INVESTIGATION OF STRONTIUM-90 IN GROUNDWATER DISCHARGING TO THE OTTAWA RIVER

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ABSTRACT

In 2007, an area of Ottawa Riverbed (about 250 m^2 in extent) was found to be a groundwater discharge zone with above-background levels of tritium (as tritiated water - HTO). The area was adjacent to the shoreline along the property boundary of Atomic Energy of Canada Limited's Chalk River Laboratories. The probable source of the HTO has been identified as a groundwater plume originating from a facility undergoing decommissioning. A discharge of strontium-90 (^{90}Sr) that occurs within a small part of the groundwater discharge zone in the study area has also been identified and is likely coming from the same groundwater plume as the HTO. Based on information from years of ongoing site and plume monitoring, a more detailed investigation was conducted to provide specific information on where the ^{90}Sr plume was emerging to the surface and to estimate contaminant flux to the river.

Work in 2009/10 revealed discharge of ^{90}Sr within a small portion of the groundwater discharge area. A land-based survey using a portable gamma spectrometer showed up to 550 total gamma counts per second above the plume's path. Based on a ^{137}Cs survey, these gamma counts were not attributable to ^{137}Cs and are likely due to the presence of ^{90}Sr and its decay product, yttrium-90 (^{90}Y). A vegetation survey of alders along the shoreline revealed gross beta levels of 1 – 70 Bq/g. Alders are known to root in wet ground and are likely pumping ^{90}Sr from the contaminated groundwater to the surface. Groundwater samples obtained from the river using mini-piezometers had gross beta levels of 3.3 – 590 Bq/L, with the highest values obtained near the area of the most contaminated vegetation. These same groundwater samples also contained elevated tritium. Seepage meters were used to calculate discharge rates to the river.

Although actions were taken to cut the source of ^{90}Sr to the aquifer, the residual ^{90}Sr plume continues to advance toward the Ottawa River. The primary goal of the current project is to quantify the discharge of ^{90}Sr that moves to the river annually at this location. The groundwater plume had been defined previously using widely spaced onshore monitoring wells 20 – 40 m from the shoreline. However, in this study, closely spaced mini-piezometers showed actual locations of ^{90}Sr discharge to the river and seepage meters provided direct measurements of discharge rates. As the project nears completion, the resulting data will be useful if remedial action is required.

1. INTRODUCTION

Groundwater is the primary method of transport for contaminants that get released into the aquatic system. The rate at which these contaminants are transported depends on the distribution coefficient (K_d), which describes how the contaminant partitions between the sediment and groundwater. Thus, some contaminants travel very rapidly through groundwater, while others can take several years to move even short distances.

Routine operations at Atomic Energy of Canada Limited (AECL)'s Chalk River Laboratories (CRL) since approximately 1945 have resulted in the formation of several plumes of contaminated groundwater. All of these plumes are monitored to determine relative contaminant migration rates and concentrations. Two radionuclides have been found to be particularly significant in groundwater flow systems: tritium, which is present as tritiated water (HTO), and strontium-90 (^{90}Sr). Both have moderately long half-lives (12.4 and 29.1 years for tritium and ^{90}Sr, respectively). Tritium, when released to the subsurface, moves at the velocity of the transporting groundwater. Several investigations at CRL have found that in many cases, ^{90}Sr moves at approximately 10% of the groundwater velocity in local overburden flow systems. Therefore, it can be transported appreciable distances, though at fairly slow velocities.

Routine monitoring is carried out near the sources of the plumes identified at CRL in order to accurately monitor plume inputs. Additionally, plume studies are carried out periodically to map the full extent of the plume and to qualitatively assess the risks of plume migration. These studies also assess the viability of the planned (or assumed) plume and source-area remediation activities. The contaminant plume that is the focus of this study originated from the Rod Bays of the National Research Experimental (NRX) Reactor, which operated from 1947 until 1992. The Rod Bays are a series of concrete pools that were used to handle and store irradiated fuel assemblies where the water in the pools would provide radiation shielding and cooling. The NRX Rod Bays were modified substantially in 1959; following these modifications, it was suspected that a leak had developed from the Rod Bays. The leak was confirmed in 1962 and several investigations followed to refine the location of the leak. Leakage to the subsurface continued until 2006, when the water was drained from the NRX Rod Bays and the source of the leak was successfully eliminated [1]. The plume has been closely monitored since the leak was discovered and extensive monitoring has continued since the elimination of the source. The main contaminants that have travelled a significant distance beyond the structure in the plume are ^{90}Sr and tritium (in the form of tritiated water – HTO); other radionuclides that are part of the plume have remained within 10 – 20 m of the Rod Bays structure. The plume begins at the NRX Rod Bays and extends 330 m to the Ottawa River. Part of the plume is intercepted by a building tile drain system [1]; the remainder of the plume continues uninterrupted to the river. The plume has been monitored by collecting samples from transects of boreholes and wells, with the last transect comprised of a line of boreholes and samplers spaced approximately 20 – 60 m apart in a line located 20 – 40 m from the Ottawa River shoreline. The current remediation strategy for this plume is to continue monitoring the movement and concentrations of the contaminants to confirm that the contamination remains at acceptable levels.

In the summer of 2007, seepage meters were placed along the Ottawa River shoreline and seepage rates up to 10 $L/m^2 \cdot day$ were measured in the area coinciding with the track of the groundwater plume. Subsequent groundwater analysis revealed elevated levels of HTO entering the river comparable to those found in the groundwater influenced by the contaminated plume. This led to a study of the sediment in the area. Sediment samples were collected in intervals of

10 m, or at 1 m intervals if contamination was detected. The results showed that gross beta levels were up to seven times greater than background in the area where the plume was projected to be discharging. The elevated gross beta measurements suggested the presence of ^{90}Sr in the area. As a result of these findings, follow-up was initiated in 2008/2009 to locate and quantify groundwater transport of ^{90}Sr to the Ottawa River along the CRL shoreline.

2. MATERIALS AND METHODS

2.1 Field work

To further characterize the study area, mini-piezometers were installed to measure hydraulic head and to obtain samples of groundwater [2]. Mini-piezometers consist of a length of polyethylene tubing with holes punched in the bottom 10 cm. These holes are then covered by a nylon screen secured to the tubing with electrical tape. To install the piezometers (Figure 1), a piece of 1/2-inch stainless steel pipe with a bolt inserted in the bottom was hammered into the riverbed. The piezometer was placed inside the pipe, screen-end first. The pipe was then removed by gently twisting and pulling up simultaneously, while ensuring the piezometer stayed in place. Once the piezometer was installed and it was determined to be responsive with a hydraulic head greater than the river level, a plastic bag was attached to the tubing to allow artesian flow to add water to the submerged bag and provide groundwater samples over varying periods of time. Rates of flow and governing gradients were used to calculate the hydraulic conductivity of the material at the screen, as described below.

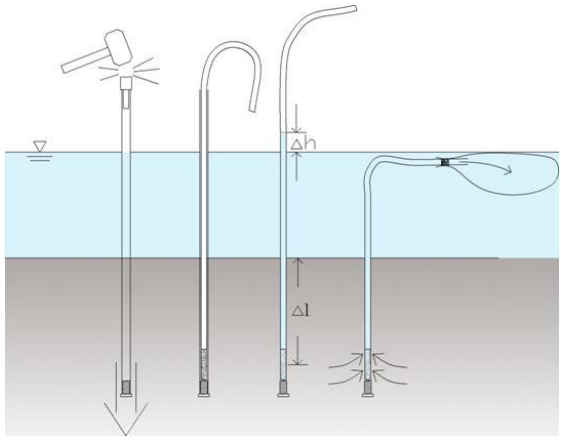

Figure 1. The steps for installation and use of a mini-piezometer. The figure was adapted from [2].

Seepage meters were used in conjunction with mini-piezometers. Seepage meters give a direct measurement of groundwater seepage flux through the riverbed [2]. They are constructed by cutting off the top or bottom fifth of a large drum and punching a hole near the rim in the top and side. The hole on the side is plugged with a rubber stopper and is only used if the water is too

shallow to allow the top hole to be used. A #5 rubber stopper with a hole punched in the center is placed in the top hole of the seepage meter. A plastic bag is attached to a piece of tubing in the rubber stopper via a piece of amber tubing. One liter of water is placed in the bag prior to attaching it as previous studies have shown that not pre-filling bags can result in an anomalous influx of water into the bags for a short period of time after they are attached [3]. The groundwater collected in the bag over a given period of time gives the rate of seepage at that location. The groundwater seepage flux and the hydraulic head from the mini-piezometer can be used to calculate the hydraulic conductivity. Figure 2 shows the set-up of the seepage meter.

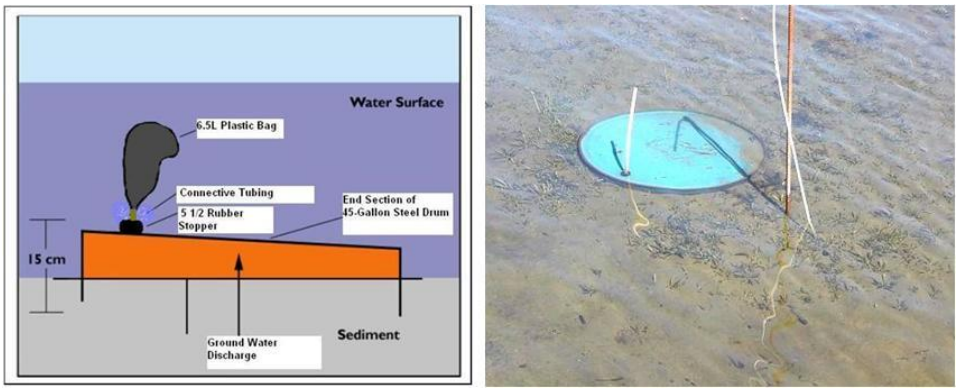

Figure 2. Set-up of a seepage meter. The figure on the left shows the components of the seepage meter and was adapted from [4]. The figure on the right shows a seepage meter and a mini-piezometer set up together.

When the piezometers were not in use, they were secured in an upright position by attaching them to a piece of rebar with a twist-tie. The hydraulic head was measured before the bag was attached to the piezometer, as well as after the water sample was taken. The second head measurement was taken after allowing sufficient time for the piezometer to re-equilibrate in the upright position. The time was recorded as soon as the plastic bags were attached to the seepage meters and mini-piezometers. Once the sampling period was over, the time was again recorded and the volume of the groundwater samples was measured. For the samples taken from the mini-piezometers, a 1-L aliquot was poured into a sample bottle. In the summer of 2010, water levels were extremely low and a number of the piezometers were no longer in water (the river level had dropped below the elevation of the riverbed where the instruments had been installed). Therefore, groundwater was pumped from the piezometers using a Geopump.

To calculate groundwater flux, the volume of water collected from the seepage meters was divided by the period of collection time in days. This yielded a value in cm^3/day. This was then divided by the area of the seepage meter to obtain the flux in $cm^3/m^2 \cdot day$. To determine the hydraulic conductivity, Hvorslev's equation [5] was used:

$$K_h = \frac{Q \ln[\left(\frac{mL}{D}\right) + \left(1 + \left(\frac{mL}{D}\right)^2\right)^{0.5}]}{2\pi L H_c} \qquad (1)$$

Where k = hydraulic conductivity (cm/sec)

Q = seepage flux (cm^3/sec)

D = diameter of intake tube (cm)

H_c = change in hydraulic head ($H_{after} - H_{before}$, or ΔH)

L = length of mini-piezometer (cm)

m = transformation ratio (assumed equal to 1)

To supplement the groundwater data, samples of sediment, vegetation and river water under the ice sheet were also collected and analyzed. Sediment samples were collected from the study area in 2010 and 2011 using piston cores [6]. Briefly, a core tube was fitted with a piston unit, comprised of alternating rubber and steel washers, in the bottom and secured by a rope through the core tube to a 2 x 4 piece of board so that it would not move during coring. The core tube was pounded into the riverbed; as this occurred, the piston remained fixed in place, creating suction and drawing the sediment into the core tube. Once the desired depth was reached, the core tube was removed from the sediment and the apparatus was disassembled to retrieve the core of sediment. The sediment was sectioned at the desired intervals in the lab. Each interval was dried and split until 0.4 g remained.

Water samples were also collected from underneath the ice sheet. Holes were drilled in the ice using an auger and a 1-L sample of the water directly beneath the ice was collected.

A vegetation survey was performed by collecting new growth twigs from alders (~90%), as well as sumac and maple (~10%), growing in the study area. A radiation survey was completed by manually carrying a portable gamma spectrometer (Exploranium Model GR460) with the detector held 10 – 20 cm above the soil in the study area.

2.2 Sample analysis

Prior to anion analysis, the water samples were filtered. Before being analyzed for gross beta, the filtered groundwater samples and water samples from under the ice were acidified with 1% concentrated nitric acid. Electrical conductivity, gross beta activity of groundwater samples and the chloride concentrations are presented as averages of the 2009 and 2010 field seasons.

2.2.1 Electrical conductivity

Electrical conductivity of the samples was measured using a Thermo Scientific Orion 3 Star portable conductivity meter.

2.2.2 Gross beta

To determine the gross beta levels in the vegetation, samples were dried, ashed, mounted on planchettes and counted on a Tennelec LB5100 α/β counter.

To determine gross beta of groundwater samples and the water samples collected from under the ice, 500 ml of water from each sample was evaporated onto a stainless steel planchette and fixed with collodion (10% collodion, 90% acetone). The planchettes were then counted on a Tennelec LB5100 α/β counter.

To determine gross beta of sediment samples, 0.4 g of each interval were mounted on a stainless steel planchette and fixed with collodion (10% collodion, 90% acetone). The planchettes were then also counted on the Tennelec LB5100 α/β counter.

2.2.3 Chloride

The water samples were analyzed for chloride using ion chromatography (Dionex ICS-1500).

3. RESULTS AND DISCUSSION

Figure 3 shows the NRX Rod Bays plume and the study area in the river. As evident in the left-hand panel of Figure 3, although the plume is wide, there is one specific zone where the discharge contains higher levels of gross beta activity as compared to the rest of the plume. The right-hand panel of Figure 3 shows the location of the mini-piezometers and seepage meters. Wherever possible, seepage meters and mini-piezometers were placed together. The area that is expected to be most influenced by the plume, and that was the focus of this study, includes the cluster of instruments that starts with 11, 43, 44 and 45 at the shoreline and extends offshore to the location of the stilling well (Figure 3).

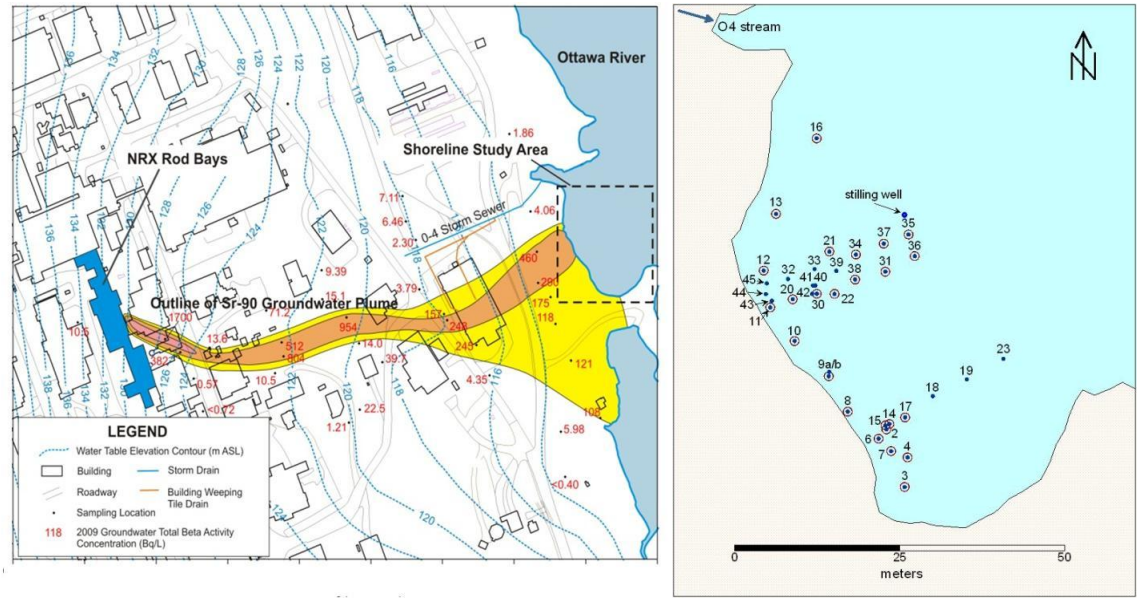

Figure 3. Map of the groundwater discharge study area. The left-hand panel shows the track of the NRX Rod Bays groundwater plume as determined by monitoring wells on land and the shoreline study area. The right-hand panel shows a zoomed-in view of the study area and the locations of the mini-piezometers (blue dots) and seepage meters (red circles).

3.1 Seepage flux and hydraulic gradient

Seepage flux was calculated at each of the seepage meter locations. Calculations in 2010 were sparse due to abnormally low water levels that prevented the use of seepage meters for the majority of the field season. In general, flux in 2010 was higher than that measured in 2009 (

Table 1). Negative seepage flux was obtained in two locations in 2009. The reason for this is unknown; however, possibilities include groundwater recharge or technical error.

Table 1. Average seepage flux measured in the 2009 and 2010 field seasons.

	Seepage Flux ($cm^3/m^2 \cdot day$)	
Seepage Meter	**2009**	**2010**
SM-02-09	11116	22537
SM-03-09	2827	n/a
SM-04-09	8760	n/a
SM-06-09	11517	83774
SM-07-09	5869	12260
SM-08-09	11310	n/a
SM-09-09	9337	n/a
SM-10-09	32297	n/a
SM-11-09	18590	n/a
SM-12-09	2565	n/a
SM-13-09	11499	n/a
SM-17-09	2246	12162
SM-18-09	n/a	4945
SM-20-09	-5315	n/a
SM-21-09	-6925	n/a
SM-22-09	2198	n/a

Hydraulic conductivity values were calculated at the mini-piezometer locations using Equation 1 (Section 2.1). Hydraulic conductivity is a measure of how easily water moves through pore spaces, fractures in rock or sediment. These values can vary by several orders of magnitude. In the Ottawa River, the site geology is complex and includes crystalline bedrock, organic sands, glacial till, sand and gravel. The conditions are hydrogeologically difficult because the permeability of these materials varies by a factor of 10^6 or greater. Thus, due to this and the fact that the Ottawa River sediment is not homogenous, the hydraulic conductivity varies extensively. All of the measurements in this study were done in riverbed sands. Even so, considerable variation in hydraulic conductivity was noted. In 2009, the values ranged from 6E-07 to 1.18E-01 cm/s. In 2010, with limited data due to excessively low water levels, the values for hydraulic conductivity ranged from 3E-05 to 2.84E-03 cm/s.

3.2 Electrical conductivity

Electrical conductivity is a measure of the total dissolved solids in a sample. The electrical conductivity of the Ottawa River water is approximately 0.06 mS/cm. Electrical conductivity in the groundwater samples from the mini-piezometers reached up to 1.45 mS/cm at MP-11 and

was lowest at MP-35 and MP-36, falling to 0.08 mS/cm (Figure 4, left panel). Overall, there was no obvious trend and electrical conductivity did not seem to be dependent on the mini-piezometer's location. Groundwater monitoring from wells on land approximately 20 m from the shoreline gave electrical conductivity values generally ranging from 0.4 – 1.6 mS/cm, with a high of 2.57 mS/cm, and the high values are attributed to the use of road salt on site [1]. The levels measured in this study fall within this range and thus are also likely to be attributable to the infiltration of road salt into the groundwater on site. This conclusion was supported by also measuring the electrical conductivity in the samples taken from beneath the ice sheet. These samples also showed higher electrical conductivity in the near-shore area where plume discharge would occur, measuring approximately 0.2 – 0.4 mS/cm compared to 0.06 – 0.1 mS/cm further from shore (Figure 4, right panel).

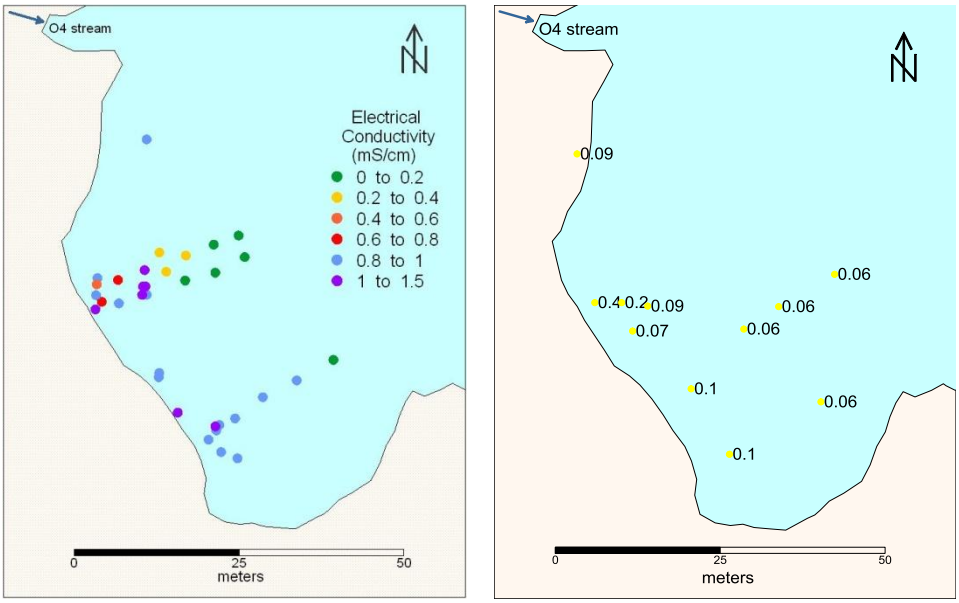

Figure 4. Average electrical conductivity measurements in groundwater collected from the mini-piezometers (right) and grab samples of river water collected under the ice sheet (left). All values are in mS/cm. The Ottawa River itself has an electrical conductivity of 0.06 mS/cm.

3.3 Gross beta

3.3.1 Vegetation/land survey

The radiation survey of the land near the shoreline registered 550 total counts/second (Figure 5, left panel). Given the elevated total counts, a follow-up survey for ^{137}Cs was completed. The survey revealed that the elevated counts were not attributable to ^{137}Cs, as this radionuclide was not detectable in the area. Thus, given these results and the history of the site, the elevated counts are likely due to the presence of ^{90}Sr and its daughter product, ^{90}Y. Although both of these radionuclides are pure beta emitters, some of the beta particles, especially from ^{90}Y, have

enough energy to emit low-energy gamma rays as Bremsstrahlung radiation [7]. This gamma can then be detected by the portable gamma spectrometer.

The land in the study area is covered primarily by alder trees (*Alnus rugosa*) and these elevated counts are likely a result of a surface manifestation of biological pumping of ^{90}Sr to the surface by the phyreatophytic alders. Alder trees are known to root in wet ground. Thus, if ^{90}Sr-contaminated groundwater is flowing and rising into the capillary fringe, it would be taken up by the alders. Since ^{90}Sr has similar biological behavior to calcium, capillary rise and transpiration could move ^{90}Sr from the groundwater to the plant and into leaves that accumulate on the land surface. The 2011 vegetation survey supported the conclusions made based on the land survey. The gross beta results of the vegetation revealed levels up to 58 Bq/g (dw) along the shoreline where the plume is expected to be approaching the river (Figure 5, right panel). This also agreed well with a vegetation survey completed in 2009, which revealed levels up to 70 Bq/g gross beta, with the highest values occurring in the same location as the 2011 survey.

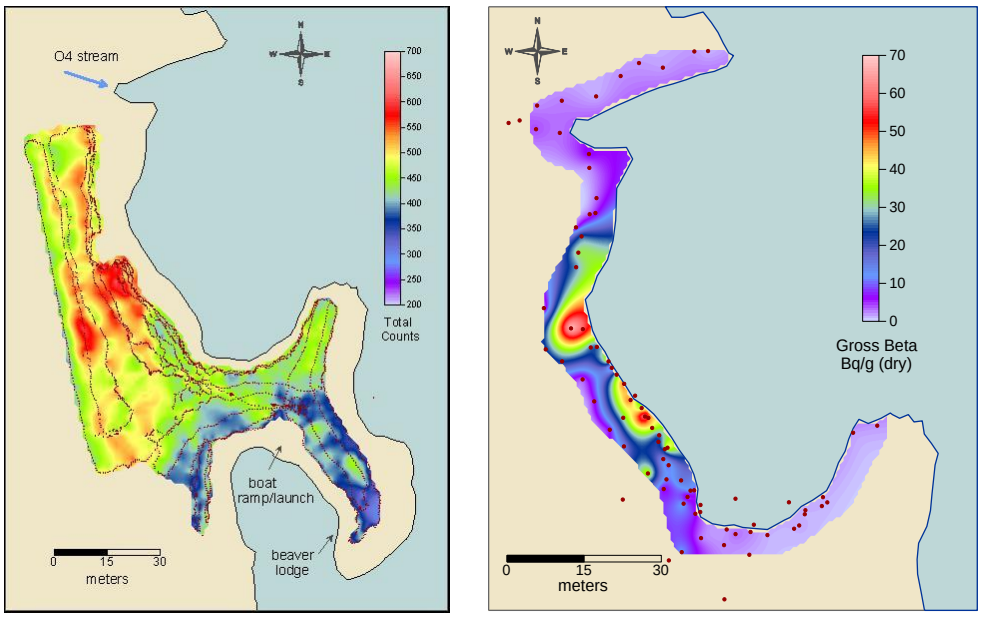

Figure 5. Land-based and vegetation surveys of the shoreline where the plume discharges to the river. The land-based survey in the left panel shows radiation levels in total counts per second. The vegetation survey in the right panel shows gross beta levels in Bq/g (dw).

3.3.2 Groundwater

Gross beta activity in the discharging groundwater was elevated in the shoreline area directly downgradient of where the highest concentrations of beta activity were observed in groundwater monitoring wells. Levels reached as high as almost 600 Bq/L in the groundwater collected from the mini-piezometer closest to shore (Figure 6, left panel). As mentioned, a previous gamma spectrometer survey carried out on the sediment in this area revealed elevated gamma counts not attributable to ^{137}Cs, but likely due to ^{90}Sr/^{90}Y. Concentrations of gross beta decreased as distance from shore increased. Concentrations were lower in the lower section of the study area,

peaking at about 200 Bq/L (Figure 6, left panel). Although the plume discharge zone extends to this area, the gross beta activity is this portion of the plume is lower (Figure 3, left panel). Analysis of groundwater from the monitoring wells located on land to track the plume in question had slightly higher concentrations of gross beta in 2009 as compared to previous years, with a peak value of 460 Bq/L [1]. Thus, the data obtained from monitoring wells on land lines up relatively well with the data obtained from instrumentation in the river along the shoreline. All of the elevated levels are attributable to the contaminated groundwater plume.

Water samples were also collected from the river through the ice in March. Groundwater is known to discharge near the river's edge in order to get to an area of low elevation head. Thus, it was theorized that the discharging groundwater would move upwards towards the ice sheet and as a result, increased gross beta levels would be observed. This was in fact the case, as seen in the right panel of Figure 6. The gross beta levels of the water from under the ice where the plume discharges near shore were significantly higher compared to other locations further offshore, reaching as high as 49 Bq/L compared with values of 0.2 – 16 Bq/L elsewhere.

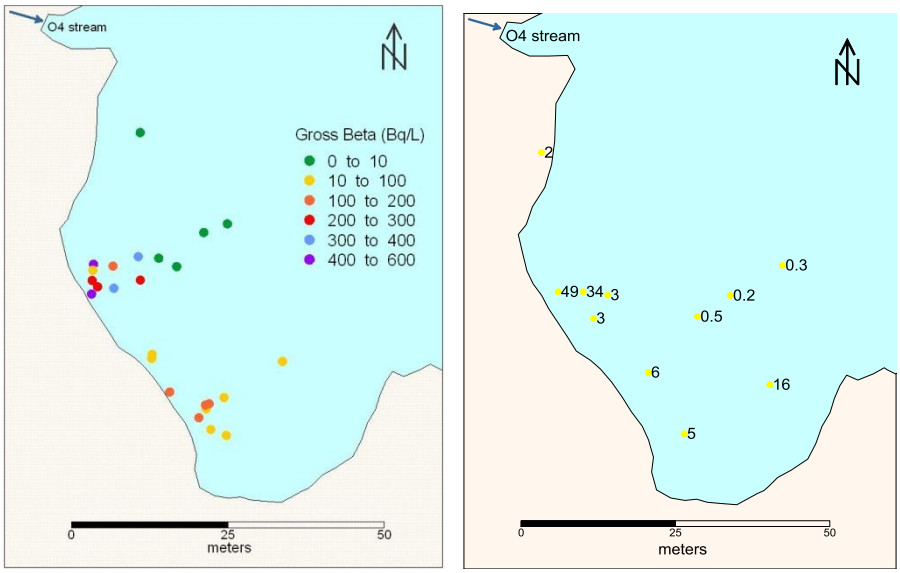

Figure 6. Average gross beta concentrations in the groundwater collected from the mini-piezometers (left) and grab samples of water from under the ice sheet (right). All values are in Bq/L.

Taken together, the data provides evidence that the ^{90}Sr plume is discharging directly into the Ottawa River.

3.3.3 Sediment

To support the conclusions based on the land and groundwater data, sediment was sampled by piston coring in the area where groundwater analyses have revealed elevated levels of gross beta (Figure 7). The depth of the cores varied, depending on whether the core tube was able to penetrate deep into the sediment or whether it encountered any rocks or other obstacles in the

riverbed. PC-11-01 to PC-11-05 were collected as part of a separate study and therefore, the results are not discussed here.

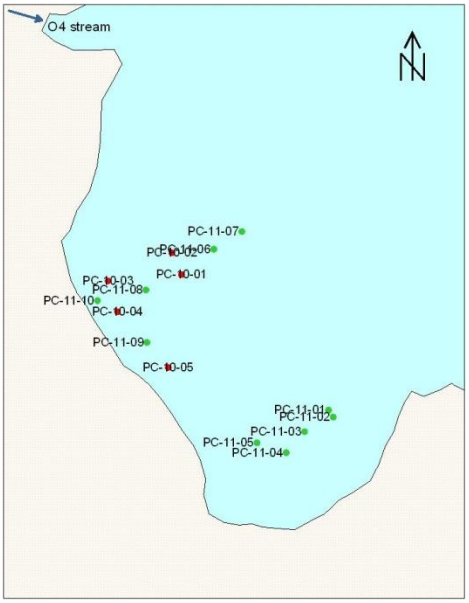

Figure 7. Piston coring sample locations.

The cores collected in 2010 revealed elevated levels of gross beta, with the exception of PC-10-1 and PC-10-2, which hovered close to background (~1.5 Bq/g) and were collected further offshore (Figure 8, left panel). The highest levels occurred at depths of 10 cm or greater and were closer to shore. This is consistent with groundwater flow paths in which discharge occurs close to the shoreline and thus, higher contaminant levels are observed in these locations [8]. The plume becomes more diluted as it moves offshore, resulting in lower contaminant concentrations. More cores were taken in 2011 once the area in question had been further characterized. Again, the results showed elevated gross beta levels in sediment samples taken close to shore (Figure 8, right panel). These levels decreased to background as distance from shore increased. It should be noted that the concentrations were lower at some of the locations added in 2011, which is likely an indication that they are not situated in direct line with the highest plume discharge zone. Thus, this data also helps to delineate the boundaries of the plume discharge.

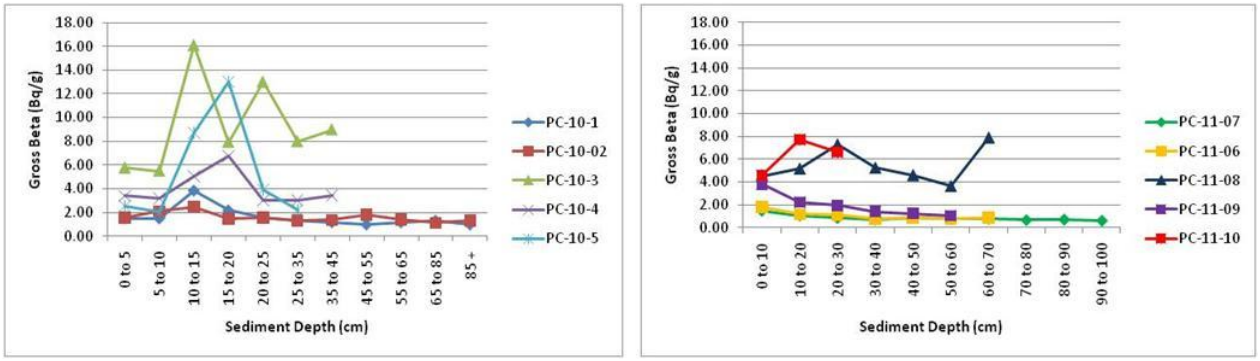

Figure 8. Levels of gross beta (Bq/g) with depth in sediment samples collected from the study area in 2010 (left panel) and 2011 (right panel).

3.4 Chloride

Major anions were measured in the groundwater samples as an indicator of water quality. Sulfate, fluoride and nitrate were not found to be elevated. Of the anions measured, chloride was present in the highest concentrations. Chloride was found in concentrations of up to 300 mg/L in the groundwater sampled closest to shore (Figure 9, left panel). The concentrations decreased as distance from shore increased. Chloride was also detected in the samples collected from under the ice sheet (Figure 9, right panel). The elevated chloride is likely a result of run-off from the use of road salt on site, another line of evidence that groundwater is entering the Ottawa River from the CRL site at this location.

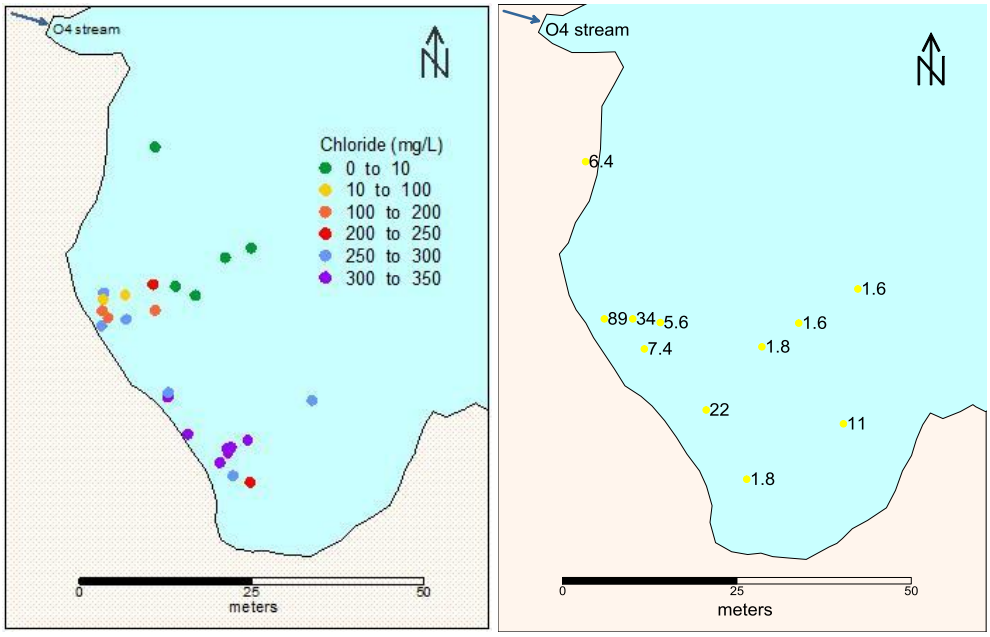

Figure 9. Average chloride concentrations in groundwater collected from the mini-piezometers (left) and grab samples of water from under the ice sheet (right). All values are in mg/L.

4. CONCLUSIONS

Taken together, the gross beta levels in the vegetation, the groundwater and the sediment provide strong evidence that the contaminated plume from the NRX Rod Bays is transporting ^{90}Sr directly into the Ottawa River. Elevated levels of electrical conductivity and chloride from road salt provided supporting evidence of plume discharge to the river. Although the gross beta concentrations are high near-shore, the levels decrease quickly as distance from shore increases. This, along with the fact that the study area is in a rugged embayment adjacent to a nuclear facility, decreases the chances that the public would come into contact with the contamination. Based on the data collected from the monitoring wells on land, the current remedial strategy is to continue monitoring the plume to ensure that the contamination remains at acceptable levels. The characterization data obtained in this study from instrumentation in the Ottawa River, as

well as a planned human health and ecological risk assessment of the area, will help determine if the remedial strategy is still adequate or if further action is required.

5. REFERENCES

1. Audet, M., "CRL Groundwater Monitoring Program Annual Report for 2009", 2011 March, 3611-01612-REPT-008. R0.
2. Lee, D.R., Cherry, J.A., "A field exercise on groundwater flow using seepage meters and mini-piezometers", *Journal of Geological Education*, Vol. 27, 1978, pp.6-10.
3. Shaw, R.D., Prepas, E.E., "Anomalous, short-term influx of water into seepage meters", *Limnology and Oceanography*, Vol.34, Iss.7, 1989, pp.1343-1351.
4. Lee, D.R. "A device for measuring seepage flux in lakes and estuaries", *Limnology and Oceanography*, Vol. 22, Iss.1, 1977, pp.140-147.
5. Hvorslev, M.J., "Subsurface Exploration and Sampling of Soils for Civil Engineering Purposes", Report on a Research Project of the American Society for Civil Engineers, 1949, Waterways Experiment Station, Vicksburg, Mississippi.
6. Munch, J.H., Killey, R.W.D., "Equipment and methodology for sampling and testing cohesionless sediments", *Ground Water Monitoring Review*, 1985, Winter, pp.38-42.
7. Killey, D. Personal Communication. 2011.
8. Lee, D.R., Cherry, J.A. and J.F. Pickens, "Groundwater transport of a salt tracer through a sandy lakebed", *Limnology and Oceanography*, Vol.25, Iss.1, 1980, pp.45-61.