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Ecological half-times of ^{137}Cs and ^{90}Sr in forest and freshwater ecosystems

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Abstract. More than 200 data sets from the literature of measured time series of activity concentrations and transfer factors concerning: (1) soil, green plants, mushrooms, and game in forests and (2) water, particulate matter, and fish in rivers, reservoirs and lakes in Europe have been analyzed. We used a sum of up to 3 exponential functions to evaluate ecological half-times (T_{eco}).

In *fresh water systems* we fitted lognormal distributions of T_{eco} for ^{137}Cs (e.g. in the water of 32 European lakes, $T_{eco1} = (0.3*2.2^{\pm 1})$ years and $T_{eco2} = (5.7*3.9^{\pm 1})$ years as geometric mean value and standard deviation). Pronounced seasonal variations were observed in suspended matter of Lake Lugano, reflecting the bio-mass cycle of this lake.

In *forests*, ecological half-times for ^{137}Cs can also be fitted by a lognormal distribution. For Central Europe $T_{eco} < 12$ years. Extraordinarily high ^{137}Cs activity concentrations were observed recently in wild boar in Germany. For Eastern Europe the physical half-time is dominant and in some components of the ecosystem the activity concentration is still increasing.

1. INTRODUCTION

The time-dependency of the radionuclides ^{137}Cs and ^{90}Sr in ecosystems can be characterized by the ecological half-time (T_{eco}), which is an integral quantity combining all processes which cause a decrease of radioactivity in a medium besides physical decay. Examples of such processes are leaching, fixation, resuspension and others. These processes are specific for the medium considered (e.g. in soil the leaching of radionuclides into deeper soil horizons, or in lakes the sedimentation of radiocuclides attached to suspended matter in lake water). This report is based on parts of a recent data evaluation [1].

2. METHODOLOGY

2.1 Fit-function

The general expression used to describe the time-dependency of the activity concentration A(t) in Bq/kg is the following:

$$A = \exp\left(-\ln 2 \cdot \frac{t}{T_{phys}}\right) \cdot \left(\sum_{n=1}^{3} a_n \cdot \exp\left(-\ln 2 \cdot \frac{t}{T_{eco_n}}\right)\right) \cdot \left(1 - b \cdot \sin(\omega \cdot t + \varphi)\right) \quad (1)$$

where T_{phys} is the physical half-time, which is 30.07 years for ^{137}Cs and 28.79 years for ^{90}Sr, and T_{eco} is the ecological half-time. The sum of the inverse of these half-times gives the definition of the effective half-time defined as

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$$\frac{1}{T_{eff}} = \frac{1}{T_{eco}} + \frac{1}{T_{phys}} \tag{2}$$

Uncertainties of ecological half-times and amplitudes are determined from error propagation of the uncertainties of the fit parameters.

The sinusoidal function in (1) is used to describe seasonal effects (e.g. as observed in lake water and in suspended matter). A sum of up to 3 exponential functions is generally sufficient to describe the time scales after a spike contamination which can be characterized by an outflow from the medium within days, fixation processes within years, and an approach to the equilibrium within decades [2, 3].

An accumulation of radionuclides in a medium is taken into account in (1) by using an additional factor in order not to modify the structure of formula (1)

$$(1 - e^{-\lambda_0 \cdot t}) \tag{3}$$

2.2 Fitting procedure

Empirical data on ecological compartments are usually log-normally distributed. The probability density function of the log-normal distribution is defined as

$$f_{log}(x) = \frac{1}{\sqrt{2\pi} \cdot \sigma \cdot x} \cdot e^{-(\ln x - \mu)^2 / 2\sigma^2}, x > 0 \tag{4}$$

This is the normal distribution of the logarithms of the quantity x with median μ ($\exp(\mu)$ is the geometric mean of the empirical data x) and σ is the standard deviation of the distribution of the "ln(x)" values, which is calculated as

$$\sigma = \sqrt{\frac{\sum_{i=1}^{n} (\ln x_i - \mu)^2}{n-1}} \tag{5}$$

If the measured values are distributed log-normally and if their standard deviation is known then the fit of the exponential function to the empirical data can be achieved with the maximum likelihood method which gives for a statistical sample the probability to obtain values x_i as

$$\prod_{i=1}^{n} p_\pi(x_i) = p_\pi(x_1) \cdot p_\pi(x_2) \cdot ..... \tag{6}$$

because of the assumed independence of empirical data. This expression can be maximized with respect to parameter π. In the case of the lognormal distribution function π is μ and σ.

In the software used in this work (STATISTICA 5.5, StatSoft, USA) the maximum of the logarithm of this probability, the log-likelihood function, was determined

$$L(x_1, ..., x_n, \pi) = \log(\prod_{i=1}^{n} p_\pi(x_i)) \tag{7}$$

Then the product of probabilities becomes a sum in the log-likelihood function in which π was determined in a way to make L a maximum.

If the variability of empirical data points is unknown (e.g. if only one data point per year was measured) then a least square fit of up to 3 exponential functions, un-weighted or weighted, was applied. However, in principle this method is allowed only when empirical data points are distributed normally.

3. RESULTS FOR ^{137}CS

3.1 Forest ecosystem

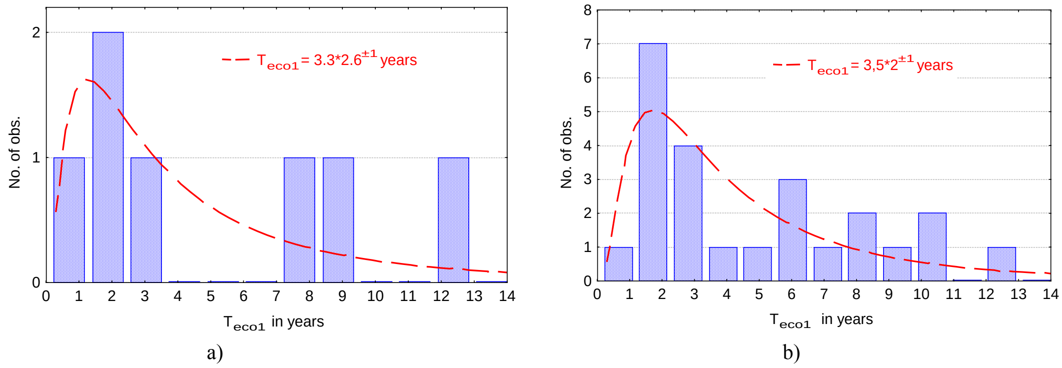

Figure 1. Ecological half-times (T_{eco1}) for ^{137}Cs in (a) roe deer, and (b) in wild boar, chamois, grazing plants and *Xerocomus badius.*

In Fig. 1a, ecological half-times (T_{eco1}) for ^{137}Cs in roe deer from different forests in Austria and Germany are distributed in the range of 1 to 13 years. Different values in T_{eco1} are due to differences in ecosystem (e.g., soil type, climate, season, contamination level, etc.) and/or differences in fitting procedure (e.g., if 1 or more exponential functions are used). In Fig. 1b, ecological half-times (T_{eco1}) for ^{137}Cs in wild boar, deer, chamois, grazing plants and *Xerocomus badius* are shown. They lie in the same time window as those of roe deer shown in Fig. 1a. The empirical data can be described by a log-normal distribution with a median and standard deviation of $T_{eco1} = 3.5{*}2^{\pm1}$ years.

An example for the time-dependency of ^{137}Cs in wild boar from Bavaria, Germany, is shown in Fig. 2a. The median within each year is decreasing with $T_{eco1} = (10.5 \pm 1.6)$ years but the variability within the year is increasing by about 2 magnitudes.

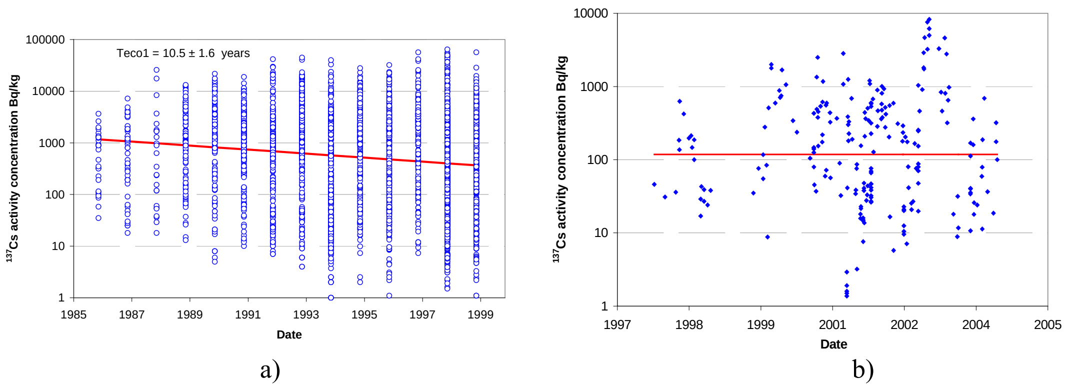

Figure 2. ^{137}Cs in wild boar from Germany a) Bavaria; b) Bad Waldsee.

Data from more recent years were taken in the forest Bad Waldsee in Fig. 2b where the increase in variability with time is confirmed. The increase in the first half of the year 2003 in Fig. 2b is probably caused by the increased consumption of deer truffles, *Elaphomyces Granulatus Fr* [6].

Concerning forests of Eastern Europe few data sets were accessible (e.g. in Ukraine for ^{137}Cs in roe deer T_{eco1} = (128 ± 123) years; for mushroom 2 years < T_{eco1} < $5*10^5$ years; for spruce bark 9 years < T_{eco1} < 14 years; and in spruce needles, shots and hard wood ^{137}Cs activity concentrations are still increasing with time [1]).

3.2 Freshwater ecosystem

Ecological half-times have been determined and are collected for 31 European lakes in Fig. 3. In Fig. 3, the logarithms of the empirical values of T_{eco} are described by a normal distribution and median and standard deviation of T_{eco} are given

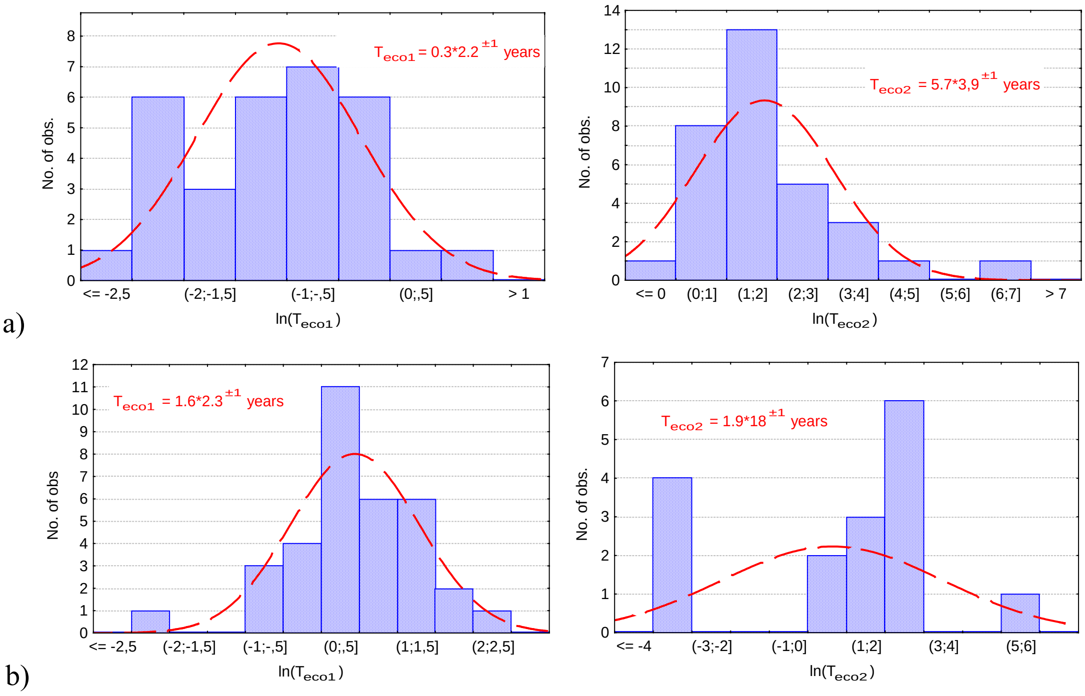

Figure 3. Ecological half-times of ^{137}Cs in (a) lake water and (b) fish.

An example for the ^{137}Cs activity concentration in water and fish of a small eutrophic lake is given in Fig.4. The oscillations in ^{137}Cs activity of the water are caused by radionuclide cycling between sediment and water body, based on ammonium production and caesium release in the sediment due to microbial decomposition of organic matter and a flux of ^{137}Cs incorporated into organic detritus into the lake [4].

In [5] a model is presented to describe the time dependency in water and fish. In this model ^{137}Cs activity concentration of lake water is described and predicted by a simple exponential "transfer function" model for the runoff of activity from contaminated catchments. Furthermore lake water residence times and ^{137}Cs uptake by sediments are taken into account. The fit-function used in this work follows from these ideas [2, 3, 5].

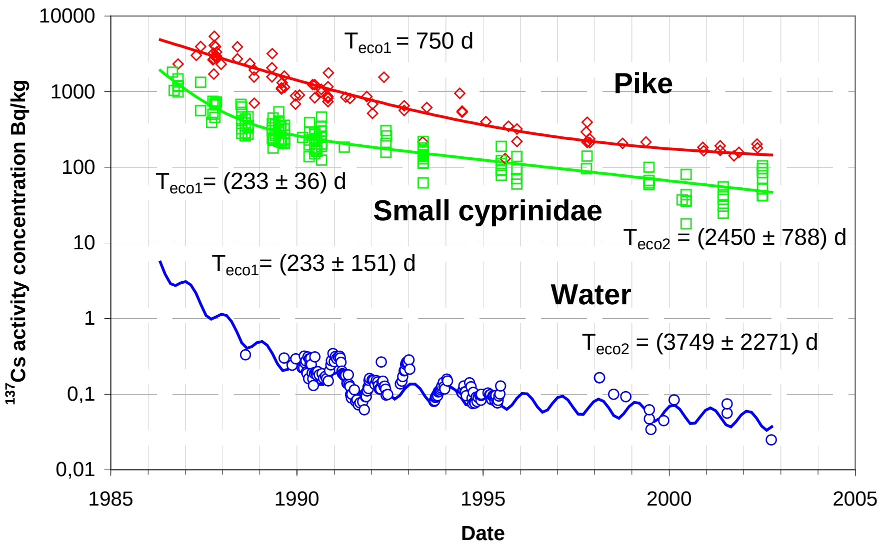

Figure 4. ^{137}Cs activity concentration in lake water and fish from Lake Vorsee, Germany.

In Fig. 5, empirical values of the ^{137}Cs activity concentration of seston of Lake Lugano, Switzerland, are shown for the time before and after the accident in Chernobyl, resulting in a factor of 10^3 increase in activity concentration of the seston in May, 1986. The seston was trapped in a depth of 79 m, about 6 m above ground. A modulation of the activity concentration can be seen with a period of 1 year and a phase which is about the same as in the lake water in Fig. 4. Only the empirical data points after May 1, 1986 are described by the fit. Standard deviations of T_{eco} could not be determined for this fit because of strong correlations between the parameters used.

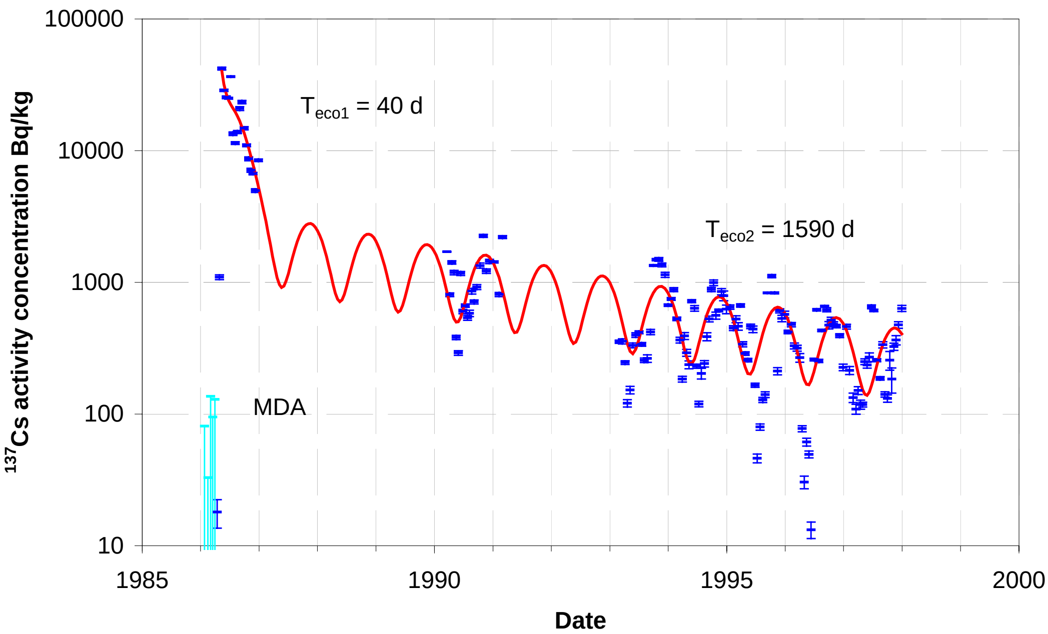

Figure 5. ^{137}Cs in seston taken in a depth of 79 m in Lake Lugano, Melide, Switzerland.

4. RESULTS FOR ^{90}SR

Time series in forests were accessible only for antlers of deer from Austria [T_{eco1} = (22 ± 4) years] [1]. Concerning fresh water only original data from lake water [T_{eco1} = (2 ± 2.1) years and T_{eco2} = (19 ± 48) years] and fish-bone [T_{eco1} = (6.3 ± 1.5) years] from Lake Berdenish, Russia, were accessible. Data for lakes in Italy (7 years ≤ T_{eco2} ≤ 93 years), rivers in Belarus, Russia and Ukraine (6 years ≤ T_{eco2} ≤ 25 years); reservoirs in Russia and Ukraine (8 years ≤ T_{eco2} ≤ 19 years) were taken from the literature [1].

5. CONCLUSIONS

In freshwater systems, a sum of up to 3 exponential functions can be used to evaluate ecological half-times (T_{eco}) of ^{137}Cs. Time-series of at least one decade for T_{eco2} and another one for T_{eco3} are needed to achieve a thorough description. Pronounced seasonal variations of ^{137}Cs activity concentrations are observed for lake water and suspended matter. T_{eco} for lake water and fish are described by lognormal distributions.

In forests, T_{eco1} of ^{137}Cs can be described by a lognormal distribution. For Central Europe, T_{eco1} < 12 years. In wild boar, the variability of ^{137}Cs activity concentration – in 1998 it varied over 5 orders of magnitude in Bavaria, Germany- is still increasing with time.

Concerning ^{90}Sr, very few data sets are available. Due to the higher mobility of ^{90}Sr as compared to ^{137}Cs, more data sets are urgently needed to give advice to the population in order to minimise doses from ^{90}Sr.

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References

[1] G. Pröhl, I. Fiedler, E. Klemt, G. Zibold, S. Ehlken, Erfassung ökologischer Halbwertszeiten von ^{90}Sr und ^{137}Cs in terrestrischen und aquatischen Ökosystemen, Abschlussbericht St.Sch. 4276, Bundesamt für Strahlenschutz, 2004.

[2] J.T. Smith, D.R.P. Leonard, J. Hilton, and P.G. Appleby, *Health Phys.* **72**, 880-892 (1997).

[3] L. Monte, *J. Environ. Radioactivity* **26,** 71-82 (1995).

[4] S. Kaminski, T. Richter, M. Walser, G. Lindner, Redissolution of cesium radionuclides from sediments of freshwater lakes due to biological degradation of organic matter. *Radiochimica Acta* **66/67** (1994) pp. 433-436.

[5] G. Zibold, S. Kaminski, E. Klemt & J. T. Smith, Time-dependency of the ^{137}Cs activity concentration in freshwater lakes, measurement and prediction. Proceedings of the international congress on the radioecology of continental and estuarine environments. *Radioprotection-Colloques*, **37**, C1-75-80 (2002).

[6] Putyrskaya V., Klemt E., Paliachenka H., Zibold G. Accumulation of ^{137}Cs in elaphomyces granulatus fr. and its transfer to wild boar. XXXIII ESNA Conference Proc. Viterbo, Italy 27.08.03- 31.08.03 Working Group 3: Soil-to-Plant-Relationships, N. Mitchell, V. Licina & G. Zibold (Eds.) ISSN 1611-9223, pp. 1-5, Weingarten, 2003.