

## Natural and Fission-produced Radioactivity in Four Indiana Soils<sup>1</sup>

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Continued testing of nuclear weapons and increasing developments in reactors for use in power production have caused growing concern about the levels of fission-produced radioactivity in the earth's surface. In order to establish reference points for the levels of natural and fission-produced radioactivity in Indiana, bulk samples of the 0-6" layer of four well-characterized soils were collected for determination of gamma ray activities. Periodic checking of samples from these sites will provide information on the build-up of radioactivity.

### Introduction

Naturally occurring radioisotopes in soils include the thorium and uranium series and K<sup>40</sup>. The amounts of these isotopes depend on materials from which the soils are developed and the extent of weathering and leaching.

The gamma-ray spectra emitted by present-day surface soils reveal the presence of several lines which do not pertain to the thorium and uranium series or to K<sup>40</sup>. These additional gamma-ray lines come from radioactive fission products in fall-out (2).

Fission-produced radioisotopes in fall-out include Zr<sup>95</sup>-Nb<sup>95</sup>, Sr<sup>90</sup>, Rh<sup>106</sup>-Ru<sup>106</sup>, I<sup>131</sup>, Cs<sup>137</sup>, Ba<sup>140</sup>-La<sup>140</sup>, and Ce<sup>144</sup>-Pr<sup>144</sup>. Of these, Cs<sup>137</sup> and Sr<sup>90</sup> are of greatest concern because of their relatively long half-lives and their entrance into the food chain. Since the behavior of Cs<sup>137</sup> and Sr<sup>90</sup> in soils and plants is somewhat similar to that of K and Ca, respectively, it is of considerable concern to be able to detect and measure these radioisotopes.

In natural environments, radioactive fission products can get into plants both through the aboveground portions of the plant from external surface contamination (direct absorption) and through the roots by uptake from the contaminated soil.

Sr<sup>90</sup> has probably received the greatest attention because of its accumulation in the bones of the human body. Kulp and Schuler (3) have made detailed studies of Sr<sup>90</sup> in man and as a result have developed an equation which makes it possible to predict the average number of micromicrocuries of Sr<sup>90</sup> per gram of calcium in milk when data on rate of fallout in rain and cumulative deposition in soils are available.

Thus, measurements of the cumulative deposition of fission-produced radioisotopes in soils and the amount of fallout in precipitation become very important in making predictions of levels of Sr<sup>90</sup> in the milk supply as well as in assessing other possible harmful radiation effects.

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The measurement of naturally occurring and fission-produced radioisotopes generally requires cumbersome radiochemical separation procedures. Gustafson et al. (2) and Mortensen (6) have recently applied scintillation spectrometry for the detection and measurement of gamma emitters in soils and plants. Gustafson (1) has particularly emphasized the desirability of using  $Cs^{137}$  as a monitor for  $Sr^{90}$  in soils.

Since  $Cs^{137}$  is a gamma-emitter it can be measured by gamma-ray spectrometry.  $Sr^{90}$ , on the other hand, is a beta-emitter and its direct determination requires time-consuming and costly chemical separation processes.

During the fission process, approximately 1.76 atoms of  $Cs^{137}$  are formed for each atom of  $Sr^{90}$ . When the half-lives of these two are considered, the  $Cs^{137}/Sr^{90}$  activity ratio is found to be 1.83. This ratio should be universally prevalent in fallout if no fractionation occurs from the time of detonation to the time of deposition on the ground (1).

After the radioisotopes are deposited on the soil surface fractionation is quite likely to occur with  $Cs^{137}$  being more strongly retained than  $Sr^{90}$ . If the soil is sampled to a depth of 6 inches, virtually all radioactive fission products should be included and the  $Cs^{137}/Sr^{90}$  ratio should be fairly constant.

Gustafson (1) examined the  $Cs^{137}/Sr^{90}$  ratio data for numerous samples of rain water and surface soils. He found the ratio in the upper 2 inches of the soils to vary from 1.3 to 1.8 and concluded that the  $Sr^{90}$  concentration could be estimated within  $\pm 20$  percent by measuring the  $Cs^{137}$  and dividing by 1.6.

#### Experimental Procedure

Bulk soil samples of 80-100 lbs. were taken from the 0-6" layer of adjacent virgin and cultivated sites. The locations and sampling

TABLE 1. Location of Sample Site and Sampling Date.

Plainfield sand—Porter County, E $\frac{1}{2}$ NE $\frac{1}{4}$ NE $\frac{1}{4}$ Section 12, T 37 N, R 5 W. Sampled May 3, 1961.
Crosby silt loam—Hancock County, NW $\frac{1}{4}$ NE $\frac{1}{4}$ Section 20, T 16 N, R 7 E. Sampled May 17, 1961.
Clermont silt loam—Franklin County, SW $\frac{1}{4}$ Section 32, T 8 N, R 2 W. Sampled May 17, 1961.
Zanesville silt loam—Dubois County, S $\frac{1}{4}$ SW $\frac{1}{4}$ SE $\frac{1}{4}$ Section 21, T 3 S, R 5 W. Sampled May 16, 1961.

dates are given in Table 1. The samples were air-dried, crushed and passed through a 2 mm. sieve.

The gamma-ray spectra were taken with a scintillation counter which consisted of 5 x 4-inch NaI(Tl) crystal mounted on a DuMont 6364 photomultiplier tube enclosed in stainless steel. A 2 kg. soil sample was placed in a special stainless steel container which allowed a sample thickness of 1 inch to be distributed around and over the scintillation crystal. Background was reduced by the use of thick steel shielding around the sample and detector. Pulse-height analysis was carried out with an Argonne type 256 channel analyzer.

The concentration of fission and natural radioactivity in the soil samples was determined by using sources of known activity incorporated in a mock soil consisting of  $\text{Na}_3\text{PO}_4$ .

In preliminary measurements the activities of the virgin and cultivated samples from a given site were found to be essentially the same. The measurements reported are the average for the pair. Activities were corrected for decay to sampling date.

Gustafson et al. (2) showed that 97 percent of fission activity in soil occurred at a depth of 0-3". The use of samples from 0-6" depth should thus insure sampling of all fission activity that had accumulated up to the time of sampling.

### Results and Discussion

The levels of naturally occurring gamma-ray activity are shown in Table 2. The values agree well with those given by Gustafson et al.

TABLE 2. Naturally Occurring Gamma-ray Activity in Four Indiana Soils (0 - 6 Inches).

Soil	U+	Th+	K <sup>40</sup>
	x 10 <sup>-3</sup> g./kg.	x 10 <sup>-3</sup> g./kg.	g./kg.
Plainfield s	0.76	1.74	15.1
Clermont sil	3.95	9.36	14.2
Crosby sil	3.05	7.76	12.3
Zanesville sil	3.17	6.14	14.1

(2) and Mortensen (6). It is perhaps significant that the Clermont soil is one of the older soils in the state and its content of U+ and Th+ is the highest of the four soils examined.

The levels of fission-produced gamma-ray activity are shown in Table 3. The Plainfield soil has the lowest levels of the four soils; this suggests the possibility that some loss by leaching may occur on this very porous soil. The values are in good agreement with those of Gustafson et al. (2).

TABLE 3. Fission-produced Gamma-ray Activity and Estimated Sr<sup>90</sup> Content in Four Indiana Soils (0-6 Inches).

Soil	Cs <sup>137</sup>	Ru <sup>106</sup> -Rh <sup>106</sup>	Ce <sup>144</sup> -Pr <sup>144</sup>	Estimated Sr <sup>90</sup> <sup>1</sup>
	millicuries per square mile			
Plainfield s	35	76	65	22
Clermont sil	50	108	170	31
Crosby sil	58	150	170	36
Zanesville sil	43	157	177	27

1. Estimated by dividing the Cs<sup>137</sup> activity by 1.6 (Gustafson, Sci. 130:1404-1405, 1959).

The last column gives the estimated concentration of  $\text{Sr}^{90}$  using the activity ratio  $\text{Cs}^{137}/\text{Sr}^{90}$  of 1.6 as suggested by Gustafson (1). This is also in good agreement with prevailing  $\text{Sr}^{90}$  values for this geographic area.

The equation for predicting  $\text{Sr}^{90}$  content of milk as given by Kulp and Schulert (3) is as follows:

$$Q_m = AX + BY$$

where  $Q_m$  is the average number of micromicrocuries of  $\text{Sr}^{90}$  per gram of calcium in milk;  $X$  is the average rate of deposition of  $\text{Sr}^{90}$  in mc./mi.<sup>2</sup> per 6 months for a particular growing season;  $Y$  is the cumulative deposit of  $\text{Sr}^{90}$  in mc./mi.<sup>2</sup> measured at the midpoint of the growing season;  $A$  is the number of micromicrocuries of  $\text{Sr}^{90}$  per gram of calcium in milk, due to direct absorption, divided by the number of mc./mi.<sup>2</sup> of  $\text{Sr}^{90}$  deposited during the growing season; and  $B$  is the number of micromicrocuries of  $\text{Sr}^{90}$  per gram of calcium in milk, due to the cumulative deposit, divided by the total mc./mi.<sup>2</sup> of  $\text{Sr}^{90}$  deposited to the midpoint of the growing season since the start of nuclear testing. The coefficients  $A$  and  $B$  have the magnitude of 0.65 and 0.12, respectively.

In the fall of 1958 Soviet testing introduced about 1.3 megacuries of  $\text{Sr}^{90}$  into the Northern Hemisphere. This produced fallout of 14 mc./mi.<sup>2</sup> from April 1 to October 1, 1959 and a total deposition to July 1 of about 16 mc./mi.<sup>2</sup> (3). In the recent Soviet series of September-October 1961, it is estimated that 2.5 megacuries of  $\text{Sr}^{90}$  were introduced. Assuming that the rate of transfer of debris was about the same for the 1961 tests as in 1958, this would have produced fallout of 27 mc./mi.<sup>2</sup> from April 1 to October 1, 1962 and total deposition to July 1, 1962 of about 31 mc./mi.<sup>2</sup>. The recent report of Kuroda and Nix (4) suggests that this is a very good approximation. Resumption of tests by the United States will increase this even more.

Taking the Crosby silt loam soil as an example, and assuming fallout of 27 mc./mi.<sup>2</sup> and total deposition of 31 mc./mi.<sup>2</sup> we can estimate the number of micromicrocuries of  $\text{Sr}^{90}$  per gram of calcium in milk as follows:

$$\begin{aligned} Q_m &= AX + BY \\ &= 0.65 \times 27 + 0.12 \times 67 = 25 \text{ micromicrocuries} \\ &\qquad\qquad\qquad \text{of } \text{Sr}^{90} \text{ per gram of} \\ &\qquad\qquad\qquad \text{calcium in milk.} \end{aligned}$$

This estimated value is close to the average value of about 20 micromicrocuries of  $\text{Sr}^{90}$  per gram of calcium in milk for this region.

It would thus appear that the use of  $\text{Cs}^{137}$  as a monitor for  $\text{Sr}^{90}$  in soils would make possible rapid and extensive reconnaissance surveys of  $\text{Sr}^{90}$  concentrations in soils as well as in milk.

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