Time-dependency of the ^{137}Cs activity concentration in freshwater lakes, measurement and prediction

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Abstract. 14 years after the nuclear reactor accident in Chernobyl, activity concentrations of lake water are measured as 0.06 mBq/l, 2 mBq/l, and 60 mBq/l in Lake Constance, Lake Lugano and Lake Vorsee. These values can be explained by differences in limnological character of these lakes. To describe and predict the data of lake water contamination we use (1) a simple exponential "transfer function" model for the runoff of activity from contaminated catchments, and assume that runoff water determines lake water contamination. In a second approach (2) lake water residence times and ^{137}Cs uptake by sediments are taken into account together with runoff. With their blind predictions both models describe the ^{137}Cs contamination in lake water of the three lakes quite well. Based on the second approach, a fish model is used to blindly predict the fish contamination in Lake Constance and Lake Vorsee with a deviation smaller than factor 10 from measured values. Attempts to use these models also for a number of other lakes in Europe and in the CIS countries near to the site of Chernobyl are promising with respect to a generalisation of our results to predict ^{137}Cs contamination of freshwater lakes for radiation protection purposes in case of emergency.

1. INTRODUCTION

Following a fallout event, radionuclides are removed from a lake through the outflow and to bottom sediments. Transfers to bottom sediments occur as a result of attachment to and settling of suspended particles and via direct diffusion across the sediment/water interface. In the longer term, radionuclides like ^{137}Cs may potentially be remobilised from bottom sediments, under anoxic conditions, or resuspended by biological and physical turbation. In addition to these mechanisms, tributaries transport radionuclides from the contaminated catchment in the lake. In the runoff model this transport out of the catchment is considered only.

2. RESULTS AND DISCUSSION

2.1 Runoff model

In this model [1-3] is assumed, that the activity concentrations of lake water and of water flowing in the lake from the catchment are the same. For this study we will use an exponential "transfer function" model [4, 1] for the runoff of activity from contaminated catchments as described by Monte 1995 and Smith et al. 1999. This model assumes three components to the transfer:

- a short term (timescale, $\tau \sim 30$ d) transfer of recently deposited activity,
- a medium term exponential decline ($\tau \sim$ years) as a result of changing availability of the radionuclide in catchment soils and
- a long-term ($\tau \sim$ decades) "equilibrium" value.

C_R (Bq.m^{-3}), the radionuclide concentration in runoff water is given by:

$$C_R(t) = D\left(\alpha\, e^{-(\lambda+k_1)t} + \beta\, e^{-(\lambda+k_2)t} + \gamma\, e^{-(\lambda+k_3)t}\right) \qquad (1)$$

where λ is the decay constant of the radionuclide and D is the radionuclide fallout (Bq.m^{-2}) on the catchment. $\alpha,\ \beta,\ \gamma$ (m^{-1}) and $k_1,\ k_2\ k_3$ (y^{-1}) are empirically determined constants.

From extensive data sets of ^{137}Cs in runoff water the following parameter values resulted [2]:

$\alpha = 0.3\ m^{-1}$ $\qquad\qquad\qquad\qquad\qquad$ $k_1 = 18\ y^{-1}$

$\beta = 0.003(f_{min}) + 0.05(f_{org})\quad m^{-1}$ $\qquad$ $k_2 = 0.41\ y^{-1}$

$\gamma = 0.0002(f_{min}) + 0.007(f_{org})\ m^{-1}$ $\qquad$ $k_3 = 0.02\ y^{-1}$

Where f_{org} is the fractional coverage of the catchment by organic, boggy soils and f_{min} (= 1 - f_{org}), the fractional coverage by non-organic soils. This set of parameter values is used in the following models to make blind predictions for the ^{137}Cs activity concentration in water and fish of three different lakes. Required input data for the "Runoff-model" is the deposition, D, and the catchment coverage by organic, boggy soils, f_{org}.

In Fig. 1 the result of the runoff model is given for Lake Constance, Lake Vorsee and Lake Lugano. The following input data have been used:

Table 1: Input data of the runoff model for lake water from catchments having fractional coverage by organic soils f_{org} and ^{137}Cs inventory D and decay constant λ.

Lake	f_{org}	D [kBq/m^2]	λ [a^{-1}]
Constance	0	7,6	0,02295
Vorsee	0,2	28	0,02295
Lugano	0	24	0,02295

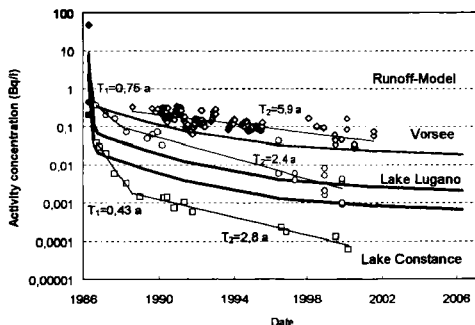**Figure 1:** Time-dependency of ^{137}Cs activity concentration of water in three prealpine lakes, measured data with trend lines and ecological half-lives T$_1$ and T$_2$. Runoff model calculated until 2006. Full symbols give initial values.

In Fig. 1 data measured [5-8], are presented together with trend lines for these data points. For the year 2000 a good fit of the data is achieved for lakes Vorsee and Lugano. For Lake Lugano activity concentrations given at the same sampling date are for different water depth, and they increase in value with water depth. For Lake Constance measured values are a factor 10 smaller than fitted values. It is known [9, 10] that the catchment of Lake Constance is rich in clay minerals, e.g. illite, which can fix ^{137}Cs, so that activity concentration of runoff water is smaller for this catchment as compared to the catchments of the other two lakes. The initial decline in contamination in lakes is determined not by runoff but by in-lake processes, thus $\ln 2/k_1$ is different from the experimental T_1, which is the lake Cs residence time.

2.2 Lake model

This model [1-3] takes into account runoff from the catchment, outflow and sedimentation of suspended particulates. By consideration of mass balance, we can write a differential equation for C_L (Bq·m^{-3}), the total lakewater concentration of the radionuclide, dissolved in the water and adsorbed to suspended particulates:

$$\frac{dC_L}{dt} = \frac{C_R}{\tau_w} - \frac{C_L}{\tau_w} - \frac{C_L}{\tau_L} - \lambda C_L \qquad (2)$$

C_R (Bq·m^{-3}) is the activity concentration of inflowing streams as estimated by Equation (1). τ_w the water residence time constant is defined under the assumption that discharges of water through the outflow and inflow are about the same. τ_L the residence time constant for uptake by sediments represents transport to the sediments via attachment to and settling of suspended particulates, diffusion is not considered. λ (y^{-1}) is the radioactive decay constant.

The model for prediction of ^{137}Cs in lake water is [1-3]:

$$C_L = C_L(0)e^{-(K+\lambda)t} + \frac{De^{-\lambda t}}{\tau_w}\left[\frac{\alpha(e^{-k_1 t} - e^{-Kt})}{K - k_1} + \frac{\beta(e^{-k_2 t} - e^{-Kt})}{K - k_2} + \frac{\gamma(e^{-k_3 t} - e^{-Kt})}{K - k_3}\right] \qquad (3)$$

$C_L(0)$ (Bq·m^{-3}) is the initial mean lakewater concentration (shown in Fig. 1 and Fig.2 as open symbols). α, β, γ, k_1, k_2, k_3 are the same parameters and have the same values as in the runoff model above. D is the deposition to the catchment, also as in the runoff model above. K, $C_L(0)$ and τ_L are given by:

$$K = 1/\tau_w + 1/\tau_L \qquad\qquad C_L(0) = \frac{D_L}{\delta} \qquad\qquad \tau_L \approx \delta / f_p v_p \qquad (4)$$

where D_L (Bq.m^{-2}) is the deposition per square metre of lake surface, δ (m) is the mean depth of the lake, v_p (m·y^{-1}) is the particulate settling velocity, f_p is the dimensionless fraction of activity sorbed to suspended particles, determined by K_d (m^3·kg^{-1}), the solids-aqueous distribution coefficient, and s (kgm^{-3}) the suspended solids concentration as

$$f_p = \frac{sK_d}{1 + sK_d}.$$

Measured data and input values to the model are collected in table 2

In Fig. 2 the result of the lake model calculation is presented together with measured values. Especially for Lakes Constance and Lugano, agreement between measurement and modelling is satisfying . For Lake Vorsee calculated activity concentrations are too small by factor 3. The values of K_d in table 2 are measured with respect to the exchangeable ^{137}Cs [11, 12]. For the chosen values of parameters the lake

model underestimates the contamination. For Lake Vorsee a reported seasonal redissolution of ^{137}Cs from the sediment [13], not considered in the model, could add to this underestimation. It is known that ^{137}Cs is bound tightly in the sediment of Lake Constance [10], whereas less tightly in that of Lake Lugano [14].

Table 2: Characteristics of lakes studied

Properties	Lake Constance	Lake Vorsee	Lake Lugano
Lake surface (km²)	572	0,09	48,9 (total)
Max. water depth (m)	254	2,2	288
Catchment area A_c (m²)	11487 E+06	1,27 E+06	615 E+06
Potassium concentration [K⁺] (10^{-6} Eq./l)	25	50	10

Values used in the model	Lake Constance	Lake Vorsee	L. Lugano South. Bas.
Mean water depth δ (m)	85	0,6	55
Time constant τ_w (a)	4,1	0,24	2,5
Deposition on lake surface D_L (kBq/m²)	17	28	24
Suspended solids concentration s (kg.m⁻³)	0,001	0,005	0,0007
Particulate settling velocity v_p (m. a⁻¹)	2370	401	401
Distribution coefficient K_d (m³.kg⁻¹)	33	1	5
Fraction of activity sorbed to particles f_p	0,032	0,005	0,0035
Rate K (a⁻¹)	1,14	7,5	4,25
Initial activity concentration $C_L(0)$ (Bq/m³)	200	46667	400
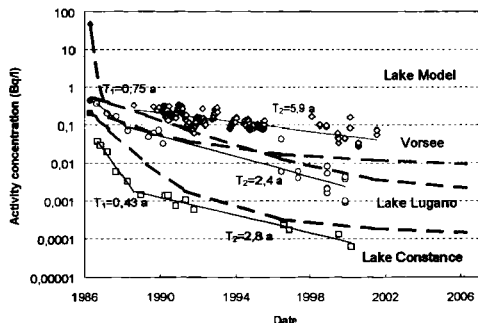
Figure 2: Time-dependency of ^{137}Cs activity concentration of water in three prealpine lakes, measured data with trend lines and ecological half-lives T_1 and T_2. Lake model (dashed) calculated until 2006. Full symbols give initial values.

2.3 Comparison of ecological half-lives of European lakes

Smith et al. [2, 3] compared the time-dependency of the ^{137}Cs activity concentration in water of several European lakes. In Fig. 3 some of their data and the results for the three lakes of this study are given for the ecological half-lives T_1 and T_2 for 17 and 20 lakes respectively. Fig. 3 indicates that ecological half-

lives of lakes are distributed log-normally and geometric mean values are $T_{1gm} = 0,41$ years and $T_{2gm} = 2,7$ years. Deviations from the log-normal distribution for small T_1 are probably due to overlap with ecological half-times belonging to a time window of even smaller values. These values are not included in the geometric mean values given in Fig.3.
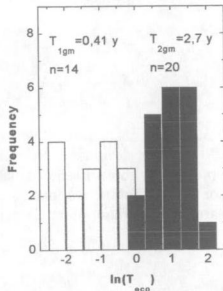

Figure 3: Frequency distribution of the logarithm of ecological half-lives T_{eco} and geometric mean values T_{1gm} and T_{2gm} of the ^{137}Cs activity concentration in European lakes [1-7].
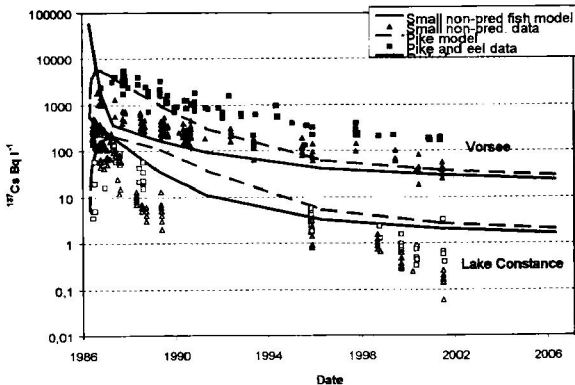
2.4 Fish contamination in Lake Constance and Lake Vorsee

Figure 4: Time-dependency of ^{137}Cs activity concentration of fish in 2 pre-alpine lakes. measured data and fish model blind prediction until 2006.

On the basis of the "lake model", Smith et al. [15] proposed a "fish" model which takes into account that the fish-water concentration factor of ^{137}Cs is inversely proportional to potassium concentration as the uptake rate of ^{137}Cs in fish is negatively correlated with lake potassium concentration. We used values for potassium and other parameters as given in table 1 and our data [8, 10] for a blind prediction of fish contamination in Lake Constance and Vorsee shown in Figure 4. Again, as in the case of lake water, the model underestimates the contamination of fish in Lake Vorsee and overestimates that in Lake Constance. However, deviations are smaller than a factor 10, and the general trend of our measurements is well reproduced by the model.

3. SUMMARY

Simple models allow to blindly predict the ^{137}Cs activity concentration of water and fish of different lakes as a function of time for about 15 years since 1986 with a deviation from measured values smaller than factor 10. Results of these models are useful to predict the contamination of lake water and fish for other lakes, and for the calculation of the distribution of ^{137}Cs in bottom sediments [13]. Ecological half-lives of ^{137}Cs in lake water of about 20 European Lakes are distributed log-normally in several time windows.

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