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1 SOILS, SEC # • RESEARCH ARTICLE

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3 **Quantification of vertical solid matter transfers in soils during pedogenesis by a multi-**
4 **tracer approach**

5

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18

19 **Abstract**

20 *Purpose* - Vertical transfer of solid matter in soils (bioturbation and translocation) is responsible
21 for changes in soil properties over time through the redistribution of most of the soil constituents
22 with depth. Such transfers are, however, still poorly quantified.

23 *Materials and methods* - In this study, we examine matter transfer in four eutric Luvisols
24 through an isotopic approach based on ^{137}Cs , $^{210}\text{Pb}_{(\text{xs})}$, and meteoric ^{10}Be . These isotopes
25 differ with respect to chemical behaviour, input histories and half-lives, which allows us
26 to explore a large time range. Their vertical distributions were modelled by a diffusion-
27 advection equation with depth-dependent parameters. We estimated a set of advection and
28 diffusion coefficients able to simulate all isotope depth distributions and validated the resulting
29 model by comparing the depth distribution of organic carbon (including $^{12/13}\text{C}$ and ^{14}C isotopes)
30 and of the 0-2 μm particles with the data.

31 *Results and discussion* – We showed that i) the model satisfactorily reproduces the organic
32 carbon, ^{13}C and ^{14}C depth distributions, indicating that organic carbon content and age can be
33 explained by transport without invoking depth-dependent decay rates; ii) translocation partly
34 explains the 0-2 μm particle accumulation in the Bt-horizon; and iii) estimates of diffusion
35 coefficients that quantify the soil mixing rate by bioturbation are significantly higher for the
36 studied plots than those obtained by ecological studies.

37 *Conclusions* - This study presents a model capable of satisfactorily reproducing the isotopic
38 profiles of several tracers and simulating the distribution of organic carbon and the translocation
39 of 0-2 μm particles.

40

41 **Keywords** 0-2 μm fraction • Bioturbation • Clay translocation • Cosmonuclides • Fallout
42 radionuclides • $\delta^{13}\text{C}$ • Organic carbon • Numerical modelling

43

44

45 **1 Introduction**

46 Vertical matter transfer processes in soils, such as bioturbation and 0-2 μm particle
47 translocation, are responsible for the evolution through time of the ecosystem services provided
48 by the soils through the redistribution of most of their components with depth, especially the
49 finest particle fractions and organic matter, which are responsible for the main properties of soil
50 surface layers. Progressive transfer of matter with depth leads to the development of texture-
51 contrasted soils (Jamagne and Pedro 1981; Bockheim and Gennadiyev 2000) and to the
52 redistribution of organic carbon with depth. Indeed, a recent state-of-the-art model of soil
53 organic carbon dynamics (e.g., Koven et al. 2013) concludes with the need to estimate carbon
54 vertical transport.

55 Despite their importance in soil formation processes and in the control of the organic carbon
56 depth distribution, the dynamics of the vertical transfer of soil matter are still poorly
57 constrained and modelled (Samouelian and Cornu, 2008). Salvador-Blanes et al. (2007)
58 reported that most of the existing models of pedogenesis failed at describing the horizon
59 formation because they did not consider the redistribution of matter with depth. To improve
60 modelling of these processes, there is a need to better quantify the transfer of matter in soils
61 and to constrain their kinetics. To this end, the measurement of tracers that are supplied to
62 the soil surface through meteoric deposition and that have a strong affinity for soil
63 constituents can provide relevant information to improve our knowledge of those processes.
64 Fallout radionuclides ^{10}Be , ^{137}Cs and $^{210}\text{Pb}_{(\text{xs})}$, which are characterized by different half-lives,
65 may be used as chronometers in this context.

66 Meteoric ^{10}Be (half-life = 1.39 Myr (Korschinek et al., 2010) is a cosmogenic nuclide created
67 in the upper atmosphere by the interaction of primary and secondary cosmic ray particles with
68 oxygen and nitrogen atoms (Lal and Peters 1967). It is delivered to the terrestrial surface
69 through wet/dry fallout. ^{137}Cs (half-life = 30.2 years) was released and dispersed during

70 thermonuclear atmospheric tests that occurred between 1953 and the 1970s and during nuclear
71 power plant accidents (Chernobyl in 1986 or Fukushima Dai-ichi in 2011). Finally, ^{210}Pb (half-
72 life = 22.3 years) is naturally present as it decays from its ultimate parent ^{238}U , and it constitutes
73 a daughter product of gaseous ^{222}Rn . Some ^{222}Rn may escape from the soil and then decay in
74 the atmosphere, producing ^{210}Pb , which then returns to the surface of the Earth through wet
75 fallout. This isotope is therefore referred to as unsupported or excess ($_{\text{xs}}$) ^{210}Pb , in contrast to
76 supported ^{210}Pb , which is naturally present in the soil. Due to their different input histories and
77 half-lives, these isotopes provide temporal constraints ranging from the duration of soil
78 formation (^{10}Be) to the duration of human activity (^{137}Cs and $^{210}\text{Pb}_{\text{(xs)}}$).
79 ^{137}Cs and ^{10}Be are known to sorb on clay particles (Tamura and Jacobs 1960; Sawhney 1972;
80 Pavich et al. 1984; Cremers et al. 1988; You et al. 1989; Takahashi et al. 1999) and $^{210}\text{Pb}_{\text{(xs)}}$ on
81 both clay particles and organic matter (Dörr and Münnich, 1989). They were therefore used in
82 the past to trace erosion (e.g. He and Walling, 1997) and bioturbation (Jarvis et al., 2010). Their
83 depth distribution is classically described by an advection diffusion in the literature (e.g. He
84 and Walling, 1997). However, to the best of our knowledge, these three tracers were rarely used
85 in combination in the literature.

86 In previous studies, Jagercikova et al. (2014a and 2015) fitted a diffusion-advection
87 equation to the depth distributions of ^{137}Cs and $^{210}\text{Pb}_{\text{(xs)}}$ and of meteoric ^{10}Be along a Luvisol
88 sequence developed on loess with varying histories of land use and soil management. ^{137}Cs and
89 $^{210}\text{Pb}_{\text{(xs)}}$ advection velocities (Jagercikova et al. 2014a) were one order of magnitude higher than
90 those of meteoric ^{10}Be (Jagercikova et al. 2015). To understand the origin of the discrepancy in
91 the diffusion-advection coefficients obtained by Jagercikova et al. (2014a) and (2015), we
92 return in this study to model rationales and initialisation conditions to infer the major
93 discrepancies. Indeed, i) diffusion-advection coefficients used for ^{137}Cs and $^{210}\text{Pb}_{\text{(xs)}}$ varied in
94 depth with organic matter and pH (Jagercikova et al. 2014a) but decreased exponentially with

95 depth for meteoric ^{10}Be (Jagercikova et al. 2015); ii) $^{137}\text{Cs}/^{210}\text{Pb}_{(\text{xs})}$ and meteoric ^{10}Be
 96 simulations were not performed for the same time period (last 60 years for ^{137}Cs and $^{210}\text{Pb}_{(\text{xs})}$)
 97 and 15 kyr for meteoric ^{10}Be); and iii) histories of isotope input differed: ^{137}Cs and $^{210}\text{Pb}_{(\text{xs})}$ had
 98 no inherited fraction, whereas more than two-thirds of the meteoric ^{10}Be was inherited
 99 (Jagercikova et al. 2015). To integrate the information provided by both ^{137}Cs and meteoric
 100 ^{10}Be in a single model, we designed a single diffusion-advection model based on the difference
 101 in input history, including inheritance, of the different isotopes to simulate the ^{137}Cs , ^{10}Be and
 102 $^{210}\text{Pb}_{(\text{xs})}$ depth-distribution profiles along four of the Luvisols, that had been measured by
 103 Jagercikova et al. (2014a and 2015).

104 We then used the obtained coefficients to estimate the solid matter transfer of both the 0-
 105 $2\ \mu\text{m}$ fraction and organic carbon. For organic carbon depth-distribution modelling, we tested
 106 as a simplifying hypothesis that carbon mineralization, i.e., carbon decay rate, is not depth-
 107 dependent. Because organic carbon dynamics are usually constrained by isotope measurements
 108 (Koven et al. 2013; Ahrens et al. 2014), ^{13}C and ^{14}C depth distributions were also modelled.

109

110 **2 Materials and methods**

111 2.1 Numerical modelling

112 2.1.1 Diffusion-advection equation and tillage mixing term

113 Numerical modelling was performed using a diffusion-advection equation including
 114 radioactive decay (He and Walling 1997; Schuller et al. 1997; Jagercikova et al. 2014a):

$$115 \quad \frac{\partial C}{\partial t} = \frac{\partial}{\partial z} \left(D_s \frac{\partial C}{\partial z} - v_s C \right) - \lambda C \quad (1)$$

116 with C being the isotopic concentration per soil volume (isotope. cm^{-3}) obtained by
 117 multiplying the isotopic concentration by the soil bulk density ($\text{g}\cdot\text{cm}^{-3}$); z the depth (cm);
 118 t the time (yr); D_s the diffusion coefficient ($\text{cm}^2\cdot\text{yr}^{-1}$); v_s the advection velocity ($\text{cm}\cdot\text{yr}^{-1}$);

119 and λ the radioactive constant (yr^{-1}) in the case of radioisotopes.

120 Both the diffusion and advection coefficients are considered to vary with depth, as follows:

$$\begin{aligned} 121 \quad v_s(z) &= v_0 + 1/20 (v_{20} - v_0) z && \text{for } z \text{ in } [0, 20] \\ 122 \quad v_s(z) &= v_{20} \exp(-dz) && \text{for } z \geq 20 \end{aligned} \quad (2)$$

123 with v_0 being the surface advection velocity, v_{20} the advection velocity at 20 cm depth and

124 d the constant of the exponential term (cm^{-1}); and

$$125 \quad D_s(z) = D_0 \exp(-bz) \quad " \quad z \quad (3)$$

126 with D_0 being the diffusion coefficient at the surface and b the constant of the exponential

127 term (cm^{-1}). The negative exponential term in the advection and diffusion coefficients

128 determines the diminution with depth. In the case of advection, this is due to decreasing

129 hydraulic conductivity in the soil profile and leads to the accumulation of the 0-2 μm particles

130 and radioisotopes (e.g., meteoric ^{10}Be , as discussed in Jagercikova et al. 2015). The decreasing

131 diffusion coefficient reflects the decreasing organic matter content on which earthworms feed

132 (Jagercikova et al. 2014a), earthworms being the main bioturbation agent in the temperate zone

133 (Gobat et al. 2004; Wilkinson et al. 2009).

134 Finally, a tillage term is introduced into the model. This term mixes annually 85 % of the soil

135 matter homogeneously over the whole tilled layer as proposed in the Soil and Water Assessment

136 Tool (SWAT) models (Ullrich and Volk 2009 and references therein) and detailed in equation

137 4.

$$138 \quad C(z, t + \Delta t) = (1 - M_R)C(z, t) + \frac{M_R}{T_D} \int_0^{T_D} C(z, t) dz \quad \forall z \in [0; T_D] \quad (4)$$

139 with M_R being the mixing ratio (85 %) and T_D the tillage depth.

140

141 2.1.2 Soil pools subjected to advection

142 Advection corresponds to the vertical transfer of the isotopes that can either occur under solute

143 form or in association with particles. Bundt et al. (2000) showed that ^{137}Cs is mainly found
144 along preferential flow paths in soils where both the majority of particle transfers (Jacobsen et
145 al. 1997; Majdalani et al. 2007) and rapid water flow occurs (Sammartino et al., 2015).
146 Accordingly, we hypothesize that only the fraction of the isotopes surrounding macropores are
147 subjected to advection. We thus assumed that all the isotopes brought by meteoric deposition
148 after the loess was deposited are subjected to advective transfer. On the other hand, the inherited
149 isotopes are supposed to be homogeneously distributed in the initial loess deposit. Thus, only
150 20 % of them are assumed to surround the macropores and are therefore as assumed to be
151 submitted to advection. As mentioned in the introduction, ^{137}Cs has no inherited fraction,
152 whereas more than two-thirds of the meteoric ^{10}Be is inherited (Jagercikova et al. 2015).
153 To summarize, we assume for each isotope and/or element that i) the isotopes brought to the
154 soil after the loess was deposited are subjected to both diffusive and advective transfers and ii)
155 for ^{10}Be and the 0-2 μm fraction, 20 % of the initial loess content is also subjected to both
156 diffusive and advective transfers, while the remaining 80 % is subjected exclusively to
157 diffusion, because bioturbation is considered to affect the entire soil mass.

158

159 *2.1.3 Numerical implementation and parameter optimization*

160 The boundary, initial conditions and isotope input history used for the simulations are reported
161 in Table 1. The differential equation (1) is solved using an upstream finite difference
162 approximation for spatial derivatives and a semi-implicit Crank-Nicholson scheme as a time-
163 solver (implementation is detailed in Jagercikova 2014a). The obtained system of algebraic
164 equations is solved by Gaussian elimination. Simulations start at the end of the loess deposition
165 period, which coincides with the assumed beginning of pedogenesis, *i.e.*, 15 kyr BP. The
166 advection-diffusion coefficients and their standard deviations are determined with a non-linear
167 least-squares minimization procedure based on the Levenberg-Marquardt algorithm. For
168 estimating the performance of the fitting procedure, the modelling efficiency (*EF*) is calculated

169 (Loague and Green 1991; see Jagercikova et al. (2014a) for further details). All parameters
170 are deduced from both ^{137}Cs and ^{10}Be profiles. Combining two isotopes with different periods
171 and input histories allows better constraint of the equation parameters than would be possible
172 using a single isotope. The long period of ^{10}Be allows precise definition of the D_0 and d terms,
173 whereas ^{137}Cs provides better information on the advection term in the upper 20 cm. The
174 obtained diffusion and advection velocity coefficients are then used to simulate the 0-2 μm
175 fraction, organic carbon concentration and $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ isotope depth distributions for
176 the different soil profiles.

177

178 2.2 Application of the model to four Luvisol profiles in northern France

179 2.2.1 Studied soil profiles

180 Four of the nine eutric Luvisol profiles studied by Jagercikova et al. (2014a and 2015) were
181 selected. These soil profiles are part of long-term experimental sites in northern France
182 dedicated to the evaluation of the impact of different farming practices on soil characteristics
183 (Table 2, Fig. 1).

184 These Luvisols developed on Weichselian (Upper Pleniglacial) loess deposited in both the
185 Somme and the Seine River basins (northern France) during glacial eras and until 15-17 kyr BP
186 (Antoine et al. 1999, 2003). They have thus undergone pedogenesis for at least the last 15 kyr
187 and developed from the same parent material under similar climatic and topographic conditions
188 (slope < 1 %). The four studied soils have different pH levels (from 8 to 6, Fig. 2) and are
189 developed on different substrates (Table 3). In Mons, Luvisols (under cropland and grassland)
190 are developed from several metres of loess and are thus well drained, whereas in Feucherolles,
191 clay and gritstone deposits are found at approximately 1.5 m depth. Finally, in Boigneville,
192 chalk is reached at approximately 80 cm depth. Luvisols in Feucherolles exhibit features of
193 morphologic degradation due to the change of parent material with depth.

194 Soils are continuously sampled along a pit side with a sampling increment ranging from 2

195 to 10 cm and following the horizon boundaries. Sampling is stopped at 1.7 m depth or when the
196 underlying parent material is reached. Bulk densities are measured using cylinders of 500 cm³,
197 with three replicates.

198

199 *2.2.2 Sample preparation and analysis*

200 *Particle size fractionation* - Collected samples are oven-dried for a week at 40°C and sieved
201 to 2 mm as recommended in the NF ISO 11464 procedure. Particle size distribution (<2 µm; 2-
202 20 µm; 20-50 µm; 50-200 µm; 200-2000 µm) is measured by wet sieving down to 50 µm and
203 according to the Robison's pipette method below 50 µm after removal of organic matter using
204 H₂O₂ at the INRA Laboratory of Soil Analysis (Arras, France).

205

206 *Organic carbon content and isotope analysis* - Organic carbon content (hereafter referred to
207 as carbon content) is determined by dry combustion (Carlo Erba NA 1500 Elemental
208 Analyzer) of samples ground to less than 200 µm, and δ¹³C is analysed by coupled mass
209 spectrometry analysis (EA-IRMS, Delta Plus, ThermoFinnigan). The δ¹³C values are
210 reported vs V-PDB, using polyethylene IAEA CH7 (δ¹³C = -32.15 ± 0.05 ‰) as a secondary
211 standard; the sucrose standard IAEA CH6 (Coplen et al., 2006) yields a mean value
212 of -10.45 ± 0.04 ‰. When containing carbonates, the samples are pretreated with repeated
213 amounts of 2 M HCl added to 100 mg of sample. To avoid the loss of acid-extracted organic
214 carbon, the whole suspension is then dried at 60°C.

215 For ¹⁴C analyses, the CO₂ gas is prepared at the LSCE - ¹⁴C lab using a protocol adapted to
216 achieve carbonate leaching without any loss of organic carbon, with ¹⁴C activity measurement
217 performed by AMS at the French LMC14 facility (Cottureau et al., 2007). The ¹⁴C activities
218 are presented in Δ (‰) notation, with measurement and sampling year corrections as
219 recommended by Stuiver and Polach (1977).

220 $\delta^{13}\text{C}$ vertical distributions are available for the Mons grassland and Boigneville cropland
221 sites, while the $\Delta^{14}\text{C}$ vertical distribution is available for the Mons grassland site only.

222

223 *2.2.3 Application to organic carbon transfer in soil*

224 Because organic carbon is a complex ensemble of molecules with different decomposition rates,
225 the proposed model accounts for soil organic carbon dynamics by considering organic carbon
226 partitioning between pools with different residence times based on the RothC model
227 (Jenkinson and Rayner 1977; Coleman and Jenkinson 1999; Jenkinson and Coleman 2008).
228 Five carbon pools are considered (Table 4): decomposable plant matter (DPM), resistant plant
229 matter (RPM), decomposers biomass (BIO), stabilized organic matter (HUM) and almost inert
230 organic matter (IOM). All RothC parameter values are set to the default values provided by
231 Coleman and Jenkinson (1999), except the parameter driving IOM, which is kept as proposed
232 by Jenkinson and Rayner (1977) (Table 4).

233 In the proposed representation, the organic matter decomposition rate evolves with depth as a
234 function of the 0-2 μm fraction content that affects the proportion of carbon allocated to the
235 BIO (P_{BIO}) and HUM (P_{HUM}) pools (Coleman and Jenkinson 1999; Table 4). The transport
236 parameters (v and D) are the same for the five dynamic pools.

237 Organic carbon input is distributed between above-ground inputs set at 70 % of the total C-
238 input and below-ground inputs consisting of the remaining 30 %. The latter are distributed
239 according to an exponential law, with 80 % of the below-ground input found in the first 20 cm
240 for grasslands and areas with maize and wheat crops (Warembourg and Paul 1977; Balesdent
241 and Balabane 1992; Swinnen et al. 1994).

242 The $\delta^{13}\text{C}$ at each depth is calculated by assuming an enrichment of the $\delta^{13}\text{C}$ associated with
243 organic matter decomposition that is fitted to -1.00010 (the fractionation factor associated
244 with heterotrophic respiration). The $^{13}\text{C}/^{12}\text{C}$ ratios of the biodegradation products allocated to

245 BIO and HUM are then calculated by isotopic mass balance. The newly formed BIO, HUM and
246 IOM are therefore enriched in ^{13}C by 0.32‰ to 0.58‰ compared to the source pool at each
247 biodegradation step.

248

249 *2.2.4 Application to 0-2 μm particle transfer in soil*

250 Two input data are needed to simulate the 0-2 μm particle transfer in soil: i) the initial 0-2 μm
251 particle content in the loess deposit, set at 24 % (Mons and Feucherolles) and 28 %
252 (Boigneville), and ii) the pool of 0-2 μm particles surrounding macropores, which corresponds
253 to the pool submitted to advection, set at 20 % as for ^{10}Be (Table 1).

254

255 **3 Results and discussion**

256 3.1 Obtained advection and diffusion coefficients

257 Evolution with depth of the advection velocity and diffusion optimized for the four sites is
258 shown in Fig. 3 and Table 5. Comparisons between simulated data and observations (^{10}Be ,
259 ^{137}Cs , and $^{210}\text{Pb}_{(\text{xs})}$ when available) are shown for Mons grassland, Boigneville cultivation,
260 Mons cultivation, and Feucherolles cultivation in Figs. 4, 5, 6 and 7, respectively. The proposed
261 model fits the isotope depth distribution with modelling efficiencies (EF) greater than 0.9 for
262 meteoric ^{10}Be and equal or greater than 0.8 for ^{137}Cs (Table 5). These efficiencies strengthen
263 confidence in the model and its rationales. $^{210}\text{Pb}_{(\text{xs})}$ is reasonably modelled for Mons grassland,
264 the only Luvisol profile having $^{210}\text{Pb}_{(\text{xs})}$ concentrations higher than the detection limit
265 (Jagercikova et al. 2014a). The fact that we could reasonably well predict the distribution of all
266 the studied isotopes with a single set of diffusion-advection coefficients suggests that the
267 isotopes are mainly transported with solid matter in the studied soils, more than as soluble form.
268 Campforts et al. (2016) recently demonstrated using a K_d modelling approach that, for soils
269 with high K_d s (i.e., for soil with pH higher than 5), 5/6 of the isotopic redistribution in depth

270 occurs through particle transfer, even on timescales as long as 10^4 years. This is typically the
271 case of the studied soils where pH does not decrease below 5 as shown in Figure 2. Pavich et
272 al. (1986), Graly et al. (2010), Jarvis et al. (2010), Willenbring and von Blanckenburg (2010),
273 and Matisoff et al. (2011) also found that the transfer of these isotopes in soils mainly occurs
274 in association with the solid matter, especially in soils with negligible contents in dissolved
275 organic carbon, as observed in the neutral eutric Luvisols developed on loess investigated in
276 the current research. Nevertheless, chemical mobility in soluble phases cannot be ruled out, and
277 the coefficients determined here are therefore considered as proxies maximizing particulate
278 transfer.

279 The advection velocity ranges from 0.12 to 0.27 cm yr^{-1} at 20 cm depth (Fig. 3, Table 5).
280 Kaste et al. (2007) have estimated advection velocities (though not depth-dependent) to vary
281 between 0.07 and 0.2 cm yr^{-1} at their study sites by conducting a combined $^{210}\text{Pb}_{(\text{xs})}$ and ^{137}Cs
282 modelling approach. Based on the modelling of ^{137}Cs vertical distributions in seven Luvisols
283 collected in different environmental conditions (Milton et al. 2001; Al-Masri 2006; Schimmack
284 and Márquez 2006), Jagercikova et al. (2014b) have found mean advection velocities of $0.22 \pm$
285 0.06 cm yr^{-1} (not depth-dependent). All these data are of the same order of magnitude.
286 Calculated advection velocities in the first 20 cm in cultivated plots are of poor significance
287 because advection is hidden by inversion tillage in these soils. The exponential decrease below
288 20 cm modelled at all sites, which matches the meteoric ^{10}Be accumulation in the Bt-horizon
289 (Figs. 4b, 5b, 6b and 7b), can be attributed to the decreasing hydraulic conductivity measured
290 at these sites (data not shown), leading to the subsequent accumulation of 0-2 μm particles.
291 Advection velocities become lower than 0.001 cm.yr^{-1} at 82 cm for grassland and at depths
292 ranging from 42 to 73 cm for cultivation sites (Table 5, Fig. 3), which correspond to Bt-
293 horizons. The mean advection velocities vary down to these depths between 0.03 and 0.09
294 cm.yr^{-1} (Table 5).

295 The surface diffusion coefficient is equal to $3.2 \pm 1.0 \text{ cm}^2.\text{yr}^{-1}$ at Mons grassland plot.
296 Although their value was not depth-dependent, Kaste et al. (2007) also estimated the diffusion
297 coefficient at $2.1 \pm 0.4 \text{ cm}^2.\text{yr}^{-1}$ for a grassland site in Tennessee valley (California). In
298 cultivated plots, the surface diffusion coefficient varies from 0.26 ± 0.06 to $40 \pm 39 \text{ cm}^2 \text{ yr}^{-1}$
299 (Table 5). This very large range of values is attributed to the prevailing effect of tillage mixing
300 on diffusion that renders the diffusion coefficient difficult to quantify. In agricultural plots,
301 below the surface layer, diffusion becomes negligible ($D_s < 0.01 \text{ cm}^2.\text{yr}^{-1}$). The associated mean
302 diffusion coefficients are difficult to quantify (Table 5). Diffusion in terms of bioturbation will
303 therefore be further discussed for the grassland plot only.

304

305 3.2 Application of transfer coefficients to modelling of the organic carbon vertical distributions

306 – Validation of the model

307 Comparisons of modelled and observed carbon contents ^{13}C and ^{14}C (when available)
308 are shown in Fig. 8 for Mons grassland and cultivation and in Fig. 9 for Boigneville and
309 Feucherolles cultivation sites. The transfer coefficients determined using ^{137}Cs and ^{10}Be are
310 applied to vertical distributions of organic carbon. The amount of the inherited carbon set by
311 estimation of the C-horizon (Table 1) has a limited effect on final simulation results and only
312 affects deep layers, because the inherited carbon is exhausted. Indeed, less than 1 % of initial
313 C remains, according to the mineralisation rate constants (Table 4). Modelling efficiencies
314 greater than 0.6 are obtained for carbon concentrations. The *EF* for $\Delta^{14}\text{C}$ is 0.87 for the
315 grassland plot in Mons, and those for $\delta^{13}\text{C}$ exceed 0.80 for the grassland in Mons and the
316 Boigneville plot (Table 6). The *EF* values show that the model is valid for the modelling of
317 organic carbon depth distribution.

318 In detail, the simulation of C, ^{13}C and ^{14}C is excellent between 30 and 70 cm in all soils (Figs.
319 8 and 9). Below this depth, the model tends to systematically underestimate the organic C

320 concentrations and $\delta^{13}\text{C}$, but not $\Delta^{14}\text{C}$. We can therefore conclude that the hypothesis of
321 constant decay rates (not depth-dependent) down to 70 cm is consistent with the observed
322 profiles. Underestimation of carbon content below 70 cm can be explained by i) an
323 underestimation of the deep root input by the exponential rooting function; ii) an improper
324 approximation of the continuum of decay rates by discrete pools (Table 4), which is intrinsic to
325 the RothC model and neglects carbon intermediate between HUM (100 yrs) and IOM (2000
326 yrs); iii) an overestimation of the decay rate or an underestimation of the initial IOM; or iv) an
327 underestimation of the advective movement in deep layers. Further analysis is needed to
328 distinguish among these different hypotheses,.

329 Furthermore, the agreement between the simulation of the carbon and the 0-2 μm particles
330 corroborates our vertical transfer model. Indeed, it illustrates the link between the carbon
331 content and the 0-2 μm fraction, as most of the organic carbon content is contained in the 0-2
332 μm fraction (Supplementary Data file).

333

334 3.3 Diffusion coefficient as an indicator of the bioturbation rate

335 The diffusion coefficient rapidly decreases with depth (Fig. 3). Diffusion coefficients are
336 classically used in the literature to approximate bioturbation (Wilkinson et al., 2009; Jarvis et
337 al., 2010). To compare the modelled diffusion obtained for the pasture plot to bioturbation data
338 based on biological indicators provided in the literature, we evaluated the mean bioturbation
339 rates for the upper 30 cm layer of the soil, which corresponds to the depth classically considered
340 in ecological studies. To this end, we used the method described in Jarvis et al. (2010) and
341 Jagercikova et al. (2014a), taking into account the mean diffusion for the upper 30 cm of the
342 pasture plot, the soil bulk density ($1.32 \text{ g}\cdot\text{cm}^{-3}$) and the earthworm length (8 cm at the grassland
343 site). The mean diffusion coefficient ranges from 1.4 to $2.9 \text{ cm}^2\cdot\text{yr}^{-1}$ for the pasture plot, and
344 the calculated bioturbation rates vary from 693 to $1,473 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$. When considering vertical

345 movement, ecological studies estimate that the bioturbation induced by anecic worms
346 dominates in temperate climates (Gobat et al. 2004; Persson et al. 2007). According to Bouche
347 (1981), anecic species consume approximately 200 times their biomass of dry matter per year,
348 thereby generating bioturbation rates ranging from 6 to 400 t.ha⁻¹.yr⁻¹ for grasslands
349 (Jagercikova et al. 2014a). The estimation provided by the multi-tracer approach is thus
350 significantly higher than those based on anecic worm biomass. In both cases, the estimated
351 ranges are very wide, and significant uncertainties remain regarding the quantification of
352 bioturbation. Our estimation based on a diffusion coefficient includes i) all processes of
353 pedoturbation, not just earthworm activity, including the processes associated with rooting (fall
354 of soil in dead root pores) or swelling and shrinking; ii) some solute transport. Accordingly,
355 this estimation provides a maximized value of (bio)turbation in soils. The discrepancies
356 observed between the two approaches must be further investigated and better understood to
357 improve the estimation of bioturbation in soils.

358

359 3.4 Application of transfer coefficients to modelling of the vertical distributions of the 0-2 µm 360 particle-size fractions

361 The determined transfer coefficients (Table 5) are applied to the vertical distribution of the
362 0-2 µm fraction in the uppermost metre of the soil profiles. Model efficiencies range from 0.88
363 to 0.97 (Table 6), according to the site. The simulations reproduce, within the range of
364 measurement uncertainties, i) the upper and lower 0-2 µm concentrations, except at the
365 Feucherolles site, where the simulated 0-2 µm concentrations in the upper part of the profile
366 are higher than the measured values, and ii) the depth of the inflexion points in the 0-2 µm
367 concentrations within a precision of 5 cm (10 cm for cropland at Boigneville) (Fig. 10).
368 Nevertheless, concentrations measured in the 0-2 µm bulge are underestimated by the model
369 (Fig. 10). The model underestimates the 0-2 µm particles accumulation in the bulge by 76 % at

370 the grassland plot. This result is, however, obtained i) for an initial 0-2 μm particle content of
371 24 % in the loess parent material, which is higher than the contents generally found in loess
372 (Table 7); ii) considering that the advection-diffusion coefficient represents particle-bound
373 transfer only, although the occurrence of some transfer under soluble form may not be entirely
374 excluded. The model underestimation of the 0-2 μm particles accumulation in the bulge is thus
375 minimal. Other processes, such as clay neo-formation, probably contributed to the formation of
376 this horizon. In addition, long-term erosion is not considered in our modelling approach, while
377 soil truncation at the surface due to human activity from the Neolithic times cannot be ruled
378 out.

379 Finally, the temporal evolution over the last 15 kyr of the 0-2 μm fraction distribution is
380 reconstructed for the grassland plot (Fig. 11 and video in Supplementary Data). After 30 years,
381 a 0-2 μm fraction bulge is formed. With time, this bulge migrates deeper and widens due to the
382 accumulation of 0-2 μm particles and the occurrence of diffusive processes. It reaches the
383 dimension and position of the present-day Bt-horizon after approximately 5 kyr. Then its
384 migration almost stagnates as it reaches a depth range where the advection velocity is low, and
385 the 0-2 μm fraction distribution does not significantly change after 9 kyr of simulation (i.e.,
386 over the last 6 kyr). This simulated age of the Bt-horizon agrees with the estimated ^{14}C age of
387 greater than 3.4 kyr. The 0-2 μm depth distribution does not significantly evolve over the last
388 5 kyr because both diffusion and advection processes are still ongoing and compensate for each
389 other. The system can be considered as having reached a steady state. Nevertheless, in this
390 model, the decarbonation and Ca^{2+} desaturation of the parent loess material are not considered.
391 These processes may take several thousands of years to occur (Finke and Hutson 2008; Quénard
392 et al. 2011) and may impede 0-2 μm particle translocation (Jamagne 1973; Quénard et al. 2011).
393 Independent simulation with SoilGen (Finke 2012) at this site with the same parent material
394 hypothesis led to estimates of 5 kyr for decarbonation of 0.5 m of loess and 6 kyr for

395 decarbonation of 1 m of loess. Because the steady state persists over at least 6 kyr in the
396 simulation, the estimated duration for decarbonation remains relevant to the duration required
397 to form the Bt-horizon generated by our simulation. At last, the steady state is disturbed by the
398 introduction of agriculture. Indeed, a noticeable change occurs in the 0-20 cm layer after the
399 introduction of tillage practices, considered to have occurred between 1700 and 1939 AD for
400 this plot.

401

402 **4 Conclusions**

403 Redistribution of ^{137}Cs , meteoric ^{10}Be , and $^{210}\text{Pb}_{(\text{xs})}$ with depth in the studied soils is
404 simulated using a diffusion-advection equation with a single set of vertically variable
405 coefficients. The resulting depth evolutions of the two coefficients are consistent with the soil
406 characteristics controlling particle mobilization and/or transfer in soils under both particulate
407 and soluble forms. The mean advection velocities vary to depths between 0.03 and 0.09 $\text{cm}\cdot\text{yr}^{-1}$
408 ¹. To be fully comprehensive, additional processes such as lateral mixing (between macropores
409 and the soil matrix), compaction, erosion and alteration should be added to the current model
410 as well as explicit consideration of both solute and particulate transfers. These improvements
411 might be achieved by including new datasets from other disciplines (past erosion rates, parent
412 material compositions and atmospheric isotopic fluxes).

413 Based on the obtained advection-diffusion coefficients, we satisfactorily model the organic
414 carbon, $\delta^{13}\text{C}$ and ^{14}C depth distributions in the studied Luvisols. The model, however, slightly
415 underestimates the carbon content below 70 cm, likely due to the organic carbon representation,
416 which is split into the 5 RothC pools. This independent verification confirms the validity of the
417 defined depth-dependent advection velocity and diffusion parameters.

418 The surface diffusion coefficient is equal to $3.2 \pm 1.0 \text{ cm}^2 \text{ yr}^{-1}$ at the Mons grassland plot.
419 When used as a proxy for bioturbation, the modelled diffusion term leads to significantly higher

420 estimates than those found in the literature and those derived from anecic and endogeic worm
421 biomass data under similar climatic conditions. Further studies should therefore be performed
422 to better understand the processes involved and to reconcile both approaches. The fact that
423 carbon profiles in the top 70 cm can be reproduced by the same solid matter transport
424 parameters as ^{137}Cs also supports the suggestion that carbon concentrations could be used with
425 ^{137}Cs to constrain the bioturbation intensity and depth in these environments. This finding is in
426 line with the common observations of field soil scientists, who diagnose the intensity of
427 bioturbation through the observation of the carbon profile.

428 Regarding clay translocation, we conclude that i) it is possible to simulate the formation of
429 a Bt-horizon at the appropriate depth by restricting modelling to vertical solid transfers and ii)
430 the 0-2 μm fraction bulge is rapidly formed (within a few decades after the beginning of the
431 simulation), demonstrating that clay translocation is a rapid process. We also highlight that
432 vertical solid matter transfer explains a maximum of 24 % of the 0-2 μm accumulation in the
433 Bt-horizon, the rest being due to other processes, such as *in situ* weathering of primary minerals
434 and clay neo-formation.

435 In conclusion, this study presents a model capable of simultaneously reproducing the
436 isotopic profiles of several tracers and simulating the organic carbon distribution down to a
437 depth of 70 cm and the 0-2 μm fraction translocation. We also coupled the 0-2 μm fraction and
438 the organic carbon depth evolution; such a coupling could improve long-term organic carbon
439 modelling.

440

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455

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627

628 **Figure captions**

629

630 **Figure 1:** Location of the study sites in Northern France (red asterix – campaign in 2011; open asterix -
631 campaign in 2012).

632

633 **Figure 2:** Measured depth distributions of a- the 0-2 μm fraction, b- organic carbon content and c- the $\text{pH}_{\text{H}_2\text{O}}$ for
634 all sampled soils. CT stands for cultivation

635

636 **Figure 3:** Evolution of a- advection velocity and b- diffusion with soil depth, as determined by the multi-isotopic
637 modelling performed on ^{137}Cs and meteoric ^{10}Be data. CT stands for cultivation.

638

639 **Figure 4:** Measured (green) and simulated (blue curves) vertical isotopic distributions at Mons grassland: a- ^{137}Cs ;
640 b- meteoric ^{10}Be ; and c- ^{210}Pb (xs)

641

642 **Figure 5:** Measured (green) and simulated (blue curves) vertical distributions at Boigneville cultivation: a- ^{137}Cs ;
643 b- meteoric ^{10}Be . Pb-210 (xs) measurements were below the detection limit

644

645 **Figure 6:** Measured (green) and simulated (blue curves) vertical distributions at Mons cultivation: a- ^{137}Cs ; b-
646 meteoric ^{10}Be . Pb-210 (xs) measurements were below the detection limit

647

648 **Figure 7:** Measured (green) and simulated (blue curves) vertical distributions at Feucherolles cultivation: a-
649 ^{137}Cs ; b- meteoric ^{10}Be . Pb-210 (xs) measurements were below the detection limit

650

651 **Figure 8:** Measured (green) and simulated (blue curves) vertical isotopic distributions at Mons of a- organic carbon
652 concentrations and RothC compartments (RPM for resistant plant matter, BIO for decomposers biomass, HUM
653 for stabilized organic matter and IOM for almost inert organic matter), with the blue curve representing the sum
654 of all compartments; b- $\delta^{13}\text{C}$ and c- $\Delta^{14}\text{C}$ for the grassland plot; and d- organic carbon concentrations and RothC
655 compartments, with blue curve representing the sum of all compartments for the cultivation plot

656

657 **Figure 9:** Measured (green) and simulated (blue curves) vertical distributions of a- organic carbon concentrations
658 and RothC compartments (RPM for resistant plant matter, BIO for decomposers biomass, HUM for stabilized
659 organic matter and IOM for almost inert organic matter), with the blue curve representing the sum of all
660 compartments and b- $\delta^{13}\text{C}$ for Boigneville cultivation; c- organic carbon concentrations and RothC compartments
661 for Feucherolles cultivation, with blue curve representing the sum of all compartments

662

663 **Figure 10:** Measured (green) and simulated (blue curves) of the 0-2 μm fraction vertical distributions considering
664 the transfer coefficient reported in Table 5: a and b- Mons grassland and cultivation, respectively; c- Feucherolles
665 cultivation; and d- Boigneville cultivation

666

667 **Figure 11:** Temporal evolution over 15,000 years of the 0-2 μm fraction vertical distribution at Mons
668 grassland based on the transfer coefficients determined in Table 5. Tillage is simulated from year 14,700 to

669 year 14,939

670

671

Table 1: Parameters used when modelling the studied isotopes (initial concentrations, input histories, and fraction of the inherited concentration subjected to advection).

Isotope and 0-2 μm	Input history reconstruction	Initial concentration or composition	Fraction of the inherited concentration subjected to advection
^{137}Cs	Nuclear weapon tests (Cambray et al., 1989) and the Chernobyl accident in 1986 (Roussel-Debel et al., 2007)	None	None
^{210}Pb (xs)	$93 \pm 8 \text{ Bq m}^{-2} \text{ yr}^{-1}$ (Jagercikova et al., 2014)	None	None
Carbon	After Davis et al., 2003, Verbruggen et al. (1996), Meteo France data and site's data	0.005 g g^{-1}	
$\delta^{13}\text{C}$	Francey et al. (1999); White and Vaughn (2011); Schmitt et al. (2012)	-23‰ (Mons and Feucherolles) and 24‰ (Boigneville)	None
$\Delta^{14}\text{C}$	Natural flux (Reimer et al., 2009) and nuclear weapon tests (Hua et al., 2013)	275.7‰ (Reimer et al., 2009)	
Meteoric ^{10}Be	Constant flux of $1.15 \times 10^6 \text{ atoms cm}^{-2} \text{ yr}^{-1}$ (Jagercikova et al., 2015)	$3.4 \times 10^8 \text{ atoms cm}^{-3}$ (Mons); $4.2 \times 10^8 \text{ atoms cm}^{-3}$ (Feucherolles); $7.6 \times 10^8 \text{ atoms cm}^{-3}$ (Boigneville) (Jagercikova et al., 2015)	20%
0-2 μm	None	240 g kg^{-1} (Mons and Feucherolles) 280 g kg^{-1} (Boigneville)	20%

Table 2: General descriptions of the study sites.

Site	Mons	Feucherolles	Boigneville
Experiment name	Essay system – ORE-ACBB ^a Mons	ORE-PRO ^b -Qualiagro	Wheat monoculture
Managing institution	INRA	INRA	Arvalis (Plant institute)
Geographic coordinates	49°52'01"N - 3°01'53" E	48°53'49"N - 1°58'19"E	2°22'58"E - 48°19'30"N
Elevation	88 m	120 m	116 m
Mean annual rainfall	680 mm	660 mm	630 mm
Mean annual temperature	11 °C	11.2 °C	10.4 °C
Considered land uses	Cultivation Grassland (since 1939)	Cultivation	Cultivation
Crop rotation	Wheat – corn – sugar beet rotation	Corn – wheat rotation	Wheat (continuous)
Liming	Not since 1986 for cultivation and 1939 for pasture	Not since 1998	Not since 1970
Sampling date	March 2011	April 2011	March 2012

^a- Observatory for Environmental Research on Agrosystems, Biogeochemical Cycles and Biodiversity

Table 3: Synthetic soil description of the different plots and sites.

Sites	Plots	Horizon						
		A	E	Degraded Bt	Bt	C/Bt	C	II
Mons	Cultivation	Silt-loam, 10YR4/4 ^a : 0-28 cm: tillage horizon, crumbly, no earthworms. 28-38 cm: base of the ancient tillage, polyhedral, few earthworms.	None	None	38-130 cm: silty clay-loam, prismatic, 10YR4/4 ^a with 7.5YR4/4 coatings, few earthworms.	130-150 cm: silt- loam, continuous, 10YR5/6, few earthworms.	> 150 cm: same characteristics as C/Bt, no earthworms.	None
	Grassland ^b	Richer in organic matter and earthworm activity, exhibiting darker colours (10 YR 3/2) and frequent earthworm casts						
Feucherolles	Cultivation	0-25(27) cm: tillage horizon, silt loam, crumbly to polyhedral, 10YR4/4 ^a . 25(27)-32(35) cm: base of the former tillage, poorly polyhedral.	32(35)-45(50) cm, silt loam, poorly polyhedral, 10YR4/6 ^a .	45(50)-75(80) cm: silty clay loam, prismatic, 7.5YR4/6 ^a with 10YR4/6 tongues.	75(80)-95(100) cm: silty clay loam, 10YR5/6 ^a with 7.5YR4/6 coatings.	None	95(100)-160 cm: silt loam, 10 YR5/6 ^a , water- logged at its base during sampling; 160-168(172) cm: carbonated Ck.	> 168 (172) cm: clays with gritstone gravels.
Boigneville	Cultivation	0-20(25) cm: tillage horizon, silt loam, crumbly, 10YR4/4 ^a ; 20(25)-35 cm, former tillage, compact, more clayey, 10YR3/3.5	None	None	34(50)-73(80) cm: silty clay loam to silty clay, prismatic, 10YR4/4 ^a with 10YR3/4 coatings.	None	None ^c	>73(80) cm: transition to another parent material.

^a According to the Munsell Soil Color Chart, 2000.

^b Only changes compared to the reference cropped profile at the same site are described.

^c A fresh loess sample could not be collected at Boigneville, where it was entirely pedogenetised.

Table 4: Proportion of the organic carbon compartment (P) and its decay rate k used for modelling the carbon dynamics in soil. All parameters are the RothC default values from Coleman and Jenkinson (1999) except P_{IOM} and k_{IOM} .

Parameters	Mons Grassland	Mons CT ^a	Feucherolles CT ^a	Boigneville CT ^a
P_{DPM}	0.4	0.59	0.59	0.59
P_{RPM}	0.6	0.41	0.41	0.41
P_{BIO}	0.099 for $z=0$ ^b	0.098 for $z=0$ ^b	0.093 for $z=0$ ^b	0.104 for $z=0$ ^b
P_{HUM}	0.116 for $z=0$ ^b	0.115 for $z=0$ ^b	0.110 for $z=0$ ^b	0.122 for $z=0$ ^b
P_{IOM}	0.001208	0.001208	0.001208	0.001208
$1/k_{DPM}$ yr	0.15	0.13	0.17	0.21
$1/k_{DPM}$ yr	5.11	4.50	5.52	7.03
$1/k_{BIO}$ yr	2.32	2.04	2.51	3.20
$1/k_{HUM}$ yr	76.6	67.5	82.8	105.5
$1/k_{IOM}$ yr	2042	3372	2759	2812

^a CT stands for cultivation

^b P_{BIO} and P_{HUM} are functions of 0-2 μm fraction content and are thus depth-dependent: Only their surface values are reported in the table.

Table 5: Advection and diffusion coefficients and modelling efficiencies obtained by fitting the ^{137}Cs and ^{10}Be vertical distributions.

Site	D_0 $\text{cm}^2 \text{yr}^{-1}$	b cm^{-1}	v_0 ($z=0$ cm) cm yr^{-1}	v_1 ($z=20$ cm) cm yr^{-1}	d cm^{-1}	$EF_{^{137}\text{Cs}}$	$EF_{^{10}\text{Be}}$
Mons							
Grassland	3.22 ± 1.02	0.030 ± 0.004	0.18 ± 0.08	0.27 ± 0.64	0.090 ± 0.004	0.78	0.99
Cultivation	0.26 ± 0.06	0.001 ± 0.003	0.03 ± 0.15	0.14 ± 0.01	0.230 ± 0.020	0.89	0.99
Feucherolles							
	1.85 ± 0.87	0.043 ± 0.008	0.00 ± 1.28	0.12 ± 0.02	0.090 ± 0.002	0.97	0.98
Boigneville							
	40.00 ± 38.65	0.139 ± 0.037	0.05 ± 0.08	0.14 ± 0.09	0.155 ± 0.017	0.97	0.89

Table 6: Modelling efficiencies (EF) of the 0-2 μm fraction, organic carbon and isotope depth redistributions, considering the transfer coefficients reported in Table 5.

Site	EF			
	0-2 μm	C	$\delta^{13}\text{C}$	$\Delta^{14}\text{C}$
Mons				
Grassland	0.95	0.60	0.89	0.87
Cultivation	0.91	0.87	NA	NA
Feucherolles	0.88	0.96	NA	NA
Boigneville	0.80	0.90	0.8	NA

Table 7: Data compilation of worldwide 0-2 μm fraction and carbonate contents in loess.

Only studies providing both loess characteristics are reported.

Reference	Place	0-2 μm	CaCO₃ %	0-2 μm % after carbonate dissolution
Antoine (pers. com.)	Beugnatre (France)	15-18	12-15	17-21
Antoire et al. (1999)	St. Pierre les Elbeuf (NW France)	14	12	16
Antoine et al. (2003)	Villiers Adam (Val d'Oise, France)	15	18	18
Bettis et al. (2003)	Columbia Plateau (USA)	12	20	15
Joret and Malterre (1947)	Mons region (France)	14	12	16
Sterckeman et al. (2006)	Nord Pas de Calais (France)	18	13	21
Pécsi (1990)	Typical loess	5-25	1-20	6-31























