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Noble gas diffusion through variably-saturated rock: implications for verification of subsurface nuclear events

John P. Ortiz, Chelsea W. Neil, Scott Broome, Phil Stauffer, Hakim Boukhalfa, Robert Roback, Pat Brug
Los Alamos National Laboratory, Computational Earth Science (EES-16), Los Alamos, NM

Earth & Environmental Sciences

Abstract

Gas transport through variably-saturated geologic media has important applications for nuclear nonproliferation, as noble gas detection is one of the best candidates for the verification of clandestine underground nuclear events. The transport properties of porous geologic media with respect to such gases are of fundamental importance in developing accurate predictive transport models to determine the origin of detected nuclear signatures. We therefore aim to characterize the diffusion of gaseous krypton, xenon, and sulfur hexafluoride (SF6) through intact porous rock with varying degrees of liquid saturation in order to quantify vapor diffusion coefficients and relate them to changes in rock saturation. We conducted a series of diffusion cell experiments using intact tuff cores at varying saturations. We fit the experimental results using Finite Element Heat and Mass (FEHM) numerical model simulations to calculate effective vapor phase diffusion coefficients for each gas and saturation. Results will be used to develop more accurate transport models for subsurface fission product releases, which have implications for treaty verification, repository science, and radioactive contaminant transport.

Goal: To determine transport properties of variably-saturated rock and improve transport models.

Background

Noble gases produced by nuclear detonations interact very little with soil, making them key components for monitoring compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

In the case of prompt gas release following a UNE, yields of radioxenon isotopes are close to the independent yields produced by nuclear fission. If explosion is well-contained (i.e. slow to vent), the decay of parent radionuclides contributes to cumulative yield – this has potentially large impacts on measured isotopic ratios. Diffusive transport must be considered over these timescales.

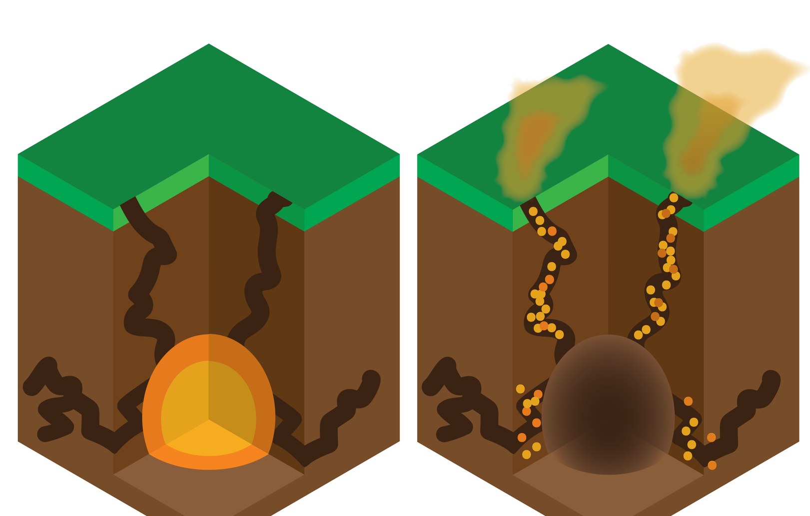

Fig. 1: Over time, barometric pumping in fractured media can bring gaseous subsurface contaminants to the surface.

Transport mechanisms are further complicated by variable saturation in the subsurface. Gases partition into pore water in different proportions, which can lead to increased storage times (right).

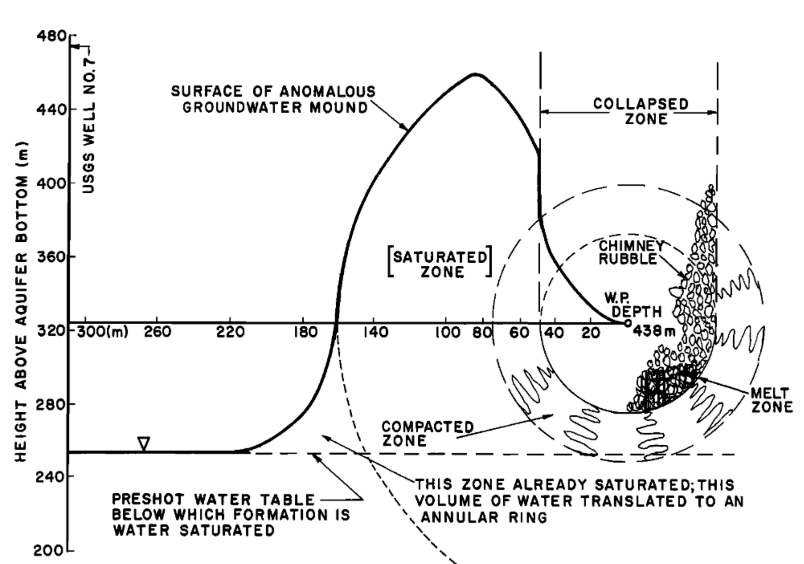

Fig. 2: Approximation of anomalous groundwater mound created by a UNE test (from Knox *et al.*, 1965).

Furthermore, the UNE itself can alter local hydrogeology (above). Detonations sometimes create a cone of elevated hydraulic heads that can persist for weeks or months (Knox *et al.*, 1965). Such groundwater changes are expected to significantly alter the transport conditions for fission products.

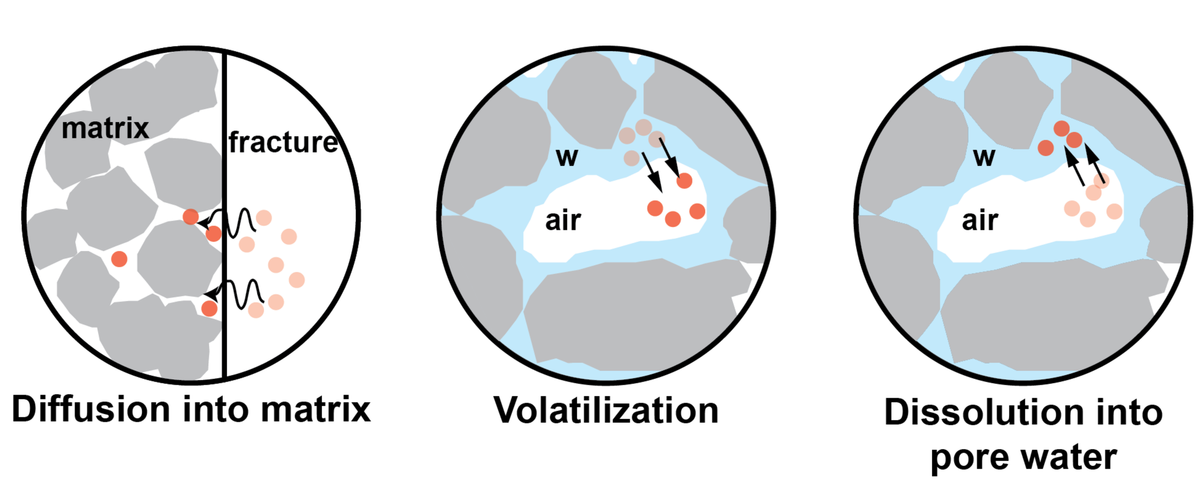

Fig. 3: Conceptual diagram of gas diffusion and partitioning between vapor and liquid phases within porous media.

Experimental setup

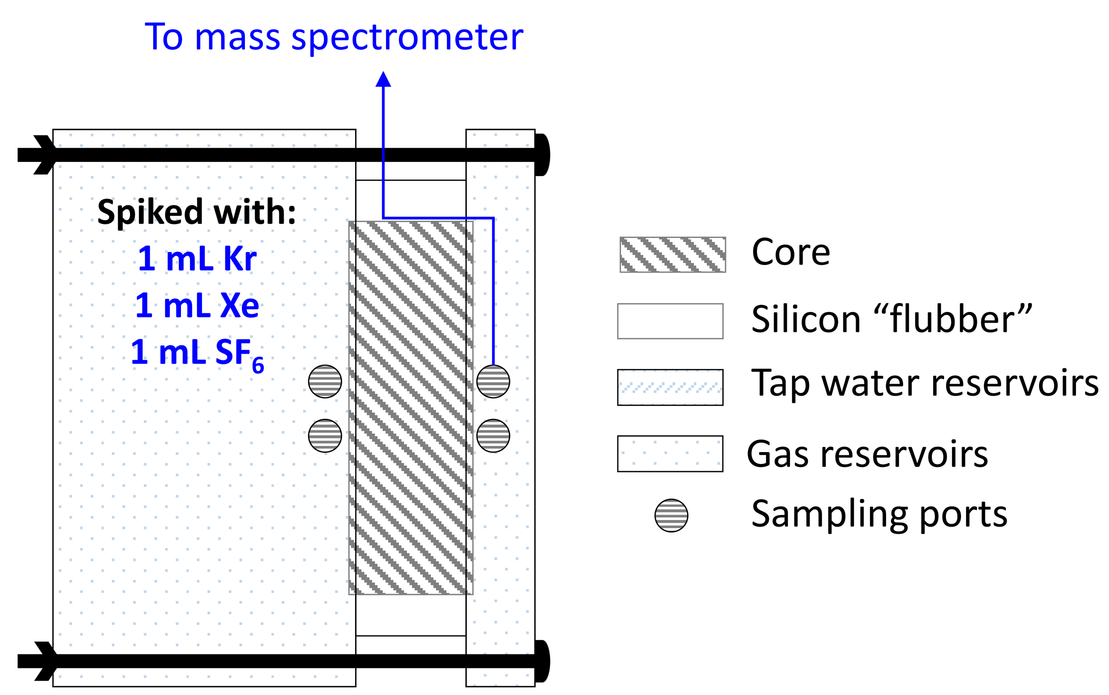

Fig. 4: Schematic of gas diffusion cell. Diagram by Chelsea Neil.

Numerical investigation

Model was built in the FEHM (Finite Element Heat and Mass) porous-flow simulator, developed at LANL.

Model Domain

- 2D radial mesh geometry
- Tuff core: radius = 3.175 cm, length = 5.1 cm
- Reservoir volume = 500 mL
- Sampling chamber volume = 75 mL
- 25,351 nodes (101 nodes in x-dirxn, 251 nodes in y-dirxn)
- Uniform discretization in the radial (x) dirxn
- Discretization in y-dirxn is uniform within each region (reservoir chamber, core, sampling chamber) (see Fig. 4, inset)

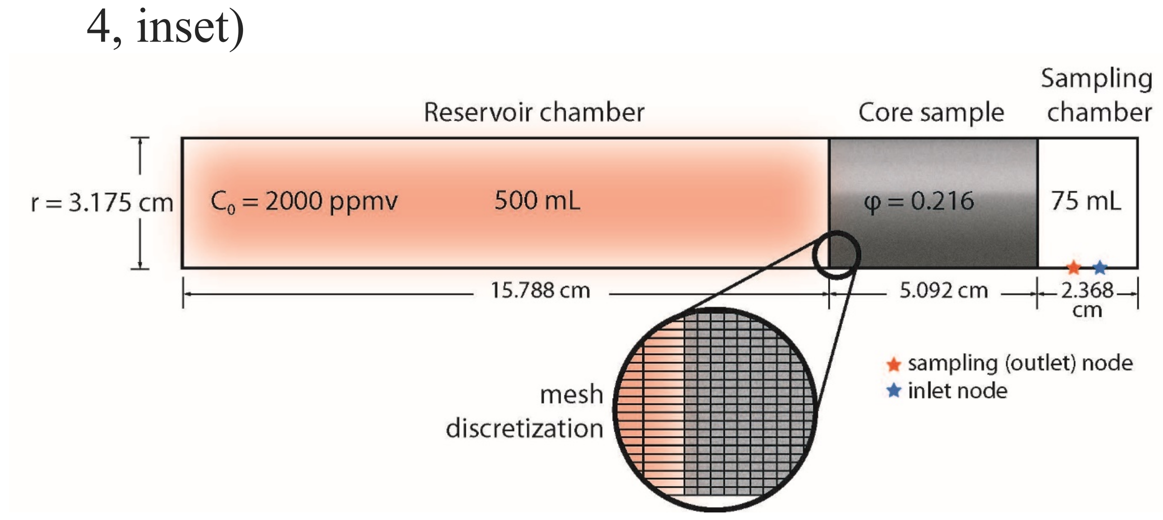

Fig. 5: Schematic of core diffusion cell setup showing the model domain with mesh discretization.

Material Properties

- Average tuff sample porosity (ϕ) = 0.216 m^3/m^3
- Intrinsic tuff permeability (k) = 1×10^{-14} m^2 (10 millidarcies)

Boundary and Initial Conditions

Initial conditions

- Pressure = atmospheric conditions everywhere
- Gas concentration = 2000 ppmv in reservoir chamber, 0 ppmv everywhere else

Boundary Conditions

- No-flux b.c.s (both flow and transport) on all exterior boundaries except for outflow (sampling) and inflow tubes (Fig. 4)
- Neumann constant fluid flux b.c.s for sampling and inflow nodes = $\pm 6.125 \times 10^{-10}$ kg/s of air; represents the average rate of suction from sampling chamber into mass spectrometer

Table 1. Selected solute parameters used in model.

	D_m (in an excess of water) [m^2/s]	$k°_H$ [mol/kg bar]
SF_6	1.20×10^{-09}	0.00024
xenon	1.84×10^{-09}	0.0043
krypton	1.47×10^{-09}	0.0025

Contact information

John P. Ortiz
Doctoral Student GRA
jportiz@lanl.gov
(+1) 505-665-5901

Results

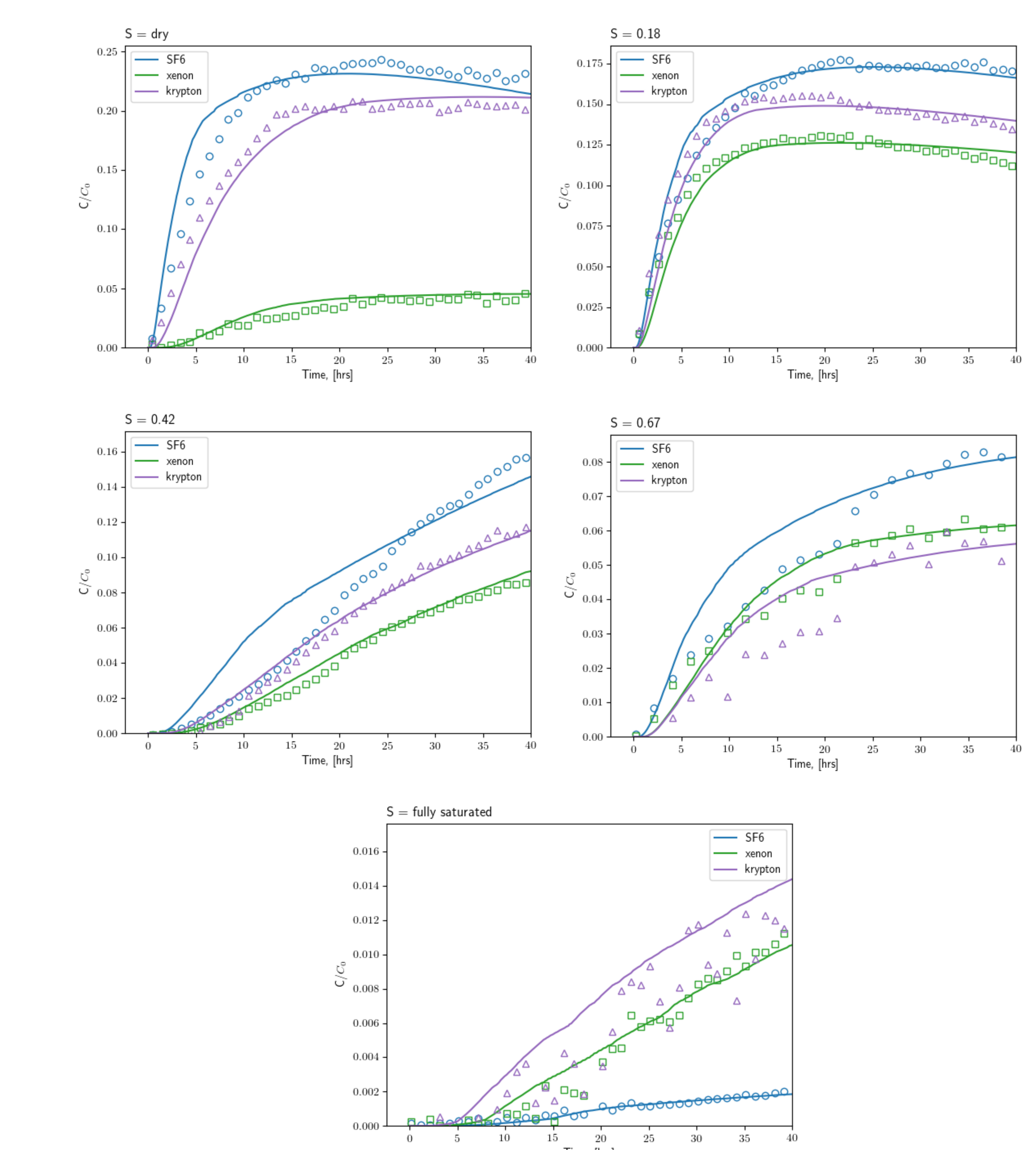

Fig. 6: Tracer breakthrough curves, where circles represent experimental results (not corrected for dilution) and lines are results produced by numerical simulation.

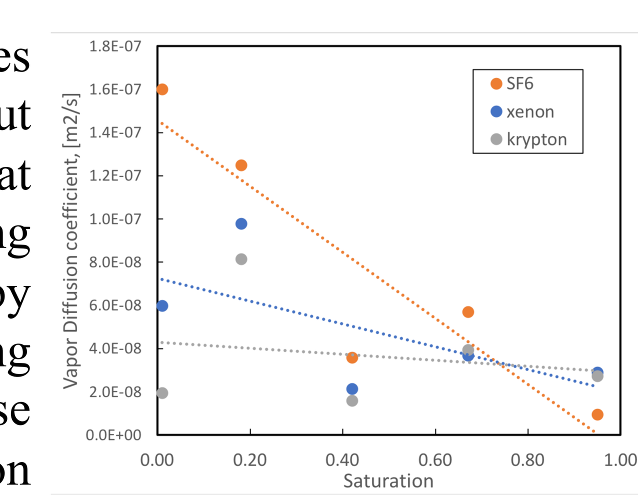

Fig. 7: Vapor D_m versus saturation for each gas.

Model generally reproduces experimental results, but indicates in some cases that more dilution is taking place than is prescribed by the mass spec sampling rate. Of note is the increase in vapor diffusion coefficient for both xenon and krypton going from 1% to 18% saturation, which contradicts the expected relationship. We aim to investigate whether this can attributed to selective adsorption of these gases by zeolites – if so, a small degree of saturation may fill these pores and cause reduced adsorption and, thus, faster transport.

References

Haynes, W. M. (1994). *CRC handbook of chemistry and physics*. Boca Raton: CRC Press.

Knox, J. B., Rawson, D. E., and Korver, J. A. (1965), Analysis of a groundwater anomaly created by an underground nuclear explosion, *J. Geophys. Res.*, 70(4), 823– 835.