



Cesium accumulation by fish following acute input to lakes: a comparison of experimental and Chernobyl-impacted systems

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ABSTRACT

An uptake parameter u ($\text{L kg}^{-1} \text{d}^{-1}$) and a loss rate parameter k (d^{-1}) were estimated for the patterns of accumulation and loss of ¹³³Cs by three fish species following an experimental ¹³³Cs addition into a pond in South Carolina, USA. These u and k parameters were compared to similar estimates for fish from other experimental ponds and from lakes that received ¹³⁷Cs deposition from Chernobyl. Estimates of u from ponds and lakes declined with increasing potassium concentrations in the water column. Although loss rates were greater in the experimental ponds, the times required to reach maximum Cs concentrations in fish were similar between ponds and lakes, because ponds and lakes had similar retentions of Cs in the water column. The maximum Cs concentrations in fish were largely determined by initial Cs concentrations in the water column. These maximum concentrations in fish and the times required to reach these maxima are potentially useful indicators for assessments of risks to humans from fish consumption.

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1. Introduction

Because fish consumption can be an important source of ¹³⁷Cs exposure to humans (e.g., Travnikova et al., 2004), the ¹³⁷Cs concentrations of fish in contaminated lakes have received extensive study. In situations where the ¹³⁷Cs concentrations in lake water are approximately constant over time, several analyses have derived simple equations to predict concentration factors (C_f) and elimination rates (Reichle et al., 1970; Blaylock, 1982; Ugedal et al., 1992; Rowan and Rasmussen, 1994, 1995; Smith et al., 2000; NCRP, 2007) using variables such as the size and trophic positions of fish and the temperature, pH and K concentrations of lake waters.

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Such simple predictive equations are less well developed for conditions that are dynamic and not at steady state, such as those following the pulsed inputs of Cs isotopes from the Chernobyl accident. This lack is especially noteworthy given that ¹³⁷Cs concentrations in fish 1) may not reach their maximum concentrations until months after the initial input (Grimas, 1989; Hakanson et al., 1992; Monte et al., 1993; Sundbom et al., 2003; Saxen and Sundell, 2006), and 2) may retain these high concentrations for some months thereafter (Jonsson et al., 1999; Smith et al., 2000, 2002; Sundbom et al., 2003; Franic and Marovic, 2007). These delayed maxima and continuing high concentrations reflect 1) the time required for Cs to pass through lake food chains leading to fish, 2) continued elevated Cs concentrations in the water column due to either continuing Cs inputs from the watershed (Smith et al., 1997; Hakanson, 2004) or the recycling of Cs from lake sediments (Alberts et al., 1979; Evans et al., 1983), and 3) the often low rate of elimination of Cs by fish (Ugedal et al., 1992; Rowan and Rasmussen, 1995).

This paper examines Cs accumulation in fresh water fish under nonsteady-state conditions resulting from 1) the experimental additions of Cs into relatively small ponds and 2) the contamination

of larger lakes by Chernobyl releases. The purposes of this analysis are to further illustrate the potential utility of a simple modeling approach employed by Smith et al. (2002), and to identify factors that may be useful in determining the appropriate parameters for this modeling approach in newly contaminated systems. The following sections of the paper 1) describe the modeling approach, 2) derive estimates of the model parameters from the experimental addition of ^{133}Cs to Pond 4, a small, shallow reservoir in the southeastern United States, 3) derive model parameters from previously reported experimental studies by Kolehmainen et al. (1967) and Hakonson and Whicker (1975), 4) compare the parameter estimates from the experimental ponds with those reported by Smith et al. (2002) for European lakes impacted by Chernobyl releases, and 5) examine potential relationships between model parameters and basic lake metrics, such as pH, K concentrations, water retention times and mean depths, that may be useful in establishing preliminary estimates of parameter values. The models are also used to estimate 1) the time to maximum concentrations in fish and 2) the concentrations occurring at that time.

2. Modeling approach

2.1. Model structure

To model the transient behavior of ^{137}Cs concentrations in fish, several researchers and modelers have employed the simple uptake and loss model

$$\frac{F(t)}{dt} = \mu W(t) - kF(t) \quad (1)$$

where μ is an uptake constant with units $\text{L kg}^{-1} \text{d}^{-1}$ and k is a first-order loss rate constant with units d^{-1} , $W(t)$ is the dissolved Cs concentration in water, and $F(t)$ is the Cs concentration in whole fish or specific fish tissues such as muscle. For this analysis where data on stable ^{133}Cs and radioactive ^{137}Cs will be compared, all concentrations will be expressed as mass of the Cs isotope per volume of water or wet mass of fish tissue. Thomann (1981) used a similar approach for modeling ^{239}Pu and ^{137}Cs in aquatic food chains. Smith et al. (2002) used Eq. (1) to examine the temporal dynamics of ^{137}Cs in Chernobyl contaminated lakes, and Smith et al. (2006) and Smith (2006) have contrasted the approach of Eq. (1) with the use of models based on C_r for nonsteady-state river systems.

The approach of Eq. (1) has been used to predict Cs movement through lake food webs (Monte et al., 2003) in the models AQUASCOPE (Smith et al., 2005) and ECOPRAQ (Comans et al., 2001). Other, more mechanistic models (e.g., Hakonson, 2000) exist to predict Cs behavior in lake food chains, but Eq. (1) is employed in these analyses because the purpose is not so much to predict movement as it is to summarize the pattern of movement into a few simple parameters that may be used to readily compare patterns among lakes and ponds. Where these parameters are used for prediction, such as predicting the maximum Cs concentration in fish, they are used because the required input parameters are not available to compute predictions from more sophisticated models for all the lakes in question.

The parameters μ and k in Eq. (1) will be referred to as uptake and loss rather than absorption and elimination because their estimated values may measure more than just physiological processes. Two separate interpretations of μ are illustrated in Fig. 1. For producer components (P), such as periphyton, which absorb Cs directly from the water column (W), μ is an estimate of absorption rate, and the transfer $t_{1,2}$ between the water and the producer

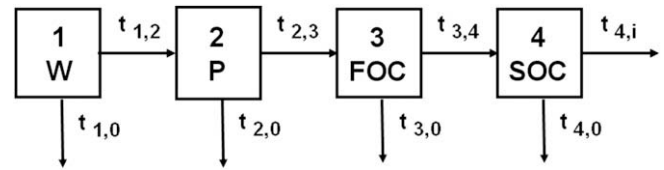


Fig. 1. Illustration of a simple transfer pathway for Cs where W = water, P = producer, FOC = first-order consumer, and SOC = second-order consumer. The $t_{i,j}$ indicates transfers where $t_{1,0}$ indicates loss of Cs from the water column, $t_{1,2}$ = absorption of Cs from the water column by the producers, $t_{i,0}$ for $i \geq 2$ indicates loss from the biota due to excretion as well as losses due to mortality, emigration and other causes, and $t_{i,i+1}$ for $i \geq 2$ indicates consumption by higher trophic levels.

component in Fig. 1 may be expressed as $\mu W(t)$. For the consumer compartments (C) in Fig. 1, μ is not a measure of absorption from the water column or absorption from ingestion of the precursor compartment. Rather it is a measure of the transfer through the pathways from the water to the i th component, and μ increases with the magnitudes of the transfers $t_{i-1,i}$ and declines with the magnitudes of the transfers $t_{i-1,0}$. The factors affecting μ become increasingly complex with increases in the number of trophic levels and increases in the number of alternative pathways from the water to the consumer. For this reason, μ may decline as trophic position increases despite an increase in assimilation efficiencies of Cs absorption from ingested material at higher trophic levels. This interpretation of μ for fish assumes that Cs is primarily absorbed from ingested materials which is consistent with studies showing that relatively little Cs is directly absorbed from water by fish (Hewett and Jeffries, 1976, 1978; Morgan et al., 1993; Topcuoglu, 2001; Malek et al., 2004).

The parameter k in Eq. (1) is a measure of loss that incorporates both the transfers $t_{i,0}$ and $t_{i,i+1}$ in Fig. 1. The $t_{i,0}$ loss involves excretion as well as losses due to mortality. Because $F(t)$ is expressed as a concentration, k may also include the effects of growth dilution (Thomann, 1981) and recruitment of relatively uncontaminated new individuals in short-lived populations. The effects of transfers $t_{i,i+1}$ on k for the i th component are affected by the pattern of consumption. Where concentrations are uniform among individuals, consumption may have little impact on concentrations and k . Where consumption preferential occurs on individuals with large concentrations, k is increased. Where individuals with small concentrations are preferentially consumed, k is decreased. Because k measures more than just physiological elimination, the estimated k could be expected to differ from physiological elimination rates predicted by the procedures of Rowan and Rasmussen (1995) and Ugedal et al. (1992).

2.2. Procedures for estimating μ and k in experimental systems

The estimation of μ and k in small ponds receiving experimental additions of Cs isotopes involved a two step process. First, the declines in Cs concentration in water following the experimental addition were approximated using a multi-component exponential equation (Whicker and Shultz, 1982) of the form:

$$W(t) = \sum_i a_i e^{-b_i t} \quad (2)$$

where $W(t)$ = the dissolved Cs concentration in water, a_i = the initial concentration of the i th component and b_i = the rate constant for exponential decline in the i th component. For the purposes of discussion, these components will be arranged in order of decreasing b_i and termed the first, second and third components. In the second step, estimates of μ and k are obtained by fitting the

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