COMMENTS ON “RADIATION-DAMAGE RESISTANCE IN PHYLLOSILICATE MINERALS FROM FIRST PRINCIPLES AND IMPLICATIONS FOR RADIOCESIUM AND STRONTIUM RETENTION IN SOILS” BY M. SASSI, K.M. ROSSO, M. OKUMURA, AND M. MACHIDA

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INTRODUCTION

Sassi et al. (2016) used ab initio molecular dynamics calculations to obtain threshold displacement energy (TDE) values for Mg, Si, Al, O, and H atoms within vermiculite. Then they used the TDE values “to estimate the probability of Frenkel-pair creation by direct electron–ion collision” upon β decay of $^{137}$Cs, $^{90}$Sr, and $^{90}$Y. In a context of concern about the long-term effects of fallout radionuclides, they used these probability values to imply substantial radiation damage to phyllosilicates in fallout-contaminated soils and sediments: “For $^{137}$Cs and $^{90}$Sr, the calculated probability is ~36%, while for $^{90}$Y the probability is much greater at ~89%. The long-term retention picture that emerges is that decay will progressively alter the clay interlayer structure and charge, probably leading to delamination of the clay, and re-release of residual parent isotopes.”

The TDE values may be useful for understanding the effects of radiation in vermiculite contaminated by high-level nuclear waste, but in soils and sediments contaminated by radioactive fallout the expected radiation damage owing to Frenkel-pair formation will be so little and so well dispersed that it will have no appreciable effect on retention of fallout radionuclides. That such radiation damage will be little is evident from comparison of the number density of radioactive atoms in fallout-contaminated soil ($<2 \times 10^{13}$ cm$^{-3}$) with the typical number density of unit cells ($\sim 1 \times 10^{21}$ cm$^{-3}$) in the 2:1 phyllosilicate minerals that hold most of the fallout radioactivity. That such radiation damage will be well dispersed is evident from the typical range in common silicates of $\beta^-$ particles having sufficient energy to displace most kinds of atoms in the minerals (from $\sim 0.1$ mm to $\sim 1$ mm), which is much greater than the dimensions of clay particles.

DISCUSSION

Concentration of fallout radioactivity in soils

The most heavily contaminated soil sample examined by Steinhauser et al. (2013, table S1), from a radioactivity hot spot 1.5 km from the damaged Fukushima I power plant, contained $6.7 \times 10^{12}$ radioactive atoms of Cs and Sr (predominantly $^{137}$Cs, $4.6 \times 10^{3}$ Bq g$^{-1}$) per g of dried soil. The radioactivity was much less in 16 soil samples representative of typical (not extreme) radioactivity in a belt of high fallout that runs to the northwest of the Fukushima accident site, corresponding to $1.6 \times 10^{11}$ atoms g$^{-1}$ for the most contaminated sample and to $8 \times 10^{10}$ atoms g$^{-1}$ on average (Sahoo et al., 2016). For comparison, there are $\sim 7 \times 10^{20}$ unit cells g$^{-1}$ of vermiculite. Because, on average, only about one atomic displacement should occur per $^{137}$Cs decay event (Hess et al., 2000), Frenkel-pair formation owing to β radiation should affect only a minute fraction of the unit cells of the clay minerals that hold most fallout radioactivity.

Most important for this discussion is that Kaneko et al. (2015) found $\sim 3 \times 10^{5}$ Bq g$^{-1}$, $\sim 4 \times 10^{12}$ atoms g$^{-1}$, of $^{137}$Cs in a $<1 \mu$m fraction separated from severely contaminated soil in which the clay held at least 78% of the $^{137}$Cs. Because particle volume in this fraction must have ranged downward from $\sim 1$ μm$^3$, the number of particles was $>4 \times 10^{11}$ g$^{-1}$. Fewer than 10 radioactive atoms per clay particle were in this severely contaminated soil clay.

Sassi et al. (2016) were aware that radionuclides are dilute at the scale of whole particles, but they asserted that radionuclides could become concentrated locally in clay structures: “A key element of the dose rate is the local concentration of emitters that accumulate in the structure by means of exchange. Early models focused on edge sites of phyllosilicates, not only as key entry points for exchange but also because locally increased concentrations of interlayer cesium could be found spectro-microscopically, probably due to the increased degrees of structural freedom available in these locations (McKinley et al., 2004). More recently, however, and at much higher spatial resolution, transmission electron microscopy studies have shown that cesium also easily...”

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DOI: 10.1346/CCMN.2017.064076