Temporal changes in radionuclide transfer to biota in Canadian Shield lakes receiving chronic inputs: Reconstruction of radionuclide exposure to non-human biota in Perch Lake over a 40 year period

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Abstract. Temporal changes in radionuclide transfer factors were evaluated in forage fishes over a 40- to 50-year period. In general, 90Sr, 60Co and 137Cs transfer factors fairly consistent over time, as indicated by strong correlations between measured and estimated values. This conclusion was further strengthened by comparing radionuclide transfer factors measured in Perch Lake biota relative to those reported for other Canadian Shield lakes, since similar values were found. Despite these similarities, however, within-lake differences of up to an order of magnitude were observed for a given species collected in a given year, suggesting that within lakes, there may be factors influencing biota exposure to radionuclides and/or bioavailability.

1. INTRODUCTION

Changes in environmental contaminant levels are often assessed using non-human biota as biomonitors, due to their propensity to accumulate trace elements found at low levels in natural systems over time [1]. This is often accomplished through direct comparisons of contaminant concentrations in indicator species with those present in surface waters, in the form of transfer factors [1-5]; however, transfer factors can vary by orders of magnitude between species, between systems, and possibly within systems. In addition, transfer factors are applied under a steady state assumption, whereby the relationship between contaminant concentrations in biota tissues and concentrations in abiotic media in the surrounding environment are assumed constant through space and time. Depending upon the attributes of the species being considered and the conditions in a given system, it is possible that lags in transfer between biota and their environment may contribute to the large variability inherent in contaminant transfer factors. As a result, quantification of temporal changes in transfer factors of contaminants, such as radionuclides, in aquatic systems receiving chronic inputs can provide useful insight as to whether variability in transfer factors is related to changes in contaminant fluxes through a lake (i.e. whether transfer factors are constant over time within a system).

1.1 Study objectives

The objective of this study was to estimate 90Sr, 137Cs and 60Co transfer to Perch Lake forage fishes over the past 40 to 50 years based on fluctuations in surface water radionuclide concentrations;
2. METHODOLOGICAL APPROACH

2.1 Study system

Perch Lake (Chalk River, Ontario) represents an ideal system to assess whether radionuclide transfer factors remain constant over time within a system, since it has received chronic inputs of $^{60}$Co, $^{137}$Cs, $^{90}$Sr, and $^3$H that have been changing over a 40 to 50 year period [6]. Under typical conditions, larger fishes and other aquatic biota are prevented from entering or leaving the lake due to the presence of weirs on inflowing and outflowing streams. Therefore, in general, Perch Lake can be considered an open system with respect to radionuclides and a closed system with respect to large aquatic biota.

2.2 Methods

Forage fishes, including cyprinids and pumpkinseeds, were collected in Perch Lake in the mid- to late-1990s and subsequently analyzed for radionuclides. Archived samples were also processed and measured for radionuclide levels and historical data on radionuclides in water were compiled. Radionuclide data for biota and water were then plotted to evaluate inter-annual changes in radionuclide concentrations in these phases between the 1950s and the late 1990s. Radionuclide concentrations in Perch Lake biota collected in a given year were compared to a subset of values for biota collected in 1996, since a comprehensive survey was carried out in 1996. This involved tabulation of radionuclide transfer factors based on a subset of the 1996 values, and back-calculation to estimate radionuclide levels in biota in other years, as well as in the remaining 1996 samples, based on concentrations in surface water and transfer factors, as follows:

$$C_b = TF \cdot C_w \quad \text{(Eq. 1)}$$

or

$$TF = \frac{C_b}{C_w} \quad \text{(Eq. 2)}$$

where $C_b$ is the radionuclide concentration in biota (in Bq/kg fresh weight); $TF$ is the radionuclide transfer factor (in L/kg fresh weight); and $C_w$ is the radionuclide concentration in unfiltered Perch Lake surface water (in Bq/L).

Mean, minimum and maximum measured radionuclide levels and estimated values in biota were then compared graphically. Since a transfer factor represents the ratio of the concentration in biota relative to the water, the slope of a regression depicting radionuclide concentrations in biota tissues (Y) and concentrations in water (X) should be approximately equal to the transfer factor. Therefore, in cases where radionuclide concentration data were available over a range of surface water concentration values, the slope of a plot of concentration in biota relative to water was compared to the measured 1996 radionuclide transfer factors. Transfer factor data measured in Perch Lake were then compared to values measured in other Canadian Shield Lakes, wherever possible.

3. RESULTS AND DISCUSSION

3.1 Temporal changes in $^{90}$Sr transfer to Perch Lake forage fishes

In general, there was strong agreement between estimated and measured $^{90}$Sr levels in Perch Lake forage fishes (Table 1). Mean transfer factors fell within 1.0- and 1.3-fold of values estimated for pumpkinseeds and cyprinids, respectively, based on the slope of the line between whole fish $^{90}$Sr levels and those of Perch Lake surface waters (corresponding to percent differences of 0.053% and 13%, respectively) (Figures 1 and 2).
Table 1. Comparison of $^{90}$Sr transfer factors measured for a subset of 1996 samples and $^{90}$Sr transfer factors estimated based on the slope of a plot of $^{90}$Sr concentration in biota versus concentration in Perch Lake surface waters.

<table>
<thead>
<tr>
<th>Forage Fish Species</th>
<th>Sr-90 Transfer Factor (L/kg fresh weight)</th>
<th>$^a$ Estimated</th>
<th>$^b$ % Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pumpkinseed</td>
<td>945</td>
<td>944 (585 – 1831)</td>
<td>0.053%</td>
</tr>
<tr>
<td>Blacknose Shiner</td>
<td>429 (128 – 1229)</td>
<td>561</td>
<td>-13.3%</td>
</tr>
</tbody>
</table>

n.a. – not available

$^a$ Estimated Transfer Factor = Slope (of $^{90}$Sr Concentration in Biota versus $^{90}$Sr in Perch Lake surface water)

$^b$ %Difference = ($\text{TF}_{\text{measured}} - \text{TF}_{\text{estimated}}$) $\times$ 100%.

Figure 1. Comparison of $^{90}$Sr concentrations in whole pumpkinseeds relative to surface water. Data points represent mean measured data ($\pm$ SE), whereas the dashed line represents the linear regression line.

Figure 2. Comparison of $^{90}$Sr concentrations in whole cyprinids relative to surface water. Data points represent mean measured data ($\pm$ SE), whereas the dashed line represents the linear regression line.
2.2 Temporal changes in $^{60}$Co transfer to Perch Lake forage fishes

Measured $^{60}$Co concentrations in Perch Lake forage fish tissues also corresponded with estimated values (Table 2). For example, measured versus estimated $^{60}$Co transfer factors differed by approximately 1.2-fold for Perch Lake pumpkinseeds (Figure 3), although data were insufficient to make similar comparisons for cyprinid species occupying the lake. In addition, $^{60}$Co transfer factors concurred with literature values reported for Canadian Shield lakes, showing differences of approximately 1.0- to 1.2-fold (Table 3).

### Table 2. Comparison of $^{60}$Co transfer factors measured for a subset of 1996 samples and $^{60}$Co transfer factors estimated based on the slope of a plot of $^{60}$Co concentration in biota versus concentration in Perch Lake surface waters.

<table>
<thead>
<tr>
<th>Species/Medium</th>
<th>Co-60 Transfer Factor (L/kg fresh weight)</th>
<th>$^a$ Estimated</th>
<th>$^b$ % Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Measured</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pumpkinseed</td>
<td>85.9 (20.9 – 104)</td>
<td>71.5</td>
<td>9.15%</td>
</tr>
<tr>
<td>Blacknose Shiner</td>
<td>128 (121 – 135)</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
</tbody>
</table>

n.a. – not available

$^a$ Estimated Transfer Factor = Slope (of $^{60}$Co Concentration in Biota versus $^{60}$Co in Perch Lake surface water)

$^b$ %Difference = \( \frac{\text{TF measured} - \text{TF estimated}}{\text{TF measured} + \text{TF estimated}} \times 100\% \).

### Table 3. Comparison of transfer factors of $^{60}$Co radioisotopes for Perch Lake (PL) relative to Lake 226 (L226) non-human biota (based on data collected as part of this study, as well as data from Bird et al. [7]).

<table>
<thead>
<tr>
<th>Species</th>
<th>$^{60}$Co Transfer Factor (L/kg dry)</th>
<th>Perch Lake</th>
<th>Lake 226 (ELA)</th>
<th>% Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forage Fishes</td>
<td></td>
<td>430</td>
<td>530</td>
<td>10%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>640</td>
<td>530</td>
<td>9%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>535</td>
<td>530</td>
<td>0.45%</td>
</tr>
</tbody>
</table>

Figure 3. Comparison of $^{60}$Co concentrations in whole pumpkinseed tissue relative to surface water. Data points represent mean measured data (± SE), whereas the dashed line represents the linear regression line.
3.3 Temporal changes in $^{137}$Cs transfer to Perch Lake forage fishes

Unlike for $^{90}$Sr and $^{60}$Co, only limited historical data were available for $^{137}$Cs in Perch Lake forage fishes, making it necessary to estimate temporal changes in $^{137}$Cs concentrations in fish tissues. This was accomplished using empirical models that had been developed by Rowan and Rasmussen [8] for non-piscivorous fishes, which required input of $^{137}$Cs and K concentration data for surface waters. In general, Perch Lake water shows K concentrations that tend to fall on the low end of the range compared with other freshwaters systems, with values of 0.9 to 1 mg/L. Measured $^{137}$Cs values fell within the estimated range for Perch Lake fishes, although data were not available during the period prior to 1980 when $^{137}$Cs levels were highest in the lake (Figure 4).

Cesium-137 transfer factors for Perch Lake fishes 1.5- to 3.3-fold of values reported for other Canadian Shield lakes [7]. Therefore, estimated values fell within the variability produced by Rowan and Rasmussen’s [8] empirical model, suggesting again that radionuclide transfer in Perch Lake is similar to values measured in other Shield lakes.

Table 4. Comparison of transfer factors of Cs radioisotopes for Perch Lake (PL) relative to Lake 226 (L226) non-human biota (based on data collected as part of this study, as well as data from Bird et al. [7]).

<table>
<thead>
<tr>
<th>Species</th>
<th>Cs Radioisotopes Transfer Factor (L/kg dry)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Perch Lake</td>
</tr>
<tr>
<td>Forage Fishes (Pumpkinseeds)</td>
<td>919</td>
</tr>
<tr>
<td>Forage Fishes (Blacknose Shiners)</td>
<td>425</td>
</tr>
<tr>
<td>Forage Fishes (general)</td>
<td>672</td>
</tr>
</tbody>
</table>

Figure 4. Comparison of measured $^{137}$Cs concentrations in Perch Lake non-piscivorous fish relative to estimated values for non-piscivorous fishes. Triangles represent measured data points, whereas lines represent modelled values.
4. SUMMARY OF WORK

In general, radionuclide transfer factors for forage fishes were fairly consistent over time, as indicated by strong correlations between measured and estimated values. This conclusion was further strengthened by comparing radionuclide transfer factors measured in Perch Lake biota relative to those reported for other Canadian Shield lakes [7], since similar values were found. Despite these similarities, however, within-lake differences of up to an order of magnitude were observed for a given species collected in a given year, suggesting that within lakes, there may be factors influencing biota exposure to radionuclides (possibly related to proximity to source and mixing) and/or bioavailability (possibly due to spatial differences in speciation in lakes). Further work is required to gain insight as to the role of such factors in influencing the transfer of radionuclides to aquatic biota.

References