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# **Long term decrease of $^{137}\text{Cs}$ bioavailability in French pastures: results from 25 years of monitoring**

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## 1 **1. Introduction**

2 The atmospheric nuclear weapons testing and the nuclear accidents such as Chernobyl and  
3 Fukushima Daiichi disasters in 1986 and 2011 respectively led to the release of huge  
4 quantities of radionuclides into the atmosphere and among them cesium 137 ( $^{137}\text{Cs}$ ), which in  
5 turn contaminated the terrestrial and aquatic environments (Ashraf et al., 2014; UNSCEAR,  
6 2000). The long term behavior of  $^{137}\text{Cs}$  is still ongoing topic of research due to its highly  
7 persistent nature, and bioavailability for vegetation uptakes(Comans et al., 1989; Cremers et  
8 al., 1988; Gillham et al., 1980; Smolders et al., 1997). The soil of pastures still represents an  
9 important sink for  $^{137}\text{Cs}$  in the contaminated territories, and thereby cesium may readily enter  
10 the food chain and reach the human body through the soil-grass-cow's milk pathway (Strebl  
11 et al., 2002).

12 Few years after a nuclear accident,  $^{137}\text{Cs}$  is retained in the surface layer of soil despite being  
13 subjected to various loss processes. Loss mechanisms at long term include radioactivity decay  
14 (radioactivity decay constant:  $2.3 \times 10^{-2} \text{ year}^{-1}$ ), uptake by plants, leaching and physical mass  
15 transport (e.g. watershed runoff and erosion) (Ehlken and Kirchner, 2002; IAEA-TECDOC-  
16 472, 2010; Mück, 2003). In addition to those processes, it is well known that the  
17 bioavailability of radiocaesium in soil decreases with time due to its diffusion into the illite  
18 lattice (Absalom et al., 1995; Smith et al., 1999), a process commonly known as "aging".  
19 Aging is a crucial process in the assessment of environmental risks as it leads to the decrease  
20 of the contamination levels in grass and thereby in the food chain.

21 Information on the decline of  $^{137}\text{Cs}$  bioavailability in pasture ecosystem components and their  
22 mathematical modeling are crucial for an accurate assessment of long term impacts after a  
23 nuclear accident. In this respect, several models were developed and applied during the last  
24 two decades in order to predict the fate and transport of radionuclides in pastures. Some

25 examples of such models are the semi-mechanistic model of Absalom et al. (2001) which  
26 predicts the activity concentrations of radiocaesium transferred to plants from the  
27 contaminated soil, the radiological risk assessment tool (ASTRAL) to account for the dose  
28 ambient rates in agricultural and forest systems (Calmon and Mourlon, 2002), and also the  
29 European Decision Support System (RODOS) for nuclear emergencies (Ehrhardt and Weis,  
30 2000). These models differ largely by their complexity, structure, assumptions, and  
31 mathematical formalisms but a common problem of determining the best sets of model  
32 parameters remains an issue (Almahayni et al., 2019). One key parameter for most of these  
33 radiological models is the effective half-life parameter which is defined as the time during  
34 which the radionuclides activity content, in an environmental component, is reduced to half  
35 by the action of all processes including the physical decay (IAEA-TECDOC-472, 2010).  
36 These parameters are often required as inputs in assessment tools and determination of their  
37 values as accurately as possible is, therefore, crucial for optimum prediction results.  
38 Estimating the values of effective half-life for  $^{137}\text{Cs}$  in pasture ecosystem components has  
39 been well treated in literature but only a few studies such as those by Corcho-Alvarado et al.  
40 (2016) and Roussel-Debet et al. (2007) have estimated their values by measurements followed  
41 on a time scale of more than 15 years after a nuclear accident. Studies over greater time may  
42 therefore be required for more realistic estimations because of the quite high persistence of  
43  $^{137}\text{Cs}$  in the environment. Additionally to this, there is a little or no data about the uncertainty  
44 associated with these parameters although this is essential information to perform  
45 probabilistic assessments. Here we present a study examining the long term effective half-  
46 **lives** values in different pasture components as a first step towards more robustness and  
47 reliability of radiological risks assessment models. The main goal was to use more than  
48 twenty five years monitoring data, collected at dozen of French pasture sites by the IRSN  
49 (Institute for Radiological and Protection Nuclear Safety), in order to estimate the average

50 values of the long term effective half-lives with their associated uncertainty in soil-grass-  
51 cow's milk components. An additional goal was then to evaluate the decrease of  $^{137}\text{Cs}$   
52 bioavailability rates that are required in radiological assessment models.

## 53 **2. Monitoring data**

### 54 **2.1. Monitoring sites**

55 Locations of the ten monitoring sites are shown in Fig.1. These sites are distributed in eastern,  
56 south-eastern and northern regions of France which are regarded as the most contaminated  
57 areas by  $^{137}\text{Cs}$  atmospheric fallouts over the French territory. Eight of the sampled sites are  
58 grazing areas located around the Electricité de France nuclear power plants (CHO, CAT,  
59 NOG, FES, BUG, CRM, CRU, TRI), while the two additional investigation sites, Mercantour  
60 (MER) and Beaune-Le-Froid (BLF), are both located in permanent prairie areas. The main  
61 sources of  $^{137}\text{Cs}$  contamination in the monitoring sites were the depositions from nuclear  
62 weapons tests and Chernobyl fallouts (Roussel-Debet et al., 2007). Additionally to these  
63 depositions, a very minor and negligible amount was also received after the Fukushima  
64 accident in spring 2011 (Masson et al., 2011; Parache et al., 2011). The contribution of  $^{137}\text{Cs}$   
65 originating from nuclear reactor releases to the total  $^{137}\text{Cs}$  content in terrestrial environment  
66 can be ignored (Duffa et al., 2004). All sites are used for cow grazing and therefore grass and  
67 cow's milk sampled from these sites are used to assess the transfer of  $^{137}\text{Cs}$  from soil to grass  
68 and from grass to milk. In winter cows are kept in barns and fed by hay harvested during the  
69 summer. Cows also receive very small amounts of supplement that contain vitamins and  
70 proteins (usually flour corn or soya). Overall, at grazing as for indoor feeding, the feeding  
71 rates did not change over time. Table 1 gives the names and the corresponding abbreviations  
72 of the sites of interest, their geographical location, the periods of observation, the target  
73 component and the corresponding ranges of  $^{137}\text{Cs}$  activities. The granulometry and soil organic

74 content in the studied sites have been summarized in Table 2. The soil is loamy with low  
75 organic matter content in all studied sites except for Nogent and Mercantour in which clay  
76 soil and organic soil predominate respectively (IAEA classification, (IAEA-TECDOC-472,  
77 2010)). The monitoring sites are, in general, all located in areas where oceanic climate  
78 prevails except for Mercantour for which high-mountain climate prevails (Pourcelot et al.,  
79 2003).

80 .

## 81 **2.2. Sampling and analytical methodology**

82 At each of the selected pastures plot (surface ranges between 2,000 and 10,000 m<sup>2</sup> about) soil  
83 from the upper 5 cm layer was taken following methodology described in Roussel-Debet et al.  
84 (2007). At the same plot, grass (stems and shoots) covering one to two square meters was  
85 collected using shears, 2-5 cm above the ground in order to prevent any contamination by soil  
86 particles. At the same time, about 5 L of cow's milk was also sampled at the farm when cow's  
87 herd was grazing in the fields. Back to the laboratory, soil sample was first sieved to remove  
88 >2 mm material. Then soil and grass samples were dried at 80 °C for 5 days. Milk sample was  
89 dried by lyophilization. Finally dried samples of grass and milk were ashed at 480°C for 24 h  
90 and crushed prior to <sup>137</sup>Cs analyses. Low level gamma spectrometry allows us to quantify  
91 <sup>137</sup>Cs activity, following methodology given by Bouisset and Calmet (1997).

## 92 **2.3. Available data and selection criteria**

93 Data used for this study were aggregated from the results of long term radioactivity  
94 monitoring programs conducted during the period from 1992 to 2017 at various monitoring  
95 sites in France. Overall, we had access to about 600 measurement data of <sup>137</sup>Cs activities  
96 across the monitoring sites continuously sampled at the same areas. Measurements were  
97 collected either from only one plot or from several plots that were located within area  
98 delineated by a circle having a diameter of less than 30 km. Given the fact that these data

99 were obtained from various studies and monitoring programs, some were incomplete or not  
100 suitable for this study purposes due to the relatively short period of their observation which  
101 was not long enough to reveal a significant decline in  $^{137}\text{Cs}$  content. To avoid potential  
102 misleading results and ensure that our parameters estimation was as reliable and accurate as  
103 possible, we set criteria for data selection. Hence, for  $^{137}\text{Cs}$  half-life estimations, the data sets  
104 that covered a time period of at least ten years with at least ten measurements were only  
105 selected. We applied later an additional criterion: the simple exponential model explained in  
106 section 3, to which our data were fitted, should be able to explain at least 50% of the data  
107 variation (i.e. the determination coefficient value  $R^2$  derived from a least squares analysis  
108 should be equal or greater than 0.5). In the light of this analysis, the simultaneous availability  
109 of data sets on  $^{137}\text{Cs}$  measurements in soil, grass and milk was limited to three sites (BLF,  
110 CHO and CRM) whereas data of the remaining sites were limited to one or two of these  
111 components (see Table 1 and Fig.1).

### 112 **3. Data modeling**

#### 113 **3.1. Theory**

114 Following a nuclear accident, contamination of grass vegetation can result from either wet/dry  
115 atmospheric deposition onto the aerial part (i.e. foliage) or by root uptake of bioavailable  
116 radiocesium from the soil layer.

117 However, the contribution of the foliar pathway becomes negligible after some years, while  
118 uptake by roots becomes the major process controlling the radiocesium activity concentration  
119 in grass but also in cow s' milk, as the ingested grass is the major source of contamination in  
120 milk at the sites investigated here (i.e. the contribution of other feedstuffs and drinking water  
121 is negligible). On the long term, it is usually assumed that radiocesium activity concentrations

122 in grass and milk are in equilibrium with the activity concentration of bioavailable  
123 radiocesium in soil (denoted  $C_{av}$  hereafter).

124 On the long term, the decrease of bioavailable radiocesium in soil after deposition can be  
125 described as a two-phase process, with a rapid initial decrease during the first few years,  
126 followed by a much slower decrease with a characteristic time greater than a decade typically.  
127 The decrease is caused mainly by the combined effect of: fixation of radiocesium on specific  
128 and non-specific sites, leaching of the upper rooting layer by downward infiltration, soil  
129 erosion and plant uptake. Mathematically, this can be expressed as follows:

$$C_{av}(t) \propto P_{fast} \cdot \exp(-(k_{fast} + \lambda) \cdot t) + (1 - P_{fast}) \cdot \exp(-(k_{slow} + \lambda) \cdot t) \quad (1)$$

130 Where  $C_{av}(t)$  is the sum of the labile and non-labile bioavailable radiocesium concentrations  
131 in the root layer at time  $t$  (in years),  $\lambda$  is the physical decay rate ( $2.31 \times 10^{-2} \text{ year}^{-1}$ ) and  $k_{fast}$   
132 and  $k_{slow}$  (in  $\text{year}^{-1}$ ) are the apparent first order kinetic rates for the fast ( $P_{fast}$ ) and slow ( $1-$   
133  $P_{fast}$ ) declining fractions respectively. Absalom et al. (1999) used a similar equation to predict  
134 the soil-to-plant transfer of radiocesium using soil characteristics. Mück (2003) had reported  
135 long term measurements of radiocesium activity in cow's milk from 4 European countries  
136 after the Chernobyl accident which showed effectively a two phase decline.

137 In case of long term monitoring that started after the initial phase has ceased (i.e.  
138  $\exp(-(k_{fast} + \lambda) \cdot t) \rightarrow 0$ ), equation (1) shows that  $C_{av}$  decays exponentially at a rate equal to  
139  $(k_{slow} + \lambda)$ . The latter can be quoted as an effective half-life of bioavailable radiocesium (in  
140 years) as follows:

$$T_{\text{eff-slow}}^{\text{av}} = \frac{\ln(2)}{k_{\text{slow}} + \lambda} \quad (2)$$

141 On the other hand, the long term rates of change of the total radiocesium content in soil (i.e.  
142 the sum of bioavailable and non-bioavailable fractions) is commonly described by assuming



143 that the decline in radiocesium concentration in soil  $C_s$  is exponential, and the corresponding  
144 effective half-life can therefore be given as follows:

$$T_{\text{eff-slow}}^{\text{soil}} = \frac{\ln(2)}{\lambda_{\text{soil}}} \quad (3)$$

145 Here,  $\lambda_{\text{soil}}$  is greater than  $\lambda$  and refers to the radiocesium loss mainly caused by physical  
146 decay, plant uptake, erosion and leaching processes. If the contribution of the environmental  
147 processes other than physical decay is small compared to the contribution of adsorption and  
148 fixation mechanisms, the total radiocesium concentration in soil would then change with a  
149 rate close to the physical decay (i.e.  $C_s \propto \lambda$ ).

150 In this work the hypothesis of exponential decline in the three different pasture components  
151 soil, grass and milk was tested by analyzing the long term decrease-of radiocesium activity  
152 concentrations in sample collected at the studied sites.

### 153 **3.2. Mathematical and statistical data analysis**

154 We estimated the values of kinetic rate constants ( $k_{\text{slow}}, \lambda_{\text{soil}}$ ) with their associated  
155 uncertainties at confidence intervals of 95% using the multiple  
156 linear regression tool of Excel software (Microsoft, USA, 2010). The coefficient of  
157 determinations ( $R^2$ ) was used as indicator of the goodness of fit. The fitting of ( $k_{\text{slow}} + \lambda$ )  
158 required for equation (2) was performed by either the slope of logarithmically transformed  
159 concentrations of  $^{137}\text{Cs}$  in grass versus time ( $\rightarrow T_{\text{eff-slow}}^{\text{av}} = T_{\text{eff-slow}}^{\text{grass}}$ ) or from the slope of  
160 logarithmically transformed concentrations of  $^{137}\text{Cs}$  in milk versus time ( $\rightarrow T_{\text{eff-slow}}^{\text{av}} =$   
161  $T_{\text{eff-slow}}^{\text{milk}}$ ). The kinetic rate  $\lambda_{\text{soil}}$  required for equation (3) was determined from the slope of  
162 logarithmically transformed concentrations of total  $^{137}\text{Cs}$  in soil versus the elapsed time.  
163 To evaluate the long term decrease of the bioavailable pool of  $^{137}\text{Cs}$ , the characteristic time of  
164 the decrease of cesium bioavailability ( $T^{\text{slow}}$ ) was calculated as follows:

$$\frac{1}{T_{\text{slow}}} = \frac{1}{T_{\text{eff-slow}}^{\text{grass}}} - \frac{1}{T_{\text{eff-slow}}^{\text{soil}}} \quad (4)$$

165 Where  $T_{\text{eff-slow}}^{\text{grass}}$ ,  $T_{\text{eff-slow}}^{\text{soil}}$  are the distributions of effective half-lives values for grass and soil  
 166 respectively averaged over the sites where experimental data are available (i.e. BLF, CHO,  
 167 CRM, FES, NOG for grass and BLF, BUG, CAT, CHO, CRM, CRU, NOG for soil). The soil-  
 168 to-grass transfer factor (TF) defined as the ratio of the  $^{137}\text{Cs}$  content in grass ( $\text{Bq} \cdot \text{kg}_{\text{dm}}^{-1}$  where  
 169 “dm” stands for dry mass) to that in soil ( $\text{Bq} \cdot \text{kg}_{\text{dm}}^{-1}$ ), was also calculated. TF for a site of  
 170 interest was calculated for each pair of soil and grass samples taken on the same monitoring  
 171 year. TF calculations were limited to the sites where experimental data were available (i.e.  
 172 BLF, CRM, CHO, NOG). Statistical analysis of the results was performed by Matlab software  
 173 package (Mathwork, USA) using the one analysis of variance (ANOVA,  $p < 0.05$ )  
 174 (Mohammed et al., 2016) in order to assess whether significant differences existed between  
 175 the distributions of  $T_{\text{eff-slow}}$  values estimated for  $^{137}\text{Cs}$  in soil, grass and milk at each of the  
 176 studied sites.

## 177 4. Results and discussion

### 178 4.1. Temporal trends in $^{137}\text{Cs}$ activity concentrations

179 Fig.2 shows the long term temporal changes observed in the activity concentrations of  $^{137}\text{Cs}$   
 180 in soil, grass and milk for the BLF (Fig.2a), CHO (Fig.2b) and CRM (Fig.2c) sites. Results  
 181 for the remaining sites are presented in Fig.A.1 in Appendix. Overall, it could be seen from  
 182 these measurements that activity concentrations for all investigated sites tend to decrease  
 183 slowly with time. The rates of decline of  $^{137}\text{Cs}$  activity concentrations in soil are generally  
 184 slower than observed in milk or grass with also smaller fluctuations over time for all studied  
 185 sites. Such observations in fact may be likely attributed to the slow irreversible fixation of  
 186  $^{137}\text{Cs}$  in soil that is frequently reported as a limiting factor of bioavailability and mobility of

187  $^{137}\text{Cs}$  (Absalom et al., 1995; Smith et al., 1999). We may also notice that the relatively large  
188 spatial variability in the measurements of  $^{137}\text{Cs}$  activity concentrations at CRM, NOG, BUG  
189 sites (where soil samples were collected from several observation plots) surprisingly does not  
190 affect the emergence of clear decline trends. However, in contrast to the trends observed in  
191 the root layer, the activity concentrations in milk and grass reveal temporal and spatial  
192 fluctuations at all investigated sites. Moreover, it is noted that the ratio between the activity  
193 concentrations measured in grass to that in milk varies largely between sites. While the  
194 activity concentrations in grass are approximately 10 and 20 times higher than those recorded  
195 in milk at the two sites CHO and CRM sites (Fig. 2b and Fig. 2c respectively), it is only two  
196 times higher at BLF site (Fig.2a). Potential reasons to explain these variations cannot be  
197 derived from this study.

#### 198 **4.2. Estimation of long term effective half-lives ( $T_{\text{eff-slow}}$ )**

199 When possible, the distributions of  $T_{\text{eff-slow}}$  values for milk, grass and soil at each investigated  
200 site were calculated. They are displayed in Fig.3. These distributions reflect, indeed, the  
201 uncertainties that were caused by both the selected model (i.e. the simple exponential  
202 equation) and measurement errors including the spatial variability in the data used. In fact,  
203 results from all studied sites demonstrate that the long term loss of  $^{137}\text{Cs}$  for the three  
204 components can be described to a good degree of significance with a simple exponential  
205 equation (see Fig. 3) as evidenced by the relatively high values of coefficient of  
206 determinations ( $R^2 > 0.65$  with p-values much below 0.05 in most cases). A good consistency  
207 could be observed between the probability distributions of  $T_{\text{eff-slow}}^{\text{milk}}$  obtained at all studied  
208 sites (see Fig.3a), except for the two sites TRI and CRU in which the uncertainties were  
209 relatively large as indicated by the ninety-five percent confidence limits. The median values  
210 are found to range between 6.1 and 12.6 years. Similar results could also be observed in the  
211 case of grass (Fig.3b) but with uncertainties larger over all investigated sites, particularly in

212 FES where the long term distributions of  $T_{\text{eff-slow}}^{\text{grass}}$  values ranged from 12.0 to 52.9 years with  
213 a median value of 19.8 years. With the exception of FES, the median values in grass ranged  
214 from 5.9 to 12.6 years which are very close to those obtained in milk. The results obtained for  
215 soil were also consistent over all sites (Fig.3c) with median values ranging between 12.0 and  
216 24.9 years (i.e. corresponding to variations of a factor of  $\sim 2$  similar to those in milk and  
217 grass). The highest uncertainties at BUG and CRU sites may be attributed to the relatively  
218 large spatial variability in the used data. In overall, it is noted that whatever the investigated  
219 site or the environmental component, the median  $T_{\text{eff-slow}}$  values estimated in soil, grass or  
220 milk are significantly lower than the physical decay (30 years), indicating therefore that the  
221 environmental conditions and their complex interactions are the factors driving the long term  
222 fate and transfer of radiocesium in pasture ecosystems.

### 223 **4.3. Comparison of $T_{\text{eff-slow}}$ at monitoring sites**

224 The variances between the distributions of effective half-life values in milk, grass and soil  
225 were evaluated independently for each studied site by means of one way analysis of variance  
226 tests (ANOVA,  $p < 0.05$ ). These tests were limited to the sites where at least 2 compartments  
227 were monitored (i.e. BLF, CRM, CRU, NOG and CHO). The  $p$  values yielded by the  
228 statistical tests are all summarized in Table A.1 in Appendix. For BLF and CRM sites,  
229 independently, significant differences appeared between the distributions of effective half-life  
230 values estimated in milk ( $T_{\text{eff-slow}}^{\text{milk}}$ ) or grass ( $T_{\text{eff-slow}}^{\text{grass}}$ ) against that estimated in soil  
231 ( $T_{\text{eff-slow}}^{\text{soil}}$ ) (ANOVA,  $p < 0.05$ ). In contrast, no such significant differences were found  
232 between  $T_{\text{eff-slow}}^{\text{milk}}$  and  $T_{\text{eff-slow}}^{\text{grass}}$  (ANOVA,  $p > 0.05$ ). Similar results were also found at CRU  
233 site with respect to the difference between grass and soil, and at CHO site with respect to the  
234 difference between grass and milk. Indeed, these statistic findings, of no significant  
235 differences between grass and milk but significant differences with respect to soil, support the

236 view that the bioavailable pool of  $^{137}\text{Cs}$  is likely controlled by the slow irreversible fixation of  
237 cesium to the soil matrix. This, in turn, limits its bioavailability for vegetation uptake and its  
238 transfer to milk for cows that are fed with locally produced vegetation. Moreover, these  
239 statistic findings are also in agreement with the conclusion frequently reported in other  
240 previous studies that the milk could be a robust indicator of contamination in grass (e.g. Mück  
241 and Gerzabek, 1995; Pourcelot et al., 2007). However, results inconsistent with these findings  
242 suggesting differences between grass and soil were found for CHO and NOG sites. These  
243 differences may be related to the variability across studied sites so that the fixation process at  
244 those two sites was less important in affecting  $^{137}\text{Cs}$  bioavailability compared to the  
245 aforementioned sites.

246 In order to evaluate the difference in half-lives values between the three environmental  
247 components over all sites, the distributions of  $T_{\text{eff-slow}}$  values averaged over all investigated  
248 sites were calculated for each environmental component using the geometric mean. They are  
249 presented in Fig.4 (bars denoted with the letter F). Calculations were made by Monte-Carlo  
250 simulations procedure using as inputs the related distributions calculated at each site.  
251 Gaussian distributions were assumed with mean and standard deviation values equal to those  
252 derived through linear regression. Hence, the distributions yielded by this procedure reflect  
253 the average variability of  $T_{\text{eff-slow}}$  values among sites. Results show that the median values of  
254 these distributions decrease in the following order soil  $\gg$  grass  $>$  milk with corresponding  
255 values of 17.0, 11.0, 9.0 years respectively. The larger uncertainties of  $T_{\text{eff-slow}}$  values in soil  
256 and grass are related to the large uncertainties in input distributions.

257 Based on joint analysis of exponential decline rates in soil versus in grass, the characteristic  
258 time of the decrease of  $^{137}\text{Cs}$  bioavailability in soil ( $T^{\text{slow}}$ ) was then calculated according to  
259 equation (4). Its median value was found to be in the order of 26 years with corresponding  
260 bioavailability decrease rate ( $k_{\text{slow}} = \ln(2)/T^{\text{slow}}$ ) of  $0.026 \text{ year}^{-1}$  (the 95 % CI ranges between

261 0.008 and 0.044 year<sup>-1</sup> with corresponding characteristic times from 13 to 61 years in soil).  
262 The impact of aging of bioavailable pool of <sup>137</sup>Cs was also shown when the soil-to-grass  
263 transfer factor (TF) values calculated within different decades were compared. The obtained  
264 TF values were found to decrease with time and to range from 1.3 x 10<sup>-2</sup> to 65.8 x 10<sup>-2</sup> with a  
265 median value of 7.7 x 10<sup>-2</sup> within the first decade after Chernobyl accident (i.e. t ≤ 1996),  
266 from 0.6 x 10<sup>-2</sup> to 5.7 x 10<sup>-2</sup> with a median value of 2.5 x 10<sup>-2</sup> within the second decade after  
267 Chernobyl accident (1997 - 2006 ) and from 1.03 x 10<sup>-2</sup> to 1.36 x 10<sup>-1</sup> with a median value of  
268 2.1 x 10<sup>-2</sup> within the third decade after Chernobyl accident (2007 - 2016 ). All TF values are  
269 in the lower bound of the range of 10<sup>-2</sup>-5.0 given by IAEA-TECDOC-472, (2010). These can  
270 be explained by the “old” contamination of monitoring sites since our <sup>137</sup>Cs measurements  
271 were performed many years after the initial deposit. It is important to stress here that our  
272 findings regarding the evolution of bioavailable fraction of <sup>137</sup>Cs with time were based on  
273 long term monitoring data (i.e. ~ 6-30 years from the Chernobyl accident) and therefore it is  
274 impossible at present time, due to absence of measurements, to predict whether the decrease  
275 in the bioavailable fraction in the very long term (i.e. >30 years) will still continue as in the  
276 long term or a steady state can be reached. More investigations are still required.

#### 277 **4.4. Comparison to other findings**

278 Roussel-Debet et al. (2007) calculated  $T_{\text{eff-slow}}^{\text{soil}}$  based on <sup>137</sup>Cs measurements collected at  
279 BLF, BUG, CAT, CHO and CRM sites through the period from 1991 to 2004. The  
280 reported  $T_{\text{eff-slow}}^{\text{soil}}$  values are given in Table 3. Although measurement have been made on the  
281 same plots, comparison of these values with our values makes obvious that our median  
282  $T_{\text{eff-slow}}^{\text{soil}}$  values are higher by approximately 1.6 to 2.8 times at all sites except at BLF where  
283  $T_{\text{eff-slow}}^{\text{soil}}$  value is lower by 7 years. This remarkable difference may be partly explained by the  
284 low goodness of fit index reported by Roussel-Debet et al, (2007)(R<sup>2</sup> of 0.2, 0.3, 0.6, 0.4, 0.4  
285 for BLF, BUG, CAT, CHO, CRM respectively).

286 Our results are then compared with the long term  $T_{\text{eff-slow}}$  values reported in the literature.  
287 Table 3 summarizes the values of  $T_{\text{eff-slow}}$  reviewed in literature from 6 different studies in  
288 cow's milk, 5 in grass and 4 in soil. For a consistent comparison, all reviewed studies were  
289 carried out in pasture plots located in Western Europe where  $T_{\text{eff-slow}}$  values were estimated by  
290 means of  $^{137}\text{Cs}$  data originated from both the nuclear weapons tests and the Chernobyl  
291 accident like in the previous section. For each environmental component, the distribution of  
292 the  $T_{\text{eff-slow}}$  values averaged over the reviewed studies were calculated by Monte Carlo  
293 simulations. For comparison purpose, the averaged long term  $T_{\text{eff-slow}}$  values yielded by these  
294 calculations were also presented in Fig.4 (bars denoted with the letter L). Similar to our  
295 findings, the averaged  $T_{\text{eff-slow}}$  using data from literature were found to decrease in the order  
296 soil >> grass > milk with median values of 15.0 , 9.0 and 8.0 years respectively and with a  
297 global tendency to underestimate slightly the values derived in this study. This slight  
298 difference of about 2-3 years may be caused by several reasons. In addition to the  
299 aforementioned reason of low goodness of fit, this difference may be attributed to the  
300 difference in the length and starting time of time series used in  $T_{\text{eff-slow}}$  estimation. Indeed,  
301 despite the finding by Mück, (2003) that the effective slow decline rate of  $^{137}\text{Cs}$  begins only 6-  
302 7 years after Chernobyl accident (1993), many studies estimated  $T_{\text{eff-slow}}$  values from time  
303 series observed before this date (Table 3). Moreover, some reported  $T_{\text{eff-slow}}$  were derived from  
304 time periods that ceases even earlier than 1993 such as the reported values by Pröhl. et al.  
305 (2006) in grass, by Fesenko et al. (1995) in milk and by Isaksson et al. (2001) in soil. Also  
306 with respect to this point, the effective half-lives given by these two latter authors of 11.7 and  
307 13.0 years respectively seemed to be not reliable since they were estimated by fitting to  
308 experimental data measured only for short time periods of about 3 years which are much less  
309 than their reported  $T_{\text{eff-slow}}$  values. In contrast to this,  $T_{\text{eff-slow}}$  values yielded by this work were  
310 estimated by data collected over longer period of almost 25 years since about 1993. Another

311 possible reason is that the  $^{137}\text{Cs}$  content in pastures might decrease progressively with time  
312 (i.e. by multiple decline phases rather than only two, fast and slow, phases) and therefore it is  
313 reasonable to assume that the longer time series led to larger  $T_{\text{eff-slow}}$  values. However, this  
314 hypothesis can be rejected by our measurements that displayed only one long term decline  
315 rate of  $^{137}\text{Cs}$  as shown in Fig. 2. Additionally to the above mentioned reasons, one may argue  
316 that this difference may be related to the difference in sampling strategy between different  
317 works. However, this was found to have no or minor influence in our case.

## 318 **5. Summary and conclusions**

319 The long term effective half-lives of radiocesium together with their associated uncertainties  
320 were estimated and analyzed by using of more than twenty five years monitoring data  
321 collected at ten of French pasture sites. Estimations were performed by a simple exponential  
322 model for describing the long-term radiocesium transfer processes in a pasture contaminated  
323 by atmospheric fallouts. On average, it was found that the median values of effective half-  
324 lives of radiocesium  $T_{\text{eff-slow}}$  were in the order of 17.0, 11.0 and 9.0 years for soil, grass and  
325 milk respectively and which were slightly greater than literature values by about 2-3 years  
326 (Fig.4). The rates of decline of radiocesium observed in milk closely correspond to those  
327 observed in grass. Based on  $T_{\text{eff-slow}}$  values obtained in grass and soil, the decrease rate of  
328  $^{137}\text{Cs}$  bioavailability in the long term was found to range between 0.008 and 0.044  $\text{year}^{-1}$ . We  
329 found that the soil to grass transfer factor values were concentrated in the lower bound of the  
330 range of values given by IAEA-TECDOC-472, (2010) and this was explained by the old  
331 contamination of monitoring sites. The question of whether the decrease in the bioavailable  
332 fraction in the very long term (i.e. >30 years) will still continue as in the long term or reach a  
333 steady state is still unsolved at the present and requires further investigations. Future work, in  
334 our opinion, should be devoted to testing of the reliability of model parameters linked to the  
335 fast decline phase  $P_{\text{fast}}$ ,  $k_{\text{fast}}$  and to estimate the uncertainties associated with their values using



336 field data collected in the first years after the Chernobyl or Fukushima accidents to enable a  
337 more accurate prediction of radiocesium contamination impacts.

338

339 **Acknowledgements**

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344

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438

439

1 **Table 1:** Main characteristics of the investigated sites

Selected sites			Observation period			<sup>137</sup> Cs activity range (min-max)		
Site	Abbreviation	Coordinates X,Y (°)	soil	grass	milk	soil (Bq.kg <sup>-1</sup> )	grass (Bq.kg <sup>-1</sup> )	milk (Bq.L <sup>-1</sup> )
Beaune-Le-Froid	BLF	2.918, 45.589	1993-2016	1993-2013	1993-2016	14.5-51.0	0.2-20.4	0.1-1.3
Bugey	BUG	5.267, 45.800	1995-2016	-	-	9.3-48.7	-	-
Cattenom	CAT	6.267, 49.417	2005-2016	-	-	9.2-18.5	-	-
Chooz	CHO	4.817, 50.100	1992-2016	1998-2016	1992-2016	8.9-32.0	0.1-1.1	0.01-0.2
Creys-Malville	CRM	5.467, 45.800	1995-2016	1996-2016	1996-2016	8.4-48.0	0.1-5.4	0.01-0.1
Cruas	CRU	4.800, 44.633	1999-2016	-	1994-2016	15.2-28.7	-	0.01-0.1
Fessenheim	FES	7.567, 47.917	-	1999-2015	-	-	0.1-0.4	-
Mercantour	MER	7.083, 44.167	-	-	1999-2017	-	-	0.1-6.2
Nogent	NOG	3.533, 48.517	1997-2012	1997-2014	-	5.7-20.7	0.1-0.4	-
Tricastin	TRI	4.732, 44.330	-	-	2000-2015	-	-	0.02-0.05

2

3 **Table 2:** Soil granulometry and organic matter content in the different investigated sites

Site	Clay (%)	Silt (%)	Sand (%)	Organic matter (%)
Beaune-Le-Froid	27.0	41.5	31.5	9.4
Bugey	18.0	23.2	58.5	5.2
Cattenom	39.2	46.3	14.5	6.4
Chooz	16.9	44.1	39.0	5.5
Creys-Malville	15.1	31.5	53.4	4.4
Cruas	10.6	37.7	52.1	2.3
Fessenheim	16.2	35.2	48.6	4.5
Mercantour	33.4	35.8	30.8	30 - 68
Nogent	35.0	38.2	26.8	8.1
Tricastin*	-	-	-	-

4 \*: No data was available for Tricastin site



5 **Table 3:** Reported long term  $T_{\text{eff-slow}}$  values in literature for  $^{137}\text{Cs}$  in milk, grass and soil of  
6 pastures

Component	$T_{\text{eff-slow}}$ [years]	Observation period	Study area	Source
<i>Milk</i>				
	5.6	1994-1998	Austria	Mück, 2003
	2.4-38.6 <sup>a</sup>	1994-2013	Switzerland	Corcho-Alvarado et al., 2016
	2.6-11.7 <sup>a</sup>	1990-1992	Russia	Fesenko et al., 1995
	7.1-9.1 <sup>a</sup>	1993-2007	Austria	Lettner et al., 2009
	4.0-6.7 <sup>a, b</sup>	1990-2003	Finland	Kostiainen, 2005
	4.4 <sup>b</sup>	1989-1999	Germany	Pröhl. et al., 2006
	(9.9) <sup>d</sup>	(1992-2017)	France	(This study)
<i>Grass</i>				
	5.6-25.1 <sup>a</sup>	1994-2013	Switzerland	Corcho-Alvarado et al., 2016
	4.1-10.0 <sup>a, b</sup>	1989-1999	Russia	IAEA, 2009
	4.1- 6.0 <sup>a, b</sup>	1990-1997	Sweden	Andersson et al., 2001
	0.8-5.7 <sup>a, b</sup>	1988-1991	Poland	Pröhl. et al., 2006
	6.4	1992-1999	Austria	Strebl et al., 2002
	(12.5) <sup>d</sup>	(1993-2016)	France	(This study)
<i>Soil</i>				
	17.8-28.2 <sup>a</sup>	1994-2013	Switzerland	Corcho-Alvarado et al., 2016
	8.0-10.0	1988-1998	Sweden	Isaksson et al., 2001
	19.4	1990-1997	Sweden	Andersson et al., 2001
	13.0	1989-1990	Italy	Astori et al., 1999
	28.8 <sup>c</sup> (21.5)	1991-2004 (1993-2016)	BLF	
	7.7 <sup>c</sup> (21.5)	1992-2004 (1995-2016)	BUG	Roussel-Debet et al., 2007
	6.7 <sup>c</sup> (14.2)	1991-2004 (2005-2016)	CAT	
	5.1 <sup>c</sup> (13.1)	1992-2004 (1992-2016)	CHO	
	7.3 <sup>c</sup> (12.0)	1992-1999 (1995-2016)	CRM	
	(18.7) <sup>d</sup>	(1992-2016)	France	(This study)

7 (<sup>a</sup>)These values have been averaged over several monitoring plots, locations, soil types or soil textures thus they  
8 may differ from those found directly in the cited papers. The two given numbers are the ninety-five percent

9 confidence limits for the average of reported values along with their associated uncertainties in the cited paper.  
10 Calculations were established based on  $10^6$  Monte-Carlo simulations and a Gaussian probability distribution.

11 (<sup>b</sup>)When the cited paper indicated that the action of the physical decay of cesium ( $T_r=30.2$  years) was not  
12 included in the calculations of their reported values ( $T_{\text{ecological-slow}}$ ), the corresponding  $T_{\text{eff-slow}}$  values were then  
13 derived as follows:  $1/T_{\text{eff-slow}}=1/T_{\text{ecological-slow}}+1/T_r$

14 (<sup>c</sup>)The data reported by Roussel-Debet et al. (2007) in soils of the same sites investigated by this work were not  
15 included in calculation of the averaged  $T_{\text{eff-slow}}$  values of literature (i.e. those presented in Fig. 4) . For  
16 comparison purpose, the median of the corresponding  $T_{\text{eff-slow}}$  values with their observation periods obtained by  
17 this study were given in brackets

18 (<sup>d</sup>) For a comparison purpose, the median of the distributions of  $T_{\text{eff-slow}}$  values averaged over all investigated  
19 sites are also given in the above table.

20

## 1 **Figure captions**

2 **Fig. 1:** Location of the selected sites with indication of the environmental component targeted  
3 by long term monitoring (i.e. root soil/grass/cow's milk). The sites are denoted as follows:  
4 CHO: Chooz; CAT: Cattenom; NOG: Nogent; FES: Fessenheim; BUG: Bugey; CRM: Creys-  
5 Malville; BLF: Beaune-le-Froid; MER: Mercantour; CRU: Cruas ; TRI:Tricastin.

6 **Fig. 2:** Time series of  $^{137}\text{Cs}$  activity concentrations in soil grass and milk at three sites a)  
7 Beaune- Le-Froid (BLF), b) Chooz (CHO) and c) Crey-Malville (CRM).  $^{137}\text{Cs}$  activity  
8 concentrations in milk are given in  $\text{Bq.L}^{-1}$  whereas  $^{137}\text{Cs}$  activity concentrations in grass and  
9 soil are given in  $\text{Bq.kg}_{\text{dm}}^{-1}$ . Lines represent the result of the linear regression analysis of the  
10 data assuming a simple exponential decline. Long term effective half-life quoted refers to the  
11 median values of these parameters (i.e. 50% confidence intervals). The error bars indicate one  
12 standard deviation.

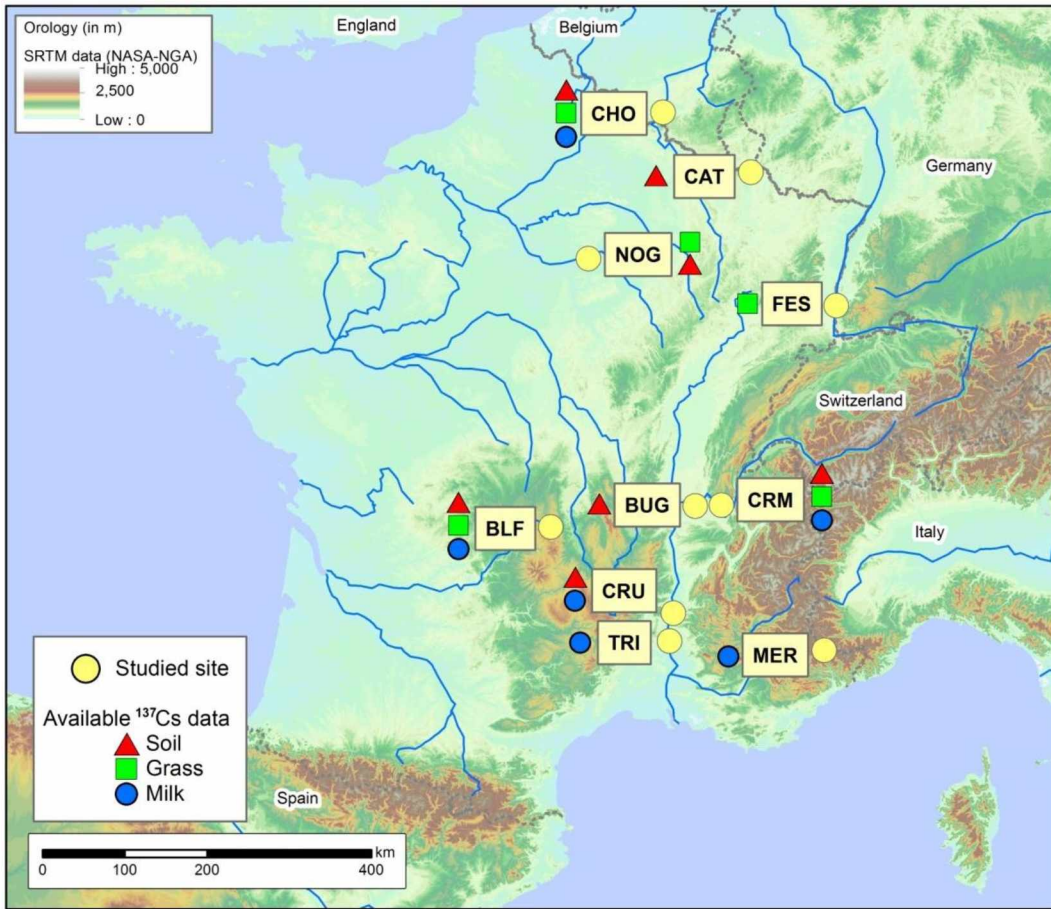
13 **Fig. 3:** Estimated long term effective half-lives ( $T_{\text{eff-slow}}$ ) of  $^{137}\text{Cs}$  in a) milk, b) grass and c)  
14 soil over the studied sites. Median values of  $T_{\text{eff-slow}}$  as well as the 5 and 95 percentiles are  
15 given to right of bars. The  $n$  value corresponds to the number of observation whereas  $R^2$  value  
16 is the goodness of fit of a simple exponential equation to the data accompanied with p-values  
17 in brackets.

18 **Fig. 4:** Comparison between the distributions of averaged  $T_{\text{eff-slow}}$  values obtained from the  
19 literature (denoted with the letter L) against those estimated in France (denoted with the letter  
20 F). Median values of  $T_{\text{eff-slow}}$  as well as the 5 and 95 percentiles are given in numbers.

21

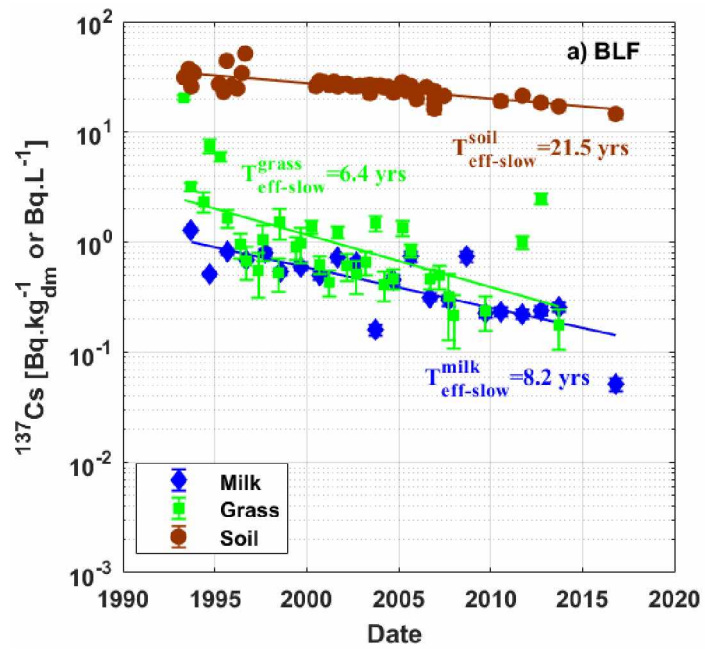
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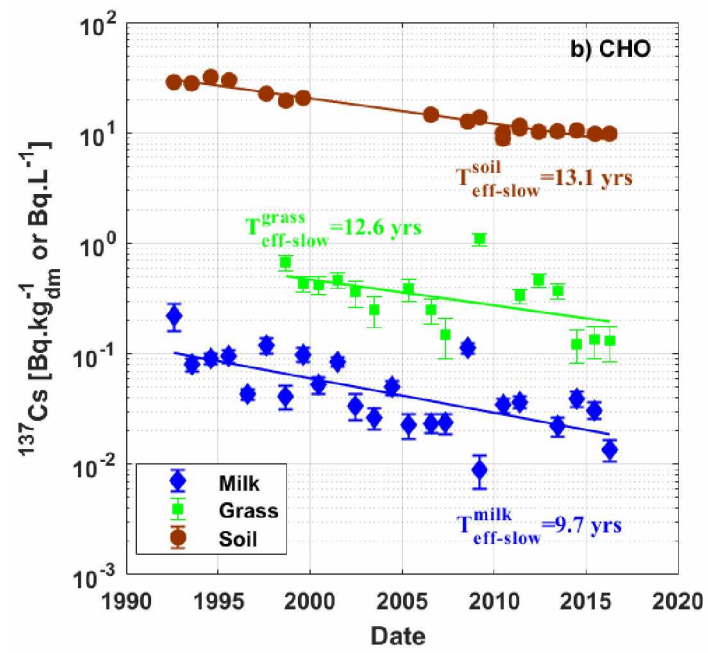


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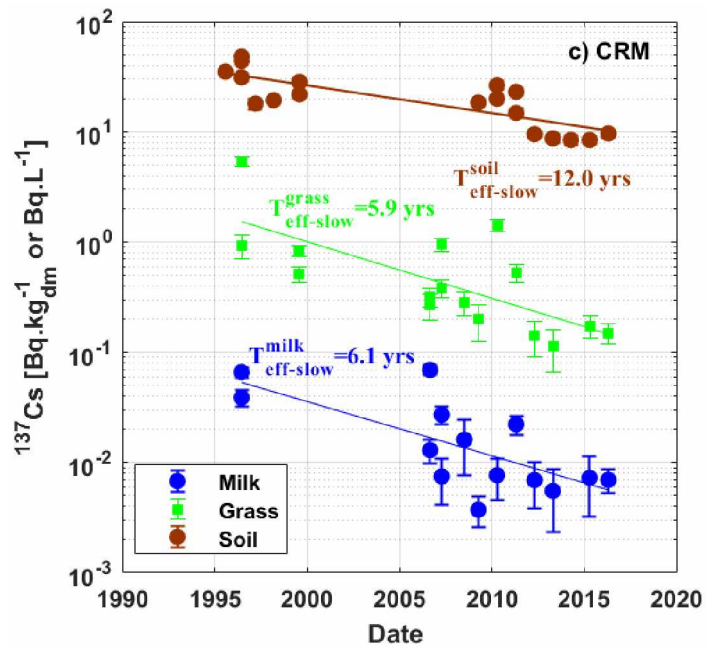
**Fig. 1**



31



32

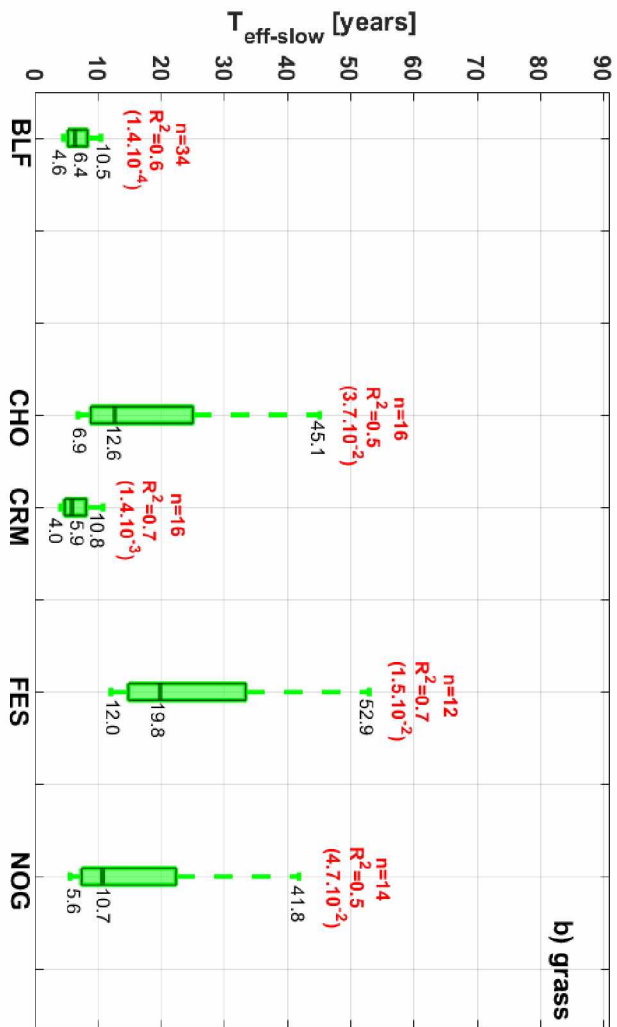
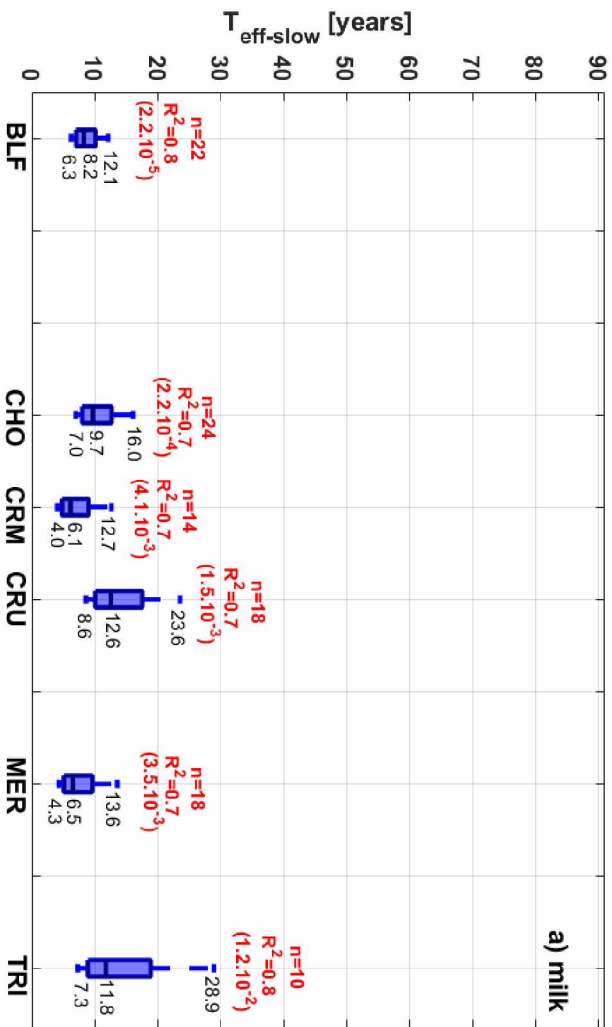


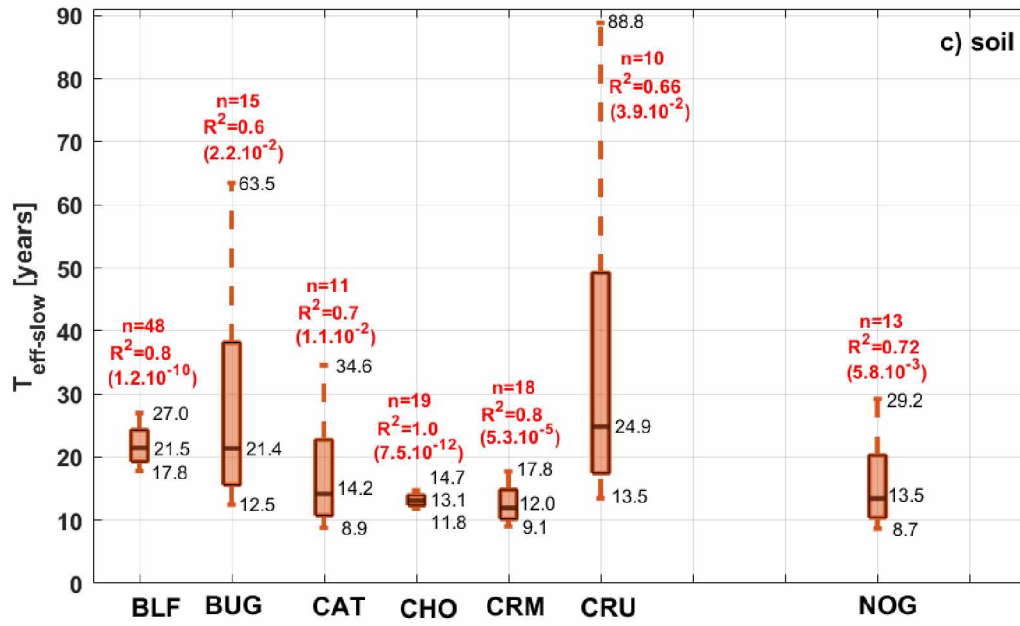
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34 Fig. 2 (a, b, c)

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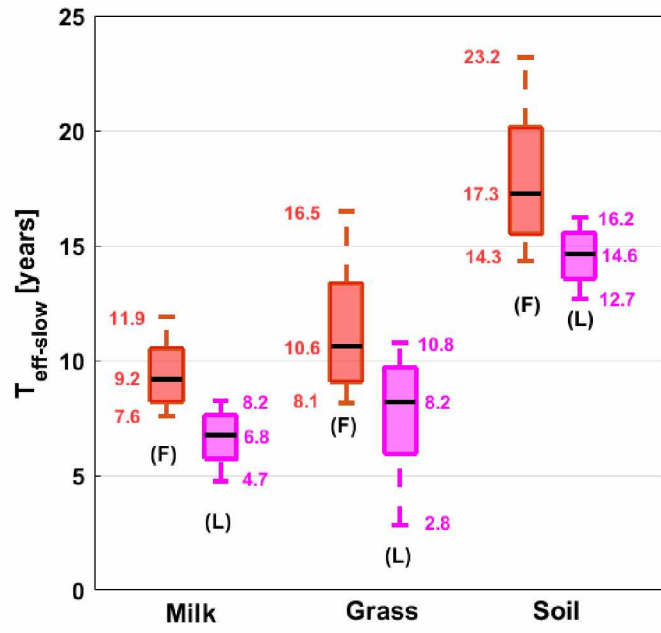
42 Fig. 3 (a, b, c)

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45



46

47 **Fig. 4**

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49

50

## **Appendix**

### **Long term decrease of $^{137}\text{Cs}$ bioavailability in French pastures: results from 25 years of monitoring**

Khaled Brimo, Marc André Gonze, Laurent Pourcelot

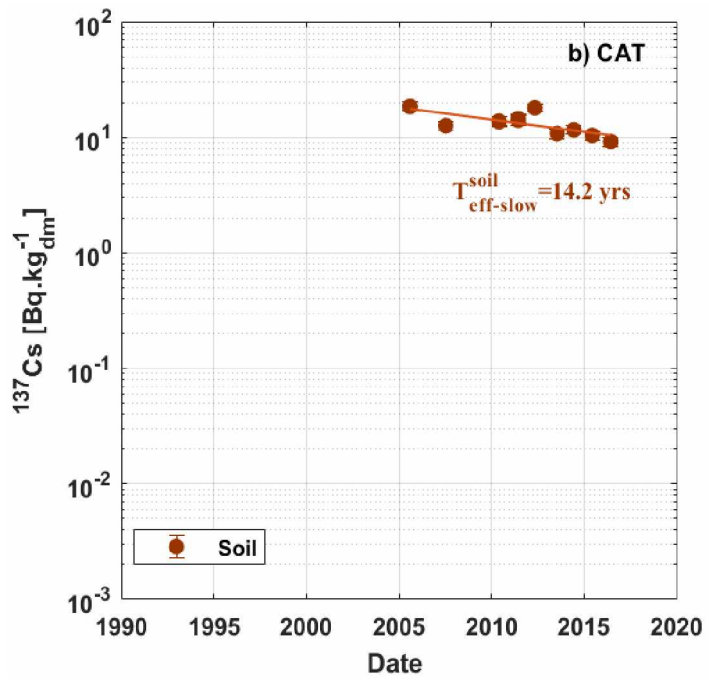
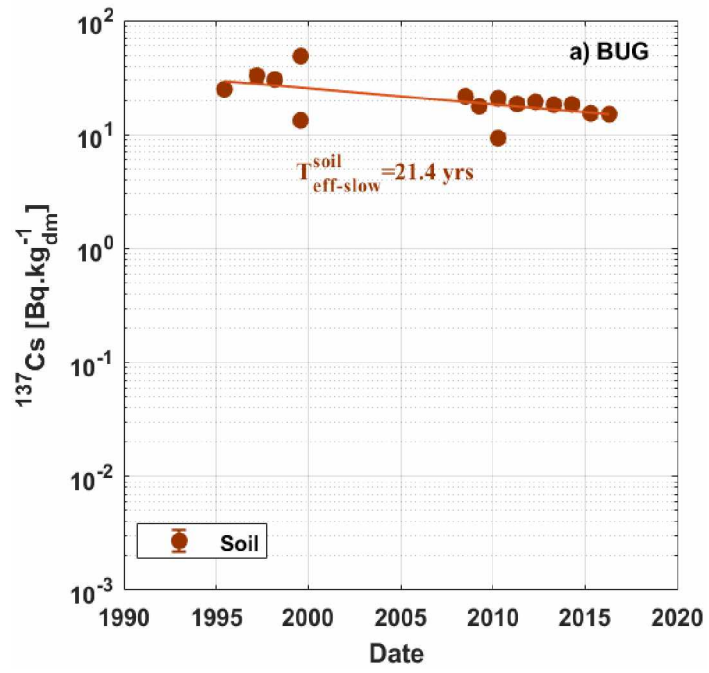
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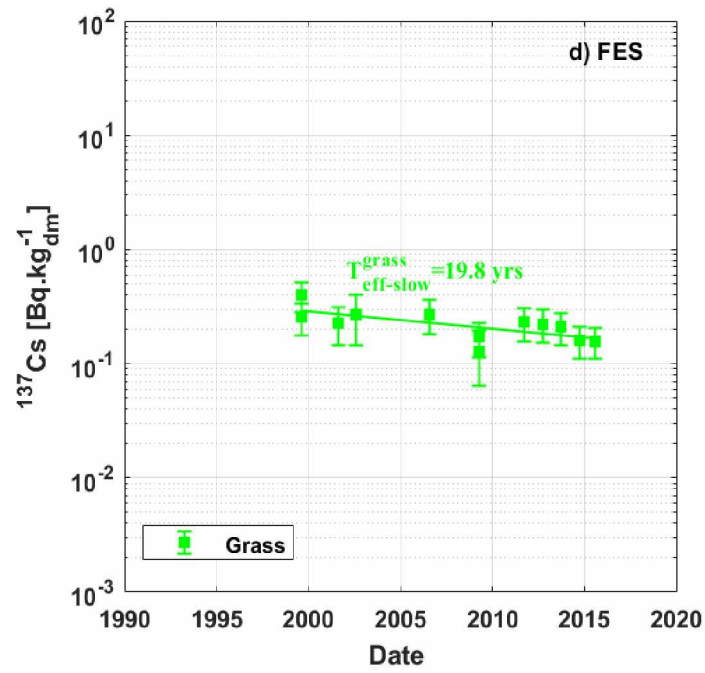
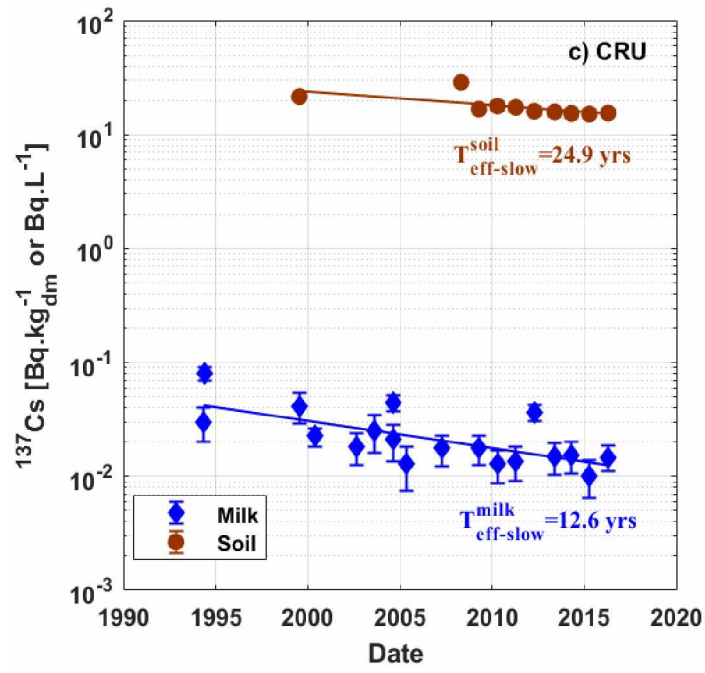
**Table A.1:** The  $p$  values yielded by one-way ANOVA for testing at each site the variances between the distributions of  $T_{\text{eff-slow}}$  values in milk, grass and soil

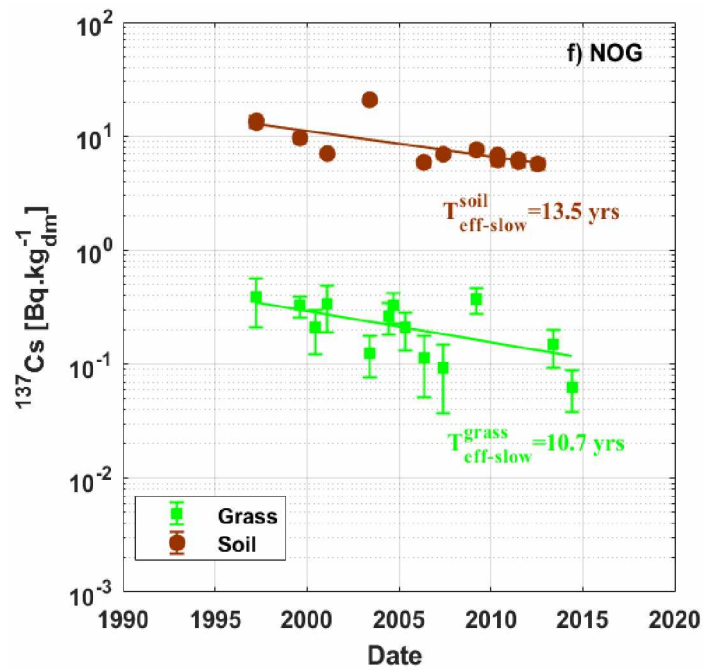
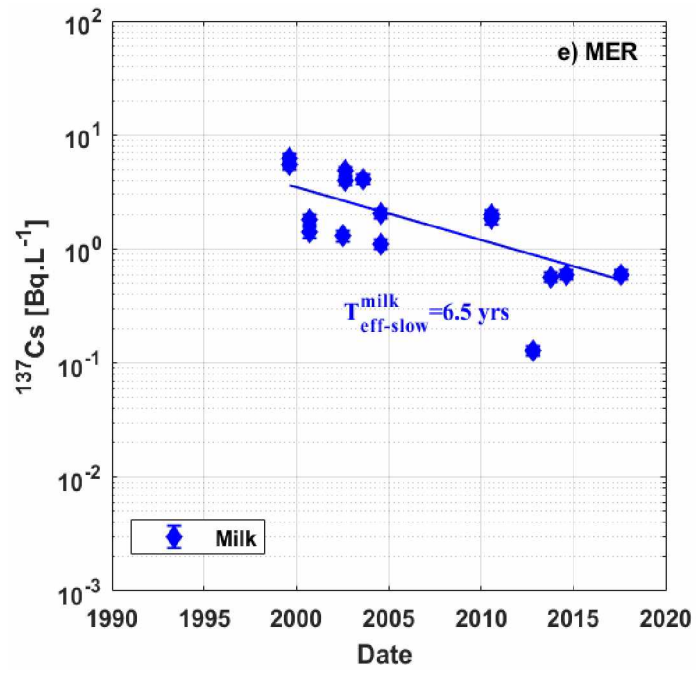
Site	$p$ value*		
	$T_{\text{eff-slow}}^{\text{grass}}$ vs. $T_{\text{eff-slow}}^{\text{milk}}$	$T_{\text{eff-slow}}^{\text{grass}}$ vs. $T_{\text{eff-slow}}^{\text{soil}}$	$T_{\text{eff-slow}}^{\text{milk}}$ vs. $T_{\text{eff-slow}}^{\text{soil}}$
BLF	0.1899	<b>0.0016</b>	<b>0.0015</b>
BUG	nd	nd	nd
CAT	nd	nd	nd
CHO	0.3368	0.9487	0.0943
CRM	0.8615	<b>0.0146</b>	<b>0.03</b>
CRU	nd	nd	<b>0.043</b>
FES	nd	nd	nd
MER	nd	nd	nd
NOG	nd	0.584	nd
TRI	nd	nd	nd

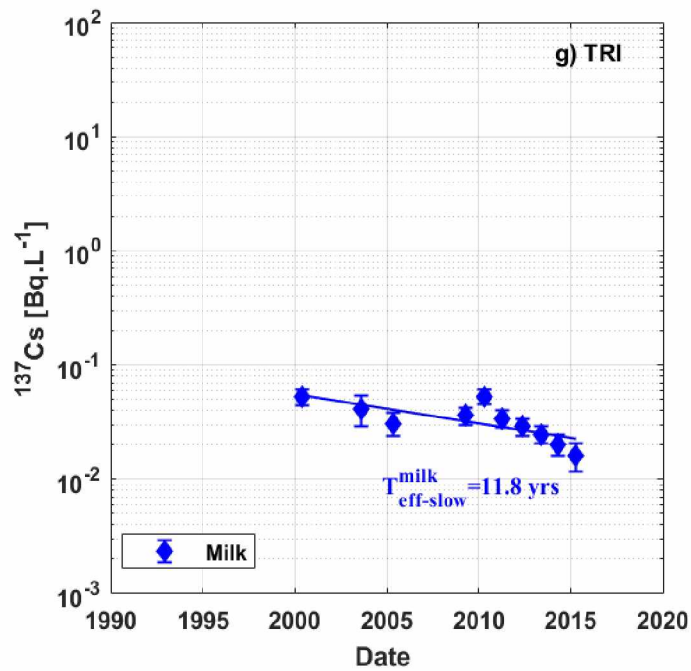
(\*) bold value indicates to a significant difference ( $p < 0.05$ )

nd: not determined because of insufficient data









**Fig. A.1:** Series of  $^{137}\text{Cs}$  activity concentrations in soil grass and milk at three sites a) Bugey (BUG), b) Cattenom (CAT), c) Cruas (CRU), d) Fessenheim (FES), e) Mercantour (MER), f) Nogent (NOG) and g) Tricastin (TRI).  $^{137}\text{Cs}$  activity concentrations in milk are given in  $\text{Bq.L}^{-1}$  whereas  $^{137}\text{Cs}$  activity concentrations in grass and soil are given in  $\text{Bq.kg}_{\text{dm}}^{-1}$ . Lines represent the result of the linear regression analysis of the data assuming a simple exponential decline. Long term effective half-life quoted refers to the median values of these parameters (i.e 50% confidence intervals). The error bars indicate one standard deviation.