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Long term decrease of ^{137}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}Cs), which in
5 turn contaminated the terrestrial and aquatic environments (Ashraf et al., 2014; UNSCEAR,
6 2000). The long term behavior of ^{137}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}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}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}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}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}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}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}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}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}Cs
65 originating from nuclear reactor releases to the total ^{137}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}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}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 m² 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 ¹³⁷Cs analyses. Low level gamma spectrometry allows us to quantify
91 ¹³⁷Cs 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 ¹³⁷Cs 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}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}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}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), λ is the physical decay rate ($2.31 \times 10^{-2} \text{ year}^{-1}$) and k_{fast}
132 and k_{slow} (in 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:

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

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, λ_{soil} is greater than λ 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_{slow} , λ_{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}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}Cs in milk versus time ($\rightarrow T_{\text{eff-slow}}^{\text{av}} =$
161 $T_{\text{eff-slow}}^{\text{milk}}$). The kinetic rate λ_{soil} required for equation (3) was determined from the slope of
162 logarithmically transformed concentrations of total ^{137}Cs in soil versus the elapsed time.
163 To evaluate the long term decrease of the bioavailable pool of ^{137}Cs , the characteristic time of
164 the decrease of cesium bioavailability (T^{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}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}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}Cs activity concentrations**

179 Fig.2 shows the long term temporal changes observed in the activity concentrations of ^{137}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}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}Cs in soil that is frequently reported as a limiting factor of bioavailability and mobility of

187 ^{137}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}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}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 ~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}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}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}Cs bioavailability in soil (T^{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 year^{-1} (the 95 % CI ranges between

261 0.008 and 0.044 year⁻¹ with corresponding characteristic times from 13 to 61 years in soil).
262 The impact of aging of bioavailable pool of ¹³⁷Cs 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 10⁻² to 65.8 x 10⁻² with a
265 median value of 7.7 x 10⁻² within the first decade after Chernobyl accident (i.e. t ≤ 1996),
266 from 0.6 x 10⁻² to 5.7 x 10⁻² with a median value of 2.5 x 10⁻² within the second decade after
267 Chernobyl accident (1997 - 2006) and from 1.03 x 10⁻² to 1.36 x 10⁻¹ with a median value of
268 2.1 x 10⁻² within the third decade after Chernobyl accident (2007 - 2016). All TF values are
269 in the lower bound of the range of 10⁻²-5.0 given by IAEA-TECDOC-472, (2010). These can
270 be explained by the “old” contamination of monitoring sites since our ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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)(R² 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}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}Cs begins only 6-
302 7years 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}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}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}Cs bioavailability in the long term was found to range between 0.008 and 0.044 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_{fast} , k_{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

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344

345 **References**

- 346 Absalom, J.P., Young, S.D., Crout, N.M.J., 1995. Radio- caesium fixation dynamics: measurement in
347 six Cumbrian soils. *Eur. J. Soil Sci.* 46, 461–469. <https://doi.org/10.1111/j.1365->
348 2389.1995.tb01342.x
- 349 Absalom, J.P., Young, S.D., Crout, N.M.J., Nisbet, A.F., Woodman, R.F.M., Smolders, E., Gillett,
350 A.G., 1999. Predicting soil to plant transfer of radiocesium using soil characteristics. *Environ.*
351 *Sci. Technol.* 33, 1218–1223.
- 352 Absalom, J.P., Young, S.D., Crout, N.M.J., Sanchez, A., Wright, S.M., Smolders, E., Nisbet, A.F.,
353 Gillett, A.G., 2001. Predicting the transfer of radiocaesium from organic soils to plants using soil
354 characteristics. *J. Environ. Radioact.* 52, 31–43.
- 355 Almahayni, T., Beresford, N.A., Crout, N.M.J., Sweeck, L., 2019. Fit-for-purpose modelling of
356 radiocaesium soil-to-plant transfer for nuclear emergencies: a review. *J. Environ. Radioact.* 201,
357 58–66. <https://doi.org/10.1016/j.jenvrad.2019.01.006>
- 358 Ashraf, M.A., Akib, S., Jamil, M., 2014. Cesium-137: radio-chemistry, fate, and transport,
359 remediation, and future concerns. *Crit. Rev. Environ. Sci. Technol.* 44, 1740–1793.
360 <https://doi.org/10.1080/10643389.2013.790753>
- 361 Astori, E., Bianchi, D., Cammarata, E., Gastaldo, S., Marzolla, N., Rabbia, G., Vescovi, A., Priano, L.,
362 1999. Surface contamination of radiocesium measured and calculated in South Piemonte (Italy).
363 *J. Environ. Radioact.* 45, 29–38. [https://doi.org/10.1016/S0265-931X\(98\)00071-X](https://doi.org/10.1016/S0265-931X(98)00071-X)
- 364 Bouisset, P., Calmet, D., 1997. Hyper pure gamma-ray spectrometry applied to low-level
365 environmental sample measurements, in: *Proceedings of the International Workshop on the*
366 *Status of Measurement Techniques for the Identification of Nuclear Signatures.*
- 367 Calmon, P., Murlon, C., 2002. Equations et paramètres du logiciel ASTRAL v2. IRSN. Rapport
368 DPRE/SERLAB/LMODE no 02-29, p. 99.
- 369 Comans, R.N.J., Middelburg, J.J., Zonderhuis, J., Woittiez, J.R.W., Lange, G.J. De, Das, H.A.,

370 Weijden., C.H. Van Der, 1989. Mobilization of radiocaesium in pore water of lake sediments.
371 Nature 339, 367–369.

372 Corcho-Alvarado, J.A., Balsiger, B., Sahli, H., Astner, M., Byrde, F., Röllin, S., Holzer, R.,
373 Mosimann, N., Wüthrich, S., Jakob, A., Burger, M., 2016. Long-term behavior of ⁹⁰Sr and
374 ¹³⁷Cs in the environment : Case studies in Switzerland. J. Environ. Radioact. 160, 54–63.
375 <https://doi.org/10.1016/j.jenvrad.2016.04.027>

376 Cremers., A., Elsen., A., Preter., P. De, Maes., A., 1988. Quantitative analysis of radiocaesium
377 retention in soils. Nature 335, 247–249. <https://doi.org/10.1038/335247a0>

378 Duffa, C., Masson, M., Gontier, G., Claval, D., Renaud, P., 2004. Synthèse des études
379 radioécologiques annuelles menées dans l'environnement des centrales électronucléaires
380 françaises depuis 1991. Radioprotection 39, 233–254.

381 Ehlken, S., Kirchner., G., 2002. Environmental processes affecting plant root uptake of radioactive
382 trace elements and variability of transfer factor data: a review. J. Environ. Radioact. 58, 97–112.
383 [https://doi.org/10.1016/S0265-931X\(01\)00060-1](https://doi.org/10.1016/S0265-931X(01)00060-1)

384 Ehrhardt., J., Weis., A., 2000. RODSOS: Decision support for off-site nuclear emergency management
385 in Europe. European commission, Brussels, Report EUR 19144.

386 Fesenko, S. V., Alexakhin, R.M., Spiridonov, S.I., Sanzharova, N.I., 1995. Dynamics of ¹³⁷Cs
387 concentration in agricultural products in areas of Russia contaminated as a result of the accident
388 at the Chernobyl nuclear power plant. Radiat. Prot. Dosimetry 60, 155–166.
389 <https://doi.org/10.1093/oxfordjournals.rpd.a082712>

390 Gillham, R.W., Cherry, J.A., Lindsay, L.E., 1980. Cesium distribution coefficients in unconsolidated
391 geological materials. Health Phys. 39, 637–49. [https://doi.org/10.1097/00004032-198010000-](https://doi.org/10.1097/00004032-198010000-00006)
392 00006

393 IAEA-TECDOC-472, 2010. Handbook of parameter values for the prediction of radionuclide transfer
394 in terrestrial and freshwater. Vienna.

395 Isaksson, M., Erlandsson, B., Mattsson, S., 2001. A 10-year study of the ^{137}Cs distribution in soil and
396 a comparison of Cs soil inventory with precipitation-determined deposition. *J. Environ. Radioact.*
397 55, 47–59. [https://doi.org/10.1016/S0265-931X\(00\)00186-7](https://doi.org/10.1016/S0265-931X(00)00186-7)

398 Masson, O., Baeza, A., Bieringer, J., Brudecki, K., Bucci, S., Cappai, M., Carvalho, F.P., Connan, O.,
399 Cosma, C., Dalheimer, A., Didier, D., Depuydt, G., Geer, L.E. De, Vismes, A. De, Gini, L.,
400 Groppi, F., Hammond, À.D., Hanley, O.O., Hole, K., Gudnason, K., Gurriaran, À.R., Hainz, D.,
401 Homoki, Z., Ioannidou, A., Isajenko, K., Jankovic, M., Katzlberger, C., Kettunen, M., Kierepko,
402 Q.R., Kontro, R., Kwakman, P.J.M., Lecomte, M., Vintro, L.L., Lepp, A., Lind, X.B., Lujanienė,
403 G., Ginnity, P.M., Mahon, C.M., Mal, H., Manenti, S., Manolopoulou, M., Mattila, A., Mäuring,
404 A., Mietelski, J.W., Möller, B., Nielsen, S.P., Nikolic, J., Overwater, R.M.W., Reis, M.C.,
405 Ringer, W., 2011. Tracking of airborne radionuclides from the damaged Fukushima Dai-Ichi
406 nuclear reactors by European networks. *Environ. Sci. Technol.* 45, 7670–7677.

407 Mohammed, G., Trolard, F., Bourrié, G., Gillon, M., Tronc, D., 2016. A long-term data sequence (
408 1960-2013) to analyse the sustainability of hay quality in irrigated permanent grasslands under
409 climate change. *Am. J. Agric. For.* 4, 140–151. <https://doi.org/10.11648/j.ajaf.20160406.11>

410 Mück, K., 2003. Sustainability of radiologically contaminated territories. *J. Environ. Radioact.* 65,
411 109–130. [https://doi.org/10.1016/S0265-931X\(02\)00091-7](https://doi.org/10.1016/S0265-931X(02)00091-7)

412 Mück, K., Gerzabek, M.H., 1995. Trends in caesium activity concentrations in milk from agricultural
413 and semi-natural environments after nuclear fallout. *Die Bodenkultur J. für Landwirtsch. Forsch.*
414 46, 337–353.

415 Parache, V., Pourcelot, L., Orjollet, D., Leblanc, F., Soria, C., Gurriaran, R., Renaud, P., Masson, O.,
416 2011. Transfer of ^{131}I from Fukushima to the vegetation and milk in France. *Environ. Sci.*
417 *Technol.* 45, 9998–10003.

418 Pourcelot, L., Louvat, D., Gauthier-Lafaye, F., Stille, P., 2003. Formation of radioactivity enriched
419 soils in mountain areas. *J. Environ. Radioact.* 68, 215–233.

420 Pourcelot, L., Steinmann, P., Froidevaux, P., 2007. Lower variability of radionuclide activities in
421 upland dairy products compared to soils and vegetation : Implication for environmental survey.
422 Chemosphere 66, 1571–1579. <https://doi.org/10.1016/j.chemosphere.2006.08.008>

423 Pröhl., G., Ehlken., S., Fiedler., I., Kirchner., G., Klemt., E., Zibold., G., 2006. Ecological half-lives of
424 ⁹⁰Sr and ¹³⁷Cs in terrestrial and aquatic ecosystems. J. Environ. Radioact. 91, 41–72.
425 <https://doi.org/10.1016/j.jenvrad.2006.08.004>

426 Roussel-Debet, S., Renaud, P., Métivier, J., 2007. ¹³⁷Cs in French soils : Deposition patterns and 15-
427 year evolution. Sci. Total Environ. 374, 388–398. <https://doi.org/10.1016/j.scitotenv.2006.12.037>

428 Smith, J.T., Fesenko, S. V., Howard, B.J., Horrill, A.D., Sanzharova, N.I., Alexakhin, R.M., Elder,
429 D.G., Naylor, C., 1999. Temporal change in fallout ¹³⁷Cs in terrestrial and aquatic systems : A
430 whole ecosystem approach. Environ. Sci. Technol. 33, 49–54. <https://doi.org/10.1021/es980670t>

431 Smolders, E., Van Den Brande, K., Merckx, R., 1997. Concentrations of ¹³⁷Cs and K in soil solution
432 plant availability of ¹³⁷Cs in soils. Environ. Sci. Technol. 31, 3432–3438.
433 <https://doi.org/10.1021/es970113r>

434 Strebl, F., Ringer, W., Gerzabek, M.H., 2002. Radiocaesium contamination of meadow vegetation -
435 Time-dependent variability and influence of soil characteristics at grassland sites in Austria. J.
436 Environ. Radioact. 58, 143–161. [https://doi.org/10.1016/S0265-931X\(01\)00063-7](https://doi.org/10.1016/S0265-931X(01)00063-7)

437 UNSCEAR, 2000. Sources and effects of ionizing radiation. New York.

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439

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

| Selected sites | | | Observation period | | | ¹³⁷ Cs activity range (min-max) | | |
|-----------------|--------------|---------------------|--------------------|-----------|-----------|--|---------------------------------|-------------------------------|
| Site | Abbreviation | Coordinates X,Y (°) | soil | grass | milk | soil (Bq.kg ⁻¹) | grass (Bq.kg ⁻¹) | milk (Bq.L ⁻¹) |
| 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}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 ^a | 1994-2013 | Switzerland | Corcho-Alvarado et al., 2016 |
| | 2.6-11.7 ^a | 1990-1992 | Russia | Fesenko et al., 1995 |
| | 7.1-9.1 ^a | 1993-2007 | Austria | Lettner et al., 2009 |
| | 4.0-6.7 ^{a, b} | 1990-2003 | Finland | Kostiainen, 2005 |
| | 4.4 ^b | 1989-1999 | Germany | Pröhl. et al., 2006 |
| | (9.9) ^d | (1992-2017) | France | (This study) |
| <i>Grass</i> | | | | |
| | 5.6-25.1 ^a | 1994-2013 | Switzerland | Corcho-Alvarado et al., 2016 |
| | 4.1-10.0 ^{a, b} | 1989-1999 | Russia | IAEA, 2009 |
| | 4.1- 6.0 ^{a, b} | 1990-1997 | Sweden | Andersson et al., 2001 |
| | 0.8-5.7 ^{a, b} | 1988-1991 | Poland | Pröhl. et al., 2006 |
| | 6.4 | 1992-1999 | Austria | Strebl et al., 2002 |
| | (12.5) ^d | (1993-2016) | France | (This study) |
| <i>Soil</i> | | | | |
| | 17.8-28.2 ^a | 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 ^c (21.5) | 1991-2004 (1993-2016) | BLF | |
| | 7.7 ^c (21.5) | 1992-2004 (1995-2016) | BUG | Roussel-Debet et al., 2007 |
| | 6.7 ^c (14.2) | 1991-2004 (2005-2016) | CAT | |
| | 5.1 ^c (13.1) | 1992-2004 (1992-2016) | CHO | |
| | 7.3 ^c (12.0) | 1992-1999 (1995-2016) | CRM | |
| | (18.7) ^d | (1992-2016) | France | (This study) |

7 (^a)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 (^b)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 (^c)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 (^d) 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}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}Cs activity
8 concentrations in milk are given in Bq.L^{-1} whereas ^{137}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.

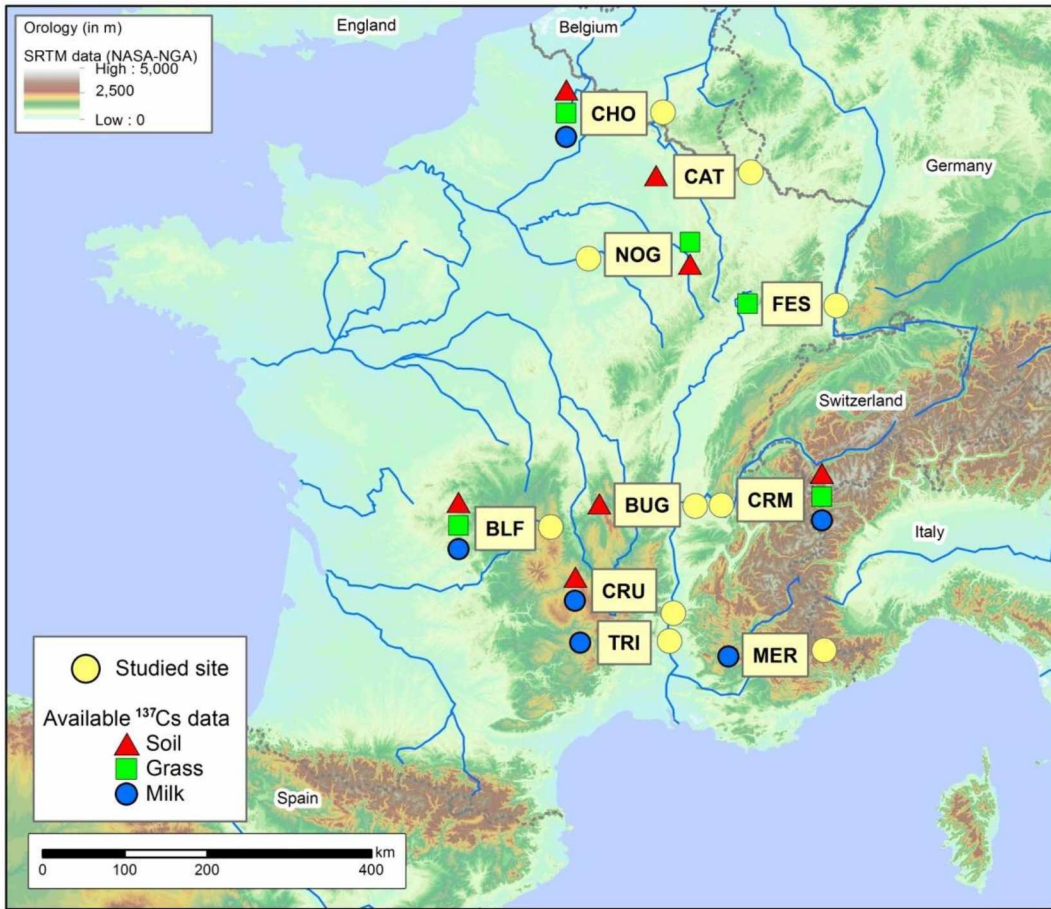
13 **Fig. 3:** Estimated long term effective half-lives ($T_{\text{eff-slow}}$) of ^{137}Cs in a) milk, b) grass and c)
14 soil over the studied sites. Median values of $T_{\text{eff-slow}}$ as well as the 5 and 95 percentiles are
15 given to right of bars. The n value corresponds to the number of observation 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

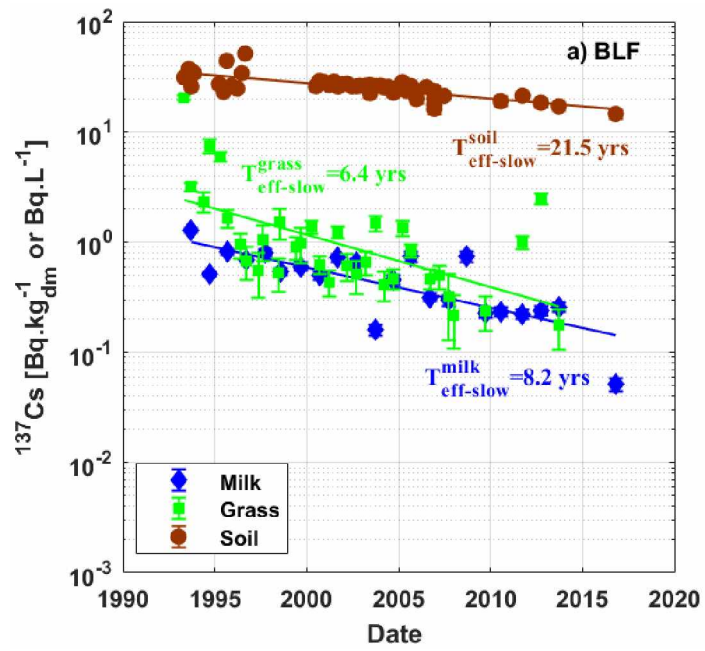
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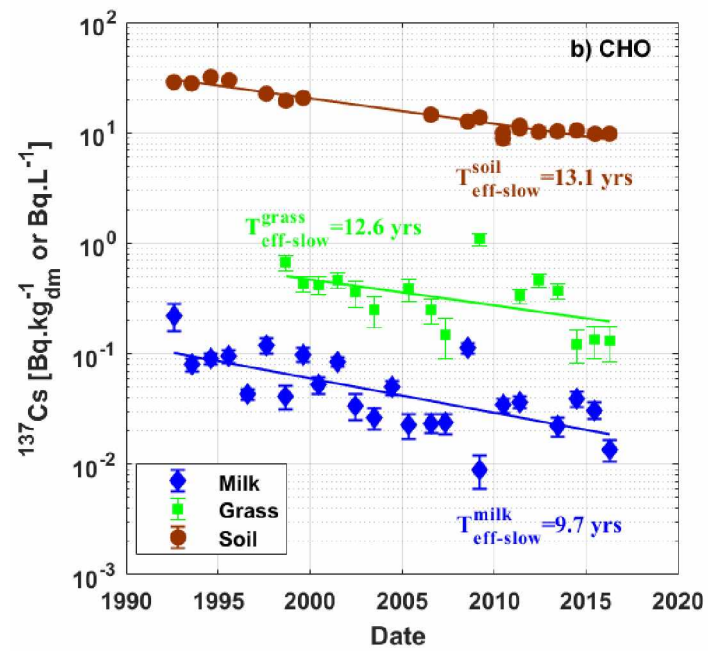


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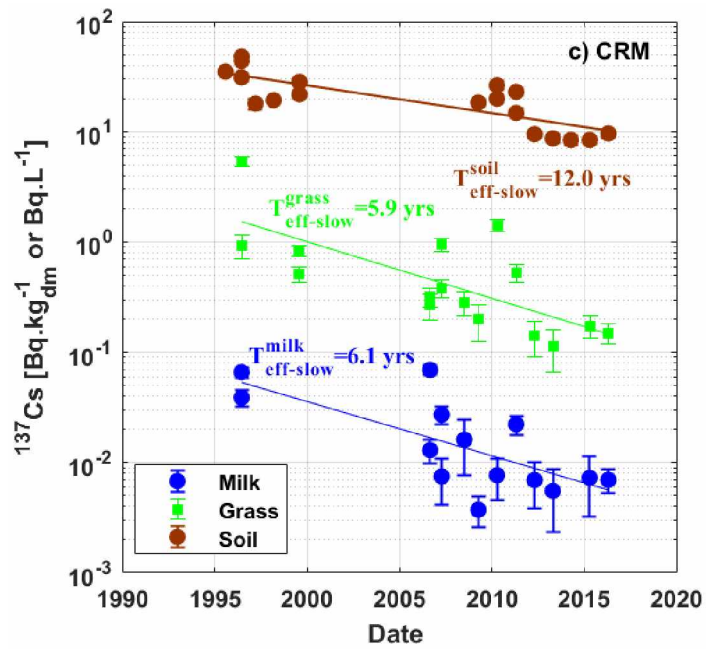
Fig. 1



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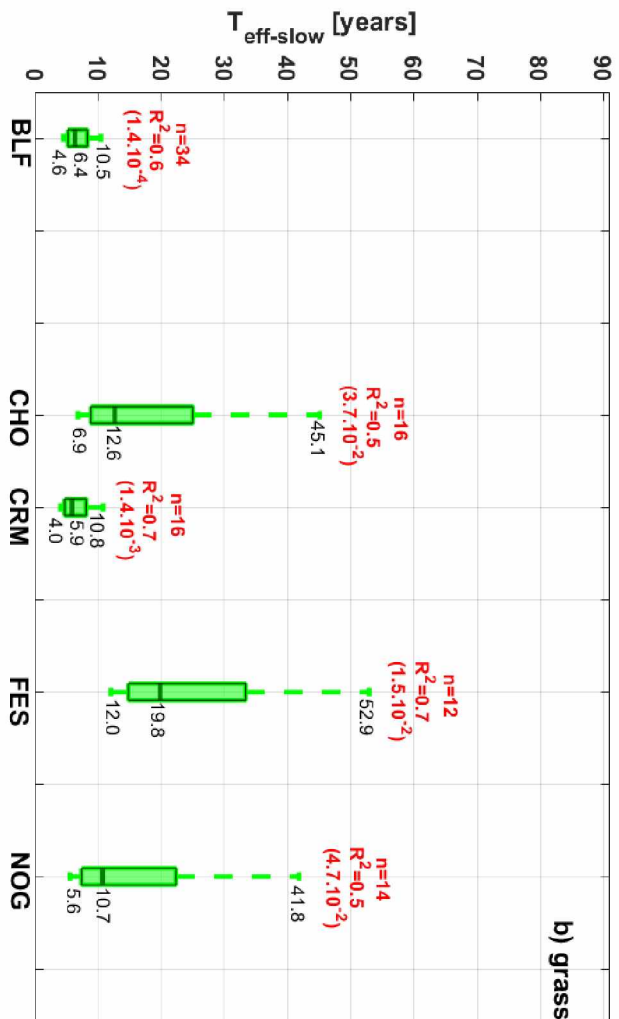
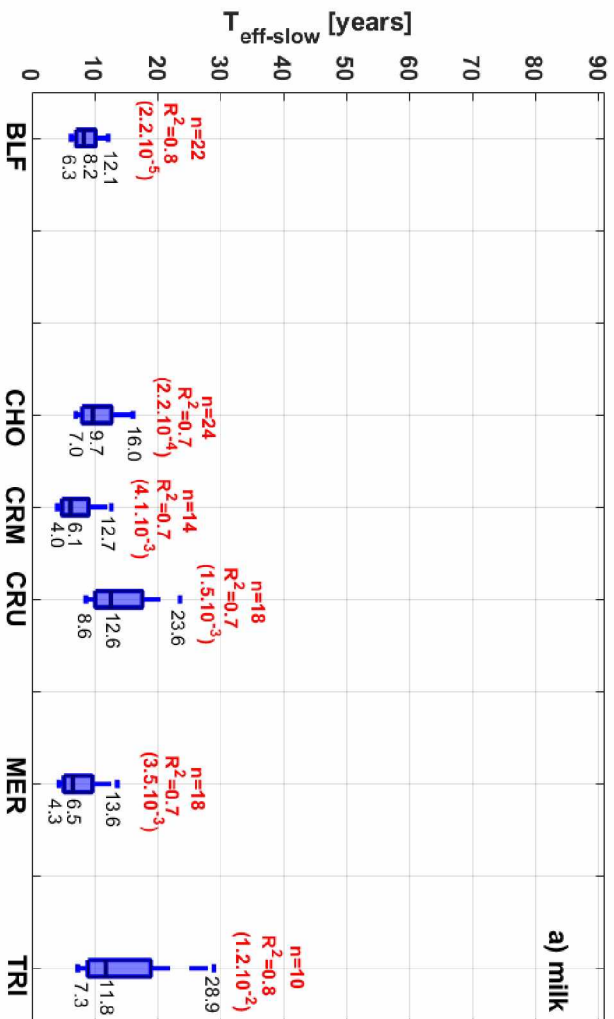


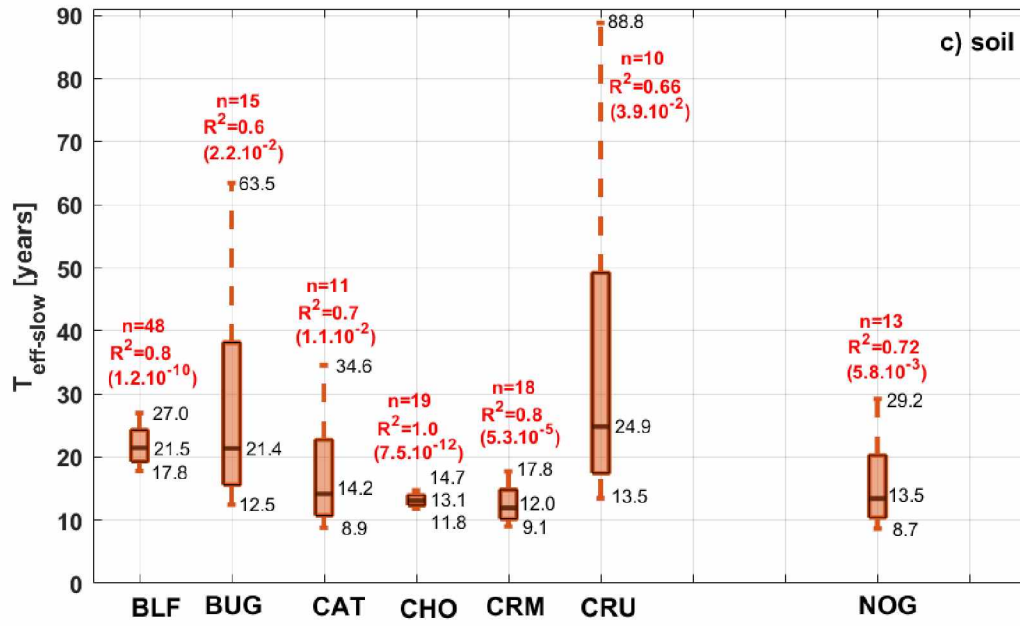
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34 Fig. 2 (a, b, c)

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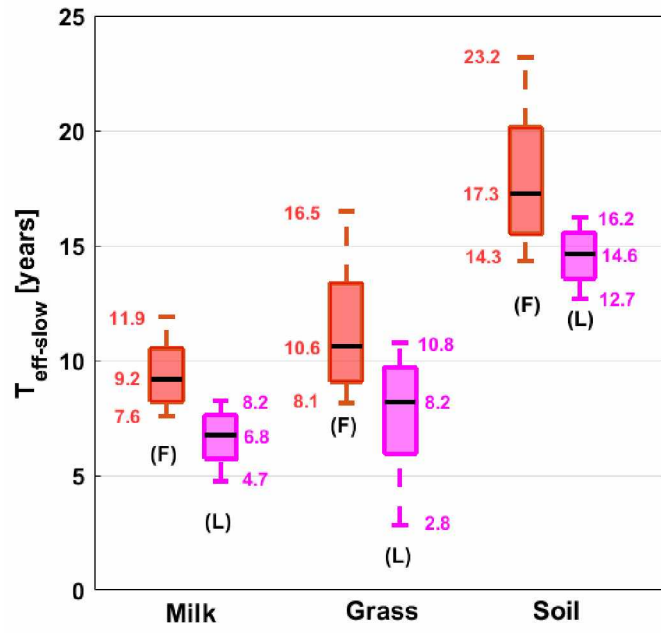
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42 **Fig. 3 (a, b, c)**

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45



46

47 **Fig. 4**

48

49

50

Appendix

Long term decrease of ^{137}Cs bioavailability in French pastures: results from 25 years of monitoring

Khaled Brimo, Marc André Gonze, Laurent Pourcelot

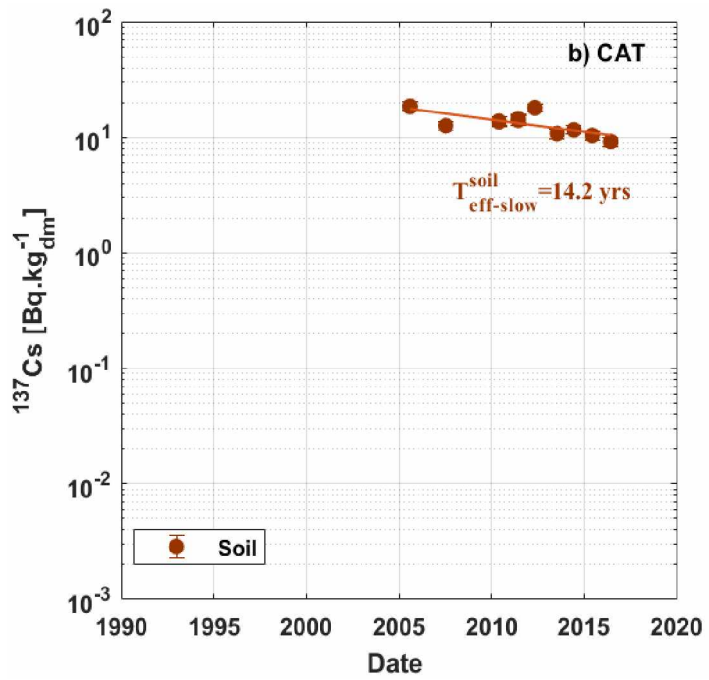
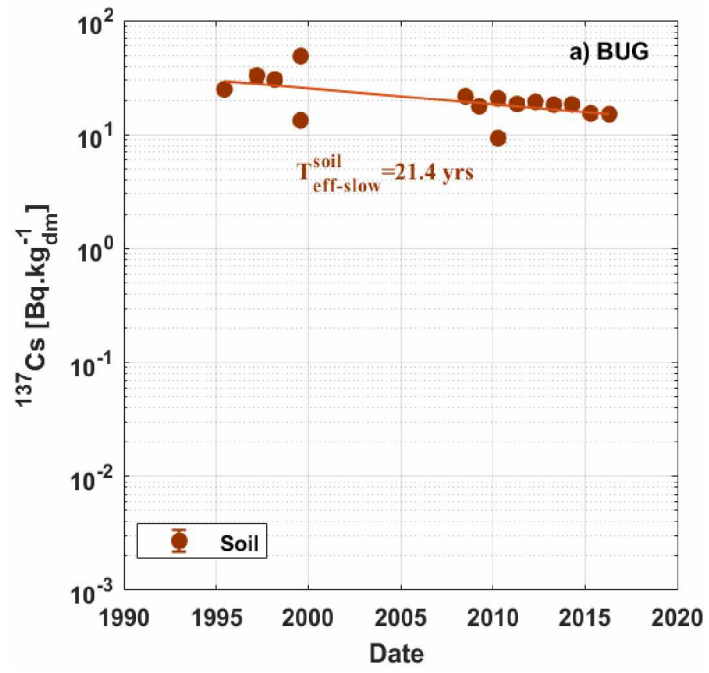
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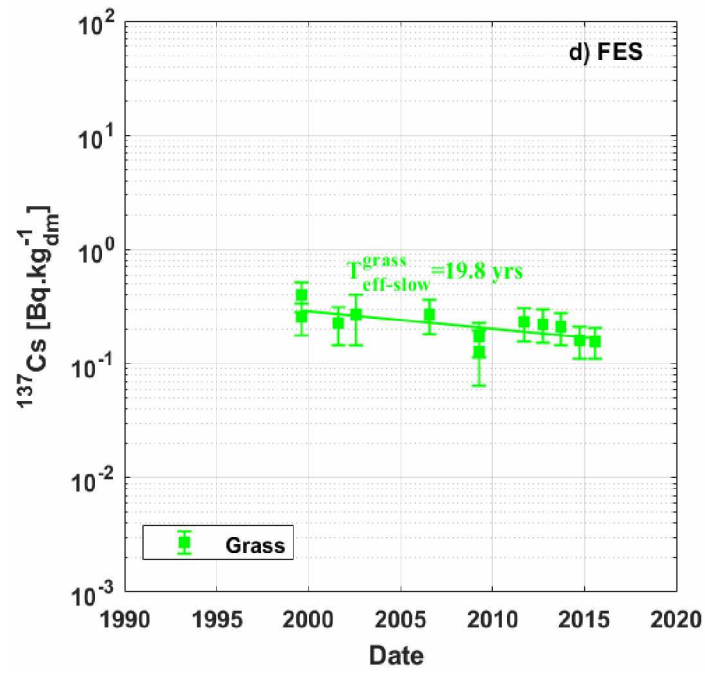
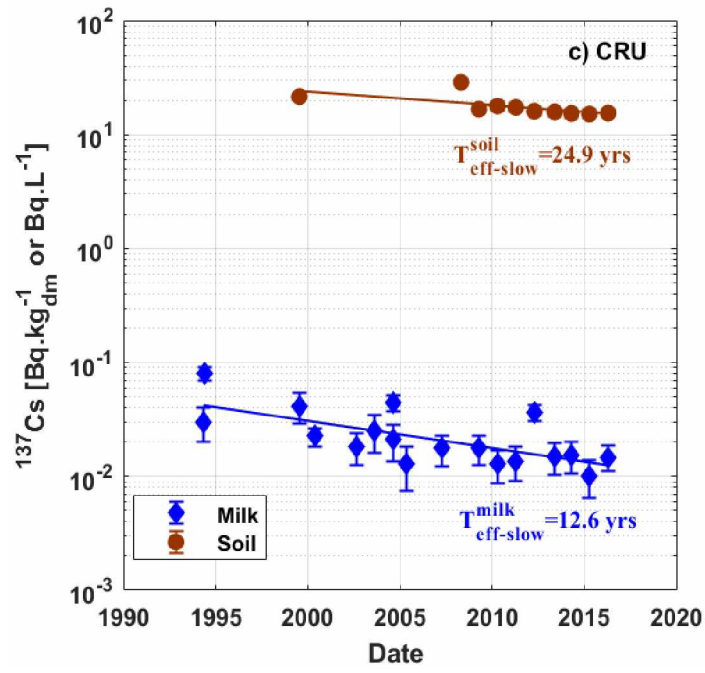
Table A.1: The p values yielded by one-way ANOVA for testing at each site the variances between the distributions of $T_{\text{eff-slow}}$ values in milk, grass and soil

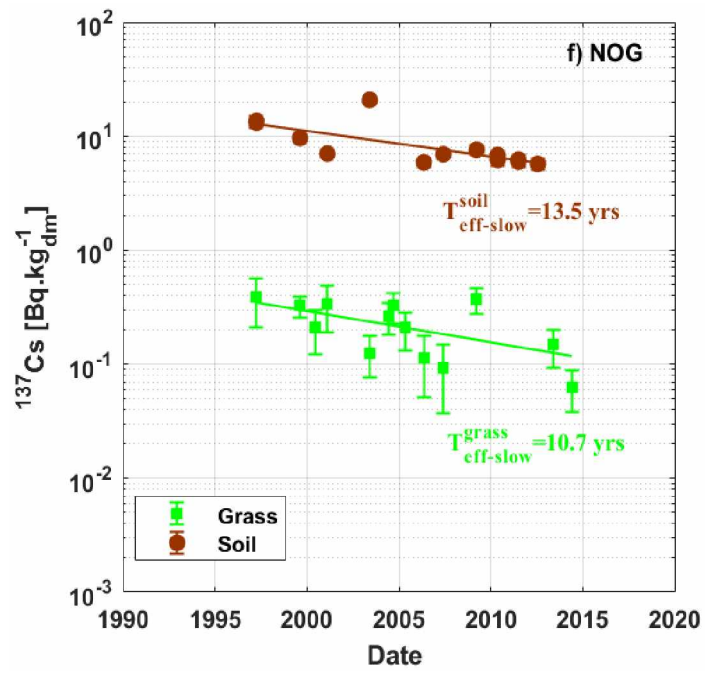
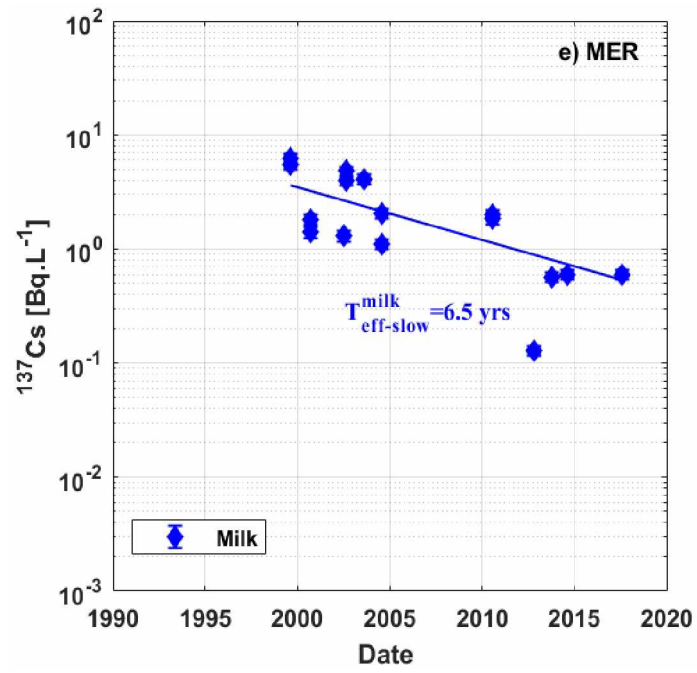
| Site | p value* | | |
|------|--|--|---|
| | $T_{\text{eff-slow}}^{\text{grass}}$ vs. $T_{\text{eff-slow}}^{\text{milk}}$ | $T_{\text{eff-slow}}^{\text{grass}}$ vs. $T_{\text{eff-slow}}^{\text{soil}}$ | $T_{\text{eff-slow}}^{\text{milk}}$ vs. $T_{\text{eff-slow}}^{\text{soil}}$ |
| BLF | 0.1899 | 0.0016 | 0.0015 |
| BUG | nd | nd | nd |
| CAT | nd | nd | nd |
| CHO | 0.3368 | 0.9487 | 0.0943 |
| CRM | 0.8615 | 0.0146 | 0.03 |
| CRU | nd | nd | 0.043 |
| 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







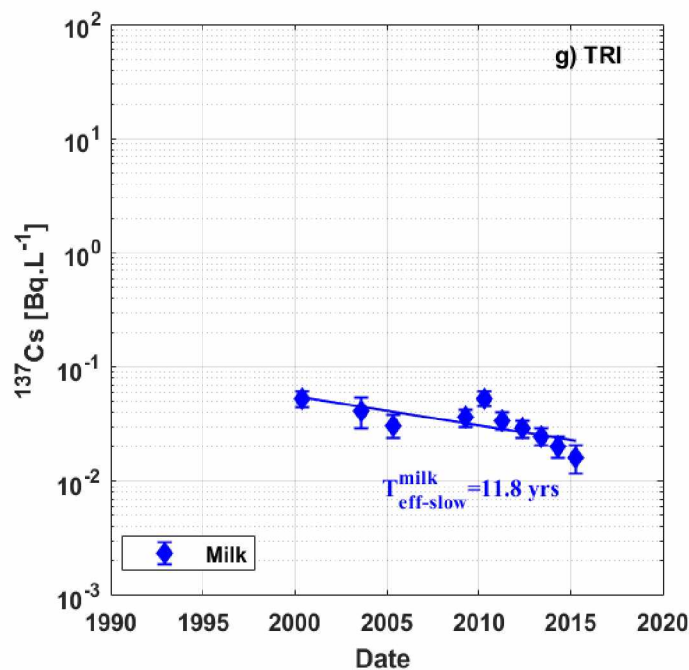


Fig. A.1: Series of ^{137}Cs activity concentrations in soil grass and milk at three sites a) Bugey (BUG), b) Cattenom (CAT), c) Cruas (CRU), d) Fessenheim (FES), e) Mercantour (MER), f) Nogent (NOG) and g) Tricastin (TRI). ^{137}Cs activity concentrations in milk are given in Bq.L^{-1} whereas ^{137}Cs 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.