

## Modelling of Cs adsorption in natural mixed clays and the effects of ion competition

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Cesium-137 (half-life, ~30 years) is an important fission product from the irradiation of uranium-based fuels and it has been released in the past to soils and waters as a result of nuclear accidents or weapon testing. Radiocesium is particularly relevant from an environmental point of view because it always exists as the monovalent cation Cs<sup>+</sup>, which presents very high solubility.

Cesium migration in the environment is mainly controlled by sorption onto mineral surfaces; in particular, it is rather strongly adsorbed onto clays by ionic exchange. Thus, with the objective of designing a geochemical reactive barrier to treat Cs-137 pollution in a salt marsh zone, several natural clayrocks were analysed as possible sorbent. The composition of the rocks was variable and within the clayey fraction the mayor clay minerals were illite, smectite and kaolinite.

The high salinity of natural waters presents in the zone obliged the understanding and quantification of the the effects of competitive ions, which may hinder cesium adsorption. Furthermore the water chemical composition variability leads to significant differences in distribution coefficients ( $K_d$ ).

The semi-empiric approach in the analysis of experimental data, above all when sorption is non-linear (as in the case of Cs) is unsatisfactory.

Thus, the development of sorption models, validated by experimental data, obtained under conditions representative of the site is needed. A multisite cation exchange model, considering both the solid (mixed clays) and water composition was used to successfully explain cesium sorption in the analysed natural clayrocks and the effects of highly competitive ions present in the aqueous phase, in particular K<sup>+</sup>, NH<sub>4</sub><sup>+</sup> and Na<sup>+</sup>.

## Removing the “heavy mineral effect” to obtain a new Pb isotopic value for the upper continental crust

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Knowing the average Pb isotopic composition of the continental crust is crucial to constrain the evolution of the Earth since its formation. Current estimates are rare. Some come from Earth evolution models but most are based on data acquired from river or oceanic sediments. The latter assume that sediments are representative of their continental sources but bias can be introduced if sedimentary mineral sorting is ignored. Several authors already demonstrated that Hf isotopes are fractionated by the so-called “zircon effect”, i.e. preferential concentration of unradiogenic Hf-rich zircons in coarse sediments which produces finer sediments with much more radiogenic Hf isotopes than their continental source. Because heavy minerals have sometime extremely radiogenic Pb isotopes, we evaluate the impact of the “heavy mineral effect” on Pb isotopes of sediments.

Here, we report Pb isotopic compositions of Himalayan river sediments as well as those of several grain-size fractions and pure mineral separates. We demonstrate that Pb isotopes of both bedloads and suspended loads are biased towards more radiogenic values than their source rocks due to a “heavy mineral effect” caused by mineral sorting during fluvial transport on continents. The sparse zircons, monazites and allanites present in all samples (< 1 wt%), including suspended loads, generate a Pb isotopic variability ( $37.85 < {}^{208}\text{Pb}/{}^{204}\text{Pb} < 43.16$  ;  $15.62 < {}^{207}\text{Pb}/{}^{204}\text{Pb} < 16.18$  ;  $17.83 < {}^{206}\text{Pb}/{}^{204}\text{Pb} < 22.58$ ) as large as that observed in the Earth’s mantle. Our new data suggest that mineralogical effects must be corrected for sediment Pb isotopes to be used as provenance and anthropogenic tracers. After correction of the mineralogical effect, we propose an average value for the composition of the upper Himalayan crust and a new Pb isotopic value for the Earth’s upper continental crust i.e.  ${}^{208}\text{Pb}/{}^{204}\text{Pb} = 39.16$ ;  ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 15.73$  ;  ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 18.95$ .