Groundwater Transport of Radionuclides in a Crystalline Geosphere

John Avis
Geofirma Engineering, Ottawa, Ontario, Canada
Erik Kremer
Nuclear Waste Management Organization, Toronto, Ontario, Canada

ABSTRACT
The Nuclear Waste Management Organization (NWMO) is responsible for the implementation of Adaptive Phased Management, the federally-approved plan for the safe long-term management of Canada’s used nuclear fuel. Under this plan, used nuclear fuel will ultimately be placed within a deep geological repository in a suitable host rock formation.

The NWMO recently completed a study illustrating the postclosure safety of a hypothetical used fuel repository in a crystalline host formation. The Base Case scenario assumes that undetected defects in used fuel containers eventually lead to container failure and release of radionuclides to the geosphere. Detailed groundwater flow and transport modelling was used to support the safety assessment by calculating transport of radionuclides from the repository to the biosphere.

The relative importance of different transport processes is important in terms of defining site characterization requirements for actual repository sites. Sensitivity cases addressing geosphere transport parameters provided initial insight into dominant transport processes. Probabilistic simulations were used to further investigate relative importance of various transport processes and the sensitivity to parameters describing the processes.

1 INTRODUCTION
The Nuclear Waste Management Organization (NWMO) has undertaken postclosure safety assessments of hypothetical geological repositories for used fuel hosted in crystalline and sedimentary rock formations. Before specific sites have been identified for detailed examination, generic case studies are used to illustrate the long-term performance and safety of the multi-barrier repository system within various geological settings (NWMO 2012, 2013). These postclosure safety assessments are part of a federally approved Adaptive Phased Management plan for the safe long-term management of Canada’s used nuclear fuel. NWMO (2017) describes the recently completed Sixth Case Study (6CS), an assessment of a revised repository and EBS design, situated at a depth of approximately 500 metres below ground surface (mBGS) within a hypothetical complex faulted crystalline geosphere. The Base Case scenario assumes that undetected container defects eventually lead to container failure, to groundwater making contact with used nuclear fuel, to consequent release of radionuclides and transport to the geosphere. The scenario considers the primary route to the biosphere to be a deep water-supply well.

The role of different processes in radionuclide transport within crystalline rock is well understood at a conceptual level. Advection and related dispersive processes are important at higher hydraulic conductivities and/or hydraulic gradients, while diffusion dominates where advective flow is stagnant or non-existent. However, the threshold at which the dominant process...
changes from advection to diffusion is not well defined. In a crystalline rock geosphere, thresholds will be site specific as hydraulic gradients are dependent upon local and regional topography and the extent and permeability of large scale fracture systems.

As the NWMO program moves into siting activities at volunteer communities, site specific site characterization programs will be designed. Hydraulic conductivity testing will be an integral component of any characterization program. Testing of extremely low hydraulic conductivity rock, as described in Beauheim et al (2014), is complex and expensive. Knowledge of the required lower limit of testing, below which only diffusive transport is expected, will be useful in ensuring that technically appropriate hydraulic test programs are planned. The assessment approach described here can be applied to future potential sites before field programs are undertaken to assist in planning field programs.

2 MODELLING APPROACH

This section describes numeric modelling tools used in this study, and the design and construction of the flow and transport models.

2.1 Numeric Tools and Computational Platform

All groundwater flow and transport simulations were conducted using FRAC3DVS-OPG v1.3, a control-volume finite-element code (Therrien et al, 2010). FRAC3DVS-OPG has been used extensively by the NWMO in previous safety assessments (NWMO 2012, 2013).

paCalc, developed by Geofirma Engineering, is a model execution framework that supports probabilistic assessments using detailed numeric models (Avis and Sgro, 2013) and also provides extensive statistical post-processing capabilities.

mView is a model pre- and post-processor, also developed by Geofirma Engineering, that was used for grid generation, property assignment, and all post-processing and model results presentation. Initial model set-up (gridding, property assignment) is performed using the Windows GUI version of mView. A command line version is called by paCalc to execute realization-specific pre-processing, and post-processing realization results into a form suitable for reading by paCalc.

All computations were performed on a local cluster machine with five dual-quad Xeon processor computers. Each computer was equipped with 48 GB of RAM.

2.2 Flow and Transport Models

Flow and transport modelling was performed using a nested-models approach. Two model scales are relevant to the current paper: 1) the Subregional Flow Model (SRF), a watershed scale model, which calculates a steady-state groundwater flow system, and 2), the Repository Transport model, which calculates transport of radionuclides from a source within the repository footprint to a water-supply well.

The SRF model does not contain any repository features, while the reduced domain Repository Transport model includes a detailed representation of the waste placement rooms. It uses fixed-head boundary conditions extracted from the SRF model on all vertical sides to maintain fidelity with the subregional groundwater flow system. Surface topography, surface water features, repository layout and model boundaries are shown in Figure 1.

The models assume a low-permeability geosphere where bedrock hydraulic conductivity decreases with depth. Fractures are included using an equivalent porous medium (EPM) approach where fracture element properties are adjusted to reflect a blended conductivity tensor and porosity scalar. Figure 2 shows hydraulic conductivity on a vertical slice through the Repository Transport model. Repository placement rooms are located on either side of the main vertical fracture that bisects the repository.

3 DETERMINISTIC MODEL RESULTS

The deep water-supply well and the location of hypothetical container failures (“sources”) in the Repository Transport model were selected in such a way as to maximize transport to the well, using the procedure described in Avis and Kremer (2017). Four consequential well and associated source locations were determined. This paper concentrates on the Main fracture well location that serves as the Base Case in NWMO (2017).
at 1000 a, with subsequent failures every 100 ka thereafter. All containers were conservatively assumed to be located at the specified source location, chosen to minimize the travel time to the well, rather than being randomly distributed throughout the repository footprint. Simulations presented in this paper are for the transport of Iodine-129, a long-lived radionuclide with a significant contribution to dose. Iodine is a conservative species with respect to transport, with no sorption expected. Other, shorter-lived and/or sorbing species were considered in the assessment but are not presented here.

3.1 Base Case Results

Figure 3 presents time-series results showing the defective container source term and the simulated uptake at the water supply well.

![Figure 3. Base Case transport source term and results.](image)

The water supply well is effective in capturing released I-129, with approximately 90% of the source term activity captured by the well over the duration of the simulation. Figure 4 is a three-dimensional representation of the I-129 concentration at the time of peak transport to the well.

![Figure 4. Base Case transport at 25 ka](image)

3.2 Geosphere Parameter Sensitivity Case Results

Geosphere parameters evaluated in sensitivity cases include hydraulic conductivity, dispersivity, and effective diffusion coefficient, as listed in Table 1. A single parameter is varied for each case, with all other parameters remaining at the Base Case values.

<table>
<thead>
<tr>
<th>Case ID</th>
<th>Case Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>High K</td>
<td>Base case hydraulic conductivity x 10</td>
</tr>
<tr>
<td>Low K</td>
<td>Base case hydraulic conductivity /10</td>
</tr>
<tr>
<td>High Disp</td>
<td>Base case dispersivity x 5</td>
</tr>
<tr>
<td>Low Disp</td>
<td>Base case dispersivity / 2</td>
</tr>
<tr>
<td>High Diff</td>
<td>Base case effective diffusion coefficient x 10</td>
</tr>
</tbody>
</table>

The hydraulic conductivity sensitivity case results indicate that the Base Case model is within an advection-dominated transport regime. This was determined by examining the impact of hydraulic conductivity (see Figure 5): the peak well transport occurs much earlier for the High K, while the response is delayed for the Low K case. In a diffusion-dominated transport system this would not happen. The reduction in magnitude for the High K case is caused by reduced well capture, as the increased hydraulic conductivity in the shallow bedrock (less than 150 mBGS) allows more mass to bypass the well and discharge at surface.

![Figure 5. Hydraulic conductivity sensitivity cases](image)

Dispersion and diffusion sensitivity results confirm this conclusion (see Figure 6). Increased diffusivity has a minor impact on the peak value but does not affect arrival time at the well. The effect of dispersivity is as expected for an advective transport regime. The peak for the High Disp case arrives slightly earlier at the well, and the magnitude is somewhat decreased, corresponding to an advanced leading edge of the plume, but with increased overall spreading. In contrast, the Low Disp case results in a higher well peak transport due to reduced spreading of the plume.

![Figure 6. Hydraulic conductivity sensitivity cases](image)
3.3 Transport Process Indicators

Peclet numbers have been used as a means of determining the transport characteristics of a groundwater flow regime. Bear (1979) refers to Péclet numbers of less than 0.4 being representative of diffusion-dominated transport. Huysmans and Dassargues (2004) reviewed this approach and found no less than 10 different methods to calculate the Péclet number, with a wide variation in results for the same flow system. Clearly, not all approaches are always appropriate. Figure 7 presents Péclet numbers on a vertical slice through the Base Case Repository model. In this figure, the characteristic length is 1.0 m, the value used in NWMO (2012). Transport within the intermediate bedrock surrounding the repository appears to be in the transition region between advection- and diffusion-dominated flow.

Freeze and Cherry (1977) proposed instead to compare the ratio of dispersive to diffusive flux. Walsh and Avis (2010) used this approach in evaluating radionuclide transport under glaciation conditions. Results of this calculation are shown in Figure 8. According to this approach, transport is clearly advection dominated. Note that increasing the characteristic length for the Péclet calculation from 1.0 m to 20 m will cause the Péclet calculation results to be similar to the ratio presented in Figure 8.

4 PROBABILISTIC MODELLING RESULTS

Dominant transport processes can also be determined from statistical analyses. In this approach, univariate and multivariate statistics are used to determine the significance of individual parameters to a metric defining system performance.

4.1 Background and Approach

Avis and Sgro (2013) applied probabilistic sampling of geosphere and engineered barrier system (EBS) parameters to create parameter set realizations for transport of radionuclides using the detailed models from a previous crystalline geosphere assessment (NWMO 2012). Transport simulations were conducted for each realization and metrics describing peak transport at the well were extracted from simulation results. The significance of the independent variables (the sampled geosphere parameters) was assessed in terms of their correlation and contribution to variability in the dependent variables (well-transport peak value and time).

This approach has been applied here with a parameter set limited to the geosphere transport sensitivity parameters described previously. Hydraulic conductivity and the effective diffusion coefficient are assigned log-uniform distributions, while longitudinal dispersivity is assigned a uniform distribution. Base case and sampled distributions for each parameter are listed in Table 2.

Table 2. Sampled Parameter Values

<table>
<thead>
<tr>
<th>Case ID</th>
<th>Deterministic Base Case</th>
<th>Parameter Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Minimum</td>
</tr>
<tr>
<td>Hydraulic Conductivity (m/s)</td>
<td>4 × 10^-11</td>
<td>1 × 10^-13</td>
</tr>
<tr>
<td>Longitudinal Dispersivity (m)</td>
<td>20</td>
<td>5</td>
</tr>
<tr>
<td>Effective Diffusion (m²/s)</td>
<td>3.6 × 10^-13</td>
<td>1 × 10^-11</td>
</tr>
</tbody>
</table>

Sampled values are varied for the intermediate depth bedrock unit within which the repository is located. All other variables are fixed, or calculated from sampled parameters by fixed multipliers, using ratios defined for the Base Case model. For example, deep bedrock hydraulic conductivity is defined as 0.25 times the intermediate bedrock value, while transverse dispersivity...
is 10% of longitudinal dispersivity. One modification was made to the ratios, with shallow bedrock hydraulic defined as 5 times that of intermediate, rather than 50 times specified for the Base Case. This was done to reduce the confounding impact of reduced well capture at higher shallow bedrock permeabilities (evident in the reduced peak value of the High K case in Figure 5).

Samples were calculated using a Latin Hypercube Sampling (LHS) routine (Iman and Shortencarier, 1984) to ensure complete coverage of the distribution domain. No variable correlations were specified. Figure 9 shows sample values for 100 realizations, with Base Case and deterministic sensitivity values for comparison.

Figure 9. Sampled intermediate bedrock geosphere parameters.

The source term was simplified to a constant magnitude $10^6$ Bq/a release at the source container location for a 1000 year period (total release $10^9$ Bq). This provides a cleaner signal for analyses than the time-varying 10 container release shown in Figure 3.

4.2 Simulation Results

One hundred realizations were simulated and well transport metrics were extracted. Three simulations failed to reach peak values within the time limits enforced by the executive code. Figure 10 presents overall well transport, peak transport rates/times for each simulation, and results of deterministic runs with varying hydraulic conductivity analogous to the High K and Low K cases presented in Figure 5.

Figure 10. Well transport for 97 completed simulations

Two dependent variable metric values are available: peak transport time or peak transport value. The ranked correlation coefficient of the dependent variables is -0.97, indicating a very high negative correlation (e.g., low peak transport times are strongly associated with high peak transport rates). This means that either metric is a suitable choice for further study, providing equivalent information. Peak transport time was selected for all subsequent analyses.

4.3 Univariate Assessment

Figure 11 shows scatter-plot correlations for each independent variable to peak transport time. Hydraulic conductivity has a meaningful correlation, driven by the near linear response for values greater than $10^{-11}$ m/s, although spreading of the points below this value indicates the increased importance of the other independent variables. Correlations to the other variables are not significant when applied over the entire sampled data set.

Figure 11. Scatter plot correlations for independent variables

Evaluation of the data can be improved by examining variable relationships in subsets of the data. The data set was divided into groups corresponding to realizations where hydraulic conductivity was within an order of magnitude of a specified value, with statistics then calculated on that data group. This approach is illustrated for four non-overlapping intervals with univariate ranked correlation statistics calculated for each varied parameter. Results shown in Figure 12 now clearly show the variation in parameter significance, with $D_{eff}$ highly correlated to transport time at hydraulic conductivity values below $10^{-12}$ m/s, and hydraulic conductivity correlations high above $10^{-11}$ m/s.

Figure 12. Variable correlations over ranges of K
Extending the approach by applying it to overlapping data groups, the hydraulic conductivity domain is partitioned into 100 overlapping log-cycle intervals and the univariate ranked correlation results are calculated on each interval. For example, the first interval extends from $10^{-13}$ m/s to $10^{-12}$ m/s, the second from $1.07 \times 10^{-13}$ to $1.07 \times 10^{-12}$, and the third from $1.14 \times 10^{-13}$ to $1.14 \times 10^{-12}$. Correlations are calculated on these “rolling” data groups (generally each containing 25 points) and are plotted at each group’s log-mean hydraulic conductivity ($3.39 \times 10^{-13}$ m/s, $3.63 \times 10^{-13}$ m/s, and $3.89 \times 10^{-13}$ m/s, respectively).

As shown in Figure 13, for hydraulic conductivity less than $10^{-12}$ m/s, diffusion is clearly dominant. For hydraulic conductivity greater than $10^{-12}$ m/s, dispersivity starts to become significant with a peak correlation of approximately 0.6 at $3.2 \times 10^{-12}$ m/s, while the significance of $D_{\text{eff}}$ steadily moves closer to zero. The correlation coefficient of hydraulic conductivity exceeds (in an absolute value sense) $D_{\text{eff}}$ for hydraulic conductivity values of $5 \times 10^{-12}$ m/s or greater, marking the transition from diffusion-dominant to advection-dominant transport.

The transition region is arbitrarily defined as the period over which the correlation coefficients of hydraulic conductivity and effective diffusion differ by less than 0.20. In Figure 13 the transition region extends from $3.8 \times 10^{-12}$ m/s to $6.6 \times 10^{-12}$ m/s.

Figure 13. Variable correlations for overlapping ranges of K

4.4 Multivariate Assessment

The overlapping-interval extraction approach described above can be further extended to use more sophisticated statistical analyses. Iman et al. (1980) describe multivariate correlation and regression approaches used for performance assessment. Ranked partial correlation coefficients (WIPP, 1995a) shown in Figure 14 provide a more nuanced interpretation than the univariate statistics. The transition region has expanded, showing significance for both parameters for hydraulic conductivities from $1.1 \times 10^{-12}$ m/s to $7.6 \times 10^{-12}$ m/s.

In this case, hydraulic conductivity has a minor significance (albeit less so than $D_{\text{eff}}$) at conductivities below $10^{-12}$ m/s, while the significance of dispersivity is greater at higher hydraulic conductivity than indicated by the univariate correlations.

As a final approach, ANOVA calculations calculated by stepwise regression (WIPP, 1995b) show the contribution to metric variance for each parameter. Figure 15 clearly shows the transition between hydraulic conductivity and $D_{\text{eff}}$ at $5 \times 10^{-12}$ m/s, while dispersivity is largely unimportant.

Figure 14. Multivariate ranked partial correlation on overlapping intervals.

4.5 Application to Alternate Well/Source Simulations

As mentioned in Section 2.2, four high consequence well and source location combinations were identified for the 6CS. We now apply the probabilistic approach to the North West (NW) well/source, for which peak transport is visualized in Figure 16.

Figure 15. Multivariate ranked ANOVA on overlapping intervals.

Figure 16. North West well/source Base Case transport at 15 ka
The NW case is a more direct transport pathway than the Main fracture case due to the much shorter fracture transport path. Transport through intact bedrock before intersecting the fracture system is similar in both cases. Results from probabilistic simulations (Figure 17) are consistent with the Main fracture case (Figure 10) although with earlier minimum arrival times.

Figure 17. Well transport for NW simulations

Multivariate statistics (Figure 18 and 19) are also very similar to the Main fracture case, although both figures show greater significance to dispersivity in the advective transport region. This is likely due to the increased magnitude of dispersivity relative to the overall transport pathway length.

Figure 18. Multivariate ranked partial correlation on overlapping intervals

Figure 19. Multivariate ranked ANOVA on overlapping intervals.

Results for the NW well/source are consistent with those for the Main fracture. Geosphere performance within the vicinity of the repository is likely similar across the footprint.

5 CONCLUSIONS

The relative significance of advective or diffusive processes in the transport of radionuclides through a crystalline rock geosphere is dependent on the site-specific hydrogeologic setting. Assuming hydraulic gradients are determined by topography and fracture networks, the hydraulic conductivity of the intact bedrock is the primary control of advective processes in the near vicinity of the repository. Existing approaches to determining dominance of transport processes are ambiguous.

A rigorous probabilistic approach to evaluating significance of transport processes has been developed. The approach provides a clear indication of the onset of diffusion dominance. In the example hypothetical geosphere, advection contributes to radionuclide transport at hydraulic conductivities down to approximately $10^{-12}$ m/s, which should therefore form the lower bound for hydraulic testing requirements in this particular geosphere.

6 ACKNOWLEDGEMENTS

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REFERENCES


