^{137}Cs in Soil Profiles in NE Estonia

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Introduction

Radiocaesium contamination in soil was produced by two fallouts: a global fallout caused by nuclear weapons testing in the atmosphere (1950s to 1970s) and a regional one - during/after the Chernobyl accident (1986). The areal distribution of the ^{137}Cs fallout after the 1986 accident was extremely uneven in Estonia. Our previous studies [Realo et al., 1994, 1995] showed that about 60% of its total deposition with the countrywide mean of 2 kBq m^{-2} occurred predominantly in the north-eastern part of Estonia. In this region the maximum deposition approached the values of 40 kBq m^{-2}. This is the reason why soil profiles in this region have been of interest. We have also determined the ^{137}Cs activity concentrations as a function of soil depth during 1991-2000. Attempts have been made to model the found depth-distributions of radiocaesium concentration and to find the possible time-dependent behaviour of these distributions in soil [Realo et al., 1994]. The migration of radiocaesium into soil is dependent on several factors: soil properties, vegetation, mode of deposition, etc. For this reason, averaged distribution parameters for both total and Chernobyl ^{137}Cs have been used for deriving general trends of the migration processes.

Sample collection and preparation

Depth distributions of the ^{137}Cs activity concentration were studied for undisturbed soil profile samples collected at more than 20 locations in NE Estonia in 1998 and 1999. Some of the sampling locations (including also sampling sites from 1991-1993) are listed in Table 1. In this report analysis data are presented together with our preliminary modelling results.

Soil profiles down to a depth of ~ 20 cm were collected by means of a 9 cm diameter steel corer. Soil cores were cut into 2-3 cm thick slices, dried, homogenised and put into metallic beakers. A low-background HPGe gamma spectrometer with a 42% efficiency and 1.7 keV resolution (1.3 MeV) was applied for the analysis of both ^{134}Cs (604 keV) and ^{137}Cs (662 keV). The Chernobyl fallout fractions in the total ^{137}Cs concentrations were calculated using the analysis data for ^{134}Cs.

Table 1. Sampling data of soil profiles.

Soil profile No	Location N/E	Short description	No of the soil profile	Location N/E	Short description
#87	59.202 27.343	Väike-Pungerja	#121	59.264 27.903	Eesti SEJ
#90	59.382 27.84	Sinimäe	#122	59.275 27.914	Eesti SEJ
#94	59.003 27.722	Vasknarva	#123	59.281 27.911	Eesti SEJ
#98	59.448 27.723	Sillamäe	#124	59.298 27.913	Balti SEJ
#117	59.391 28.081	Olgina	#126	59.354 28.071	Balti SEJ
#118	59.329 27.932	Eesti SEJ	#129	59.372 28.104	Soldina
#119	59.405 28.173	Narva	#133	59.351 28.138	Balti SEJ

Results and Discussion

Typically the depth-dependent behaviour of ^{137}Cs activity concentration along an undisturbed soil profile is successfully described by using an exponential distribution of the following type [UNSCEAR, 2000]:

$$C(x) = C_0 \exp(-\alpha x), \qquad (1)$$

where C(x) is the radionuclide concentration (Bq/kg) at depth x (cm), C_0 is the surface concentration C(x = 0) and α (cm^{-1}) is the characteristic coefficient of the distribution, the reciprocal of the relaxation length. Unfortunately, in many cases of long-term radionuclide transport the above simple distribution is a poor approximation to the real situation. When analysing our soil profiles, the same conclusion can be drawn. Considering the radionuclide transport in soil as a diffusion-convection process has lead to multiple models and their modifications, resulting in the depth-distributions of the Gaussian or lognormal types [Isaakson et al., 1998].

In the present report, we use the lognormal distribution of the form:

$$C(x) = \frac{A}{\sqrt{2\pi}\, wx} \exp\left[-\frac{(\ln x - \ln xc)^2}{2w^2}\right], \qquad (2)$$

where its parameters are mean, *ln xc,* and variance, w^2, of the distribution. According to theory these parameters are time-dependent functions of the diffusion coefficient and the mass flow transport velocity, respectively [Konschin, 1992]. In principle, the actual dependence of these parameters on time can be derived from fitting of the depth-distributed activity concentrations in soil profiles collected at different times.

Some typical results on the depth distributions of the measured ^{137}Cs concentration in soil profiles collected in NE Estonia in 1991, 1993, 1998 and 1999 are presented in the Figures 1, 2, 3 and 4, respectively. All activity concentrations are recalculated to the reference date of May 1, 1986. A notable site-specific variability is seen for depth-distributions in profiles collected in different locations.

In the profiles collected in 1991 - 1993 the maximum ^{137}Cs concentration was observed in the top-most layer and no sub-surface maximums were found. Radiocaesium concentrations decrease rather monotonously with increasing depth in these sampled soil profiles and the simple exponential model (1) described above was a satisfactory approximation [Realo et al., 1994, 1995]. The average mass relaxation length values determined for about 25 profiles all over Estonia (incl. those from the NE part) were 42 ± 15 kg m^{-2} (~ 3 cm) and 110 ± 30 kg m^{-2} (~ 10 cm) for the Chernobyl and the nuclear weapons test fallout, respectively.

In almost all profiles collected in 1998-1999 the highest concentrations of the total and Chernobyl radiocaesium were not observed in the top layer, but at the depth of 3 - 5 cm below ground surface. As the sub-surface maximums were found, this feature determined the search for a different model. Notable variations in the depth- dependent concentrations of different soil type, vegetation, water composition, etc. were also found. As a result, we choose the lognormal model (2) and together with the data for the profiles collected, revisited the former depth-distribution data collected in 1991-1993. Both total and Chernobyl ^{137}Cs concentration in profiles were fitted by using the SCIENTIST 2.0 software. As a rule, the lognormal distribution gave a reasonable fit to the measured data. The fitting parameters, xc and w, were determined for each profile and their mean values for the samples collected during one year were calculated.

Figure 5 demonstrates time-dependence of the mean parameters values, xc and w, for total ^{137}Cs in the measured profiles. The same dependence for the Chernobyl ^{137}Cs is presented in Figure 6. A rather weak increase in both parameter values with time is observed for both cases. That means a broadening of the distribution and a shift of its maximum to deeper soil layers in time. For total ^{137}Cs, where both contamination components coexist, a linear fit describes the observed time dependence of the yearly mean parameters in the profiles as follows: xc ~ 0.4 t and w ~ 0.05 t, where t is time (y) and xc is given in cm. For the Chernobyl radiocaesium component a slightly weaker dependence of these parameters on time is obtained: xc ~ 0.24 t and w ~ 0.029 t in the time interval of 1991 - 1999. It is seen that these time-dependencies have not been linear over the whole time period since 1986.

The observed weak time-dependences of the ^{137}Cs depth-distribution parameters demonstrate a rather slow migration rate of radiocaesium to deeper soil layers and its strong retention in the near-surface soil. This conclusion drawn for the neighbouring Nordic countries [Isaakson et al., 1999] supports our former assumption made on comparison of the nuclear weapons test and Chernobyl radiocaesium depth-distributions in Estonian soils [Realo, et al., 1995]. At the same time, shapes of the depth-dependencies in profiles collected in different times shows significant differences.

Our profile results can be further evaluated by means of the other mathematical models describing the time-dependent transport of radionuclides in soil. We have started the compartmental modelling approach, which has been successfully used by other authors for describing the time-dependent activity in different soil layers and predicting the future behaviour [Kirchner, 1998]. For this purpose a simple model has been compiled for modelling using SCIENTIST. In addition, in order to eliminate site-specific variability and to enable better comparison with the model new samplings in the former locations have been performed. At present both modelling attempts as well as analysis of these profiles are in progress.

Conclusions

Most sample profiles collected in NE Estonia in 1998-1999 show a different depth-dependent behaviour of the Chernobyl ^{137}Cs activity concentration in comparison with our results obtained in 1991-1993. Sub-surface maximums of the ^{137}Cs activity concentrations are characteristic for the soil profiles collected in later years, which can not be explained by a monoexponential model any more. Our preliminary modelling has demonstrated that a lognormal distribution can be satisfactorily used for fitting of the found ^{134}Cs / ^{137}Cs (Chernobyl) depth-dependencies and to derive the time-dependent migration parameters in Estonian soils. A weak time-dependence of the fitting parameters confirms slow effective migration rates and a strong retention of radiocaesium in upper soil layers. Additional samplings and analysis for different soil types and locations are in progress.

References

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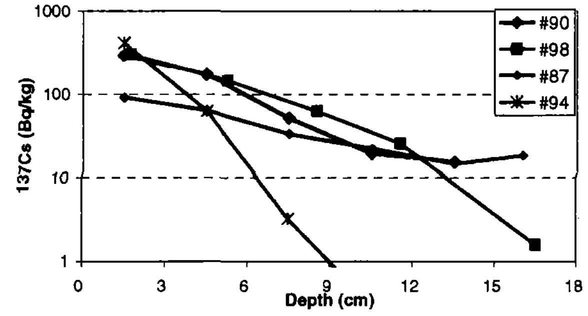

Fig. 1. The vertical distribution of 137Cs in the soils in NE Estonia in 1991.

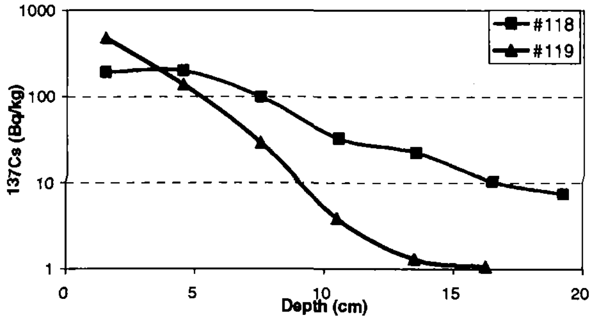

Fig. 2. The vertical distribution of 137Cs in the soils in NE Estonia in 1993.

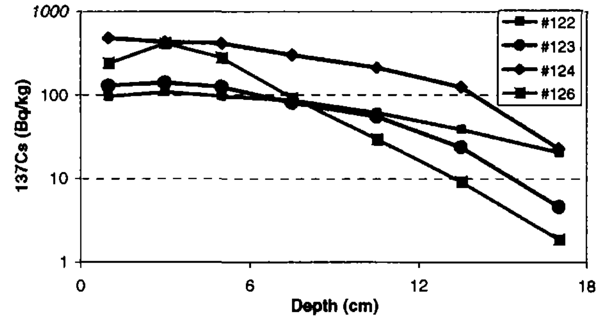

Fig. 3. The vertical distribution of 137Cs in the soils NE Estonia in 1998.

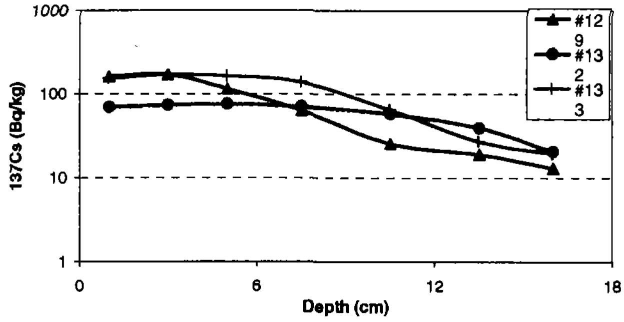

Fig. 4. The vertical distribution of 137Cs in the soils NE Estonia in 1999.

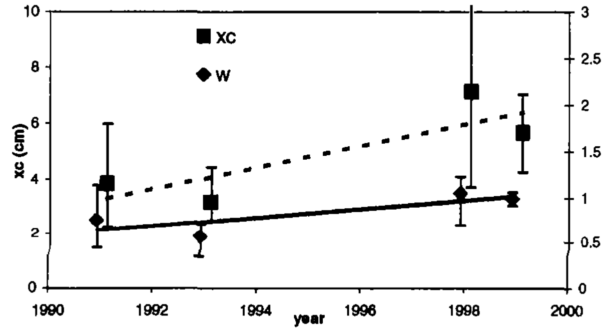

Fig. 5. Mean fitting parameters xc and w for total ^{137}Cs in soil profiles.

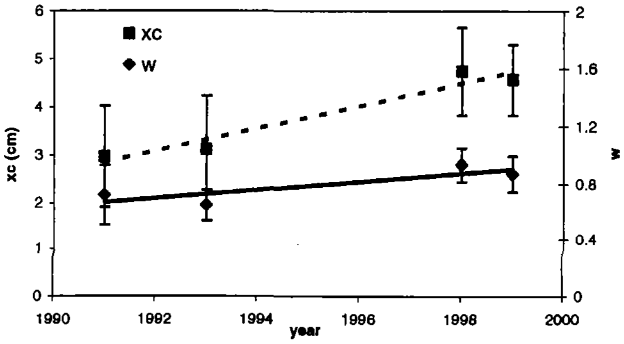

Fig. 6. Mean fitting parameters xc and w for the Chernobyl ^{137}Cs (^{134}Cs) in soil profiles.