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THE IMPORTANCE OF ENVIRONMENTAL MONITORING DATA IN ENVIRONMENTAL RISK ASSESSMENT: AN ECOSYSTEM APPROACH

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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 surface waters 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 the water, especially if assessing the success of remediation activities. The current study has focused on estimating concentrations of ^{3}H, ^{90}Sr, ^{137}Cs and ^{60}Co in 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 radionuclide activities have declined in the lake over the last 40 years, particularly for ^{137}Cs and ^{60}Co, which are much lower than they had been in the past. Tritium and ^{90}Sr are measurable in the lake water and water represents a key route of exposure for these radionuclides.

INTRODUCTION

Water can represent a major route of contaminant exposure for aquatic non-human biota. In the case of radionuclides, concentrations in the water contribute both directly towards external dose, as well as indirectly towards internal dose to aquatic organisms. The external dose from water is dictated by: the proportion of time a given type of organism spends in contact with or immersed in the water relative to other environmental compartments; the concentration of radionuclides in the water (which is proportional to exposure); and, the relationship between exposure concentrations and the dose received by the organism (i.e. the dose conversion factor, DCF) [1-7].

Radionuclides are usually carried into an aquatic system via surface water, groundwater, precipitation, and/or dry deposition of particulates [4, 8-10]. The concentration of radionuclides in the water is influenced by the flux of radionuclides entering the system via these routes relative to total loss from the system by surface water, groundwater, and/or evaporation, the tendency of each radionuclide to partition into the water relative to other environmental phases, and the physico-chemical properties of the basin [8, 9, 11-15]. The total external dose to biota, which represents the sum of external dose from all radionuclides present in the system, is proportional to exposure as described by the DCF, and is influenced by the type of habitat typically utilized by organisms in the system.

The habitat-use factor is a parameter that represents the relative amount of time an organism spends in a given type of habitat or "environmental compartment" [6, 7, 16]. This parameter reflects the "life-style" and behaviour of the organism, and the types of habitats occupied by the organism. It has an influence on exposure and subsequent dose to non-human biota because each compartment has a characteristic tendency or capacity to concentrate or store radionuclides to which resident biota are exposed to varying degrees depending on their life-style and behaviour. For aquatic organisms, water, sediments and aquatic vegetation often represent key compartments where individuals spend their time [6, 7, 16]. Therefore, the relative amount of time spent in each of these types of habitat, as well as the radionuclide concentrations in these compartments, will influence the external dose received by individual organisms.

Internal radionuclide dose-to-biota tends to be more pronounced than external dose [7]. Like external dose, internal dose is also influenced by radionuclide concentrations in surface waters, but in a more indirect way. In general, internal dose is driven by the propensity of an organism or species to accumulate radionuclides from the surrounding environment. Organisms accumulate contaminants, such as radionuclides, over time through uptake from water, food and particulates [e.g. 15, 17]. Radionuclides can partition from water into these environmental compartments creating the opportunity for radionuclide incorporation into individuals in a population. Temporal trends in radionuclide concentrations in water are important for determining internal dose to aquatic biota, since environmental radionuclides are integrated into the tissues of biota throughout their lifespans.

In order to assess risk to aquatic organisms following exposure to environmental radioactivity, it is important to accurately quantify radionuclide concentrations in the water to which aquatic biota have been exposed. Fortunately, in some cases, it is possible to utilize effluent monitoring data, which have been consistently collected over extended time periods at specific locations, to examine: i) site-specific, and/or ii) temporal trends in contaminant exposure to humans, as well as aquatic non-human biota.

To illustrate this approach, a study was conducted on temporal trends in the exposure of non-human biota to radionuclides in Perch Lake, a small, dystrophic, Canadian Shield lake at AECL's, Chalk River Laboratories site (Figure 1). Perch Lake has received ^{3}H, ^{90}Sr, ^{137}Cs and ^{60}Co input from two upstream Waste Management Areas (i.e. Areas A and B) and/or atmospheric deposition through radioactive fallout [18]. Radionuclides are removed from the lake through evaporation, losses to sedimentation and surface water outflow via Perch Creek [3, 8, 9, 18, 19]. By comparing the inputs and losses of radionuclides from the lake, it is possible to estimate the concentrations of radionuclides in the lake and how they have changed over time (Figure 2). Environmental monitoring data for radionuclides in surface water, which have been measured in inflow and outflow streams in the Perch Lake watershed, have been compiled over a 40-year period. These data were then used, in addition to environmental transfer rate constants, to estimate the radionuclide concentrations in Perch Lake. The study objectives were:

- to estimate the concentrations of ^{3}H, ^{90}Sr, ^{137}Cs and ^{60}Co in Perch Lake from concentrations measured in atmospheric deposition, as well as in the inflowing streams and downstream in Perch Creek over a 40-year period; and
- to relate temporal trends in radionuclide concentrations in Perch Lake water to exposure to aquatic biota inhabiting the lake.

DATA COLLECTION AND TREATMENT

Routine Monitoring of Radionuclide Influx into Perch Lake

Weekly surface water samples have been collected for total beta and tritium analyses since the early 1960s at Inlets 1 and 2, as well as downstream of Perch Lake in Perch Creek (Figure 1) [20-56]. Prior to 1962, water samples were collected intermittently in the Perch Lake watershed for total beta analysis [57]. Surface water was also collected at monthly intervals in the Perch Lake watershed for analysis of gamma-emitters, including ^{137}Cs and ^{60}Co.

Atmospheric deposition of radionuclides has been measured in nearby Deep River, Chalk River and Rolphton on a monthly basis since the early 1950 [58-60]. These data have been compiled to assess temporal trends in radionuclide input to Perch Lake via atmospheric deposition.

Compilation of Monitoring Data to Assess Temporal Trends in Perch Lake Radionuclide Levels

For ^{3}H and ^{90}Sr, measured data for surface water outflow from Perch Lake were assumed to represent concentrations of these radionuclides in the lake itself. However, this was not possible for ^{137}Cs or ^{60}Co, since these radionuclides were often not detectable in Perch Lake surface waters or downstream of the lake, particularly during the 1990s [20-57]. Despite their low-levels in Perch Lake water, ^{137}Cs and ^{60}Co were measurable in surface streams flowing into the lake. Using source term data, environmental transfer constants and a mass balance model, ^{137}Cs and ^{60}Co concentrations in Perch Lake surface waters have been estimated over a 40-year period. Temporal trends in radionuclide exposure to non-human biota were then quantified.

RESULTS

Summary of ^{90}Sr and ^{3}H Concentrations in Perch Lake from Environmental Monitoring Data

Based on environmental monitoring data for ^{90}Sr measured at Perch Lake outlet, peak ^{90}Sr levels occurred in the lake during the mid-1960s (Figure 3). Since then, there has been an overall decline in Perch Lake ^{90}Sr concentrations except for a slight peak in the late 1980s (Figure 3).

Tritium measurements are available for Perch Creek from the early 1960s until the present. Examination of temporal trends over this period suggests that current ^{3}H concentrations in Perch Lake are similar to or may be slightly higher than values observed in the 1960s, with lower levels occurring in the lake in the late 1970s (Figure 3). During the 1990s, it appears that there may have been a slight increase in Perch Lake ^{3}H levels; however, this trend may be an artifact, since the sampling site for the Perch Lake outflow was changed from Perch Lake outlet to a location further downstream at Perch Creek weir in 1988. Although a correction was made to account for this change in sampling location, it may have contributed to the observed increase in the 1990s.

Estimation of ^{137}Cs Concentrations in Perch Lake using Source and Loss Term Data

Between the 1950s and the 1980s, the primary source of ^{137}Cs to Perch Lake was atmospheric deposition due to global weapons testing in the late 1950s and early 1960s, with relatively little input from upstream water sources for the lake [4, 20-57; 59, 60].

Over this period, observed trends in atmospheric deposition concurred with regional and global trends reported in other studies [e.g. 4, 5, 61]. From the 1980s until the present, atmospheric deposition has been negligible and the main source of ^{137}Cs to Perch Lake has been via Inlet 2 [58]. However, overall, there has been little ^{137}Cs added to the lake following the inputs due to the period of atmospheric weapons testing (Figures 4 and 5), which represented 91% of the total ^{137}Cs added to the lake since the early 1950s.

Cesium-137 is potentially lost from Perch Lake via radioactive decay, surface outflow and complexation to the sediments. The half-life of ^{137}Cs is 30.2 years, representing a loss of 0.023 /a by radioactive decay. Loss via surface outflow is a function of the flushing rate or residence time of the lake. Perch Lake has a relatively short residence time of approximately 0.5 years, which corresponds to a loss of ^{137}Cs from the lake of 2 /a via surface outflow. Finally, ^{137}Cs loss to the sediments is a function of its tendency to partition into the sediments, as opposed to the water. This loss can be represented by the water-to-sediment transfer rate [3]. This parameter has a geometric mean value of 1.8±2.6 /a for ^{137}Cs, but can range from 0.2 to 6.3 /a [3]. On this basis, total annual loss of ^{137}Cs from Perch Lake is expected to be approximately 3.8 /a.

Cesium-137 source data and the estimated losses from Perch Lake have been used to estimate the ^{137}Cs concentration in the lake using the mass balance model developed by Lerman (Figure 6a) [62]. Although approximately 60% of the modeled values fall within 3-fold of the measured values (Figure 6b), ^{137}Cs concentrations in Perch Lake appear to be slightly overestimated using the Lerman approach (Figure 6a) [62]. This may be due to an underestimation of one of the loss terms. For example, as discussed above, the water-to-sediment transfer rate can greatly vary between lakes. Therefore, it is possible that the apparent discrepancy in the modeled values for lake ^{137}Cs concentration are due to site-specific differences in the water-to-sediment transfer rate. In addition, the ^{137}Cs concentration in surface waters upstream of the lake was often not detectable. As a result, in some cases it was necessary to use the lower limit of detection to approximate ^{137}Cs flux into the lake. Since the expected ^{137}Cs concentration was also fairly low, it is possible that a better fit of the modeled data to the measured data may have been achieved had samples been run until exact concentrations could be measured.

Estimation of ^{60}Co Concentrations in Perch Lake using Source and Loss Term Data

The Lerman model was also used to estimate ^{60}Co concentrations in Perch Lake water between the late 1950s and the present on the basis of influx data and estimated ^{60}Co loss from the lake [62]. Unlike ^{137}Cs, the contribution of ^{60}Co to Perch Lake via fallout was negligible. Surface water entering the lake at Inlet 2 represents the main source of ^{60}Co to the lake, with negligible contributions of <1% of the total ^{60}Co from Inlet 1 [20-57]. Most of the ^{60}Co entered Perch Lake between the 1960s and the mid-1980s (Figure 7).

However, since then, ^{60}Co influx into the lake has substantially declined. As a result of this decline and the relatively high rate of annual ^{60}Co loss from the water, ^{60}Co water concentrations are much lower than they had been during the time of peak input in the early 1970s (Figure 8a).

The annual ^{60}Co losses from Perch Lake are 2 , 0.13 and 1.9 /a for losses to surface water outflow, radioactive decay and sorption to sediments, respectively [3, 18, 63]. On this basis, the expected total annual loss of ^{60}Co from Perch Lake surface water is approximately 4 /a.

Overall, the ^{60}Co concentrations estimated using the Lerman model show very strong agreement with the measured values (Figure 8a) [62]. Approximately 98% of the modeled ^{60}Co concentrations in Perch Lake water fall within a factor of two of the measured values (Figure 8b). As a result, it is possible to use the Lerman model to estimate ^{60}Co levels in Perch Lake with confidence, even when they are not detectable at the lake outflow [62].

DISCUSSION AND CONCLUSIONS

Overall, it was found that the peak inputs to Perch Lake of most radionuclides considered in this 40-year period of record occurred in the mid-1960s to mid-1970s. The exception was ^{3}H which has shown relatively consistent influxes over time. During this 40-year period, ^{90}Sr, ^{3}H, ^{60}Co and ^{137}Cs migrated to Perch Lake from upstream Waste Management Areas. Radioactive fallout from weapons testing in the late 1950s and early 1960s also contributed ^{90}Sr, ^{3}H and ^{137}Cs to the lake, and represented the major source of ^{137}Cs. Since then, there have been significant declines in ^{137}Cs and ^{60}Co entering the lake.

These temporal trends have had a major impact on exposure of biota to these radionuclides via the water in Perch Lake for several reasons. First, the hydrologic residence time, or flushing time, for Perch Lake is only approximately 0.5 years. This means that if input concentrations significantly decline, “relatively contaminated” water in Perch Lake can be replaced with cleaner water over a short time period. In addition, both ^{137}Cs and ^{60}Co tend to partition into the sediments, which serve as a sink for these radionuclides. As a result of the substantial losses of ^{137}Cs and ^{60}Co from the system each year, the concentration of these radionuclides in Perch Lake surface waters is dictated by their total annual flux into the lake, and >98% of inflowing ^{60}Co and ^{137}Cs is lost from the lake within a year of input. Of this 98%, most partitions into the lake sediments, which are highly organic in nature and act as reservoir for these radionuclides. Because of this, currently, water does not represent an important exposure pathway for ^{137}Cs and ^{60}Co in comparison to the sediments.

Strontium-90 concentrations in Perch Lake are also currently lower than they had been in the past, but have not declined as quickly as ^{60}Co and ^{137}Cs. Unlike ^{60}Co and ^{137}Cs, ^{90}Sr has a relatively lower tendency to partition into the sediments than the water. Therefore, although exposure of aquatic biota to ^{90}Sr has declined, water still remains an important exposure pathway. Similarly, ^{3}H exposure via the water is very important to aquatic organisms, especially since ^{3}H has a lower water-to-sediment transfer rate than any of the other radionuclides under consideration in this study.

In conclusion, ^{3}H, ^{90}Sr, ^{60}Co and ^{137}Cs concentrations have been estimated in Perch Lake on the basis of concentrations measured in atmospheric deposition, as well as in inflowing and outflowing streams over a 40-year period, using the Lerman model [62]. Overall, the Lerman model provided reasonable estimates of radionuclide concentrations in Perch Lake over the 40 years, with most modeled values falling within 2-to-3 fold of measured values for ^{60}Co and ^{137}Cs, respectively. During this time, there has been a general decline in radionuclide concentrations in the lake. As a result, the concentrations of ^{137}Cs and ^{60}Co, which tend to partition into the sediments, are much lower in the lake water than observed in the past, and water is now not as important an exposure pathway for these radionuclides compared to the sediments. In comparison, direct contact with the water phase remains an important exposure pathway for ^{90}Sr and ^{3}H, which have not declined as much as ^{137}Cs and ^{60}Co and which tend to be more mobile in the environment. These trends are attributable to variations in the source terms, the short hydrologic residence time for Perch Lake and the different tendency of each radionuclide to partition into the sediments. Further work is currently being conducted to assess temporal trends in radiation dose to biota inhabiting Perch Lake on the basis of these and other data.

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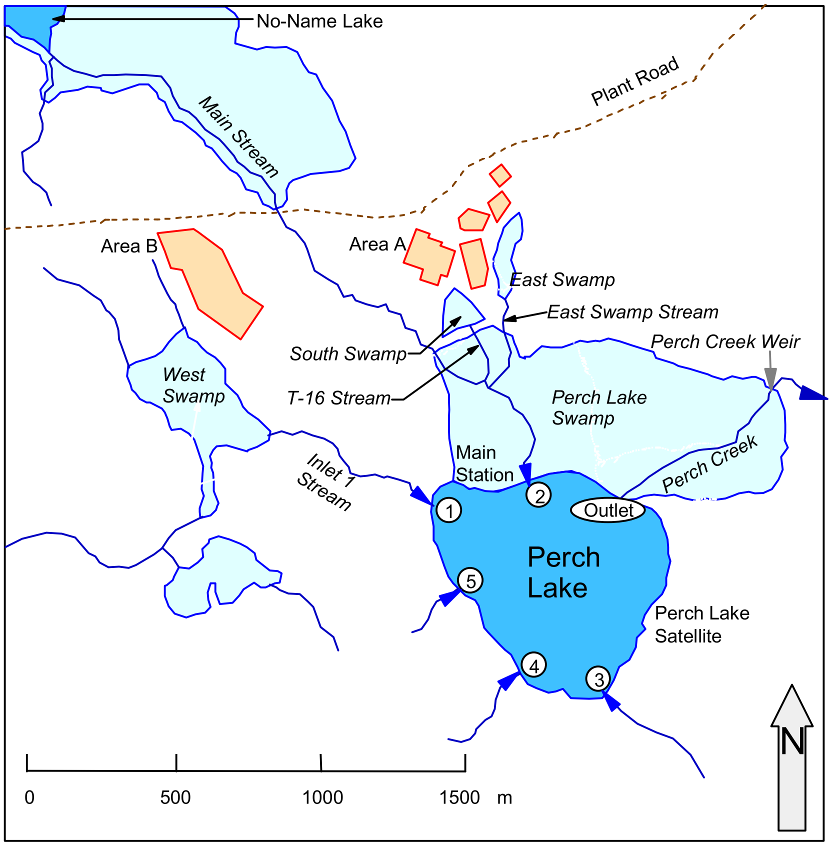

Figure 1: Map of the lower Perch Lake watershed depicting sources of radionuclide input into Perch Lake. Radionuclide sources are located north Perch Lake, originating from Areas A and B (as represented by grey boxes on the map). Surface waters exit via Perch Creek, which is located on the north-east corner of the lake.

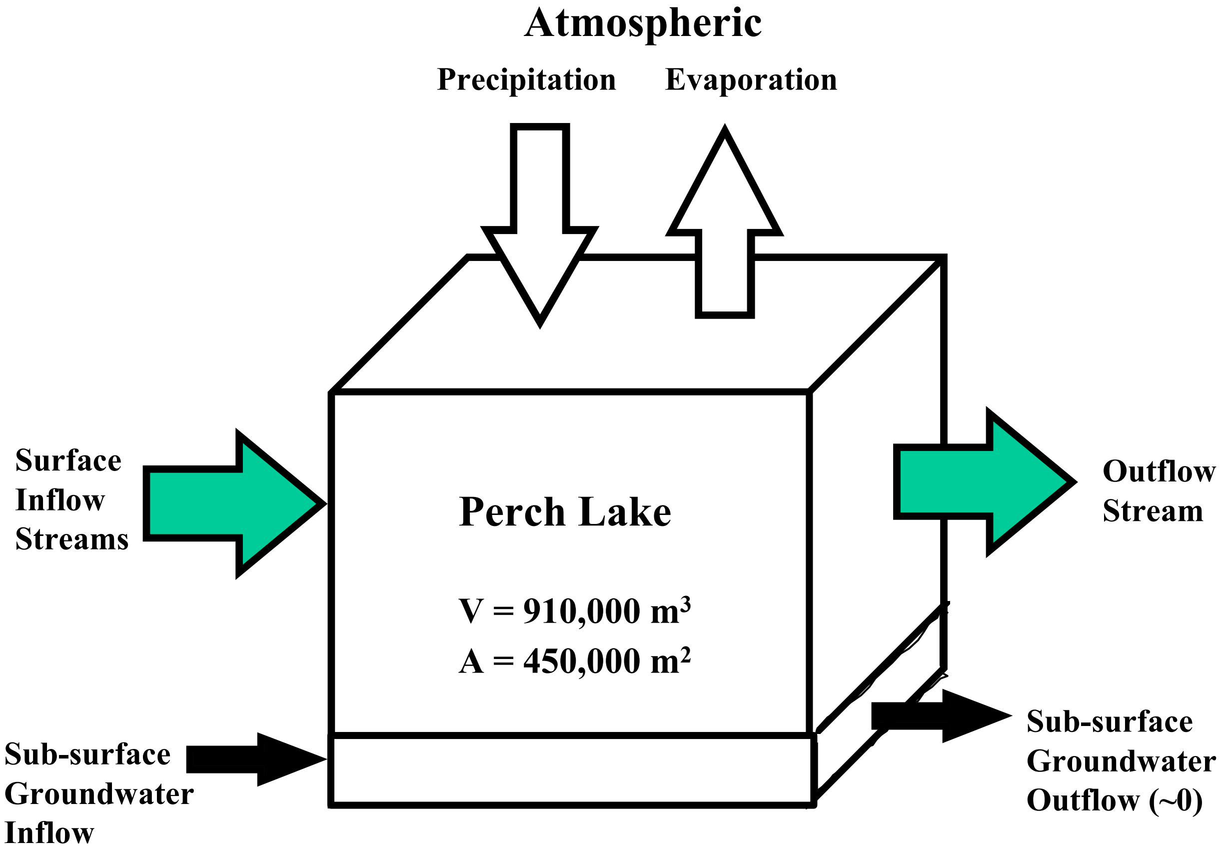

Figure 2: Mass balance diagram depicting key water flow pathways in the Lower Perch Lake watershed.

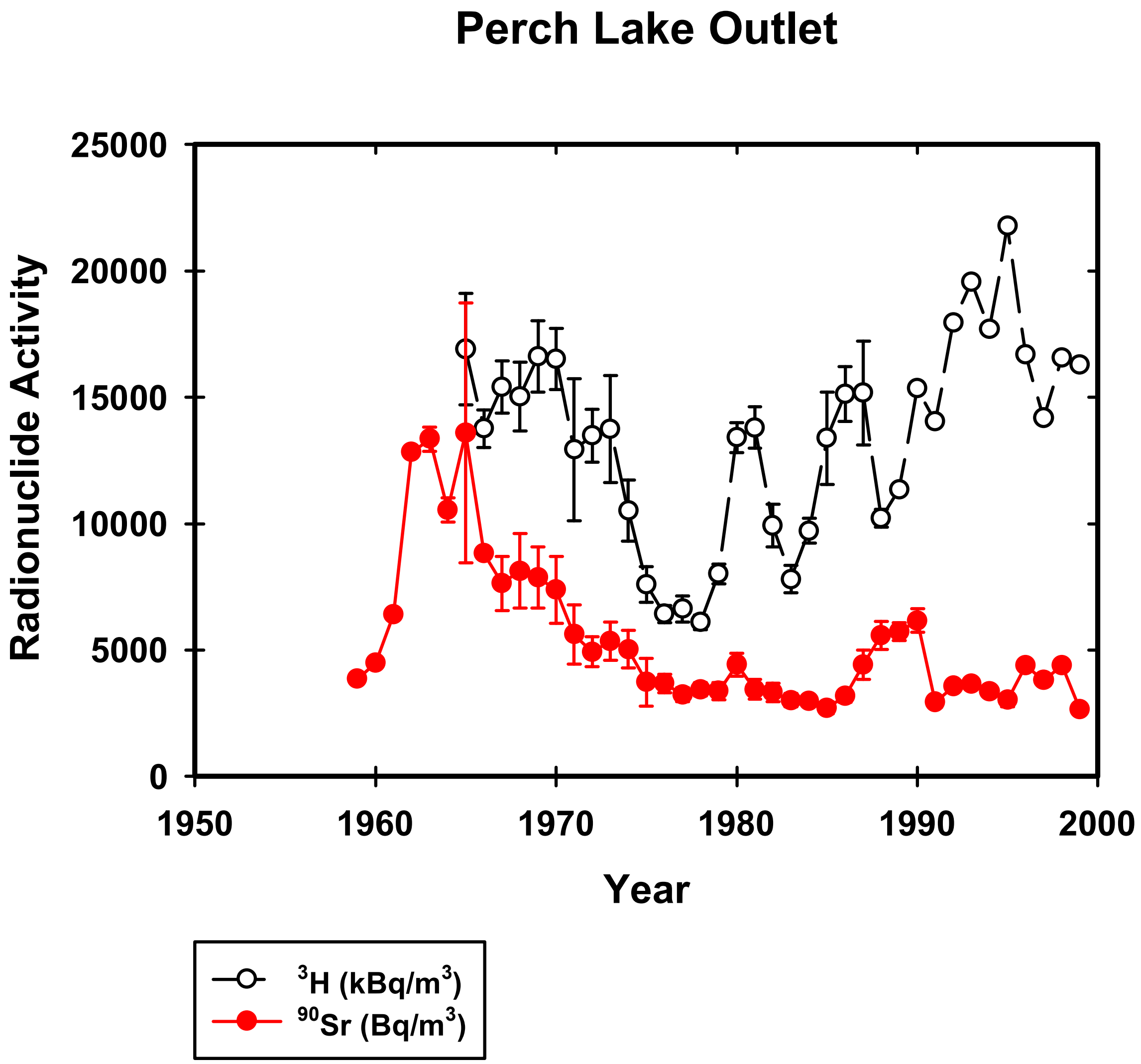

Figure 3: Mean ^{3}H and ^{90}Sr concentrations (± SE) in surface waters depicting temporal trends at Perch Lake outlet.

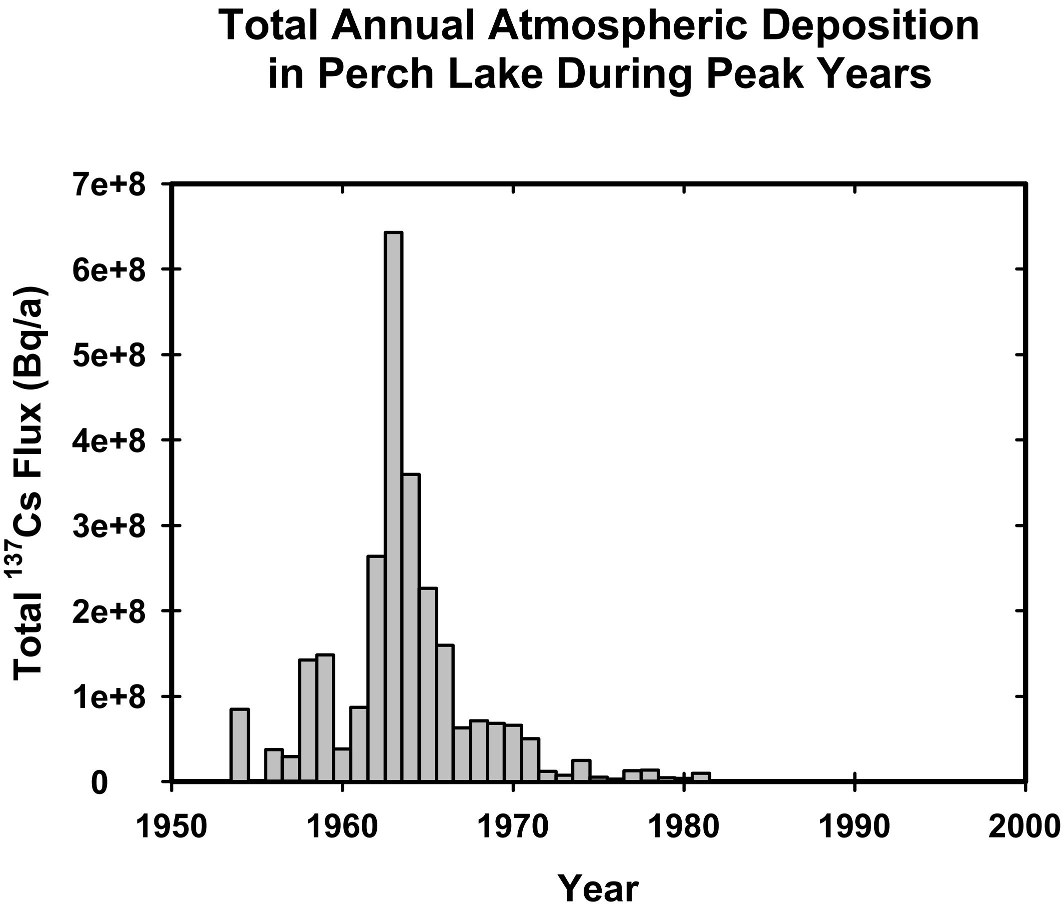

Figure 4: Total annual atmospheric deposition of ^{137}Cs in the vicinity of Perch Lake during peak years of deposition.

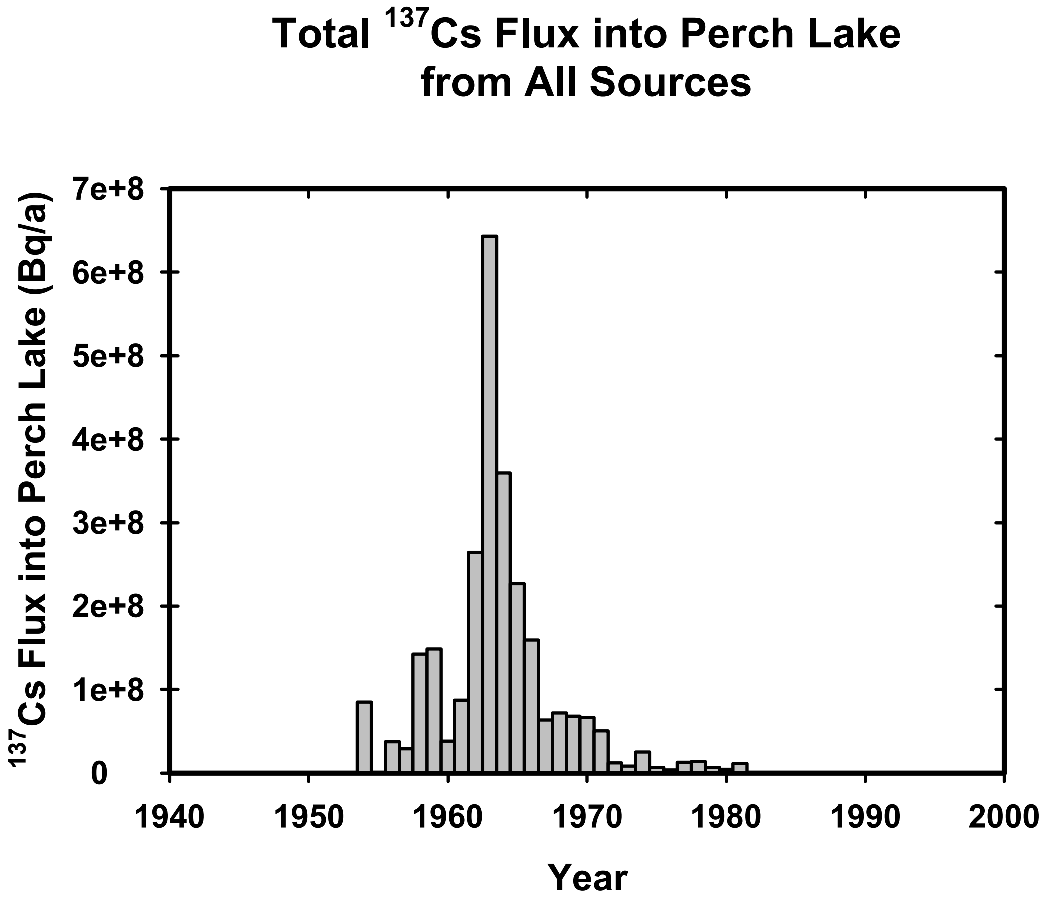

Figure 5: Temporal trends in total ^{137}Cs flux into Perch Lake via atmospheric deposition and surface inflow.

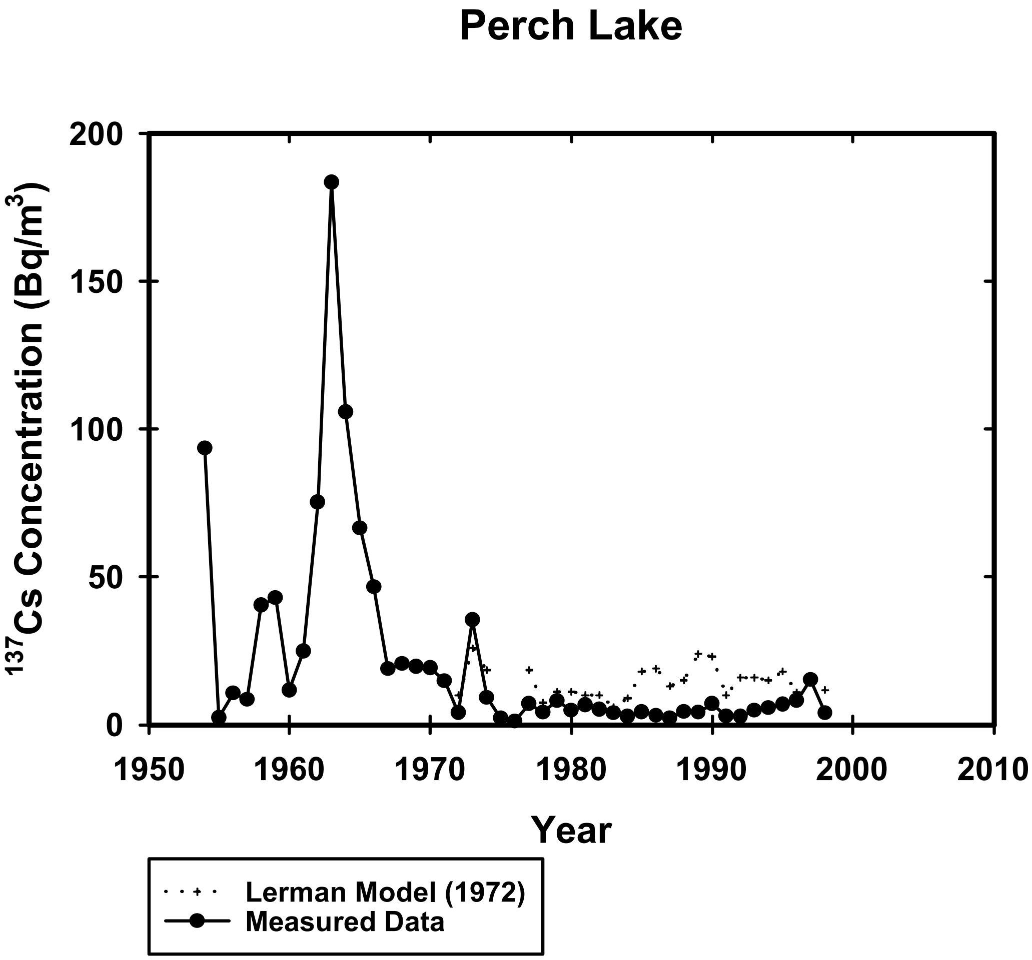

Figure 6a: Comparison of measured and modeled data depicting temporal trends in ^{137}Cs concentrations in Perch Lake surface waters.

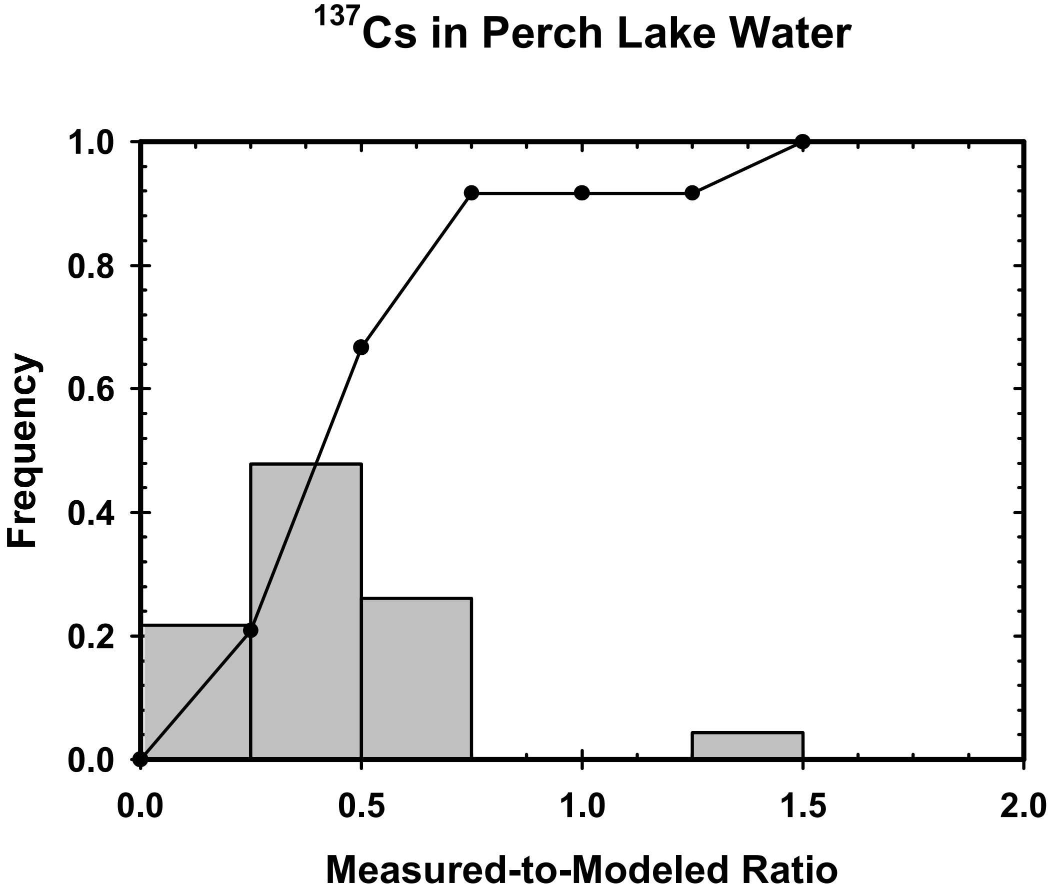

Figure 6b: Frequency histogram and cumulative distribution of measured-to-modeled ratio for ^{137}Cs concentration in Perch Lake water.

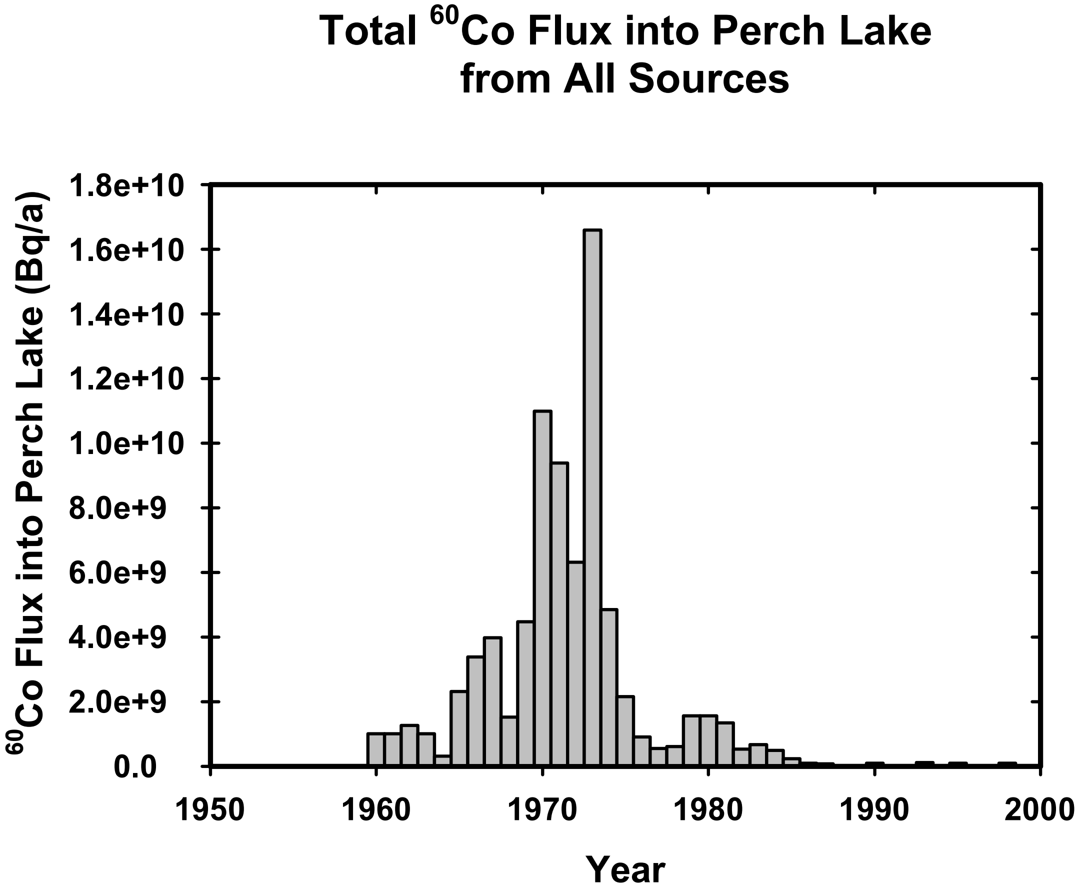

Figure 7: Temporal trends in total ^{60}Co flux into Perch Lake via surface inflow.

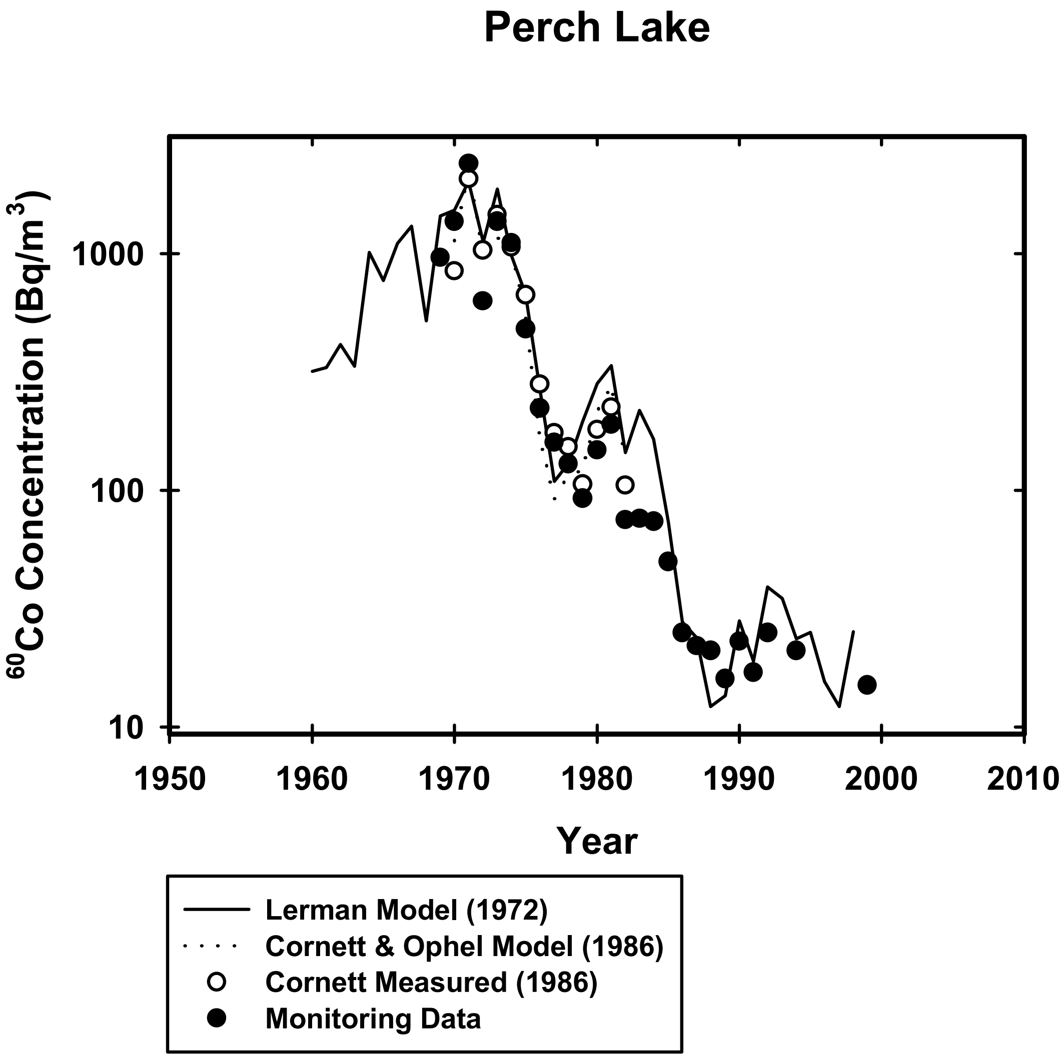

Figure 8a: Comparison of measured and modeled data depicting temporal trends in ^{60}Co concentrations in Perch Lake surface waters.

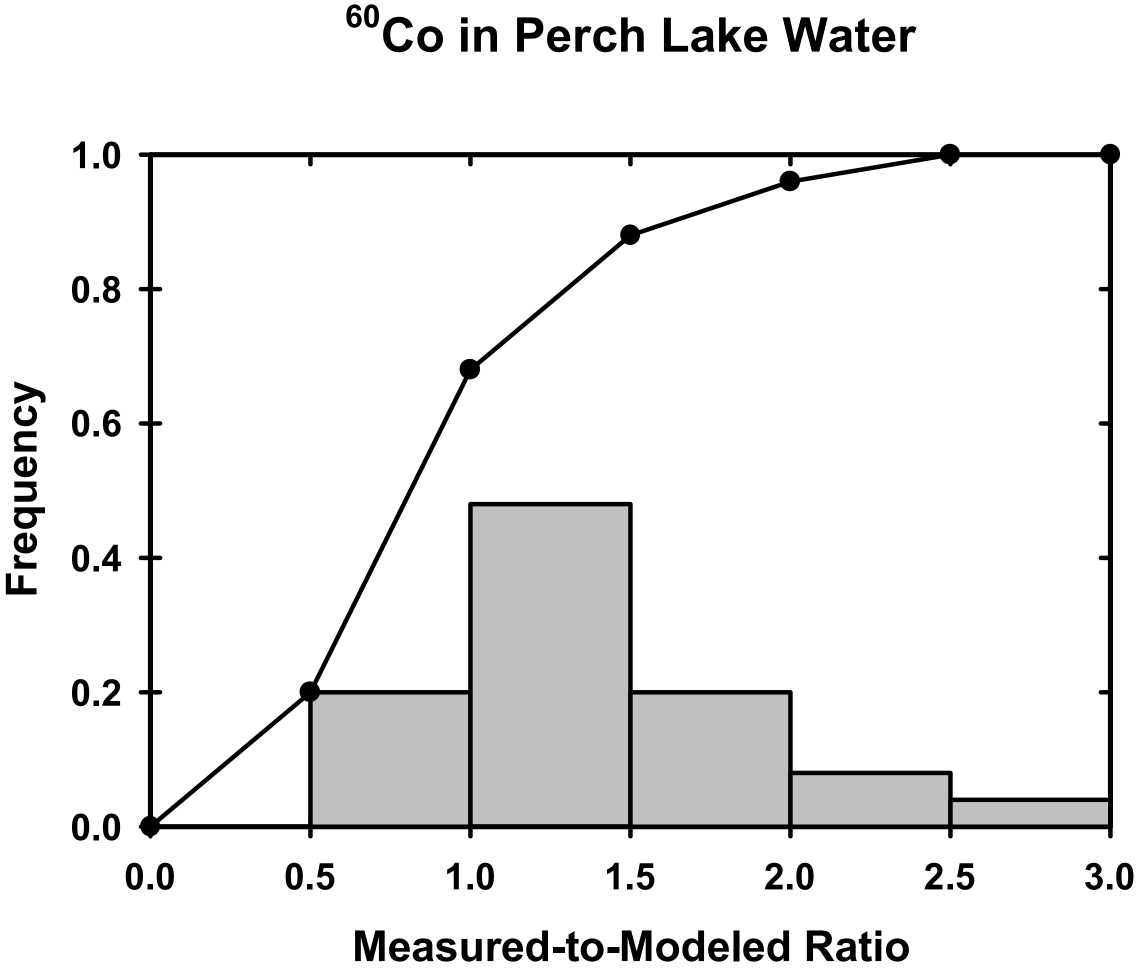

Figure 8b: Frequency histogram and cumulative distribution of measured-to-modeled ratio for ^{60}Co concentration in Perch Lake water.