

Sorption of Noble Gases in Porous Media and the Impact on Nuclear Explosion Signatures

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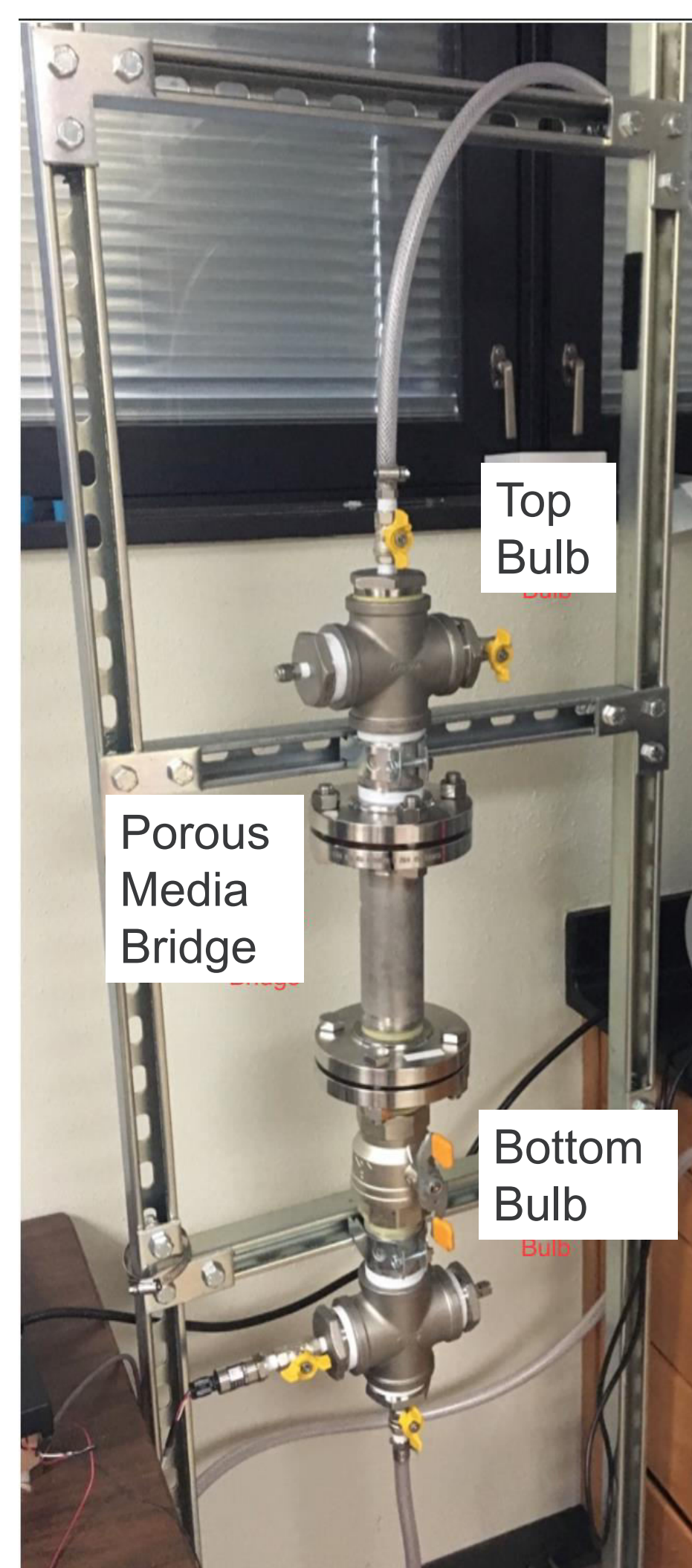
Radioactive isotopes of the noble gases xenon (Xe) and argon (Ar) are the “smoking gun” of an underground nuclear explosion in that detection of these isotopes is nearly unequivocal proof that a nuclear explosion occurred, especially if they are detected in the predicted ratios. Traditionally in the context of large-scale environmental transport, such as in the aftermath of a nuclear explosion, the noble gases are largely assumed to be non-reactive. However, results of past tracer experiments have illuminated the possibility that small pore-scale noble gas interactions in geologic media can have a significant aggregate effect on migration behavior over large distances. Presented here are ongoing efforts to understand the potential impact of noble gas sorption in porous geologic media on underground nuclear explosion signatures.

Goals and Objectives

The 2013 Noble Gas Migration Experiment yielded unexpected differences between Xe, Ar, and the chemical surrogate SF₆. Work presented here is aimed at trying to better understand these differences and their potential impact on nuclear explosion signatures and surface gas arrival times.

- ▶ Identify mechanisms that may selectively dilute Xe in contrast with other tracer gases (notably SF₆)
- ▶ Conduct controlled laboratory experiments to study the influence of subsurface water on Xe and SF₆ gas migration
- ▶ Measure effects of surface adsorption on tracer in various geologic materials and assess the potential impact of adsorption on field scale transport

Methods



Work presented here considers a simple, controlled flow-column experiment setup consisting of:

- ▶ A “two-bulb” transport vessel instrumented with sample ports, temperature, pressure and humidity sensors
- ▶ Gas Chromatographic-Mass Spectrometer used in-line for automated, regular sample analysis
- ▶ Bulbs have well-known volume
- ▶ Axial symmetry throughout the system
- ▶ Vertical orientation means no buoyancy driven flow

Experiments were performed on dry and wet compacted Ottawa sand.

- ▶ Known quantities of Xe, Kr, and SF₆ were injected at low pressure in bottom bulb (Ar analysis proved difficult and is not included)
- ▶ Concentration measurements were made at both top and bottom bulb over time
- ▶ Equilibrium concentration was estimated numerically using maximum likelihood estimation of exponential parameters

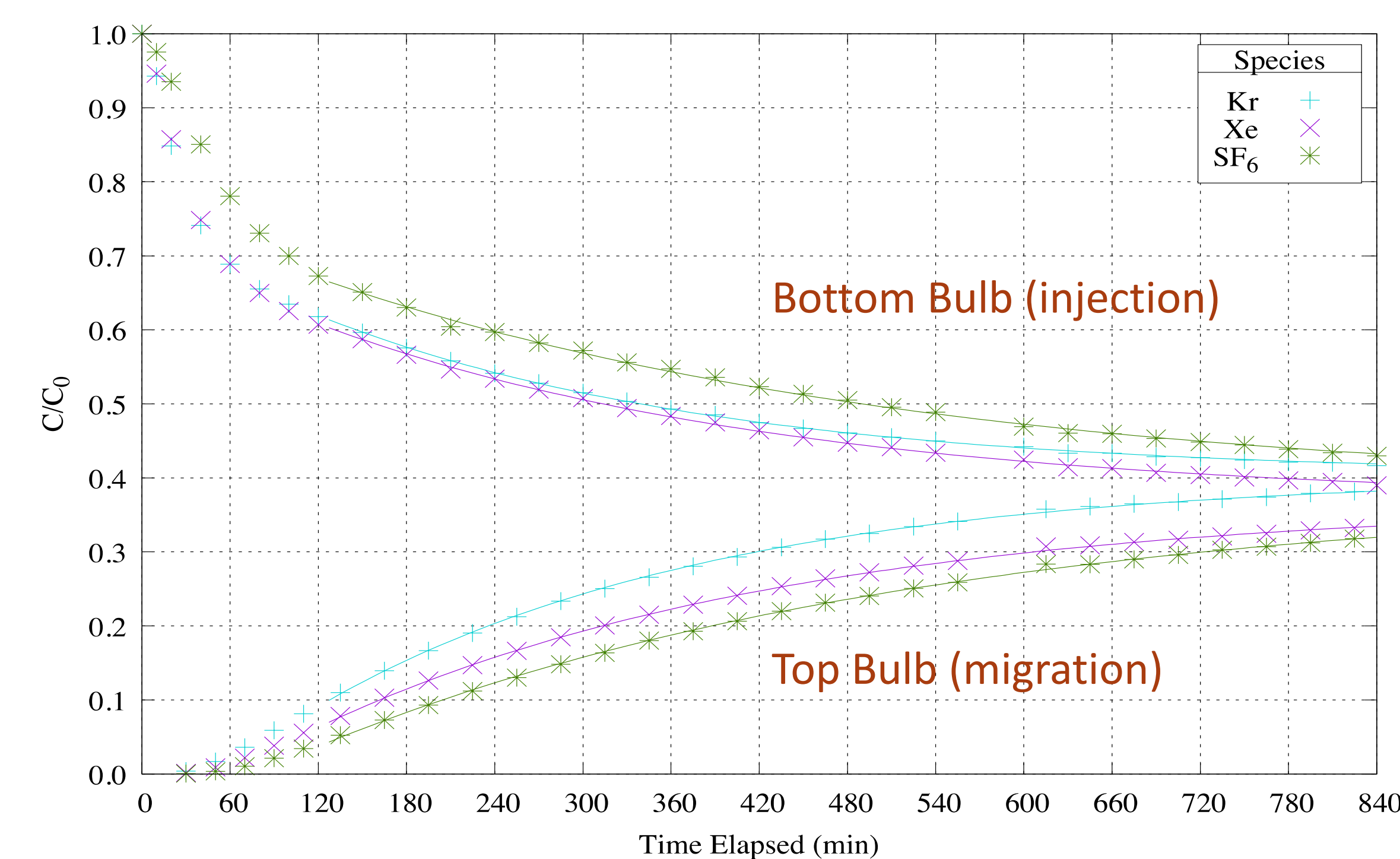
- ▶ Reactive-diffusive transport equation for gas transport for stagnant adsorbed and absorbed phases:

$$\frac{\partial c_i}{\partial t} = \frac{\mathcal{D}_i}{\kappa_i} \nabla^2 c_i - \lambda_i c_i$$

- ▶ Complete solution of the reactive-diffusive equation for a radioactive tracer where the Thiele modulus is greater than ~2

$$c_i(x, t) = \frac{c_0}{2} e^{-\sqrt{\frac{\kappa_i \lambda_i}{\mathcal{D}_i}} |x|} \left(\operatorname{erfc} \left(\frac{|x| \sqrt{\kappa_i}}{2\sqrt{\mathcal{D}_i t}} + \sqrt{\lambda_i t} \right) + \operatorname{erfc} \left(\frac{|x| \sqrt{\kappa_i}}{2\sqrt{\mathcal{D}_i t}} - \sqrt{\lambda_i t} \right) \right)$$

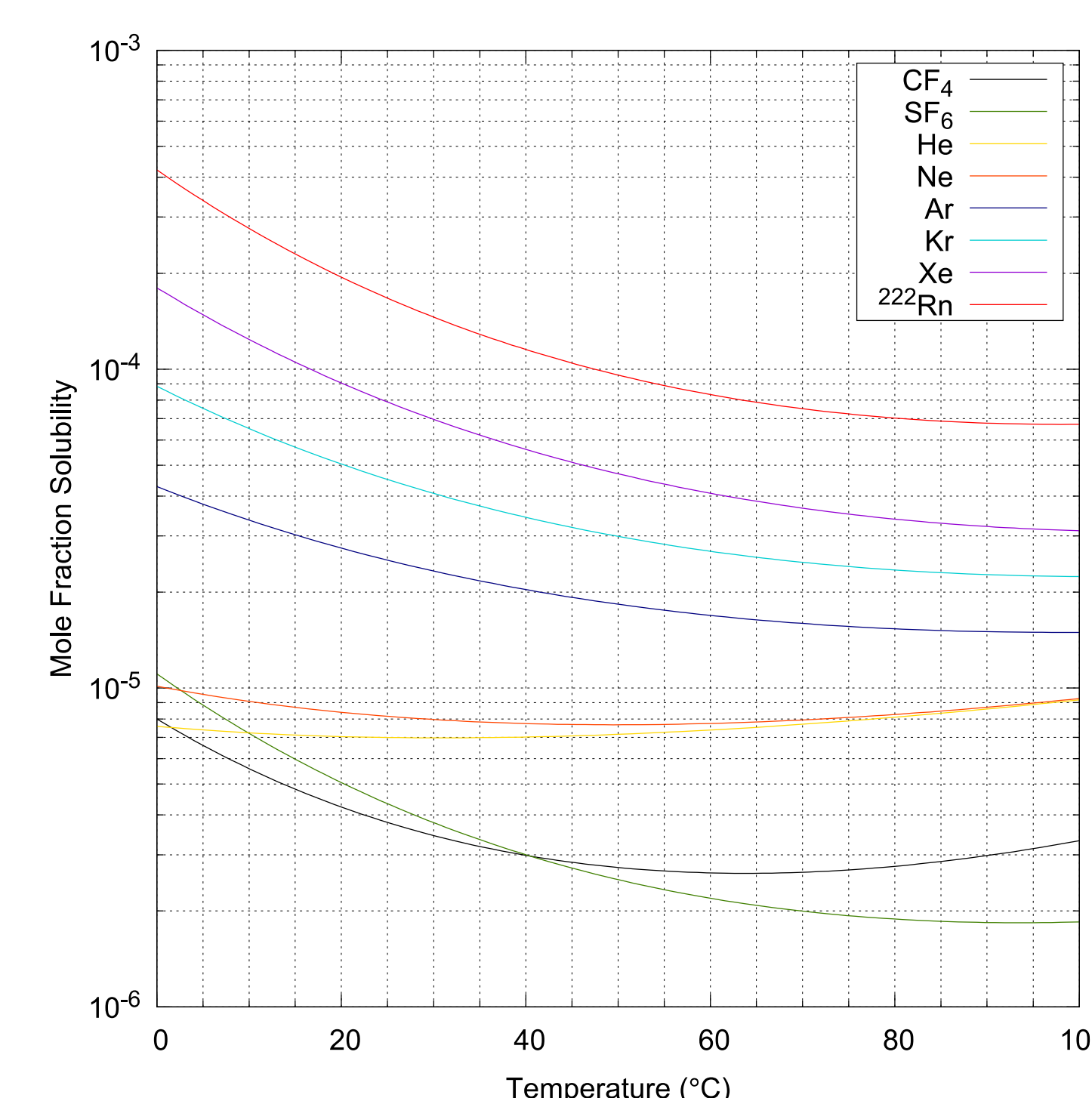
Results and Discussion



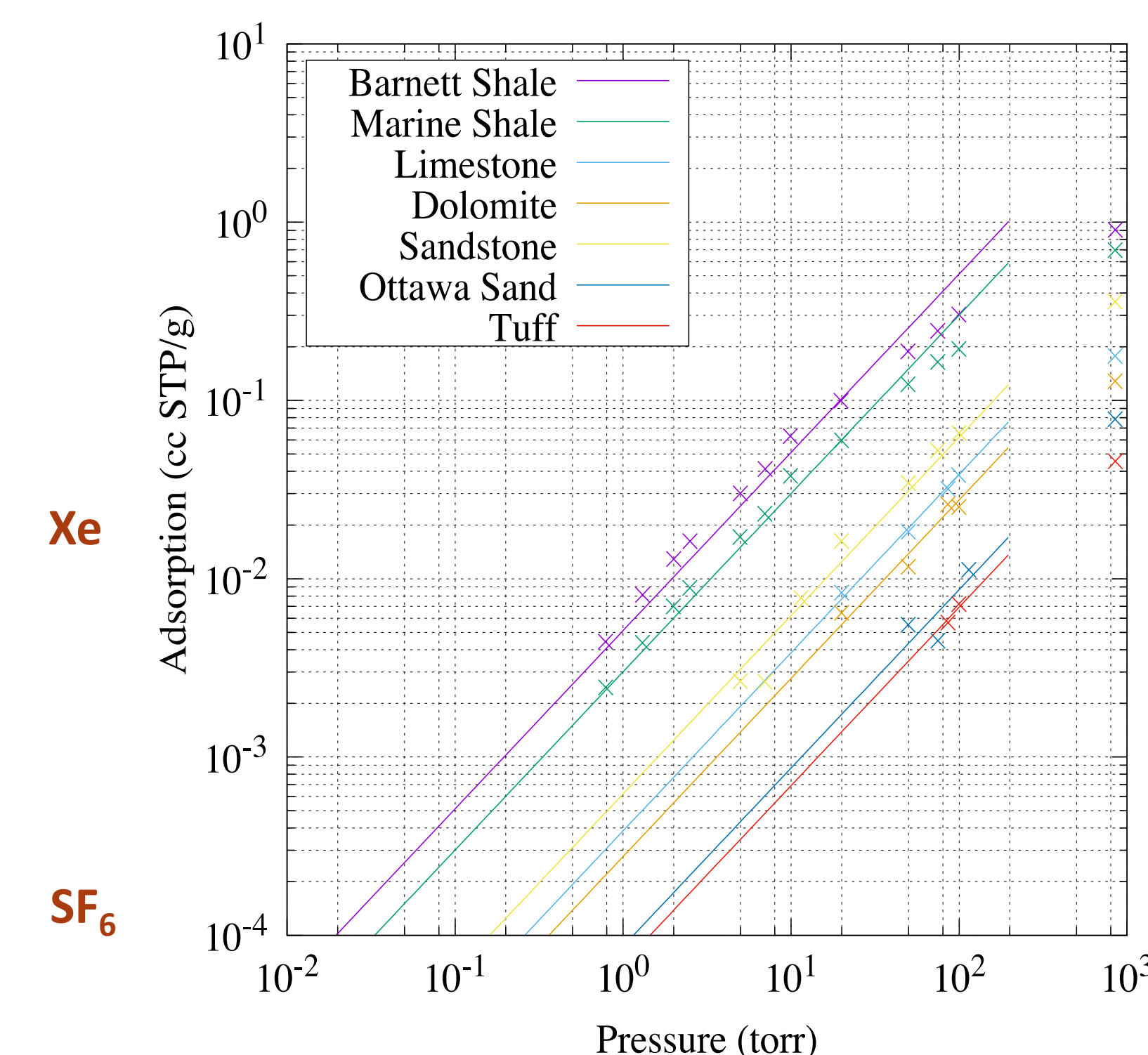
C/C_0 in contrast with time for the noble gases Kr, Xe, and stable SF₆ demonstrating the change in rate at which they reach asymptotical concentration

Results of two-bulb testing:

- ▶ Xe and the common surrogate SF₆ diffuse at substantially different rates, even in dry geology (so it is not only a difference in solubility)
- ▶ The difference in diffusion rates between Xe and SF₆ is greater in the presence of water
- ▶ Both Xe and SF₆ diffusion are slowed by water, but not at an equal rate



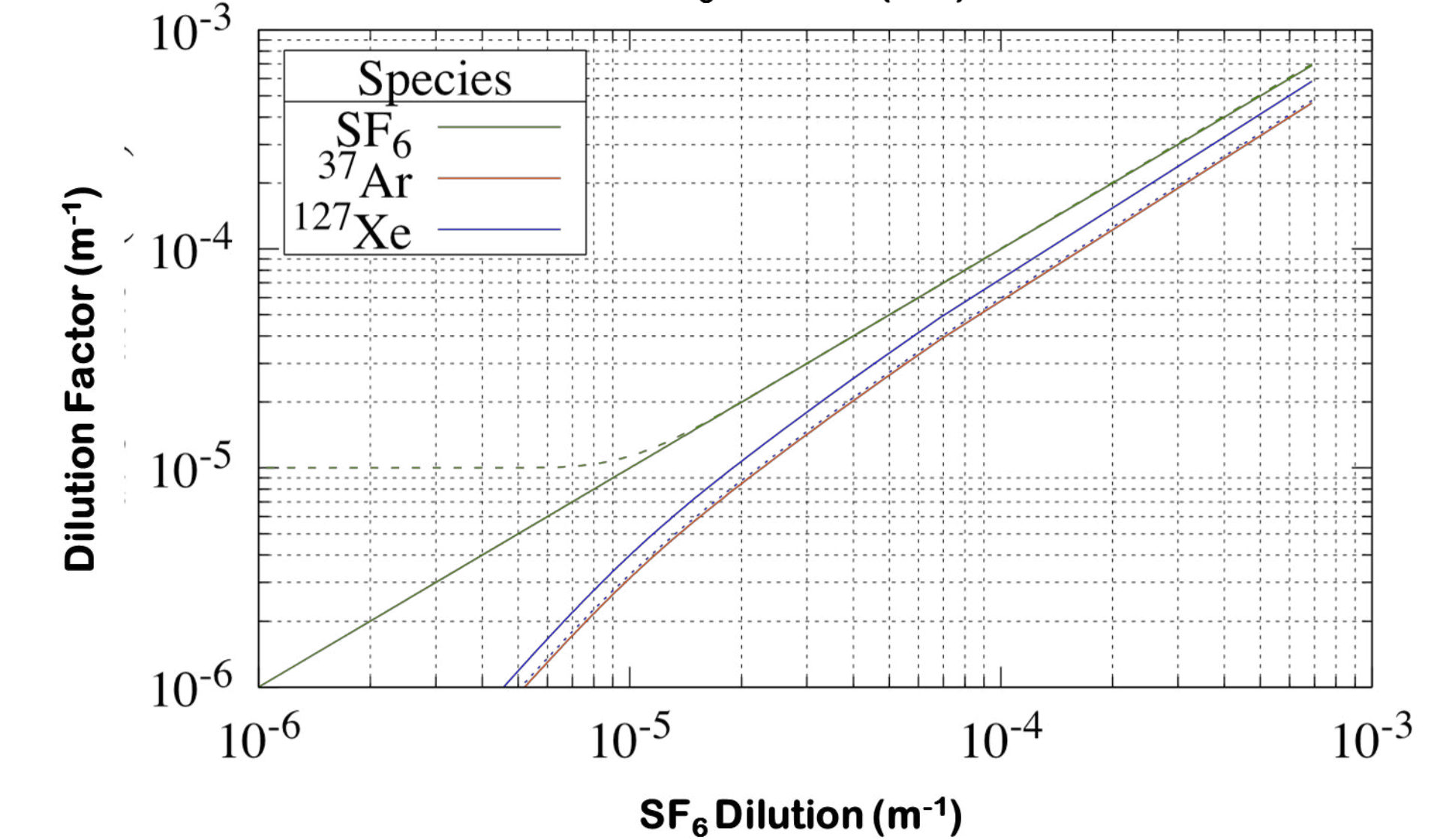
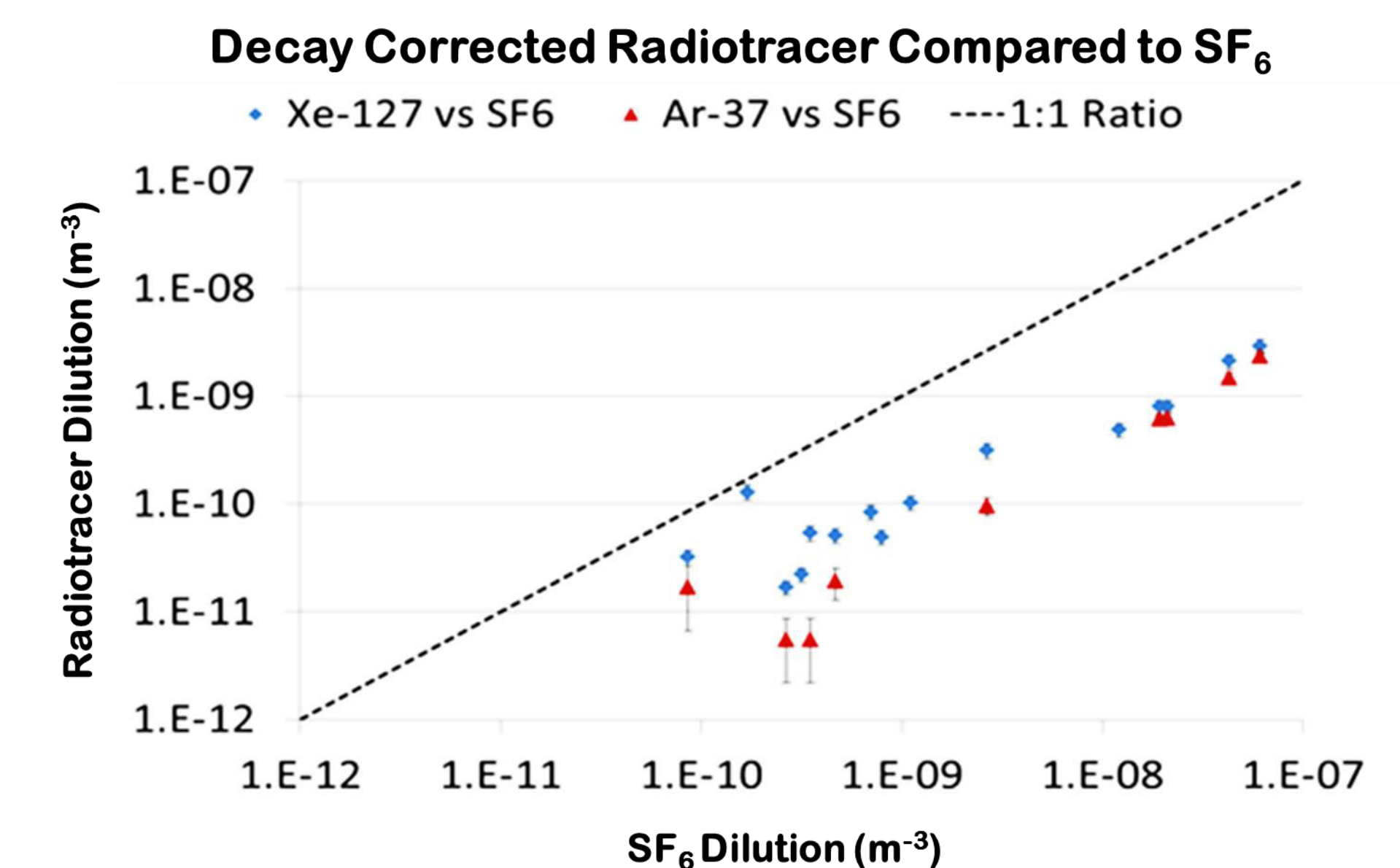
Tracer gas solubility in water



Xe isotherms in various geologies

- ▶ Noble gases are increasingly soluble with increasing atomic number
 - Fluorides (SF₆, CF₄) are poorly soluble by comparison
- ▶ Xe can exceed gas phase Xe in most materials in the adsorbed phase

Conclusions



Field results of the 2013 Noble Gas Migration Experiment (top) and ratio predictions from an analytical model of dilution based on diffusion, water absorption, and surface adsorption experiments (bottom)

- ▶ Substantial differences in noble gas and SF₆ diffusion was observed along with differential sorption effects
- ▶ Benchmarked adsorption effects were found to be significant enough to potentially explain the gas ratio deviations observed in 2013 Noble Gas Migration Experiment
- ▶ In an on-site inspection scenario under the Comprehensive Nuclear-Test-Ban Treaty, this difference in migration behavior could result in differences between noble gas arrival times of weeks
- ▶ Noble gases of interest should be used in treaty-relevant gas migration research whenever possible

Acknowledgements

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