

VERTICAL DISTRIBUTION OF RADIOACTIVE PARTICLES IN OTTAWA RIVER SEDIMENT NEAR THE CHALK RIVER LABORATORIES

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ABSTRACT

Previously, we described an area of above-background levels of radioactivity in the bed of the Ottawa River near the Chalk River Laboratories. The area was about 200 m wide by 400 m long and in water 8 to 30 m deep. The source of the radioactivity was associated with the location of cooling-water discharge [1]. Particles of radioactive material were later recovered from the upper 10-15 cm of sediment and were determined to be sand-sized grains of nuclear fuel and corrosion products [2].

This report provides an examination of the vertical distribution of radioactive particles in the riverbed. Twenty-three dredge samples (representing 1.2 m² of riverbed) were collected near the Process Outfall. Each dredge sample was dissected in horizontal intervals 1-cm-thick. Each interval provided a 524 cm³ sample of sediment that was carefully examined for particulate radioactivity. Approximately 80% of the radioactivity appeared to be associated with discrete particles. Although the natural sediment in the general area is cohesive, silty clay and contains less than 10% sand, the sediment near the Outfall was found to be rich in natural sand, presumably from sources such as winter sanding of roads at the laboratories. The radioactive particles were almost entirely contained in the top-most 10 cm of the river bed. The majority of the particles were found several centimetres beneath the sediment surface and the numbers of particles and the radioactivity of the particles peaked 3 to 7 cm below the sediment surface.

Based on the sediment profile, there appeared to have been a marked decrease in the deposition of particulate radioactivity in recent decades. The vertical distribution of radioactive particles indicated that sedimentation is resulting in burial and that the deposition of most of the particulate radioactivity coincided with the operation of Chalk River's NRX reactor from 1947 to 1992.

1. INTRODUCTION

The presence of radioactive particles and an elliptical pattern of sedimented radioactivity in the riverbed near the cooling water outfall were reported by Ophel in 1959 [3]. Careful reading of Ophel's work suggests that the accident of December 12, 1952 released particulate radioactivity which was deposited in deep water near the laboratory site. The lateral and vertical distribution of residual, particulate radioactivity remained largely ignored until 2001 when we conducted a reconnaissance survey of the riverbed and delineated a pattern of radioactivity generally similar to Ophel's [1]. Our reconnaissance map showed a footprint of above-background radioactivity that was approximately 400 m long in the direction of river flow and about 200 m wide and in water 8 to 30 m deep (Figure 1). The highest levels of radioactivity were near the cooling water outfall at the upriver end of the footprint. The bulk sediment in this area contained above-background levels of radioactivity in the topmost 10 to 15 cm of the sediments [2]. Discrete, sand-sized fragments of radioactive material were identified as bits of nuclear fuel and corrosion products [2].

This paper provides the results of a centimetre-scale examination of the vertical distribution of radioactive particulate matter in the central part of the footprint and within 110 m of the Process Outfall. This paper also compares the apparent sedimentation rate near the Outfall to the natural sedimentation rate. The results provide a basis for understanding sediment behavior at this location and the likely chronology of radioactive particles released and settled near the Outfall. The results may aid in making future decisions as to whether and where mitigation may be necessary and, if so, how it might be achieved.

2. PHYSICAL SETTING

A built-up area, now known as Chalk River Laboratories (CRL), is situated on the Ontario side of the Ottawa River. It has been the site of Canada's primary nuclear research and development activities for the past 66 years. The Ottawa River adjacent to CRL is up to 1 km wide and more than 30 m deep. The current is slow, e.g. [4], and, based on core analysis, has not prevented accumulation of cohesive sediment in water over 8 m deep [1]. Sediments in the Ottawa River in the somewhat lake-like area near CRL are comprised of clay (~65%), silt (~35%) and sand (10% or less) [1].

The literature indicates that alluvial sands were deposited during post glacial times when the Ottawa River was at higher elevations [5]. An estimated seasonal maximum base-flow discharge on the order of 200,000 m³/s prevailed in this part of the Ottawa River channel 9,600 to 8,300 years before present (BP) [6]. Such discharges would have washed the channel, leaving bedrock, stiff glacial till, washed till, boulders, gravels and sand on the floor of the paleo-riverbed. During collaborative work with personnel of the Geological Survey of Canada in 1984 and 1985, the stratigraphy of riverbed sediments was visualized using sub-bottom sonar. Acoustically transparent sediment, up to 9-m thick, blanketed a relatively smooth, acoustically opaque layer. This reflector was interpreted as "sand" [7], and it is probable that this sub-bottom reflector represents the washed channel, left by the last major post-glacial flood of about 8,300 years BP. Based on the sub-bottom sonar, the flow of the river after 8,000 years BP appears to have allowed the accumulation of soft sediments comprised of clay, silt and minor amounts of sand in areas where the channel was wide and deep. Our gravity cores have allowed us to sample only the uppermost 0.5 m of riverbed where, in water deeper than 8 m, the sediment is silty clay.

However, because the sonar did not show a change in the nature of the sediment below the upper 0.5 m and above the “smooth-looking reflector,” one might estimate that the silty clay, which we have observed near the sediment water interface, represents the predominant material throughout the entire acoustically transparent layer.

If the above estimate is correct, then the smooth, basal surface can be used to derive the rate of sedimentation (the acoustically transparent layer) since the catastrophic flood of about 8,300 years ago. Because the maximum thickness of this layer above the smooth-looking reflector is about 9 m where the river is 25 to 60 m deep, the 8,300 year average sedimentation rate in that area would appear to be about 1 mm/yr. The thickness of the sediment in the centre of the footprint area, in 15 to 25 m of water, is about 5 m. This suggests a long-term average sedimentation rate in the footprint of about 0.5 mm/year. While additional work is required to draw firm conclusions on the long-term rates of sedimentation, it may be sufficient at this time to expect variable sedimentation rates at various locations, as it may be possible to do based on the thickness of the acoustically opaque layer that drapes the smooth-looking reflector.

Using the maximum recorded spring flow of $4,800 \text{ m}^3/\text{s}$, which occurred in 1979, it is likely that the average flow of the river at CRL can, for flood periods, be as high as 0.2 - 0.3 m/s. Hydrodynamic modeling, which takes into account the channel morphology, water level elevations and flow, has shown that riverbed shear stresses in the footprint, produced by historically maximum flows, may be insufficient to erode the sediment [8]. The presence of cohesive silty clay sediments in the footprint is evidence that the area is a net depositional region.

The discharge through the Process Outfall in recent years has been about $1 \text{ m}^3/\text{s}$. The discharge enters the river at a depth of 18 m and is forced through upwardly-directed diffuser pipes. The force of this discharge pushes water and suspended solids to the surface of the river. This creates a visible turbulence and keeps a circular area, more than 10 m in diameter, ice-free in winter. At the river's surface, the turbulent water moves somewhat radially from the centre of the upwelling and broadens the dispersal area. Because river current is typically less than 0.1 m/s, sand and sand-sized solids in the discharge would tend to descend and accumulate nearby. If particles of nuclear fuel were occasionally present in the discharge, their relatively high density would result in a settling velocity higher than that of natural sand and this would decrease the distance they could move before contacting the riverbed. We therefore expected to find sand-sized particulate matter and a higher density of sand-sized fragments of nuclear material within the cohesive sediment near the Outfall.

3. METHODS

Samples of the upper 10 cm of sediment were obtained for grain size analysis within 30 m upriver, within 20 m downriver and between 65 and 110 m downriver from the Outfall using a Petite Ponar dredge. The samples were analyzed for grain size using sieves to determine the sand fraction and a hydrometer to determine the silt and clay fractions [9].

Previous work on the vertical distribution of man-made radioactivity used 0.5 g samples of sediment and had, therefore, focused on the bulk sediment, not on the relatively infrequent occurrence of radioactive particles in the sediment. Analysis of the bulk sediment from cores had revealed vertical variations in radioactivities which were large and variable, suggesting the presence of discrete radioactive particulate matter [1]. However, except for identifying the upper

10 -15 cm as containing most of the radioactivity, no report had focused on the vertical distribution of particulate radioactivity. Previous work using cores, having a cross-sectional area of 11.9 cm^2 , rarely encountered significant particulate radioactivity. Therefore, a larger area sampler was used to allow a depth-wise enumeration of the particulate radioactivity.

For the analysis reported here, we used an Ekman dredge, which samples an area of 524 cm^2 . The Ekman dredge was modified so that it could be dissected in 1-cm thick horizontal slices. The modification involved 1) replacement of the original top bracket with a taller bracket, 2) addition of bolts with wing nuts screwed through the sides of the bracket to ensure that the top would close when the dredge was being raised from the riverbed, and 3) the addition of lead weights to the sides of the dredge to improve penetration into the cohesive sediment (Figure 1).

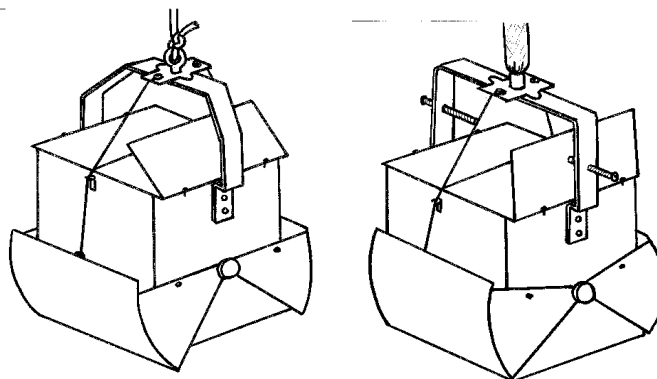


Figure 1. Original (left) and modified (right) Ekman dredges. Lead weights (not shown) were attached to the vertical sides of the top bracket of the modified dredge.

A laundry tub of river sediment was used to test the accuracy of top-down dissection in assessing particle depths. Coloured 2-mm bits of plastic wire were mixed into samples of sediment such that each centimetre layer in the container would have its own distinctly labeled particles. A spatula with bent edges was used to improve recovery of sediment from each 1-cm-thick slice. Test results showed that 70% of the labeled particles were recovered from their correct 1-cm depth and most of the remaining 30% were observed one centimetre above or below their correct depth.

To investigate the depth distribution of radioactive particles within 110 m of the Outfall, 19 Ekman dredge samples were obtained from locations downriver of the Outfall in 2005. In 2008, 4 additional samples were collected upriver and within 30 m of the outfall for a total of 23 dredge samples and a total sampling coverage of 1.2 m^2 of riverbed. The locations are shown in relation to the Outfall in figure 2. The riverbed slope in the sampling area was about 0.2 and the sample depths ranged from 17 to 23 m. Global positioning and a sonar depth finder were used to select locations and depths for sampling.

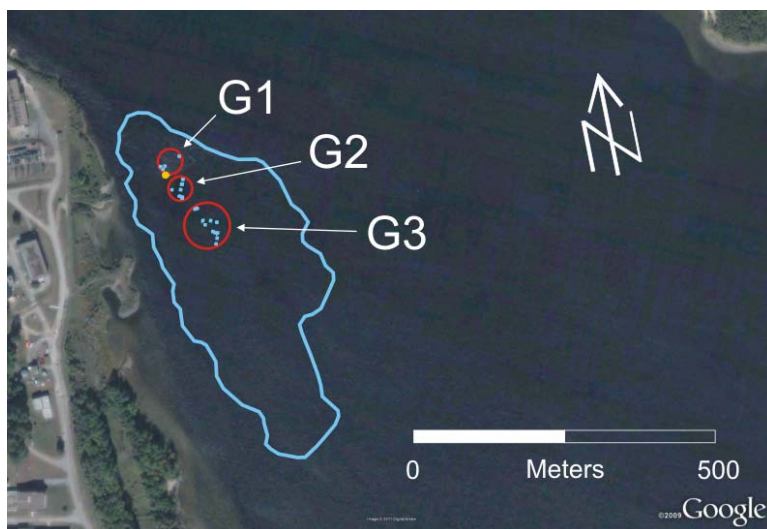


Figure 2. Outline around the above-background radioactivity in the sediments of the Ottawa River in water 8 to 30 m deep [10]. The location of each dredge sample and the three groups of samples (G1, G2 and G3) are shown in relation to the Outfall location (gold dot).

Anchors were not dropped during sampling because this would have disturbed the bottom and affected subsequent investigations. The river water on the top of each dredge sample was siphoned off using ¼-inch ID tubing. A peristaltic Geopump and ¼-inch ID tubing was used to recover the top centimetre of sediment by suction and this was transferred to a 4 L jug. This worked so well that the second one-centimetre layer was also transferred in the same way to a second jug. A 6-cm wide, stainless steel spatula with one side bent upward was used to remove successive, 1-cm-thick layers of sediment from the dredge. Once the top 9 cm of sediment had been removed and placed in plastic bags, the remaining sediment was placed in another plastic bag. The upper 2 cm of sediment contained more water than the deeper sediments. These were evaporated at 75°C until they attained a sticky consistency, similar to that of the deeper samples. During evaporation, residual mud, hardening and adhering to the inside of the container, was mixed into the wet mud. Each 1-cm layer sample of mud (about 530 cm³) was placed in a plastic bag and flattened in a large tray to a thickness of < 1 cm. The flattened sediment was scanned to locate active particles and to obtain a contamination meter reading on each. Scanning was performed by moving the pancake detector slowly across the surface of the plastic bag. The scanning was done using a Ludlum M-12 Ratemeter with 44-9 Geiger-Mueller pancake detector. Active particles were arbitrarily defined as anything over 0.7 kilo counts per minute (kcpm) through the plastic bag. This number of counts was about ten times higher than background levels. A circle was drawn on the bag with a marker to indicate the locations of each of the more radioactive locations. For the purpose of this investigation, we assumed that each radioactive location represented a discrete particle. This assumption was based on previous work at this site where particles were similarly identified but then removed, isolated and found to be single particles. Certainly 2 or more particles having an aggregate radioactivity greater than 0.7 kcpm could have been enumerated as a single particle.

3.1 Bulk sediment contamination

Bulk sediment contamination was measured at 20 to 25 random locations on each of the plastic bags of radioactive sediment. We used these measurements to compare bulk sediment contamination of each dredge sample to the radioactive particle contamination of each dredge sample. We examined location to location variations using the sums of bulk and particle-associated radioactivity in each dredge.

4. RESULTS

4.1 Grain size distribution

Grain size distribution of 3 samples of river sediment within 150 m of the Outfall is shown in Figure 3. The sandy nature of the sediments near the Outfall had been noted qualitatively during previous work, so it came as no surprise that the grain size analysis presented here would indicate the same thing. The sand content of the riverbed near the Outfall showed that sand-sized particles had settled near the source (Figure 3). Even 30 m upriver, the sand content of the sediment was 24%, and this is 3 to 5 times the natural percentage of sand at those depths in that area. As the usual sand content at these depths and current regimes is 5 to 10%, the observed increase in sand upriver was probably due to sand in the Process Outfall. Work by Merritt [4] had shown that the soluble tracer did not contact the riverbed until at least 440 m of downriver distance. Merritt also showed that upriver winds were capable of driving surface water from turbulent swirl more than 100 m upriver before moving downriver. Therefore, we attributed the presence of abnormal amounts of sand somewhat upriver and downriver from the Outfall to the settlement of sand originating in the outfall discharge.

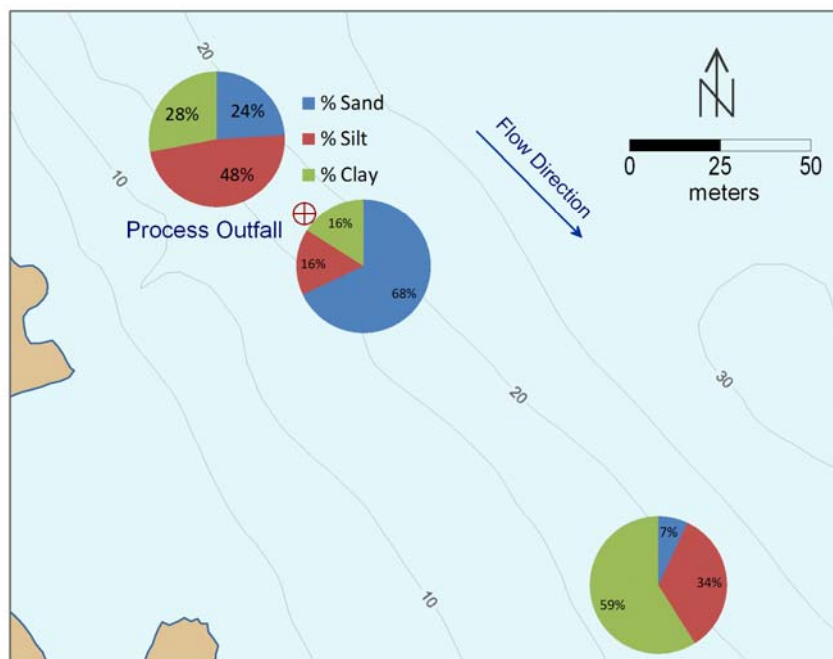


Figure 3. Sand, silt and clay in surficial river sediment along the 20-m depth contour near the Process Outfall. Each pie chart was plotted with its centre at the location of the sample.

4.2 Bulk sediment radioactivity

Bulk sediment readings, taken randomly on the bags of sediment, showed less dredge to dredge variation than that due to the radioactivity associated with particles (Figure 4).

Bulk sediment radioactivity measurements tended to parallel the particulate radioactivity among the dredge samples, but the readings of bulk sediment radioactivity were lower and less variable from sample to sample than the measurements of particle-associated radioactivity.

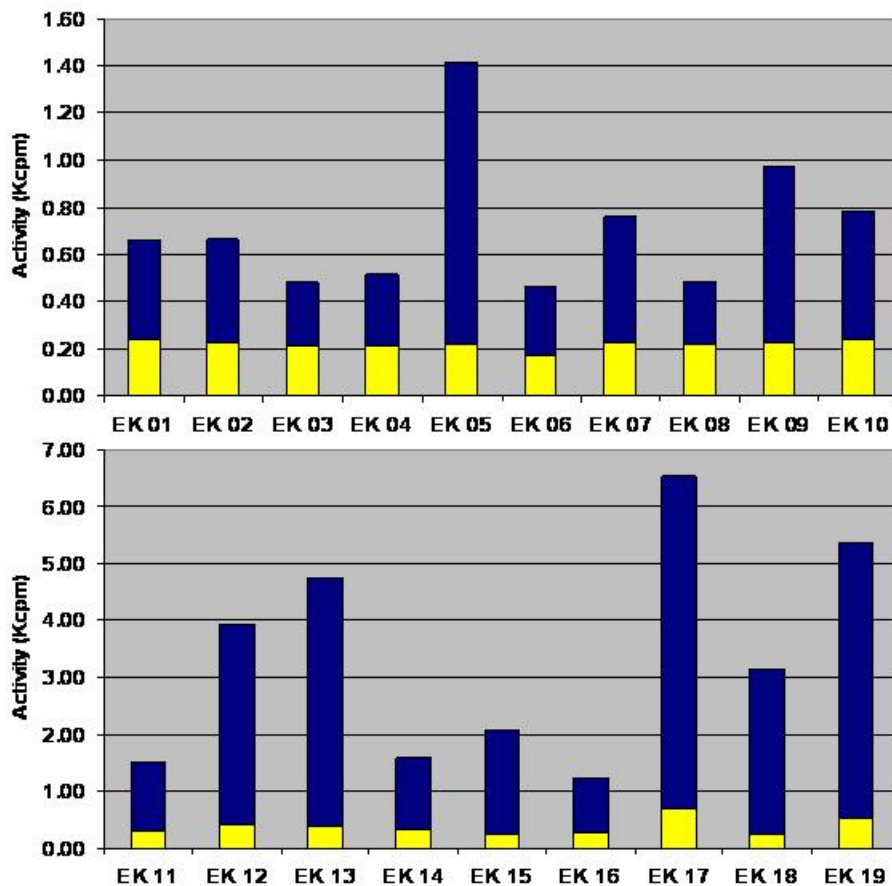


Figure 4. Contamination meter readings of radioactivity (in kcpm) in individual Ekman dredge samples. Dredge samples EK01 to EK10 (top graph) were taken 65 to 110 m downriver of the Outfall. Samples EK11 to EK19 (bottom graph) were taken within 40 m downriver of the Outfall. Blue bars show the total contamination (in kcpm) due to discrete active particles, as measured through the 1-cm thick bags of sediment. Yellow bars represent the average bulk sediment contamination meter readings, which were taken at 20 – 25 random locations on the 1-cm thick bags of sediment.

4.3 Particle radioactivity

To investigate the depth distribution of radioactive particles within 110 m of the Outfall, we obtained 23 Ekman dredge samples at 3 locations (G1, G2 and G3 - Figure 2). G1, was comprised of 4 samples and the total area of riverbed sampled at that location was 0.21 m². G2 was comprised of 8 samples and the total area sampled was 0.42 m². These samples were collected within 40 m downriver of the Outfall. G3 was comprised of 11 dredge samples and the total area sampled was 0.58 m². Group 3 samples were taken between 65 and 110 m downriver of the Outfall.

The results at G2 were from 8 Ekman dredge samples. The 0 – 1 cm layers were not scanned or the readings were lost for dredges EK-11 and EK-12 in G2. In a few other dredge samples, the dredge did not penetrate to 11 cm and the depths were not scanned. The 2 - 8 cm depth intervals were from 8 Ekman dredge samples. The 8 – 9 cm depth intervals were from 7 Ekman dredge samples. The 9 – 10 cm depth interval was derived from 4 Ekman dredge samples. The > 10 cm interval was derived from only 1 Ekman dredge sample in G2. We considered the above to have no effect on the overall value of the results.

Because the number of samples differed among groups, results are presented in terms of average numbers of particles and as the average particulate radioactivity per Ekman dredge sample (Figure 4).

Table 1 provides a summary of the lateral (group to group) variation in the number and activity of radioactive particles, per square metre of riverbed.

Ninety-six percent of the radioactive particles were found in the upper 10 cm of sediment. The number of active particles and the radioactivity of those particles decreased with increasing distance from the Outfall. The pattern was similar to that of the decreasing sand content with distance from the Outfall (Figure 3).

G1 samples (Figure 5) showed little variation in the number of radioactive particles or total particulate radioactivity with depth; the profiles were surprisingly uniform. This could have resulted from the small area (only 4 dredges) sampled in G1. G2 contained the largest number of particles and more than 5 times the radioactivity as compared with that found in G1 and G3 (note horizontal scale in Figure 6). In both G2 and G3 areas, the majority of active particles was found 2 to 7 cm below the surface of the sediment.

Within 60 m of the Outfall, the peak radioactivity was observed 3 cm below the sediment surface (Figure 6, G2). In G3 samples, between 70 and 110 m below the Outfall, the peak radioactivity was 4 to 6 cm below the sediment surface (Figure 7, G3). In both groups (Figure 6 and Figure 7), we observed a stepwise decline in particulate contamination, from peak values from midway in the profile, to the surface. Thus, the sediment record showed a temporal decrease in the accumulation of particulate radioactivity.

Using the vertical distribution as an indicator of sediment deposited overtime, the upward decline in particulate radioactivity occurred in the past 2 or 3 decades. This pattern was not observed in samples above the Outfall (Figure 5), perhaps because only 4 dredge samples were collected there.

There was a marked decrease from G2, near the Outfall, to G3 farther downriver, but the lateral variations between dredge samples was larger terms of the particulate activity. This was due to the large variations in particle.

Table 1. Lateral variation in numbers and activities of radioactive particles per square metre of riverbed at 3 locations. G1 was within 30 m but slightly upriver of the Outfall. G2 was within 40 m of the Outfall and G3 samples were collected between 65 and 110 m below the Outfall (Figure 2).

	Location		
	G1	G2	G3
Total Number of Active Particles/m ²	374	431	181
Total Particle Activity/m ² (kcpm)	975	8,853	1,623

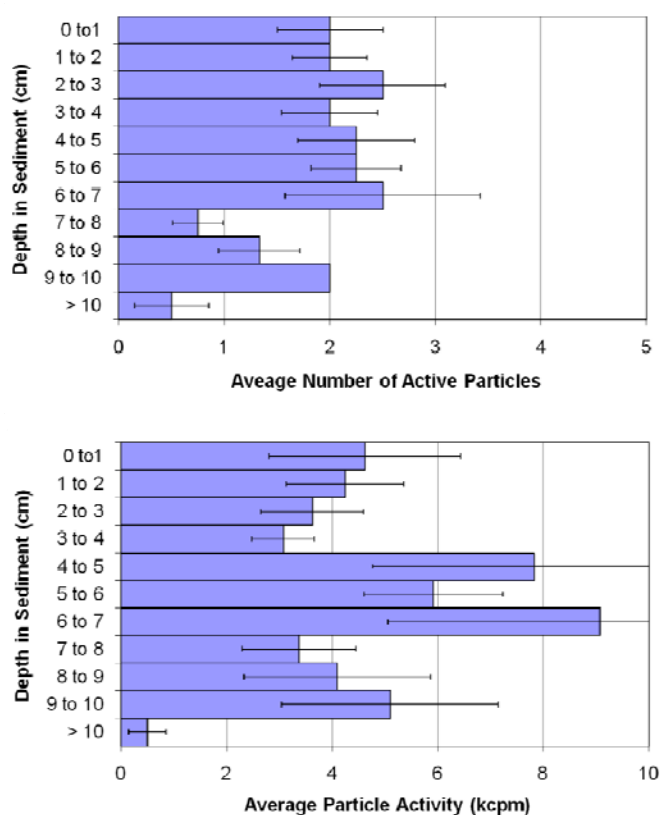


Figure 5. Radioactivity found as discrete particles (i.e. >0.7 kcpm) in G1. The bars and lines show mean and standard deviation values for all dredge samples in G1. These samples were taken just above the outfall. The 0 – 8 cm depth interval data were from 4 Ekman dredge samples. The 8 – 9 cm depth intervals were from only 3 Ekman dredge samples. The 9 – 10 cm and greater than 10 cm depth intervals were from only 2 Ekman dredge samples.

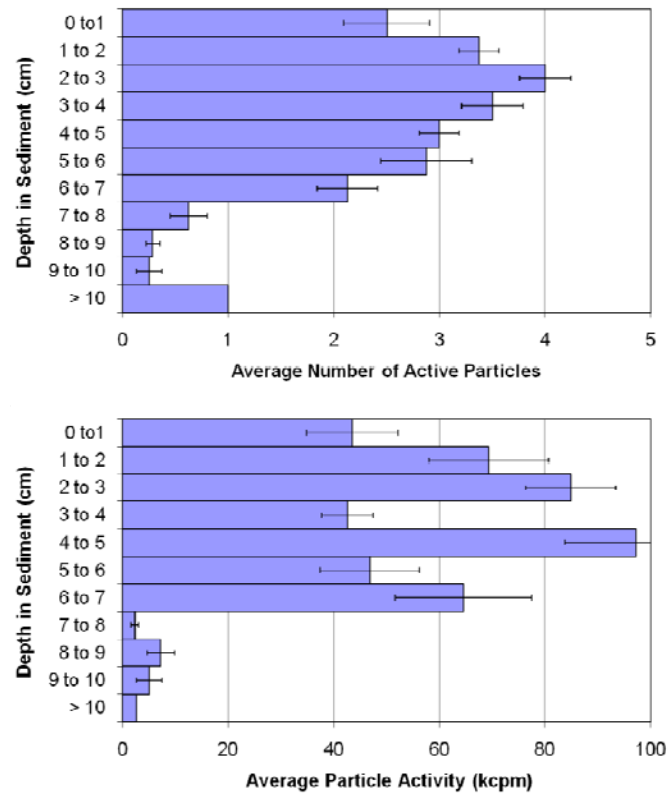


Figure 6. Radioactivity found as discrete particles (i.e. >0.7 kcpm) in G2. The bars and lines show mean and standard deviation values for all dredge samples in G2. These samples were taken from just below the Outfall.

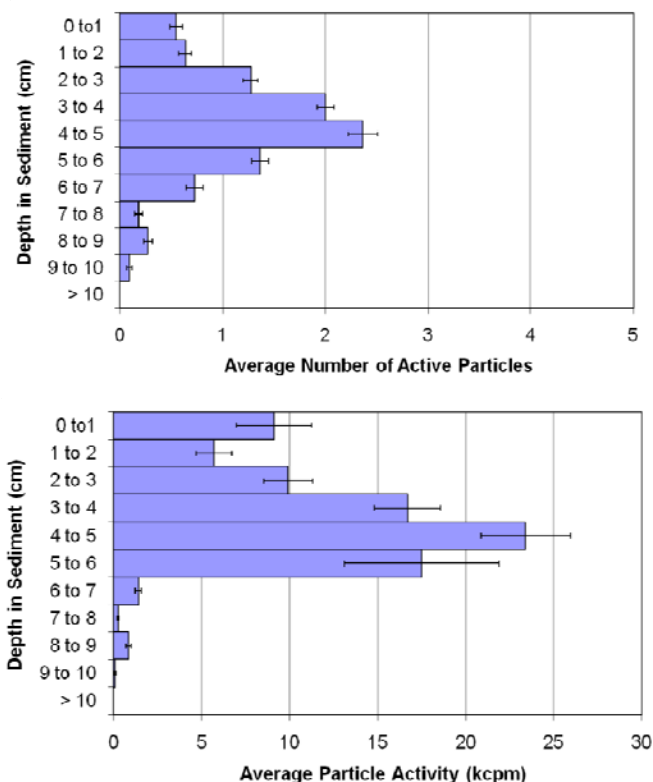


Figure 7. Radioactivity found as discrete particles (i.e. >0.7 kcpm) in G3. The bars and lines show mean and standard deviation values for all dredge samples in G3. These samples were taken from 65 to 110 m below the Outfall. The 0 – 1 cm depth interval was derived from 11 Ekman dredge samples. The greater than 10 cm interval was based on sediment from 6 Ekman dredge samples.

4.4 Lateral variation in radioactivity associated with particle in riverbed sediment

With increasing horizontal distance from the Outfall, there was a substantial decrease in the active particle content and particle-associated radioactivity of the sediments in the area of riverbed investigated by dissection of dredge samples. In the immediate vicinity of the Outfall (Figure 2 and Table 1, G2), the total number of active particles was about 400 per m² and this dropped to less than 200 per m² within 110 m downriver (Figure 2 and Table 1, G3). The total particle-associated radioactivity near the Outfall was about 8.9×10^6 cpm per m² and this dropped to 1.6×10^6 cpm per m² within 110 m downriver (Figure 2 and Table 1).

5. DISCUSSION

The grain size results of the sediment samples clearly shows that the Outfall has been a major source of sand to the riverbed at these depths. Because of particle settling and slow downriver current, the sand content decreases with distance from the Outfall, just as the particulate radioactivity was observed to decrease with distance from the Outfall (Figure 6 and Figure 7).

We have noticed qualitatively that sand from the land blows along the surface of the ice in winter, becomes entrained in the ice and snow and sinks to the riverbed as the ice melts. This process probably accounts for the minor amounts (<10%) of sand in the deepwater sediments of the river in general, but it would not be reasonable to attribute the elevated amounts of sand near the Outfall to aeolian or ice-blown sand. Almost certainly it is due to sand in the Process Outflow.

Near-background levels of particulate radioactivity were observed deeper than 9 cm into the riverbed. In fact, the majority of the particles were found above the 6-cm depth. Particles such as these do not diffuse and therefore, they may act as tracers that can be used to estimate sedimentation rates. Certainly the localized nature of the particulate radioactivity suggests that downriver transport has not been a significant process in the years since the labs began operation, and presumably began releasing radioactivity. If, for the sake of discussion, the depth of 9 cm corresponds to the year 1952, which is when the present Outfall pipe was built, then 90 mm of sediment formed in 53 years, giving an average rate of 1.7 mm/yr. The expected sedimentation rate at this site, based on the 8,300 year average was about 0.5 mm/yr (see Section 2). The higher rate of sedimentation in recent years is almost certainly the result of sand being discharged from and settling near the Outfall. Immediately below the Outfall (Figure 6), the average particle activity peaked at a depth of 4 to 5 cm in the sediment. The decrease in radioactivity near the top of the sediment column indicated decreases in both the number of radioactive particles and the total radioactivity in the last few decades.

Sediment disturbances might explain the presence of a few particles below the 8 cm depth interval. These include sampling disturbances or artifacts, repetitive core and dredge sampling of the sediment, dragging of probes laterally through the sediment and bioturbation. Sampling artifacts during sectioning of the dredge samples as described in the methods section resulted in 30% of the particles being found 1 cm above or below the layer in which they were initially located. Therefore, it was possible that particles actually situated in the 3 - 4 cm layer, for example, were included in the 2-3 or 4-5 cm layer. Two towed probe lines, dragged across the riverbed in the 5,000 m² area of investigation, would have mixed the sediment to a depth of 4 cm over a 20 m² area, but would not have compromised the sediment profile. Repetitive anchorage by non-AECL people and AECL coring would have created some sediment disturbances. Finally, the larvae of the mayfly Hexagenia live within 40 m or so of the Outfall, where the sediments apparently are not excessively sandy. Although their numbers are low, these larvae create 3- to 5-cm deep U-shaped burrows and cause bioturbation that would move particles from layers where they had initially settled.

The NRX reactor operated from 1947 to 1992. It was cooled directly by river water and thus any release of failed fuel and corrosion products could have found their way to the river. Thus, the NRX timeline coincides with the presence of the majority of the particulate radioactivity 4 to 6 cm below the sediment surface and a substantial decrease in the number of particles in the upper 3 cm of sediment.

6. SUMMARY AND CONCLUSIONS

An Ekman dredge was modified so it could be sampled from the top in centimetre thick slices for this investigation of particulate radioactivity in the sediment profile. A total of 23 dredge samples were retrieved within the 5,000 m² area of greatest riverbed radioactivity within 110 m of the Process Outfall. The samples were collected along the central axis of an ellipsoid that

outlined the “footprint” of the above-background radioactive sediment. Each centimetre thick layer from each Ekman dredge sample was placed in a plastic bag, flattened and scanned with a contamination meter. In order to collect sufficient “hits” in this study, radioactive particles were defined as any location-measurement having contamination-meter readings greater than 0.7 kcpm.

Dissection of the riverbed sediment at 1-cm intervals and contamination meter scans for radioactive particles showed that discrete particles of radioactivity probably account for the majority of the above-background radioactivity below CRL.

The grain size analysis of the sediment showed that the Process Outfall has been a source of sand to the riverbed, increasing the sand content by a factor of about 10 within a 25 m radius of the discharge. Because of particle settling and slow downriver current, the sand content of the sediment decreased to near normal amounts within 110 m downriver from the Outfall, just as the particulate radioactivity was observed to decrease with distance.

Results showed that, in addition to sand, sand-sized fragments of radioactive material have also accumulated near the effluent Outfall in 17 to 23 m of water. The accumulation has been greatest near the Outfall and decreased significantly within 110 m downriver.

Most of the active particles were found 20 to 70 mm below the sediment surface and virtually none were found deeper than 90 mm into the riverbed. When a radioactive particle was defined as any location on a sediment sample producing more than 0.7 kcpm, the number of radioactive particles per square metre of riverbed was $431/\text{m}^2$ within 60 m downriver of the Outfall. Such particles numbered $181/\text{m}^2$ at locations between 65 and 110 m downriver of the Outfall.

Based on a preliminary assumption that the sub-bottom acoustic profiling revealed the thickness of Holocene sediment, accumulated since the last catastrophic flood, the natural, long-term average sedimentation rate at the footprint location has been on the order of 0.5 mm/yr. Based on the fact that the laboratories started up in 1944 and the assumption that radioactive and non-radioactive particulate sedimentation began about the same time, the sedimentation rate near the Outfall may have been as high as 1.7 mm/yr in the past 6 decades. If the sedimentation rate is, in fact, in the range of 1 to 2 mm/yr, then the majority of the particles were probably emitted several decades ago. The steady rise and fall of particle-associated radioactivity in the sediment profile was consistent with a sedimentary environment that is relatively stable in terms of downriver sediment movement.

Our analysis of the vertical distribution of sand-sized fragments of particulate radioactivity suggests that the releases likely occurred primarily during the time of operation of the NRX reactor, 1947 to 1992, and have been declining ever since.

7. ACKNOWLEDGEMENTS

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8. REFERENCES

1. Lee, D. R., D. S. Hartwig. "Analysis of riverbed sediments near the Chalk River Laboratories. Extended Abstract", In: Proceedings of the 2nd International Symposium on Contaminated Sediments: Characterization, Evaluation, Mitigation/Restoration, Management and Performance. Quebec City, Quebec, May 26 - 28, 2003, Sponsored by: American Society of Testing and Materials, Canadian Geotechnical Society, Canadian Society of Civil Engineering, Chapitre St. Laurent SRA-SETAC and Environment Canada.
2. Lee, D. R., R. Rao, W.H. Hocking, D.S. Hartwig, "Particulate radioactivity in Ottawa Riverbed sediments near the Chalk River Laboratories", Abstract. In: Proceedings 4th International Conference on Remediation of Contaminated Sediments, January 22-25, 2007 Savannah, GA, USA. Sponsored by Battelle.
3. Ophel, I.L., "Monitoring of fresh waters used for dispersal of radioactive wastes in Atomic Energy of Canada Limited", Papers presented by AECL to the IAEA Conference on the Disposal of Radioactive Wastes, Monaco, 1959, AECL-987, p.324-327.
4. Merritt, W.F., "Studies of dilution in the Ottawa River using rhodamine B-2. CRNL to Pembroke", Atomic Energy of Canada Limited, Chalk River, Ontario, 1964, AECL-2030.
5. Catto, N.R.; Patterson, R.J. and W.A. Gorman. "The late Quaternary geology of the Chalk River region, Ontario and Quebec", *Canadian Journal of Earth Sciences*, 1982, 19: 1218-1231.
6. Lewis, C. F. M., T. W. Anderson, "Oscillations of levels and cool phases of the Laurentian Great Lakes caused by inflows from glacial Lakes Agassiz and Barlow-Objibway", *J. Paleolimnol.*, 2, 99-145, 1989.
7. Shilts, W., Personal communication, Shilts was with the Geological Survey of Canada at the time of his work at CRL in 1984 and 1985. He later took a position as director of the Illinois Geological Survey, Champaign IL, USA.
8. Lee, D.R, C. Jones, "Hydrodynamic Model Predictions Regarding Downriver Transport of Radioactive Sediment". Paper and abstract presented at 6th International Conference on Remediation of Contaminated Sediment, New Orleans, LA Feb. 7-10, 2011. http://www.battelle.org/conferences/sediments/pdfs/preliminaryprogram_techpresentations.pdf
9. ASTM, D 422 - 63 (2007) Standard Test Method for Particle-Size Analysis of Soils.
10. Lee, D. R. and D. S. Hartwig. 2011. In Preparation: In Situ Method for Mapping Above-background Radiation: Sediments of the Ottawa River Near the Chalk River Laboratories.