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Deep Borehole Disposal Isolation Strategy

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Panel #81: Deep Borehole Disposal of Radionuclides (focused on SNF and HLW)

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Deep Borehole Disposal Isolation Strategy

Questions:

- **How do hypersaline brines form?**
- **What is the significance for waste isolation?**
- **What does ancient age tell us about potential contaminant transport times to the biosphere?**

Groundwater Survey Example (Gascoyne 2004)

Near-surface waters + 86 fracture water samples from permeable zones in 53 boreholes, up to 1000 m deep in the Lac du Bonnet batholith (SE Manitoba)

- **Near-surface (< 200 m) waters are dilute, and appear modern, meteoric (^{3}H, ^{14}C, warm-climate $^{2}H/^{18}O$)**
- **Transition (200 to 400 m) waters resemble glacial melt, increasingly alkaline (apparent age 10^3 to 10^5 a from ^{14}C)**
- **Deep (> 500 m) waters are Na–Ca–Cl–SO_4 type, salinity to 50 g/L, pre-glacial (apparent age ~1 Ma; warm-climate $^{2}H/^{18}O$)**
- **Porewaters from URL excavation (~ 500 m) are saline (90 g/L), nearly pure Ca-Cl (apparent age 10^1 to 10^3 Ma; ^{18}O deficient, He and Ar enriched, ingrowth of fission-product ^{36}Cl and ^{129}I)**

→ ***Increasing water-rock interaction, possible marine or evaporite early origin for deep waters (Ca/Na, Cl/Br, $\delta^{34}S$)***

Gascoyne, M. 2004. "Hydrogeochemistry, groundwater ages and sources of salts in a granitic batholith on the Canadian Shield, southeastern Manitoba." Applied Geochem. 19, pp. 519–560.

Kotzer, T., M. Gascoyne, M. Mukai, J. Ross, G. Waito, G. Milton and R.J. Cornett 1998. "Cl-36, I-129 and noble gas isotope systematics in groundwaters from the Lac du Bonnet Batholith, Manitoba, Canada." Radiochim. Acta 82, pp. 313–318.

Possible Origins of Hypersaline Crystalline Basement Brine:

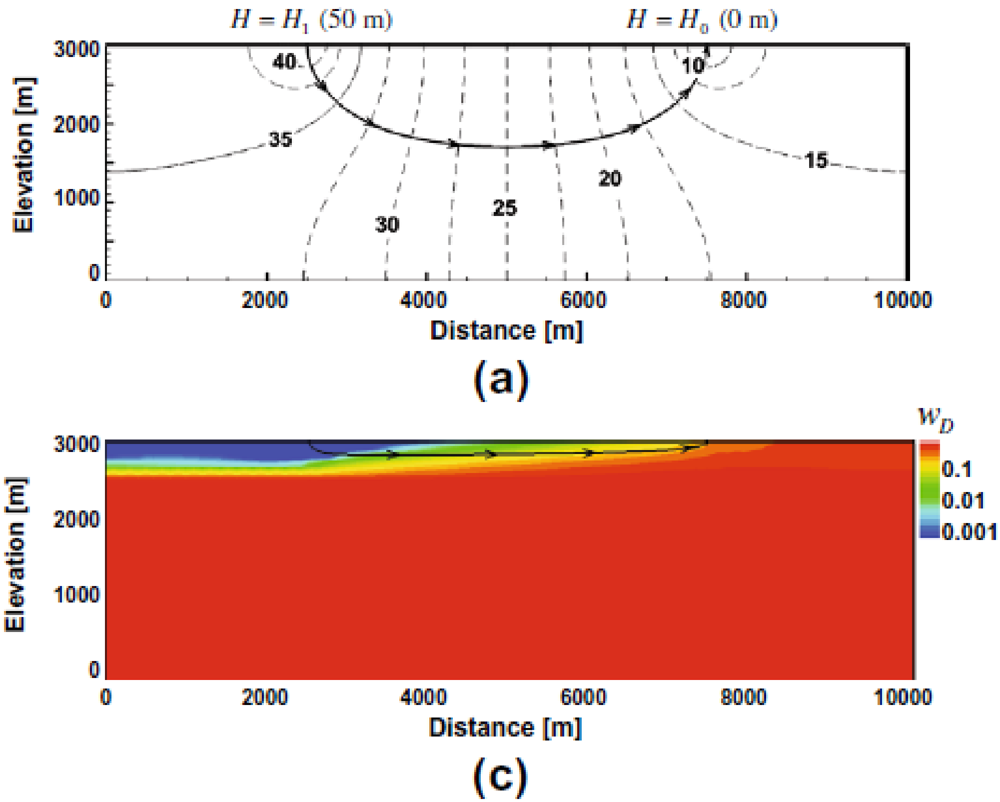
- **Marine (need some concentration or augmentation process to concentrate)**
- **Evaporite dissolution (has distinctive high Cl/Br ratio, and salt beds may be far away or low in the geologic section)**
- **Connate fluids (sedimentary pore fluids that are residues of evaporite precipitation)**
- **Cryogenic (requires previous marine transgression at the time of brine formation)**
- **Rock-water interaction**
  - H_2O consumption by mineral alteration
  - Fluid inclusions as source for chloride

Interpretations of brine origin are site specific and uncertain…

but brine formation has occurred over geologic time scales, and its occurrence is ubiquitous, so it is evidently stable.

Significance of Basement Brines to Waste Isolation

- **Density stratification → Mixing stability**
- **Verified by numerical studies *for a homogeneous earth (Park et al. 2009)***



FRAC3DVS advective-dispersive simulation from Figure 6 of Park et al. (2009), comparing source-to-sink flowpaths (ΔH = 50 m) for:

(a) uniform groundwater, and
(c) stratified groundwater (i.e., ρ = 1.2 below 2500 m),

in a homogeneous earth.

Park et al. 2009. "Effects of shield brine on the safe disposal of waste in deep geologic environments." Advances in Water Resources 32, pp. 1352–1358.

- **Permeability decrease with depth**
  - Stober & Bucher 2007. "Hydraulic properties of the crystalline basement." *Hydrogeology Jour.* 15, pp. 213–224.
  - Manning & Ingebritsen 1999. "Permeability of the continental crust: Implications of geothermal adata and metamorphic systems." *Rev. Geophys*. 37, pp. 127-150.

Groundwater Surveys, continued

- **Some Other Recent Studies:**
  - J. Lippmann, et al. 2003. "Dating ultra-deep mine waters with noble gases and ^{36}Cl, Witwatersrand Basin, South Africa." *GCA* 67(23), pp. 4597–4619.
  - Greene, S., et al. 2008. "Canadian Shield brine from the Con Mine, Yellowknife, NT, Canada: Noble gas evidence for an evaporated Palaeozoic seawater origin mixed with glacial meltwater and Holocene recharge." *GCA* 72, pp. 4008–4019.
  - Holland, G., et al. 2013. "Deep fracture fluids isolated in the crust since the Precambrian era." *Nature* 497, pp. 357-362.
  - Kietavainen, R., et al. 2014. "Noble gas residence times of saline waters within crystalline bedrock, Outokumpu Deep Drill Hole, Finland." *GCA* 145, pp. 159–174.
  - Multiple studies by Bottomley, Lehmann, Bethke, Torgerson, Fritz, Frape, Davis, Moran, and their collaborators
- **Themes: groundwater model age from:**
  - Long-lived environmental tracers (^{81}Kr)
  - Noble gas concentrations and isotopics (He, Ar; also Ne, Xe)
  - Fission product concentrations (^{36}Cl, ^{129}I)
- **Supported by brine origin and evolution hypotheses based on:**
  - Source fingerprinting and rock-water interaction (Cl/Br, Ca/Na, ^{2}H, ^{18}O, $^{6}Li/^{7}Li$, $^{87}Sr/^{86}Sr$, etc.)
- **Characterization technology is evolving**

Noble Gas Interpretation

$Ng_{tot} = Ng_{eq\text{-}atm} + Ng_{excess} + Ng_{radogenic} + Ng_{fission} + Ng_{terrestrial} + Ng_{mantle}$

$Ng_{eq\text{-}atm}$ Atmospheric equilibrium (e.g., ^{4}He, ^{3}He)

Ng_{excess} From air entrained in recharge water

$Ng_{radogenic}$ Most important in situ source (e.g., ^{4}He from U, Th α decay; ^{3}He from $^{6}Li(n, \alpha)^{3}H(\beta-)^{3}He$; ^{40}Ar from ^{40}K decay)

$Ng_{fission}$ Products of spontaneous fission of natural U in situ (e.g., certain Kr and Xe isotopes)

$Ng_{nucleogenic}$ Reactions with neutrons from spontaneous fission of U (e.g., certain Ne isotopes)

$Ng_{terregenic}$ Crustal flux (combining different production mechanisms, mostly radiogenic)

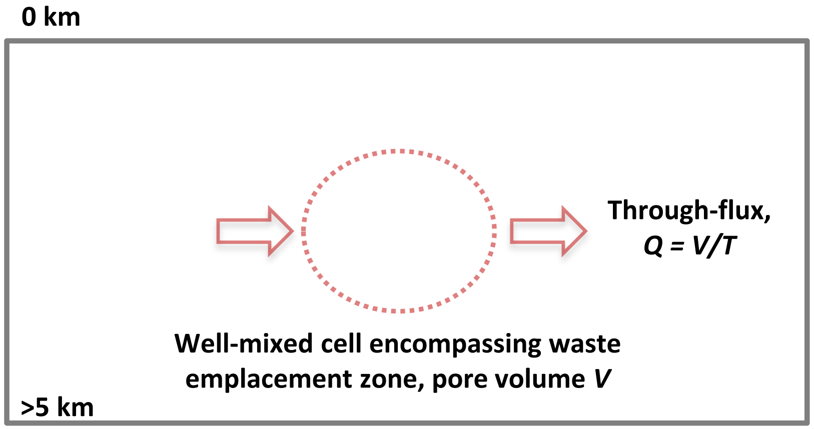
Ng_{mantle} Mantle flux

Estimating Radionuclide Travel Time from Groundwater Model Age (1/4):

- **Simplest residence time interpretation:**
  - If *T* is model age for groundwater in the control volume; and
  - Basement brines contains no indication of mixing with younger water (e.g., nuclear-age ^{3}H and ^{85}Kr, glacial $^{18}O/^{2}H$, or cosmogenic ^{81}Kr; and
  - Chemical retardation occurs in the basement; then
  - Containment time is the smaller of *T* compared to effective diffusion time (including retardation)
- **Possible problems:**
  - Model age could represent a dynamic steady state with flow in the basement
  - Site may not have an impermeable sedimentary overburden, so that interaction between near-surface and deep basement must be considered
  - Occurrence of basement flow, esp. brine discharge ("moose licks")

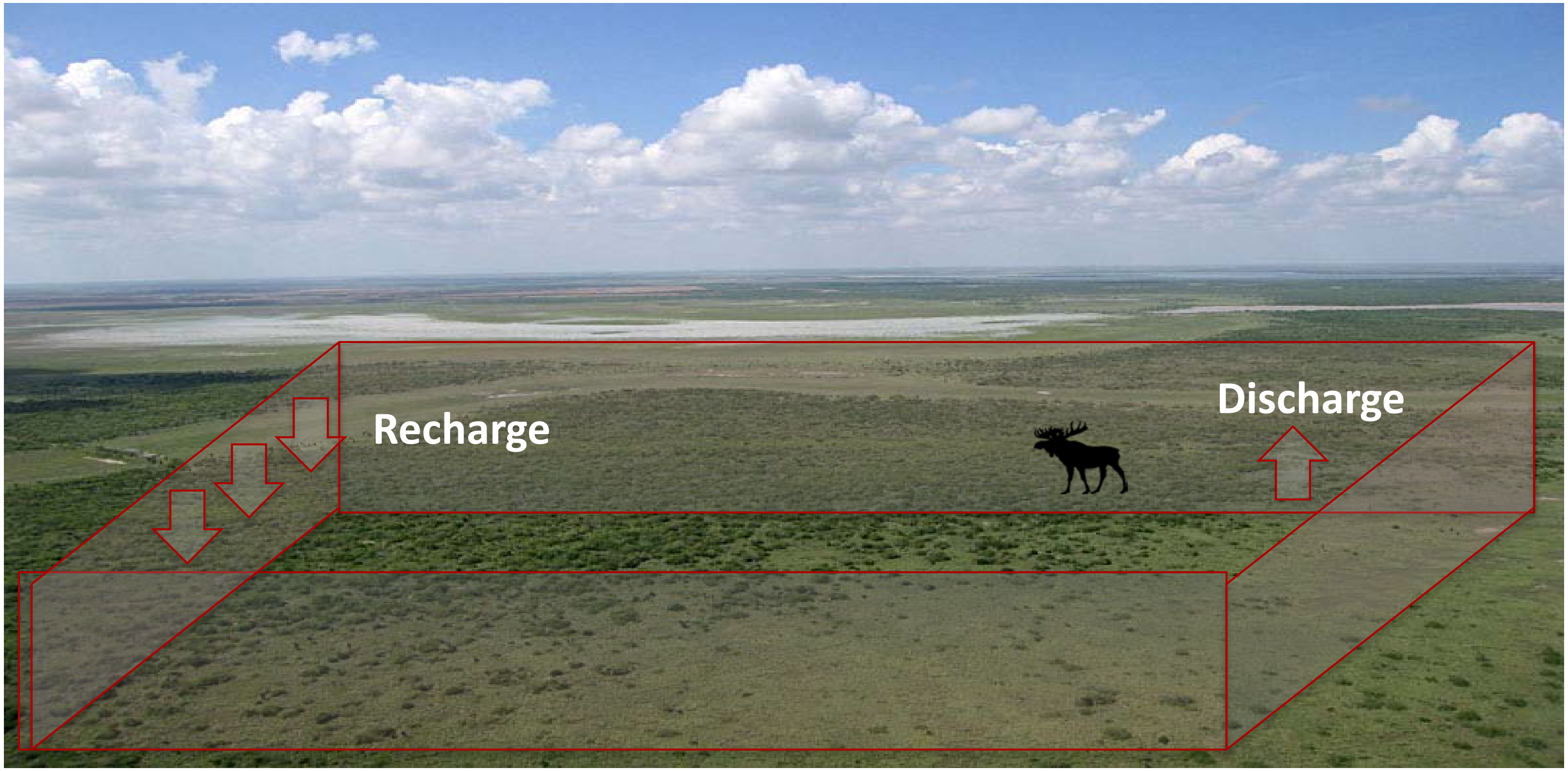
Estimating Radionuclide Travel Time from Groundwater Model Age (2/4):

- **Mixing cell interpretation:**
  - If *T* is model age for groundwater in the defined mixing volume; and
  - Basement brine contains no indication of mixing with younger water; then
  - Fractional release rate for mobile radionuclides → *1/T* (yr^{-1})
- **Possible problems :**
  - Even for model age of 10^7 to 10^9 yr, *1/T* for non-sorbing radionuclides may not be slow enough to meet waste isolation performance objectives
  - Existence of evidence for mixing of younger water with deep brine



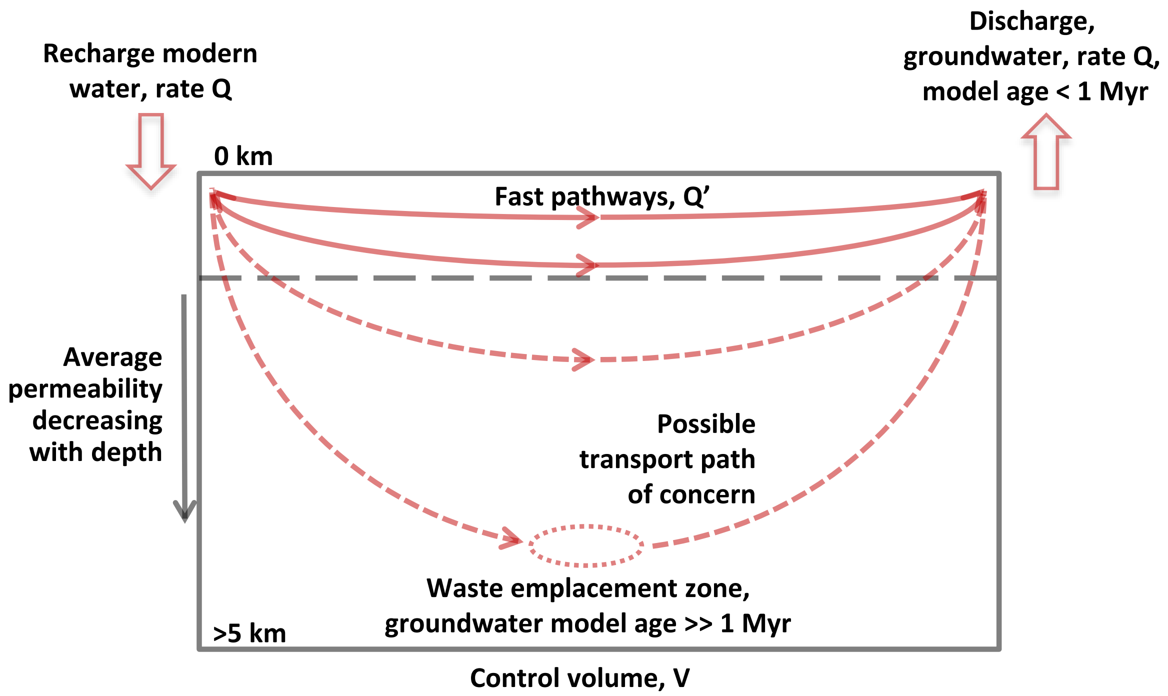
Estimating Radionuclide Travel Time from Groundwater Model Age (3/4):

- **Control volume interpretation**
  - Characterize near-surface hydrology → Identify recharge and discharge areas
  - Sample/analyze discharge for brine constituents
  - Define control volume (include recharge and discharge out to a radius representing very long travel times)



Estimating Radionuclide Travel Time from Groundwater Model Age (4/4): Control Volume Approach, cont.

- **Focus hydrologic characterization on shallow objectives (typically 500 m):**
  - Characterize recharge/discharge areas and fast pathways connecting them
  - Fast pathways have $V' < V$ and Q' approaching total Q
  - Redraw the control volume outside of fast pathways so that $T \approx (V-V')/(Q-Q')$
  - Evaluate V, V', Q and Q' from hydrologic characterization
  - Compare to T from isotopic characterization; use differences in composition between discharge and deep basement brine (e.g., $^{18}O/^{2}H$)
- **Possible problems:**
  - Inconsistent hydrochemical/ isotopic observations
  - Uncertainty in hydrologic model
- **Another alternative: Characterize hydrologic structure and flow conditions to disposal depth, and model flow throughout the basement**



Deep Borehole Field Test Borehole Sampling Objectives:

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Analyte	Sample Requirement
Water stable isotopes (e.g., ^{2}H, ^{18}O)	1 mL
Drilling fluid tracer (e.g., fluorescein or iodide)	A few mL
Major anions/cations (e.g., Na^{+}, Cl^{-}, Ca^{2+}, SO_4^{2-})	10 mL
Trace elements (e.g., Li, Sr, U)	10 mL
Dissolved inorganic and total carbon	50 mL
Other isotopic ratios for dissolved species (e.g., Li, C, N, S, Sr, U)	100's of mL
Radiogenic in situ tracers (e.g., ^{3}He, ^{4}He, ^{40}Ar)	Whole-rock samples and/or 1 to 10 L
Cosmogenic tracers (e.g., ^{81}Kr)	100 L
Scarce in situ fission products (e.g., ^{36}Cl, ^{129}I)	100's of L
Scarce terrigenic and in situ tracers (e.g., ^{3}He)	100's of L
Rare inert cases (e.g., Ne, Xe isotopes)	100's of L

Deep Borehole Field Test Borehole Sample Types:

Fluid/Gas	Solids
Drilling fluid (surface samples, also gas separator)	Cuttings
Porewater (from core: centrifuged, squeezed, flushed)	Cores (up to 150 m)
Borehole fluid (wireline sampler)	Preserved cores (a few m)
Pumped groundwater (zone isolated by packers)	