

## Implications of grainsize heterogeneity on Sr-90 adsorption

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The fission product strontium-90 is a major contaminant at nuclear sites worldwide. Sr-90 transport is sorption controlled and the geochemical processes affecting adsorption to <2mm fraction have been widely studied. The complex subsurface underlying Sellafield (UK nuclear site) is characterised by Quaternary glacial deposits with grain sizes ranging from clay to gravel. Understanding the geochemistry of >2mm fraction is thus essential for contaminant modelling to predict the fate of Sr-90. Here we used bulk sediments in batch experiments to quantify the effect of increasing grain size on  $K_d$  and to characterise the geochemistry of >2mm fraction in terms of pH and ionic strength.

The  $K_d$  values for five heterogeneous sediments relevant to Sellafield shows a fourfold increase from 10 L kg<sup>-1</sup> to 46 L kg<sup>-1</sup>. To mitigate the risk of overestimating adsorption to coarse material large scale experimentation is needed, or the sample should be sieved to <8mm to provide  $K_d$  within range of the  $K_{d_{bulk}}$  (18 L kg<sup>-1</sup>). Gravel corrections have been applied to heterogeneous sediment to determine the efficacy of estimating the  $K_d$  of the bulk sample from the <2mm fraction [1]. Surface area gravel correction using radii estimations provides the closest estimation of  $K_{d_{bulk}}$  (17.9 L kg<sup>-1</sup>). This correction includes an estimation of a fine-grained coating on the quartz grains, supported by SEM images.

The effect of grain size heterogeneity on pH sorption edge has been demonstrated. A broad edge (pH 5-7) was observed for coarse material, compared to the sorption edge of fine sands at pH 4. Consequently,  $K_d$  of >2mm sediments is only valid at the measured pH, due to increased mineral heterogeneity and presence of Fe-oxides. The effect of sea level rise on desorption of Sr-90 was studied. When exposed to high ionic strength (0.8 – 1.0 mol L<sup>-1</sup>), sorption of Sr-90 converges to 10% for all grain sizes. Desorption at high IS is independent of grain size because of surface saturation by Na<sup>+</sup>.

Overall, our results show that contaminant modelling should use a surface area based gravel correction and consider pH effects in the case of co-contaminants (eg HNO<sub>3</sub>).

[1] Kaplan et al., (2000), Groundwater 38, 192-197.