

The importance of the sediment pathway in the radionuclide dose received by aquatic non-human biota: Reconstruction and mapping of spatiotemporal partitioning in Perch lake sediments over a 40-year period

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Abstract. There is a growing interest in assessing risk to non-human biota following exposure to radionuclides. However, before dose to aquatic biota can be determined, it is critical to have a solid understanding of radionuclide concentrations in key environmental phases due to their importance in determining external and internal dose. Further understanding of a system can be reached by monitoring temporal changes in radionuclide levels in these phases, especially if assessing the success of remediation activities. The current study has focused on estimating inputs of ^{90}Sr , ^{137}Cs and ^{60}Co to the sediments of a small, Canadian Shield lake located downstream of two Waste Management Areas at AECL's, Chalk River Laboratories site over a 40-year period. Overall, it was found that ^{137}Cs and ^{60}Co inputs have declined in the lake over the last 40 years, which resulted in a net depletion of these radionuclides from the sediments over time. Strontium-90 inputs have remained fairly constant over this time period with ^{90}Sr retention in lake sediments of approximately 15%. It is expected that benthic biota will receive approximately a 2- to 6-fold higher radionuclide dose than pelagic organisms, on average, particularly when feeding in the depositional zone of the lake.

1. INTRODUCTION

Sediments often represent an important reservoir for contaminants, such as radionuclides, in aquatic systems [1-4]. Consequently, lake sediments can potentially be significant contributors to the total radionuclide dose received by non-human biota that are in contact with them. The proportion of sediment-related dose received by biota relative to the dose received through other aquatic pathways is dependent upon several factors, including net radionuclide inputs to a system through time, the tendency of each radionuclide to partition into the sediments relative to other environmental compartments, the spatial distribution of radionuclides in the sediments, and the behaviour or life-style of the biota inhabiting the lake, which can affect their overall contact with the sediments [e.g. 2, 4-7]. With increased understanding of these parameters and their interactions, it becomes possible to improve estimates of the radionuclide dose received by aquatic non-human biota through the sediment pathway.

The current study focuses on radionuclide transfer and spatial partitioning patterns in the sediments of Perch Lake (Chalk River, Ontario), a small, dystrophic-eutrophic Canadian Shield lake which has received ^{90}Sr , ^{137}Cs , ^{60}Co and ^3H inputs from two upstream Waste Management Areas (WMAs), as well as through atmospheric deposition due to global weapons testing. Environmental monitoring data for radionuclides in Perch Lake surface waters have been compiled for each radionuclide, and subsequent transfer to sediments has been estimated over a 40-year period using a mass balance modeling approach. The study objectives were:

1. To estimate ^{90}Sr , ^{137}Cs and ^{60}Co transfer to Perch Lake sediments over the past 40 years;
2. To approximate the ranges and spatial partitioning of these radionuclides in the lake sediments; and
3. To relate spatiotemporal trends in radionuclide transfer and partitioning to the radionuclide dose received by resident non-human biota through the sediment pathway.

2. DATA COLLECTION AND TREATMENT

2.1 Mass Balance Models: Approaches and Assumptions

Radionuclide inputs to Perch Lake sediments were assessed over a 40-year period using mass balance models and subsequently validated using measured data. For ^{90}Sr , it was possible to estimate inputs to lake sediments by subtracting radionuclide losses from the inputs to the lake, since this radionuclide was detectable at lake inflows and outflows throughout the 40-year study period. Cesium-137 and ^{60}Co concentrations were not typically detectable in Perch Lake surface outflows after the early 1980s and early 1990s, respectively [4]. Consequently, it was not possible to compare inputs versus outputs for these radionuclides to estimate their contribution to Perch Lake sediments, making it necessary to estimate inputs to lake sediments based on source term data.

Radionuclides can enter the sediments through deposition from the water column or through ingrowth from radioactive precursors [2, 8-9]. In the case of the radionuclides under consideration in Perch Lake, the ingrowth term is equal to zero [10]. Although some radionuclides, such as ^3H and possibly ^{60}Co , can enter the lake via the groundwater, it is assumed that there is negligible sorption of these radionuclides to the sediments and that any groundwater inputs directly enter Perch Lake surface water. As a result, groundwater inputs to the lake are considered in the mass balance model for Perch Lake surface water, which later contributes to the sediments through radionuclide deposition. Based on these assumptions, the radionuclide mass balance for Perch Lake sediments can be described by the solution of the differential equation:

$$\frac{dM_{sed}^i(t)}{dt} = \alpha_i \cdot M_1^i(t) + \lambda^{i-1} \cdot M_{sed}^{i-1}(t) - \lambda^i \cdot M_{sed}^i(t) \quad \text{Eq. 1}$$

The first term of Equation 1 represents the inputs into the sediments from the overlying water, where α_i is the water-to-sediment transfer rate for a given radionuclide (a^{-1}) and $M_1^i(t)$ is the amount of radionuclide in the overlying water at time t (Bq). The water-to-sediment transfer rates for the radionuclides that are present in Perch Lake and their stable analogues have been taken from Bird et al. [9]. Temporal trends in radionuclide concentrations in Perch Lake surface waters have been provided in Yankovich et al. [4]. The second term in the equation represents inputs into the sediments through radioactive decay of precursors, where λ^{i-1} is the radioactive decay constant for radioactive precursors and $M_{sed}^{i-1}(t)$ is the amount of precursors in Perch Lake sediments. Again, this term is equal to zero for these radionuclides. The third term represents a loss term due to radioactive decay. Radioactive decay constants for the radionuclides found in Perch Lake have been summarized in Friedlander et al. [10]. It is assumed that there are no other radionuclide losses from Perch Lake sediments. Using this approach, net annual and cumulative ^{137}Cs and ^{60}Co inputs to sediments have been estimated over the past four decades.

2.2 Spatial Partitioning of Radionuclides in Perch Lake Sediments

Following the estimation of total radionuclide inputs to Perch Lake sediments, the spatial partitioning of radionuclides in the lake was mapped out. To do this, bulk sediments and core samples were collected at 20 locations in the Perch Lake basin in February 1997. Coring locations were chosen on the basis of sediment type, water depth, proximity to radionuclide point sources in the lake, locations of the depositional, transitional and erosional zones, and locations of historical sampling sites in the lake to facilitate comparisons between current radionuclide activities in the sediments and those measured in the past [5, 11]. Sediment sampling was conducted when the lake was ice-covered to ensure minimal drift of the coring equipment as it was lowered to the sediment surface, so that each sampling coordinate was as accurate as possible. Detailed technical information on sediment sampling is provided in Yankovich et al. [11].

Radionuclide concentrations were then measured in archived Perch Lake sediment samples that were collected using the same method in 1982 to allow comparison of radionuclide levels in lake sediments between these periods.

3. RESULTS AND DISCUSSION

Peak radionuclide concentrations occurred in Perch Lake between the early 1960s and the mid-1970s to early 1980s due to atmospheric deposition from weapons testing and/or inputs from upstream WMAs [4]. Strontium-90 predominantly entered Perch Lake through two inflowing streams at lake Inlets 1 and 2, which drain WMAs B and A, respectively (Figure 1) [4, 5]. Between the early 1960s and the early 1980s, the Inlet 2 surface stream represented the primary source of ^{90}Sr to the lake, after which relatively higher amounts entered the lake through Inlet 1, with a slight increase in ^{90}Sr influx to the lake after 1985 [5]. Despite this slight increase in ^{90}Sr inputs to the lake, ^{90}Sr concentrations at the lake outflow have remained relatively constant over most of this period, suggesting a net accumulation of ^{90}Sr in the lake (Figure 2). Comparison

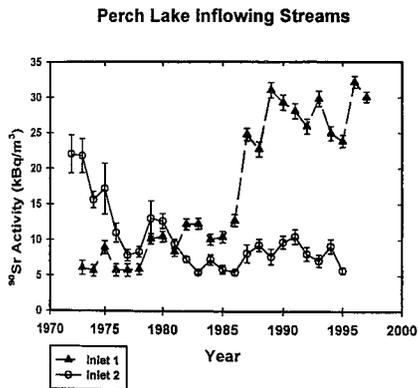


Figure 1: ^{90}Sr activity in inflowing surface water streams.

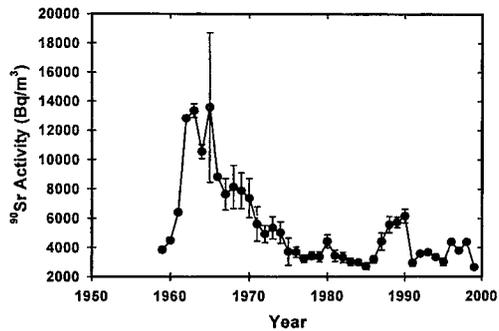


Figure 2: Temporal trends in ^{90}Sr activities in surface waters at the Perch Lake outflow.

of ^{90}Sr inputs to and outputs from Perch Lake indicates that approximately 15% of the inputs are retained by lake sediments (Figure 3). This concurs with ^{90}Sr retention values of 15% in Perch Lake, which were reported by Ophel et al. [3] in 1972. As a result, it is expected that ^{90}Sr inputs to Perch Lake sediments have been increasing over time (Figure 4). Field measurements of the ^{90}Sr inventory in Perch Lake sediments in 1982 relative to 1997 have been compared to confirm this (Figures 5 and 6). As expected, the ^{90}Sr inventory has increased between 1982 and 1997 (Table 1). Modeled inventory values were approximately equal to expected values that were estimated using available data on ^{90}Sr influx and loss from the system (Table 1).

Unlike ^{90}Sr , ^{137}Cs and ^{60}Co levels have been declining in Perch Lake sediments over time due to reductions in the source terms for these radionuclides, in addition to their losses from the sediments through radioactive decay (Table 1; Figures 7-10). Examination of ^{137}Cs and ^{60}Co sediment inventories in 1982 and 1997 indicates that measured values fall within 1.8- and 1.1-fold of expected values, respectively (Table 1). Losses of these radionuclides from lake sediments correspond to losses due to radioactive decay.

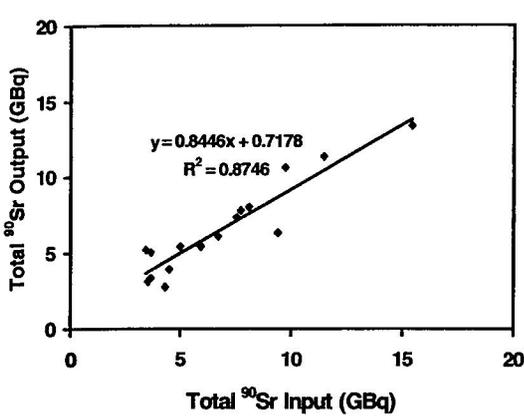


Figure 3: Relative ⁹⁰Sr inputs to and outputs from Perch Lake surface waters.

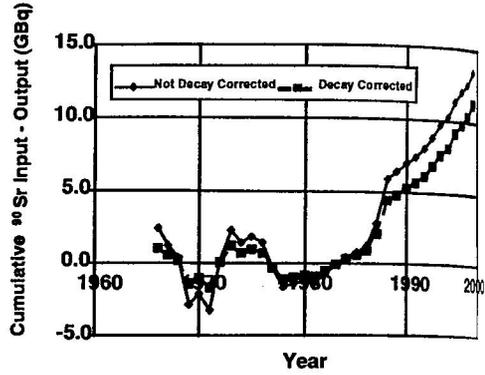


Figure 4: Cumulative ⁹⁰Sr inputs to and outputs from Perch Lake sediments.

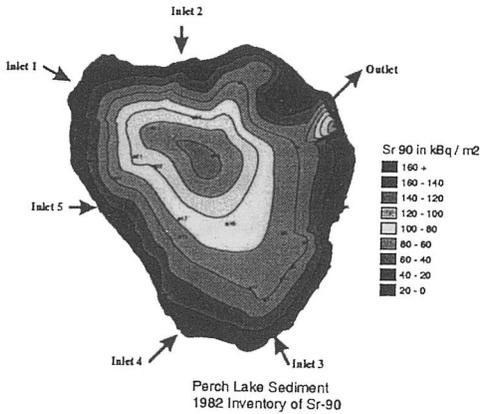


Figure 5: Spatial distribution of ⁹⁰Sr in Perch Lake sediments based on measurements made in 1982 [11].

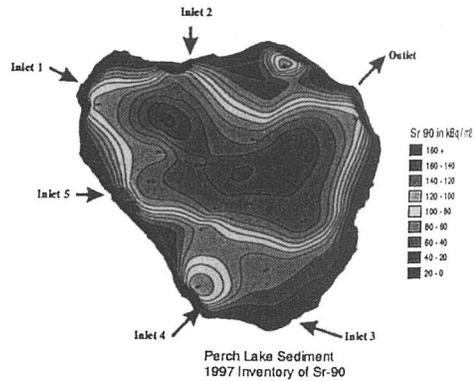


Figure 6: Spatial distribution of ⁹⁰Sr in Perch Lake sediments based on measurements made in 1997 [11].

Table 1: Comparison of measured and expected radionuclide inventories in Perch Lake sediments between 1982 and 1997.

Radionuclide	Measured Inventory (GBq)		Expected Inventory (GBq) 1997	Expected-to-Measured Inventory Ratio (1997)
	1982	1997		
⁹⁰ Sr	25.2	42.5	37.3	0.88
¹³⁷ Cs	2.8	1.1	1.98	1.8
⁶⁰ Co	6.6	3.3	1.1	1.1

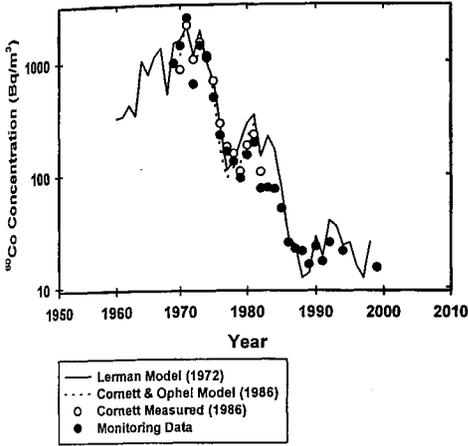


Figure 7: Temporal trends in ⁶⁰Co concentrations in Perch Lake surface waters [4].

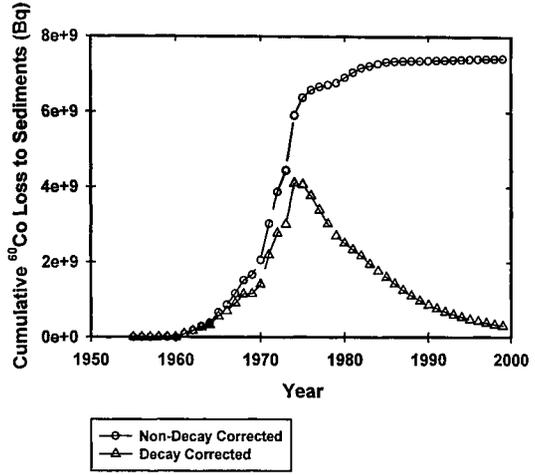


Figure 8: Temporal trends in ⁶⁰Co inputs to Perch Lake sediments.

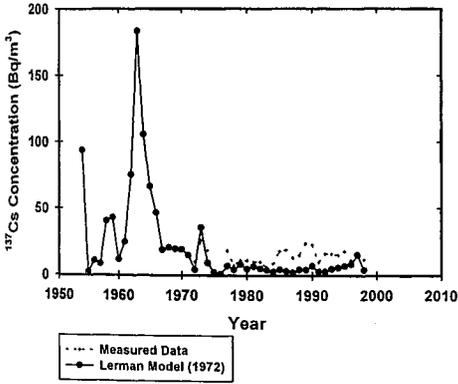


Figure 9: Temporal trends in ¹³⁷Cs concentrations in Perch Lake surface waters [4].

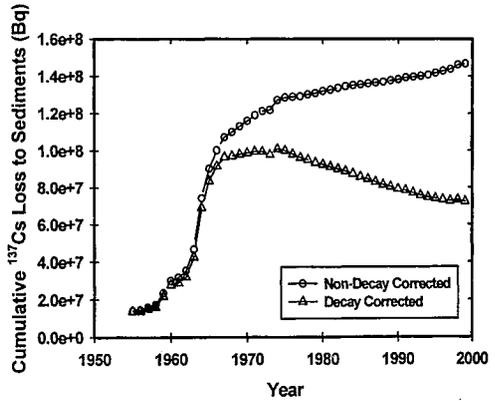


Figure 10: Temporal trends in ¹³⁷Cs inputs to Perch Lake sediments.

Exposure of biota to sediment-bound radionuclides is influenced by the spatial partitioning of radionuclides in the lake sediments relative to habitat-use by resident biota [5]. To account for this in dose calculations, aquatic non-human biota can be sub-divided into several functional groups based on their habitat-use and feeding patterns. These include littoral biota (which occupy the shallow, weedy regions along the shoreline periphery), pelagic biota (which occupy the offshore areas of the lake with limited exposure to sediments), and benthic biota (which feed in lake sediments) [5]. Since these organisms occupy different areas of the lake, a key question becomes 'how uniformly are radionuclides distributed in lake sediments in these different areas?' Once added to the lake, radionuclides do not tend to partition uniformly across the sediment surface [12]. Instead, radionuclides can migrate both horizontally and vertically in the sediments as a

different areas?' Once added to the lake, radionuclides do not tend to partition uniformly across the sediment surface [12]. Instead, radionuclides can migrate both horizontally and vertically in the sediments as a function of factors, such as sediment type, composition, hydraulic conductivity and radionuclide binding affinities. Therefore, to relate the amount of radionuclides that enter the sediments to their concentrations in the sediments, information on the spatial distribution and vertical profile of radionuclides in the sediments is required. For the purposes of this study, a 20 cm vertical depth of sediments was assumed to represent a homogeneously-mixed layer. This depth corresponds to sampling protocols used to collect archived and current bulk sediment samples. This appeared to be a reasonable approach, since measured and expected values for the radionuclide inventory in the sediments corresponded closely, as discussed above (Table 1). In terms of the horizontal distribution of radionuclides in the sediments, it was concluded that Perch Lake could be sub-divided into two zones: the erosional zone representing the shallow littoral waters along the lake shoreline (depth < 1.5 m), and the depositional zone in the offshore, pelagial areas of the lake (depth > 1.5 m). The littoral zone of Perch Lake consists of sandy and organic gyttja sediment types, whereas the pelagial zone contains only gyttja. In Perch Lake, organic gyttja covers approximately 75% of the lake surface area, with sand representing the remaining 25% of the lake. For both ^{90}Sr and ^{137}Cs , geometric mean concentrations in organic gyttja from the littoral and pelagial zones varied by only a factor of 2, whereas ^{60}Co was fairly uniformly distributed throughout the lake varying by 1.3-fold in gyttja between lake zones [5]. This suggests that biota feeding in organic sediments are receiving a similar dose (within a factor of 2, on average), regardless of where they are found in the lake. The largest differences in sediment radionuclide concentrations were observed between the two different sediment types. On average, sandy sediments were approximately 3-, 4- and 6-fold lower than organic gyttja in terms of their concentrations of ^{90}Sr , ^{60}Co and ^{137}Cs , respectively [5]. Therefore, organisms that occupy organic gyttja are expected to receive a slightly higher sediment dose than those species that tend to be found in the sand.

In summary, the spatial distribution of radionuclides in Perch Lake sediments is relatively uniform throughout the lake, although slight differences do occur between sandy and organic sediment types. Sediments potentially represent a key exposure for aquatic biota that are regularly in contact with them. In Perch Lake, the sediments serve as a sink for key radionuclides in the lake, with radionuclide levels that track inputs to the lake. Therefore, organisms, such as benthic species, which feed in the sediments and likely ingest particulates during feeding, are expected to receive relatively higher doses than pelagic species that have limited contact with the sediment phase. Further work is being conducted to examine sediment-biota interactions and their effects on dose to biota in aquatic ecosystems in more detail.

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