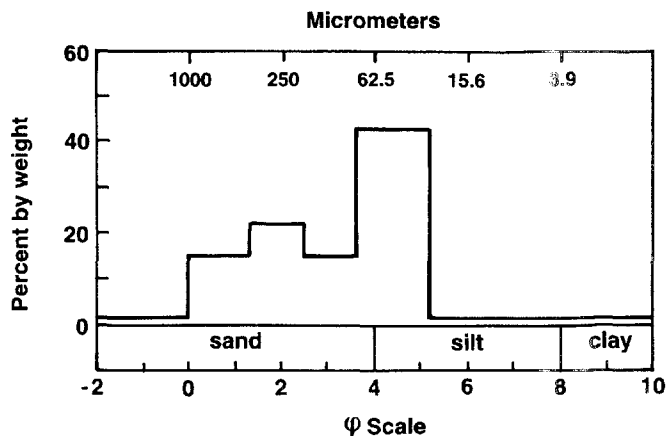


FIGURE 1. Grain-size distribution of the surface layer of a soil sample from the prairie on the Marion Campus of The Ohio State University. The grain diameters are indicated both in terms of micrometers as defined by the Wentworth scale and by the  $\phi$  scale where  $\phi$  is the negative exponent of 2 (for example,  $\phi = 2$  corresponds to  $2^{-2} = 1/4$  millimeter or 250 micrometers).

free valleys of southern Victoria Land in Antarctica (Beacon, Wright, and Taylor valleys) are likewise composed primarily of sand and silt and contain less than 5% of clay in all cases.

The X-ray diffraction pattern of the Ohio soil indicates the presence of quartz, illite, feldspar, and mixed layer-clay. Several other minerals were looked for but were not found: calcite, vermiculite, hematite, goethite, biotite, muscovite, and kaolinite. The apparent absence of these minerals means only that their abundances in the soil sample were generally less than 5%. However, the absence of calcite in surface soil is expected because this mineral has been leached by rainwater from till in central Ohio to a depth of about one meter (Place 1994).

The most abundant oxide component in the Ohio soil sample in Table 1 is silica (SiO<sub>2</sub> = 75.3%) which occurs in quartz and in alumino-silicate minerals (for example, clay minerals and feldspars). It is followed in abundance by alumina (Al<sub>2</sub>O<sub>3</sub> = 9.44%), iron oxide (Fe<sub>2</sub>O<sub>3</sub> = 3.17%), and alkali metals (Na<sub>2</sub>O = 1.18%, K<sub>2</sub>O = 1.76%). The concentrations of CaO, MgO, TiO<sub>2</sub>, MnO, P<sub>2</sub>O<sub>5</sub>, and Cr<sub>2</sub>O<sub>3</sub> are all about 1% or less. Loss on ignition (LOI = 6.6%) represents water released by clay minerals and by oxyhydroxides of iron and aluminum, as well as CO<sub>2</sub> formed by combustion of residual organic matter. In general, the chemical composition of this soil sample is compatible with its development from till of Wisconsinan age.

### Cesium-137 in Ohio Soil

The gamma-ray analysis of the surface layer of soils in Ohio in Table 2 indicates the presence of detectable amounts of <sup>137</sup>Cs, <sup>40</sup>K, <sup>208</sup>Tl, <sup>212</sup>Pb, <sup>214</sup>Pb, <sup>214</sup>Bi, and <sup>228</sup>Ac. The decay rate of <sup>137</sup>Cs in the Ohio soil at Marion is (2.2 ± 0.8) × 10<sup>-1</sup> pCi/g, but <sup>137</sup>Cs is not detectable in the soils of Antarctica. Since <sup>137</sup>Cs is a product of nuclear fission of <sup>235</sup>U and does not occur naturally, it is an anthropogenic contaminant. The decay rate of <sup>137</sup>Cs in the soil on the Marion prairie is comparable in magnitude to the results of Bajracharya and others (1998) for three soil samples in Clark County, OH, which yielded an average

decay rate of  $6.0 \times 10^{-1}$  pCi/g. The decay rate of  $(2.2 \pm 0.8) \times 10^{-1}$  pCi/g of  $^{137}\text{Cs}$  in the soil of the Marion prairie corresponds to a concentration of  $2.5 \pm 0.9$  femtograms of  $^{137}\text{Cs}$  per gram, where 1 femtogram =  $10^{-15}$  grams.

The origin of  $^{137}\text{Cs}$  in the soil of the prairie on the Marion campus of Ohio State University is not known. However, in the absence of a local source of this contaminant (for example, a nuclear reactor or a repository of spent nuclear fuel), the most likely source is the explosion of the nuclear reactor in Chernobyl in 1986, in addition to fallout deposited in the 1950s and 1960s. Fallout that was deposited on actively farmed land is diluted when it is mixed into the soil by cultivation. Prairies have the special virtue of being repositories of all kinds of particles that are deposited from the atmosphere, including not only radioactive fallout, but also terrestrial and extraterrestrial dust.

The decay rate of  $^{137}\text{Cs}$  of grass roots and other organic matter removed from the prairie soil at Marion is less than  $1 \times 10^{-2}$  pCi/g which is more than  $22 \pm 8$  times less than its decay rate in the soil. This result confirms that plants are unable to absorb cesium through their roots because of the strong sorption of this element on the surfaces of clay minerals (Roca and others 1997; Southard and Graham 1992). In contrast to cesium, potassium is an essential nutrient that enters plants through their roots. The data in Table 2 indicate that the decay rate of  $^{40}\text{K}$  in plant roots in the soil of the Marion prairie is about three times higher than that of the soil.

The other radionuclides present in the Ohio soil are all radioactive progeny of long-lived and naturally occurring  $^{238}\text{U}$  and  $^{232}\text{Th}$ . These elements occur naturally in all terrestrial rocks and in the soils derived from them (Faure 1986). Therefore, the presence of their intermediate decay products in all soils is expected. The radionuclides  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  are descendents of  $^{238}\text{U}$ , whereas  $^{208}\text{Tl}$ ,  $^{212}\text{Pb}$ ,  $^{228}\text{Ac}$ , and  $^{228}\text{Th}$  are decay products of  $^{232}\text{Th}$ .

### Cesium-137 in Antarctic Soils

The apparent absence of  $^{137}\text{Cs}$  from the surface layer of antarctic soil is consistent with the global dispersal pattern of radioactive fallout documented by Machta and others (1956), Leifer and others (1984), and Eisenbud (1987) all of which indicate that the deposition of radioactive fallout from the atmosphere decreases with increasing latitude both north and south of the source. Bartraya and others (1993) determined that only 3.1% of the global output of radioactive fallout reaches Antarctica and that the total cumulative amount of  $^{137}\text{Cs}$  deposited in Antarctica between 1955 and 1980 is about 7600 Bq. Many studies reported in the literature have failed to detect  $^{137}\text{Cs}$  in antarctic soil leading to speculation that the deposition of this radionuclide was sporadic and did not cover all parts of the continent (Strand and Holms 1993). However, the absence of  $^{137}\text{Cs}$  in the Antarctic soils may also be caused by lack of retention caused by the low abundance of smectite clays.

### CONCLUSIONS

The surface layer of a sandy loam soil on the prairie of the Marion campus of Ohio State University (latitude

$40^{\circ}\text{N}$ ) contains radioactive  $^{137}\text{Cs}$  produced by nuclear fission of  $^{235}\text{U}$  in nuclear reactors and in nuclear weapons. The  $^{137}\text{Cs}$  in the soil of the Marion prairie probably originated from the accidental explosion of a nuclear reactor in April of 1986 in Chernobyl, Ukraine, and from previous nuclear weapons tests in the northern hemisphere.

Surface layers of three soils in the ice-free valleys of southern Victoria Land, Antarctica, (latitude  $77^{\circ}\text{S}$ ) do not contain detectable  $^{137}\text{Cs}$  primarily because of nondeposition at the extreme southern latitude. This result therefore confirms that Antarctica is significantly less contaminated by deposition of radioactive fallout and, by analogy, by other industrial waste products that are transported through the atmosphere.

All of the soils included in this study contain long-lived  $^{40}\text{K}$  and the unstable progeny of naturally-occurring long-lived  $^{238}\text{U}$  and  $^{232}\text{Th}$ . The presence of these radionuclides is a natural consequence of the chemical compositions of ordinary rocks and of the soils that develop from them.

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