Contaminant Attenuation in Chalk Groundwater
A new approach using radiochemistry

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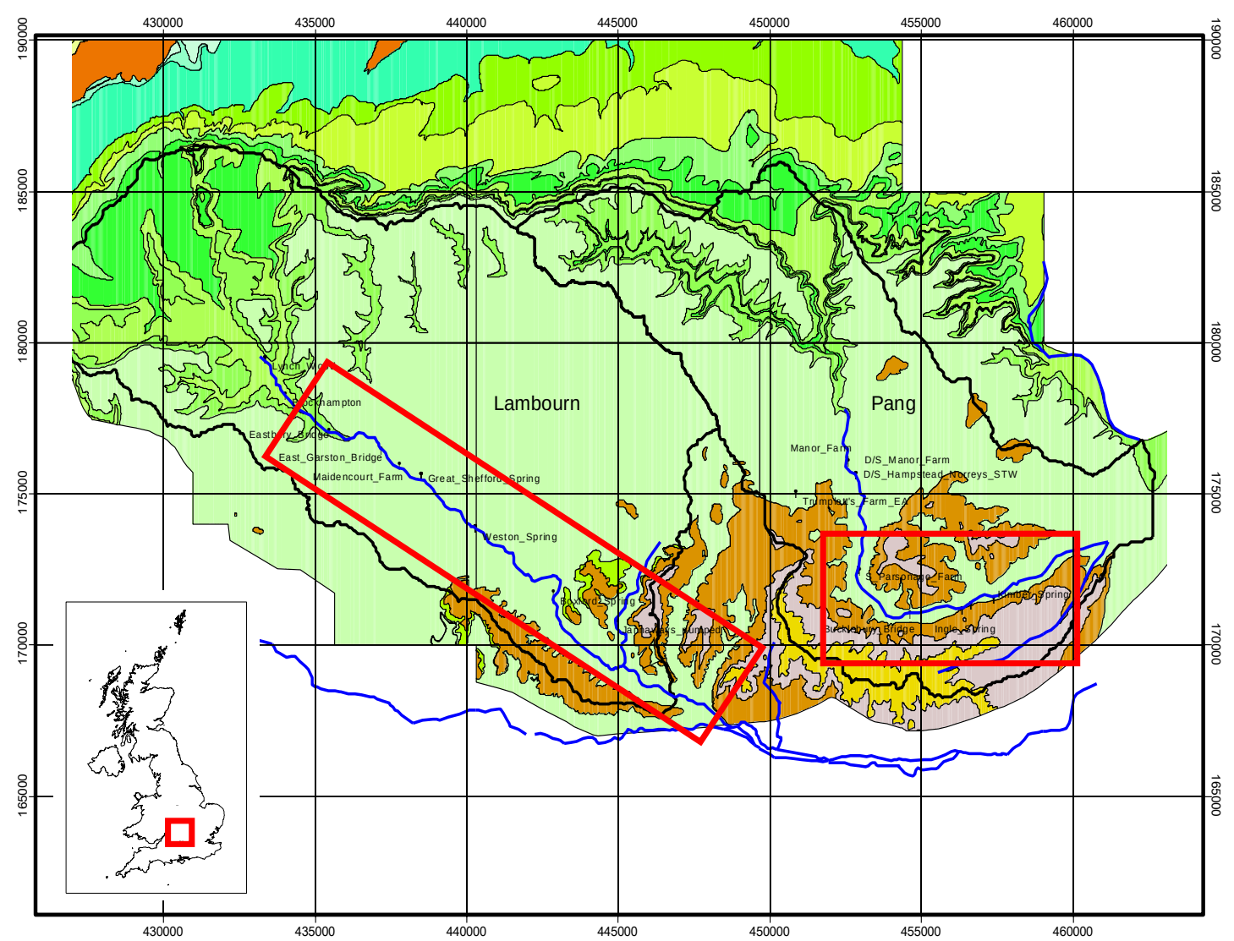
Project Summary

The aim of this NERC studentship is to investigate a potential radiochemical method for estimating the Chalk aquifer's capacity to attenuate contaminants.

Contaminant solutes are advected by groundwater flow through fractures, but are slowed and attenuated by molecular diffusion into immobile water in the pores of the Chalk (see right). Fracture apertures are the key factor controlling both advection and diffusion effects.

In principle apertures may be estimated by comparing dissolved radon (Rn) gas in fracture water with U-series isotope activities in the rock matrix, as Rn release and contaminant attenuation are both governed by similar molecular diffusion.

The project tests the robustness of a Rn-derived transport model developed by Atkinson and Barker(1) through a series of lab experiments and field observations. The results are compared against other more traditional tracer experiments.



Field Study Area – The Pang and Lambourn catchments located in West Berkshire. These catchments are underlain predominantly by Cretaceous Chalk, and both rivers are fed by the Chalk aquifer. In the south the Chalk is overlain by deposits of Palaeogene age. Superficial Quaternary deposits mainly consist of clay with flints, some of which are sandy(2).

Model Summary

Figure 1(a) sketches the dominant contaminant transport processes in fractured chalk in terms of a tracer pulse input. Groundwater flow in fractures advects the tracer across the diagram while concentrations are strongly attenuated by molecular diffusion into and out of immobile pore water in the blocks of chalk between the fractures.

These processes can be modelled in terms of two related parameters, fracture aperture, *a* and a time scale, t_{cf}, for diffusion to exchange tracer[1]. Our method estimates t_{cf} (which includes *a*) and rests on the fact that radon and contaminant concentrations are both governed by similar diffusion, see Figure 1(b).

Radon originates in the rock matrix by decay of U-series precursors. The radon concentration in fracture water is therefore controlled predominantly by diffusion through the pore space, but also by direct recoil from the fracture surfaces.

A simple mathematical model[1] relates t_{cf} to fracture ^{222}Rn activity, C_f and production rate, *P*, which in turn depends on the concentration of uranium in the matrix, *U* and the [^{226}Ra]/[^{238}U] activity ratio.

Figure 1(a) Contaminant transport processes in Chalk

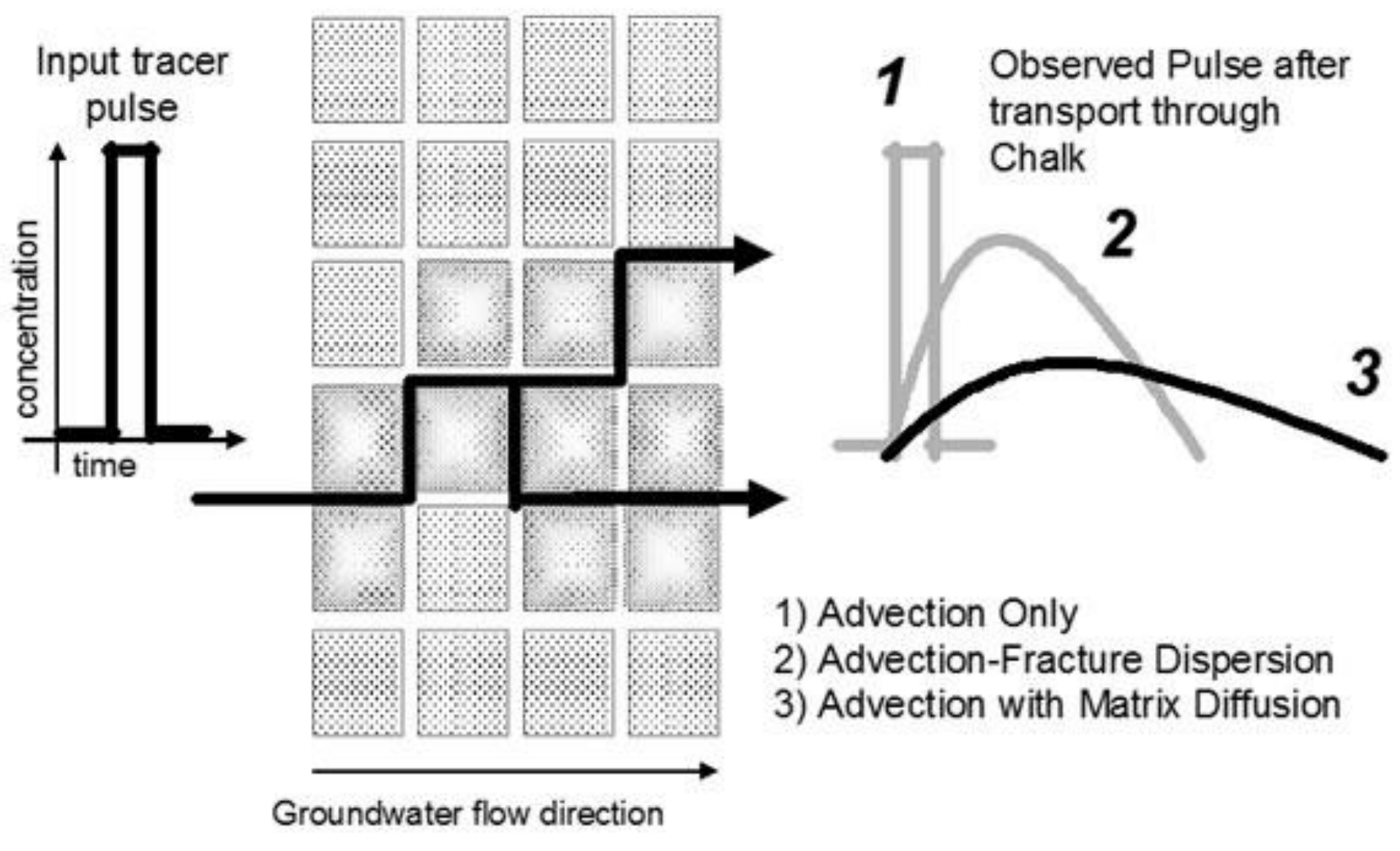
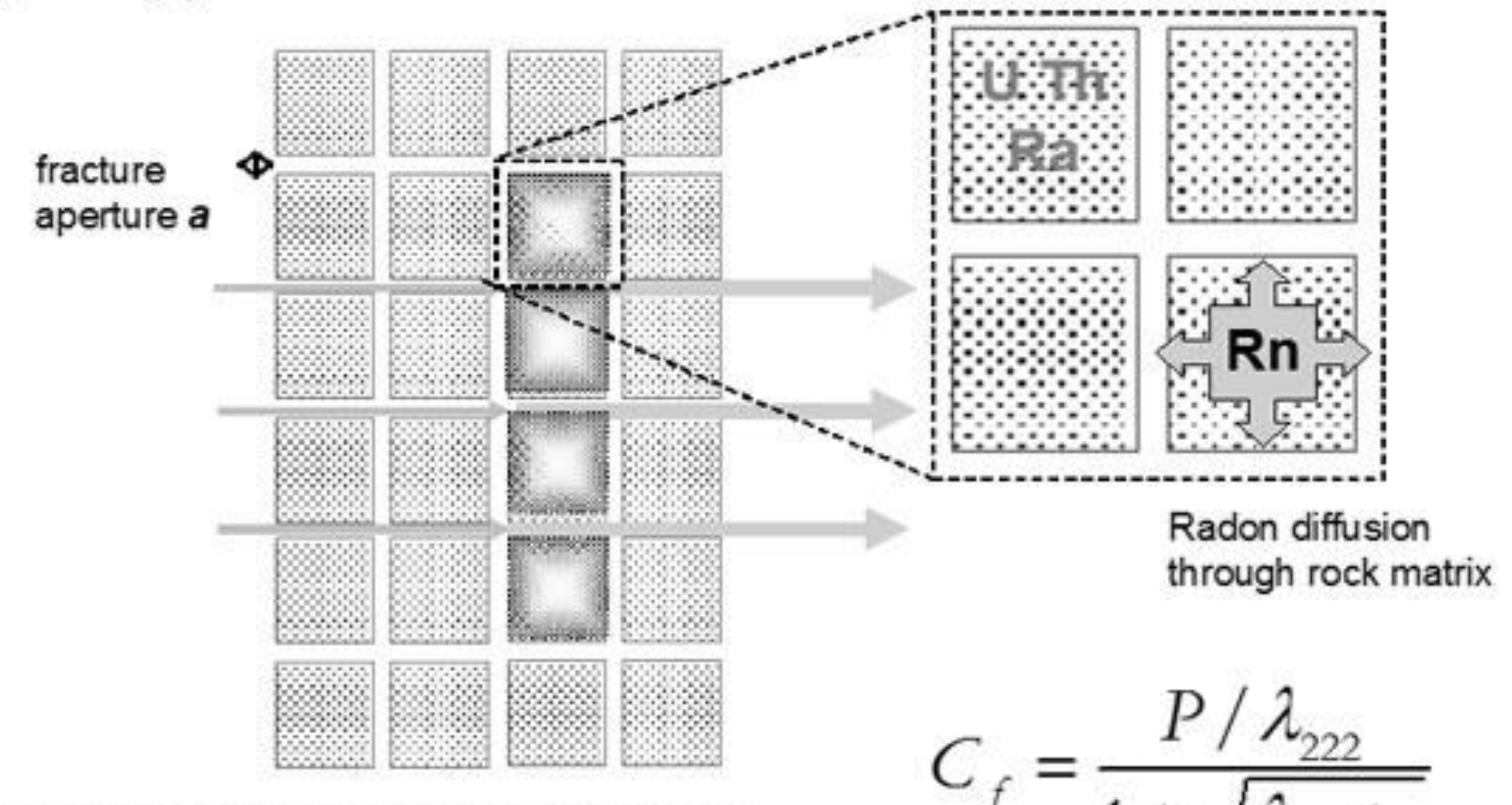


Figure 1(b) Radon emanation in Chalk by diffusion



$$C_f = \frac{P/\lambda_{222}}{1+\sqrt{\lambda_{222} t_{cf}}}$$

Fracture Water Rn Concentration (C_f), where:
P = production rate for radon
λ_{222} = decay constant for ^{222}Rn
λ_{238} = decay constant for ^{238}U
E = radon emanation factor
ρ_m = bulk density
ϕ= matrix porosity

$$P = 2.53\times10^{24}\,E\,\frac{\rho_m}{\phi}\lambda_{238}U\left[\frac{^{226}Ra}{^{228}U}\right]$$

Testing the Model Assumptions

As the model presented is relatively simple in its representation of the exchange of radon between the Chalk matrix and the fracture network a number of assumptions have had to be made. These have been tested in the laboratory and though a series of field observations.

1) **The aquifer system is in steady state with respect to radon transport from matrix to fissures.**

2) **The sample point is far enough from any boundaries (e.g. low Rn or high Rn inputs) for the Rn content of fissure water to be determined entirely by emanation of Rn from the matrix.**

Because the half life of Rn atoms is only 3.82 days, a state of equilibrium between production, diffusion and decay will occur if groundwater residence times are greater than about 25 days.

Dissolved radon concentrations have been measured at approximately monthly intervals at 10 spring locations and 6 borehole locations during 2005-2007. It is evident there are both differences in magnitude between individual locations, and that there is a consistent temporal trend throughout the year, with activity peaking in the summer months. This result is contrary to the work of others. Cuttell *et al.*(3) observed little or no variation in Lincolnshire Chalk boreholes, whereas Andrews and Wood(4) observed a decrease in summer in a more karstic Carboniferous limestone aquifer system.

Our observations suggest that water sampled at these locations (especially those samples from spring sources) may not be in steady state with the surrounding rock source and may indicate either low groundwater residence times in the shallow aquifer system or a mixing between several sources.

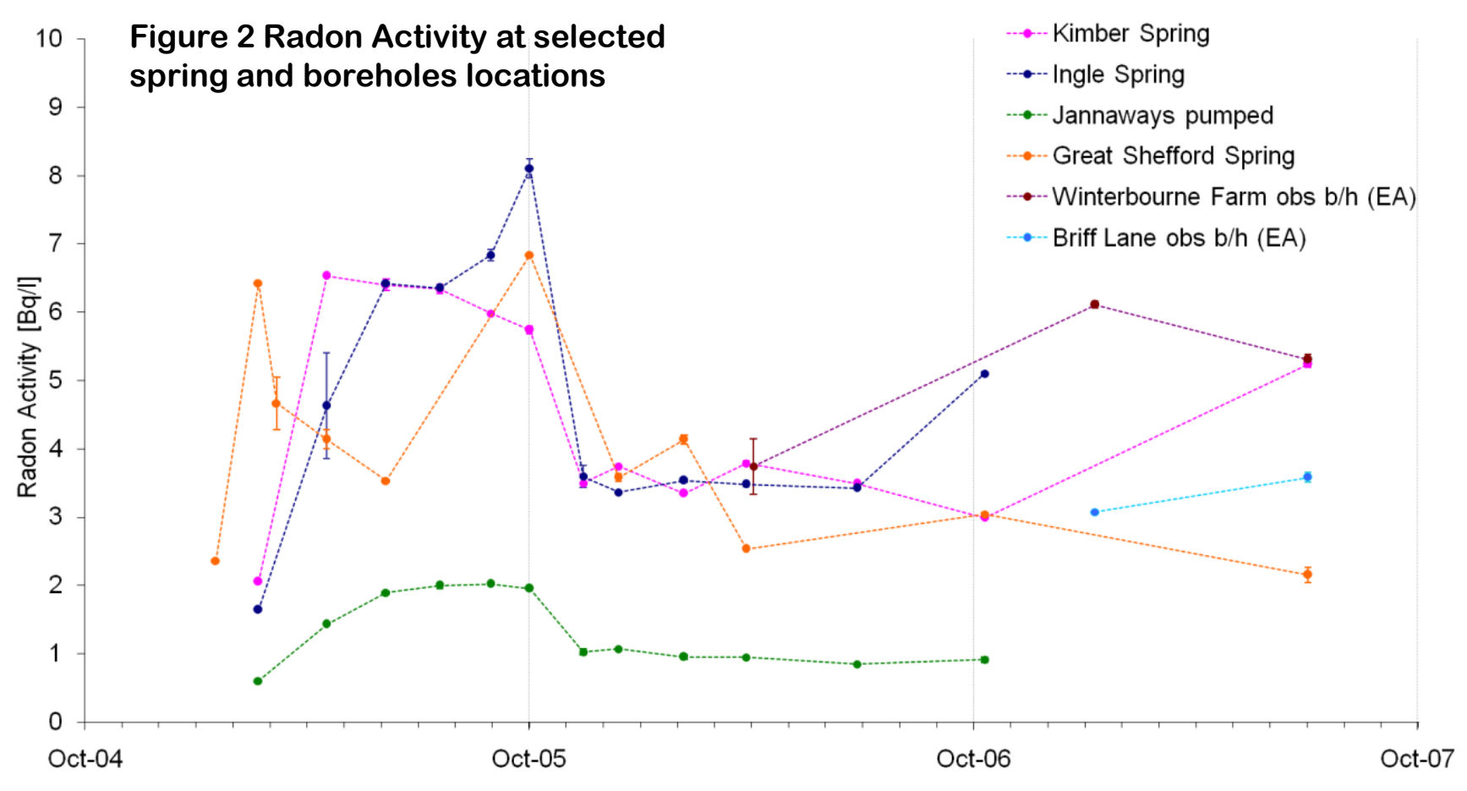


Figure 2 (left) summarizes radon activity from a selection of spring and borehole locations with the NERC LOCAR Pang and Lambourn catchments in Berkshire.

Data were collected at monthly to quarterly intervals and demonstrate significant seasonal variation. This is more pronounced at surface spring sites, where mixing of groundwater with other surface sources is considered more likely.

3) **There is a uniform source strength throughout the volume of Chalk represented by a groundwater radon measurement**

4) **Uranium concentration is an adequate and accurate surrogate for radon source strength, or ^{226}Ra concentration multiplied by an emanation factor.**

From the study of uranium and radium profiling of Chalk core (see adjacent poster for further details on this) it is clear that both radionuclides are subject to litho-stratigraphic control and that both the concentration and isotope activity ratio are subject to significant variation. Figure 3 investigates the effect that [^{226}Ra]/[^{238}U] disequilibrium would have on t_{cf}, the time scale for diffusion to exchange tracer in our current model, and demonstrates that a 3-fold range would result from the extremes of disequilibrium in existing UK Chalk rock matrix data[3,5]

From our work on subsets of solid and disaggregated core material it is also clear that the radon emanation rate from chalk particles to the matrix pores cannot be assumed to be 100%. In fact for silt particles (~63 µm) average rates of ~20% are more common. This is in line with values determined by Cuttell(3) in the Lincolnshire Chalk. Of course even this apparent emanation may be enhanced if a large amount of rapid diffusion occurs along grain boundaries, as suggested by Andrews *et al*(4), or if the immediate parent source of radon, i.e. radium, is concentrated on the surface of grains and not uniformly distributed through the material.

Assumed Radon and Rock Parameter Values				
A(Rn)	2.38 Bq.l^{-1}	E	1.0	
		ϕ	0.34	
		ρ_m	1755	kg.m^{-3}

Scoping of effect of U-series disequilibrium on model-derived contaminant transport parameter t_{cf}

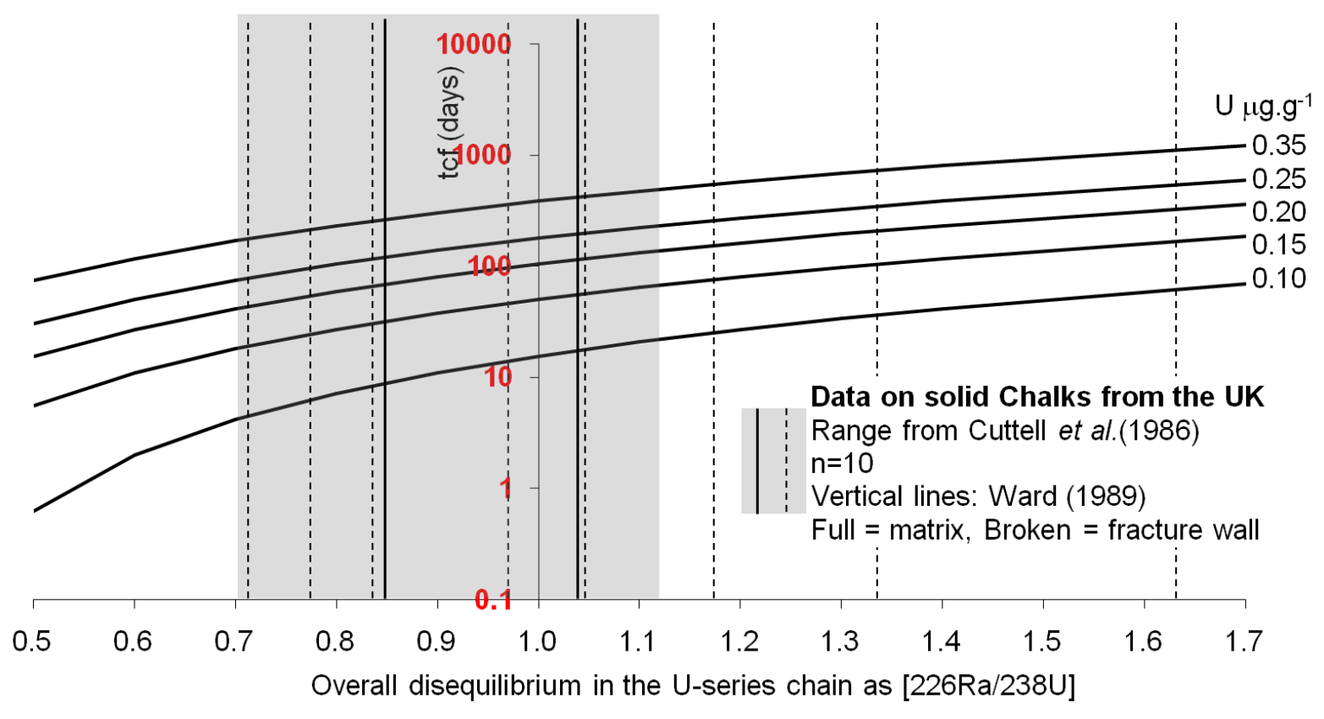


Figure 4 Matrix Radon concentration profile in fracture wall (and calculated tcf) for a known fracture radon concetraion and matrix U content , but variable emantion factor.

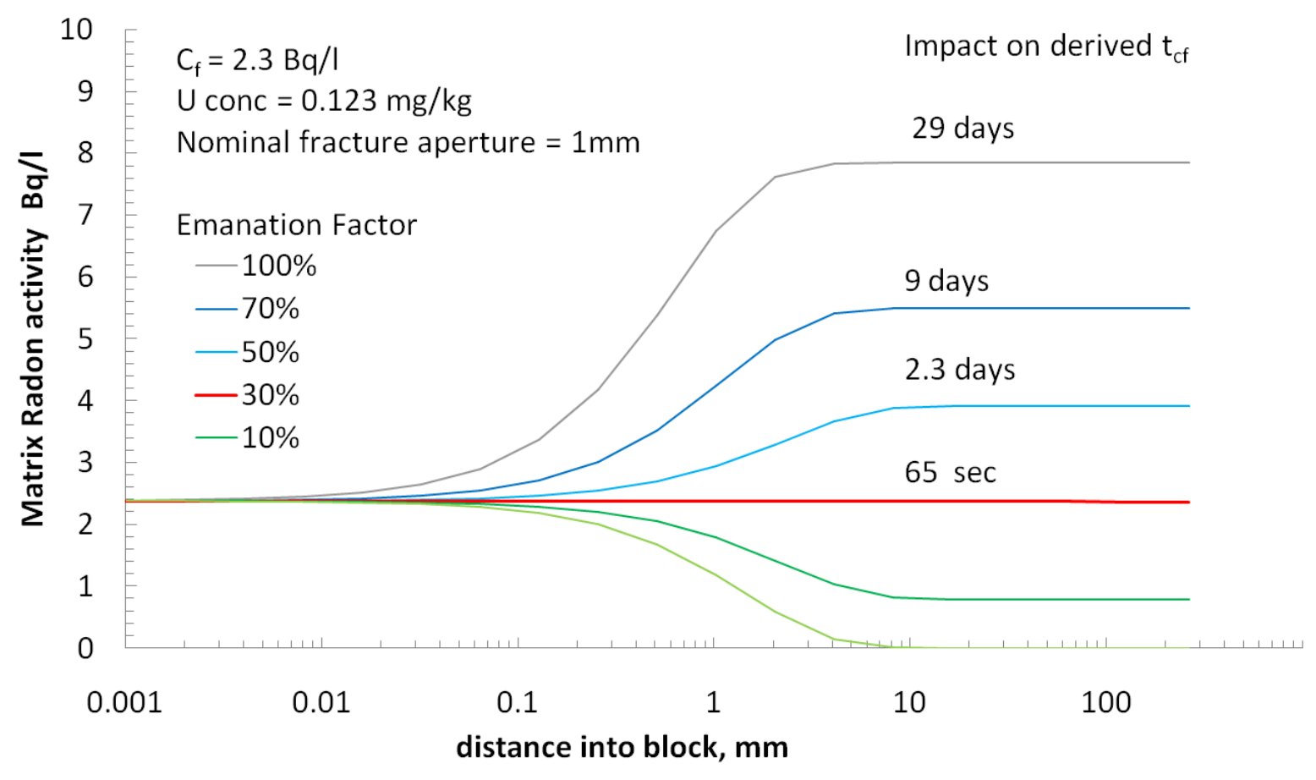
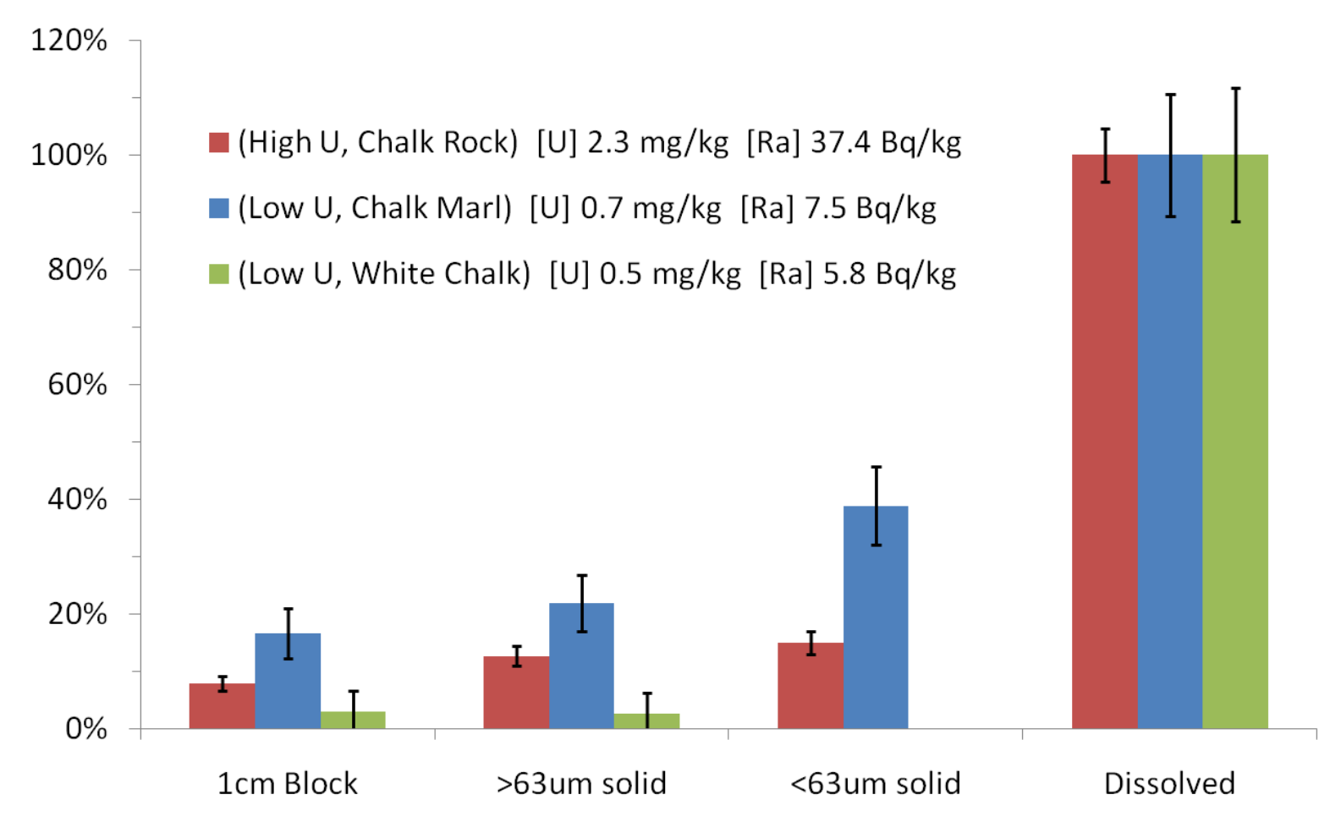


Figure 3 (above) illustrates the range of possible values of t_{cf} for a given groundwater radon content but variable matrix uranium concentration and [^{226}U]/[^{238}U] activity ratio.
Figure 4 (above right) shows the theoretical uncertainty of t_{cf} for a known groundwater radon activity and matrix uranium concentration, but with variable radon emanation factor.
Figure 5 (right) summarizes the calculation of radon emanation factors for 3 stratigraphically distinct samples of core material from the Trumplett's Farm borehole in the river Pang catchment. Each sample has also been split into a range of particle sizes.

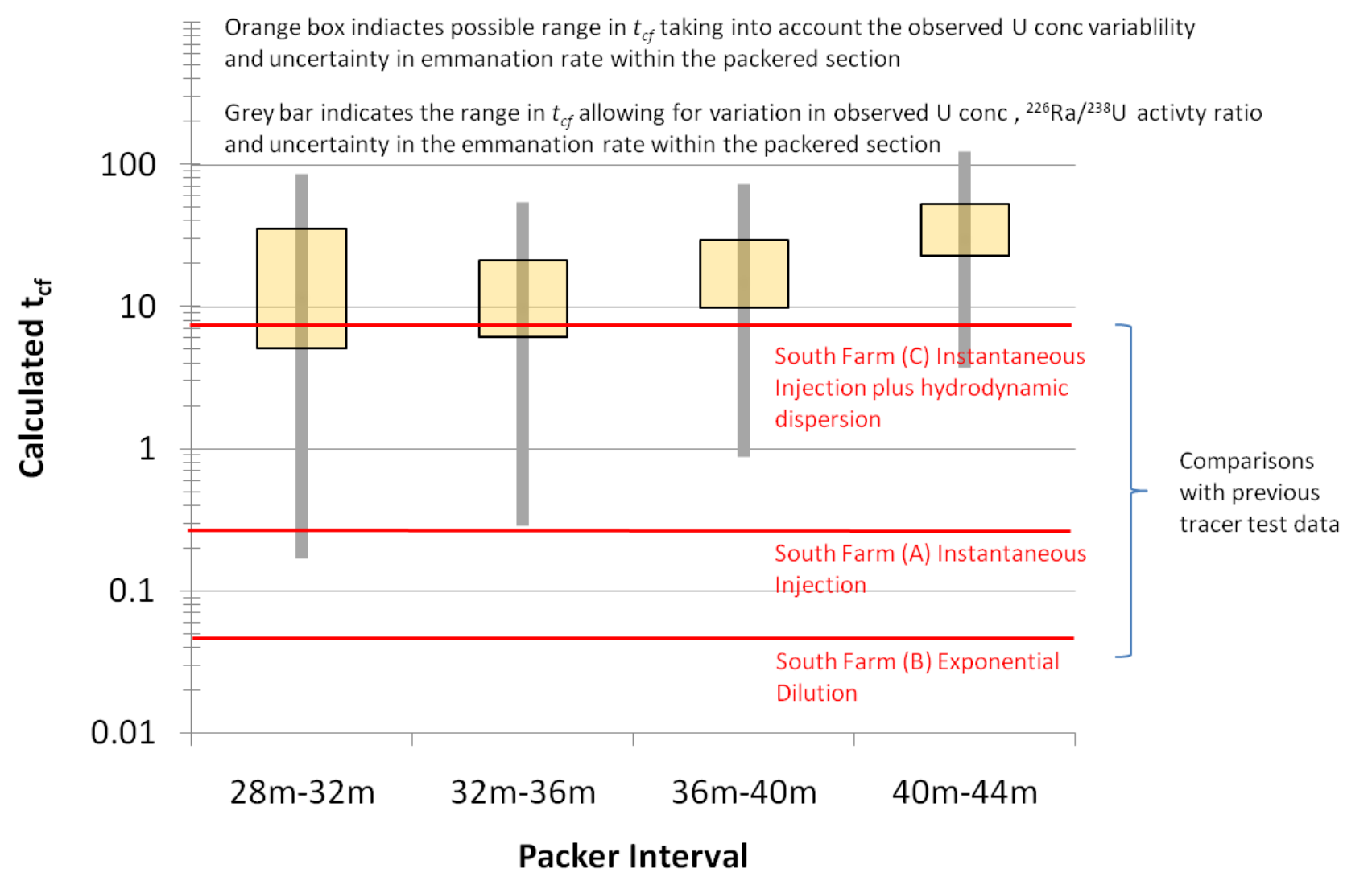
Figure 5 Radon Emanation Factors determined by lab experiment from Trumplett's Farm core material



Comparisons with other Tracer Tests

The expected value for the characteristic diffusion time t_{cf} on the basis of determined Rn and U contents of groundwater and Chalk can be determined for each set of paired measurements. Results from the analysis of core from the Trumplett's Farm site in the Pang catchment and subsequent packer testing in the same borehole are presented in Figure 6. As is clear there is significant variation in the calculated diffusion time for each packered section.

Figure 6 Calculated values of t_{cf} using combined radon activity, uranium conc, ^{226}Ra/^{238}U activity ratio and emanation rate data from Trumplett's Farm A borehole



This is due primarily to the variation in both the U concentration, the ^{226}Ra/^{238}U activity ratio within each core section and the high degree of uncertainty in the radon emanation factor. Further work to investigate the extent of U-series disequilibrium in the Upper Chalk is planned as part of this project.

The average value of t_{cf} over the four sections is ~29 days. This value is much larger than those determined from other tracer-derived tests that have been undertaken in similar Chalk aquifers(6).

These data suggest that the model in its present stage may not be appropriate for rapid determination of representative transport parameters.

Conclusions

Our original hypothesis has been tested using a mix of laboratory and field experiments. It is clear that there is a general link between Chalk matrix uranium content and fracture radon activity, which is in part due to the aquifer geometry. However, being simple, our model as it currently stands embodies several assumptions that limit its applicability for routine survey work.

At present the data requirements necessary to reduce the large uncertainty in the source term (which would then permit us to determine the contaminant attenuation properties of the aquifer) mean that this type of investigation is currently suited to more detailed work at selected sites.

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Groundwater Tracing Unit

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