MODELLING THE DYNAMICS OF FISH CONTAMINATION BY CHERNOBYL RADIOCAESIUM: AN ANALYTICAL SOLUTION BASED ON POTASSIUM MASS BALANCE

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INTRODUCTION

The Chernobyl nuclear accident provided a sudden contamination of many European water bodies with radiocaesium, followed by a rapid partial immobilization. As a result, activities and concentration factors of radiocaesium in biota fluctuated strongly over several years before reaching quasi-equilibrium, with patterns significantly differing between organisms. To model these dynamic relaxation processes, mass balance equations for ¹³⁷Cs in aquatic food chains were developed on the following basis: a) potassium acts as a biogeochemical equivalent of caesium; b) the concentration of potassium in fish and other biota is rather constant; c) the main source of potassium in freshwater fish is the dietary uptake.

MODEL DEVELOPMENT

The model describes the fate of a contamination pulse in the water and in associated food chains. The decline of the activity concentration in the water, which is considered as the secondary source of ¹³⁷Cs, can be well expressed by a two-component exponential function corresponding to a fast (**F**) and a slow (**S**) component in the immobilization of Cs:

$$A_w(t) = K_w^F \cdot \exp(-\lambda_F \cdot t) + K_w^S \cdot \exp(-\lambda_S \cdot t)$$

where \boldsymbol{t} is time, \boldsymbol{A} are activity concentrations, \boldsymbol{K} are transfer coefficients, and λ are rate constants.

The food chain considered here includes the following compartments: fish food organisms such as zooplankton and zoobenthos (**Z**), non-piscivorous fish (**NP**), and piscivorous fish (**P**). In every compartment the turnover rate of caesium is considered as constant over time. If potassium acts as a biogeochemical equivalent of caesium, the uptake of Cs in animals is proportional to the ratio of assimilation efficiencies of Cs and K, and the elimination of Cs is proportional to the ratio of turnover rates of Cs and K. Since homeostasis requires that uptake and elimination of K are equal, mass balance yields the following equation for the member **j** of a food chain:

$$\frac{d\boldsymbol{A}_{j}}{dt} + \lambda_{j}^{cs} \cdot \boldsymbol{A}_{j}(t) = \boldsymbol{T}_{j-1}^{j} \cdot \boldsymbol{A}_{j-1}(t); \quad \text{with} \quad \boldsymbol{T}_{j-1}^{j} = \boldsymbol{k}_{\lambda j} \cdot \lambda_{j}^{cs} \cdot \frac{\boldsymbol{\alpha}_{j}^{cs}}{\boldsymbol{\alpha}_{j}^{K}} \cdot \frac{\boldsymbol{C}_{j}^{K}}{\boldsymbol{C}_{j-1}^{K}} \quad \text{and} \quad \boldsymbol{k}_{\lambda j} \equiv \frac{\lambda_{j}^{K}}{\lambda_{j}^{cs}} \cdot \frac{\boldsymbol{A}_{j}^{Cs}}{\boldsymbol{\lambda}_{j}^{Cs}} \cdot \frac{\boldsymbol{A}_{j}^{Cs}}{\boldsymbol{A}_{j}^{Cs}} \cdot \frac{\boldsymbol{A}_{j}^{Cs}}{\boldsymbol{A}_{j}^{$$

where ${\bf A}$ are radiocaesium activity concentrations, ${\bf C}$ are potassium concentrations, ${\bf T}$ is a complex transfer coefficient, α are assimilation efficiencies, and λ are first order decay rate constants describing the biotic elimination (turnover) of Cs.

An analytical solution of this equation is given by:

$$\mathbf{A}_{j}\left(t\right) = \exp(-\lambda_{j}^{cs} \cdot t) \cdot \left[\mathbf{T}_{j-1}^{j} \cdot \int \, \mathbf{A}_{j-1}(t) \cdot \exp(\lambda_{j}^{cs} \cdot t) \, dt \, + \mathit{Const} \, \right]$$

where Const is determined by initial conditions:

$$Const = \begin{bmatrix} \mathbf{A}_{i} \end{bmatrix}_{t=0} - \begin{bmatrix} \mathbf{T}_{j-1}^{i} \cdot \int \mathbf{A}_{j-1}(t) \cdot \exp(\lambda_{j} \cdot t) \ dt \end{bmatrix}_{t=0}.$$

When applying this model to a food chain after combination with the water model, solving the equations adds one exponential term for each compartment. For piscivorous fish, the following equation is obtained:

$$\begin{split} \textbf{A}_{\textbf{p}}(\textbf{t}) &= \textbf{T}_{\textbf{W}}^{\textbf{p}} \cdot \left[\begin{array}{c} \textbf{K}_{\textbf{p}}^{\textbf{F}} \cdot \exp(-\lambda_{\textbf{F}} \cdot \textbf{t}) + \textbf{K}_{\textbf{p}}^{\textbf{S}} \cdot \exp(-\lambda_{\textbf{S}} \cdot \textbf{t}) \\ -\textbf{K}_{\textbf{p}}^{\textbf{Z}} \cdot \exp(-\lambda_{\textbf{Z}} \cdot \textbf{t}) - \textbf{K}_{\textbf{p}}^{\textbf{NP}} \cdot \exp(-\lambda_{\textbf{NP}} \cdot \textbf{t}) - \textbf{K}_{\textbf{p}}^{\textbf{P}} \cdot \exp(-\lambda_{\textbf{p}} \cdot \textbf{t}) \end{array} \right] \end{split}$$

where $T_{\mathbf{W}}^{\mathbf{P}}$ is the bioconcentration factor and \mathbf{K} are complex transfer coefficients.

MODEL VALIDATION

The model was tested with post-Chernobyl data from long-term studies in different European lakes [1-6]. For each compartment, a transfer coefficient (\mathbf{K} or α) and a rate constant λ was fitted iteratively to the data. An example from one lake is shown in Fig. 1. The model provided a good description of several important aspects: a) the differences in contamination levels and contamination dynamics between predatory and non-predatory species (trophic level effects); b) the dependence of ¹³⁷Cs activity in fish on the potassium concentration in the water (bioavailability); c) time scales of contamination and depuration in different fish species (prognostic tool). Evaluation also showed a significant effect of growth rate on the turnover rate (biological half-life) of ¹³⁷Cs in fish.

CONCLUSIONS

The complex contamination dynamics in aquatic food chains after an accidental release of radiocaesium can be adequately described with a physiological model, using equations that can be solved analytically. These solutions describe the ¹³⁷Cs activity concentration in every compartment as a series of exponential functions, of which some are derived from the concentration in the water (source pattern), and the others determined by the caesium turnover rate in each food chain compartment.

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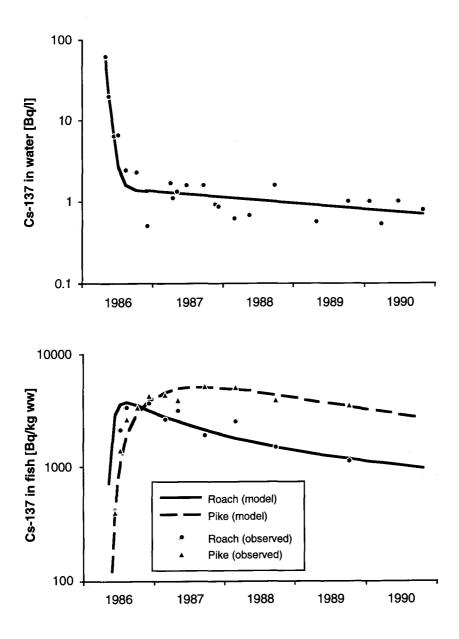


Figure 1. Observed data [1] and modelled values of ¹³⁷Cs activity concentrations in the water and in different fish species in lake Hillesjön (Sweden) after the Chernobyl fallout in 1986.