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Equilibrium and kinetic approaches for modelling sorption processes on radiocesium soil profiles in Fukushima prefecture

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The study of radionuclides (RNs) retention processes onto the solid phases is a key element for the prediction of their transfer in soils. It allows a better quantification of the persistence of radioactive contaminants on the soil surface, their availability for root uptake and their vertical transfer towards groundwater.

This work addresses the comparison between equilibrium and kinetic hypotheses of sorption processes on real post-accidental soil contamination profiles. The equilibrium-kinetic (EK) sorption model was selected as a non-equilibrium parameterization embedding the K_d approach. It supposes the existence of two types of sorption sites. The first sites are at equilibrium with solution, whereas for the second sites, kinetics of the sorption and desorption are taken into consideration.

We focused our study on four ^{137}Cs soil contamination profiles measured in a cedar stand 35 km northwest of the Fukushima Dai-ichi Nuclear Power Plant. Profiles were sampled at four different dates (between 2013 and 2018) by measuring ^{137}Cs activity in both organic (humus + litter layer) and mineral soil layers reaching a maximum depth of 20cm.

To successfully simulate the ^{137}Cs transfer throughout these soil profiles, the input flux at the mineral soil surface was reconstructed from monitored throughfall, stemflow and litterfall fluxes in the same forest stand from July 2011 to November 2016 crossed with initial deposit and dynamic of the organic layer activity.

The EK model reproduced the measured contamination profiles slightly better than the fitted K_d model. While both models were able to reproduce the overall vertical distribution throughout the profiles, the persistent contamination at the surface was closer to the measured value with the EK approach. Additionally, the fitted K_d parameters (2000 L/kg to 6500 L/kg depending on the parcel) were considerably higher than the recommended value by The IAEA for organic soils (270 L/kg). When used, this recommended K_d value produced profiles with considerably faster transfer rate between layers and shorter persistence of the contamination at the surface.

To further distinguish the models behaviors, long term simulations were conducted. EK hypotheses induced much longer residence time of the contamination at the soil surface. For instance, by 2030, the EK approach predicted that 75 % of the contamination still remained in the

0-2 cm layer due to the slow desorption rate, whereas the K_d approach predicted it to be around 51 %. This fraction becomes even smaller (8 %) when using the K_d value (270 L/kg) recommended by the IAEA for organic soils.

These results prove that the choice of the sorption model is critical in post-accidental situations. An equilibrium approach, especially when using recommended parameter values, can result in an underestimation of the RNs residence time in the surface. Whereas a kinetic approach, by distinguishing different sorption and desorption rates, is able to reproduce the slow evolution of ^{137}Cs soil profiles with time that is already observed in the case of Chernobyl contaminated areas 30 years after the accident.