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Vertical distributions of ^{137}Cs in soils: a meta-analysis

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Abstract

Purpose. The vertical distribution of ^{137}Cs – an artificial fallout radionuclide – is controlled by soil characteristics and processes that may differ among soil groups. The application of a single modelling approach to large number of soil profiles provides an original contribution to the literature and allows for comparison between these different soil groups.

Materials and methods. In order to quantify ^{137}Cs migration in soils, we compiled and modelled depth-distributed data documented in the literature between 2000–2012. The resulting database comprised ninety-nine ^{137}Cs profiles sampled in fourteen soil groups of the World Reference Base (WRB) classification (FAO 1998) under different land uses or covers and collected at various geographical locations in the Northern hemisphere between 1992 and 2007.

Results and discussion. The ^{137}Cs profiles were classified in seven different categories according to the shape and location of radiocaesium peak. Depth of the latter ranged between 0 and 12 cm (median of 2 cm) and maximal penetration of cesium reached from 12 to 60 cm. The ^{137}Cs depth distributions in these soils were fitted using a diffusion-convection equation to allow comparison between different soil groups. Diffusion coefficients ranged from 0.02 to $4.44\text{ cm}^2\text{ yr}^{-1}$ in soils (median of $0.64\text{ cm}^2\text{ yr}^{-1}$); and convection velocities varied from 0 to 0.74 cm yr^{-1} (median of 0.1 cm yr^{-1}). The model underestimated ^{137}Cs concentrations by a median value of 1.9% of the total inventory in soil samples collected below 13 cm depth.

Conclusions. Global ^{137}Cs penetration velocities ranged from 0.05 to 0.76 cm yr^{-1} (median of 0.28 cm yr^{-1}) over a 25-year period. In future, the model could be improved to better simulate deep penetration of Cs. Our results showed that modelling ^{137}Cs depth profile with a diffusion-convection equation allowed estimating the bioturbation and clay translocation velocity in a certain number of soil groups. This quantification is crucial as these processes partially control the development of soil surface characteristics and several soil services.

Keywords Bioturbation • Clay translocation • Diffusion-convection equation • Fallout radionuclides • Numerical modelling • Pedogenesis

1 Introduction

Caesium-137 is characterised by a strong affinity for fine soil particles and is therefore widely used to quantify soil erosion at the plot and hillslope scales (Ritchie and Ritchie 2007), but it is also employed to quantify the relative contribution of sources delivering sediment to rivers (Ben Slimane et al. 2013). However, its ability to trace vertical transfers of solid matter in soils was much less investigated. Despite the importance of these transfers in controlling the soil surface characteristics, the occurrence and the velocity at which those transfers operate are still poorly understood and quantified. To this end, measurement of tracers that are strongly bound to the solid phase and that were absent from the parent material before the start of pedogenesis along soil profiles could provide relevant information to improve our knowledge on those processes.

Although some limitations to this method exist (e.g. Parsons and Foster 2011), caesium-137 fulfils both conditions. It was released into the atmosphere by the nuclear weapon tests or, more recently, by nuclear accidents like Chernobyl or Fukushima. Soils may therefore be considered as free of this isotope prior to the first nuclear weapon tests in 1953. During the nuclear weapon tests, from the 1950s to the 1970s, ^{137}Cs was mixed in the stratosphere and its deposition pattern to the terrestrial surface, referred to as global fallout, reflects the atmospheric circulation and its latitude-dependence (UNSCEAR 1982). In contrast, ^{137}Cs atmospheric deposits after nuclear accidents displayed locally heterogeneous spatial patterns, mostly depending on the weather conditions that prevailed shortly after the emissions (Chartin et al., 2013; Korsakissok et al. 2013). As a result of the combination of wet and dry fallout, ^{137}Cs is delivered to the soil surface, and it may then migrate in depth at a rate depending on processes controlling this transfer and soil characteristics.

These characteristics control the fixation of ^{137}Cs on solid phases. Caesium-137 was shown to be fixed on soil constituents such as micaceous clay minerals (Tamura and Jacobs 1960;

Sawhney 1972; Konoplev et al. 1996; Bunzl et al. 1998), bearing Frayed Edge Sites (FES) which are specific sites for caesium sorption (Cremers et al. 1988). According to Vandenbroek et al. (2012), surface layers of most soils found across the world were shown to have a high selectivity for caesium adsorption, with the exception of Oxisols, Podzols and Andosols that are depleted in micas. Regarding the other soil types, Gil-Garcia et al. (2009) found high solid/liquid distribution coefficients (K_d) for caesium with mean K_d values of 530 L kg⁻¹, 5 500 L kg⁻¹ and 3 500 L kg⁻¹ for sandy, clayey and loamy soils respectively. Under these conditions, transport of caesium in dissolved form may be considered negligible after the initial fixation of this radionuclide onto mineral soil particles (Elshamy et al. 2007; Jarvis et al. 2010; Matisoff et al. 2011). Consequently, over the long-term, radionuclides are mainly transported along hillslopes with soil particles (Müller-Lemans and van Dorp, 1996; Turner et al. 2006). In this context, the presence of artificial fallout radionuclides in depth can be interpreted as resulting from a particle-borne transfer due to clay migration or to a physical mixing byurbation including bioturbation processes (Müller-Lemans and van Dorp 1996; VandenBygaart et al. 1998; Ireson and Butler 2009). Nevertheless organic soils containing more than 20 % of organic matter were shown to have a relatively lower K_d value (mean of 270 L kg⁻¹) (Gil-Garcia et al. 2009). In these soils, a significant contribution of solute transfer of ¹³⁷Cs that may be associated with DOC cannot be ruled out (AIEA 2010).

Finally, the relatively short half-life of ¹³⁷Cs ($T_{1/2}$ =30 yr) is particularly relevant for investigating processes occurring at decadal scales (Kaste et al. 2007). Although soil formation occurs over much longer periods, individual pedological processes may be characterised by temporal variations with a succession of active and inactive periods throughout the entire story of soil development. The use of ¹³⁷Cs as a tracer provides therefore a relevant tool to understand and quantify those variations during the last 50 years.

Most approaches that aimed to quantify the migration velocities of fallout radionuclides in

soils solved the diffusion-convection equation (e.g. Bossew and Kirchner 2004; Schuller et al. 2004; Schimmack and Márquez 2006), using either analytical or numerical solutions (Smith and Elder 1999; Bunzl et al. 2000; Bossew and Kirchner 2004).

In this study, we aimed at quantifying the kinetics of vertical transfers of ^{137}Cs in soils depending on their characteristics and the relative contribution of processes controlling this transfer in soils. To this end, we modelled the vertical distributions of ^{137}Cs fallout along soil profiles documented in the literature.

2 Materials and methods

2.1 Database description

The database compiles 99 profiles with detailed evolution of ^{137}Cs activities with depth retrieved from 17 publications published before 2013 (Table 1, detailed information is provided in the Electronic Annex A). We restricted our work to the investigations mentioning the soil group of the studied profiles. This reduced considerably the amount of relevant studies among the hundreds of studies dedicated to ^{137}Cs in soils that have been continuously published since the mid-1990s (Ritchie and Ritchie 2007). Land use or cover, altitude, annual precipitation, sampling date and pedological data such as pH and organic matter content were compiled when available. The depth of maximum ^{137}Cs concentration and the maximum depth reached by ^{137}Cs were also collected.

In these studies, ^{137}Cs data were expressed in different units: Bq kg^{-1} (direct measurement of ^{137}Cs activity in gamma spectrometry - 85 % of observations), Bq cm^{-3} (^{137}Cs activity per volume of soil, which requires multiplying the values in Bq kg^{-1} by the bulk density – 2 % of observations), Bq cm^{-2} (inventory per layer, which requires multiplying the values in Bq cm^{-3} by the sample thickness – 7 % of observations), or percentage of the total inventory % Bq cm^{-2} (inventory per layer divided by the total inventory - 6 % of observations). All ^{137}Cs data

were therefore converted into Bq cm^{-3} to facilitate modelling. Several pedotransfer functions exist to estimate soil bulk density (De Vos et al. 2005). However, they are generally based on the measurements of organic matter content or particle-size distributions and these data were not available in most of the compiled studies. We therefore derived a relationship between this parameter and the soil depth from the values available for 13 soil profiles.). However, We determined the duration between soil sampling and publication (mean of four years with a standard-deviation of 1 year). Therefore, when not available (6 profiles), the sampling date was estimated as the publication date minus 4 years.

Soil groups were given according to different classifications (WRB in 38 % of studies (FAO, 1998); Soil Taxonomy in 36 % of studies (Soil Survey Staff 1999) and national classification systems in the remaining studies). To allow for comparison, all soil groups were reclassified according to the WRB system (FAO 1998). Among the 30 main soil groups of the WRB classification, only 14 soil groups were represented in the database (Table 1).

The studies were conducted in 12 countries from the Northern Hemisphere that were submitted to different levels of ^{137}Cs contamination due to the Chernobyl accident, which resulted in a large variability in ^{137}Cs inventories (Table 1).

Soil profiles were collected under different land uses or covers, i.e. grassland (56 % of the cases), woodland (25 % of the cases), peatland (2% of the cases), cropland (4% of the cases), and land use or cover was not specified in 13 % of the cases. In the latter case, the sites were described as undisturbed and we hypothesised that they were under grassland.

Most studies restricted sampling to the uppermost 20-cm layer (87 %). However, in 79 % of the cases, the deepest sample had ^{137}Cs non-null concentration. The inventories estimated by the authors were therefore probably underestimated.

In some studies, only three samples were analysed along the profile. The modelling approach was therefore not applicable to such a small number of samples and these soil profiles were

removed from further analysis (16 profiles). We did not model ^{137}Cs distribution in the profiles located under cropland, as our model did not take tillage processes into account.

2.2 Modelling of radionuclide vertical distributions

The migration of fallout radionuclides in soils is often described using the diffusion-convection equation (e.g. He and Walling 1997; Schuller et al. 1997):

$$\frac{\partial A}{\partial t} = \frac{\partial}{\partial z} \left(D_s \frac{\partial A}{\partial z} - v_s A \right) - \lambda A \quad (1)$$

With A - the total ^{137}Cs activity per volume of soil (Bq m^{-3} ; obtained by multiplying the ^{137}Cs activity in Bq kg^{-1} by the soil bulk density), z - the soil depth (m), t - the time (s), D_s - the diffusion coefficients ($\text{m}^2 \text{s}^{-1}$), v_s - the convection velocity (m s^{-1}), λ - the radioactive decay constant (s^{-1}). The diffusion coefficients D_s and the convection velocity v_s were considered as spatially and temporally constant in this study. This is an over-simplification, as we know that mixing processes and vertical transfers in soil may vary in both time and depth. However, we considered that intensity of these processes remained constant during the last decades.

In the model, ^{137}C is given as a bulk concentration, hypothesising that ^{137}Cs is evenly distributed within the soil mass of the considered layer. However, as ^{137}Cs is mainly deposited as the result of wet fallout, its distribution is heterogeneous in soils and depends on soil structure, macropore distribution and preferential flow (Bundt et al. 2000). Furthermore, it evolves throughout time due to the occurrence of wetting/drying cycles and bioturbation, and it depends on the soil moisture content at the time of deposition. Therefore, we acknowledge that ^{137}Cs distribution in soils may be more complex than modelled in our study.

2.2.1 Boundary conditions

At the soil surface, we applied a boundary condition with a pulse-like input of radionuclides. This pulse-like input was obtained by multiplying the annual fallout deposition $a_0(t')$ in Bq

179 m^{-2} of the considered year t' by a Dirac delta function $a(0, t') = a_0(t')\delta(t-t')$:

$$180 \quad D_s \frac{\partial A(0, t')}{\partial z} = a_0(t')\delta(t-t') \quad (2)$$

181 Isotopes were delivered to the surface, at $z=0$, annually and input time-series were dealt as a
 182 time-dependent boundary condition, i.e. $A(0, t') = A(0, t'-1) + a_0(t')\delta(t-t')$.

183 The annual global fallout input $a_{GF}(t')$ followed the distribution calculated by Cambray et al.
 184 (1989) for various locations across the Northern Hemisphere, with a total fallout
 185 accumulation of 3018 Bq m^{-2} before the Chernobyl accident in 1986. The global fallout was
 186 latitude-dependent and it was reconstructed based on ^{90}Sr fallout recorded by UNSCEAR
 187 (1982) in different latitudinal bands (Table 2). As the $^{137}\text{Cs}/^{90}\text{Sr}$ ratio was constant over time
 188 (1.6 for global bomb fallout), ^{137}Cs fallout (Table 2) was deducted from ^{90}Sr measurements.

189 With respect to the total fallout accumulation of Cambray et al. (1989), we defined a weighted
 190 factor maximum r_{th} as a function of the latitudinal position of the sampled site (Table 2).
 191 Therefore global fallout contribution was determined as the product of the weighted factor r
 192 and the annual global fallout a_{GF} (3). In the model, the weighted factor r was allowed to vary
 193 between 0 and the maximal weighted factor r_{th} of the latitudinal band in which the sampling
 194 site was located (Table 2). The factor r was considered as an unknown variable and was
 195 estimated by the model for each profile. As ^{137}Cs input due to the Chernobyl accident (a_{Ch})
 196 strongly varied across space, it was also considered as an unknown variable and was
 197 estimated by the model. Hence, the temporal input variations were described as follows:

$$198 \quad \begin{aligned} a(0, t') &= r a_{GF}(t')\delta(t-t') & 1954 \leq t' < 1986 \\ a(0, t') &= a_{Ch}\delta(t-t') & t' = 1986 \end{aligned} \quad (3)$$

199 We applied as lower boundary condition (at 200 cm) an advective transport free of diffusion
 200 in order to avoid spurious accumulation of radionuclides and not to influence the numerical
 201 solution within the first 100 cm.

202

203 2.2.2 Discretization and resolution of the equation

204 We solved the differential Eq. (1) using upwind finite differences approximation for spatial
 205 derivatives and semi-implicit Crank-Nicholson scheme as a time-solver. The obtained
 206 algebraic system of equations was solved by Gaussian elimination. All the simulations started
 207 in 1954 and finished on the sampling year. Time-step was equal to 0.01 year. The accuracy of
 208 the numerical solution was verified by comparison with the analytical solution (4) obtained
 209 with similar initial and boundary conditions:

$$210 \quad A(z, t) = a_0 \left\{ \frac{1}{\sqrt{\pi D_s t}} \exp\left(-\frac{(z - v_s t)^2}{4 D_s t}\right) - \frac{v_s}{2 D_s} \exp\left(\frac{v_s z}{D_s}\right) \operatorname{erfc}\left(\frac{v}{2} \sqrt{\frac{t}{D}} + \frac{z}{2 \sqrt{D t}}\right) \right\} \exp^{-\lambda t} \quad (4)$$

211 with a simple pulse-like input ($a_0(t) \delta(t)$) into a semi-infinite domain and a zero-concentration
 212 when $z \rightarrow \infty$.

213

214 2.2.3 Non-linear least square minimization procedure

215 The unknown parameters (D_s , v_s , r , a_{Ch}) and their standard deviations were determined with a
 216 non-linear least-square minimization procedure based on the Levenberg-Marquardt algorithm
 217 included in Python *lmfit* package. The minimized value for each measurement A_m (in Bq cm⁻³)
 218 was calculated in an integral form, which provided a way to take different sampling intervals
 219 into account (Bossew and Kirchner, 2004):

$$220 \quad \operatorname{argmin} = \left(A_m(z_2 - z_1) - \int_{z_1}^{z_2} A(z, t) dz \right)^2 \quad (5)$$

221 In order to estimate the performance of the fitting procedure, we calculated residual sum of
 222 squares (*RSS*) for N sampled layers and modelling efficiency (*EF*):

$$223 \quad RSS = \sum_{i=0}^N \left(A_m(i)(z_{i2} - z_{i1}) - \int_{z_{i1}}^{z_{i2}} A(z, t) dz \right)^2 \quad (6)$$

$$EF = 1 - \frac{\sum_{i=0}^N \left(\int_{z_{i1}}^{z_{i2}} A(z,t) dz - A_m(i)(z_{i2} - z_{i1}) \right)^2}{\sum_{i=0}^N \left(A_m(i)(z_{i2} - z_{i1}) - \frac{1}{N} \sum_{i=0}^N A_m(i) \right)^2} \quad (7)$$

The first simulation was run to fit all 4 parameters (D_s , v_s , r , a_{Ch}). If the resulting model parameters remained in the range of possible values ($0 < r \leq r_{th}$, $0 \leq a_{Ch}$), the simulation was retained as the best fit. Otherwise, we added the following assumptions: $a_{Ch} = 0$ if the sampling site was located in a region spared by Chernobyl deposits and $ra_{GF} = 4\,527\text{ Bq m}^{-2}$ (the maximum global fallout input) if the model attributed larger global fallout input than expected, which was often the case for the sites characterised by high levels of Chernobyl contamination. All sites affected by soil erosion or soil accumulation were excluded from the database for further analysis, and these processes were therefore not simulated by the model.

3 Results and discussion

3.1 Soil bulk density within the uppermost 20 centimetres of the soil: evolution and estimation

Bulk densities reported in the literature for 13 soil profiles ranged from 0.6 to 1.41 g cm⁻³ at 5-cm-depth and from 0.9 to 1.47 g cm⁻³ at 20-cm-depth. The mean bulk density of these 13 soil profiles evolved with depth following a logarithmic function (Fig. 1a; eq. 8):

$$\rho(z) = C_1 \ln(z) + C_2 \quad (8)$$

with depth z expressed in cm. For the considered soil profiles, C_1 was equal to 0.1636 and C_2 to 0.7329 ($R^2=0.94$).

We considered a comparable evolution of the bulk density in depth for the other profiles, with the same C_1 value, whereas C_2 was estimated in order to fit the total ¹³⁷Cs inventory (I) considering the depth evolution of the activities of ¹³⁷Cs in Bq kg⁻¹, using the minimization equation (9):

$$\text{argmin} = \left(\sum_{i=0}^N \left(\left(C_1 \ln \left(\frac{z_{i2} + z_{i1}}{2} \right) + C_2 \right) A_s(i) (z_{i2} - z_{i1}) \right) - I \right)^2 \quad (9)$$

In order to check the accuracy of the obtained predictive model, bulk densities were estimated for the soil profiles for which these data were available (Fig. 1b). We obtained a reasonable estimation of the bulk density in samples close to the surface, but the density was underestimated by the model below 10 cm depth. However, as only a small proportion of the ^{137}Cs was located at this depth, the impact of bulk density underestimation on ^{137}Cs vertical distribution was considered to be negligible.

The model was applied to the 48 soil profiles for which bulk density data were not available (Electronic Annex A). C_2 varied between 0.37 and 2.53. Eighty percent of the mean bulk density of the 0-20 cm layer - estimated by the predictive model - ranged from 0.94 to 1.66 g cm⁻³, with a median value of 1.22 g cm⁻³. The estimated soil bulk densities were considered as irrelevant in 5 profiles compared to the values classically found in soils. In addition, negative values were obtained in 4 profiles. They were therefore not considered in further analysis.

3.2 Cs-137 distributions in soils

The recorded ^{137}Cs inventories (decay-corrected for 2013) ranged from 697 Bq m⁻² to 107 200 Bq m⁻². The highest inventories were measured in soils collected in the areas severely contaminated by Chernobyl fallout in central Europe and Sweden as documented in the atlas of Chernobyl contamination (De Cort et al. 1998). The depth of the maximum concentration (z_{maxC}) ranged between 0 and 12 cm (with a median value of 2 cm) and the maximal penetration depth (*i.e.* with a ^{137}Cs concentration higher than or equal to 0.001 Bq cm⁻³) ranged between 12 cm and 60 cm. Width of the peak (estimated as Half Width at Half Maximum (*HWHM*)) ranged between 1.25 and 13.7 cm (median of 5 cm). We could not identify any clear relationship between the soil groups and the depth of maximum

272 concentration, maximum depth or width of the peak.

273 Based on the depth of maximum ^{137}Cs concentration, the width of peak and the shape of
274 radiocesium profile, we determined 7 classes of depth distribution profiles (Fig. 2):

275 1- Continuous decrease in the concentrations with depth ($z_{\text{max}C} = 0$ cm and $HWHM \leq 5$ cm -
276 13 profiles);

277 2- Half bell-shaped curve with maximum concentration at the soil surface. The concentrations
278 decrease with depth following a Gaussian-like shape ($z_{\text{max}C} = 0$ cm and $HWHM > 5$ cm - 10
279 profiles);

280 3- Narrow hump-shaped curve at shallow depth with a maximum concentration comprised
281 between 0 and 2 cm depth ($0 < z_{\text{max}C} \leq 2$ cm and $HWHM \leq 5$ cm - 25 profiles);

282 4- Wide hump-shaped curve shape at shallow depth with a maximum concentration within the
283 0-2 cm depth interval ($0 < z_{\text{max}C} \leq 2$ cm and $HWHM > 5$ cm - 13 profiles);

284 5- Narrow hump-shaped curve with maximum concentration deeper than 2 cm ($z_{\text{max}C} > 2$ cm
285 and $HWHM \leq 5$ cm - 12 profiles);

286 6- Wide hump-shaped curve with maximum concentration deeper than 2 cm ($z_{\text{max}C} > 2$ cm and
287 $HWHM > 5$ cm - 18 profiles);

288 7- Constant isotopic profile concentration over depth (4 profiles).

289

290 As ^{137}Cs fate in the soil depends on the soil characteristics and processes, ^{137}Cs depth
291 distribution should be a function of the soil groups. It was however not the case (Table 3).

292 Other factors may influence the distribution profile such as the duration since the ^{137}Cs input
293 or the climate conditions.

294 VandenBygaert et al. (1998), Tyler et al. (2001), Bunzl (2002), Kaste et al. (2007), Elshamy
295 et al. (2007), Ireson and Butler (2009), Jarvis et al. (2010) and Matisoff et al. (2011)
296 demonstrated the impact of bioturbation on radionuclide redistribution in soil profiles. The

297 importance of bioturbation may be estimated from the width of the peak: highly active
 298 bioturbation leads to wider shaped curves. We would thus expect stronger bioturbation in the
 299 profile types 2, 4, 6 than in profile types 1, 3, 5. As bioturbation depends on the mean annual
 300 temperature, it should be more intense under warm climates than under cold or dry climates.
 301 Table 4 shows the climate distribution for the different profile types as it was determined
 302 from the location of the sampling site and the climate classification according to Kottek et al.
 303 (2006). However, depth distribution profiles 2, 4, 6 were not more frequent under warm
 304 temperate and humid climates (C classes of Köppen's climate classification, 32 profiles out of
 305 69), than under dry or cold climates (B and D classes of Köppen's climate classification; 8 out
 306 of 20). Land use or cover may also impact the ^{137}Cs distribution. Most of the type-1
 307 distribution profiles corresponding to Cambisols were collected under forests whereas
 308 peatlands corresponded to type 5 ($n = 2$). All the cultivated plots were attributed to type 7
 309 ($n = 4$).
 310 The soil profiles were sampled between 1992 and 2007. The classification of different soils of
 311 a same soil group according to different depth-profile types may be explained by the temporal
 312 evolution of the initial profile. Indeed, after ^{137}Cs inputs, the peak progressively migrates in
 313 depth, and its distribution profile will widen with time depending on the turbation rate. This
 314 evolution will nevertheless depend on the time elapsed after the main ^{137}Cs input that may
 315 either be due to the bomb tests or to the Chernobyl accident depending on the site. This
 316 probably explains why we could not find any relationship between ^{137}Cs depth profiles and
 317 the sampling dates. In addition, for similar sampling dates, classification according to
 318 different profile shapes may originate from differences in values of diffusion coefficient (D_s
 319 in $\text{cm}^2 \text{ yr}^{-1}$) and convection velocity (v_s in cm yr^{-1}): profile type 1 being obtained for $D_s > 0$
 320 and $v_s \approx 0$; profile type 2 for $D_s \gg 0$ and $v_s \approx 0$; profile type 3 for $D_s > 0$ and $v_s > 0$; profile
 321 type 4 for $D_s \gg 0$ and $v_s > 0$; profile type 5 for $D_s > 0$ and $v_s \gg 0$; and profile type 6 for D_s

$\gg 0$ and $v_s \gg 0$. The profiles belonging to class 7 result either from a disturbance or from a mixing of the entire profile that may have been achieved by agricultural practices such as tillage.

As a conclusion, the ^{137}Cs profile type is a complex result of the variations in soil processes affecting the diffusion and convection transport and resulting in contrasted temporal evolutions.

3.3 Diffusion and convection in soils

3.3.1 Overall quality of the simulation results

We have simulated the evolution of 71 profiles out of the 99 profiles compiled in the database (detailed results are provided in the Electronic Annex A and B). We have excluded 4 cultivated profiles, 4 profiles with thick organic horizons containing more than 30 % of the total ^{137}Cs inventory, 4 profiles with unrealistic bulk density values and 16 profiles with only 3 measurements per profile, which was insufficient for optimizing the 4 required parameters. Overall, modelling efficiency (EF) reached a mean of 0.89 ± 0.20 , and a median of 0.96 ($\max(EF) = 1$), which is fairly good. The comparison between simulated and observed ^{137}Cs concentrations in all profiles showed a reasonable agreement with some underestimation at low concentrations (Fig. 3). Scaled residuals as a function of ^{137}Cs concentrations were normally distributed. However, when considered as a function of depth (Fig. 4), residuals were normally distributed and centred at zero in the first 13 cm only. They shifted towards positive values below 13 cm and they were all positive below 30 cm. This demonstrates that the model underestimated ^{137}Cs concentrations in most samples collected below 13 cm and that it underestimated all values below 30 cm.

Finally, v_s was null for 23 profiles. These profiles exhibited large standard deviation values for all parameters. The model could therefore not differentiate the inputs due to global fallout from those due to the Chernobyl accident, and the estimation of D_s coefficient satisfied a wide

range of values.

3.3.2 Estimation of global fallout

We have compared the global fallout ra_{GF} values optimized by our model to the range of fallout estimations provided by UNSCEAR (1982) for different latitudes (Table 2). In 59 % of the cases, the modelled global fallout values were equivalent to the values estimated by UNSCEAR (1982), but they were lower in the remaining 41% of the sites. However, the values estimated by UNSCEAR (1982) may have been affected by other local factors such as rainfall. As an example, Cambray et al. (1989) provided lower values of global fallout compared to UNSCEAR (1982) for regions located in the 40-50°N band. Furthermore, extremely low values of global fallout were obtained for the sites located in arid regions in Syria, reaching even zero at several places (4 sites). The lower fallout obtained in this region compared to those estimated by UNSCEAR (1982) probably reflects the absence of precipitations (Aoyama et al. 2006).

The occurrence of soil erosion between the end of 1960s and Chernobyl accident may also explain the lower values of global fallout obtained for several profiles. However, no data (such as the history of land use or cover of the corresponding sites) were available to verify this hypothesis. In addition, erosion was probably very limited at these sites, since they were generally covered by vegetation (forest, grassland) and were therefore considered as undisturbed.

For three Swedish sites located between 62 and 64°N, the use of the global fallout of the 50-60°N band with higher fallout rates improved the modelling efficiency compared to the one obtained when taking values of the 60-70° band. Similarly, Sigurgeirsson et al. (2005) reported that ^{137}Cs deposition was underestimated for 60-70° N band by UNSCEAR (1982) in Icelandic soils. We observe the same phenomena for Chinese sites at 29°N, showing that the

latitude parallels defining the limit between two bands should be rather considered as buffer (transition?) zones.

Overall, differences between observed and simulated global fallout may be explained by: (i), a virtual absence of fallout because of the lack of precipitation in arid regions (as it was probably the case in Syria); (ii), the possible occurrence of soil erosion between the end of 1960s and Chernobyl accident.

3.3.3 Possible mechanisms behind D and v

Diffusion coefficient is widely used to estimate bioturbation in the literature (e.g. Elzein and Balesdent 1995; Schiffers et al. 2011). Bioturbation in soil is due to the activity of micro- and macro-organisms. Under temperate climates, it is mainly controlled by earthworm activity (Wilkinson et al. 2009). However, diffusion coefficient only represents a local mixing, whereas a non-negligible part of bioturbation may be due to non-local feeder earthworm species (Jarvis et al. 2010; Schiffers et al. 2011). As the diffusion coefficient is unable to simulate this non-local behaviour, alternative models were developed (Müller-Lemans and van Drop, 1996; Schiffers et al. 2011). A non-local mixing term was integrated into the general diffusion-convection equation (Jarvis et al. 2010), which improved the simulations of deep penetration of ^{137}Cs . The transfer function describing the behaviour of non-local feeders requires the implementation of other parameters (*i.e.* ingestion rate of non-local feeders, depth of ingestion), which were calibrated by Jarvis et al. (2010) for their field-site based on an ecological study of anecic and endogeic earthworm species (*i.e.*, classical non-local feeders in temperate soils). As such data were not available for the study sites, we could not model the behaviour of non-local feeders. However, Jarvis et al. (2010) showed that estimating bioturbation with a diffusion coefficient fits reasonably well with ^{137}Cs profiles, although it underestimates concentrations at depth.

The simulated diffusion coefficients D_s ranged from 0.02 to 4.44 cm² yr⁻¹ (median of 0.64 cm² yr⁻¹). Among the profiles compiled in our database, modelling had already been conducted on 35 soils using different methods for solving the diffusion-convection equation (Szerbin et al. 1999; Krstic et al. 2004; Schimmack and Márquez 2006; Zhang et al. 2008; Legarda et al. 2011). These studies obtained a median D_s value of 0.53 cm² yr⁻¹, which is slightly different from our findings. However, this discrepancy may be explained by the inclusion of ca. 40 additional profiles in our study, which may have enlarged the range of D_s values in soils. These D_s values were the highest for depth-distribution type 4 (mean of 1.4 cm² yr⁻¹). Then, they were higher for depth distribution types 2, 3, and 6 (mean ranging from 0.8 to 1.3 cm² yr⁻¹) than for depth distribution types 1 and 5 (mean of 0.5 cm² yr⁻¹; ANOVA with a Fisher test at 5 % level of confidence).

We could not observe any significant difference in D_s values as a function of land use or cover, the time elapsed after the main ¹³⁷Cs input, soil group or the climate type.

One of the difficulties associated with our approach is that D_s may account for both a diffusion process and a dispersion of the convection velocities when the convection operates. Therefore, a correlation between both parameters may indicate that D_s is not exclusively linked to bioturbation but that it is also affected by convection processes. However, we did not observe any correlation between D_s and either the precipitation or v_s when considering the entire set of profiles. When the different soil groups were considered separately (Fig. 5), D_s remained independent from v_s in Luvisols and Regosols, whereas in Yermosols, a positive linear relationship was observed between D_s and v_s ($D_s = 4.13v_s + 0.08$, $R^2 = 0.84$, $n = 8$). This relationship suggested that an important dispersive effect exists in Yermosols with increasing convection velocity. The relatively high D_s values observed in this soil group under a dry climate are therefore likely due to the convection process occurring in these soils and not to bioturbation, which is probably low under dry climate conditions. In Cambisols, i.e. the most

documented soil group ($n = 19$), three trends between D_s and v_s were observed: a variable diffusion at zero-velocity, a slight dispersion with increasing v_s and a high dispersion. Although they were not significantly different, D_s values lower than $0.2 \text{ cm}^2 \text{ yr}^{-1}$ were generally found either in the soils experiencing podzolization or gleization (Spodic Dystric Cambisols, Hypogleyic Cambisols, Podzols, Gleyic Phaeozems) or in soils collected in Northern regions (latitude $> 60^\circ\text{N}$ with the exception of a Histosol profile sampled in Hille, Sweden). Soils of the first group were acidic and thus unfavourable to bioturbating organisms, whereas in the second group bioturbation was limited because of the cold climate conditions. In Cambisols and Regosols, D_s increased with soil pH (R of 0.50 for $n = 13$ in Cambisol). For the other soil groups, the soil pH data were too scarce to draw any conclusion. Finally, the high D_s values obtained for Chernozems and Vertisols may be due to the occurrence of specific pedological processes in these soils (organic matter diffusion andurbation via shrinking and swelling, respectively), rather than to bioturbation (Table 5).

The v_s coefficient can be considered as representative of the transfer of ^{137}Cs that was either adsorbed on the clay surface - and that may therefore reflect clay translocation-, or that was dissolved in organic rich soils. In this study, v_s ranged from 0 to 0.74 cm yr^{-1} (median of 0.1 cm yr^{-1}). These values were of the same order of magnitude as those obtained for the 35 profiles that were previously modelled in the literature (Szerbin et al. 1999; Krstic et al. 2004; Schimmack and Márquez 2006; Zhang et al. 2008; Legarda et al. 2011). The v_s values ranked as follows from the highest to the lowest: types 5 and 6 (mean ranging from 0.27 to 0.35 cm yr^{-1}) $>$ type 3 and 4 (mean ranging from 0.13 to 0.15 cm yr^{-1}) $>$ type 1 and 2 (mean ranging from 0 to 0.01 cm yr^{-1}) (ANOVA with a Fisher test at 5 % level of confidence). Zero-velocity occurred in 23 profiles corresponding to different soil groups: Acrisols ($n=1$), Cambisols ($n=7$), Fluvisols ($n=2$), Histosols ($n=3$), Leptosols ($n=1$), Regosols ($n=2$), Solonetz ($n=2$), Vertisols ($n=2$) and Yermosols ($n=3$). We could not identify the key parameter responsible for

these zero-velocities. Significant differences in v_s were observed under various land uses and covers, with the highest v_s values found under peatland (0.38 cm yr^{-1} , $n=2$) and the lowest under forests (0.07 cm yr^{-1} , $n=6$), soils located under grassland being associated with intermediary v_s values (0.14 cm yr^{-1} , $n=63$).

Several authors found that migration velocity of ^{137}Cs decreased with time after the radionuclide input (Alexakhin and Krouglov, 2001). To test the impact of this parameter, we calculated the time elapsed since the main ^{137}Cs input considering the simulated respective contributions of global fallout and Chernobyl accident (all values were decay-corrected to the sampling date). The simulated v_s values for the entire set of profiles decreased with time (T) after the main ^{137}Cs input ($R=-0.32$, $n=71$; Fig. 6).

When considering the different soil groups, velocities were independent of the time after the main input in Vertisols, Solonetz, Luvisols, Fluvisols and Cambisol. For the two first soil groups, v_s were systematically lower than 0.11 cm yr^{-1} , whereas in Luvisols, velocities were relatively high ($0.22 \pm 0.06 \text{ cm yr}^{-1}$). They ranged from 0 to 0.43 yr cm^{-1} in Fluvisols and Cambisols. In Leptosols, Podzols, Regosols and Yermosols, the velocities decreased with time, whereas they increased in Histosols.

In addition, Histosols showed two populations of profiles: profiles with a high D_s value ($> 1.5 \text{ cm}^2 \text{ yr}^{-1}$) and a v_s close to 0 cm yr^{-1} or profiles with the highest v_s values ($v_s > 0.2 \text{ cm yr}^{-1}$ with $D_s < 0.2 \text{ cm}^2 \text{ yr}^{-1}$). The highest v_s values were simulated in peatland profiles, whereas the lowest velocities were obtained for Histosols under forest or grassland. In addition v_s values increased with time. As discussed in the introduction, transport of ^{137}Cs in dissolved form can partially explain the simulated v_s values in Histosols that contain more than 20% of organic matter. Their relatively lower value when Histosol were collected under grassland or forest is probably due to a partial absorption of the soluble fraction of ^{137}Cs by the vegetation. In the case of Luvisols, Acrisols, Solonetz and Cambisols, the obtained v_s values may reflect

the occurrence of clay translocation, as clay translocation is a major pedogenic process in Luvisols, Acrisols and Solonetz, and secondary pedogenic process in Cambisols (FAO 1998; Bockheim and Gennadiyev 2000). The significantly higher v_s values obtained for Luvisols (ANOVA with a Fisher test at 5 % level of confidence, Table 5) are consistent with the occurrence of clay translocation in this soil group. Clay translocation may also explain some of the few high v_s values found in Cambisols. However, some profiles corresponding to these soil groups have a v_s value equal to 0 cm yr⁻¹ (Acrisols for example) or positive but extremely low values (Solonetz with a mean v_s of 0.03 cm yr⁻¹). In these soils, clay translocation might not have been active since the beginning of the ¹³⁷Cs release into the atmosphere. This finding is consistent with the fact that this process is only active in soils where pH ranges from 5 to 6.5 (Quénard et al. 2011), except in sodic soils (Solonetz for example).

3.3.4 Global penetration velocity of caesium-137

Soils compiled in the database were submitted to different inputs of ¹³⁷Cs and were sampled at various dates. As mentioned above, these parameters control the actual depth of penetration. We modelled D_s and v_s that are responsible for the penetration of caesium in the soils, but these parameters did not provide a direct picture of the penetration velocity of caesium in the soil. To obtain it, we calculated the mean depth reached by ¹³⁷Cs after 25 years of migration, considering that ¹³⁷Cs was instantaneously injected into the soil as a Dirac distribution representing a 'spike' of concentrated ¹³⁷Cs (Table 5). Mean penetration velocity of ¹³⁷Cs over 25 years varied between 0.05 and 0.76 cm yr⁻¹ (median of 0.28 cm yr⁻¹). When calculated over 50 years, these values decreased by about 20 % in average because of diffusive processes and ranged from 0.04 to 0.71 cm yr⁻¹ (median of 0.21 cm yr⁻¹).

3.3.5 Caesium-137 content at depth below 13 cm

As mentioned before, the distribution of scaled residuals suggested that the model underestimated the deep penetration of ^{137}Cs below 13 cm. We therefore estimated the unmodelled Cs inventory and quantified its migration rate. The proportion of the inventory underestimated by the model below 13 cm ranged between -3.8 and 37.2 %, with a median value of 1.9 %. The mean transfer rate below 13 cm was then calculated by dividing the underestimated fraction by the time after the main ^{137}Cs input. It ranged from 7.5 to 44.1 years (median of 20.1 years) and the obtained rate of transfer below 13 cm varied from 0 to 0.02 yr^{-1} (median of 0.001 yr^{-1}). As discussed above, this transfer in depth could be due to the behaviour of non-local feeders. Matisoff et al. (2011) estimated that the feeding rate of non-local feeders ranged from 0.002 to 0.02 yr^{-1} depending on the soil group. As our results are of the same order of magnitude, this suggests that non-local feeders could be responsible for these deep transfers.

3.3.6 Approximations in the model: implications for estimating solid matter transfers

Initial distribution of radionuclides was modelled as an injection at the surface ($z=0$), which is an over-simplification as ^{137}Cs wet deposits are inherently heterogeneous. Cs-137 is most likely sorbed in soil macropores, and sorbing zones are affected by preferential flow (Bundt et al. 2000), which depends itself on soil structure and moisture. Prediction of initial ^{137}Cs distribution is therefore case-dependent and as such, it is difficult to reproduce in a general modelling approach.

Furthermore, in our modelling approach, we considered that the diffusion and convection velocity coefficients were constant in time and depth. However, some authors predicted a decrease in migration velocities with time (Alexakhin and Krouglov 2001). Nevertheless, as outlined by Kirchner (1998), this apparent decrease simulated by compartment models may be explained by the occurrence of the diffusive process itself. The heterogeneous distribution

of ^{137}Cs can also be a cause of this decrease: ^{137}Cs was likely initially located on the surface of macropores and aggregates and it was then progressively incorporated within the soil matrix and become therefore less mobile. Nevertheless, it has been shown that preferential flow paths (and thus the soil structure) may remain stable for more than 40 years in soils (Bundt et al. 2000). Moreover, pedogenic processes affecting the vertical solid matter transfers such as bioturbation were recognized to be depth-depend (Müller-Lemans and Van Dorp 1996). Similarly, integrating non-local feeders modelling as in Jarvis et al. (2010) has a direct influence on the soil displacement rate (corresponding to v_s in our study), which decreased linearly with depth in the feeding zone. Therefore, we expect that the inclusion of depth-variable coefficients in the model may improve simulations of ^{137}Cs distributions at depths < 13 cm. However, including spatially variable coefficients in the model would require the inclusion of new parameters (at least 2 additional factors to account for exponentially decreasing D_s and v_s), the improvement of the database or the collection of additional samples and measurements in the field.

4 Conclusions

The modelling approach proposed in this study provides an original basis for comparing ^{137}Cs depth migration in different soil groups. However, no difference of global ^{137}Cs penetration velocities was observed among soil groups. It ranged from 0.05 to 0.76 cm yr⁻¹ (median of 0.28 cm yr⁻¹), with a significantly lower velocity for soil groups poorly evolved (Regosol), and a significantly higher for soils rich in organic matter (Histosols). The model underestimated this velocity under a 13-cm depth. In the future, the model could be improved by introducing a non-local feeder term or by allowing D_s and v_s values to vary with soil depth in the diffusion-convection equation.

D_s and v_s values explained the depth profile of ^{137}Cs in soils and were shown to partly reflect

the occurrence ofurbation including bioturbation for D_s and clay translocation for v_s , except in Histosols where the transfer of ^{137}Cs in dissolved form was very likely. D_s values ranged from 0.02 to 4.44 $\text{cm}^2 \text{yr}^{-1}$ (median of 0.64 $\text{cm}^2 \text{yr}^{-1}$) and v_s varied from 0 to 0.74 cm yr^{-1} (median of 0.1 cm yr^{-1}).

Due to the limited number of documented profiles per soil group and other potential sources of variability that could not be documented (climate, land use or cover, pH), it was not possible to attribute precisely D_s and v_s variations to the occurrence of different soil processes. However, clear trends were observed for certain soil groups. Lower D_s values were simulated in acidic soils or under cold climate conditions, and they likely reflected the lower-intensity bioturbation prevailing under those pedoclimatic conditions. In addition, high v_s values were simulated in soils submitted to clay translocation (notably Luvisols). A relationship was also derived between v_s and certain land uses or covers. In this context, our results showed that modelling ^{137}Cs depth-profile with a diffusion-convection equation allowed estimating the bioturbation and clay translocation velocity in a certain number of soil groups. Although the occurrence of these soil processes is well known in most soils, they have been rarely quantified. Still, this quantification is crucial as these processes partially control the development of soil surface characteristics and several soil services such as plant nutrition, organic carbon storage or pollutant fixation. In the future, to overcome the limitations associated with the modelling of diffusion-convection parameters based on ^{137}Cs measurements, we suggest to conduct a multi-isotopic characterisation of different types of soil profiles in order to quantify the relative contribution of the different processes responsible for vertical solid matter transfer.

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References

- AIEA (2010) Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments. Technical Reports Series No. 472. International Atomic Energy Agency
- Al-Masri M (2006) Vertical distribution and inventories of Cs-137 in the Syrian soils of the eastern Mediterranean region. *J Environ Radioact* 86:187-198
- Alexakhin RM, Krouglov SV (2001) Soil as the main compartment for radioactive substances in terrestrial ecosystems. In: Bréchignac F, Howard BJ (eds) *Radioactive pollutants. Impact on the environment*, EDP Sciences, Les Ulis, France, pp 149-174
- Aoyama M, Hirose K, Igarashi Y (2006) Re-construction and updating our understanding on the global weapons test ¹³⁷Cs fallout. *J Environ Monit* 8:431-438
- Ben Slimane A, Raclot D, Evrard O, Sanaa M, Lefèvre I, Ahmadi M, Tounsi M, Rumpel C, Ben Mammou A, Le Bissonnais Y (2013) Fingerprinting sediment sources in the outlet reservoir of a hilly cultivated catchment of Tunisia. *J Soils Sediments* 13:801-815
- Bockheim J, Gennadiyev A (2000) The role of soil-forming processes in the definition of taxa in Soil Taxonomy and the World Soil Reference Base. *Geoderma* 95:53-72
- Bossew P, Kirchner G (2004) Modelling the vertical distribution of radionuclides in soil. Part 1: the convection-dispersion equation revisited. *J Environ Radioact* 73:127-150

598 Bunzl K (2002) Transport of fallout radiocesium in the soil by bioturbation: a random walk
 599 model and application to a forest soil with a high abundance of earthworms. *Sci Total*
 600 *Environ* 293:191–200

601 Bunzl K, Kracke W, Schimmack W, Zelles L (1998) Forms of fallout Cs-137 and Pu239+240
 602 in successive horizons of a forest soil. *J Environ Radioact* 39:55-68

603 Bunzl K, Schimmack W, Zelles L, Albers B (2000) Spatial variability of the
 604 vertical migration of fallout Cs-137 in the soil of a pasture, and consequences for long-
 605 term predictions. *Radiat Environ Biophys* 39:197-205

606 Bundt M, Albrecht A, Froidevaux P, Blaser P, Flühler H (2000) Impact of preferential flow
 607 on radionuclide distribution in soil. *Environ Sci Tech* 34:3895-3899

608 Cambray RS, Playford K, Lewis G, Carpenter R (1989) Radioactive fallout in air and rain:
 609 results to the end of 1988. Environmental and Medical Sciences Division, United
 610 Kingdom Atomic Energy Authority

611 Chartin C, Evrard O, Onda Y, Patin J, Lefèvre I, Ayrault S, Lepage H, Bonté P (2013)
 612 Tracking the early dispersion of contaminated sediment along rivers draining the
 613 Fukushima radioactive pollution plume. *Anthropocene* 1: 23-34

614 Cremers A, Elsen A, Depreter P, Maes A (1988) Quantitative-analysis of radiocesium
 615 retention in soils. *Nature* 335:247-249

616 De Cort M, Dubois G, Fridman SD, Germenchuk MG, Izrael YA, Janssens A, Jones AR,
 617 Kelly GN, Kvasnikova EV, Matveenkov II, Nazarov IM, Pokumeiko YM, Sitak VA,
 618 Stukin ED, Tabachnyi LY, Tsaturov YS, Avdyushin SI (1996) Atlas of caesium
 619 deposition on Europe after the Chernobyl accident. EUR Report 16733, EC, Office for
 620 Official Publications of the European Communities, Luxembourg

621 De Vos B, Van Meirvenne M, Quataert P, Deckers J, Muys B (2005) Predictive quality of
 622 pedotransfer functions for estimating bulk density of forest soils. *Soil Sci Soc Am J*

623 69:500-510

624 Elshamy M, Mathias S, Butler A (2007) Demonstration of radionuclide transport modelling
625 under field conditions: 50-year simulation of caesium migration in soil. Tech. Rep.
626 Imperial/ NRP_016, United Kingdom Nirex Limited

627 Elzein A, Balesdent J (1995) Mechanistic simulation of vertical distribution of carbon
628 concentrations and residence times in soils. *Soil Sci Soc Am J* 59:1328-1335

629 FAO (1998) World reference base for soil resources. *World Soil Resources Rep.* 84, Rome

630 Gallagher D, McGee E, Mitchell P (2001) A recent history of C-14, Cs-137, Pb-210, and Am-
631 241 accumulation at two Irish peat bog sites: An east versus west coast comparison.
632 *Radiocarbon* 43:517-525

633 Gil-Garcia C, Rigol A, Vidal M (2009) New best estimates for radionuclide solid liquid
634 distribution coefficients in soils. Part 1: radiostrontium and radiocaesium. *J Environ*
635 *Radioact* 100:690-696

636 He Q, Walling D (1997) The distribution of fallout Cs-137 and Pb-210 in undisturbed and
637 cultivated soils. *App Radiat Isot* 48:677-690

638 Ireson M, Butler A (2009) A review of soil bioturbation and soil development. Tech. Rep.
639 Imperial/NRP_018, United Kingdom Nirex Limited

640 Isaksson M, Erlandsson B, Mattsson S (2001) A 10-year study of the Cs-137 distribution in
641 soil and a comparison of Cs soil inventory with precipitation-determined deposition. *J*
642 *Environ Radioact* 55:47-59

643 Jarvis NJ, Taylor A, Larsbo M, Etana A, Rosen K (2010) Modelling the effects of
644 bioturbation on the re-distribution of ¹³⁷Cs in an undisturbed grassland soil. *Eur J*
645 *Soil Sci* 61:24-34

646 Karadeniz O, Yaprak G (2008) Geographical and vertical distribution of radiocesium levels in
647 coniferous forest soils in Izmir. *J Radioanal Nucl Chem* 277:567-577

648 Kaste JM, Heimsath AM, Bostick, BC (2007) Short-term soil mixing quantified with fallout
649 radionuclides. *Geology* 35:243-246

650 Kirchner G (1998) Applicability of compartmental models for simulating the transport of
651 radionuclides in soil. *J Environ Radioact* 38:339-352

652 Konoplev A, Bulgakov A, Popov V, Hilton J, Comans R (1996) Long-term investigation of
653 Cs-137 fixation by soils. *Radiation Protection Dosimetry* 64:15-18

654 Korsakissok I, Mathieu A, Didier D (2013) Atmospheric dispersion and ground deposition
655 induced by the Fukushima nuclear power plant accident: a local-scale simulation and
656 sensitivity study. *Atmos Environ* 70:267-279

657 Kottek M, Grieser J, Beck C, Rudolf B, Rubel F (2006) World map of the Koppen Geiger
658 climate classification updated. *Meteorologische Zeitschrift* 15(3):259-263

659 Krstic D, Nikezic D, Stevanovic N, Jelic M (2004) Vertical profile of ¹³⁷Cs in soil. *App*
660 *Radiat Isot* 61:1487-1492

661 Kruse-Irmer S, Giani L (2003) Vertical distribution and bioavailability of Cs-137 in organic
662 and mineral soils. *J Plant Nutr Soil Sci* 166:635-641

663 Kruyts N, Delvaux B (2002) Soil organic horizons as a major source for radiocesium
664 biorecycling in forest ecosystems. *J Environ Radioact* 58:175-190

665 Legarda F, Romero LM, Herranz M, Barrera M, Idoeta R, Valino F, Olondo C, Caro A (2011)
666 Inventory and vertical migration of Cs-137 in Spanish mainland soils. *J Environ*
667 *Radioact* 102:589-597

668 Lepage H, Evrard O, Onda Y, Patin J, Chartin C, Lefèvre I, Bonté P, Ayrault S (2014)
669 Environmental mobility of ^{110m}Ag: lessons learnt from Fukushima accident (Japan)
670 and potential use for tracking the dispersion of contamination within coastal
671 catchments. *J Environ Radioactiv* 130:44-55

672 Mabit L, Benmansour M, Walling DE (2008) Comparative advantages and limitations of the

673 fallout radionuclides Cs-137, Pb-210(ex) and Be-7 for assessing soil erosion and
674 sedimentation. *J Environ Radioactiv* 99:1799-1807

675 Matisoff G, Ketterer ME, Rosen K, Mietelski JW, Vitko LF, Persson H, Lokas E (2011)
676 Downward migration of Chernobyl-derived radionuclides in soils in Poland and
677 Sweden. *Appl Geochem* 26:105-115

678 Milton G, Kramer S, Watson W, Kotzer T (2001) Qualitative estimates of soil disturbance in
679 the vicinity of CANDU stations, utilizing measurements of Cs-137 and Pb- 210 in soil
680 cores. *J Environ Radioact* 55:195-205

681 Müller-Lemans H, van Dorp F (1996) Bioturbation as a mechanism for radionuclide transport
682 in soil: Relevance of earthworms. *J Environ Radioact* 31:7-20

683 Noordijk H, Van Bergeijk KE, Lembrechts J, Frissel MJ (1992) Impact of ageing and weather
684 conditions on soil-to-plant transfer of radiocesium and radiostrontium. *J Environ*
685 *Radioact* 15:277-286

686 Parsons AJ, Foster IDL (2011) What can we learn about soil erosion from the use of ¹³⁷Cs?
687 *Earth-Sci Rev* 108:101-113

688 Quénard L, Samouelian A, Laroche B, Cornu S (2011) Lessivage as a major process of soil
689 formation: A revisitation of existing data. *Geoderma* 167-168:135-147

690 Ritchie JC, Ritchie CA (2007) Bibliography of publications of ¹³⁷cesium studies related to
691 erosion and sediment deposition. Tech. rep., USDA Agricultural Research Service

692 Rosén K, Öborn I, Lönsjö H (1999) Migration of radiocaesium in Swedish soil profiles after
693 the Chernobyl accident, 1987–1995. *J Environ Radioact* 46:45-66

694 Sanchez A, Smolders E, Van den Brande K, Merckx R, Wright S, Naylor C (2002)
695 Predictions of in situ solid/liquid distribution of radiocaesium in soils. *J Environ*
696 *Radioact* 63:35-47

697 Sawhney B (1972) Selective sorption and fixation of cations by clay minerals: a review. *Clays*

698 Clay Miner 20:93-100

699 Schiffrers K, Teal LR, Travis JMJ, Solan M (2011) An open source simulation model for soil
700 and sediment bioturbation. PloS ONE 6:e28028

701 Schimmack W, Márquez FF (2006) Migration of fallout radiocaesium in a grassland soil from
702 1986 to 2001. Part II: Evaluation of the activity-depth profiles by transport models. Sci
703 Total Environ 368:863-874

704 Schimmack W, Schultz W (2006) Migration of fallout radiocaesium in a grassland soil from
705 1986 to 2001 - Part I: Activity-depth profiles of Cs-134 and Cs-137. Sci Total Environ
706 368:853-862

707 Schuller P, Bunzl K, Voigt G, Ellies A, Castillo A (2004) Global fallout Cs-137 accumulation
708 and vertical migration in selected soils from South Patagonia. J Environ Radioactiv
709 71:43-60

710 Schuller P, Ellies A, Kirchner G (1997) Vertical migration of fallout Cs-137 in agricultural
711 soils from Southern Chile. Sci Total Environ 193:197-205

712 Sigurgeirsson MA, Arnalds O, Palsson SE, Howard BJ, Gudnason K (2005) Radiocaesium
713 fallout behaviour in volcanic soils in Iceland. J Environ Radioact 79:39-53

714 Smith JT, Elder DG (1999) A comparison of models for characterizing the distribution of
715 radionuclides with depth in soils. Eur J Soil Sci 50:295-307

716 Smith JT, Fesenko SV, Howard BJ, Horrill AD, Sanzharova NI, Alexakhin RM, Elder DG,
717 Naylor C (1999) Temporal change in fallout ¹³⁷Cs in terrestrial and aquatic systems:
718 a whole ecosystem approach. Environ Sci Tech 33:49-54

719 Szerbin P, Koblinger-Bokori E, Koblinger L, Vegvari I, Ugron A (1999) Caesium- ¹³⁷
720 migration in Hungarian soils. Sci Total Environ 227:215-227

721 Soil Survey Staff (1999) Soil taxonomy: a basic system of soil classification for making and
722 interpreting soil surveys. US Department of Agriculture, Agriculture Handbook, N°

723 436, Washington DC, 2nd edition

724 Tamura T, Jacobs D (1960) Structural implications in cesium sorption. *Health Phys* 2:391-

725 398

726 Turner NB, Ryan JN, Saiers JE (2006) Effect of desorption kinetics on colloid facilitated

727 transport of contaminants: Cesium, strontium, and illite colloids. *Water Resour Res*

728 42:W12S09

729 Tyler A, Carter S, Davidson D, Long D, Tipping R (2001) The extent and significance of

730 bioturbation on Cs-137 distributions in upland soils. *Catena* 43:81-99

731 UNSCEAR (1982) Ionizing radiation: sources and biological effects report to the general

732 assembly, with annexes. Tech. rep., United Nations, New York

733 Vandebroek L, Van Hees M, Delvaux B, Spaargaren O, Thiry Y (2012) Relevance of

734 Radiocaesium Interception Potential (RIP) on a worldwide scale to assess soil

735 vulnerability to ¹³⁷Cs contamination. *J Environ Radioact* 104:87-93

736 VandenBygaart A, Protz R, Tomlin A, Miller J (1998) Cs-137 as an indicator of earthworm

737 activity in soils. *Appl Soil Ecol* 9:167-173

738 Wilkinson MT, Richards PJ, Humphreys GS (2009) Breaking ground: Pedological,

739 geological, and ecological implications of soil bioturbation. *Earth Sci Rev* 97:257-272

740 Zhang X, Long Y, He X, Fu J, Zhang Y (2008) A simplified (CS)-C-137 transport model for

741 estimating erosion rates in undisturbed soil. *J Environ Radioactiv* 99:1242-1246

742 Zhang X, Qi Y, Walling DE, He X, Wen A, Fu J (2006) A preliminary assessment of the

743 potential for using Pb-210(ex) measurement to estimate soil redistribution rates on

744 cultivated slopes in the Sichuan Hilly Basin of China. *Catena* 68:1-9

745 Zheng JJ, He XB, Walling D, Zhang XB, Flanagan D, Qi YQ (2007) Assessing soil erosion

746 rates on manually-tilled hillslopes in the Sichuan Hilly Basin using Cs-137 and Pb-

747 210(ex) measurements. *Pedosphere* 17:273-283

Table 1 Studies and soil profiles contained in the database and their main characteristics

WRB soil groups	Landuse or cover	Country	Sampling year	Nb of profiles	Inventory range Bq m ⁻²	Profil-class	Climate Köppen-Geiger classification	OM ^a range %	pH range	Paper
Acrisol	Grassland	China	2004	1	1300	2	Cwa	NA	NA	Zhang et al. (2008)
Arenosol	Forest	Germany	1999	1	36123	4	Cfb	6.3	7.7	Kruse-Irmer and Giani (2003)
	Grassland	Serbia	2001	1	11523	5	Cfb	3.3	5.2	Krstic et al. (2004)
Cambisol	Forest	Germany	1998-99	3	1465-28452	1	Cfb	6.2-19.6	4.1-4.4	Kruse-Irmer and Giani (2003)
	Grassland	Syria	2000	2	2814-6420	1; 5	Csa	NA	NA	Al-Masri (2006)
		Sweden	1998	1	1927	3	Cfb	NA	NA	Isaksson et al. (2001)
		Sweden	2007	1	55950	6	Dfb	22.0	5.2	Jarvis et al. (2010)
		Serbia	2001	4	9947-17842	3; 4; 6	Cfb	5.8-8.8	5.2-5.9	Krstic et al. (2004)
		Germany	1999	1	26302	4	Cfb	9.1	6.3	Kruse-Irmer and Giani (2003)
		Spain	2006	1	2771	6	Csb	NA	NA	Legarda et al. (2011)
		Sweden	1994	1	75147	3	Dfb	22.0	5.2	Rosen et al. (1999)
		Hungary	1995-97	7	2200-13500	1; 3; 4	Cfb	1.1-8.2	5.6-7.5	Szerbin et al. (1999)
Chernozem	Grassland	Hungary	1995	2	4200-6100	3; 6	Cfb	3.4-4.4	7.2-8.1	Szerbin et al. (1999)
Fluvisol	Grassland	Serbia	2001	2	4595-20632	3; 5	Cfb	1.0-7.3	4.8-7.0	Krstic et al. (2004)
		Hungary	1995	5	1300-7800	3; 4	Cfb	0.9-5.0	6.2-7.9	Szerbin et al. (1999)
Histosol	Forest	Germany	1998	1	5040	3	Cfb	93.0	4.0	Kruse-Irmer and Giani (2003)
	Grassland	Germany	1999	1	53712	1	Cfb	78.9	4.4	Kruse-Irmer and Giani (2003)
		Sweden	2007	1	111400	6	Dfb	46.3	5.9	Matisoff et al. (2011) ^b
		Sweden	1994	2	53463-165913	3	Dfb	64.6	5.5	Rosen et al. (1999)
	Peat	Ireland	NA ^a	2	3900-4800	5	Cfb	NA	NA	Gallagher et al. (2001)
Leptosol	Grassland	Hungary	1995-97	2	6000-21500	1; 4	Cfb	2.0-4.6	7.7-7.8	Szerbin et al. (1999)

Luvisol	Forest	Canada	NA	1	1610	6	Dfb	NA	7.7	Milton et al. (2001)
	Grassland	Syria	2000	2	2218-9647	1; 3	Csa	NA	NA	Al-Masri (2006)
		Germany	2001	5	18161-23640	3; 5	Cfb	12.4	NA	Schimmack and Schultz (2006)
Phaeozem	Grassland	Sweden	1992	1	159923	3	Dfb	1.1	6.2	Rosen et al. (1999)
Podzol	Forest	Canada	NA	1	3030	3	Dfb	NA	4.9	Milton et al. (2001)
	Grassland	Sweden	1994	1	2921	6	Dfc	3.0	4.9	Rosen et al. (1999)
Regosol	Cultivated	China	2002	3	898-1840	7	Cwa	NA	NA	Zhang et al. (2006)
	Forest	Turkey	2002	16	2974-21201	1; 2; 5; 6	Csa	3.8-11.0	5.1-7.5	Karadeniz and Yaprak (2008)
		Germany	1999	1	23486	2	Cfb	3.8	7.7	Kruse-Irmer and Giani (2003)
	Grassland	Germany	1999	1	50642	2	Cfb	8.4	7.4	Kruse-Irmer and Giani (2003)
		Sweden	1994-95	3	11214-47230	1; 3; 5	Dfc	7.2-7.5	5.4-6.3	Rosen et al. (1999)
		China	2002	1	1984	2	Cwa	NA	NA	Zhang et al. (2006)
		China	2004	1	2244	6	Cwa	NA	NA	Zheng et al. (2007)
Solonetz	Grassland	Hungary	1995-97	3	1300-4400	1; 4	Cfb	2.1-4.6	6.3-7.4	Szerbin et al. (1999)
Vertisol	Cultivated	Morocco	NA	1	952	7	Csa	NA	NA	Mabit et al. (2008)
	Grassland	Syria	2000	1	5646	4	Csa	NA	NA	Al-Masri (2006)
		Serbia	2001	3	12104-19975	2; 4	Cfb	3.9-6.0	5.7-7.5	Krstic et al. (2004)
		Morocco	NA	1	1445	3	Csa	NA	NA	Mabit et al. (2008)
Yermosol	Grassland	Syria	2000	7	2000-6477	1; 3; 4; 6	BSH; BSk; BWk	NA	NA	Al-Masri (2006)

^a OM stands for organic matter and NA for not analysed (or probived)

^b Four other profiles from this study were excluded as we were not able to determine the soil bulk density with the soil bulk density model and the inventories reported in the paper (negative values)

Table 2 Values of global ^{137}Cs fallout estimated by UNSCEAR (1982) for different ranges of latitude and simulated in this study for the different modelled sites (grouped according to their latitude)

Latitude band (degrees)	Estimation UNSCEAR ^a Bq m ⁻²	r_{th}	Global fallout values simulated: mean [range] Bq m ⁻²
60-70	2425	0.80	3999 [3954 - 4044] (n=2)
50-60	4028	1.34	3205 [815 - 4044] (n=9)
40-50	4502	1.50	3561 [0 - 4527] (n=44)
30-40	3261	1.09	1358 [0 - 3289] (n=13)
20-30	2467	0.82	2806 [1931 - 3531] ^b (n=3)

^a: calculated as ^{90}Sr deposition published in (UNSCEAR, 1982) times 1.6 (ratio $^{137}\text{Cs}/^{90}\text{Sr}$ in global fallout) and decay-corrected for 1986

^b: sites located in China (~29°N) hypothesized to be free of Chernobyl contamination

Table 3 Types of profile depth distributions attributed to the different soil groups

Profile depth distribution	Type 1	Type 2	Type 3	Type 4	Type 5	Type 6	Type 7
Acrisols	0	1	0	0	0	0	0
Arenosols	0	0	0	1	1	0	0
Cambisols	5	0	7	5	1	3	0
Chernozems	0	0	1	0	0	1	0
Fluvisols	0	0	4	2	1	0	0
Histosols	1	0	3	0	2	1	0
Leptosols	1	0	0	1	0	0	0
Luvisols	0	0	4	0	2	1	0
Phaeozems	0	0	1	0	0	0	0
Podzols	0	0	1	0	0	1	0
Regosols	2	6	1	0	5	9	3
Solonetz	2	0	0	1	0	0	0
Vertisols	0	2	1	2	0	0	1
Yermosols	3	0	1	1	0	3	0

Table 4 Types of profile depth distributions collected under different climates

Climate type	Class ^a	Type 1	Type 2	Type 3	Type 4	Type 5	Type 6	Type 7
Cold desert climate	BWk	1	0	0	0	0	0	0
Warm steppe climate	BSh	1	0	0	1	0	2	0
Cold steppe climate	BSk	1	0	1	0	0	1	0
Temperate climate	Cfb	8	4	15	11	6	2	0
	Csa	2	3	2	1	5	8	1
	Csb	0	0	0	0	0	1	0
	Cwa	0	2	0	0	0	1	3
Continental climate	Dfb	0	0	5	0	0	3	0
	Dfc	1	0	1	0	1	1	0

^a Köppen-Geiger climate classes determined according to Kottek et al. (2006).

Main climate: B- Arid, C- Warm Temperate, D- Snow

Precipitation: W- desert, S- steppe, f- fully humid, s-summer dry, w- winter dry

Temperature: h- hot arid, k-cold arid, a- hot summer, b- warm summer, c- cool summer

Table 5 Average and associated standard deviation of the diffusion coefficient (D_s), the convection velocity (v_s) and the mean migration depth of a Dirac distribution after 25 years for the different soil groups compiled in the database

Soil type	n	D_s cm ² yr ⁻¹	v_s cm yr ⁻¹	Migration depth cm
Acrisols	1	1.52	0.00	6.96
Arenosols	2	1.98±0.67	0.13±0.12	7.33±0.02
Cambisols	19	0.96±0.85	0.11±0.13 ^{ab}	6.81±3.06
Chernozems	2	1.06±0.95	0.46±0.29	13.42±8.05
Fluvisols	7	1.16±1.50	0.18±0.14 ^{ab}	8.34±2.73
Histosols	7	1.33±1.51	0.22±0.26 ^{ab}	9.86±4.16
Leptosols	2	1.37±1.61	0.22±0.30	9.18±9.16
Luvisols	7	0.41±0.18	0.22±0.06 ^a	7.31±0.88
Phaeozems	1	0.08	0.16	4.53
Podzols	1	0.09	0.06	2.60
Regosols	6	0.60±0.40	0.10±0.12 ^{ab}	5.76±1.73
Solonetz	3	0.89±0.69	0.03±0.05	5.36±2.69
Vertisols	5	1.18±0.67	0.05±0.04 ^b	6.56±1.59
Yermosols	8	0.84±0.86	0.19±0.19 ^{ab}	7.43±5.67

a, ab, b: ANOVA analysis performed for soil groups with more than 3 individuals with a Fisher test at 5 % of

confidence level. The ANOVA could not discriminate groups with the Fisher test for D_s and *migration depth*.

Figure captions

Fig. 1: Modelling of soil bulk densities: (a) Linear regression of mean bulk densities (open circles) and bulk density profiles (lines) documented in the database (n=13); error bars represent standard deviation in the data, (b) Comparison of simulated bulk densities with the bulk densities documented in the database

Fig. 2: The different depth-distribution profile types for ^{137}Cs compiled in the database. Typical examples are provided. The distributions were normalized by their maximum concentrations. Type 1: example of profile CMg of Kruse-Irmer and Giani (2003); type 2: example of the reference profile of Zhang et al. (2008); type 3: example of Möjsjövik from Rosen et al. (1999); type 4: example of Somogy from Szerbin et al. (1999); type 5: example of profile E from Schimmack and Schultz (2006)); type 6: example of Veszprém from Szerbin et al. (1999).

Fig. 3: Simulated versus observed ^{137}Cs values in the entire set of modeled profiles

Fig. 4: Scaled residuals (n=809) of all simulations and its moving average as a function of soil depth. Note that the moving average of the scaled residuals shifted towards positive values below 13 cm and that the scaled residuals remained systematically positive below 30 cm

Fig. 5: Relationship between fitted diffusion and convection velocity coefficients for selected soils groups: a, Yermosols; b, Cambisols; c, Luvisols; d, Regosols

Fig. 6: Relationship between fitted convection velocity and time after the main ^{137}Cs input (i.e., global fallout and Chernobyl accident)

Figure 1

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.