COLD WAR LEGACY: SURFACE INVESTIGATION OF UNSATURATED PRAIRIE SOIL RADIOLOGICALLY CONTAMINATED IN 1951

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ABSTRACT

In order to investigate the effect of weathering on the ground deposition of fallout from nuclear weapons releases, an aqueous, acidic solution of fission products and actinides was spread on bare ground and concrete surfaces at a remote location in southern Alberta. In July 1951, 6.7 L of this solution, containing 360 GBq of radioactive material, leaked from a storage container buried nearby. Since then, the radioactive material, created by the irradiation of 40 g of natural uranium metal in the NRX reactor, has migrated through the soil. This article describes the surface survey measurements for both residual contamination and to locate the buried storage tank.

INTRODUCTION

The unsaturated zone (UZ) extends from the land surface to the underlying water table (saturated zone) within subsurface porous media. The UZ is the interface between surface events involving hydrology and contamination, and the subsurface. This paper presents the surface measurements carried out to monitor surface contamination and locate the source of the subsurface radiological contamination in unsaturated Alberta soil.

SITE CONTAMINATION

In 1951, 6.7 L of a mixed fission product (FP) and actinide solution, buried in a storage tank located at a remote location at Canadian Forces Base Suffield, Alberta, leaked into the ground at its buried depth of 3.7 m. It is estimated that 360 GBq (9.7 Ci) of radioactivity contained in the acidic, aqueous solution, produced from the irradiation of natural uranium metal, was released into the UZ in the arid prairie soil. The resulting natural transport process of FPs and actinides has been permitted to occur undisturbed over the past 50 years, as the contaminated land in question has been restricted by the land owner, the Department of National Defence (DND), to research use only. No military training or exercise activities have been permitted there. Further, in exercising due diligence, DND has undertaken to define the status, condition and location of the FPs. Therefore, in 1999 a project was initiated to investigate the leakage of radioactivity from the buried storage tank. The challenge then is to determine the current location of the contaminants. Ultimately, decisions must be made about the long-term disposition of this contaminated site in general, and the contaminated soil in particular.

Transport of solute material in subsurface porous media occurs by the processes of advection, diffusion or a combination of both. In the case of UZ solute transport in an arid environment, the dominant transport process is diffusion, due to the low rates of soil water recharge from natural precipitation sources such as rain, dew, snow, runoff, etc. Examination of this site should eventually permit determination of values of diffusion and sorption coefficients for the most numerous and long-lived FP and actinide radionuclides present in the contamination, i.e., ^{137}Cs, ^{90}Sr, ^{144}Ce, ^{113m}Cd, ^{151}Sm, ^{129}I, ^{238}U and ^{239}Pu. This, in turn, will enable prediction of the time required for the soil concentration of specific radioisotopes to reach regulated limits at the depth of the water table at this location (45 m), to assist in determining the ultimate disposition of the site.

HISTORICAL BACKGROUND

In the late 1940's and into the 1950's there was much interest in western countries in the effects, both short-term and long-term, of contamination from radioactive fallout particles on structures and surfaces. In 1951, one such study was initiated at the DND experimental station called at that time the Suffield Experimental Station (SES) (now named Defence Research and Development Canada (DRDC) - Suffield) located northwest of Medicine Hat, Alberta. The purpose of the particular experiment germane to this study was to record the natural weathering effects on mixed FP contamination on concrete and earth surfaces. A welded stainless steel container was constructed for the radioactive solution and was buried at a depth of 3.7 m at an isolated location on the prairie. The container was designed so that the liquid FP solution could be extracted when needed and dispersed onto a number of different concrete and earth surfaces.

The mixed FP solution was produced by irradiating 40 g of natural uranium metal in the Atomic Energy of Canada Limited (AECL) Chalk River pile (NRX reactor) for 10 days in late June 1951. After irradiation, the activity was 740 GBq (20 Ci). The metal was then flown by military aircraft from Ottawa to Alberta. At Suffield, it was dissolved in a nitric acid / hydrochloric acid solution, poured into the storage container and diluted with water to a final volume of 13.1 L. On 23 July 1951, the buried storage container was found to be empty, when there should still have been liquid FP solution remaining. Of the 13.1 L of solution originally available, 6.4 L was utilized in scientific research and the remaining 6.7 L is assumed to have leaked into the soil.

WEATHERING EXPERIMENTS

As a result of the high level of radioactivity of the solution from the storage tank, samples were drawn off into glass beakers using remote handling tools. These beakers were then placed on the earth and concrete pads. Dispersal onto the surface was achieved by firing small calibre rifle rounds at the beakers. The contaminated surfaces were then monitored at varying intervals to determine the influence of weather on the residual radioactivity. It was noted that the gradual loss of radionuclides due to wind and rain was somewhat greater from the earth than from the concrete pad. For example, after 130 days, it accounted for 63 % from the earth compared to 48 % from the concrete. The dominant removal mechanism remained radioactive decay throughout the 4.5 month study.[1]

SURFACE SURVEYS

In the period between 1951 and 2000, three documented radiological surveys were conducted to determine the residual biological hazard and to observe which radionuclides were present and the radiation rates at the surface level.

1989 Defence Research Establishment Ottawa Survey.

The first documented survey of the gamma radiation levels at the surface of the site since the 1951 releases was conducted by personnel from Defence Research Establishment Ottawa (DREO).[2] The purpose of the survey was to determine the extent of existing radioactive material and any hazards resulting therefrom. The measured data included over 700 radiation readings for surface-based gamma and gamma plus beta, measured at heights of both 10 cm and 1 m above the surface. Also, 76 surface samples of mainly soil but also vegetation, concrete and wood chips, were taken and analyzed for radionuclide content. An additional four soil samples from depths up to 10 cm below the surface were obtained. The soil and vegetation samples were analyzed using a gamma-ray spectroscopy system consisting of a 5 cm x 5 cm sodium iodide (thallium) (NaI (Th)) scintillator and a high purity germanium detector.

The highest observed radiation reading was found at the center of one of the small 1 m x 1 m concrete test pads (Pad "A") where the reading on the surface was 25 μSv/h gamma and 140 μSv/h gamma and beta. This gamma dose rate reading was some 208 times the background gamma reading of 0.12 μSv/h for Suffield. However, it should be noted that the maximum reading was observed at the center of the concrete pad and that the radiation readings were indistinguishable from background levels at a distance of 1 m from the edge of the concrete pad. Overall, the site consists of a large 9 m $\times$ 9 m concrete pad, two small 1 m x 1 m concrete pads, a large 9 m $\times$ 9 m earth pad and a small 1 m x 1 m earth pad. The total area of the site is approximately 250 m x 400 m.

The surface radiation readings can be seen in Table 1 while Table 2 shows the ^{137}Cs and ^{90}Sr activities for soil samples taken for depths up to 10 cm. The soil radiation readings reveal a decrease in the activity and thus quantity of radionuclide, the deeper into the ground the sampling is made. It is also, however, evident from the specific activity values that the ^{90}Sr has been transported further into the ground.

The salient observations resulting from this survey were:

- ^{137}Cs and ^{90}Sr were found in the soil samples as expected, as these are the major long-lived fission products;
- ^{90}Sr exhibited greater mobility in terms of vertical migration than ^{137}Cs. This is expected due to its higher solubility than ^{137}Cs;

- The mean measured rates of vertical/downward migration, over the 38 year period, 1951 to 1989, were:

  ^{137}Cs 0.5 mm / year
  ^{90}Sr 1.8 mm / year.

The major recommendation of this survey was that the lateral spread of radionuclides be immobilized through the addition of uncontaminated soil to the pads where FPs were spread. It was recommended that if 25 cm of earth were added to the concrete and earth test pads, then the gamma radiation would be reduced by a factor of 20; sufficient to reduce the above-ground radiation dose rate over the most active features to prevailing background rates. This recommendation was carried out in 1990, when concrete caps were added to the concrete pads, earth pads and the site of the buried storage tank.

1999 Defence Research Establishment Ottawa Survey.

This survey was a revisit by DREO personnel 10 years after the initial survey and was conducted to observe the change in the radiation levels at the site.[3] The measurements were taken using portable NaI gamma-ray and beta spectroscopy units. It was found that there were still measurable quantities of ^{137}Cs on the surface near the pads. This was expected, as ^{137}Cs does not transport quickly into and through the soil. No significant beta readings were observed at the surface, indicating that the ^{90}Sr had migrated below the surface or had been removed from the surface through wind and weathering action.

The major recommendation of this report was that work should be undertaken to locate the remnants of the storage tank and to examine and map the distribution of radionuclides around its location.

2000 Atomic Energy of Canada Limited Survey.

This survey was conducted by the AECL Low-Level Radioactive Waste Management Office (LLRWMO) in order to determine the gross gamma radiation levels at the surface within and around the site.[4] The radiation survey was conducted using a large area gamma survey (LAGS) system, hand-held Geiger-Mueller survey meters and hand-held scintillators. The observed radiation readings from this survey are shown in Table 3. As can be seen, the average radiation readings were only slightly above the background levels for the area. This reduction in the radioactivity from that measured in 1989 was due to a combination of the effects of the shielding provided by the added concrete cap, and also to decay, and further transport into the soil.

SAMPLE COLLECTION

In October 2001, a team from the Royal Military College of Canada (RMC) sought to confirm the location of the underground storage tank (using ground penetrating radar (GPR)) and to drill boreholes and recover soil samples from the vicinity of the buried container.[5] A total of 18 boreholes were drilled using the sonic drilling technique, with 16 of the holes located in the immediate vicinity of the storage container, while 2 offsite boreholes were drilled approximately 15 m from the container location.

A total of 311 soil samples (ranging in mass from 50 to 120 g) were collected from the boreholes and were transported to RMC for radiochemical analysis. In addition, the complete soil contents of one offsite borehole were returned to RMC for use in studying solute transport by attempting to replicate what occurred in the field.

GROUND PENETRATING RADAR

As noted above, GPR was used to map the location of the underground storage tank. The GPR device is a subsurface imaging instrument that emits a weak RF signal into the ground and measures the time delay and strength of the returning echo. From this information software is used to convert the signal to a display showing a map of subsurface solid objects. GPR is effective in depths of up to 10 m of clay, the type of soil found at the site. The model of GPR used was the Noggin 1000 Smart Cart system from Sensors and Software Inc. of Mississauga, ON, and was built to transport the Noggin GPR on a 4-wheeled cart, along with the onboard digital video logger (DVL). The DVL contains software to operate the system and to record the data collected. Upon completion of GPR mapping, the collected image files were downloaded from the DVL to a computer for interpretation. The Noggin 1000 subsurface imaging instrument operates at a frequency of 1000 MHz and is capable of acquiring up to 100,000 samples per second. Figure 1 is a photograph of the Noggin Smart Cart GPR system used in this work.

Prior to conducting the GPR mapping, a 5 m x 5 m grid was established over the surface suspected of covering the storage tank. The centre of the grid was designated as the point where two steel pipes protruded from the ground, as can be seen in Figure 2. The outer boundary of the grid was oriented roughly in the Northeast - Southwest direction, with the Grid North – South line oriented 39 degrees East (Clockwise) from the True North.

Analysis of the GPR data indicated what appeared to be the signature of a solid object, located approximately 0.8 m below the surface and roughly 3 m x 3 m in size. Subsequent investigation revealed that the shallow object was a concrete pad. The top of the pad was uncovered and it was found to be 3.19 m in length in the North - South direction by 2.43 m in length in the West - East direction with a thickness of 10 cm. There was no historical documented evidence of this concrete pad in archive reports of experimental work carried out at the site.

Further, a thin and long solid object was detected near the centre of the grid at the depth corresponding to that indicated in previous reports for the buried tank. This image was presumed to be a reflection of either the piping or the remnants of the storage tank.

The GPR results did reveal the presence of the concrete pad. However, below the concrete pad it was difficult to determine with confidence the location of pipes and the storage tank. Subsequent action involved drilling boreholes through the concrete pad to take samples which could be measured to determine whether and where radioactive material was actually located in the subsurface soil.

CONCLUSIONS

A study has been initiated to determine the location and concentrations of FPs and actinides that were accidentally released into the UZ of Alberta soil in 1951 and have been allowed to be transported through the soil by natural means over the past 50 years. Gradually the surface contamination, introduced through weathering experiments, has diminished with time, and a biological shield of concrete has been added to cover high areas of radioactivity. Gamma spectroscopy has revealed the presence both on the surface and slightly subsurface of ^{137}Cs. Beta/gamma probes have also revealed the presence of a beta emitter, presumably ^{90}Sr.

FUTURE WORK

The next step in this project involves conducting soil characterization and analyzing the soil samples for the presence and concentrations of the principal radioisotopes of interest. These values, in turn, will be used to determine the respective diffusion coefficients. Subsequent transport modelling can then be used to generate concentration maps as functions of time, in order to make informed decisions about the ultimate disposition of the contaminated site.

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Table 1 - Radiation Readings at Contaminated Site

Location	10 cm above surface		100 cm above surface		^{137}Cs in soil (Bq/g)
	γ (μSv/h)	γ and β (μSv/h)	γ (μSv/h)	γ and β (μSv/h)	
Small Concrete Pad "A"	0.60 - 25	1.65 -140	0.62 -0.70	0.70 - 3.8	0.3 - 18.8
Small Concrete Pad "B"	1 - 6.1	1.4 - 58	0.3 - 0.7	1 - 2.1	0.2 - 0.8
Large Concrete Pad	0.25 - 2	0.3 - 3.1			0.3 - 9.5
Background at Pad "A"	0.20	0.22	0.18	0.20	0.09 - 0.2
Background at Pad "B"	0.20	0.20	0.18	0.18	
Soil 22 km from Site	0.12				0.04 (total activity)

Table 2 - Soil versus Depth Measurements for the Large Earth Pad

Depth Range (cm)	^{137}Cs Activity (Bq/g)	^{90}Sr Activity (Bq/g)
0 to 2.5	8.3	2.1
2.5 to 5.0	2.7	1.2
5.0 to 7.5	0.5	0.9
7.5 to 10.0	0.2	0.7

Table 3 – Surface Radiation Reading Taken by AECL in 2000

Location	Average (μSv/h)	Maximum Reading (μSv/h)
Background	0.04	
Large Concrete Pad	0.086	0.252
Small Concrete Pads (2 locations)	0.073	0.091
Large Earth Pad	0.074	0.144
Small Earth Pad	0.076	0.172
Waste Trench	0.069	0.087

Figure 1. Noggin 1000 Smart Cart Used for the GPR Readings.
Pipes are assumed to be from the buried storage tank.

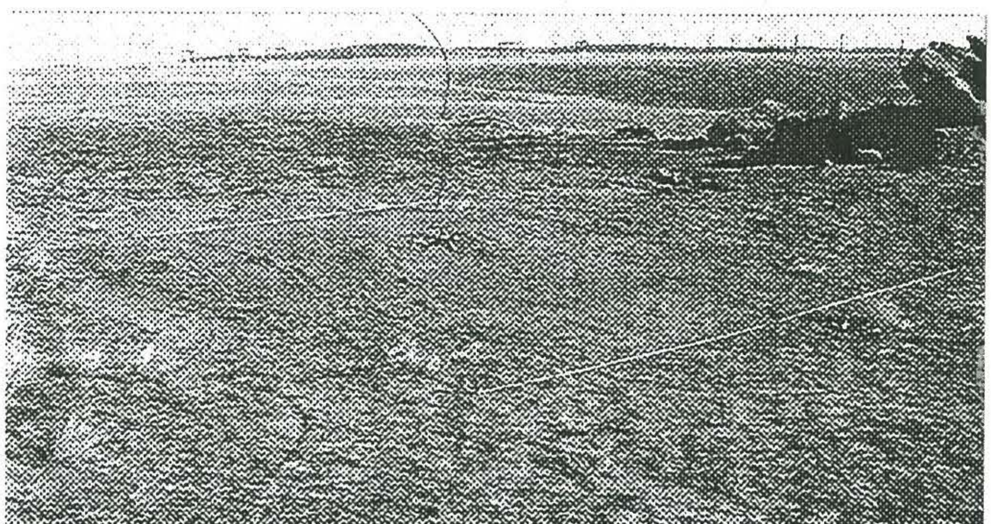

Figure 2. Initial grid over suspected tank location.
The concrete cap has been removed and rests at the right of the photo.