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

Khaled Brimo, Marc Andre Gonze, Laurent Pourcelot

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

Khaled Brimo[a]*, Marc André Gonze[a], Laurent Pourcelot[a]

[a] Institut de Radioprotection et de Sûreté Nucléaire (IRSN), LEREN, Cadarache, 13115 Saint Paul lez Durance, France

*Corresponding authors: Khaled Brimo

Tel: + 33 04.42.19.91.09

E-mail: khaled.brimo-proman@irsn.fr

1. Introduction

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

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

Information on the decline of ^{137}Cs bioavailability in pasture ecosystem components and their mathematical modeling are crucial for an accurate assessment of long term impacts after a nuclear accident. In this respect, several models were developed and applied during the last two decades in order to predict the fate and transport of radionuclides in pastures. Some examples of such models are the semi-mechanistic model of Absalom et al. (2001) which predicts the activity concentrations of radiocaesium transferred to plants from the contaminated soil, the radiological risk assessment tool (ASTRAL) to account for the dose ambient rates in agricultural and forest systems (Calmon and Mourlon, 2002),and also the European Decision Support System (RODOS) for nuclear emergencies (Ehrhardt and Weis, 2000). These models differ largely by their complexity, structure, assumptions, and mathematical formalisms but a common problem of determining the best sets of model parameters remains an issue (Almahayni et al., 2019). One key parameter for most of these radiological models is the effective half-life parameter which is defined as the time during which the radionuclides activity content, in an environmental component, is reduced to half by the action of all processes including the physical decay(IAEA-TECDOC-472, 2010). These parameters are often required as inputs in assessment tools and determination of their values as accurately as possible is, therefore, crucial for optimum prediction results. Estimating the values of effective half-life for ^{137}Cs in pasture ecosystem components has been well treated in literature but only a few studies such as those by Corcho-Alvarado et al. (2016) and Roussel-Debet et al.(2007) have estimated their values by measurements followed on a time scale of more than 15 years after a nuclear accident. Studies over greater time may therefore be required for more realistic estimations because of the quite high persistence of ^{137}Cs in the environment. Additionally to this, there is a little or no data about the uncertainty associated with these parameters although this is essential information to perform probabilistic assessments. Here we present a study examining the long term effective half-**lives** values in different pasture components as a first step towards more robustness and reliability of radiological risks assessment models. The main goal was to use more than twenty five years monitoring data, collected at dozen of French pasture sites by the IRSN (Institute for Radiological and Protection Nuclear Safety), in order to estimate the average values of the long term effective half-lives with their associated uncertainty in soil-grass-cow's milk components. An additional goal was then to evaluate the decrease of ^{137}Cs bioavailability rates that are required in radiological assessment models.

2. Monitoring data

2.1. Monitoring sites

Locations of the ten monitoring sites are shown in Fig.1. These sites are distributed in eastern, south-eastern and northern regions of France which are regarded as the most contaminated areas by ^{137}Cs atmospheric fallouts over the French territory. Eight of the sampled sites are grazing areas located around the Electricité de France nuclear power plants (CHO, CAT, NOG, FES, BUG, CRM, CRU, TRI), while the two additional investigation sites, Mercantour (MER) and Beaune-Le-Froid (BLF), are both located in permanent prairie areas. The main sources of ^{137}Cs contamination in the monitoring sites were the depositions from nuclear weapons tests and Chernobyl fallouts (Roussel-Debet et al., 2007). Additionally to these depositions, a very minor and negligible amount was also received after the Fukushima accident in spring 2011(Masson et al., 2011; Parache et al., 2011).The contribution of ^{137}Cs originating from nuclear reactor releases to the total ^{137}Cs content in terrestrial environment can be ignored (Duffa et al., 2004). All sites are used for cow grazing and therefore grass and cow‘s milk sampled from these sites are used to assess the transfer of ^{137}Cs from soil to grass and from grass to milk. In winter cows are kept in barns and fed by hay harvested during the summer. Cows also receive very small amounts of supplement that contain vitamins and proteins (usually flour corn or soya). Overall, at grazing as for indoor feeding, the feeding rates did not change over time. Table 1 gives the names and the corresponding abbreviations of the sites of interest, their geographical location, the periods of observation, the target component and the corresponding ranges of ^{137}Cs activities. The granulometry and soil organic content in the studied sites have been summarized in Table 2. The soil is loamy with low organic matter content in all studied sites except for Nogent and Mercantour in which clay soil and organic soil predominate respectively (IAEA classification, (IAEA-TECDOC-472, 2010)). The monitoring sites are, in general, all located in areas where oceanic climate prevails except for Mercantour for which high-mountain climate prevails (Pourcelot et al., 2003).

.

2.2. Sampling and analytical methodology

At each of the selected pastures plot (surface ranges between 2,000 and 10,000 m² about) soil from the upper 5 cm layer was taken following methodology described in Roussel-Debet et al. (2007). At the same plot, grass (stems and shoots) covering one to two square meters was collected using shears, 2-5 cm above the ground in order to prevent any contamination by soil particles. At the same time, about 5 L of cow's milk was also sampled at the farm when cow's herd was grazing in the fields. Back to the laboratory, soil sample was first sieved to remove >2 mm material. Then soil and grass samples were dried at 80 °C for 5 days. Milk sample was dried by lyophilization. Finally dried samples of grass and milk were ashed at 480°C for 24 h and crushed prior to ^{137}Cs analyses. Low level gamma spectrometry allows us to quantify ^{137}Cs activity, following methodology given by Bouisset and Calmet (1997).

2.3. Available data and selection criteria

Data used for this study were aggregated from the results of long term radioactivity monitoring programs conducted during the period from 1992 to 2017 at various monitoring sites in France. Overall, we had access to about 600 measurement data of ^{137}Cs activities across the monitoring sites continuously sampled at the same areas. Measurements were collected either from only one plot or from several plots that were located within area delineated by a circle having a diameter of less than 30 km. Given the fact that these data were obtained from various studies and monitoring programs, some were incomplete or not suitable for this study purposes due to the relatively short period of their observation which was not long enough to reveal a significant decline in ^{137}Cs content. To avoid potential misleading results and ensure that our parameters estimation was as reliable and accurate as possible, we set criteria for data selection. Hence, for ^{137}Cs half-life estimations, the data sets that covered a time period of at least ten years with at least ten measurements were only selected. We applied later an additional criterion: the simple exponential model explained in section 3, to which our data were fitted, should be able to explain at least 50% of the data variation (i.e. the determination coefficient value R^2 derived from a least squares analysis should be equal or greater than 0.5). In the light of this analysis, the simultaneous availability of data sets on ^{137}Cs measurements in soil, grass and milk was limited to three sites (BLF, CHO and CRM) whereas data of the remaining sites were limited to one or two of these components (see Table 1 and Fig.1).

3. Data modeling

3.1. Theory

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

However, the contribution of the foliar pathway becomes negligible after some years, while uptake by roots becomes the major process controlling the radiocesium activity concentration in grass but also in cow s' milk, as the ingested grass is the major source of contamination in milk at the sites investigated here (i.e. the contribution of other feedstuffs and drinking water is negligible). On the long term, it is usually assumed that radiocesium activity concentrations in grass and milk are in equilibrium with the activity concentration of bioavailable radiocesium in soil (denoted C_{av} hereafter).

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

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

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

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

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

On the other hand, the long term rates of change of the total radiocesium content in soil (i.e. the sum of bioavailable and non-bioavailable~~s~~ fractions) is commonly described by assuming that the decline in radiocesium concentration in soil C_s is exponential, and the corresponding effective half-life can therefore be given as follows:

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

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

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

3.2. Mathematical and statistical data analysis

We estimated the values of kinetic rate constants (k_{slow}, λ_{soil}) with their associated uncertainties at confidence intervals of 95% using the multiple linear regression tool of Excel software (Microsoft, USA, 2010). The coefficient of determinations (R^2) was used as indicator of the goodness of fit. The fitting of (k_{slow} +λ) required for equation (2) was performed by either the slope of logarithmically transformed concentrations of ^{137}Cs in grass versus time ($\rightarrow T^{av}_{eff-slow} = T^{grass}_{eff-slow}$) or from the slope of logarithmically transformed concentrations of ^{137}Cs in milk versus time ($\rightarrow T^{av}_{eff-slow} = T^{milk}_{eff-slow}$). The kinetic rate λ_{soil} required for equation (3) was determined from the slope of logarithmically transformed concentrations of total ^{137}Cs in soil versus the elapsed time.

To evaluate the long term decrease of the bioavailable pool of ^{137}Cs, the characteristic time of the decrease of cesium bioavailability (T^{slow}) was calculated as follows:

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

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

4. Results and discussion

4.1. Temporal trends in ^{137}Cs activity concentrations

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

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

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

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

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

4.3. Comparison of $T_{eff-slow}$ at monitoring sites

The variances between the distributions of effective half-life values in milk, grass and soil were evaluated independently for each studied site by means of one way analysis of variance tests (ANOVA, $p < 0.05$). These tests were limited to the sites where at least 2 compartments were monitored (i.e. BLF, CRM, CRU, NOG and CHO). The p values yielded by the statistical tests are all summarized in Table A.1 in Appendix. For BLF and CRM sites, independently, significant differences appeared between the distributions of effective half-life values estimated in milk ($T_{eff-slow}^{milk}$) or grass ($T_{eff-slow}^{grass}$) against that estimated in soil ($T_{eff-slow}^{soil}$) (ANOVA, $p < 0.05$). In contrast, no such significant differences were found between $T_{eff-slow}^{milk}$ and $T_{eff-slow}^{grass}$ (ANOVA, $p > 0.05$). Similar results were also found at CRU site with respect to the difference between grass and soil, and at CHO site with respect to the difference between grass and milk. Indeed, these statistic findings, of no significant differences between grass and milk but significant differences with respect to soil, support the view that the bioavailable pool of ^{137}Cs is likely controlled by the slow irreversible fixation of cesium to the soil matrix. This, in turn, limits its bioavailability for vegetation uptake and its transfer to milk for cows that are fed with locally produced vegetation. Moreover, these statistic findings are also in agreement with the conclusion frequently reported in other previous studies that the milk could be a robust indicator of contamination in grass(e.g. Mück and Gerzabek,1995; Pourcelot et al., 2007). However, results inconsistent with these findings suggesting differences between grass and soil were found for CHO and NOG sites. These differences may be related to the variability across studied sites so that the fixation process at those two sites was less important in affecting ^{137}Cs bioavailability compared to the aforementioned sites.

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

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

0.008 and 0.044 $year^{-1}$ with corresponding characteristic times from 13 to 61 years in soil). The impact of aging of bioavailable pool of ^{137}Cs was also shown when the soil-to-grass transfer factor (TF) values calculated within different decades were compared. The obtained TF values were found to decrease with time and to range from1.3 x10^{-2} to 65.8 x 10^{-2} with a median value of 7.7 x 10^{-2} within the first decade after Chernobyl accident (i.e. $t \leq 1996$), from 0.6 x10^{-2} to 5.7 x 10^{-2} with a median value of 2.5 x 10^{-2} within the second decade after Chernobyl accident (1997 - 2006) and from 1.03 x10^{-2} to 1.36 x 10^{-1} with a median value of 2.1 x 10^{-2} within the third decade after Chernobyl accident (2007 - 2016). All TF values are in the lower bound of the range of 10^{-2}-5.0 given by IAEA-TECDOC-472, (2010). These can be explained by the "old" contamination of monitoring sites since our ^{137}Cs measurements were performed many years after the initial deposit. It is important to stress here that our findings regarding the evolution of bioavailable fraction of ^{137}Cs with time were based on long term monitoring data (i.e. ~ 6-30 years from the Chernobyl accident) and therefore it is impossible at present time, due to absence of measurements, to predict whether the decrease in the bioavailable fraction in the very long term (i.e. >30 years) will still continue as in the long term or a steady state can be reached. More investigations are still required.

4.4. Comparison to other findings

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

Our results are then compared with the long term $T_{eff\text{-}slow}$ values reported in the literature. Table 3 summarizes the values of $T_{eff\text{-}slow}$ reviewed in literature from 6 different studies in cow's milk, 5 in grass and 4 in soil. For a consistent comparison, all reviewed studies were carried out in pasture plots located in Western Europe where $T_{eff\text{-}slow}$ values were estimated by means of ^{137}Cs data originated from both the nuclear weapons tests and the Chernobyl accident like in the previous section. For each environmental component, the distribution of the $T_{eff\text{-}slow}$ values averaged over the reviewed studies were calculated by Monte Carlo simulations. For comparison purpose, the averaged long term $T_{eff\text{-}slow}$ values yielded by these calculations were also presented in Fig.4 (bars denoted with the letter L). Similar to our findings, the averaged $T_{eff\text{-}slow}$ using data from literature were found to decrease in the order soil >> grass > milk with median values of 15.0 , 9.0 and 8.0 years respectively and with a global tendency to underestimate slightly the values derived in this study. This slight difference of about 2-3 years may be caused by several reasons. In addition to the aforementioned reason of low goodness of fit, this difference may be attributed to the difference in the length and starting time of time series used in $T_{eff\text{-}slow}$ estimation. Indeed, despite the finding by Mück, (2003)that the effective slow decline rate of ^{137}Cs begins only 6-7years after Chernobyl accident (1993),many studies estimated $T_{eff\text{-}slow}$ values from time series observed before this date (Table 3). Moreover, some reported $T_{eff\text{-}slow}$ were derived from time periods that ceases even earlier than 1993 such as the reported values by Pröhl. et al. (2006) in grass, by Fesenko et al. (1995)in milk and by Isaksson et al. (2001) in soil. Also with respect to this point, the effective half-lives given by these two latter authors of 11.7 and 13.0 years respectively seemed to be not reliable since they were estimated by fitting to experimental data measured only for short time periods of about 3 years which are much less than their reported $T_{eff\text{-}slow}$ values. In contrast to this, $T_{eff\text{-}slow}$ values yielded by this work were estimated by data collected over longer period of almost 25 years since about 1993.Another possible reason is that the ^{137}Cs content in pastures might decrease progressively with time (i.e.by multiple decline phases rather than only two, fast and slow, phases) and therefore it is reasonable to assume that the longer time series led to larger $T_{eff\text{-}slow}$ values. However, this hypothesis can be rejected by our measurements that displayed only one long term decline rate of ^{137}Cs as shown in Fig. 2.Additionally to the above mentioned reasons, one may argue that this difference may be related to the difference in sampling strategy between different works. However, this was found to have no or minor influence in our case.

5. Summary and conclusions

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

Acknowledgements

We would like to thank the Électricité de France (EdF) for funding parts of the measurements. We are grateful to Dr. Jean-Michel Méteivier for his assistance in producing Fig 1.Authors thank Dr. Marie Simon-Cornu and Dr. Gregory Mathieu for reading the manuscript prior to submission.

References

Absalom, J.P., Young, S.D., Crout, N.M.J., 1995. Radio- caesium fixation dynamics: measurement in six Cumbrian soils. Eur. J. Soil Sci. 46, 461–469. https://doi.org/10.1111/j.1365-2389.1995.tb01342.x

Absalom, J.P., Young, S.D., Crout, N.M.J., Nisbet, A.F., Woodman, R.F.M., Smolders, E., Gillett, A.G., 1999. Predicting soil to plant transfer of radiocesium using soil characteristics. Environ. Sci. Technol. 33, 1218–1223.

Absalom, J.P., Young, S.D., Crout, N.M.J., Sanchez, A., Wright, S.M., Smolders, E., Nisbet, A.F., Gillett, A.G., 2001. Predicting the transfer of radiocaesium from organic soils to plants using soil characteristics. J. Environ. Radioact. 52, 31–43.

Almahayni, T., Beresford, N.A., Crout, N.M.J., Sweeck, L., 2019. Fit-for-purpose modelling of radiocaesium soil-to-plant transfer for nuclear emergencies: a review. J. Environ. Radioact. 201, 58–66. https://doi.org/10.1016/j.jenvrad.2019.01.006

Ashraf, M.A., Akib, S., Jamil, M., 2014. Cesium-137: radio-chemistry, fate, and transport, remediation, and future concerns. Crit. Rev. Environ. Sci. Technol. 44, 1740–1793. https://doi.org/10.1080/10643389.2013.790753

Astori, E., Bianchi, D., Cammarata, E., Gastaldo, S., Marzolla, N., Rabbia, G., Vescovi, A., Priano, L., 1999. Surface contamination of radiocesium measured and calculated in South Piemonte (Italy). J. Environ. Radioact. 45, 29–38. https://doi.org/10.1016/S0265-931X(98)00071-X

Bouisset, P., Calmet, D., 1997. Hyper pure gamma-ray spectrometry applied to low-level environmental sample measurements, in: Proceedings of the International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures.

Calmon, P., Mourlon, C., 2002. Equations et paramètres du logiciel ASTRAL v2. IRSN. Rapport DPRE/SERLAB/LMODE no 02-29, p. 99.

Comans., R.N.J., Middelburg., J.J., Zonderhuis., J., Woittiez., J.R.W., Lange., G.J. De, Das., H.A., Weijden., C.H. Van Der, 1989. Mobilization of radiocaesium in pore water of lake sediments. Nature 339, 367–369.

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

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

Duffa, C., Masson, M., Gontier, G., Claval, D., Renaud, P., 2004. Synthèse des études radioécologiques annuelles menées dans l environement des centrales électronucléaires francaises deupuis 1991. Radioprotection 39, 233–254.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Component	$T_{eff\text{-}slow}$ [years]	Observation period	Study area	Source
Milk				
	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)
Grass				
	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)
Soil				
	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	Roussel-Debet et al., 2007
	7.7[c] (21.5)	1992-2004 (1995-2016)	BUG	
	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)

([a])These values have been averaged over several monitoring plots, locations, soil types or soil textures thus they may differ from those found directly in the cited papers. The two given numbers are the ninety-five percent confidence limits for the average of reported values along with their associated uncertainties in the cited paper. Calculations were established based on10^6 Monte-Carlo simulations and a Gaussian probability distribution.

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

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

([d]) For a comparison purpose, the median of the distributions of $T_{eff-slow}$ values averaged over all investigated sites are also given in the above table.

Figure captions

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

Fig. 2: Time series of ^{137}Cs activity concentrations in soil grass and milk at three sites a) Beaune- Le-Froid (BLF), b) Chooz (CHO) and c) Crey-Malville (CRM). ^{137}Cs activity concentrations in milk are given in $Bq.L^{-1}$ whereas ^{137}Cs activity concentrations in grass and soil are given in $\mathrm{Bq.kg_{dm}^{-1}}$. 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.

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

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

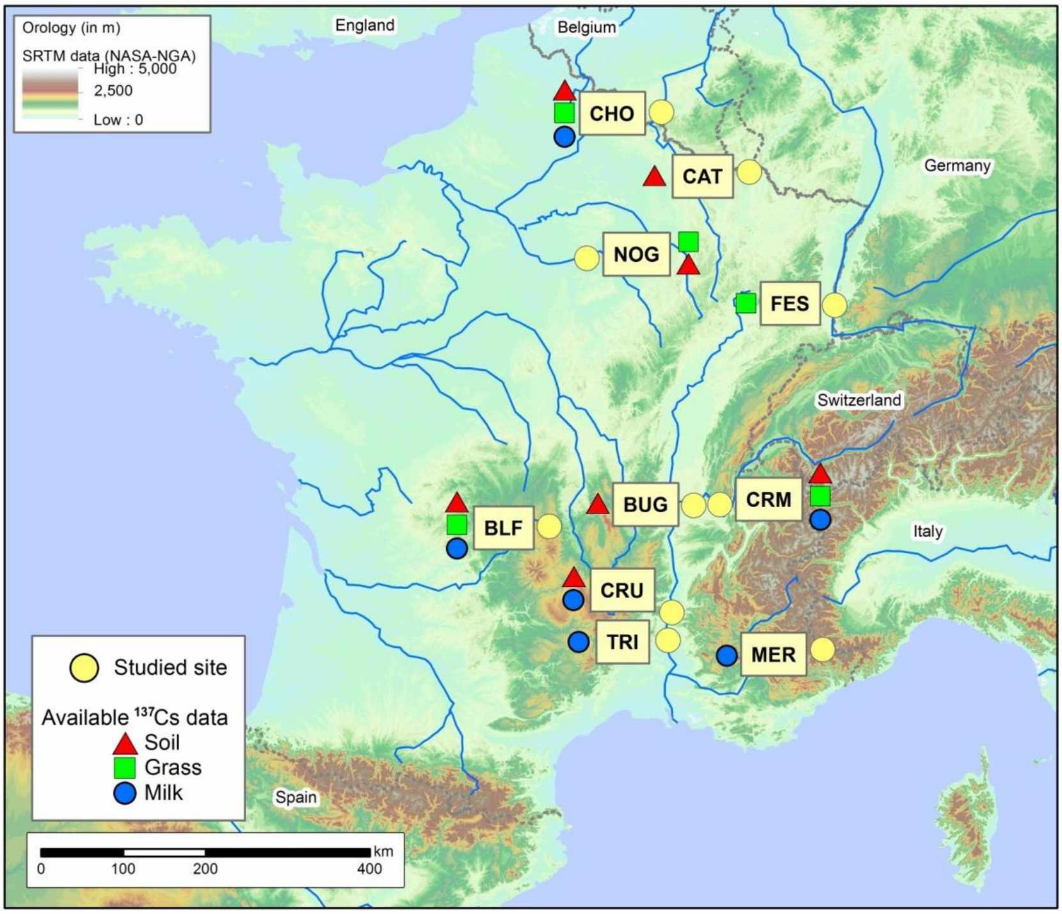
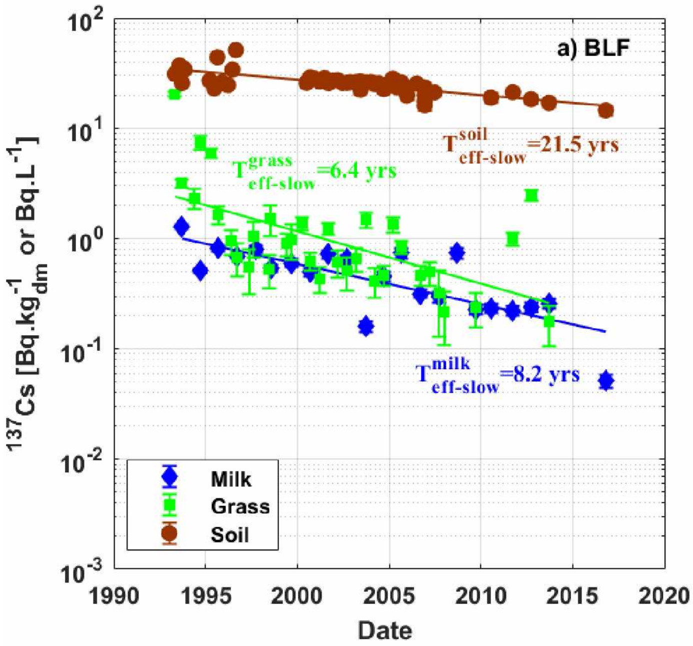
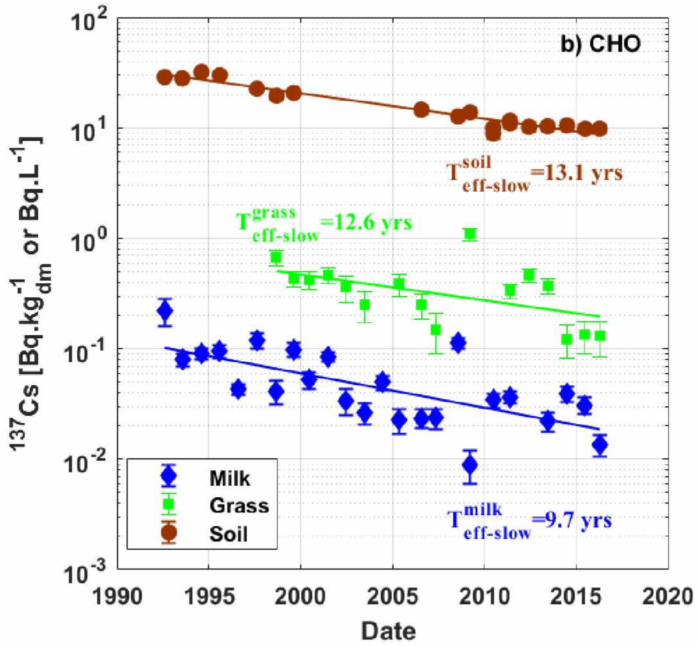

Orology (in m)
SRTM data (NASA-NGA)
High : 5,000
2,500
Low : 0
England
Belgium
Germany
Switzerland
Italy
Spain
CHO
CAT
NOG
FES
BUG
CRM
BLF
CRU
TRI
MER
Studied site
Available ^{137}Cs data
Soil
Grass
Milk
km
0
100
200
400

Fig. 1


a) BLF
$T^{soil}_{eff\text{-}slow}$=21.5 yrs
$T^{grass}_{eff\text{-}slow}$=6.4 yrs
$T^{milk}_{eff\text{-}slow}$=8.2 yrs
^{137}Cs [$Bq.kg^{-1}_{dm}$ or $Bq.L^{-1}$]
Date
Milk
Grass
Soil


b) CHO
$T^{soil}_{eff\text{-}slow}$=13.1 yrs
$T^{grass}_{eff\text{-}slow}$=12.6 yrs
$T^{milk}_{eff\text{-}slow}$=9.7 yrs
^{137}Cs [$Bq.kg^{-1}_{dm}$ or $Bq.L^{-1}$]
Date
Milk
Grass
Soil

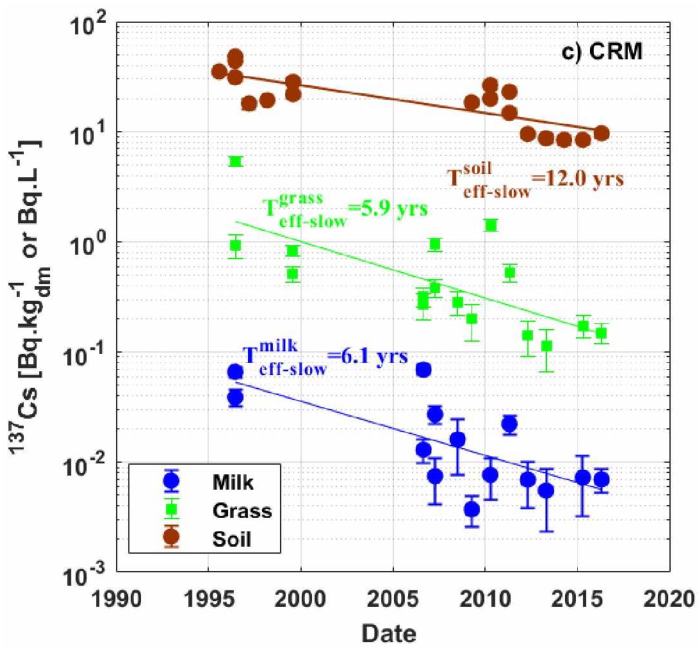
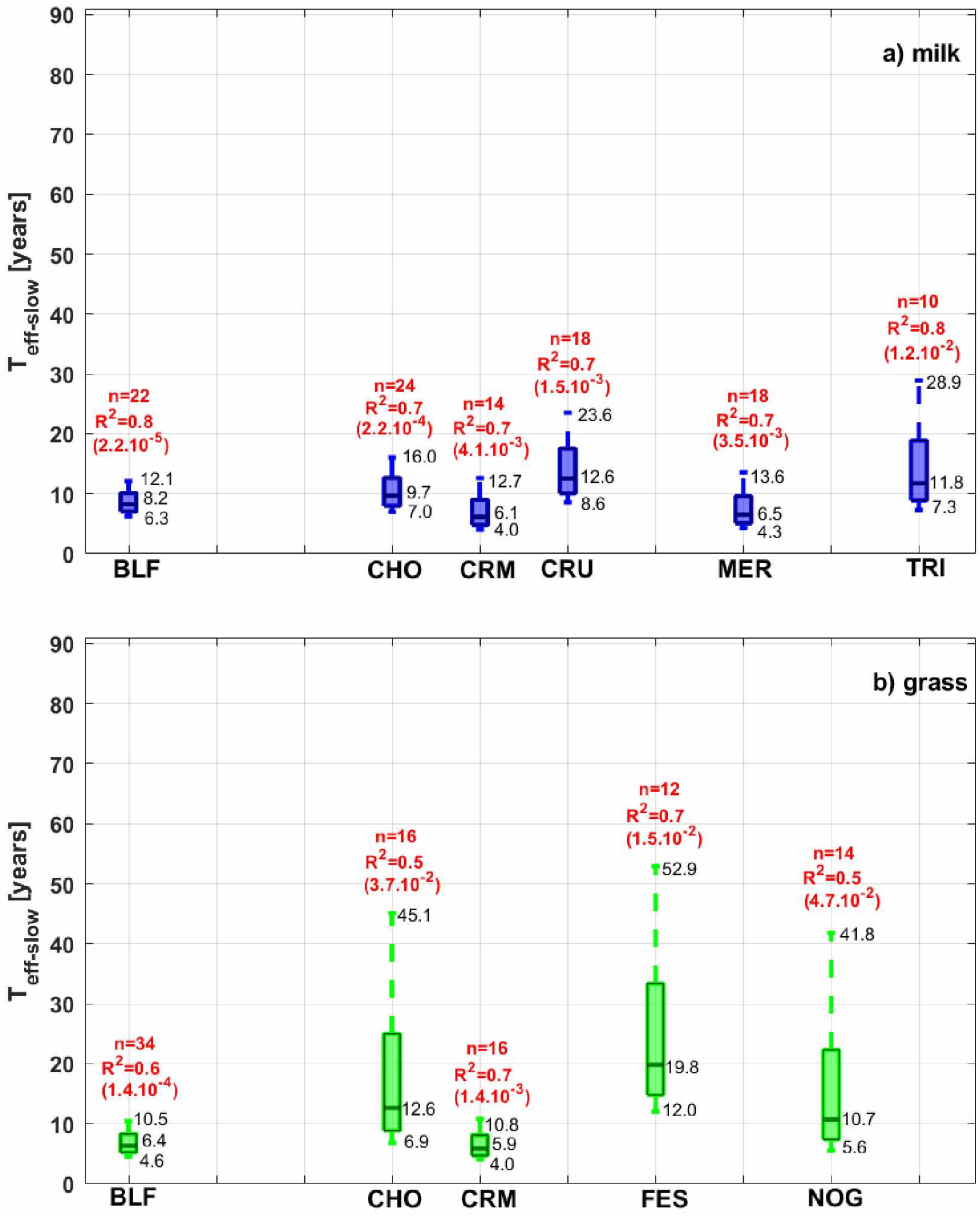

c) CRM
$T^{soil}_{eff-slow}$=12.0 yrs
$T^{grass}_{eff-slow}$=5.9 yrs
$T^{milk}_{eff-slow}$=6.1 yrs
Milk
Grass
Soil
^{137}Cs [Bq.kg$^{-1}_{dm}$ or Bq.L^{-1}]
Date
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
1990
1995
2000
2005
2010
2015
2020

Fig. 2 (a, b, c)



a) milk
$T_{eff-slow}$ [years]
n=22
R^2=0.8
(2.2.10^{-5})
12.1
8.2
6.3
n=24
R^2=0.7
(2.2.10^{-4})
16.0
9.7
7.0
n=14
R^2=0.7
(4.1.10^{-3})
12.7
6.1
4.0
n=18
R^2=0.7
(1.5.10^{-3})
23.6
12.6
8.6
n=18
R^2=0.7
(3.5.10^{-3})
13.6
6.5
4.3
n=10
R^2=0.8
(1.2.10^{-2})
28.9
11.8
7.3
BLF
CHO
CRM
CRU
MER
TRI
b) grass
$T_{eff-slow}$ [years]
n=34
R^2=0.6
(1.4.10^{-4})
10.5
6.4
4.6
n=16
R^2=0.5
(3.7.10^{-2})
45.1
12.6
6.9
n=16
R^2=0.7
(1.4.10^{-3})
10.8
5.9
4.0
n=12
R^2=0.7
(1.5.10^{-2})
52.9
19.8
12.0
n=14
R^2=0.5
(4.7.10^{-2})
41.8
10.7
5.6
BLF
CHO
CRM
FES
NOG

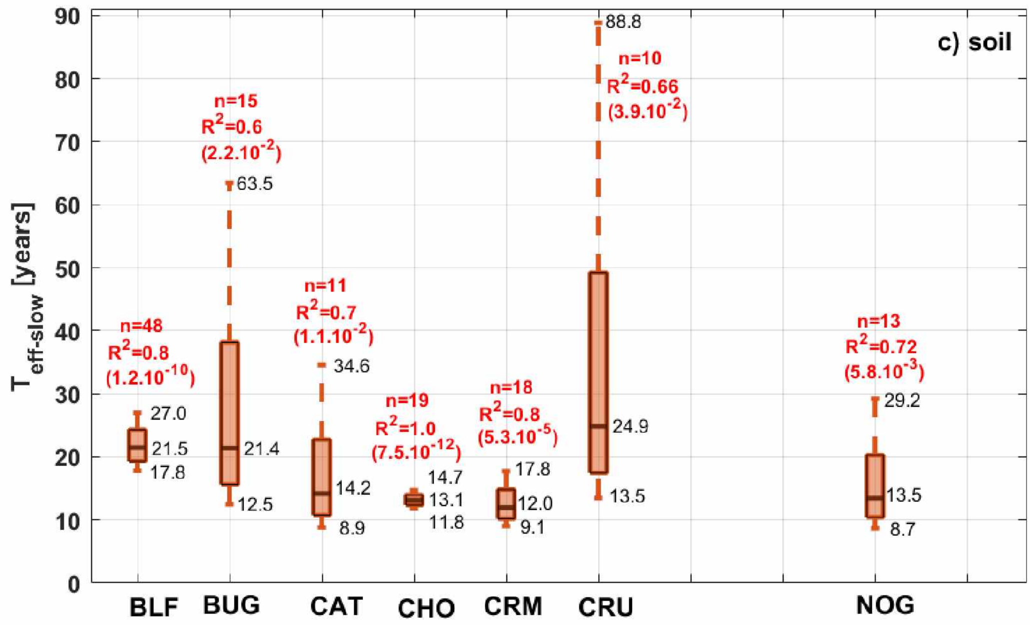
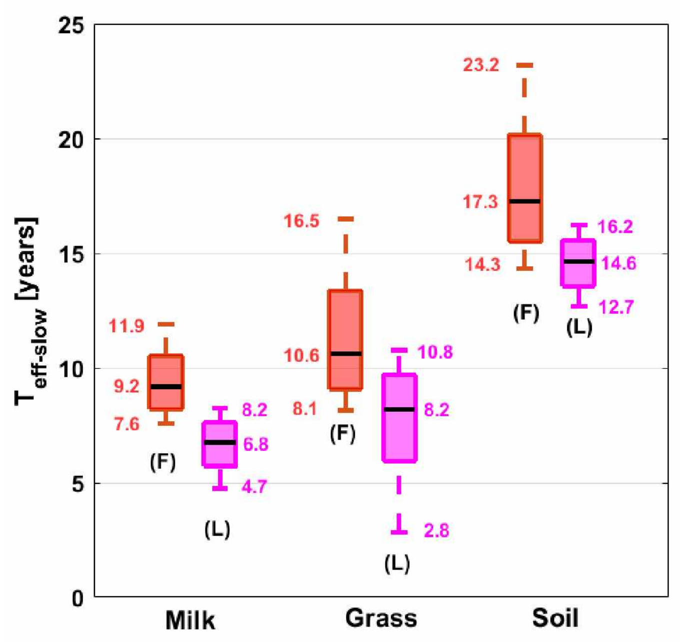

c) soil
T eff-slow [years]
0
10
20
30
40
50
60
70
80
90
BLF
BUG
CAT
CHO
CRM
CRU
NOG
n=48
R2=0.8
(1.2.10-10)
27.0
21.5
17.8
n=15
R2=0.6
(2.2.10-2)
63.5
21.4
12.5
n=11
R2=0.7
(1.1.10-2)
34.6
14.2
8.9
n=19
R2=1.0
(7.5.10-12)
14.7
13.1
11.8
n=18
R2=0.8
(5.3.10-5)
17.8
12.0
9.1
88.8
n=10
R2=0.66
(3.9.10-2)
24.9
13.5
n=13
R2=0.72
(5.8.10-3)
29.2
13.5
8.7

Fig. 3 (a, b, c)



T_eff-slow [years]
25
20
15
10
5
0
23.2
17.3
14.3
16.2
14.6
12.7
16.5
10.6
8.1
10.8
8.2
2.8
11.9
9.2
7.6
8.2
6.8
4.7
(F)
(L)
Milk
Grass
Soil

Fig. 4

Appendix

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

Khaled Brimo, Marc André Gonze, Laurent Pourcelot

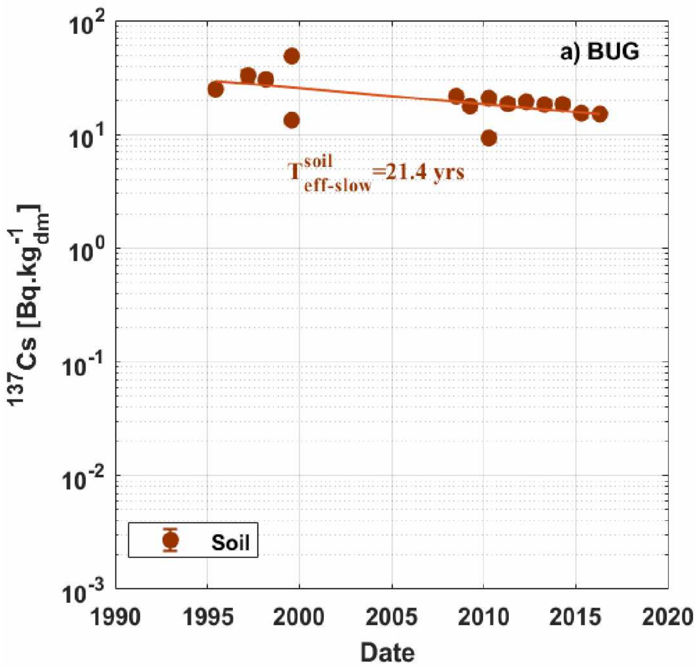
Content: Six pages, one table, one figure

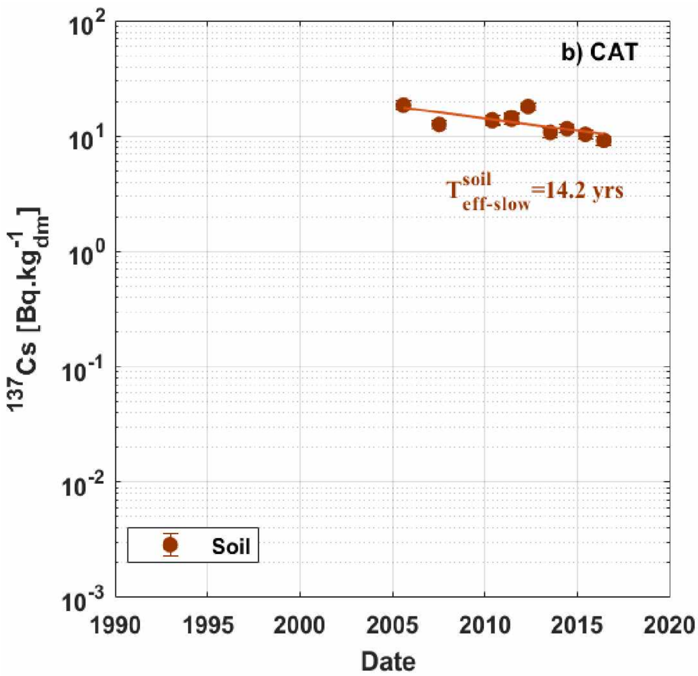
Table A.1: The p values yielded by one-way ANOVA for testing at each site the variances between the distributions of $T_{eff-slow}$ values in milk, grass and soil

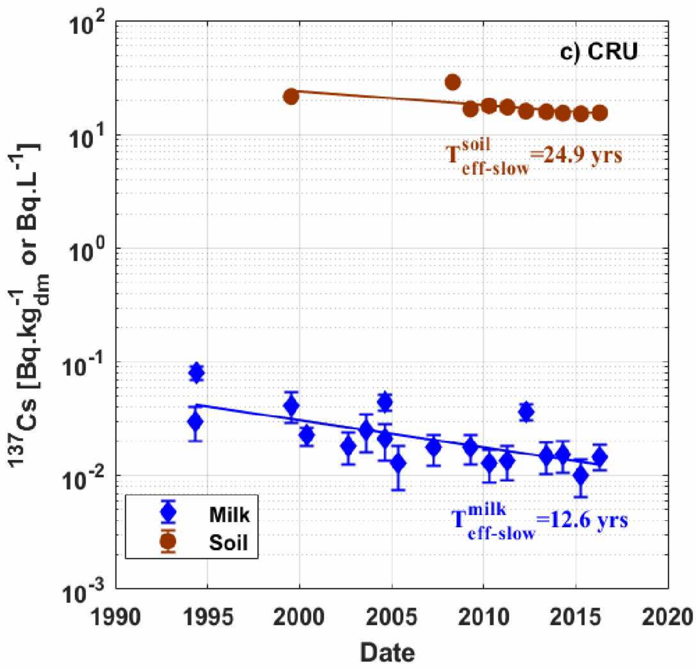
Site	p value[*]		
	$T^{grass}_{eff-slow}$ .vs. $T^{milk}_{eff-slow}$	$T^{grass}_{eff-slow}$ .vs. $T^{soil}_{eff-slow}$	$T^{milk}_{eff-slow}$ .vs. $T^{soil}_{eff-slow}$
BLF	0.1899	**0.0016**	**0.0015**
BUG	nd	nd	nd
CAT	nd	nd	nd
CHO	0.3368	0.9487	0.0943
CRM	0.8615	**0.0146**	**0.03**
CRU	nd	nd	**0.043**
FES	nd	nd	nd
MER	nd	nd	nd
NOG	nd	0.584	nd
TRI	nd	nd	nd

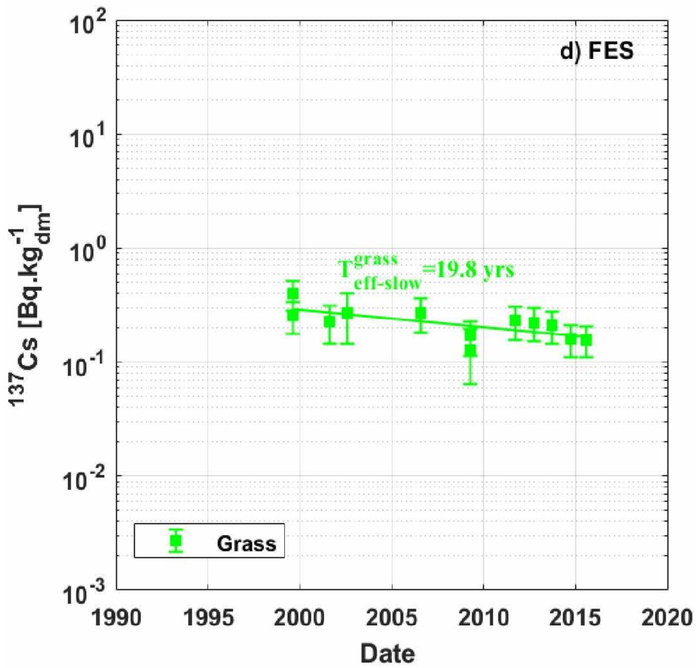
([*]) bold value indicates to a significant difference ($p < 0.05$)

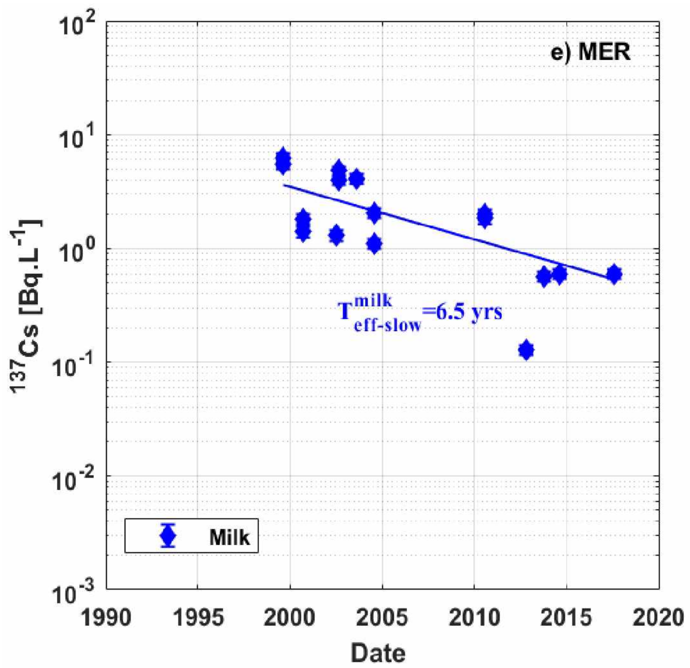
nd: not determined because of insufficient data

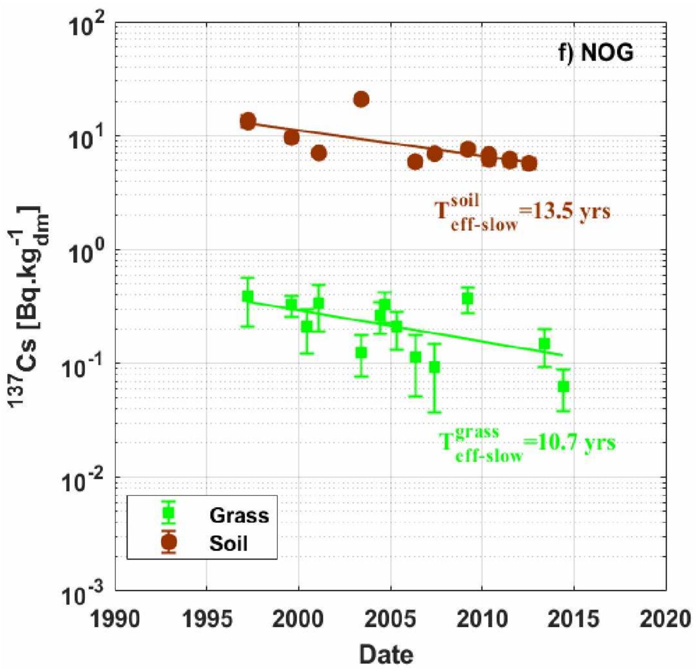

a) BUG
$T^{soil}_{eff-slow}$=21.4 yrs
Soil
^{137}Cs [Bq.kg$^{-1}_{dm}$]
Date


b) CAT
$T^{soil}_{eff-slow}$=14.2 yrs
Soil
^{137}Cs [Bq.kg$^{-1}_{dm}$]
Date


c) CRU
^{137}Cs [Bq.kg$^{-1}_{dm}$ or Bq.L^{-1}]
$T^{soil}_{eff\text{-}slow}$=24.9 yrs
$T^{milk}_{eff\text{-}slow}$=12.6 yrs
Milk
Soil
Date


d) FES
^{137}Cs [Bq.kg$^{-1}_{dm}$]
$T^{grass}_{eff\text{-}slow}$=19.8 yrs
Grass
Date


e) MER
Milk
$T^{milk}_{eff\text{-}slow}$=6.5 yrs
^{137}Cs [Bq.L^{-1}]
Date
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
1990 1995 2000 2005 2010 2015 2020


f) NOG
$T^{soil}_{eff\text{-}slow}$=13.5 yrs
$T^{grass}_{eff\text{-}slow}$=10.7 yrs
Grass
Soil
^{137}Cs [Bq.kg$^{-1}_{dm}$]
Date
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
1990 1995 2000 2005 2010 2015 2020

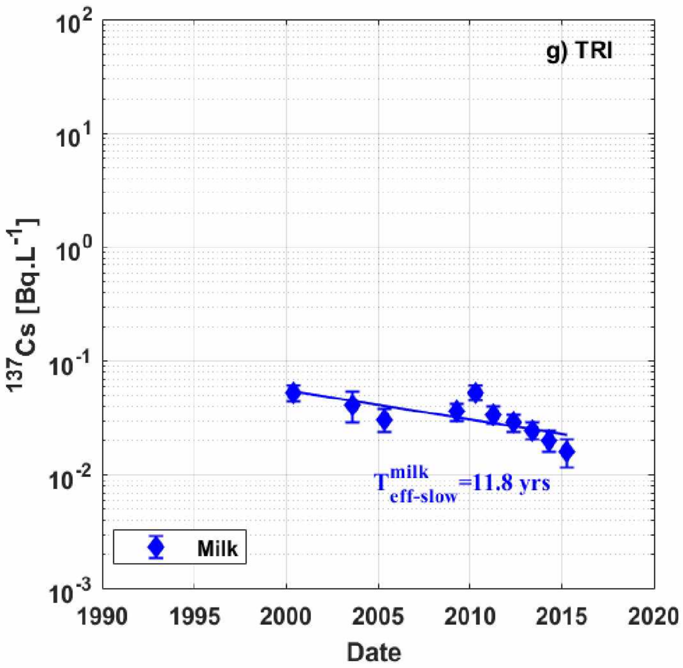


Fig. A.1: Series of ^{137}Cs activity concentrations in soil grass and milk at three sites a) Bugey (BUG), b) Cattenom (CAT), c) Cruas (CRU), d) Fessenheim (FES), e) Mercantour (MER), f) Nogent (NOG) and g) Tricastin (TRI). ^{137}Cs activity concentrations in milk are given in Bq.L^{-1} whereas ^{137}Cs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