Modelling of decay chain transport in groundwater from uranium tailings ponds

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Outline

- Introduction
- Model
- Result and discussion
- Conclusions
Introduction

- Uranium tailings ponds
- The principal radiation risk
Introduction

- Percentage of inventory of the world

Total volume: $938 \times 10^6 \text{ m}^3$
Introduction

The Uranium-238 Decay Chain

Atomic Number

82  83  84  85  86  87  88  89  90  91  92

Only main decays are shown
Gamma emitters are not indicated

Element Names
U - uranium
Th - thorium
Ra - radium
Pa - protactinium
Rn - radon
Po - polonium
Bi - bismuth
Pb - lead

Half-life units
a - years
d - days
h - hours
m - minutes
s - seconds

http://pubs.usgs.gov/
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>$4.51 \times 10^9$ years</td>
</tr>
<tr>
<td>U-234</td>
<td>$2.48 \times 10^5$ years</td>
</tr>
<tr>
<td>Th-230</td>
<td>$8.00 \times 10^4$ years</td>
</tr>
<tr>
<td>Ra-226</td>
<td>$1.60 \times 10^3$ years</td>
</tr>
<tr>
<td>Th-234</td>
<td>$6.60 \times 10^{-2}$ years</td>
</tr>
<tr>
<td>*Rn-222(g)</td>
<td>$1.05 \times 10^{-2}$ years</td>
</tr>
<tr>
<td>Pb-210</td>
<td>$2.20 \times 10^1$ years</td>
</tr>
<tr>
<td>Bi-210</td>
<td>$1.37 \times 10^{-2}$ years</td>
</tr>
<tr>
<td>Po-210</td>
<td>$3.79 \times 10^{-1}$ years</td>
</tr>
<tr>
<td>Pa-234m</td>
<td>1.17 mins</td>
</tr>
<tr>
<td>Po-218</td>
<td>3.05 mins</td>
</tr>
<tr>
<td>Pb-214</td>
<td>26.8 mins</td>
</tr>
<tr>
<td>Bi-214</td>
<td>19.9 mins</td>
</tr>
<tr>
<td>Po-214</td>
<td>0.1643 ms</td>
</tr>
</tbody>
</table>
Introduction

- Limitation
  - Number of species
  - Retardation factors
- Homogeneous and isotropic medium
- Dimensionality of model
- The source depletes only by radioactive decay
Introduction

- Development
  - Bauer developed a Laplace domain solution using a recursive form with distinct retardation factors. (2001)
  - Their recursive formula can be used to build a more complex multi-species transport solution.
Model

Schematic of Contaminant Transport

Tailings pond

Leaching

Aquifer

Infiltration

U-238, U-234, Th-230, Ra-226
Ingrowths of progenies

Advection
Hydrodynamic dispersion
Retardation and decay

Concentration

Radiological model

Dose

Well
Model

- Parent radionuclide for an inhomogeneous and anisotropic aquifer, the equation:

\[
R_1 \frac{\partial N_1}{\partial t} = \frac{\partial}{\partial x} \left( D_x \frac{\partial N_1}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_y \frac{\partial N_1}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_z \frac{\partial N_1}{\partial z} \right) - u \frac{\partial N_1}{\partial x} - v \frac{\partial N_1}{\partial y} - w \frac{\partial N_1}{\partial z} - R_1 \lambda_1 N_1
\]

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Dimension</th>
</tr>
</thead>
<tbody>
<tr>
<td>(N_1)</td>
<td>the concentration of the parent in groundwater</td>
<td>atom(L^{-3}) or mol</td>
</tr>
<tr>
<td>(D_x, D_y, D_z)</td>
<td>hydrodynamic dispersion coefficient</td>
<td>(L^2T^{-1})</td>
</tr>
<tr>
<td>(x, y, z)</td>
<td>longitudinal, lateral, vertical distance</td>
<td>(L)</td>
</tr>
<tr>
<td>(u, v, w)</td>
<td>groundwater seepage velocity</td>
<td>(LT^{-1})</td>
</tr>
<tr>
<td>(\lambda_1)</td>
<td>radioactive decay constant</td>
<td>(T^{-1})</td>
</tr>
</tbody>
</table>
Model

- Decay chain equation for $i$th member

$$R_i \frac{\partial N_i}{\partial t} = \frac{\partial}{\partial x} (D_x \frac{\partial N_i}{\partial x}) + \frac{\partial}{\partial y} (D_y \frac{\partial N_i}{\partial y}) + \frac{\partial}{\partial z} (D_z \frac{\partial N_i}{\partial z}) - u \frac{\partial N_i}{\partial x} - v \frac{\partial N_i}{\partial y} - w \frac{\partial N_i}{\partial z} - R_i \lambda_i N_i + R_{i-1} \lambda_{i-1} N_{i-1}$$

- $i > 1$ to $M$, $M$: number of the total nuclide

- Last term: ingrowths of the progenies from the preceding parent radionuclides.
Model

- Retardation factor of the radionuclide

\[ R_i = 1 + \frac{(K_{di} \rho_b)}{\theta} \]

- \( K_{di} \): the distribution coefficient of nuclide \( i \)
- \( \rho_b \): the bulk density of the aquifer material (\( ML^{-3} \))
- \( \theta \): the porosity
Model

- Hydrodynamic dispersion coefficients:

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</tr>
</thead>
<tbody>
<tr>
<td>$\alpha_L$</td>
<td>the longitudinal dispersivity</td>
<td>L</td>
</tr>
<tr>
<td>$\alpha_T$</td>
<td>the lateral dispersivity</td>
<td>L</td>
</tr>
<tr>
<td>$\alpha_V$</td>
<td>the vertical dispersivity</td>
<td>L</td>
</tr>
<tr>
<td>$\vec{V}$</td>
<td>the velocity vector</td>
<td>LT$^{-1}$</td>
</tr>
<tr>
<td>$D_m$</td>
<td>the molecular diffusivity</td>
<td>L$^2$T$^{-1}$</td>
</tr>
<tr>
<td>$\tau$</td>
<td>tortuosity</td>
<td>-</td>
</tr>
</tbody>
</table>
Model

http://www2.nau.edu/~doetqp-p/courses/env303a/lec36/lec36.htm
Model

- The release rate of radionuclides from the tailing ponds into the groundwater:

\[
\varphi_i(t) = N_i(t) K_{li} \exp\left[-(\lambda_i + K_{li})t\right]
\]

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<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Dimension</th>
</tr>
</thead>
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<tr>
<td>(\varphi_i(t))</td>
<td>the release rate of the (i)th nuclide</td>
<td>atoms (T^{-1})</td>
</tr>
<tr>
<td>(N_i(t))</td>
<td>the inventory of the (i)th nuclide(atoms) at time (t)</td>
<td>atoms</td>
</tr>
<tr>
<td>(K_{li})</td>
<td>the leach late/ fractional release rate of the (i)th nuclide</td>
<td>(T^{-1})</td>
</tr>
<tr>
<td>(t)</td>
<td>time</td>
<td>(T)</td>
</tr>
</tbody>
</table>
Model

- The leach rate from tailing ponds can be calculated as shown below:

\[
K_{li} = \frac{vS}{\theta_s \ VR_{is}}
\]

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<tr>
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<th>Dimension</th>
</tr>
</thead>
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<tr>
<td>v</td>
<td>the infiltration rate of the water from the tailing pond</td>
<td>LT(^{-1})</td>
</tr>
<tr>
<td>s</td>
<td>the surface area of the tailing pond</td>
<td>L(^2)</td>
</tr>
<tr>
<td>(\theta_s)</td>
<td>the porosity of the tailings</td>
<td>-</td>
</tr>
<tr>
<td>V</td>
<td>the volume of the tailing pond</td>
<td>L(^3)</td>
</tr>
<tr>
<td>(R_{is})</td>
<td>the retardation factor of nuclide i in the tailings pond</td>
<td>-</td>
</tr>
</tbody>
</table>
Model

- The concentration of the $i$th species at the source area:

$$N_{0i}(t) = \frac{N_i(t)}{\theta_S \ VR_{is}} \ exp[-(\lambda_i + K_{li})t]$$
Model

- The number of atoms of the parent radionuclide at the source area can be calculated using following equation:

\[
\frac{dN_1}{dt} = -\left( \lambda_1 + K_{l1} \right) N_1
\]
Model

- The number of atoms \( i \)th daughter radionuclide can be calculated using the following equation:

\[
\frac{dN_i}{dt} = \lambda_{i-1} N_{i-1} - (\lambda_i + K_{li}) N_i
\]
Result and discussion

- Parameter:
  - 316.2m x 316.2m x 20m
  - The groundwater seepage velocity : 0.03m/day
  - Porosity(θ) : 0.3
  - Bulk density : 1.5g/mL
Result and discussion

- Bauer-1D transient model

\[ t=300 \text{days} \]
\[ \lambda=(7.0, 5.0, 4.5, 3.8) \]
\[ R=(5.3, 1.9, 1.2, 1.3) \times 10^{-4} \text{day}^{-1} \]
\[ u=1 \text{m/day} \]
\[ C_0=(100, 0, 0, 0) \text{mmol} \]
\[ \theta=0.15 \]
Result and discussion

- Bauer-1D steady state model

\[ \lambda = (7.0, 5.0, 4.5, 3.8) \]
\[ R = (5.3, 1.9, 1.2, 1.3) \times 10^{-4} \text{day}^{-1} \]
\[ u = 1 \text{m/day} \]
\[ C_0 = (100, 0, 0, 0) \text{mmol} \]
\[ \theta = 0.15 \]
Result and discussion

- Bauer-3D transient model

\[ \lambda = (7.0, 5.0, 4.5, 3.8) \]
\[ R = (5.3, 1.9, 1.2, 1.3) \times 10^{-4} \text{day}^{-1} \]
\[ u = 1 \text{m/day} \]
\[ C_0 = (100, 0, 0, 0) \text{mmol} \]
\[ \theta = 0.15 \]
### Result and discussion

**Table 1**
Uranium decay chain and nuclear dependent parameters.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life, $T_{1/2}$ (years)</th>
<th>Distribution coefficient, $K_d$ (mL/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{U}$</td>
<td>$4.51 \times 10^9$</td>
<td>$5.00 \times 10^2$</td>
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<tr>
<td>$^{234}\text{Th}$</td>
<td>$6.60 \times 10^{-2}$</td>
<td>$2.00 \times 10^3$</td>
</tr>
<tr>
<td>$^{234}\text{U}$</td>
<td>$2.48 \times 10^5$</td>
<td>$5.00 \times 10^2$</td>
</tr>
<tr>
<td>$^{230}\text{Th}$</td>
<td>$8.00 \times 10^4$</td>
<td>$2.00 \times 10^3$</td>
</tr>
<tr>
<td>$^{226}\text{Ra}$</td>
<td>$1.60 \times 10^3$</td>
<td>$5.00 \times 10^2$</td>
</tr>
<tr>
<td>$^{222}\text{Rn}$</td>
<td>$1.05 \times 10^{-2}$</td>
<td>0.00</td>
</tr>
<tr>
<td>$^{210}\text{Pb}$</td>
<td>$2.20 \times 10^1$</td>
<td>$3.00 \times 10^2$</td>
</tr>
<tr>
<td>$^{210}\text{Bi}$</td>
<td>$1.37 \times 10^{-2}$</td>
<td>$3.00 \times 10^2$</td>
</tr>
<tr>
<td>$^{210}\text{Po}$</td>
<td>$3.79 \times 10^{-1}$</td>
<td>$1.50 \times 10^2$</td>
</tr>
</tbody>
</table>
Result and discussion

- Time history of $U^{238}$ and its progenies

$Rn^{222} > Po^{210} > Bi^{210} = Pb^{210} > Ra^{226} > U^{238} = U^{234} > Th^{234} > Th^{230}$
Result and discussion

- Lower C (∵ high $K_d$ value)
- Higher C (∵ low $K_d$ value and ingrowth)
- Rn-222: dissolve in groundwater and does not escape from groundwater.
Result and discussion

- U-238 as distance from tailing ponds

y=0, z=0, t=100 years
Result and discussion

- With & without decay chain
Result and discussion

- Percentage contribution: 99.75%
- $\text{Rn}^{222}, \text{Po}^{210}, \text{Pb}^{210}, \text{Ra}^{226}$
- Percentage contribution: 0.25%
Conclusions

- Low $K_d$ & ingrowth $\rightarrow$ High C
- When distance ↑, C ↓
- $C_{\text{progenies}} > C_{\text{parent}}$
- The effective dose with decay chain transport are 100 times than without decay chain transport.
- $\text{Rn}^{222}, \text{Po}^{210}, \text{Pb}^{210}, \text{Ra}^{226}$: 99.75%, and other radionuclide: 0.25%
Thank you for listening!