Evaluation of ^{137}Cs bioavailability decay in French pastures from long-term field monitoring

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Soil-to-plant transfer of ^{137}Cs to grass and crops is a crucial pathway since this is considered as a long-lasting input of radioactivity into the human food chain. A long-term radiological monitoring of remaining artificial radionuclides from atmospheric nuclear tests and Chernobyl accident has been conducted by IRSN since the 1990's at a dozen of pasture plots located in France. The sampling sites are located in areas heavily affected by the atmospheric depositions, but also in the environment of nuclear facilities. The monitoring involves measurements of ^{137}Cs activity in upper soil (0-5 cm), grass and cattle milk, continuously sampled at the same plot, leading to a total of about 600 data available for analysis. The observed values of the soil-to-plant transfer factor ("field Tf"), defined as the ratio between ^{137}Cs activities in vegetation and soil , vary by more than one order of magnitude from one site to another, with a mean value which is significantly lower than those given by IAEA (2010) (0.29 and 0.18 for sandy and clayed soils, respectively). It is often assumed that environmental parameters such as soil physical and chemical properties (clay content, but also pH, organic matter, exchangeable potassium, etc.) or plants species can explain a part of the variability observed between sites. The issue of the present research is to compare observed Tfs with theoretical values predicted from semi-mechanistic models derived mainly from experiments and available in the literature (Smolders et al., 1997; Absalom et al., 1999; Absalom et al., 2001; Tarsitano et al., 2011). "Predicted Tf" requires the determination of empirical parameters such as the labile distribution coefficient Kdl at the soil-soil solution interface and the concentration factor CF, at the soil solution-roots interface. The calculation of Kdl and CF requires knowledge of the soil physico-chemical parameters cited before. An additional dynamic parameter (namely Dt) is needed which represents the ageing of ^{137}Cs in the rooting layer, defined as the percentage of bioavailable ^{137}Cs with respect to time. It is usually assumed that Dt obeys a kinetics equation involving two kinetic rates, i.e. a fast and a slow components denoted k_{fast} (1.9 10^{-3} d^{-1}) and k_{slow} (1.9 10^{-4} d^{-1}) respectively, which values must be ascertained (Absalom et al., 1999). Special attention should be paid to the calculation of Dt and hence on the chosen values of k_{fast} and k_{slow} which drastically influence the "predicted Tf" on the long term. Thus, before any comparison between "predicted Tf" and "field Tf", an attempt is made in the present study to evaluate k_{slow} using our time observations of ^{137}Cs activities in grassland plants and milk. k_{fast} could not be evaluated because the monitoring started in the 90's, several years after deposition. The long-term time series of ^{137}Cs activity in vegetation and milk recorded at 13 plots of the French territory allows us to deduce an apparent decay rate for each plot, l_{app} (d^{-1}), from which the corresponding decay rate of ^{137}Cs activity in the upper soil layer, l_{mig} (d^{-1}), was subtracted. This enables to estimate the slow component of the bioavailability factor Dt, i.e. $k_{slow=}l_{app}$-l_{mig}, with two distinct values at each site (for grass and milk). The observed median values of k_{slow} are 1.6 10^{-4} and 1.7 10^{-4} d^{-1} for respectively grass and milk. These are close to values deduced from data acquired in Switzerland (four monitoring plots) using the same methodology, namely 1.3 10^{-4} and 2.3E 10^{-5} d^{-1} (Corcho-Alvarado et al., 2016). Our results thus corroborate the time constant used in the Tf's equations according to Absalom et al. (1999). Thus in the following of this research project a site specific value of Dt will allow us to estimate Tf for each monitored plot.

Keywords: 137Cs, soil, plants, bioavailability, modelling