Predicting transfers of ^{137}Cs in terrestrial and aquatic environments: A whole-ecosystem approach

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Abstract. It is well known that during the years after a nuclear accident the bioavailability and environmental mobility of radionuclides may change significantly, resulting in significant changes in contamination of foodstuffs and surface waters. Studies on ^{137}Cs and, to a lesser extent, ^{90}Sr, have quantified these changes in some ecosystems. However, variability in temporal changes of these radionuclides in aquatic and terrestrial systems is not yet well quantified. Estimation of such variability is a key component of any predictive model for long-term transfers of radionuclides in the environment. We have analysed measurements (from both weapons testing and Chernobyl deposits) of ^{137}Cs in runoff waters and in aquatic and terrestrial foodstuffs in order to determine temporal changes in their bioavailability and mobility. Using these empirical data, and the results of parallel modelling studies of vertical migration of radiocaesium in soils, we have quantified the relative importance of transport processes of ^{137}Cs, as compared to slow changes in its chemical availability in the soil. On the basis of these results, we have developed simple models for predicting time changes in activity concentrations of ^{137}Cs in surface water, foodstuffs and the human body during the years to decades after radioactive fallout. Importantly, we also determine uncertainties in model parameter estimates, and highlight the potential causes of this uncertainty.

1. INTRODUCTION

1.1 Pre-Chernobyl studies

Prior to the Chernobyl accident, much was already known about the movement of radionuclides (radiocaesium and radiostrontium in particular) in the environment. Numerous studies had been carried out of the environmental and foodchain transfer of global fallout from the above-ground nuclear weapons tests of the 1950's and 1960's. Laboratory studies had shown that the sorption of ^{137}Cs by soils and sediments is dominated by specific sorption to certain sites on illitic clay minerals [1]. This sorption was known to have a slow kinetic component, in which ^{137}Cs is transferred to less available sites in the mineral lattice [2], a process often referred to as "fixation". Caesium competes for binding sites on the clay mineral lattice with similarly sized K^+ and NH_4^+ ions. Thus, a common measure of the "availability" or "exchangeability" of radiocaesium sorption to soils and sediments is to carry out an extraction with ammonium acetate or ammonium chloride solution. Coughtrey & Thorne [3] report measurements by Evans & Dekker that a large proportion (around 85%) of radiocaesium in soils was in fixed form with only 15% exchangeable with a molar solution of ammonium acetate, though this fraction varied considerably with soil type. Environmental studies of global fallout from nuclear weapons testing also showed that transfers of radiocaesium to milk were much higher in the Faroe Islands than in Denmark. This was attributed to greater availability of ^{137}Cs in the organic soils of the Faroe Islands than in more mineral soils in Denmark [4].

Reports by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [5,6] documented activity concentrations of ^{137}Cs in various foodstuffs and in the human body in different countries during and after the period of weapons test fallout (UNSCEAR 1977; 1982). These data highlighted the relatively very high transfer of ^{137}Cs to humans living in arctic environments confirming the hypothesis of Aarkrog [4] that some environments were much more sensitive to high radiocaesium transfers than others.

Studies were also carried out of the change in ^{137}Cs activity concentrations over time after weapons test fallout. Aarkrog [7] observed an exponential decline in ^{137}Cs activity concentrations in milk and rye with half life 1-2 years. In a study of radiocaesium levels in humans during the weapons test fallout period, Newton and coworkers [8] tentatively hypothesised a pool of bio-available ^{137}Cs in the environment which declined even more slowly, with half-life 7.5 to 30 years.

1.2 Post -Chernobyl studies

During the weeks to months after Chernobyl, radiocaesium activity concentrations in both vegetation and surface waters were determined by short term processes. Activity concentrations in plants are determined by interception and washoff rates of the initial fallout, as well as uptake by the roots (e.g. [9]). Similarly, in rivers and lakes, activity concentrations are initially high as a result of direct deposition to the water surface and rapid runoff of ^{137}Cs before it is sorbed to catchment soils [10]. Activity concentrations then decline over a period of weeks to months as a result of reduced runoff from catchments and, for lakes, loss of ^{137}Cs through the outflow and deposition to bottom sediments.

On long time scales (years), the processes which determine radiocaesium transfers to and from many different ecosystem components (for example, between plants and herbivourous mammals) are fast in comparison with the slow decline in radiocaesium availability in soil. Thus, the change in radiocaesium activity concentration in the main environmental compartments should be controlled by slow changes in its soil-soil solution partitioning. To test this hypothesis, Smith *et al.* [11] analysed many long term field studies of temporal changes in radiocaesium in three different ecosystem components: vegetation, surface waters (dissolved phase), and milk following the Chernobyl accident. Over the first five years after

Chernobyl, these workers observed similar rates of decline of ^{137}Cs in all three ecosystem components, all having effective ecological half lives (the time taken for the radioactivity to reduce by one half by physical decay and environmental processes) in the range 1-4 years, with mean approximately 1.7 years.
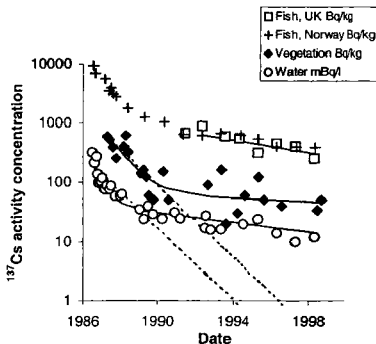
Figure 1: Long-term changes in ^{137}Cs in brown trout, Norway (data from ref. [12]), perch, terrestrial vegetation and water in Cumbria, UK. The decline in ^{137}Cs in immature fish, water and vegetation during the first five years has an effective ecological half life (T_{eff}) of 1 - 4 years [11,12] as a result of "fixation". The dotted lines demonstrate a hypothetical continuation of irreversible fixation. Fits of the two exponential model to our data, indicating reversible "fixation" are shown as solid lines. Long-term declines in all three Cumbrian systems are similar (T_{eff} = 6-30 years), and are in quantitative agreement with the Norwegian fish data [12,13].

However, as shown in Figure 1, recent data [12,13] show a two-component exponential decline (i.e. of form: $C = C_1 e^{-\alpha_1 t} + C_2 e^{-\alpha_2 t}$) in Chernobyl-derived ^{137}Cs activity concentrations in surface water, terrestrial vegetation and fish. The observations show that the effective ecological half-life in young fish, water and terrestrial vegetation has increased from 1 - 4 years during the first five years after Chernobyl to 6 - 30 years in recent years. From the observed persisting mobility of radiocaesium in the environment, and particularly the increase of T_{eff} towards the physical decay rate of [137]Cs ($T_{1/2}$ = 30.2 years), it was concluded [12] that the sorption-desorption process of radiocaesium in soils and sediments is tending towards a reversible steady–state. These results were consistent with the tentative hypothesis of Newton and coworkers [8] that there was a long-term component to the decline in ^{137}Cs activity concentrations in foodstuffs.

1.3 Comparing the behaviour of weapons test and Chernobyl fallout

The weapons test fallout occurred at differing rates over a number of years whilst the vast majority of the Chernobyl fallout was deposited during only a few days. The different input functions of the two sources thus makes direct statistical comparisons difficult. Recently, Smith and coworkers [14] have developed a method, using SAS statistical software (SAS Institute Inc.), to incorporate the time-dependent behaviour of weapons-test fallout into a statistical curve-fitting routine. This allowed comparison of the mobility of ^{137}Cs [14] and ^{90}Sr [15] from weapons test and Chernobyl fallout events. Models for the runoff of ^{137}Cs and ^{90}Sr in Finnish catchment calibrated using data from the pre-Chernobyl period were shown to give good predictions of runoff after the Chernobyl fallout. Thus, when the different input functions of the two sources was accounted for, their mobility was found to be similar.

1.4 Purpose of the present study

The purpose of the present study is to use historical data to further quantify the time dependence in ^{137}Cs and mobility following the weapons test and Chernobyl fallout events. We will carry out an example uncertainty analysis to illustrate the major sources of error in models to predict activity concentrations in the human body. The model we develop is not novel, being similar in principle to models applied previously (e.g. [5,8,10]). Rather it represents an attempt to generalise model parameters: we will show that the time dependence of radiocaesium activity concentrations is relatively invariant between different environmental conditions and ecosystem components. We will also show that parameters describing this time dependence are quantitatively applicable to both Chernobyl and weapons test fallout events.

2. METHOD

The concentration of the radionuclide in an ecosystem component, $C_x(t)$ (Bq m^{-3}) resulting from a "spike" (Chernobyl-type) deposition, D (Bq m^{-2}) is given by:

$$C_x(t) = D.T_x(A.\delta_0 + B.e^{-(\lambda+k_1)t} + C.e^{-(\lambda+k_3)t}) \qquad (1)$$

where, A, B and C are dimensionless coefficients representing, respectively, the initial rapid decline in activity concentrations as a result of short term processes, the slow decline as a result of soil fixation processes, and the very long term (possibly also slowly declining) fraction as the activity concentration tends to an equilibrium value. λ is the radioactive decay constant (0.023 y^{-1} for ^{137}Cs). The "delta function", δ_0, models the initial transfer of recently deposited activity. It causes the coefficient, A, to have a positive value during the first year following deposition, and value zero thereafter. The parameter k_2 represents an exponential decline in runoff over the first few years following the deposition as a result of fixation processes of the radionuclide to catchment soils [11]. The parameter k_3 represents a longer term decline over a period of decades. Since Eq. 1 is not a simple exponential function, the effective ecological half life (T_{eff}), calculated as the time for the activity concentration in an ecosystem component to halve, is strongly dependent on the time period after fallout over which the calculation is made.

The parameter T_x m^2 kg^{-1} represents the variation in activity concentration at a specified time, for ecosystem component, x, where x represents, for example, milk or meat or drinking water. Variation in T_x changes the predicted activity concentration of an ecosystem component, but does not influence the time dependence in activity concentrations. It is analogous to an aggregated transfer factor, but is time invariant.

For a time-dependent input function, with fallout input occuring over several years, such as that resulting from weapons testing, the activity concentration of the radionuclide in the ecosystem component in year j after the beginning of fallout, $C_x(j)$ (Bq m^{-3}), is given by:

$$C_x(j) = A.T_x.D(j) + \sum_{i=1}^{i=j} D(i).T_x(B.e^{-(\lambda+k_2)(j-i)} + C.e^{-(\lambda+k_3)(j-i)}) \qquad (2)$$

where $D(j)$ (Bq m^{-2}) is the deposition during year j. For weapons test data, $D(j)$ was estimated for different latitudes using data from [5], the temporal pattern of deposition being estimated from data in [16]. Eq. 1 is a special case of Eq. 2 whereby fallout $D(j)$ is assumed equal to zero in all except one year: this special case was used for the post Chernobyl data.

3. RESULTS AND DISCUSSION

Estimates of the parameters to predict the temporal variation in ^{137}Cs (A, B, C, k_2, k_3) were made by fitting Eqs. 1 and 2 to measurements of weapons test and Chernobyl radioactivity in rivers [14,15]. It is hypothesised that changes in average annual activity concentrations with time in different ecosystem components (e.g. milk, meat, human body) may be predicted using these parameter values. Clearly, this hypothesis does not apply (on short time scales) to ecosystem components which have relatively slow uptake and release of the radionuclide, for example, trees and predatory fish for ^{137}Cs or the human body for ^{90}Sr. For these components, the above models would need to be modified to account for slow uptake for such ecosystem components (e.g. [17]), at least for predictions in the first years after fallout.

Parameter		Best estimate	Estimated error and distribution of errors
A		0.0145	Lognormal distribution, 2 S.D. = log(3)
B		0.019	Lognormal distribution, 2 S.D. = log(3)
C		0.001	Lognormal distribution, 2 S.D. = log(3)
k_2	d^{-1}	0.5	Lognormal distribution, 2 S.D. = log(2)
k_3	d^{-1}	0.05	Equal probability of any value from 0–0.07

Table 1: Parameters describing the decline in ^{137}Cs availability over time following deposition and their estimated errors. A lognormally distributed error with 2 standard deviations = log(3) approximates a factor of 3 error range either side of the mean, 2 S.D. = log(2) approximates a factor 2 range either side of the mean.

Although it is hypothesised that time changes in ^{137}Cs may be similar in many different ecosystem components, it is well known that activity concentrations vary considerably as a result of environmental factors and different bioaccumulation in different ecosystem components. Thus the parameter T_x is expected to vary between different ecosystem components, and (for a given ecosystem component) according to environmental conditions [4].

3.1 ^{137}Cs in milk

We have fitted Eq. 2 (with A, B, C, k_2, k_3 parameters as in Table 1) to measurements of ^{137}Cs in milk in West Germany, the USA and Denmark [5,6] following weapons testing. ^{137}Cs activity concentrations in milk (per unit of fallout) in these three countries were quite similar, so a single value of T_{milk} (= 0.1 m^2 kg^{-1}) fitted all three data sets with reasonable accuracy (R^2 = 0.87). Transfers to milk in the Faroe Islands
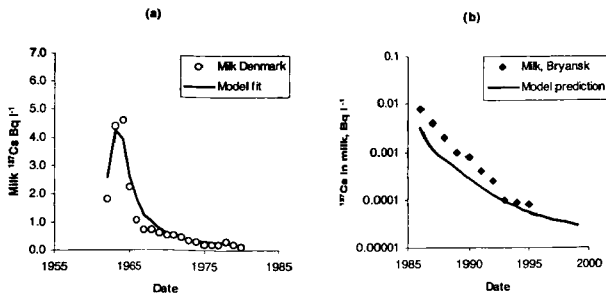

Figure 2: (a) Example of model fit to measurements of ^{137}Cs in milk in Denmark [5,6]; (b) model "blind" prediction of ^{137}Cs activity concentration in milk in the Bryansk region of Russia following Chernobyl (scaled to 1 Bq m^{-2} fallout), [18].

(as demonstrated by [4]) were more than an order of magnitude higher, having T_{milk} = 2.8 m^2 kg^{-1}, though the parameters describing the change in activity concentration over time (Table 1) fitted all the data sets well. It is clear that local environmental conditions will make large differences in ^{137}Cs activity concentrations in milk, but the time dependence of concentrations appears to be relatively invariant to environmental conditions. Further, by using appropriate T_{milk} values, it may be possible to make initial estimates of activity concentrations on a regional scale. In Figure 2(b) we show the results of a "blind" test of the model (developed from weapons test data) against measurements of ^{137}Cs in milk in the Bryansk region of Russia following Chernobyl [18]. For the prediction we assumed T_{milk} equal to that for West Germany, USA and Denmark as appropriate for the mineral soils and (in the most part) intensive agriculture in the Bryansk region.

3.2 ^{137}Cs in the human body

We have fitted Eq. 2 (with A, B, C, k_2, k_3 parameters as in Table 1) to measurements of ^{137}Cs in humans in West Germany, the USA, France, Japan, Denmark, Moscow, UK (West Cumbria and South England) [5,6,8] following weapons testing. Measurements from all of these locations could be fitted reasonably well (R^2 = 0.83, Fig. 3(a)) by a single value of T_{human} = 0.33 m^2 kg^{-1}. This parameter value also gave reasonable fits for Southern Hemisphere countries, Argentina and Australia. The southern parts of Scandinavian countries (Finland, Sweden, Norway), however, had a significantly higher value, T_{human} = 0.84 m^2 kg^{-1}, and "arctic" populations (Alaska, Murmansk, Finnish reindeer herders) two orders of magnitude higher still (T_{human} = 89.5 m^2 kg^{-1}). In most cases, data was only available for the period 1964-76, therefore slightly missing the peak of weapons test fallout in 1963.
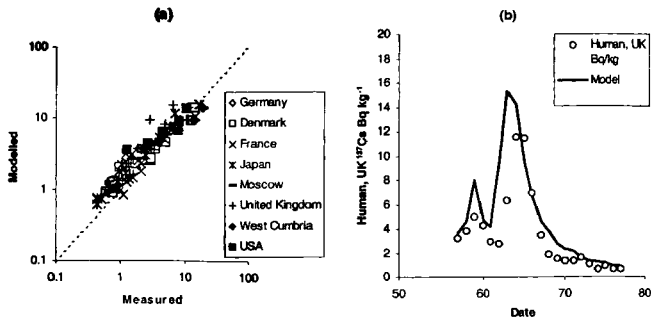

Figure 3: Model fits against measurements of ^{137}Cs in the human body (Bq kg^{-1}): (a) different "temperate" countries; (b) time series data for the UK.

Data from the UK [8], however, was available for the period 1957-77 (Fig. 3(b)) showing the "lag" of 6 months to 1 year between activity concentrations in humans and the maximum in fallout, as observed by [8]. This was believed to be due to a combination of "delays in the distribution of foodstuffs and the buffering effects in the turnover of radiocaesium by animals" [8].

3.3 Uncertainty analysis

We have carried out an analysis to assess the influence of uncertainties in the time dependence of ^{137}Cs availability on the integrated ^{137}Cs activity concentration in the human body over a 50 year period. This quantity determines the 50 year integrated internal dose to humans. Using the estimated errors in the parameters describing the time dependence of ^{137}Cs availability (Table 1) a "Monte Carlo" approach was used to estimate the variation in 50 year integrated ^{137}Cs activity concentration, C_I (Bq yrs kg^{-1}) in humans. Figure 4 shows the results of this uncertainty analysis, illustrating the differences in integrated

activity concentrations between the different populations. The large difference between the distributions for "arctic" populations and the other populations illustrate the greater relative importance of dietary and environmental factors (ie variation in T_{human}) than differences in time-dependence in determining internal dose from radiocaesium. In other words, the largest errors in prediction of doses are likely to be due to uncertainty in the value of T_{human} rather than uncertainty in the time dependent parameters. For example, the long term component of the decline ($C.e^{-(\lambda+k_{j})t}$) only accounts for approximately 20% of the overall

dose. Whilst this component is important for predicting long term contamination of the ecosystem, if the target modelling variable is integrated 50 year dose, it has relatively little influence the outcome compared to uncertainty in T_{human}.
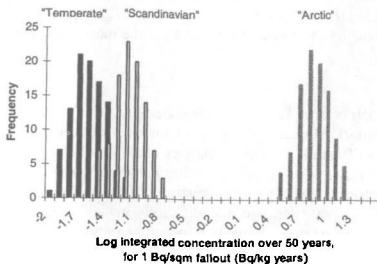
Figure 4: Variation in estimated 50 year integrated activity concentration in the human body following a Chernobyl type "spike" deposition of 1 Bq m^{-2}.

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