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Semiquantitative Measurement of Fission Produced Gamma Ray Radioactivity in Soils at Dubuque, Iowa

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Residual fission-product radionuclide contamination (fallout) in soils at Dubuque, Iowa was evaluated with an Exploranium geoMetrics gamma ray scintillometer in 1985 and after the Chernobyl accident in 1986. Anomalous amounts of gamma ray radiation were found near where downspouts discharged storm runoff from the roof of a dwelling. The 1985 residual cesium-137 activity in the soils of the area was found to be 0.2 pCi/gram with an areal contamination of 16 ± nCi/m². Activities associated with the soils near the discharge points of the downspouts ranged as high as 8 pCi/gram. This contamination seems to have occurred prior to 1970 and most probably during the period 1945 to 1963. The Chernobyl accident resulted in an additional soil activity of 0.05 pCi/gram or an areal contamination of 4 ± 0.1 nCi/m².

INDEX DESCRIPTORS: fission product contamination, fallout, cesium-137, Chernobyl

Atmospheric nuclear weapons tests conducted primarily by the United States and the Soviet Union nearly 30 years ago and the recent accident at Chernobyl resulted in the contamination of the entire globe with some 200 radionuclides of 34 elements in the mass number range of 72 to 158. Most of these radionuclides have half lives in the range of seconds to days and thus the associated radiation levels of the debris generated by the weapons tests should have approached normal background within a few weeks of the test (Holter and Glasscock, 1952). Natural background levels however, will not be reached for a good number of years due to the presence of strontium-90 and cesium-137 with half lives of 28 years and 30 years respectively. Both of these radionuclides are high yield products of the fission of uranium and plutonium and represent potential biohazards due to their affinity to biological tissues. Detection of these radionuclides in the field generally requires sensitive instrumentation and quantitative measurement requires time consuming analysis in the laboratory with expensive equipment. This presents a problem in terms of cost and time when assessing the regional contamination from a nuclear accident such as that at Chernobyl. The original purpose of this study was to explore the use of a simple and inexpensive portable hand held gamma ray scintillometer in measuring the level of radionuclide contamination from atmospheric nuclear bomb tests. The accident at Chernobyl has provided the opportunity to extend the study to nuclear reactor accidents.

Atmospheric nuclear testing was initiated in 1945 with the first atomic bomb detonation at Alamogordo, New Mexico. Atmospheric tests were conducted on a large scale by the United States and the Soviet Union until signing of the Nuclear Test Ban Treaty in 1963 (Figure 1). A low level of testing of atomic weapons in the atmosphere has been conducted since 1963 by France, The People's Republic of China, and India. In all, 300 to 350 atmospheric tests were conducted prior to the treaty and 75 since the treaty (Dennis, 1984).

The formation and meteorological distribution of the fission products of the weapons tests have been summarized by Bjornstedt and Edwarson (1965), Machta (1965), and Fowler (1960). Fission produced radionuclides are brought down with precipitation within months of their formation. Radionuclides, especially cesium-137 and strontium-90, derived from nuclear bomb tests, have been found in soils from around the world (Cox and Fankhauser, 1984, Table 2, p. 68). The various studies indicate that: 1) Areas with higher annual precipitation levels have in general higher levels of fission derived radionuclides (Mishra and Sadasivian, 1972; Kline, Colon, and Brar, 1973; Lowe, 1978; and Cox and Fankhauser, 1984); 2) The concentration of radionuclides or specific activity is greatest in the upper few centimeters of soil (Mishra and Sadasivian, 1972; Gustafson, Marinelli, and Brar, 1958; and Romney et al, 1983); and 3) The finer grained soils or those with greater amounts of clay minerals, especially the micas or mica clays, have higher concentrations of radionuclides (Lomenick and Gardiner, 1965; Kawase and Yokoyama, 1973; Francis and Brinkley, 1976; and Cox and Fankhauser 1984).

Fig. 1. Histogram showing the frequency of atmospheric nuclear weapons test by all nations for the period 1945 to 1985. Data extracted from Dennis (1984). Please note that even though the greatest number of tests occurred between 1955 and 1960 the greatest amount of fallout material was probably derived from the tests of 1960 to 1965.
Statistically a uniform rate of precipitation in an area should give the entire area an equal dose of the contaminating isotopes. However, precipitation which fell on the roof of a building would have been collected in the gutter system and transported to the ground via downspouts. The soil where downspouts discharge runoff water onto the ground should, therefore, have received a proportionately greater amount of the radionuclides thus producing a soil radiation anomaly. The overall magnitude of this soil anomaly is proportional to the areal fission product flux and the area of the roof drain by the downspout.

**EXPERIMENTAL METHOD**

The method employed here capitalizes on the concentrating effect of the roof runoff via downspouts. Systematic sets of radiation measurements were made at the discharge points of several downspouts and compared to a control set made nearby in an area that was not affected by downspout discharge or storm runoff. The difference between the data sets collected around the downspouts and the control set or background radiation is termed the excess radiation. The excess radiation was related to actual fission-product flux through the geometric considerations of the instrument and sampling net, gamma ray absorption of the soil, and detection efficiency of the instrument as employed in the field.

The site for the bulk of the study is a private, three story, wood frame residence constructed prior to 1945 which is located at Dubuque, Iowa (latitude 42° 30'N/longitude 90° 40'W). The residence was constructed upon and is surrounded by a fine-silty, mixed, mesic, typic Hapludalfs soil of the Fayette association. The characteristics of the soil are summarized in Table 2. The site receives soil pH ............................. 4.5 to 7.8* 

Organic content ... (by volume) ........... 5 to 10% (visual) 

Soil permeability .......................... 1.5 to 5.1 cm/hr* 

Soil density .......................... 1.5 gm/cm³ 

Coefficient of gamma ray absorption .... 0.045/cm

*Kuehl (1978)

Fig. 2. Plan view of the study area showing the house divided into five watersheds and the three areas adjacent to the downspouts. The downspout area for watershed “D” had been disturbed recently and was not utilized. Watershed “E” does not have a downspout.

The original set of measurements was made in July of 1985. A second set was made in July of 1986 during the evaluation of the Chernobyl accident. The second set was taken in the same way as the first, but since no markers were left after the first set, instrument sites only approximate those of the first site though the location of the grid was the same.

Four other nearby residences were checked for soil radiation anomalies with positive results. However, detailed evaluation of these anomalies was not carried out due to the inability to get permission to do such or the inability to establish a complete history of the sites in terms of downspout location and possible post 1945 soil modification from construction or gardening.

**SITE DESCRIPTIONS AND RESULTS**

**Control Area**

A control area was selected 12.8 m southeast of the dwelling on a grass covered lawn. The site is essentially flat and well away from any drainage. Two sets of data were collected in 1985; one set at 0.3 m intervals on a grid and the second, a time sequence at the same position at the center of the grid.

The set of radiation measurements taken along the grid exhibit a normal distribution with a range of 54 to 61 cps, a mean of 57 cps, a standard deviation of 1.6, and a coefficient of variation of 0.027. No pattern or trend is apparent in the spatial distribution of the data (Figure 3). Measurements made in time sequence also exhibit a normal distribution with a range of 54 to 61 cps and a mean of 57 cps. This suggests a random data collection error of approximately five percent. The spatial variance noted in Figure 3 may, therefore, be due entirely to random error. However, some of the variance could also be due to compositional differences in the soil such as buried roots, rocks, and animal burrows. The measurements from the control area suggest that normal local background radiation levels will produce instrument readings of 54 to 60 cps. The values of 60 to 61 cps may represent
above background radiation levels and will hence be considered marginally anomalous. Measurements over 61 cps indicate anomalous radiation levels. Measurements made in 1986 on the same grid gave comparable results to the 1985 measurements.

**Area “A”**

Area "A" is located at the southwest corner of the house and receives drainage from 49 m$^2$ of effective roof area. The downspout discharges onto a grass covered lawn. The surface is generally flat with a slight trough which slopes southward away from the downspout at a gradient of 2 cm/m. Discharge from the downspout during periods of high runoff moves along this trough before soaking into the ground. During periods of low discharge the water soaks immediately into the ground at the point of discharge. The current (1985-86) downspout is 0.9 m shorter than the one present during the period 1950 to 1970.

Radiation measurements are summarized in Table 2. The spatial distribution of the 1985 data (Figure 4) indicates a strong anomaly centered 0.3 m in a down gradient direction from the former terminus of the downspout. Anomalous and marginally anomalous readings occur in a down stream direction along the trough for 2.4 m before returning to background levels. The limits of these anomalous and marginally anomalous radiation levels correspond roughly to the limits of surface flow during peak discharge and Spring thaw. Radiation levels around the 1985-86 terminus of the downspout are all within the background range. Measurements made in 1986 indicated an increase in radiation near this terminus (59-62 cps).

**Area “B”**

Area "B" is situated at the east corner of the residence and receives the drainage from 55 m$^2$ of roof area. The area is shaded by two spruce trees and the ground surface is littered with spruce needles and covered by a sparse amount of grass and moss. The ground surface has no perceptible slope, but discharge from the downspout tends to flow on the surface in a southerly direction prior to soaking into the ground. The terminus of the downspout was relocated sometime after 1970 (Figure 5).

Measured radiation levels are summarized in Table II. Spatial distribution of the 1985 data (Figure 5) indicates an anomaly centered close to the old terminus of the downspout. Anomalous and marginally anomalous readings extend southward and roughly correspond to

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**Table 2. Summary of Radiation Measurements**

<table>
<thead>
<tr>
<th>Area</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>E</th>
<th>combined</th>
</tr>
</thead>
<tbody>
<tr>
<td>Roof drainage (m$^2$)</td>
<td>49</td>
<td>55</td>
<td>31</td>
<td>2</td>
<td>135*</td>
</tr>
<tr>
<td>Excess radiation (cps) (1985)</td>
<td>385</td>
<td>225</td>
<td>251</td>
<td>—</td>
<td>827</td>
</tr>
<tr>
<td>Excess radiation (cps) (1986)</td>
<td>463</td>
<td>311</td>
<td>251</td>
<td>19</td>
<td>1025*</td>
</tr>
<tr>
<td>Area contamination (cps/m$^2$) (1985)</td>
<td>7.9</td>
<td>4.1</td>
<td>7.0</td>
<td>—</td>
<td>6.1</td>
</tr>
<tr>
<td>Area contamination (cps/m$^2$) (1986)</td>
<td>9.5</td>
<td>5.7</td>
<td>8.1</td>
<td>9.5</td>
<td>7.6*</td>
</tr>
<tr>
<td>Chernobyl contamination (cps/m$^2$)</td>
<td>1.6</td>
<td>1.6</td>
<td>1.1</td>
<td>—</td>
<td>1.5</td>
</tr>
</tbody>
</table>

*excluding area E
Radioactivity in Soils

Fig. 5. Map of area "B" showing the magnitude and lateral extent of the 1985 radiation anomaly. Isopleth contour interval is 5 cps. Downspout drawn to show its current length and position; fine dotted line denotes its position prior to 1970. Dotted rectangle denotes the boundary of area "B" and is keyed to the rows and columns of the sampling grid.

the flow pattern from the old downspout terminus. The 1985-86 terminus of the downspout has radiation levels (1985) within the background range. The 1986 set indicated an increase in radiation at this terminus (58-61 cps). The two areas which correspond to the position of the two spruce trees exhibit radiation levels below background level, and probably reflect the dilution effect of the root system of the trees.

Area "C"

Area "C" lies on the northwest side of the residence and receives the runoff from 31 m² of effective roof area. The 1985-86 position of the downspout corresponds to its position during the period of nuclear tests (Figure 6). The ground surface is entirely grass covered, but being located on the northern side of the house, tends to remain frozen well into the period of Spring thaw. Downspout discharge during the Summer and Fall tends to soak in immediately, whereas during the Spring it spreads out. Furthermore, during the Winter and Spring the downspout will freeze solid thus causing overflow to accumulate behind the terminus of the pipe.

Measured radiation levels are summarized in Table II. There is an anomaly at the terminus of the downspout. This anomaly is weaker and more dispersed than the anomalies at areas "A" and "B" but does reflect the flow pattern of water discharge from the downspout.

Fig. 6. Map of area "C" showing the magnitude and lateral extent of the 1985 radiation anomaly. Isopleth contour interval is 5 cps. Downspout drawn to show its current length and position which has not changed since before 1945. Dotted rectangle denotes the boundary of area "C" and is keyed to the rows and columns of the sampling grid.

Discussion

At each station the measured level of gamma radiation can be divided into two components based upon origin: (a) that from the natural pedologic or geologic processes of the area and (b) that derived from the fission products of nuclear weapons tests or the Chernobyl accident. The pedologic/geologic component arises from the decay of potassium-40, uranium-238, and thorium-232. Potassium-40 is probably the larger in concentration of the three and is related to the potassium-bearing minerals of the soil and parent material of the soil; especially orthoclase feldspar and montmorillonite. A small portion of pedologic/geologic component can also be attributed to the cosmic background. The pre-Chernobyl fission product component should be almost entirely be due to the decay of cesium-137. The other fission product radionuclides either have half lives which are so short that their contribution to fission product component is now so small as to be considered negligible or their decay, as in the case of strontium-90, produces a radiation which is not detectable with the instrument used here. The measurements made in 1986 probably include a small component of radiation from other radionuclides (see Eisenbud, 1987, p. 375-389).

The soil within the control area, like the soil near the downspouts, should contain an intrinsic amount of fission product cesium-137 brought to the soil by rainfall. However, as a first approximation assume that the fission product component in the control area is much smaller than the pedologic/geologic component. Therefore, the total radiation in the control area is approximately equal to the pedologic/geologic component. If one assumes that the soil is homogeneous in composition throughout the immediate study area then the pedologic/geologic component for a sample near a downspout would be approximately equal to the measured gamma radiation level of the control site. Near the downspouts the measured gamma
radiation would be approximately equal to the measured gamma radiation at the control area plus that component due largely to cesium-137 activity. The cesium-137 activity near the downspouts would therefore be approximately equal to the difference between the measured gamma activities near the downspouts and the measured gamma activity of the control area.

The mapped anomalies (Figures 4, 5, and 6) represent the observed extent and magnitude of fission product component as measured in 1985. The 1985 and 1986 "excess radiation" reported in Table II for each site is the sum of the differences between the measured 1985 or 1986 gamma activities and the mean activity of the control area. The excess attributed to the Chernobyl accident is the difference between the excess radiation for 1985 and 1986.

Ideally the amount of excess radiation for each area should be proportional to the roof area being drained by the corresponding downspout given that no other processes interfere between the time of precipitation and the time of radiation measurement. Such processes include chemical leaching, both man induced or natural, and disruption of the soil by construction, gardening, or animal burrowing. The data scatter in the cross plot of excess radiation versus drainage area (Figure 7) suggests some disruption of the pre-1985 radionuclide accumulations. All data should fall on a straight line which passes through the origin where zero square meters of drainage area gives rise to 1.5 cps measured excess radiation.

The instrument used in this study was not designed for quantitative work, however it should be possible to approximate the amount of cesium-137 activity in the soil. This can be done by evaluating the geometry or spatial relationship between the detector and the soil radionuclides and the absorption of gamma rays by the soil. Consider each radiation measurement along the grids to represent the measurement of the radiation emanating from a block of homogeneous soil 30 cm by 5 cm. Assume the soil within each block to be homogeneous and assume that the fission-produced radionuclides are evenly distributed within the block. The orientation of the scintillator crystal relative to the soil block, the distance between the detector and the various parts of the soil block, the coefficient of gamma ray absorption of the soil, and the instrument detection efficiency result in the detection of approximately one percent of the emitted gamma rays.

The level of contamination from the Chernobyl accident exhibits a much closer linear relationship with the origin (Figure 7). This suggests that the apparent leaching effect noted at area "B" must operate over a longer period of time than a few months. The slope of that linear relationship suggests that one square meter of drainage area gives rise to 1.5 cps measured excess radiation.

The difference in vegetative cover at "B" as compared to "A" and "C" may account for "B" not falling on trend. Area "B" had only a sparse grass and moss cover and was littered by spruce needles. This may have given the soil a slightly lower pH which, in turn, could have affected the ability of the clays and humus of the soil to absorb and retain certain cations. Thornthwaite, Mather, and Nakamura (1960) observed that the rate of downward movement of strontium in soil was proportional to the cation exchange capacity of the soil and inversely proportional to the soil pH. Levinson (1974) reported that the cation exchange capacity increases with increased pH and that montmorillonite absorbs and retains cations at pH of six or more. Fletcher (1981) indicated that the absorption on surfaces of clays, iron and manganese sesquioxides, and organic compounds is strongly dependent upon pH, with increased absorption of cations favored by high soil pH. If two soils, which are identical in mineralogical and mineraloid makeup, differ slightly in pH then they should also differ slightly in their ability to absorb and retain cations that they come in contact with. The radionuclides at area "B" may simply have in part passed on downward in the soil while those at "A" and "C" had a greater probability of retention close to the surface where their radiation would be more accessible to detection.

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products. During the same period of time they reported 5.3 mCi/mi², 6.2 mCi/mi², and 4.9 mCi/mi² of strontium-90 for the same cities. Their report unfortunately does not give specific data on cesium-137. Since Dubuque, Iowa lies roughly in the center of the triangle formed by these three cities, the total fission product activity and strontium-90 activity may be taken as the average of the three or a total activity of 150 mCi/mi² and strontium-90 activity of 5.5 mCi/mi². Mishra and Sadasivan (1972) report that cesium-137 and strontium-90 occur at an approximate ratio of $\frac{Cs}{Sr} = 137/89 = 0.15$ for pre-1962 fallout. This ratio combined with the data extracted from the table data yielded the value of 41±8 mCi/mi² (17.5 mCi/mi² at 1985 levels) observed in 1957 at the Argonne National Laboratory in Illinois (Gustafson, Marinelli and Brar 1958). The calculated soil concentration of cesium-137 in the soils around the study area, 0.2 pCi/gram, corresponds to world wide data reported in Cox and Fankhauser (1984, Table 2) and with their cross-plot of cesium-137 activity versus mean annual rainfall (where 60 cm per year of precipitation corresponded to a cesium-137 activity of 0.10 pCi/gram). The relationship between rainfall and cesium-137 activity depicted by Mishra and Sadasivan (1972, Figure 2) correlates 60 cm of rainfall with 25 nCi/m² which is also in reasonable agreement with that measured in the study area. As a final comparison, the estimated fission yield from all atmospheric tests is about 200 megatons of TNT equivalent of which 90% occurred before 1963 (Eisenbud, 1987, Figure 13-3, p. 316). The approximate cesium-137 yield from one megaton of fission is 0.16 megacuries (Eisenbud, 1987, Table 12-2, p. 280). The cesium-137 yield from all the atmospheric tests combined would be 32 megacuries. Given the area of the Earth as 5.1X10¹² m²; the world wide average contamination would have been 63 nCi/m² or roughly 31 nCi/m² in 1985.

The Chernobyl accident released approximately 2 megacuries (MCi) of cesium-137 and 21 MCi of iodine-131 to the atmosphere (see Marshall, 1986 and Levi, 1986). This implies a world average contamination of 4 nCi/m² of cesium-137. This compares very favorably with the 4 ± 0.1 nCi/m² reported here through this value may seem quite high given the distance from the accident site. The apparent high fallout flux was probably related to the local weather. From May 13 to May 17, the period of time corresponding to the passing of the radiation cloud over the Midwest, the site of investigation experienced a series of severe thunderstorms and received 10.1 centimeters of rain with one storm on the thirteenth producing 1.3 cm in 15 minutes. Sometime after 1970 the downspouts at "A" and "B" were relocated. Atmospheric testing of nuclear weapons has continued since 1970 by France, the People's Republic of China, and India. The lack of above background levels (1985) of radiation at the current ends of these downspouts indicates that these more recent tests have not added significant amounts of fission products to the soils of the area. Slightly above background measurements were observed at the current ends of the downspouts during the 1986 survey.

The method utilized in this study should be applicable in other similar situations. The other surveys mentioned above utilized a gamma ray spectrometer to analyze specific soil samples for radionuclides. This instrument allows for direct determination of the specific activity of the nuclides of interest, but has the disadvantage of requiring a large sample or multiple samples for statistical reliability, and generally necessitates time consuming analysis in the laboratory plus the high expense of the equipment. The advantage of the method utilized here is that a good semiquantitative measure of the activity can be had in the field with a minimal time commitment, one hour per site, and with relatively inexpensive and simple to operate equipment. The main requirements besides the scintillometer are that sites are documented as to downspout discharge locations and soil disruption history. Downspout discharge should be onto flat ground with a clay rich soil and situated well away from brick walls. Control areas must have a soil that is similar to that at the discharge sites and also must be situated away from brick walls and not affected by runoff from any source. For the investigation of reactor accidents such as that at Chernobyl one either needs to know the soil activity character prior to the event as was known in this study or measurement must be limited to discharge sites located after 1970 and not effected by pre-1970 storm drainage.

CONCLUSIONS

1) The method employed here does allow for a semiquantitative measurement of fission-product soil contamination. The accuracy of such measurements is at least within an order of magnitude and probably closer though additional evaluation is required to verify this.

2) Fission product radionuclides were found concentrated near where downspouts discharge roof runoff water onto the soil. The magnitude of such anomalous concentrations is proportional to the area being drained by the downspout. The maximum observed concentration near a downspout is 8 pCi/gram of soil.

3) Fission product radionuclides, generated during atmospheric testing of nuclear weapons in the 1950s and early 1960s, were brought down by precipitation in the Dubuque, Iowa area. The current residual fission product gamma ray activity in the soil was inferred to be 0.2 pCi/gram. This corresponds to an areal contamination in 1985 of 16 ± 3 nCi/m². The majority of the contamination is attributed to the radionuclide cesium-137.

4) No significant amount of fission products has impinged upon the area since the 1970s and prior to July of 1985, even though France, the People's Republic of China, and India have continued the atmospheric testing of nuclear weapons.

5) Fission product radionuclides from the Chernobyl accident were brought down by precipitation at Dubuque, Iowa resulting in an increase in the soil gamma ray activity of 0.05 pCi/gram or an areal flux of 4 ± 0.1 nCi/m².

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