Implications of grainsize heterogeneity on Sr-90 adsorption

Georgia Barker

Dr Ian Burke, Dr Jared West, Dr James Graham (NNL)

NATIONAL NUCLEAR LABORATORY

UNIVERSITY OF LEEDS

eegrb@leeds.ac.uk

Rationale and Aims of experimental studies

NATIONAL NUCLEAR LABORATORY

UNIVERSITY OF LEEDS

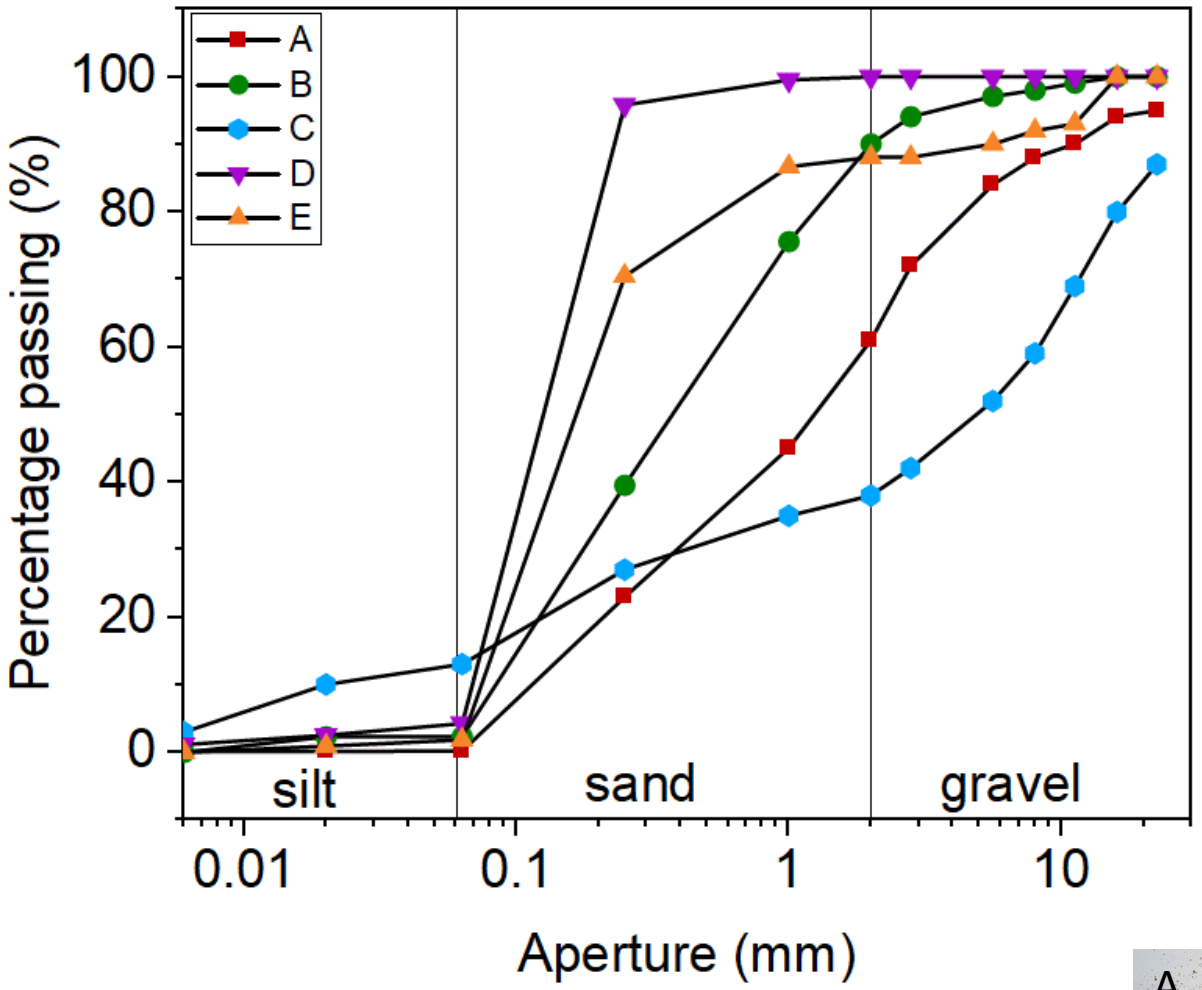
- Major contaminant at nuclear sites worldwide
- Contaminant plume under Sellafield Separation Area
- Sorption controlled transport in groundwater

- Effect of grainsize heterogeneity on adsorption of Sr-90
- Characterise geochemistry of >2mm grainsizes
- Can we assume >2mm fraction is inert?
- Improve contaminant modelling

Samples collected

Glacial sediments from
Nethertown
Seascale
Drigg Beach

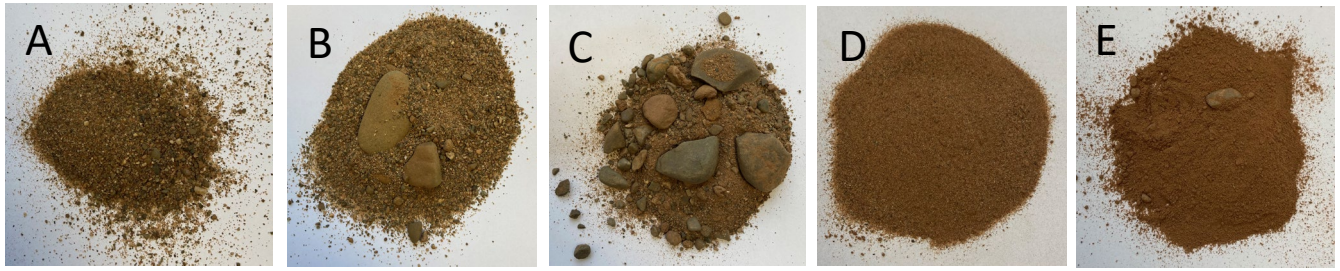
Sediment characterisation



CEC (Ross and Ketterings, 1995)
BET N_2 gas adsorption method

Sample	D_{50} (mm)	CEC (meq/100g)	SSA_{BET} (m^2/g)
A	0.40	0.78	3.73
B	1.20	1.04	2.61
C	4.00	1.24	2.90
D	0.14	6.10	5.28
E	0.18	5.28	9.56

BS ISO 11277:2009 (BS, 2009)



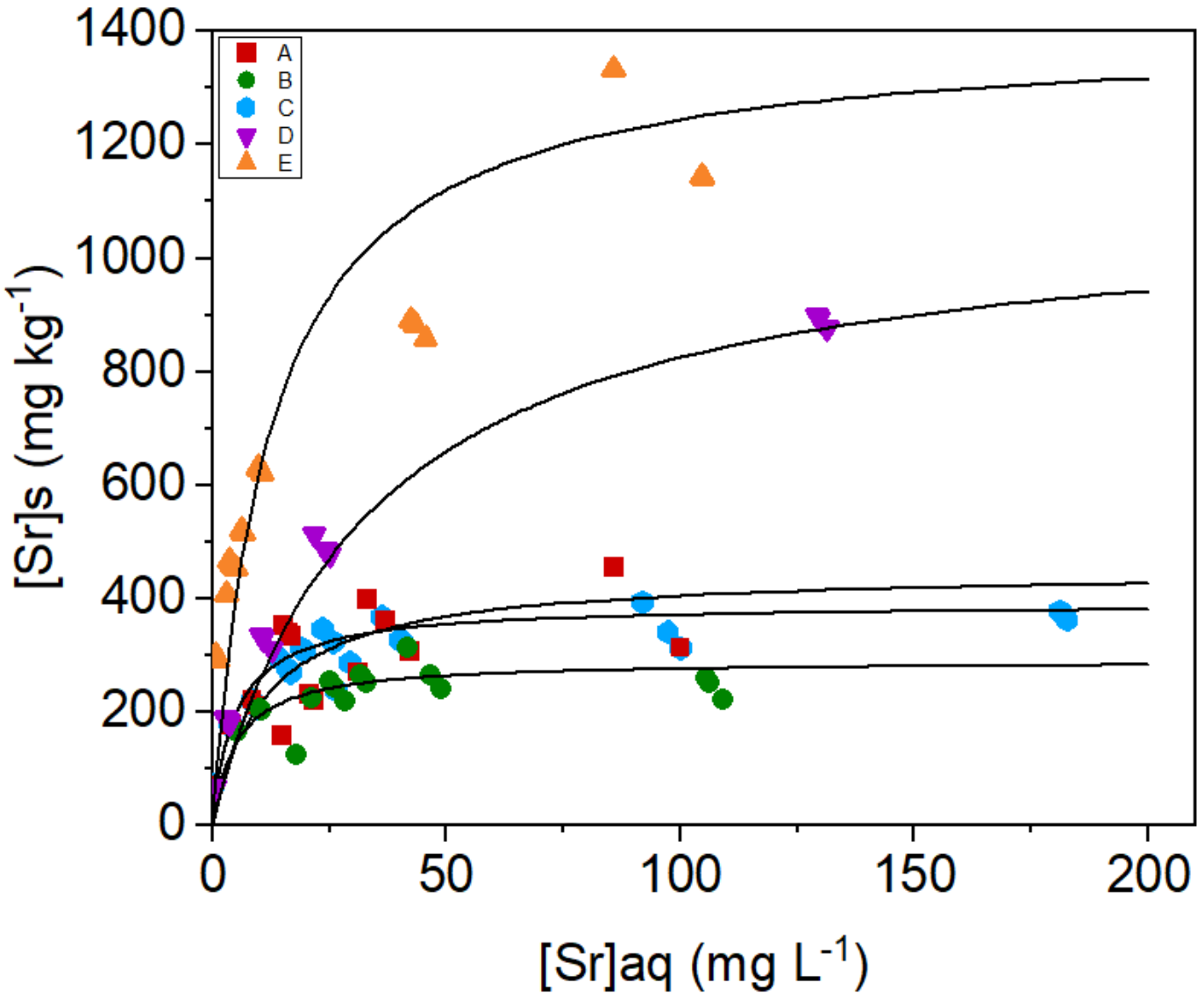
Batch adsorption tests : Isotherm

$$K_d = \frac{C_i - C_e}{C_e} \cdot SSR$$

$$[Sr\ sorbed] = \frac{q_{max} K_L [Sr]_{aq}}{1 + K_L [Sr]_{aq}}$$

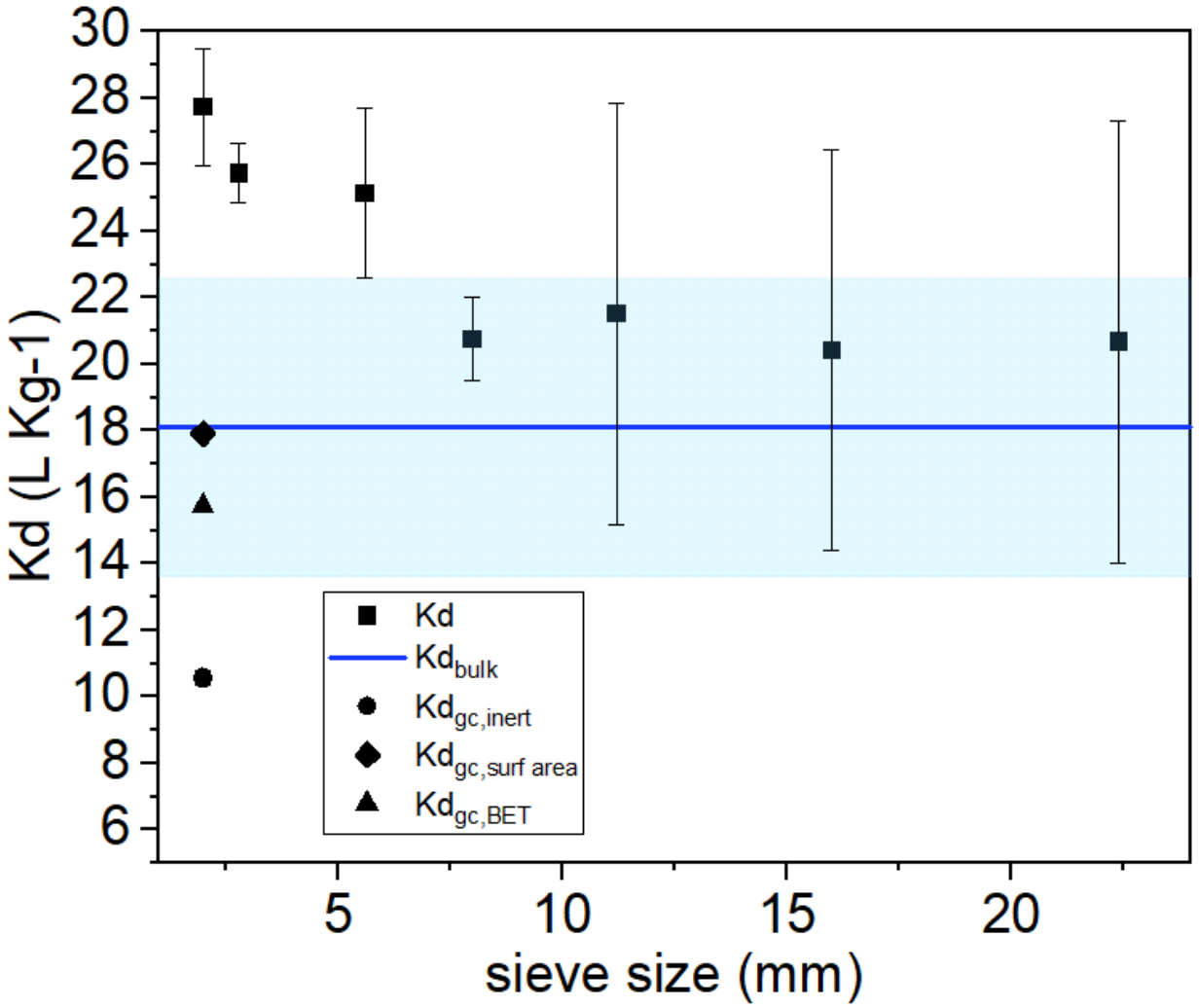
Sample	*K*d (L kg^{-1})	q_{max} (mg kg^{-1})	n
A	15	510	13
B	11	300	15
C	18	400	26
D	36	1200	9
E	46	1400	12

[Sr] 2.5 x 10^{-3} - 8.3 x 10^{-5} mol L^{-1}
[NaCl] 2.5 x 10^{-3} mol L^{-1}
10g L^{-1} Solid Solution Ratio
pH 6.5 - 7



Gravel Corrections

Standard practise sieves bulk sample to <2mm



Assumes >2mm **inert** ●

$$K_{d\,gc,inert} = (1 - f)K_{d<2mm}$$

Surface area correction using **SSA** ▲

$$K_{d\,gc,BET} = (1 - f)K_{d<2mm} + f\left[(K_{d<2mm})\left(\frac{SA_{>2mm}}{SA_{<2mm}}\right)\right]$$

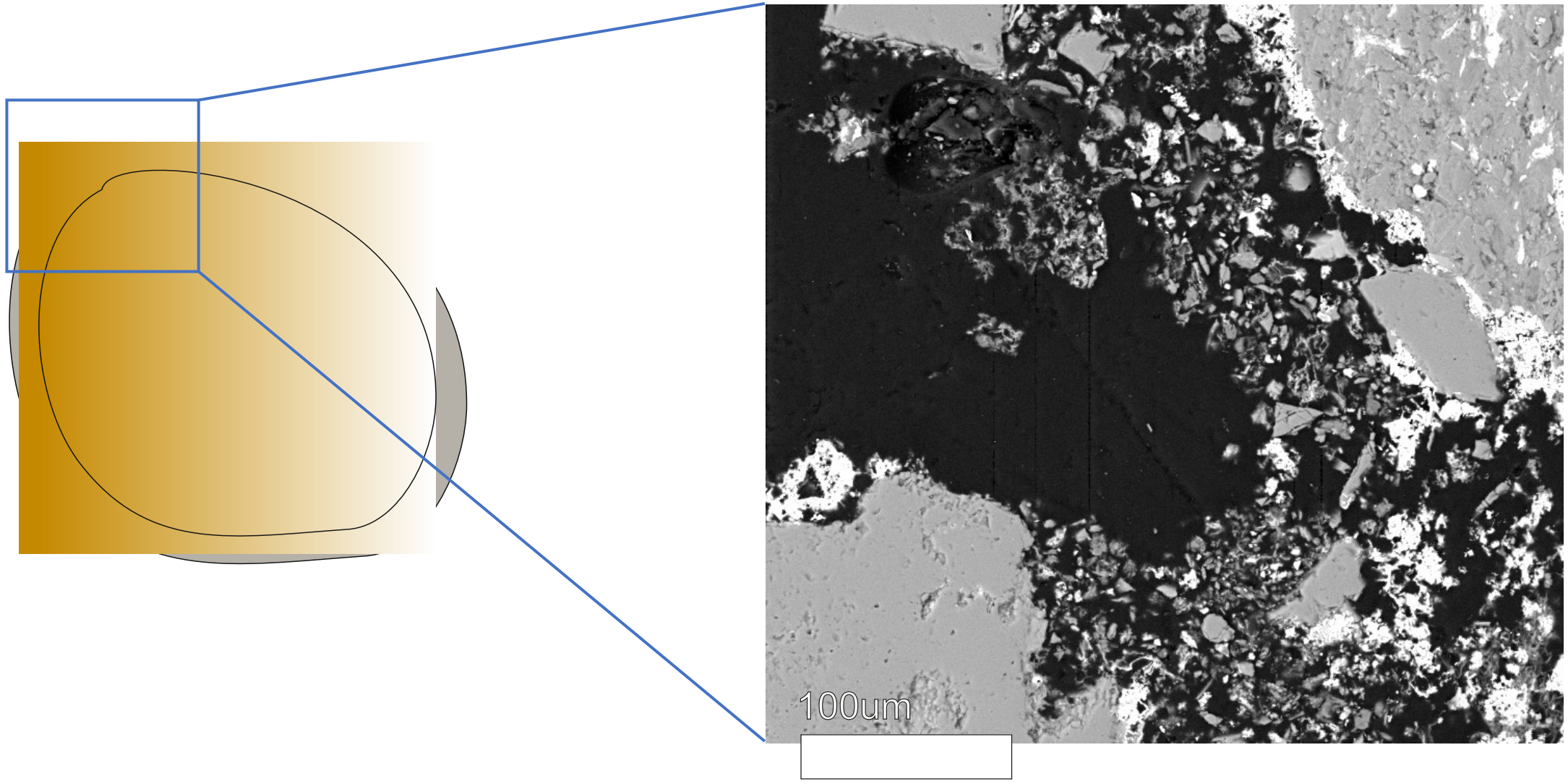
Surface area correction using **radius** estimations ◆

$$K_{d\,gc,SA} = (1 - (f \times 0.9))K_{d<2mm} + f\left[(K_{d<2mm})\left(\frac{r_{<2mm}}{r_{>2mm}}\right)\right]$$

$$r_{>2mm} = 3mm$$

(Kaplan et al., 2000)

SEM : topography of >2mm grains

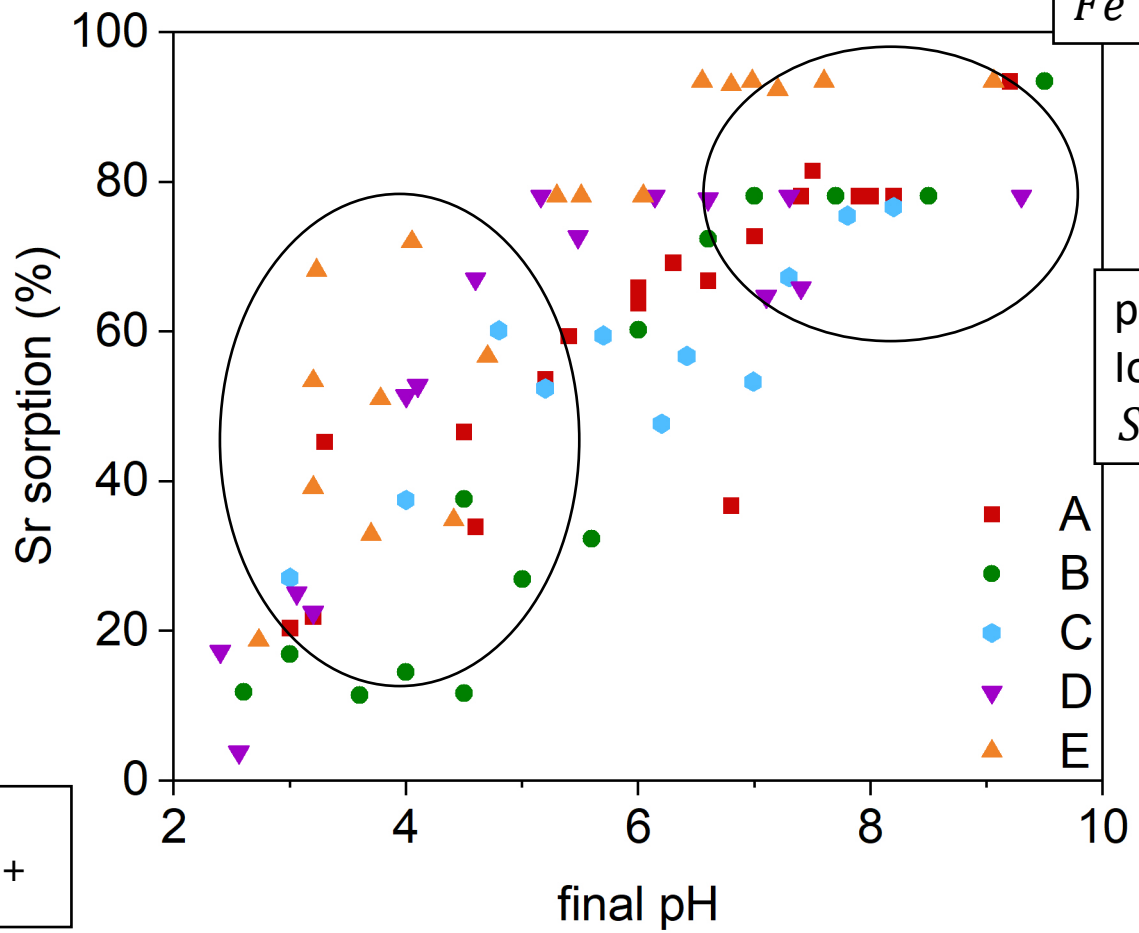


pH sorption edge

Fine sands
- Sorption edge pH 4
- Kd applicable pH >5

Coarse sediments
- Sorption edge pH 5 – 7
- Kd applicable only at pH measured at

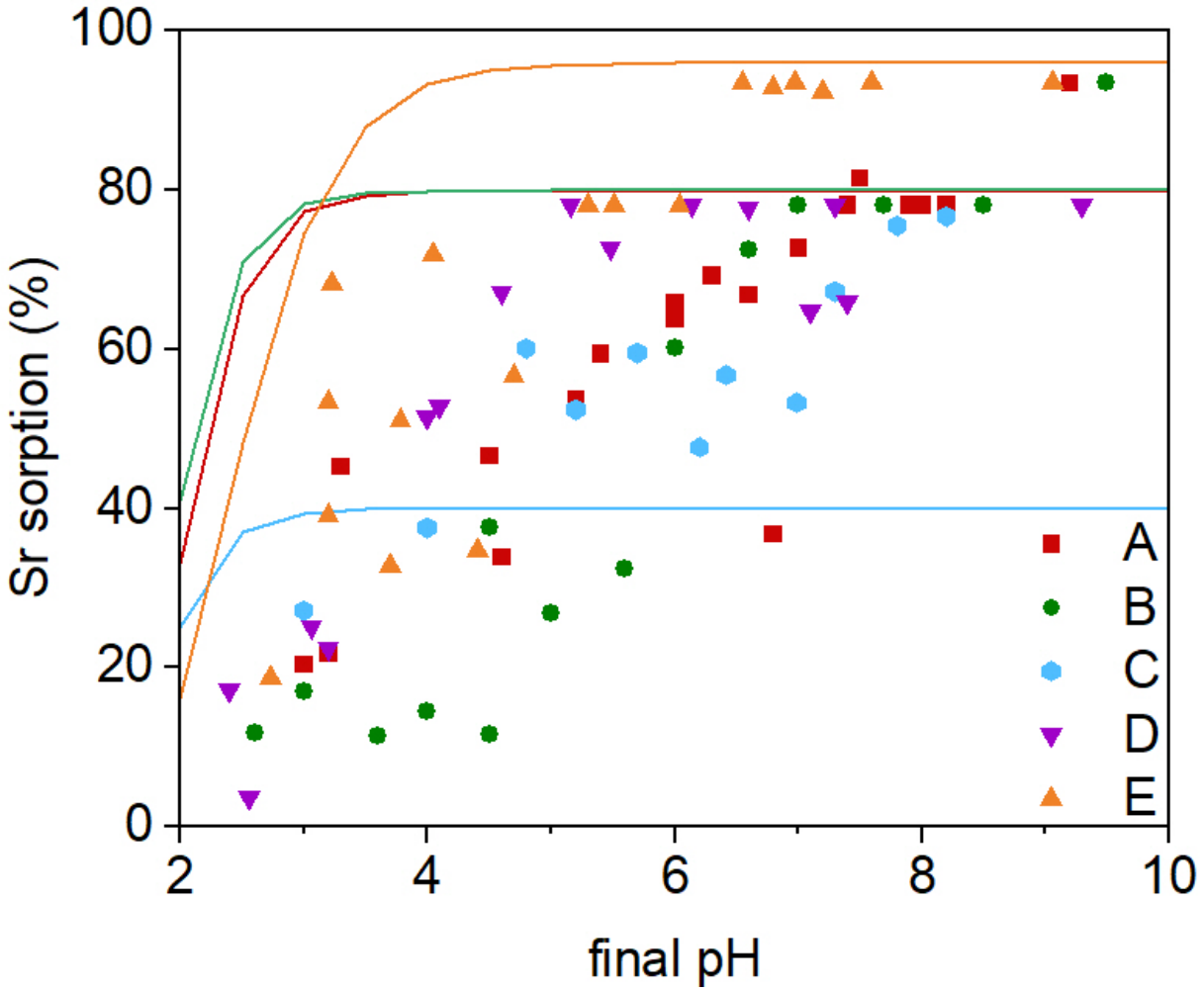
Clays
$Al - OH \leftrightarrow Al - O^- + H^+$



Fe oxides
$Fe - OH \leftrightarrow Fe - O^- + H^+$

pH 6 -7
Ion exchange
$Surf - Sr + 2H^+ \rightarrow Surf - H + Sr^{2+}$

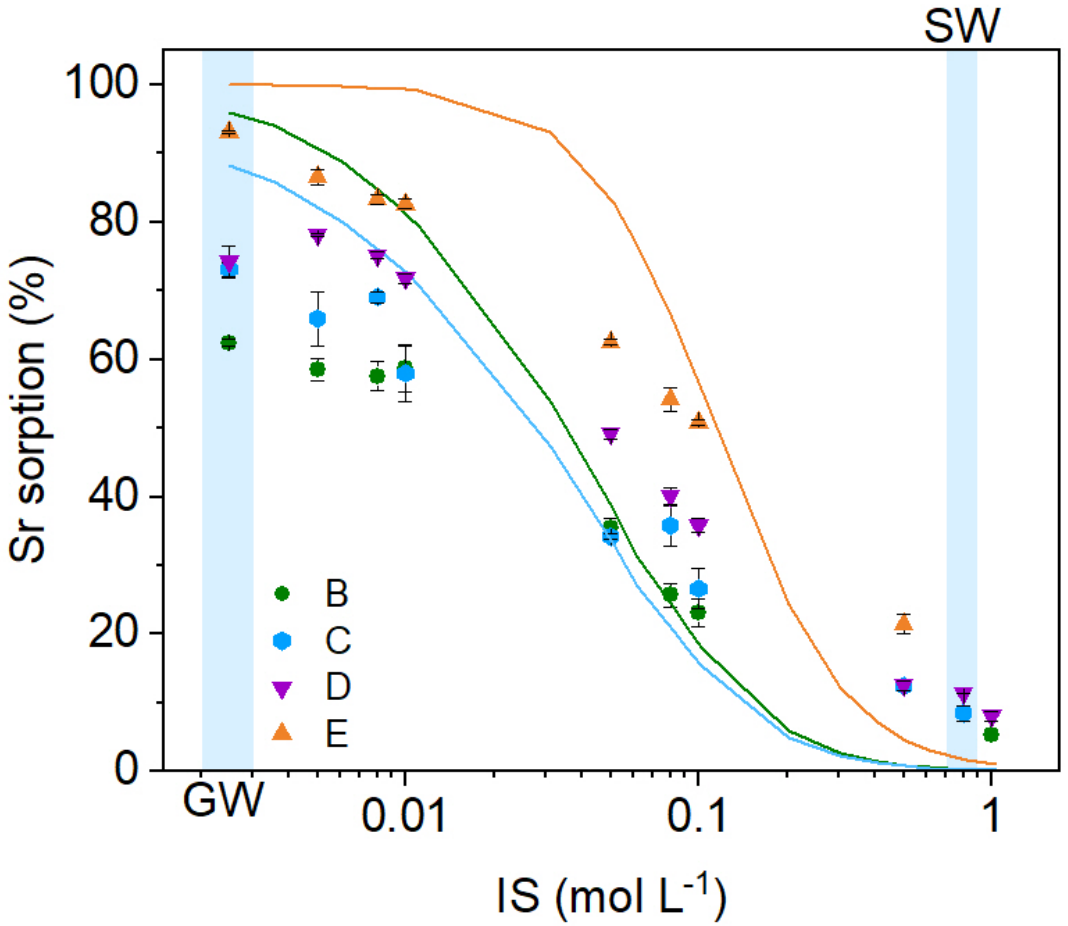
pH sorption edge



Lines show ion exchange model (PHREEQC)
Uses CEC values (D and E has the same model)

- Ion exchange model
- Sorption edge – pH 3
- Doesn't account for pH effects on mineral heterogeneity

Ionic strength : simulating sea level rise



Lines show ion exchange model (PHREEQC)
Uses CEC values (D and E has the same model)

Saline inundation
- Converges at high IS (0.8 mol L^{-1}, seawater)
- 10% Sr sorption for all grainsizes

Saline intrusion (0.1 mol L^{-1})
- Some difference in adsorption capacities

Cation exchange model
- Also converges at high IS
- Models Na^+ ion exchange well
- Underestimates Sr sorption at SW by 10%
- Assumes complete desorption of Sr^{2+} by Na^+

[Sr] 5 x 10^{-4} mol L^{-1}
[NaCl] 2.5 x 10^{-3} mol L^{-1}
10g L^{-1} Solid Solution Ratio
pH 6.5 - 7

Conclusion

1. Heterogeneous gravels provides an intermediate adsorption capacity due to the fines coatings on quartz >2mm grains.

2. Current method of assuming >2mm fraction is inert underestimates *K*d values. Gravel correction based off surface area provides the closest estimation of *K*d calculated for the bulk sample.

3. *K*d measured for heterogeneous coarse samples is only applicable at the pH it is measured at, this should be considered in contaminant modelling in case of co contaminants.

4. Desorption of Sr-90 under high ionic strength is independent of grainsize. Under saline conditions caused by sea level rise, 10% of Sr-90 would remain adsorbed to all sediments.

Thank you for listening

NATIONAL NUCLEAR LABORATORY

UNIVERSITY OF LEEDS