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

D.J. SIMS, W.S. ANDREWS*, Z. WANG and K.A.M. CREBER

Department of Chemistry and Chemical Engineering, Royal Military College of Canada, PO Box 17000 Station Forces, Kingston, ON K7K 7B4, Canada

* Corresponding author (tel: (613) 541-6000 ext 6052, fax: (613) 542-9489, e-mail: andrews-w@rmc.ca)

ABSTRACT

An unintentional release of fission products (FPs) from a buried storage tank in 1951 resulted in 6.7 L of liquid, bearing radioactive material, being spilled into unsaturated prairie soil at a depth of 3.7 m. Since then, the site has been undisturbed. In October 2001, boreholes were drilled and soil samples were recovered for analysis. Gamma well logging showed higher than background radiation readings at a depth of 3.5 m (corresponding to the storage container location) and a peak reading at 4.7 m (attributed to the breakthrough curve). The soil was determined to be predominantly lean clay with a silty sand layer between 4.4 and 5.1 m. Future work includes radiochemical analysis, soil column simulation, determination of distribution coefficients and transport modelling.

INTRODUCTION

In July 1951, 6.7 L of an acidic, aqueous solution of dissolved uranium and fission products, with a radioactivity of 360 GBq (9.7 Ci) leaked from a buried storage container at a military research facility at Suffield, Alberta. The leaked solution has remained undisturbed in the unsaturated soil surrounding the container for more than 50 years. In 2001, a team of researchers from the Royal Military College of Canada (RMC) in Kingston, Ontario went to the site to locate the buried container using ground penetrating radar (GPR) and then to collect soil samples by drilling boreholes.[1] These samples would then be taken to RMC for radiochemical analysis, with the results being used to both determine the extent of radiological contamination and eventually to characterize the migration of the various radionuclides through the soil.

SOIL SAMPLE COLLECTION AND BOREHOLE GAMMA SURVEY

The borehole drilling and soil sample collection were carried out in October 2001. In all, 18 holes were drilled using a sonic drilling technique, each to a depth of about 6.1 m. The sonic drill used high frequency mechanical oscillations, developed in the drill head, to transmit resonant vibrations and rotary power through the drill tooling to the drill bit. The drill stem and sampler barrel vibrated vertically at frequencies between about 50 and 180 Hz. This sonic vibratory action fluidized the soil particles, allowing clean, rapid and smooth penetration of overburden formations. Some of the characteristics of sonic drilling were its ability to perform core sample drilling with speed, precision, and an absolute minimal amount of disturbance and compaction, attributes not found in more traditional equipment using split spoons or augers. Sonic drilling can produce continuous core samples from underground formations with significant detail and accuracy.

Gamma well logging was conducted in the boreholes before they were refilled with non-contaminated material. The well logging results revealed the spread of radiological contamination and that some subsurface soil (at a depth of 3 to 4 m) was still radiologically contaminated. Due to the relatively long half lives of ^{90}Sr (28.8 a) and ^{137}Cs (30.5 a), coupled with a decay time of 50 years, these two radioisotopes, along with natural uranium are presumed to be the major contaminants. Consistent with previously conducted surface surveys,[1] no evidence of surface contamination in the vicinity of the buried tank was found. However, the metal pipes coming out of the ground above the tank were still slightly radioactive.

A crude concrete cap had been placed over the site of the underground storage container more than ten years previously. When it was removed prior to drilling, the metal pipes referred to above were exposed. Further, just prior to drilling, a concrete pad some 20 cm below the surface (and previously covered by the concrete cap) was discovered. When drilling was conducted, the pad was found to be approximately 10 cm thick. It is noteworthy that there was no written record found of the existence of this pad. When drilling was completed, the boreholes were refilled and the pad was recovered with soil.

Figure 1 shows the template used for the 16 boreholes drilled through the concrete pad. Also, the location of the two off-pad background boreholes are shown, located over 15 m from the concrete pad. It can be seen that 9 of the boreholes were drilled within 45 cm of the centre of the concrete pad while 7 boreholes were drilled between 45 cm and the outer radius of 75 cm.

The gamma survey was conducted by lowering an Eberline SPA-6 scintillator probe gradually down each hole. The results of the survey are given in Table 1. The highest gamma readings were recorded in boreholes 8, 20, 21 and 22. The highest recorded gamma reading was 203 μSv/h, (over 2,000 times background) in borehole 20 at a depth of 4.7 m. It is felt that the leakage from the container probably occurred from its south-east corner, due to the high underground radiation readings in this area. A plot of the gamma radiation readings for the bore holes showing the highest gamma dose rate readings can be seen in Figure 2. Two maxima can be seen in the curve, the one at 3.5 m depth corresponds to the documented buried depth of the storage container. The higher maximum is at a depth of 4.7 m below the surface level, probably due to the front of the breakthrough curve of gamma-emitting fission products being transported from the storage tank.

Metallic fragments were found in four sample holes, 1, 3, 18 and 24 and were probably from the storage tank and piping system. These particular four boreholes are on/within the 45 cm radius circle from the grid centre and, based on the location of the metal fragments, the container was probably oriented west-east.

For each sonic drilling sample tube removed from a borehole, soil samples were taken for radiochemical analysis at intervals along the sample tube varying between 10 cm to 100 cm, depending on the radiation reading of the sample. Where the radiation readings were significantly higher than background, the sampling interval was reduced. Both gamma and beta radiation surveys were conducted on each sample tube prior to soil sampling, using hand-held monitors. Samples varying in mass between 50 to 120 g were taken from each sonic drilling borehole sample tube.

SOIL CHARACTERIZATION

The soil characterization study found that the soil at this site has a large quantity of fine-grained clay. It was characterized using the USCS system as being lean clay, while between depths of 4.4 to 5.1 m the soil was found to have a high content of sand and was classified as silty sands and silt.

FUTURE WORK

The next, or laboratory, phase consists of:

a. the radiochemical analysis of samples using gamma-ray spectroscopy, neutron activation analysis (NAA), liquid scintillation counting (LSC) and mass spectroscopy;
b. a physical simulation using a soil column;
c. the determination of distribution coefficients; and
d. soil transport computer modelling.

Radiochemical Analysis

Radiochemical analysis is being used to determine the activity of FPs within the unsaturated soil surrounding the storage container. A high performance germanium (HPGe) gamma-ray spectroscopy system has been used to determine the specific radioactivity for the long-lived gamma emitting radionuclides within the samples. NAA has been conducted using the RMC SLOWPOKE nuclear reactor for neutron irradiation, followed by gamma spectroscopy using the HPGe. The LSC is being used for detecting beta-emitting radionuclides, such as ^{90}Sr. Mass spectroscopy using a high resolution quadropole inductively coupled plasma mass spectrometer will complement the other techniques.

Soil Column Simulation

Two laboratory replica soil columns have been designed, built, erected and filled with prairie soil for the depth below the storage container (i.e., depth > 3.7 m). The columns are 1.22 m high and have an inside diameter of 10 cm. Into one column a mixture containing non-radioactive or naturally occurring elements (^{133}Cs, ^{86}Sr, ^{140}Ce, ^{152}Sm, ^{110}Cd, ^{127}I and ^{238}U) has been added. The solution was introduced aat the top of the column and soil samples will periodically be taken and analyzed using techniques mentioned above. This column will allow monitoring the rate of migration of inert nuclides of the same elements present in the fission product mix. A second column will be used to replicate the leak at the contaminated site by using a reduced quantity of irradiated natural uranium, complete with the associated fission products. The quantity, however, will be more modest, being only 2 g of irradiated natural uranium metal. The metal, to be acquired from Cameco, will be irradiated in the RMC SLOWPOKE - 2 reactor facility. In a similar manner as the first column, samples will be taken periodically and analyzed. Comparisons will be made between the results of the two columns and the field results.

Separate Effects Experiments

In addition to the above, a series of separate effects experiments is being conducted using uncontaminated soil collected near the contaminated site. These experiments include examining the diffusion of solute contaminants by varying each of the following:

- concentration of solute contaminant
- soil temperature
- deep percolation ground recharge rate
- rate of solute contaminant addition
- soil porosity

The results of the separate effects experiments will provide information and measured parameters regarding the transport of key radionuclides in UZ prairie soil.

Determination of Distribution Coefficients

The transport of solute in soil is dependent on many factors, with one of these being the retardation of the solute by the soil due to adsorption. The key parameter for measuring the degree of adsorption of a solute by the surfaces of the soil is the distribution coefficient (K_d, also known as the sorption or partitioning coefficient). It will be necessary to conduct batch lab experiments in accordance with ASTM D 4319 - 93 to determine the value of K_d for the elements being studied. The concentration of the element in the liquid phase for these tests will be determined through NAA. The K_d values will be used in the subsequent computer transport modelling.

Soil Transport Modelling

The results of the radiochemical analysis will be used to derive diffusion coefficients that then can be used in commercial numerical software programs for modelling and predicting transport in the soil. Of the codes available, two have been chosen:

a. VS2DTI (Variably-Saturated 2-Dimensional flow and solute Transport Interactive), developed by the US Geological Services (USGS), is a finite difference model;[2] and
b. MOTIF (Model Of Transport In Fractured/porous media), developed by Atomic Energy of Canada Limited (AECL),[3] is a finite element model.

CONCLUSIONS

An examination has been undertaken of a site in Alberta radiologically contaminated by the accidental release from a buried storage tank of 6.7 L of an acidic, aqueous solution containing 360 GBq (9.7 Ci) of radioactive fission products and actinides. The initial aspects have involved a gamma survey of the boreholes and determination of the physical and chemical characteristics of the soil. These measurements will subsequently be used to estimate the extent of the radiological contamination of the site. They will also assist in investigating the transport aspects of the various radionuclides by determining the appropriate individual diffusion and distribution coefficients in the unsaturated prairie soil matrix. To date, borehole drilling, soil sampling, gamma well logging and soil characterization have been conducted. Sixteen boreholes were drilled through the concrete pad overlaying the area where the stainless steel container was buried, and an additional two boreholes were drilled at least 15 m away from the pad for background uncontaminated soil samples.

The gamma well logging study revealed that gamma-emitting radionuclides of significant quantity exist at approximately 4.7 m below the surface. These readings are probably due to the front of the breakthrough curve of contamination being transported from the storage tank. There is also a smaller maximum quantity of radioactivity at a depth of 3.5 m, corresponding to the reported depth for the storage container.

Further studies will include radiochemical analysis, soil column simulation, distribution coefficient determination and soil transport modelling.

Ultimately, this work is to provide sufficient detailed knowledge of the state and location of the contaminants present in the soil to assist in arriving at a decision regarding appropriate remedial or containment action. Secondary goals will be the determination of a set of diffusion and distribution coefficients for the some radionuclides in unsaturated prairie soil.

ACKNOWLEDGEMENT

The authors wish to express their appreciation and gratitude to the personnel at Defence Research Development Canada-Suffield and at Canadian Forces Base Suffield, as well as the financial support and encouragement from Dr Stefan Kupca of the Director General Nuclear Safety at National Defence Headquarters in Ottawa.

REFERENCES

1. Sims, D.J., Andrews, W.S., Creber, K.A.M., Hounsell, G., and Chenaf, D.,"Cold War Legacy: Surface Investigation of Unsaturated Prairie Soil Radiologically Contaminated in 1951", Proceedings of 8th International Conference on CANDU Fuel, Honey Harbour ON, 22-24 September 2003.
2. Healy, R.W., " Simulation of Solute Transport in Variably Saturated Porous Media with Supplemental Information on Modifications to the U.S. Geological Survey's Computer Program VS2D", U.S. Geological Survey Water-Resources Investigation Report 83-4025, Denver, USA, 1990.
3. Chan, T., Scheier, N.W. and Stanchell, F.W., "MOTIF Version 3.2, Verification and Validation Report", AECL Report Number 06819-REP-01200-10026-R00, March 2000.

Table 1. Borehole gamma radiation readings (Boreholes 0 to 24)

Depth (m)	Borehole 0 (μSv/h)	1	2	3	4	5	6	7
0		0.125	0.125	0.109	0.123	0.08	0.125	0.098
1	0.128	0.139	0.143	0.143	0.166	0.116	0.139	0.138
2	0.132	0.163	0.16	0.266	0.323	0.136	0.132	0.123
3	0.135	1.03	1.15	0.317	6.29	0.583	0.125	0.316
3.1		1.9	1.7		9.16	0.621	0.142	0.344
3.2		4.11	3	0.64	15.9	0.728	**0.173**	0.366
3.3		7.21	8.61		29.3	0.851	0.156	0.303
3.4		**13.3**	**20**	0.99	36.8	1.39	0.098	**0.318**
3.5	0.146	10.6	15.1		**40**	**1.90**	0.112	0.276
3.6			4.6			1.60		0.216
4	0.15		0.37					

Depth (m)	Borehole 8 (μSv/h)	17	18	19	20	21	22	23	24
0	0.087	0.111	0.145	0.118	0.103	0.096	0.096	0.0941	0.589
1	0.137	0.15	0.14	0.128	0.155	0.14	0.127	0.0932	0.471
2	0.163	0.205	0.151	0.181	0.46	0.458	0.33	0.208	0.95
2.5		0.3	0.2	0.28					0.94
3	1.16				4.05	0.75	0.26	0.150	13.4
3.1	1.42				5.64	1.14	0.331	0.163	17.9
3.2	1.76				8.4	1.78	0.43	0.169	26.0
3.3	2.23				12.8	2.35	0.66	0.175	36.9
3.4	3.2				22	2.71	1.07	0.229	66.9
3.5	4.84				28	2.83	1.4	0.241	49.2
3.6	5.67				23.5	2.86	1.85	0.248	28.5
3.7	5.42				13.8	3.43	2.2	0.217	14.2
3.8	5.81				10.7	3.84	1.98	0.213	6.85
3.9	7.34				10.9	5.20	1.99	0.222	4.85
4	10.5				11.4	6.17	2.96	0.278	3.71
4.1	15				13.5	7.35	4.43	0.312	3.31
4.2	25				20.0	9.34	6.83	0.316	3.54
4.3	44.8				28.4	15.1	10.6	0.326	3.56

Depth		Borehole				
4.4	70.2	42.3	16.3	15.6	0.305	4.56
4.5	108	84.3	38.3	25.8	0.471	6.21
4.6	168	148	**70.7**	58.1	0.705	7.85
4.7	**173**	**203**	70	**103**	0.829	**13.0**
4.8	92	130	25	57	0.719	8.59
4.9	38	46.1	8.7	20.6	0.559	4.47
5	12	13.3	4.9	6.4	0.473	1.84
5.1	7	8.13	30.1	3.0	0.509	1.07
5.2	3.2	4.06	14.1	1.3	0.335	0.64
5.3	2.7	2.46		0.74	0.255	0.44

NOTES:

	Highest gamma reading in the Borehole

NOTE: Background Levels are 0.1 μSv/h at ground level on the concrete pad where the boreholes were drilled.

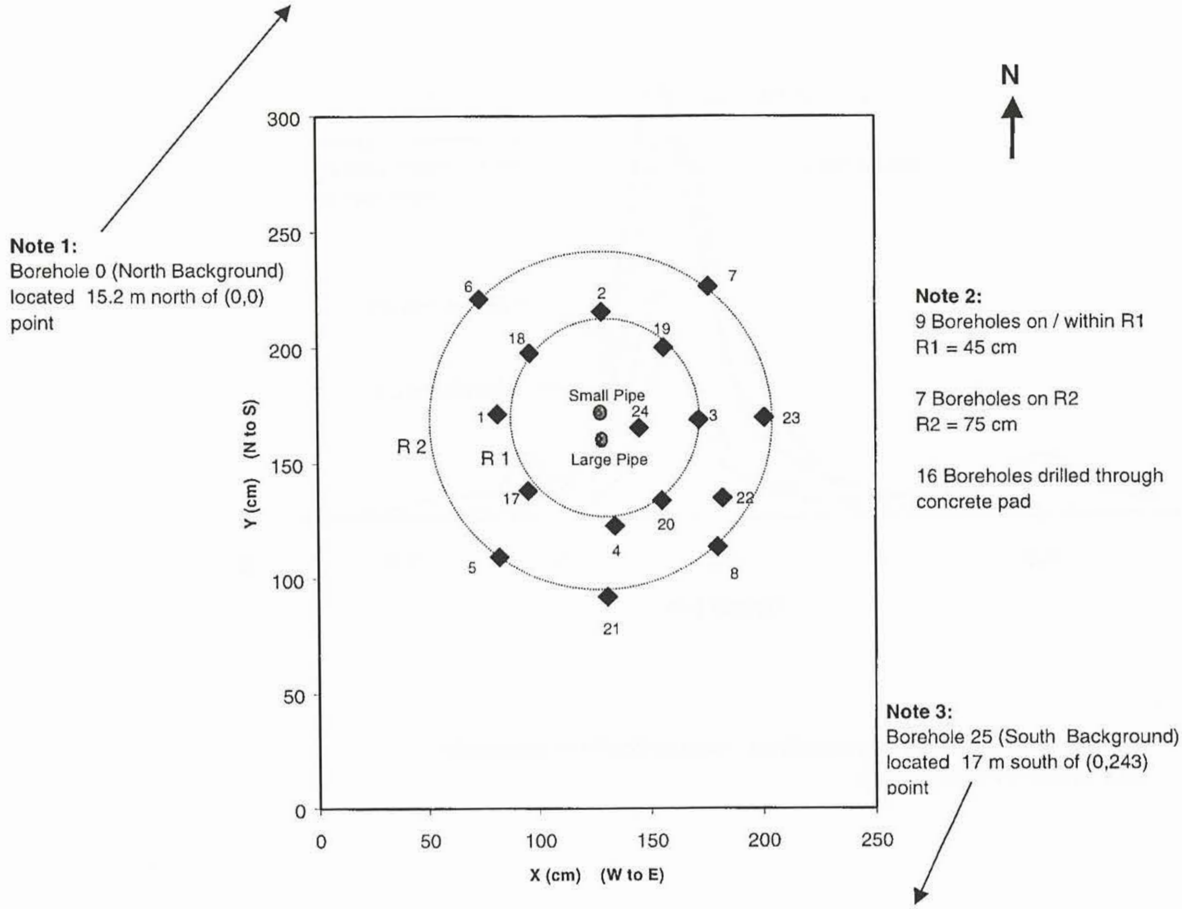

Figure 1 - Location of boreholes.

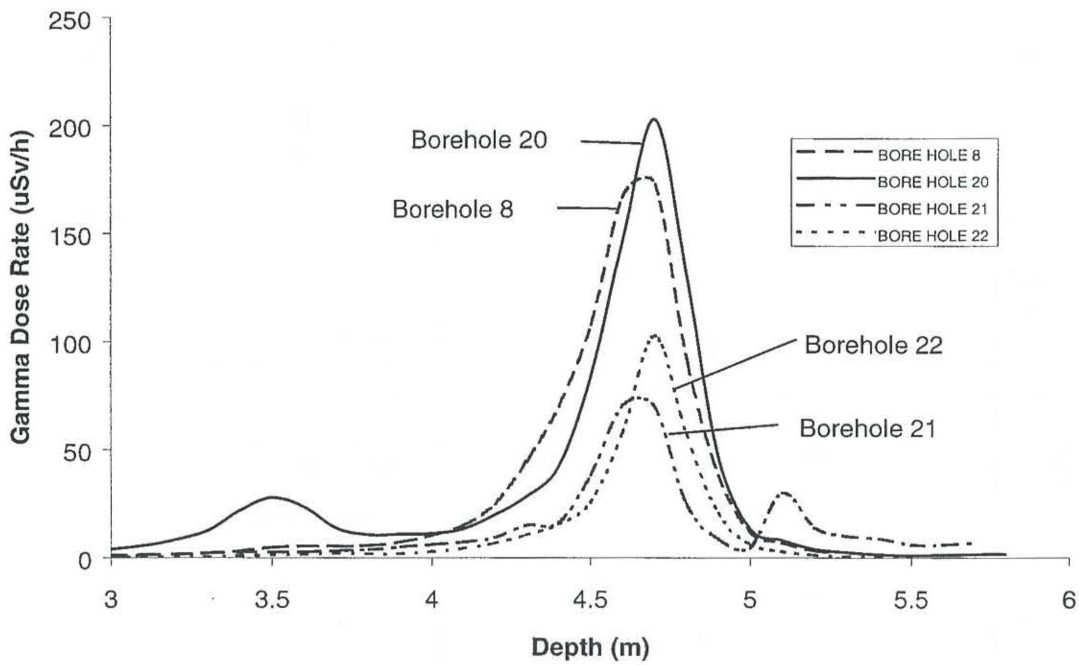

Figure 2 - Plot of gamma radiation readings versus depth in borehole (for selected boreholes).