Evaluation of the radiocaesium behaviour in agricultural Japanese soils based on potassium fertilisation, zeolite amendment and clay mineralogy

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High rates of potassium (K) fertilisation are used in arable soils affected by the Fukushima Daichii Nuclear Power Plant accident of 2011, in order to further reduce the uptake of radiocaesium (RCs) by plants. Additionally, zeolite has been applied to decrease soil solution RCs following topsoil removal. However, there is uncertainty on the role of zeolite in the uptake of RCs in Japanese soils. In this study, we compared RCs sorption in three soils with major differences in clay mineralogy: a Cambisol rich in vermiculite with strong retention of monovalent cations; an Andosol with very low 2:1 phyllosilicate content and with low K and Cs affinity; and a clay-rich, lowland smectitic Gleysol with high water holding capacity. We elucidated their solid-liquid distribution of K and \textsuperscript{133}Cs (as a proxy for RCs) in response to K addition as simulating K fertilisation, and also to zeolite (clinoptilolite) addition. The Radiocaesium Interception Potential (RIP), which is a key parameter that determines the RCs selectivity in soil and its phytoavailability, was analysed by spiking 1 g of soil with 1-2 KBq of \textsuperscript{134}Cs followed by a determination of solution \textsuperscript{134}Cs. The data were used to predict the soil-to-plant transfer factor (TF) based on a simplified version of the current RCs TF model. Our results showed that the vermiculitic soils had the lowest increase in exchangeable K (ex-K) at a given K dose, given its strong fixation in the 2:1 phyllosilicate layers, as opposed to the Andosol. Zeolite addition was shown to increase most of the soils RIP and thus proved its ability to adsorb RCs. Besides, zeolite addition also diminished both soil solution Cs (Cs_{ss}) and K (K_{ss}) concentrations for the allophanic Andosol. The K and Cs selectivity of the soil increased by zeolite addition, thus K_{ss} crucial for RCs uptake, consequently declined. This decrease observed for K_{ss} would be a reason for the reported ineffectiveness of zeolite application in previous studies. Solid-liquid distribution coefficients for exchangeable Cs (ex-Cs) suggest that the extraction determined
by with 1M ammonium acetate does not constitute a reliable proxy for RCs as compared to $C_{SS}$. At low $K_{SS}$ range ($<$0.1 mmol·L$^{-1}$), our findings for the vermiculitic and smectitic soils showed a rapid increase of $C_{SS}$. This sharp increase was not foreseen in the currently defined RCs model of Absalom et al. (1999 and ulterior). It entails, according to our predictions, a clear underestimation of the TF in the model and therefore a higher risk of RCs transfer to crops than expected in the Fukushima Prefecture vicinities - if the ongoing K fertilisation scheme is discontinued. Additionally, our comparisons of the predicted TF based on $K_{SS}$ and ex-K showed that $K_{SS}$ may be used as a more precise parameter to assess zeolite amendments in Japanese soils.