

**MODELING OF RADIOXENON PRODUCTION AND RELEASE PATHWAYS**

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**ABSTRACT**

Models have been developed to calculate the diffusion of radioxenon through porous environmental media such as sand and soil. Initial experiments have also been conducted to determine the feasibility of radioxenon transport through soil and sand columns. The initial studies investigated method detection limits for xenon detection. These results show that stable xenon may be detected through prompt gamma activation analysis (PGAA) with detection limits in the 10-ppm range. New experiments are being designed to measure the diffusion of xenon in a nondestructive temporal manner. The initial goal is to measure an overall diffusion coefficient for each porous medium. The secondary goal is to measure the isotopic fractionation resulting from the process.

### **OBJECTIVES**

The two most recent nuclear weapon tests were conducted by the Democratic People's Republic of Korea (DPRK) in October 2006 and May 2009. Prior to the DPRK tests, India and Pakistan went through a series of nuclear tests in May 1998. It is important to recognize that all of the nuclear tests for more than 12 years have been conducted by countries developing nuclear weapons. The nuclear test yields were relatively small (Nuclear Explosion DataBase, 2010) and were conducted underground.

Radionuclide releases from these nuclear explosions have been minimal. Some of this may be attributed to the low yield of the devices tested, and some may be attributed to underground containment. Radionuclide releases from the DPRK tests have provided only radioxenon measurements from the first test conducted in 2006. Radioxenon measurements showed atmospheric concentrations that at times were close to background levels (Ringbom et al., 2009).

The objective of this work is to develop a better understanding of radioxenon transport after an underground nuclear explosion. The initial source signature of a nuclear weapon may be calculated with previously developed codes. However, there is a considerably larger error in the understanding of how the source signature is perturbed as radionuclides transport in an underground environment. Both chemical fractionation and isotopic fractionation may occur. This work focuses on the initial diffusion of xenon through porous media (e.g., soil). The transport times and isotopic fractionation are calculated, and plans are under way to measure this processing utilizing PGAA.

### **RESEARCH ACCOMPLISHED**

The research accomplished may be broken into three areas. The first is the calculation of a radionuclide source term from an underground nuclear explosion. The second area is calculation of diffusion for Xe isotopes. The last area is the initial calculations of detection limits for the measurement of Xe via PGAA.

#### **Source-Term Calculation**

As part of this work, models have been developed to calculate the radionuclides produced in a 1-kt nuclear explosion. The code MCNP is utilized to model the neutron transport, while ORIGEN 2.2 is utilized to calculate the production and decay of fission products, activation products, and transuranics resulting from the calculated neutron flux profile. MONTEBURNS is a perl script that couples the MCNP and ORIGEN 2.2 calculations.

The model used for the source-term calculation is fairly simple. A weapons-grade fissile sphere ( $k_{\text{eff}} \sim 1.0$ ) is surrounded by a soil or rock shell. For the uranium core case, an 8.7-cm-radius spherical core enriched to 94%  $^{235}\text{U}$  was utilized; for the plutonium case, a 5.1-cm-radius spherical core enriched to 93.80%  $^{239}\text{Pu}$  was used. MCNP was used to determine the thickness of soil or rock necessary to accurately model the attenuation and capture of neutrons within the media. For most of the soil and rock samples used, a thickness of 150 cm around the core was sufficient, such that less than 1% of neutrons escaped the system. For all cases studied, a soil/rock thickness of 2 m yielded on the order of 0.1% neutrons escaping.

Using MONTEBURNS, a database was compiled to include the resulting concentration and activity of every isotope calculated for both a  $^{235}\text{U}$  and a  $^{239}\text{Pu}$  sphere normalized to a burn of 1 kt. Two different soil compositions and three different rock compositions were utilized in order to model a range of environments and effectively determine a range of resulting activities for the isotopes of concern. The resulting concentrations were then decayed in ORIGEN 2.2 to yield activities at post-burn times from 5 minutes up to 100 days. The results for the radioisotopes of concern are summarized in Tables 1 and 2.

**Table 1. Activities in Ci of xenon radioisotopes following modeled 1-kt explosion at various post-burn decay times, assuming both uranium and plutonium cores**

		Initial	5 min	1 hr	10 hr	1 day	10 days	30 days	100 days
Uranium Core	<sup>131m</sup> Xe	1.361E-01	1.384E-01	7.948E-01	2.386E+01	5.972E+01	3.555E+02	2.638E+02	7.439E+00
	<sup>133</sup> Xe	1.964E+02	3.016E+02	6.481E+03	8.824E+04	1.709E+05	1.127E+05	8.120E+03	7.805E-01
	<sup>133m</sup> Xe	1.560E+03	1.566E+03	1.984E+03	7.379E+03	1.208E+04	1.743E+03	3.122E+00	7.441E-10
	<sup>135</sup> Xe	1.109E+05	1.804E+05	6.651E+05	2.000E+06	1.329E+06	1.843E-01	2.334E-17	0.000E+00
Plutonium Core	<sup>131m</sup> Xe	1.811E-01	1.846E-01	9.736E-01	2.705E+01	6.791E+01	4.147E+02	3.094E+02	8.740E+00
	<sup>133</sup> Xe	6.991E+02	8.669E+02	7.171E+03	8.886E+04	1.717E+05	1.134E+05	8.178E+03	7.861E-01
	<sup>133m</sup> Xe	5.446E+03	5.452E+03	5.831E+03	1.078E+04	1.490E+04	1.906E+03	3.425E+00	8.163E-10
	<sup>135</sup> Xe	2.648E+05	3.722E+05	9.930E+05	2.269E+06	1.459E+06	1.988E-01	2.518E-17	0.000E+00

**Table 2. Activities of <sup>37</sup>Ar in Ci resulting from modeled 1-kt explosion and subsequent neutron activation in various surrounding soil and rock concentrations**

	Soil Type	Initial	10 hr	1 day	10 days	30 days	100 days
Uranium Core	Wedepohl	1.014E+03	1.005E+03	9.937E+02	8.316E+02	5.597E+02	1.401E+02
	Taylor	9.455E+02	9.377E+02	9.270E+02	7.757E+02	5.222E+02	1.307E+02
	Mason	4.616E+02	4.579E+02	4.526E+02	3.788E+02	2.549E+02	6.380E+01
	Bowen	3.488E+02	3.460E+02	3.420E+02	2.862E+02	1.926E+02	4.820E+01
	Vinogradov	3.477E+02	3.448E+02	3.409E+02	2.853E+02	1.920E+02	4.805E+01
Plutonium Core	Wedepohl	1.760E+03	1.745E+03	1.725E+03	1.444E+03	9.717E+02	2.432E+02
	Taylor	1.502E+03	1.489E+03	1.472E+03	1.232E+03	8.294E+02	2.075E+02
	Mason	7.241E+02	7.182E+02	7.100E+02	5.941E+02	3.999E+02	1.001E+02
	Bowen	5.021E+02	4.980E+02	4.922E+02	4.119E+02	2.773E+02	6.938E+01
	Vinogradov	5.011E+02	4.970E+02	4.913E+02	4.111E+02	2.767E+02	6.925E+01

### Diffusion Coefficients for Xe Isotopes

Although the molecular transport of xenon *in* air would be more accurately modeled as a multicomponent diffusion process, we will use first a binary diffusion scenario to analyze the transport of xenon stable isotopes through a porous medium. Assuming that the molecular motion of each isotope is independent from the presence of other xenon nuclides makes it possible to employ Fick's first law of diffusion in our analysis. This assumption is valid when the amount of all but one species in the gas mixture is small (Deen 1998). Nitrogen will be used as the solvent gas in our analysis because it is an inert gas and is the most abundant element in air. For a nonpolar binary gas mixture at low pressure, the binary diffusion coefficient can be approximated using the correlation proposed by Bird et al. (2007). This formula is presented in Equation 1:

$$2.745 \times 10^{-4} \left[ \frac{T}{(T_{cXe} T_{cN_2})^{1/2}} \right]^{1.823} = \frac{p D_{XeN_2}}{(p_{cXe} p_{cN_2})^{1/3} (T_{cXe} T_{cN_2})^{5/12} \left( \frac{1}{M_{Xe}} + \frac{1}{M_{N_2}} \right)^{1/2}} \quad (1)$$

where

p = Total absolute pressure of mixture [atm]

T = Temperature of mixture [K]

$D_{XeN_2}$  = Binary diffusion coefficient of xenon in nitrogen or vice versa.

Table 3 provides a range of diffusion coefficients as calculated by Equation 1 for  $^{129}\text{Xe}$  and  $^{131}\text{Xe}$  in a binary mixture with  $\text{N}_2$ . Both  $^{129}\text{Xe}$  and  $^{131}\text{Xe}$  are stable isotopes with natural abundances of 26.4% and 21.2%, respectively (Molnar, 2004). These two isotopes are chosen for this model because of their higher detection probability through PGAA. Table 3 shows that there is about a 0.1% difference between the diffusion coefficients of  $^{129}\text{Xe}$  and  $^{131}\text{Xe}$ . While that is not a huge difference, it will result in differential transport over time and distance. Such differences will result in isotopic fractionation and changes in the radionuclide isotopic signature.

**Table 3. Binary diffusion coefficients of  $^{129}\text{Xe}$  and  $^{131}\text{Xe}$  in a binary gas with  $\text{N}_2$**

				$^{129}\text{Xe}$	$^{131}\text{Xe}$
Ma	Mb	T [K]	P [atm]	$D_{XeN_2} [\text{cm}^2 \text{s}^{-1}]$	$D_{XeN_2} [\text{cm}^2 \text{s}^{-1}]$
131	28.013	290	0.9	0.13502	0.13484
131	28.013	290	1	0.12152	0.12135
131	28.013	290	1.1	0.11047	0.11032
131	28.013	290	1.2	0.10127	0.10113
131	28.013	290	1.3	0.09348	0.09335
131	28.013	290	1.4	0.08680	0.08668
131	28.013	290	1.5	0.08101	0.08090
131	28.013	290	1.6	0.07595	0.07585
131	28.013	290	1.7	0.07148	0.07138
131	28.013	290	1.8	0.06751	0.06742
131	28.013	290	1.9	0.06396	0.06387

### Analytical Model

The analytical model for the experiment developed for this paper assumes the following:

- The diffusion of xenon isotopes occurs at uniform and constant pressure and temperature and small concentrations.
- The molecular diffusion of xenon isotopes takes place in only one direction (X direction).
- The diffusion coefficient of xenon in nitrogen is independent of the mixture composition.
- The external convection effects are negligible.
- Each xenon isotope is treated as a different species, but all isotopes' molecular diffusion is treated as in a binary system isotope-nitrogen.
- There is no chemical reaction that produces/consumes the xenon isotopes.
- Both xenon and nitrogen have ideal gas behavior.

Based on these assumptions, the molar average velocity is zero, and the equation of continuity of each isotope in molar basis is simplified to

$$\frac{\partial C_{\alpha Xe}}{\partial t} = D_{\alpha XeN_2} \frac{\partial^2 C_{\alpha Xe}}{\partial x^2} \quad (2)$$

where  $C_{\alpha Xe}$  is the molar concentration of the isotope  $\alpha Xe$ , and  $D_{\alpha XeN_2}$  is the effective diffusion coefficient. The effective diffusion coefficient takes into account the porosity and tortuosity of the porous medium. Assuming a porosity of 0.3, a tortuosity factor of 1.96 was calculated using the experimental correlation obtained by Matyka et al. (2008). From these two factors, the effective diffusion coefficients for  $^{131}Xe$  and  $^{129}Xe$  obtained are 0.01857 and 0.01854 [cm<sup>2</sup> s<sup>-1</sup>] respectively.

We will assume that the length of the duct where diffusion occurs is long enough to consider a semi-infinite medium. For this experiment, the initial and boundary conditions are

$$\begin{aligned} \text{IC:} \quad & t = 0, x > 0 \quad C_{\alpha Xe} = 0 \\ & t = 0, x = 0 \quad C_{\alpha Xe} = C_0 \end{aligned}$$

$$\begin{aligned} \text{BC1:} \quad & t > 0, x = 0 \quad \frac{\partial N_o}{\partial t} = A \cdot D_{\alpha XeN_2} \frac{\partial^2 C_{\alpha Xe}}{\partial x^2} \\ \text{BC2:} \quad & t, x \rightarrow \infty \quad C = 0 \end{aligned}$$

where  $N_o$  is the initial number of moles of the xenon isotope under analysis ( $N_o = C_o V_o$ ),  $C_o$  is the initial concentration of the isotope of interest at  $x = 0$ , and  $A$  is the cross-sectional area at  $x = 0$ .

Equation 3 presents the solution for these preliminary determinations of epicenter (PDEs), and initial and boundary conditions (Takeda et al., 2008):

$$\frac{C_{\alpha Xe}(x, t)}{C_o} = \exp\left(\frac{A^2 D_{\alpha XeN_2} t}{V_o^2} + \frac{Ax}{V_o}\right) \cdot \operatorname{erfc}\left(\frac{A \sqrt{D_{\alpha XeN_2} t}}{V_o} + \frac{x}{2 \sqrt{D_{\alpha XeN_2} t}}\right) \quad (3)$$

Figures 1 and 2 show the variation of the xenon concentration as a function of time and distance. Figures 3 and 4 show the variation of the ratio of molar concentrations  $^{131}Xe$  and  $^{129}Xe$  with respect to position and time, respectively, for diffusion of 0.0053 kg of natural xenon in a duct of 0.0070 m<sup>2</sup>. According to these results, an important ratio between the concentration of  $^{131}Xe$  and  $^{129}Xe$  could be measured at the diffusion. Conversely, at this location, the ratio  $C_{\alpha Xe}/C_o$  is very small. For example, at  $t = 1.2$  h, a ratio of  $C_{^{131}Xe}/C_{^{129}Xe} = 0.7995$  can be measured at  $x = 1.5$  m (the isotopes ratio in natural xenon is 0.803); however, at this location and time,  $C_{^{129}Xe}/C_o = 5.749 \times 10^{-5}$  and  $C_{^{131}Xe}/C_o = 5.725 \times 10^{-5}$ . These small concentrations will make the analysis of differential diffusion at the diffusion front difficult, as will be seen in the following section.

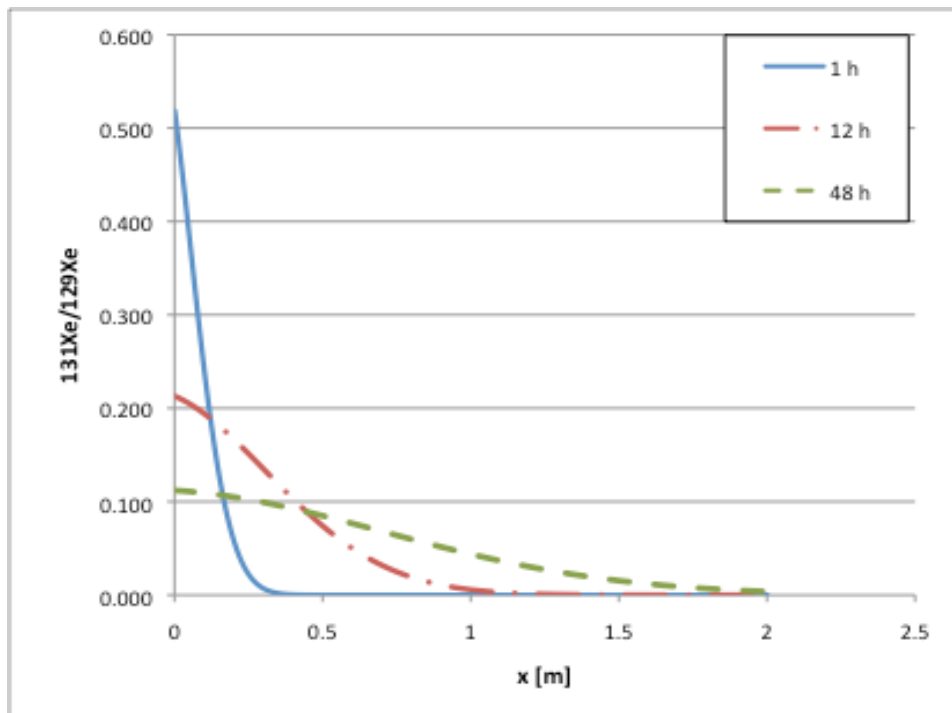


Figure 1. Variation of the Xe concentration profile at 1 h, 12 h, and 48 h.

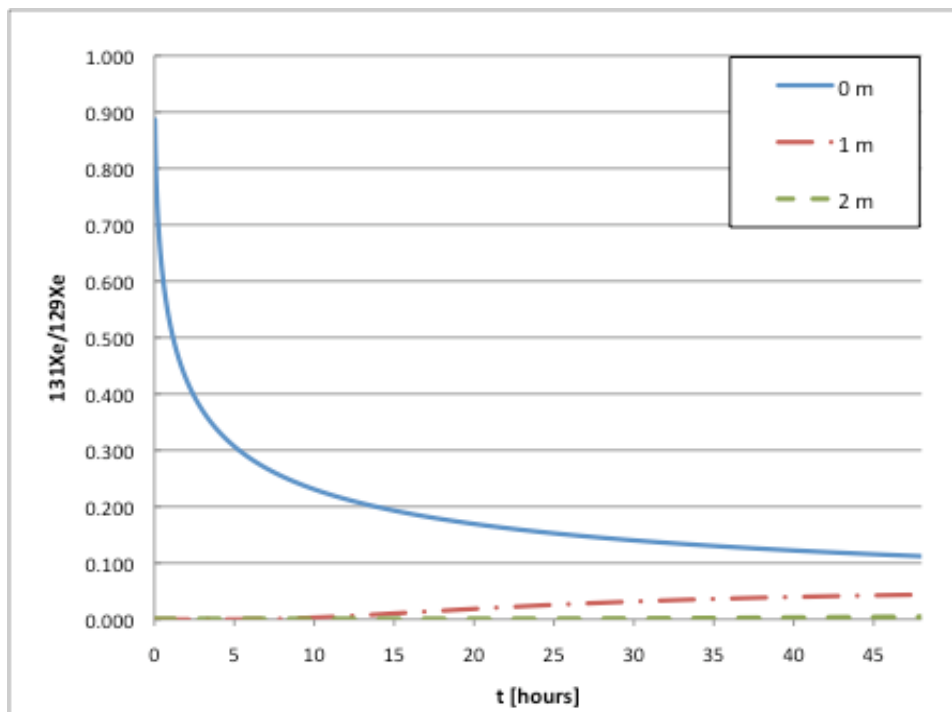


Figure 2. Variation of the Xe concentration with respect to time.

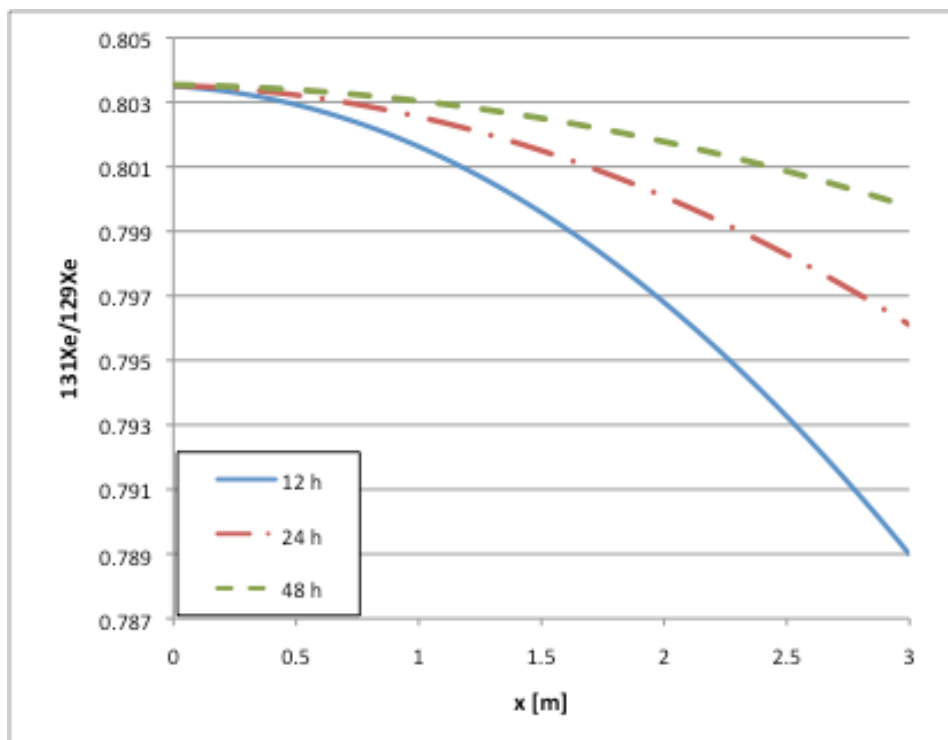


Figure 3. Variation of the ratio  $^{131}\text{Xe}/^{129}\text{Xe}$  with respect to position.

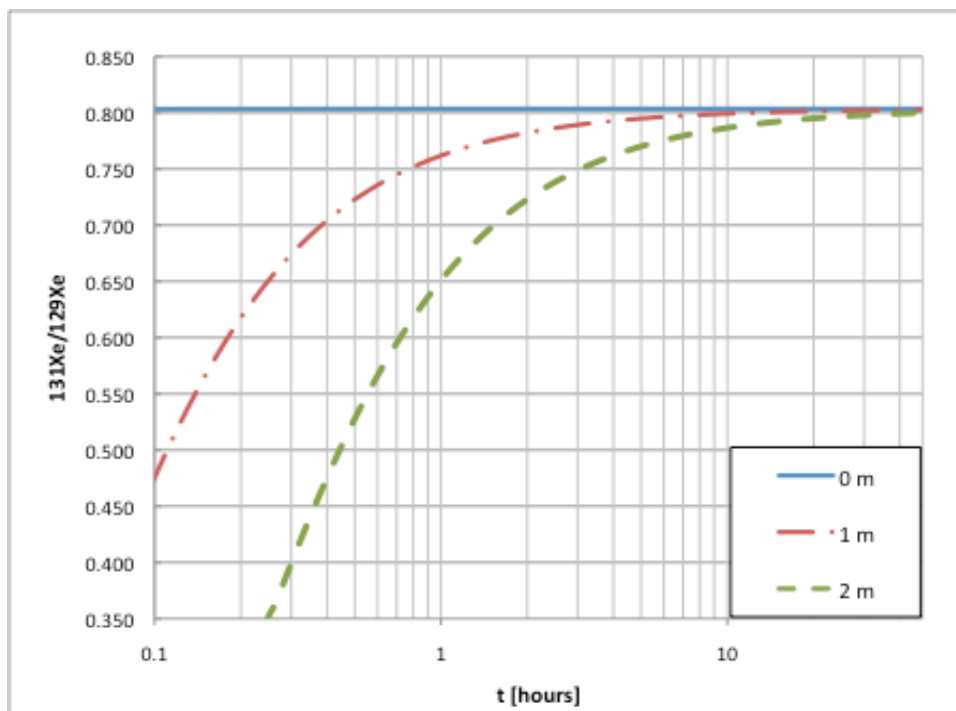


Figure 4. Variation of the ratio  $^{131}\text{Xe}/^{129}\text{Xe}$  with respect to time.

## Experimental Validation

This section evaluates the possibility of measuring the differential diffusion of  $^{131}\text{Xe}$  and  $^{129}\text{Xe}$  in a porous medium such as soil utilizing PGAA. First, the minimum amount of xenon needed for detection is calculated. Second, the effective volume of the sample is approximated. Finally, the minimum concentration of xenon required is obtained, and the feasibility of a xenon differential diffusion testing facility is discussed.

The minimum amount of xenon required to detect  $^{131}\text{Xe}$  and  $^{129}\text{Xe}$  is calculated from the molar detection limit ( $L_d$ ) of the equipment and the isotopic composition of the inert gas. Detection limits of  $6.3 \times 10^{-5}$  and  $7.7 \times 10^{-5}$  moles were calculated for  $^{129}\text{Xe}$  at 536 keV and  $^{131}\text{Xe}$  at 630 keV, for a total efficiency of 0.00145 (This efficiency was estimated from the counting rates analysis of 100% and 20% xenon samples during various time spans). The approximate percentages of  $^{131}\text{Xe}$  and  $^{129}\text{Xe}$  in natural xenon are 26.4% and 21.2%, respectively. There are two sample volumes that are important for a PGAA: the active and the effective sample volumes (Molnar, 2004). The following assumptions are made to calculate a first approximation of the effective volume of the sample:

- The attenuation of the neutron beam along the sample is negligible (both elements have small (n- $\gamma$ ) cross section).
- The detector has a collimator of 70 mm in diameter (the end-cap diameter of the detector has to be 83 mm or larger in order to have an efficiency of 65% [Rèvy et al., 2007]).

It has been reported that the neutron beam cross section at the sample location is about  $2.5 \times 1 \text{ cm}^2$  (Rèvy et al., 2007). From this, and from the assumptions listed before, the active volume of the sample could be approximated as  $6.5 \text{ cm} \times 2.5 \text{ cm} \times 1 \text{ cm}$ , i.e.,  $16.25 \text{ cm}^3$  (see Figure 5). Taking into account the geometric efficiency and the nonuniform intensity of the neutron flux, the effective sample volume is estimated to be  $12 \text{ cm}^3$ .

From the results calculated above, the minimum molar concentration of xenon required for a PGAA analysis is estimated as  $2.5 \times 10^{-5} \text{ mol}\cdot\text{cm}^{-3}$  (to detect 500 events in 1000 s). Assuming that at  $x = 0$  we have pure xenon gas at 1 atm (molar concentration of  $4.5 \times 10^{-5} \text{ mol}\cdot\text{cm}^{-3}$ ), the  $C_{a_{Xe}}/C_o$  minimum detectable is 0.556.

From Equation 3, the ratio of  $C_{a_{Xe}}/C_o = 0.556$  could be obtained at  $x = 0$  and  $t = 45 \text{ min}$ . Therefore, an accurate analysis at  $x = 0 \text{ m}$  would be difficult to conduct in a noninvasive way since the detection time for the  $L_d$  predicted is 1000 s, and the average rate of change of  $C_{a_{Xe}}/C_o$  from  $t = 30 \text{ min}$  to  $t = 45 \text{ min}$  is about  $-6 \times 10^{-5} \text{ 1/s}$ .

The analysis of a separated sample would allow us to conduct the PGAA for longer times with smaller detection limits and therefore more accuracy. For example, the  $L_d$  for  $^{129}\text{Xe}$  at 536 keV in a 7-h PGAA analysis is  $2.5 \times 10^{-6}$  moles, and the  $C_{a_{Xe}}/C_o$  minimum detectable in the gas mixture is 0.022. This ratio can be obtained from a sample extracted at  $x = 0$  after more than  $t = 150 \text{ h}$  from initiating the diffusion experiment; i.e., a sample of gas extracted during the first 150 h has larger concentrations of  $^{129}\text{Xe}$  than the  $L_d$ . Also, at  $t = 150 \text{ h}$ , the ratio  $C_{^{131}\text{Xe}}/C_{^{129}\text{Xe}}$  has changed by 0.07%. Although this change may seem very small, the analysis of the extracted gas may be prolonged in order to reach the desired accuracy without interfering with the diffusion experiment.



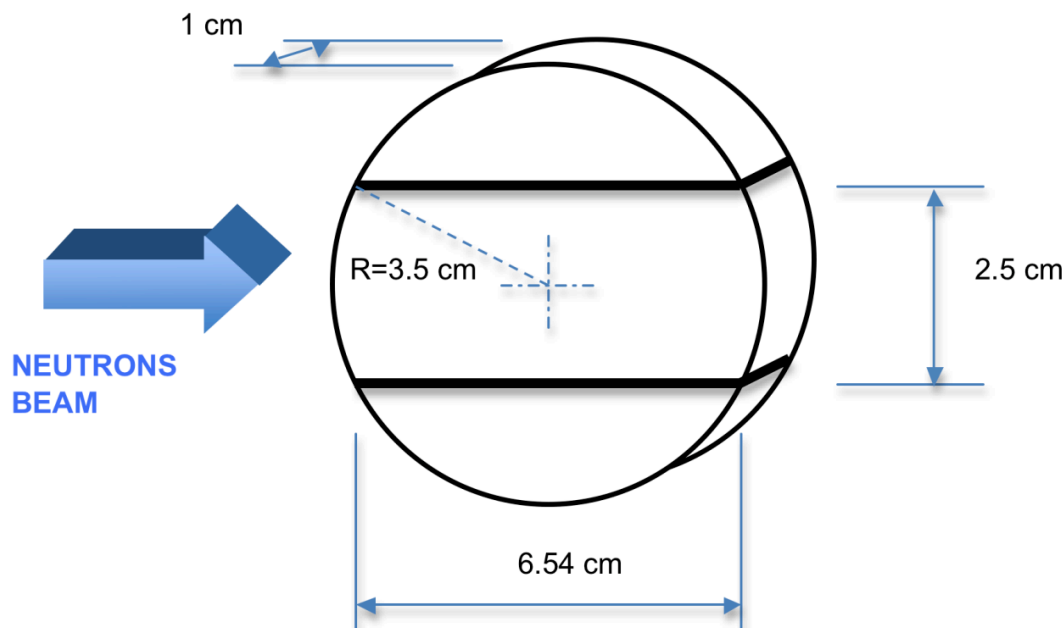


Figure 5. Effective sample volume, assuming a collimator of 7 cm in diameter.

## CONCLUSIONS AND RECOMMENDATIONS

The results shown herein lay a foundation for the modeling of Xe transport within a natural soil or sand matrix. The computational results indicate that isotopic fractionation occurs during such transport. This is fundamentally important to understanding the nuclear forensic signature of radioxenon emissions from an underground nuclear explosion.

Experiments are being set up to measure the Xe diffusion through sand and soil media utilizing PGAA. The goal would be to measure the isotopic fractionation that occurs during the diffusion process. However, initial calculations show that measuring the differential diffusion will be a challenge. At a minimum, diffusion coefficients for Xe and the models may be benchmarked with such experiments.

## ACKNOWLEDGEMENT

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