Aquifer Transport of Th, U, Ra, and Rn in Solution and on Colloids

- G. J. Wasserburg (PI)
- D. Porcelli (Co-I)
- A. Tricca

California Institute of Technology

- M. Baskaran

Texas A &M at Galveston

1. Introduction

Aims of the Study-

- To understand the **migration** of naturally-occurring **U, Th, Ra, and Rn** isotopes in the subsurface;
- To use these nuclides as **analogues for waste nuclides** and predict migration rates of waste nuclides in potentially impacted aquifers.

This project includes-

- Measuring the evolution of groundwater nuclide concentrations **along aquifer flow lines**;
- Determining nuclide **input rates** along flow paths.
- Determining nuclide **sorption characteristics** on aquifer materials.

Outline

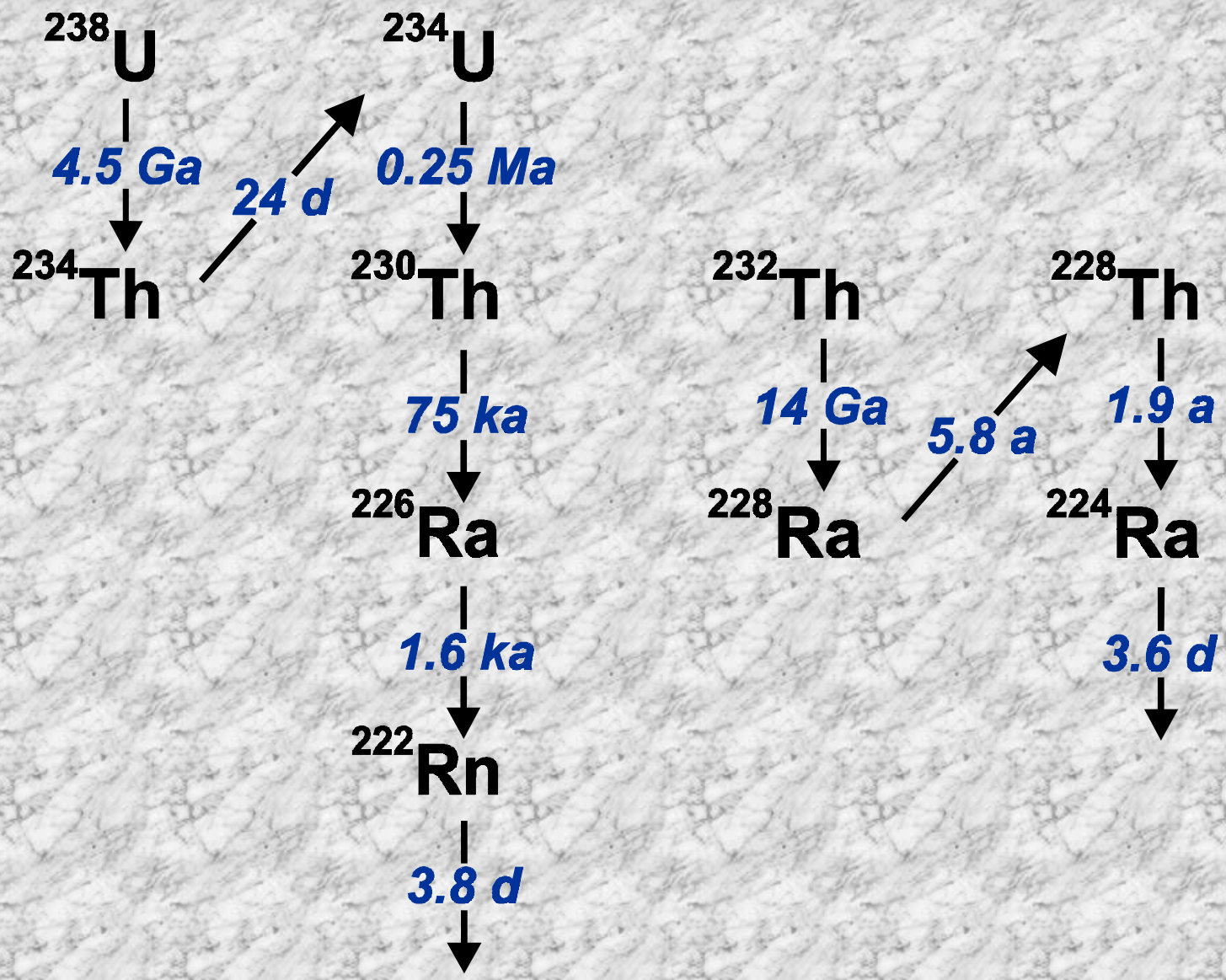
- Section 1: Modeling
  - Develop a nuclide migration model that includes likely input and removal processes
- Section 2: Aquifer Study
  - Sample groundwaters along flow lines at well-characterized sites
  - Measure U, Th series nuclide concentration profiles
  - Compare aquifer profiles with model results to identify likely controls on nuclide transport
  - Evaluate the importance of colloids in nuclide transport
- Section 3: Future Work

^{238}U and ^{232} Decay Series Nuclides

The ^{238}U and ^{232}Th decay series include U, Th, Ra, and Rn isotopes with a wide range of half-lives. This provides

- a basis for determining input rates of each nuclide by decay
- a connection between the distribution of nuclides of different elements

The ^{238}U and ^{232}Th Series



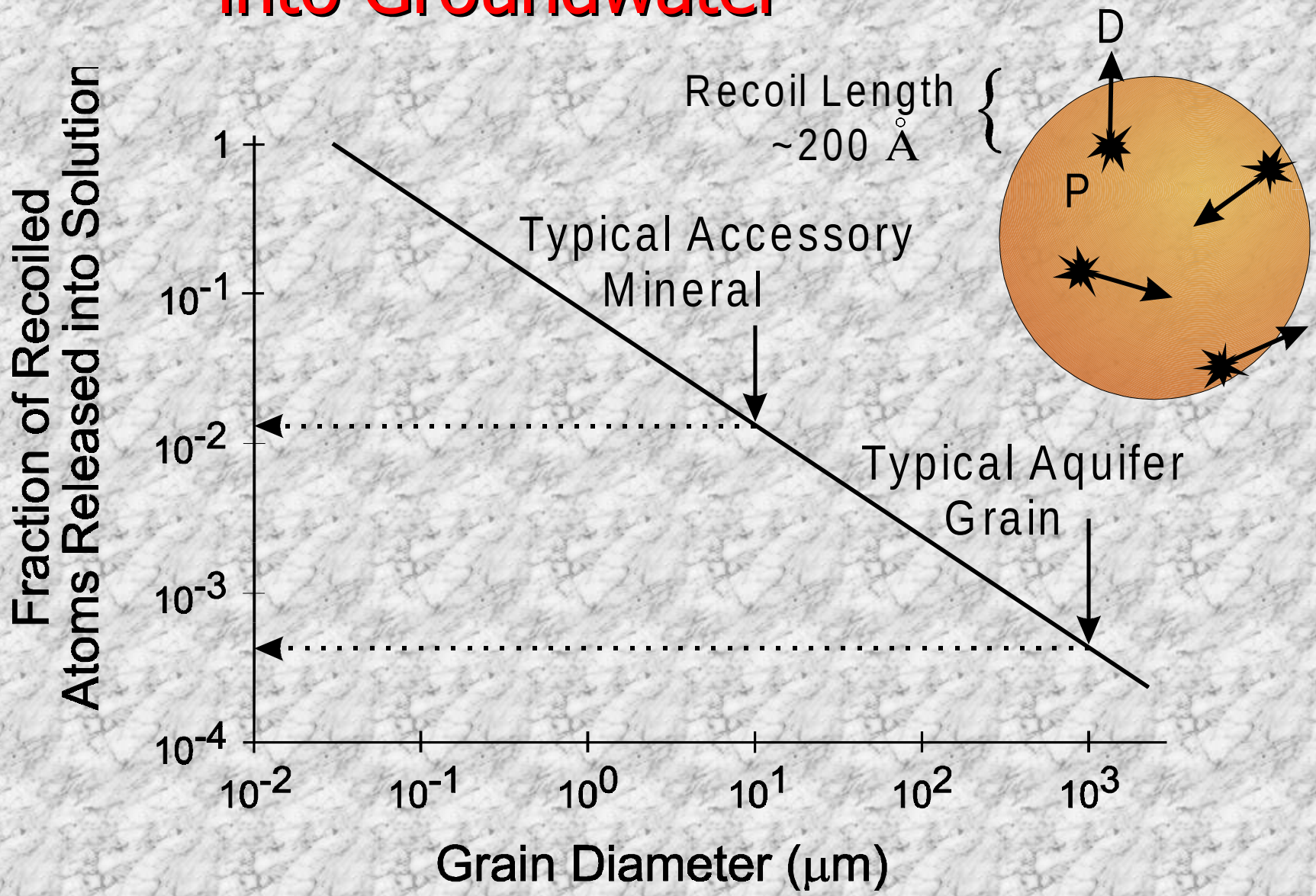
2. Radionuclide Transport Model

To obtain the variation in concentrations of each radionuclide i along a flow line the model considers:

- **3 Phases**
  - **Aquifer Mineral Grains** subject to losses by weathering and recoil of daughter nuclides
  - An **Active Surface Layer** for nuclide sorption
  - **Groundwater**

- **Removal** of nuclides from groundwater by:
  - **Decay** in solution
  - **Sorption** onto the surface layer
- **Input** of nuclides into groundwater by:
  - **Weathering** of mineral grains
  - **Recoil** from decay of parent in mineral grains
  - **Desorption** from the surface layer
  - **Recoil** from decay of parent in surface layer
  - **Decay** of nuclide in solution
  -

Recoil of Nuclides from Mineral Grains into Groundwater



The General Model Equations

- The **General Groundwater Transport Eqn.** is

Weathering *Recoil* *Desorption* *Decay of P*

$$\left(\frac{d\,{}^{i}C_{GW}}{dx}\right)\cdot v = \frac{\rho_r \cdot {}^{i}W \cdot {}^{i}C_r}{\rho_{GW}\cdot e} + \frac{\rho_r \cdot {}^{i}\varepsilon \cdot \lambda_p \cdot {}^{p}C_r}{\rho_{GW}\cdot e} + \frac{\rho_{SL}\cdot {}^{i}\hat{k}_{-1}\cdot S}{\rho_r \cdot e}\cdot {}^{i}C_{SL} + \lambda_p \cdot {}^{p}C_{GW}$$

Decay of P in SL *Decay of i* *Sorption*

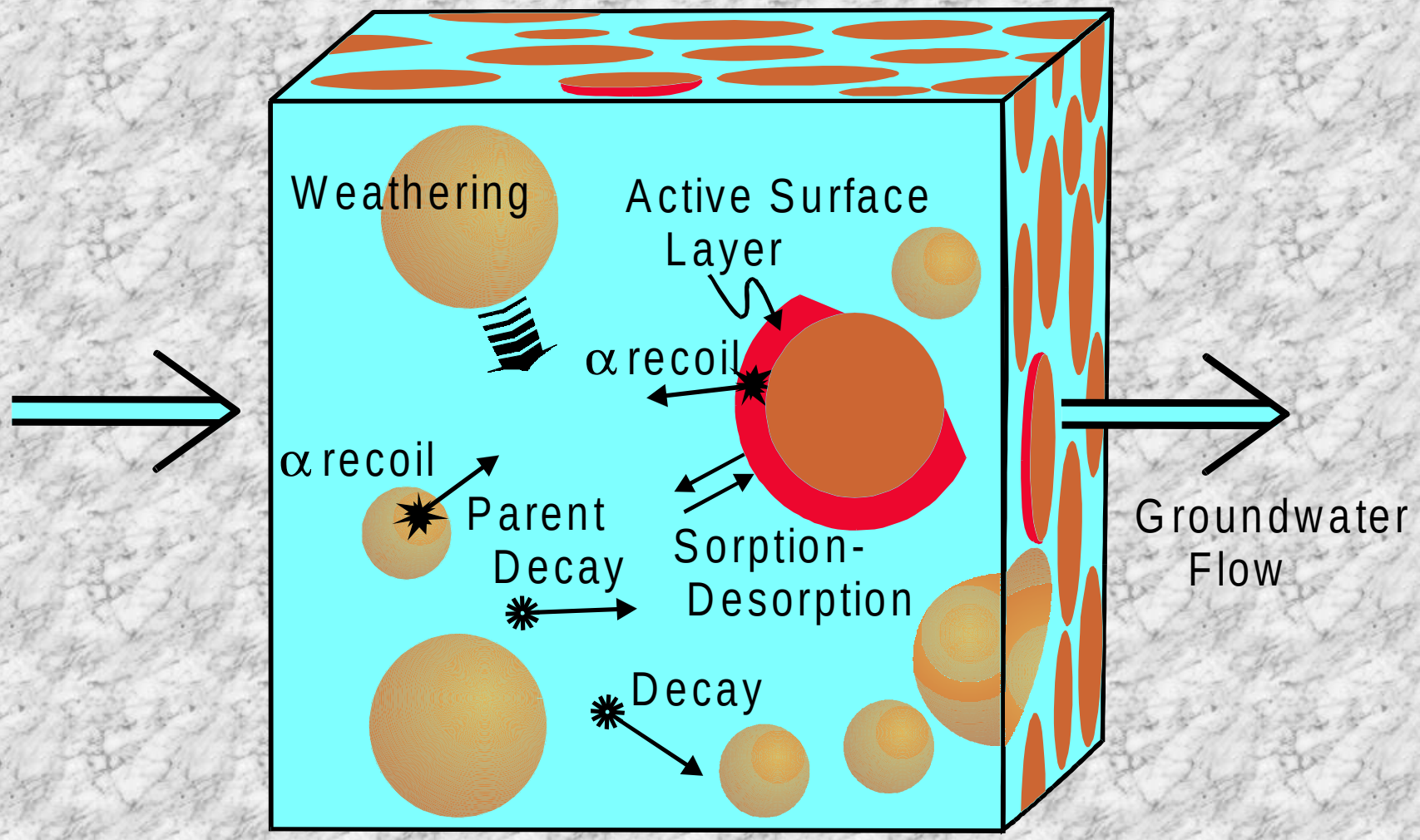
$$+\frac{\rho_{SL}\cdot f_i \cdot S \cdot \xi \cdot \lambda_p}{\rho_{GW}\cdot e}\cdot {}^{p}C_{SL} - \lambda_i \cdot {}^{i}C_{GW} - \frac{{}^{i}\hat{k}_1 \cdot S}{e}\cdot {}^{i}C_{GW}$$

- The **General Surface Layer Eqn.** is:

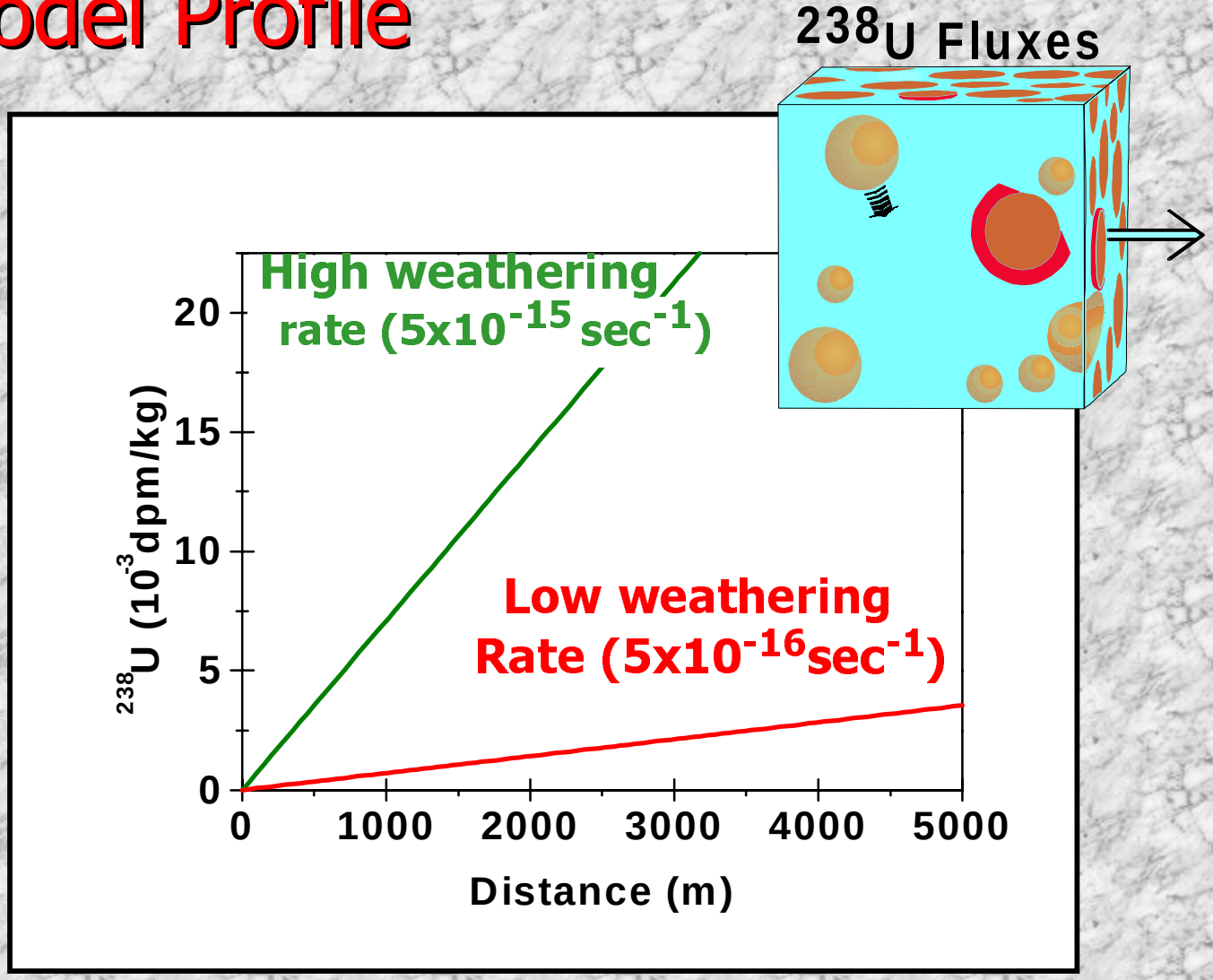
$${}^{i}C_{SL} = \frac{\rho_{GW}}{\rho_{SL}}\left(\frac{{}^{i}\hat{k}_1}{{}^{i}\hat{k}_{-1} + \lambda_i \cdot \xi}\right){}^{i}C_{GW} + (1 - f_i)\lambda_P\left(\frac{\xi}{{}^{i}\hat{k}_{-1} + \lambda_i \cdot \xi}\right){}^{P}C_{SL}$$

P-Parent Nuclide	${}^{i}W$-weathering rate(sec^{-1})	S-specific surface layer area
ρ_j-density of j	${}^{i}\varepsilon$-emanation fraction	ξ-surface layer thickness
v-water velocity	f_p-release during decay from SL	${}^{i}\hat{k}_1$- sorption rate constant (cm/sec)
e-void ratio	${}^{i}C_j$-concentration of i in j	${}^{i}\hat{k}_{-1}$- desorption rate constant (cm/sec)

Radionuclide Fluxes

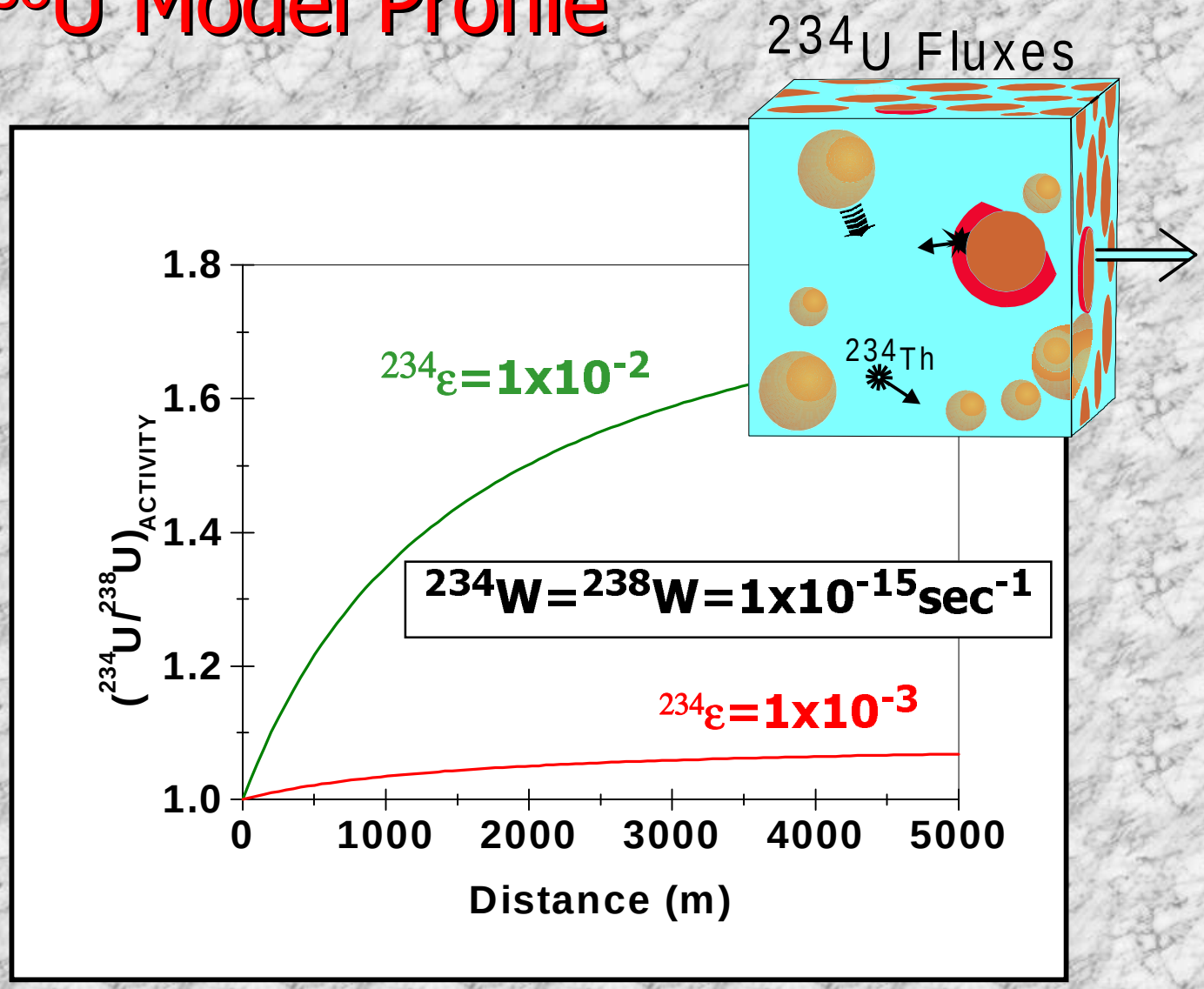


^{238}U Model Profile



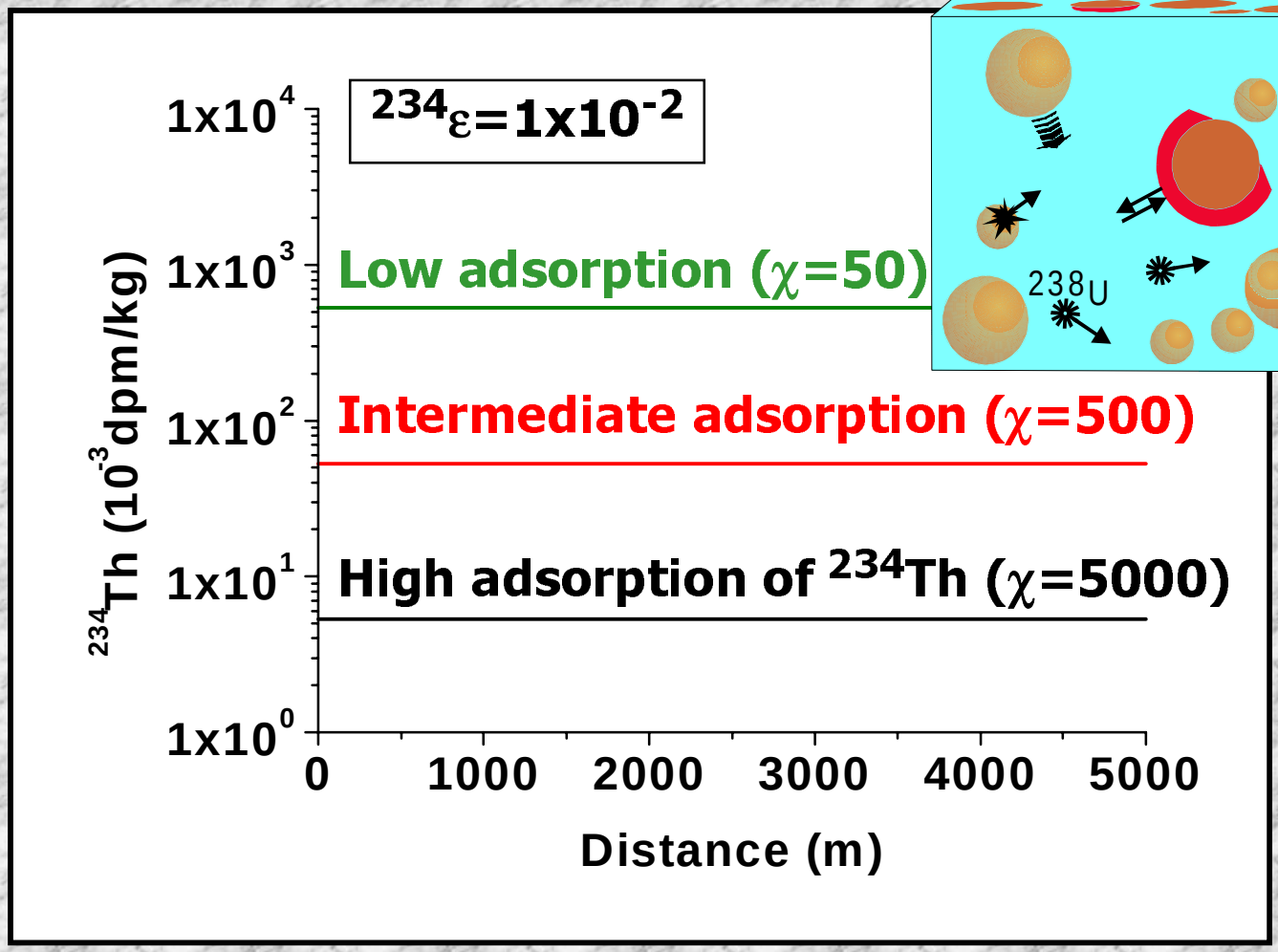
^{238}U growth is linear and **is controlled by ^{238}W**- the weathering rate constant. U is assumed to not adsorb.

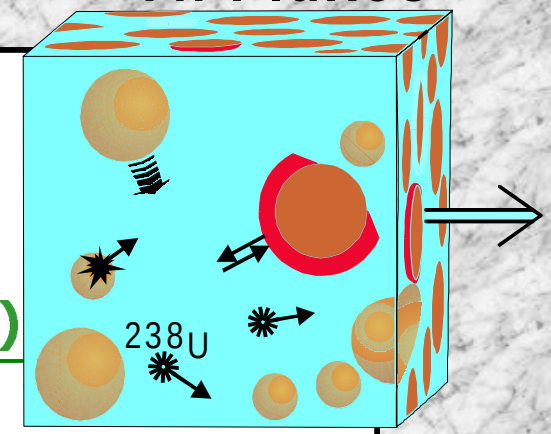
$^{234}U/^{238}U$ Model Profile



$\delta^{234}U$ increases and for a given value of ^{238}W **is controlled by $^{238}\varepsilon$, the recoil fraction**.

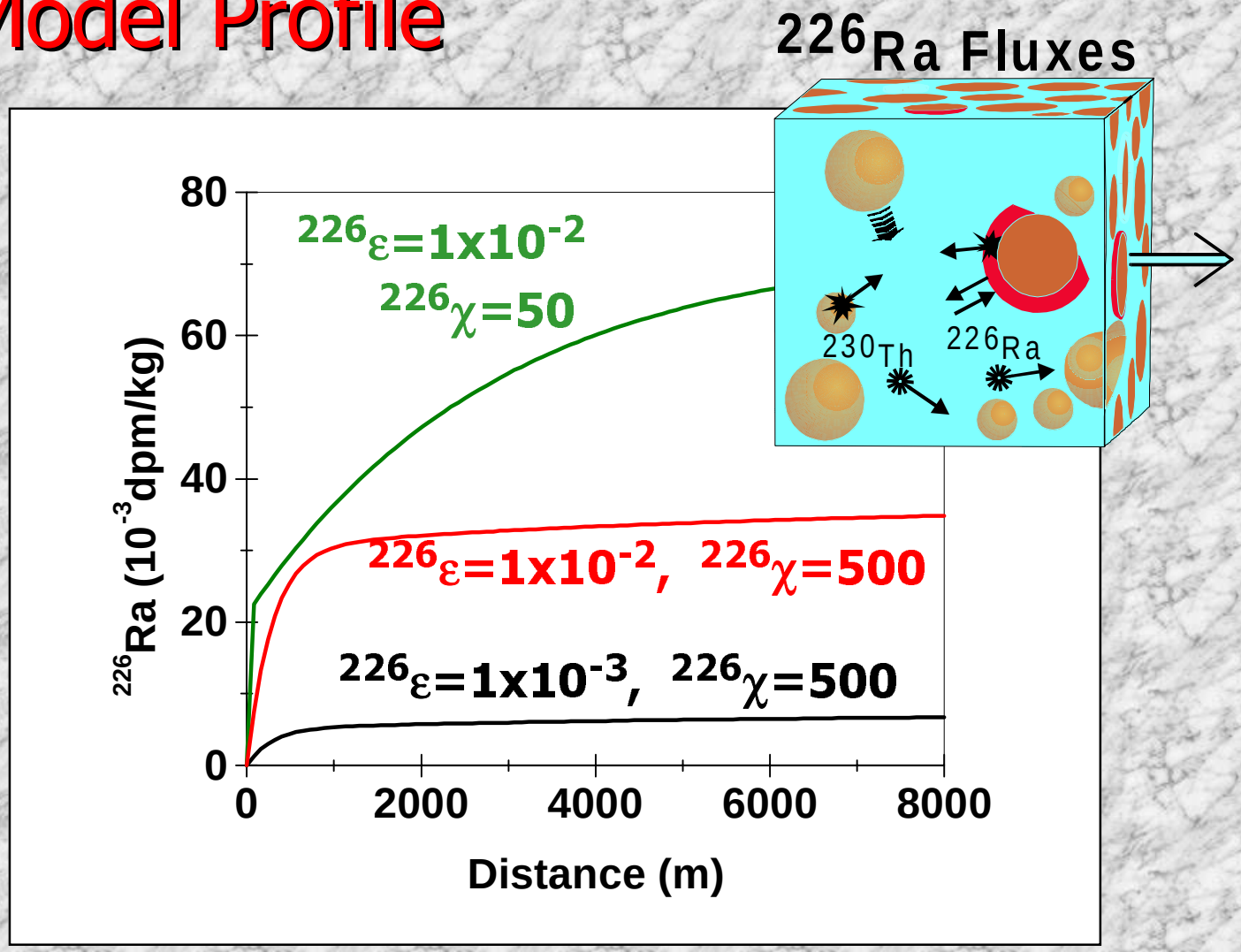
^{234}Th Model Profile





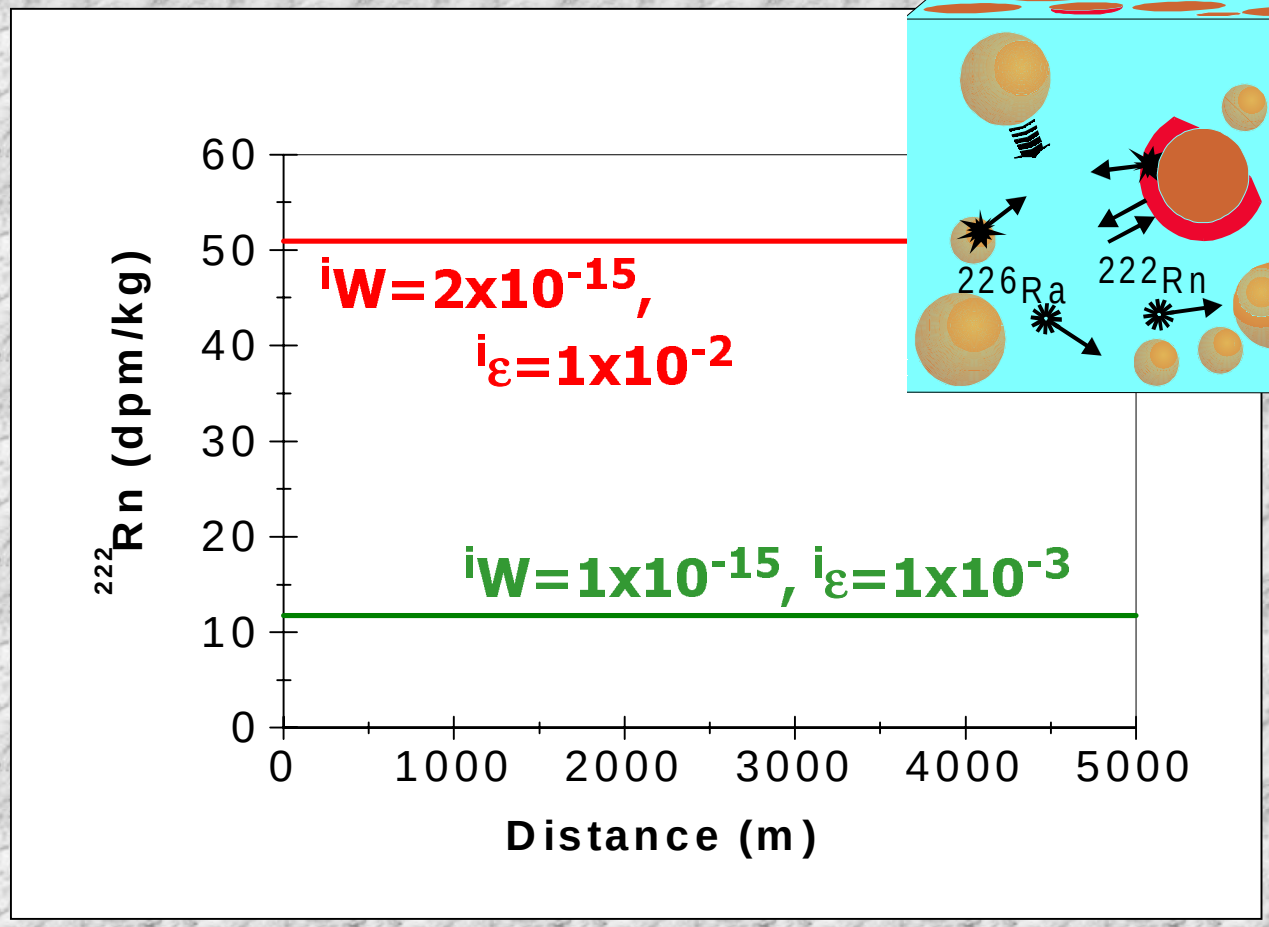
^{234}Th is constant, and for a given value of $^{234}\varepsilon$ **is controlled by χ (atoms on the surface layer/atoms in solution)**.

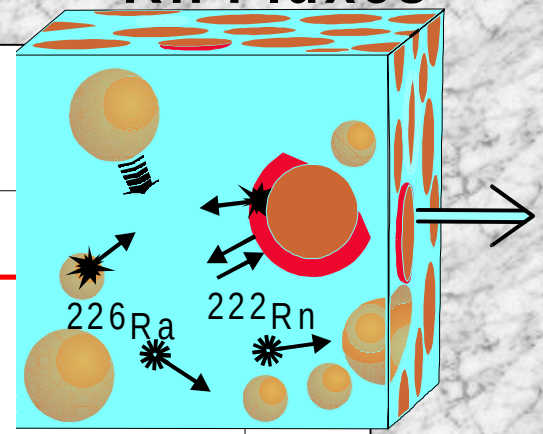
^{226}Ra Model Profile



^{226}Ra increases to a constant value, and **is controlled by $^{226}\varepsilon$ (the recoil fraction) and $^{226}\chi$ (the atoms on the surface layer/atoms in solution)**

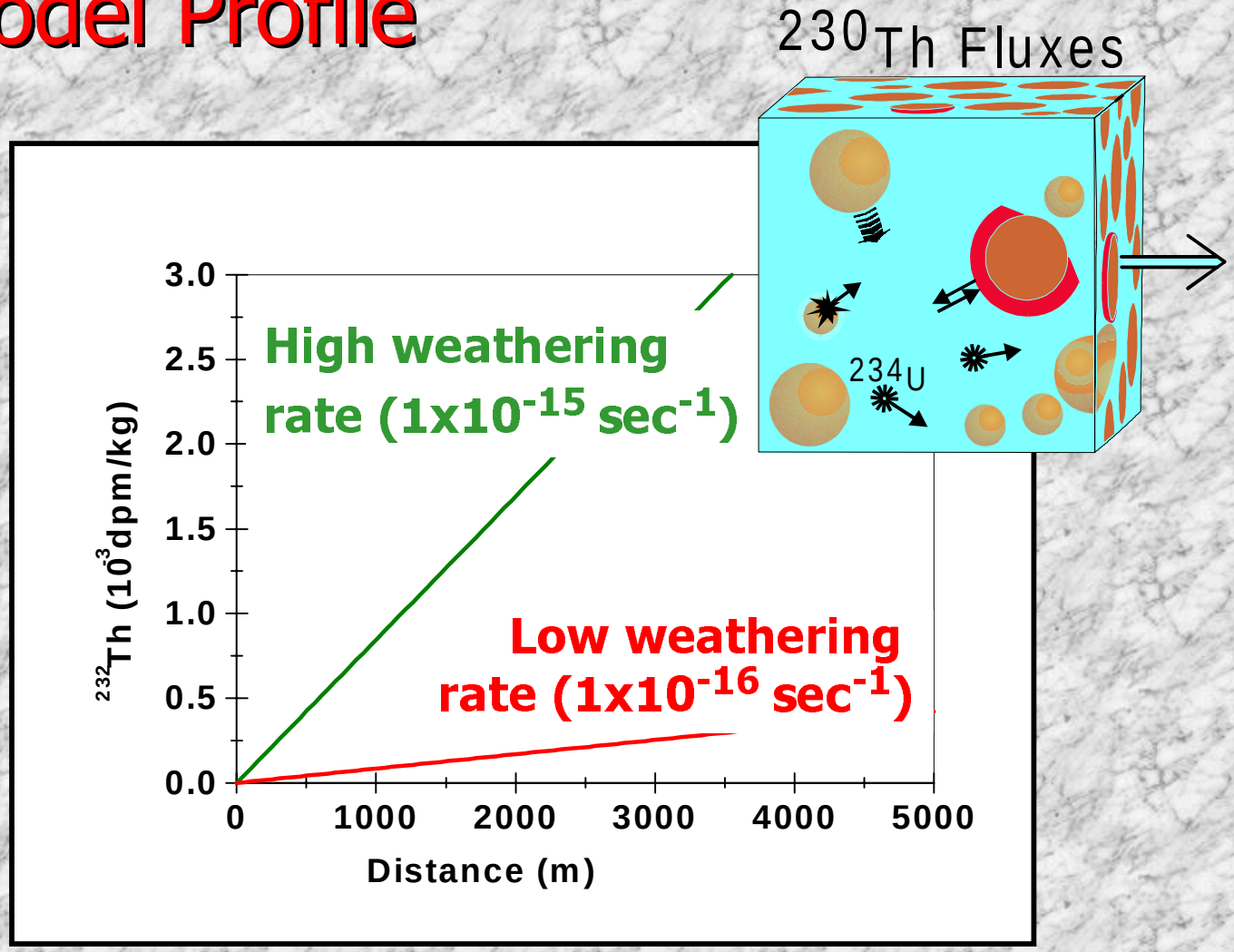
^{222}Rn Model Profile





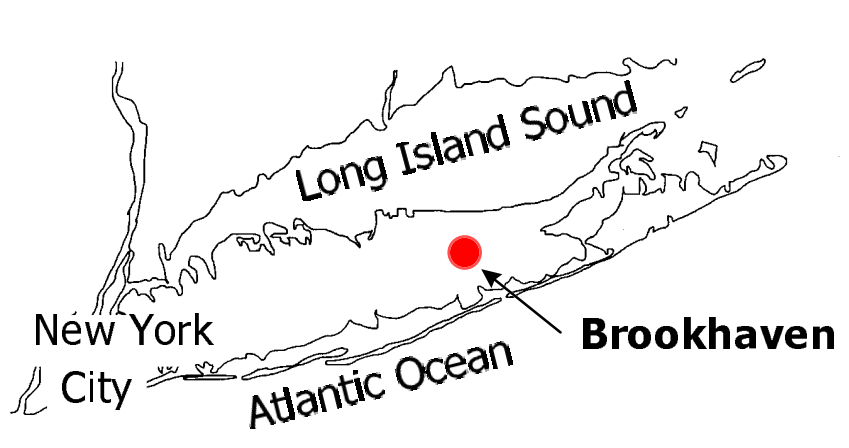
Rn is constant and largely reflects the supply of ^{230}Th and ^{226}Ra to aquifer surfaces by weathering, as well as the emanation fraction of ^{222}Rn and the other nuclides.

^{232}Th Model Profile



^{232}Th growth is linear and **is controlled by ^{232}W**- the weathering rate constant.

3. Study Site: Brookhaven National Laboratory

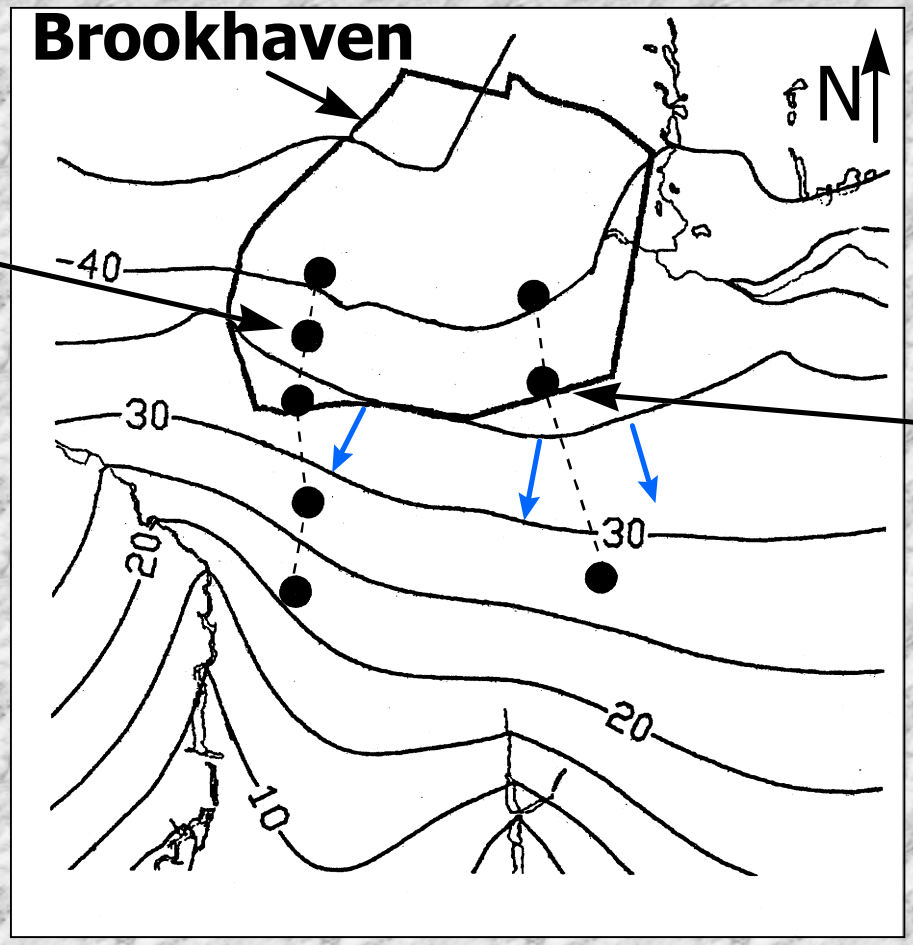


Upper unconfined aquifer at BNL chosen for study:

- Unconsolidated sandy aquifer used extensively for water supply
- Well-characterized hydrology
- Extensive groundwater monitoring network

BNL Groundwater Contour Map

Upper Glacial Unconfined Aquifer



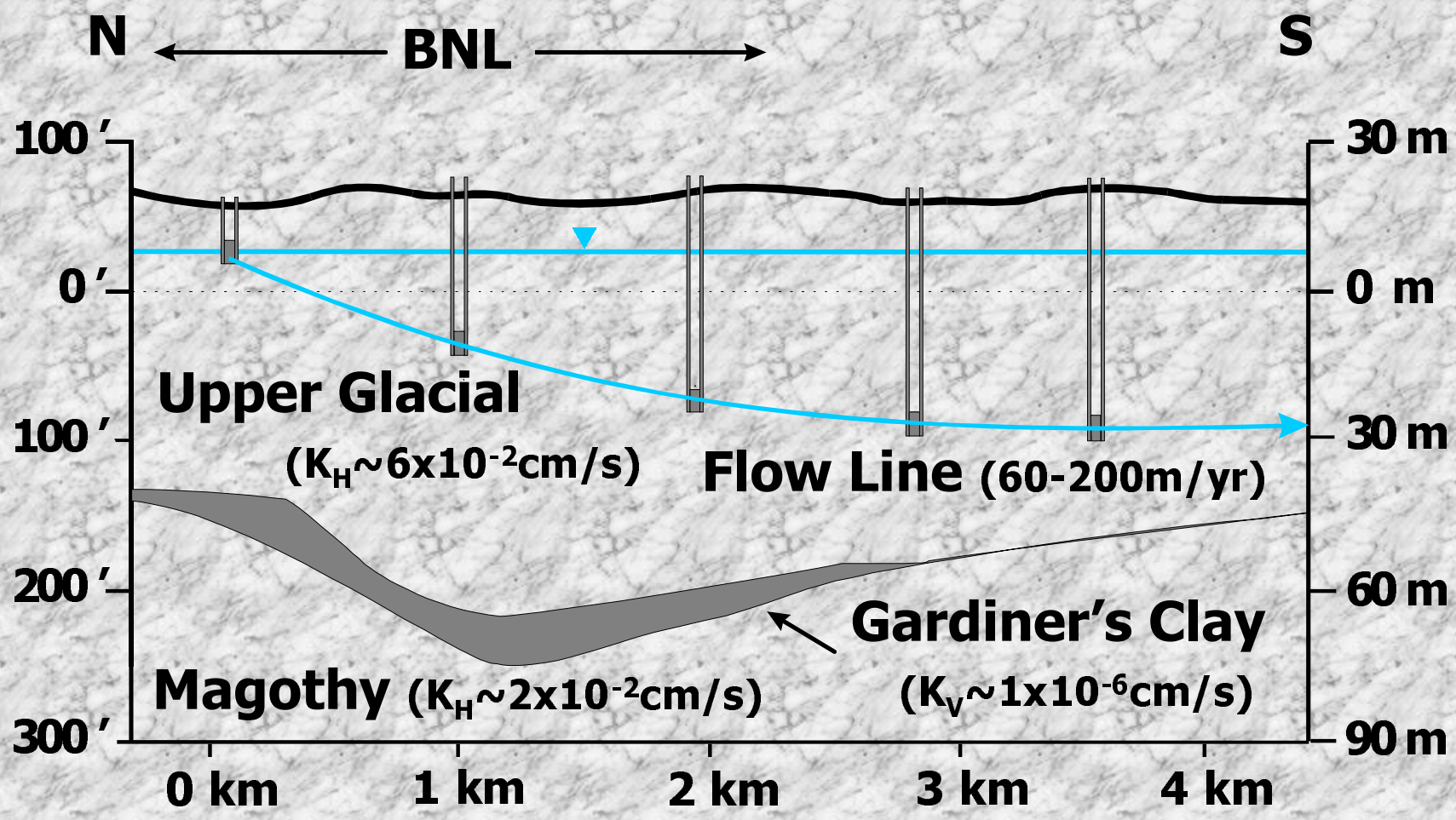
Western Line of Monitor Wells

Eastern Line of Monitor Wells

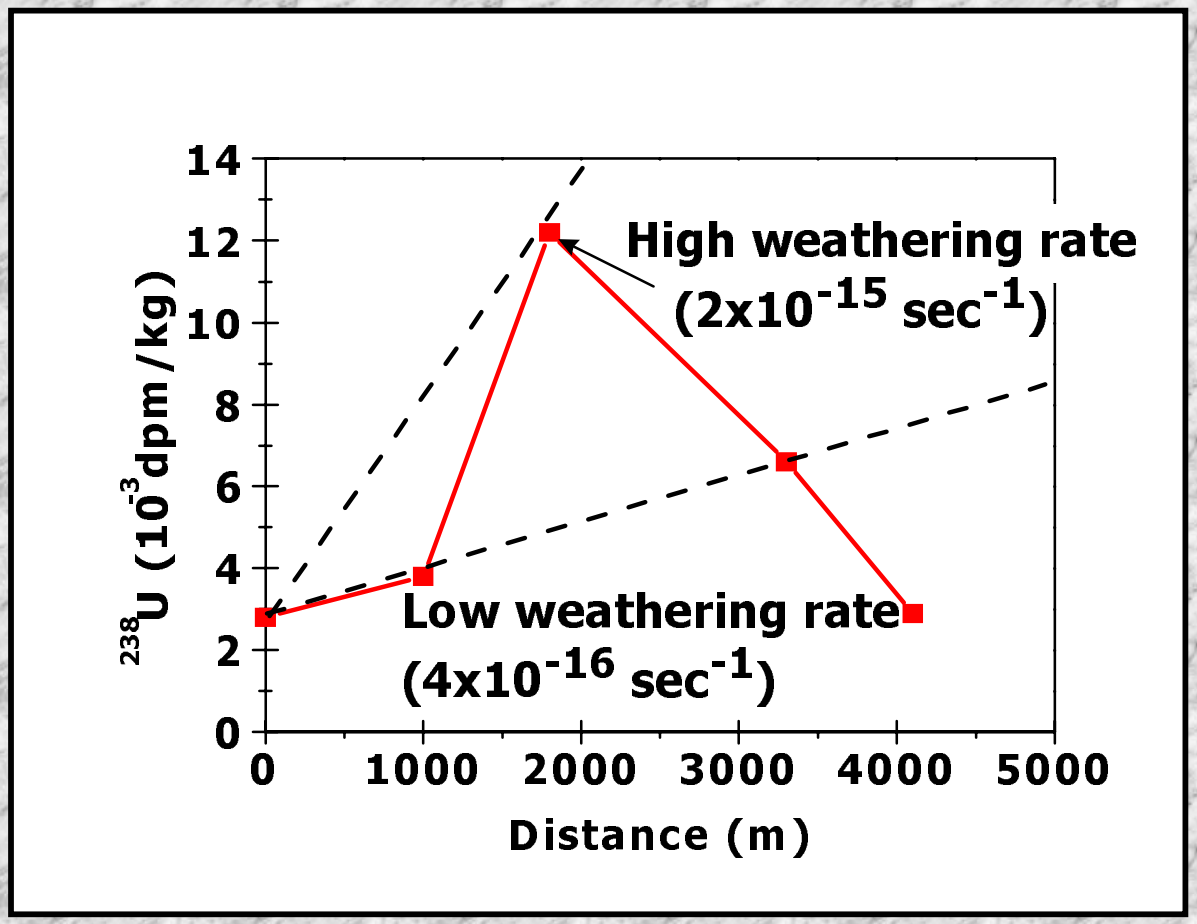
Contours in feet above mean sealevel

3 km

BNL Cross Section

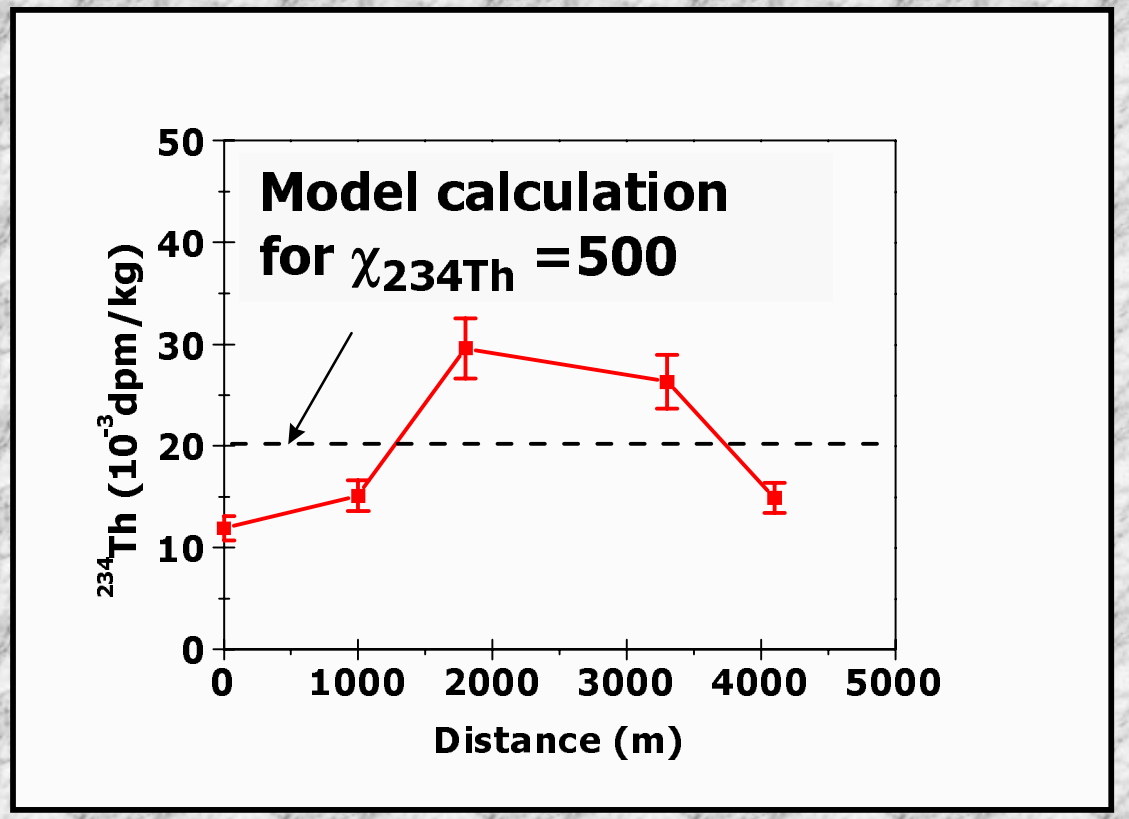


^{238}U at Brookhaven



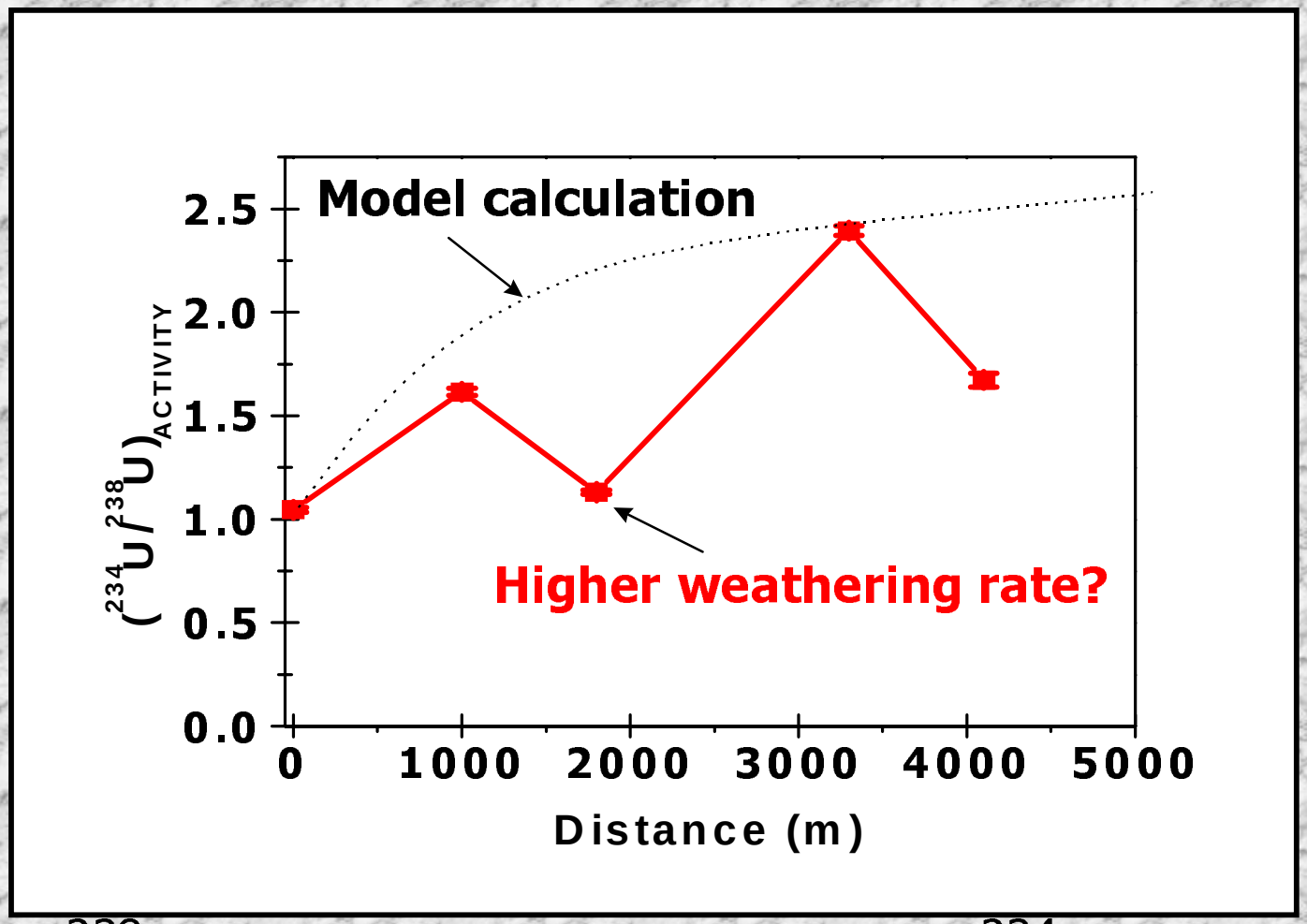
- Data is generally within model values for reasonable ^{238}W.
- Non linear concentration profile indicates either samples not from one flow line with constant ^{238}W or U is not conservative.
- The vadose zone is an important source of U to the water table.

^{234}Th at Brookhaven



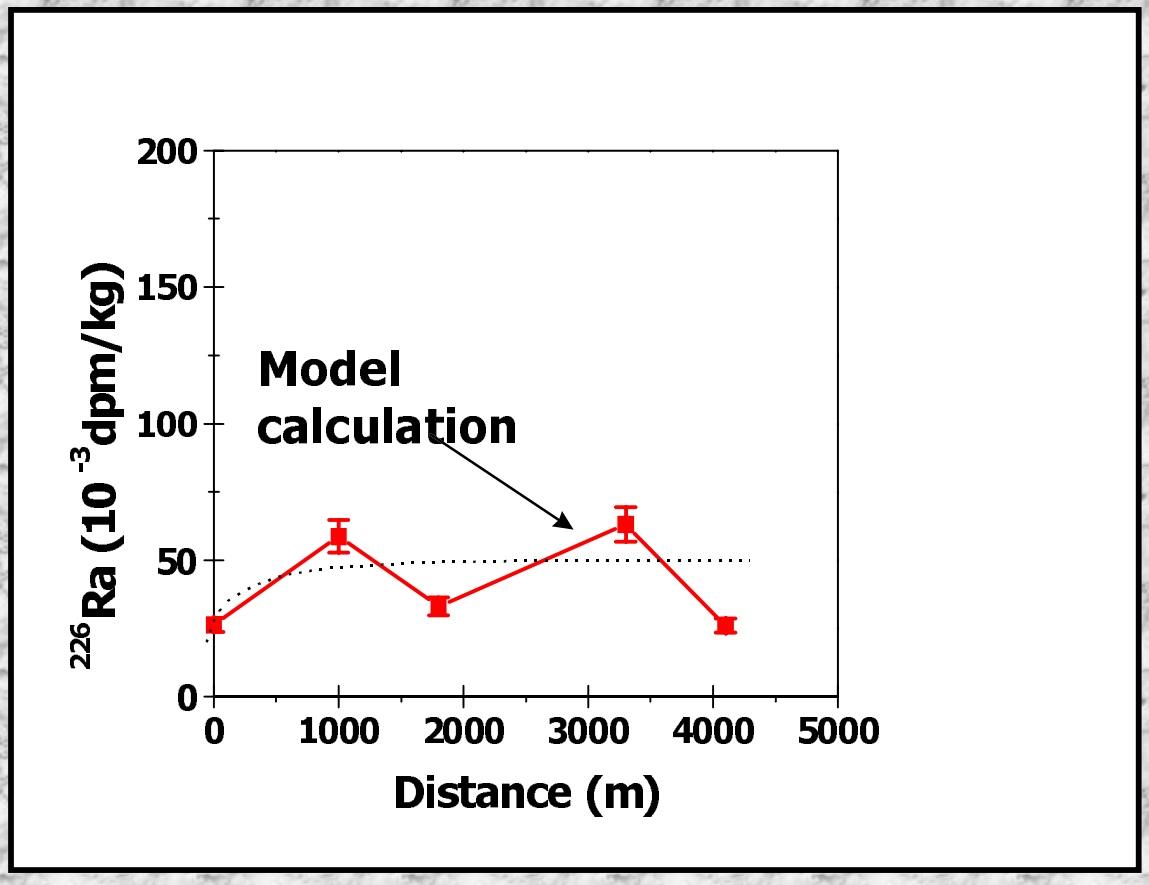
The ^{234}Th concentrations are similar along the flowline and correspond to a model value for χ (=atoms in solution/atoms on surfaces) of 500.

$^{234}U/^{238}U$ at Brookhaven



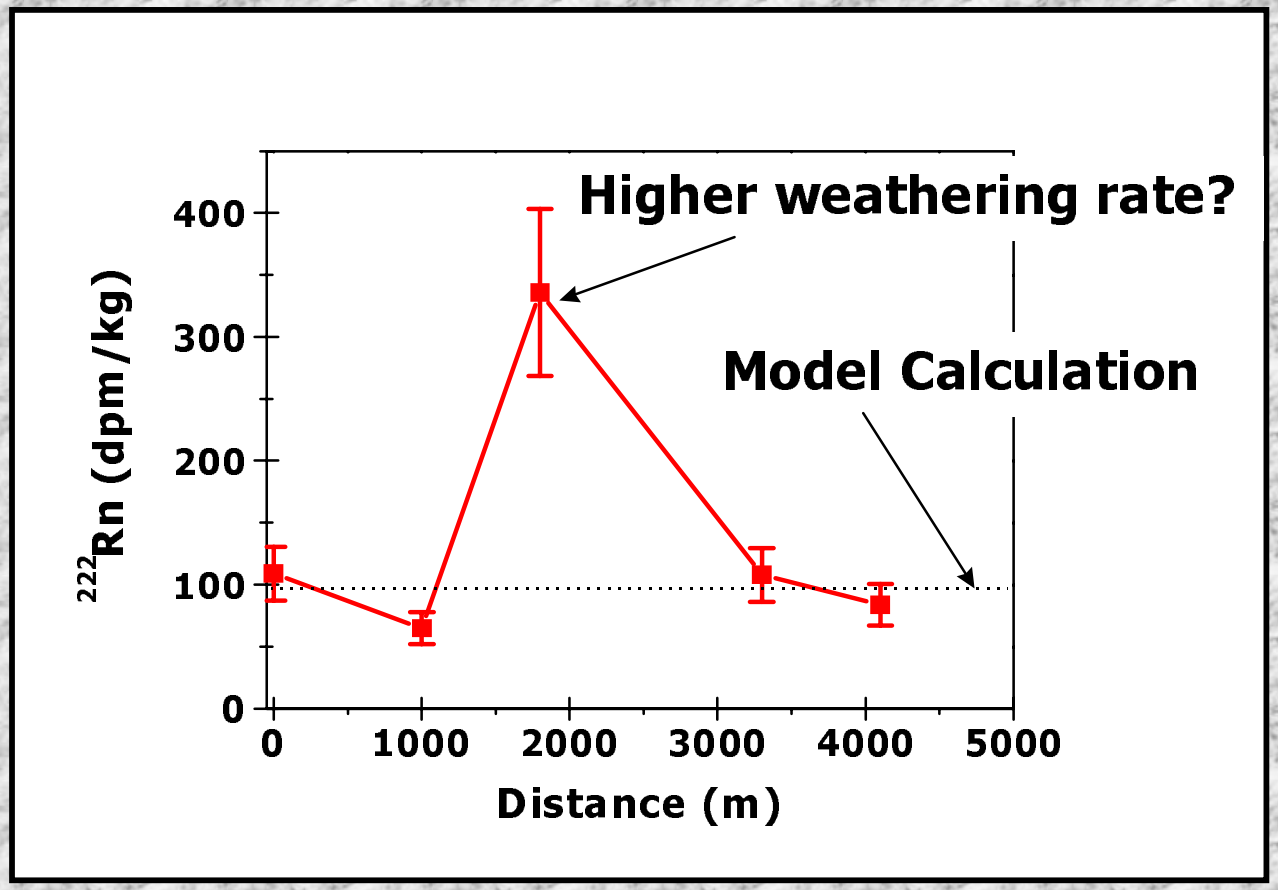
The $^{234}U/^{238}U$ ratios correspond to a model ^{234}Th emanation coefficient of $5x10^{-3}$. This implies that U is concentrated in small grains. The lower ratio may reflect a higher weathering rate.

^{226}Ra at Brookhaven



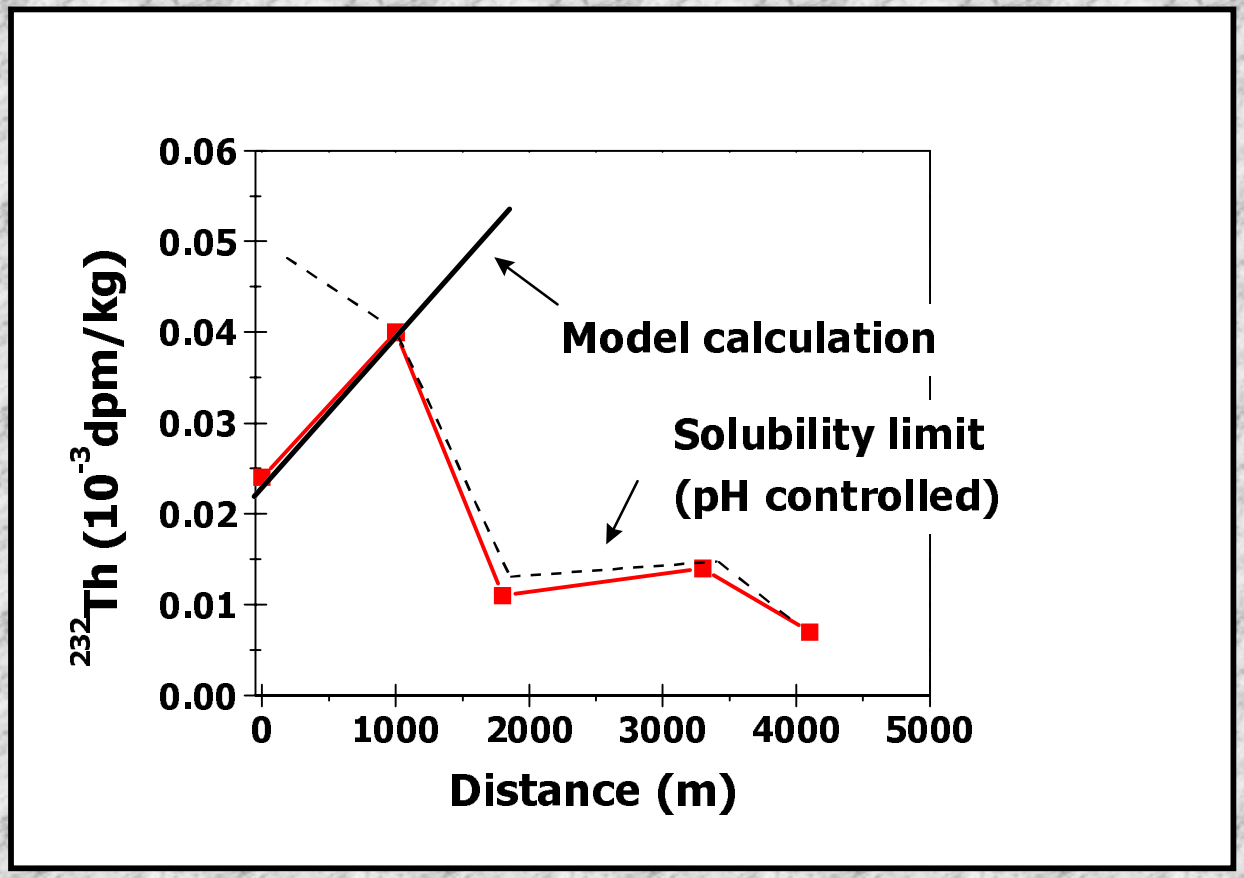
^{226}Ra concentrations are similar along a flowline and correspond to the model calculation with a χ_{226Ra} (atoms in solution/atoms on surfaces) value of 300

^{222}Rn at Brookhaven



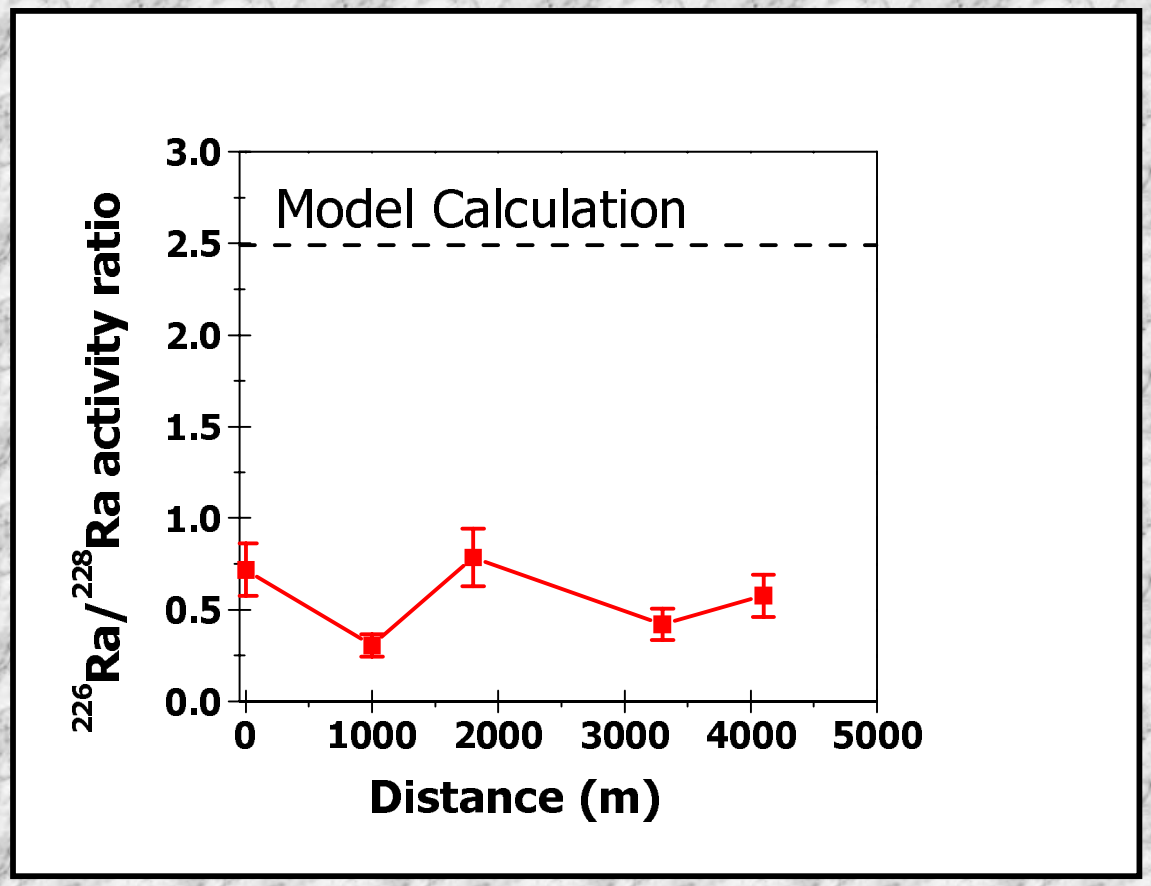
^{222}Rn concentrations are generally compatible with the model calculations. The highest ^{222}Rn value may be due to a higher weathering rate supplying ^{230}Th and ^{226}Ra to aquifer surfaces.

^{232}Th at Brookhaven



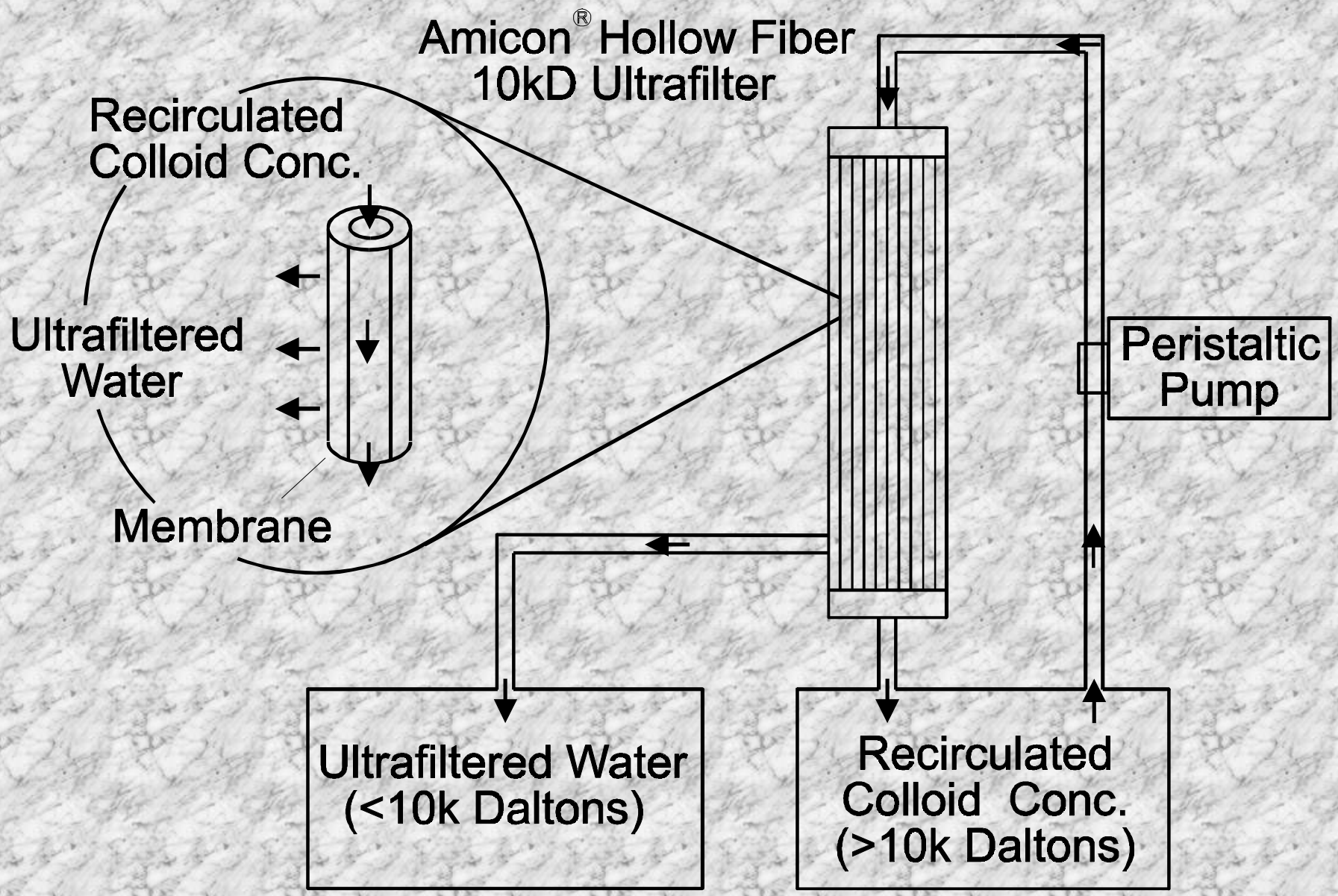
^{232}Th concentrations do not fall on a model profile, but appear to be controlled by solubility limits.

$^{226}Ra/^{228}Ra$ at Brookhaven

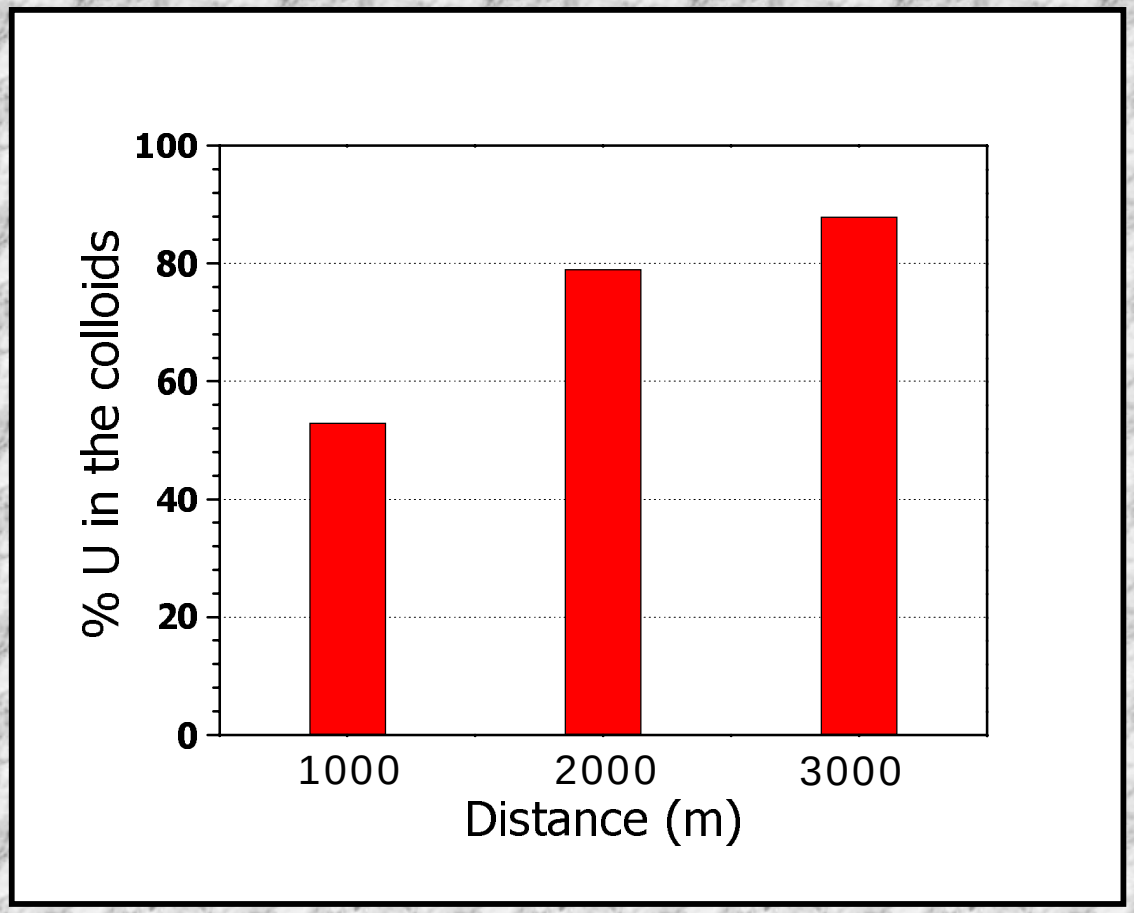


$^{226}Ra/^{228}Ra$ activity ratios are similar along a flow line, but much lower than the model calculation. This may be due to a higher production of ^{228}Ra from decay of ^{232}Th precipitated on surfaces.

Colloid Separation by Ultrafiltration



^{238}U on colloids



- A dominant fraction of U (50 to 88 %) is associated with colloids.

Conclusions

- The aquifer transport model
  - utilizes the full solutions for transport equations of U and Th series nuclides along a flow line
  - provides a framework for identifying the processes and parameter values that may be controlling aquifer nuclide concentrations.

- For BNL
  - the model provides reasonable values for average recoil and weathering rates as well as for partitioning of Th and Ra onto surfaces
  - the processes causing some variations in nuclide concentrations require further evaluation
  - water table nuclide concentrations indicate that the vadose zone is an important source of U, Th, and Ra.
  - Colloids are important for nuclide transport at BNL and require further investigation.
  -

4. Future Work

- Role of Colloids
  - Measurements of groundwater colloid Th and Ra concentrations at BNL.
  - Explicit modeling of colloid nuclide transport.
- Nuclide Supply Rates
  - Measurements of nuclides in BNL aquifer grains.
  - Lab experiments of nuclide weathering and recoil release rates from different minerals.
  - Comparison of lab results with aquifer rates inferred from model calculations and groundwater measurements.

- Nuclide Adsorption Processes
  - Measurements of nuclides on BNL aquifer surfaces
  - Lab experiments of isotopic exchange rates with waters
- Vadose Zone Transport
  - Analyses of vadose waters and nuclide transport modeling of the sandy unsaturated zone at BNL
- Groundwater Transport Modeling
  - Inclusion of U adsorption, solubility limitations, and changes in water chemistry in transport models.
- Other Aquifer Conditions
  - Investigation of oxic and anoxic waters in the Edwards limestone aquifer.

- Further Work at BNL
  - Characterization of variability of groundwaters at water table and between flow lines
  - Relating results to available information regarding on-site contaminants
  - Evaluating possible effects of inflows to the aquifer from underlying deposits
  -