U and Th radionuclides in a sandy aquifer

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Objectives:

To understand the transport of natural radionuclides in ground water

Measurements of full decay series provide information on water-rock interactions

We used a two-sided approach to understand the mechanisms:

- **experimental: data collected from the Long Island Aquifer**
- **theoretical approach: theoretical model of activities along a flow line**

The sampling field

The aquifer at Brookhaven National Laboratory

The unconfined Upper Glacial aquifer at Brookhaven National Laboratory was chosen for study:

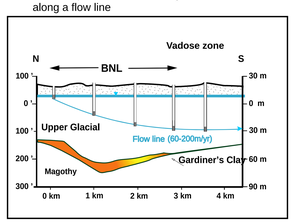
it is an unconsolidated sandy aquifer;

it has a well-characterized hydrology;

it has an extensive groundwater monitoring network

The cross section beneath shows

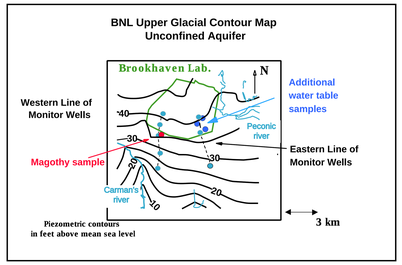
- The vertical sequence including the vadose zone, Upper Glacial and Magothy Aquifers. Note that both aquifers are separated by a layer of clay minerals, the Gardiner's Clay
- The water table and a flow line
- An example of well lines designed to collect water along a flow line

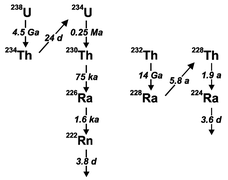


The samples

Samples from shallow and deeper wells have been collected along two well lines (Western and Eastern)

A sample from the confined Magothy aquifer has also been collected





In a closed system, all nuclides of the same series have the same activities

The decay series include isotopes with a wide range of half-lives that reflect **different time-scales**

different elements with different **chemical properties**

nuclides of different elements that are **connected by decay reactions**

The Model

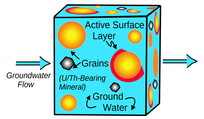
Assumptions of the model

The theoretical model used to interpret the data considers one dimensional flow along a flow line without dispersion.

The initial activity at the water table is inherited from interaction of the water with the vadose zone

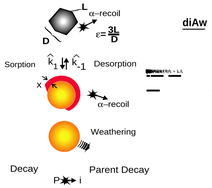
The reactions and the resulting equations for the model aquifer are shown below.

3 Phases



Groundwater

Outputs **Inputs**



Transport Equations

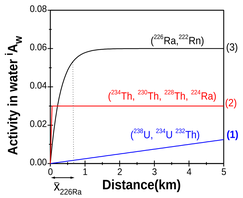
diAw =

diAw

Solutions to the model equations

3 patterns may occur depending on the half-life and the sorption properties of the nuclide

- linear evolution with distance (^{238}U, ^{234}U, ^{232}Th) if the removal time-scaleof the nuclide or its parent (half-life and adsorption time-scale) is greater than ground water age (age = distance/ velocity)
- constant activity (^{234}Th, ^{230}Th, ^{228}Th, ^{224}Ra), if the removal time-scale is very short compared to ground water age (age = distance/ velocity)
- exponential evolution towards $^{i}A_{winf}$ (^{226}Ra, ^{222}Rn) if the removal time-scale of the nuclide or its parent is comparable to ground water age (age =distance/ velocity)



Results

Uranium in the vadose zone

The U activity is variable in the vadose zone and indicates high weathering rates.

The $^{234}U/^{238}U$ activity ratios are also variable and proportional to $\varepsilon_{234Th}/W_{238U}$

U in the vadose zone is the dominant input for the U in the aquifer.

Uranium in the aquifer

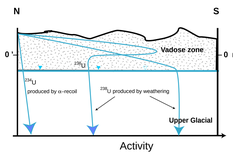
U activity

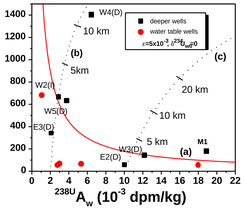
- ^{238}U activities indicate a small added contribution from the aquifer (low weathering rates)
- ^{238}U and ^{234}U activities are variable and reflect input from the vadose zone
- the wide variations of ^{238}U activities accross the site indicate that dispersive mixing is not important
- there is no evidence of U adsorption (Th/U ratio in the water ~10^{-3}crustal ratio)

U isotopic composition

- the U isotopic composition reflects the ratio recoil/weathering input rates ($\varepsilon_{234Th}/W_{238U}$)
- the increase in $^{234}U/^{238}U$ shows evidence of recoil contribution within the aquifer for waters with low U activity input from the vadose zone

Only high δ^{234}U and low ^{238}U activities or low δ^{234}U and high ^{238}U activities can be generated in the aquifer at flow distances < 100km. δ^{234}U and ^{238}U activities monitor the aquifer input over long distances





Thorium

^{232}Th activity is at the saturation limit of ThO_2, the ^{232}Th activities higher than the saturation limit are associated with colloids.

the Th/U ratio in the water ~10^{-3}crustal ratio, indicates that Th adsorbs strongly

The Th isotopic ratios reflect the ratios of the input rates: weathering for ^{232}Th and ^{228}Th, recoil for ^{230}Th and ^{234}Th

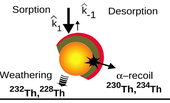
The ^{230}Th/^{232}Th atomic ratio is ~5×10^{-5} (10 times the crustal ratio) due to recoil

Th adsorbs quasi irreversibly onto the surface coating. The ratio of number of atoms in the surface coating to those in the water is ~10^{7}

The ^{234}Th activities indicate a residence time in the water is ~1h

The ^{228}Th/^{234}Th activity ratio is lower than unity and indicates slow desorption kinetics.

The residence time of Th in the surface layer ~6000 year



References for Brookhaven National Laboratory:

CopenhaverS.A., Krishnaswami S., Turekian K.K., Epler N., Cochran J.K. (1993)

de LagunaW. (1963)

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Tricca A., Wasserburg G.J., Porcelli D., Baskaran M. (2000)

Tricca A., Porcelli D., Wasserburg G.J., (2000)

Radon requires the presence of a Th-rich surface layer

^{222}Rn activities are 10^3 times higher than those of parent ^{226}Ra in the water.

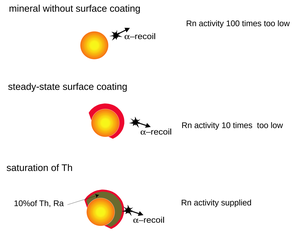
^{222}Rn activities in the water (100-300 dpm/kg) represent ~5% of the rock production by α–recoil.

Such high ^{222}Rn activities cannot be produced by a homogenously distributed aquifer solid of grain size >10 μm

The ^{226}Ra necessary to produce the ^{222}Rn measured in the water must be located on the surface coating or at grain boundaries

This cannot be attained by reversible precipitation of Ra; the alternative, irreversible precipitation of Ra, is not compatible with other Ra data

As Th is at saturation, we show that th is precipitated irreversibly. We infer that the surface coating in the aquifer contains ThO_2 (~10% of the Th rock inventory) deposited in earlier weathering cycles. This forms the major source for ^{222}Rn.



Radium

Ra sources

- Ra in the vadose zone is provided by weathering and α–recoil
- Ra in the aquifer is provided by α–recoil

The Ra nuclides are associated with the surface coating that is in local equilibrium with the water

The atom ratio of the surface coating to the water is ~10^4

The Magothy aquifer

The Magothy aquifer solid is similar to that of the Upper Glacial.

The water of the Magothy shows different characteristics:

1) the pH value is higher (6.77 against 4-5.5 for the Upper Glacial)
2) the water is less oxidizing
3) the solution contains very high amount of Fe-rich colloids

The chemical behaviour of the other radionuclides in the Magothy are the same as for the Upper Glacial

U activity is similar to that of the vadose zone, there is no evidence of important aquifer contribution

The low δ^{234}U value shows a recoil comparable to that in the Upper Glacial

The very high ^{232}Th activity (10^3 above the solubility limit) is associated with colloids (the Th/U ratio equals the ratio in the crustal rock)

Conclusions:

- Uranium

the data provide a monitor for water-rock interaction in the aquifer over long time-periods (>2000years)

the U activity reflects dominantly the vadose zone input

$^{234}U/^{238}U$ isotopic composition depends on the vadose zone input and on aquifer mineral grains input and travel time.

- Radon

^{222}Rn is a monitor for local processes on very short time-scale

the high activity levels reflect the importance of the surface coating

to provide the measured ^{222}Rn, a ^{232}Th and ^{230}Th-rich surface coating (containing ~10% of the rock inventory) must have been deposited during a previous weathering cycle

- Thorium

Th is at ThO_2 solubility limit in absence of colloids

^{234}Th and ^{228}Th activities provide a residence time in the water ~1h and in the surface coating ~ 6000 years

- Radium

Ra is strongly adsorbed on the mineral surface and is in exchange equilibrium with the water

^{224}Ra, ^{226}Ra, ^{228}Ra activities input occurs by α-recoil from the rock in the aquifer, by weathering in the vadose zone