MODELING OF RADIOXENON PRODUCTION AND RELEASE PATHWAYS

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ABSTRACT

Since the Limited or Partial Test Ban Treaty in 1963, nuclear explosion tests have largely been conducted in underground facilities. One of the main motivations for underground testing is to contain the fission product releases from the nuclear explosion. To monitor the emissions from underground nuclear tests, the world community relies upon atmospheric monitoring for radioxenon among other technologies.

When an atmospheric radioxenon signal is observed, the isotopic ratios are examined to see if they match the expected values for nuclear explosions. These isotopic ratios are utilized to distinguish between nuclear explosion sources of radioxenon and other anthropogenic sources such as the commercial nuclear industry and the radiopharmaceutical industry. Current methods to predict the various isotopic ratio signatures have largely focused on modeling the production source. While this is a good first order approximation, it does not account for the chemical and isotopic fractionation that occurs during environmental transport of radioxenon and its parent radionuclides. This fractionation causes a significant change in the isotopic ratios from their point of creation to the point where they are collected in the atmosphere.

The goal of this work is to develop transport models for xenon and its parent radionuclides for underground nuclear explosions, commercial light water reactors, and radiopharmaceutical production facilities.

OBJECTIVES

Radioxenon may be produced through one of two methods. The first method is through the fission process and the second method is through an activation reaction. All xenon isotopes with mass greater than or equal to 129 are fission products (except ¹³⁰Xe). Very high mass radioxenons like ¹⁴³Xe and ¹⁴⁴Xe are produced in fission, but they have half-lives on the order of one second so they are not very relevant to this work. Stable xenon isotopes including ¹³¹Xe, ¹³²Xe, ¹³⁴Xe, and ¹³⁶Xe are also fission products but they are not relevant to this work since they do not decay. The main fission products of interest include ¹³¹Me, ¹³³Me, ¹³³Me, ¹³⁵Xe. The ²³⁵U fission yields and half-lives for these radionuclides are listed in Table 1. The combination of high fission yields and relatively short half-lives make these radionuclides prime candidates for detection of underground nuclear explosions.

	^{131m} Xe	^{133m} Xe	¹³³ Xe	¹³⁵ Xe					
Half-Life	11.9 days	2.19 days	5.24 days	9.09 hours					
²³⁵ U Fission Yield Per 100 Fissions									
Direct (%) – Thermal	3.48 x 10 ⁻⁷	1.89 x 10 ⁻³	6.66 x 10 ⁻⁴	7.85 x 10 ⁻²					
Direct (%) – Fast	2.41 x 10 ⁻⁷	4.23 x 10 ⁻³	1.45 x 10 ⁻³	1.20 x 10 ⁻¹					
Cumulative (%) – Thermal	4.05 x 10 ⁻²	1.95 x 10 ⁻¹	6.70	6.54					
Cumulative (%) – Fast	4.51 x 10 ⁻²	6.72	1.98 x 10 ⁻¹	6.60					

Figure 1 shows a diagram of the ¹³⁵Xe production process through nuclear fission. Similar production paths exist for each radioxenon fission product. The two sources for fission product atoms are direct production from fission and the decay of parent radionuclides along the same mass chain. The probability of direct production from fission is known as the direct yield. This value is listed in Table 2 for ²³⁵U. The direct yield is a function of both the atom that fissioned as well as the energy of the neutron initiating the fission. Fission yields from spontaneous fission are purely a function of the atom undergoing fission since there are no incident particles. The cumulative yield is the probability of production both from direct production and from the decay of parent fission products. The two loss mechanisms for atoms shown in Figure 1 are decay and neutron absorption. For the majority of these nuclides, decay is the primary loss mechanism. However, neutron absorption has a high probability of occurrence in atoms such as ¹³⁵Xe due to its exceptionally large 2,943,100 b cross-section.



Figure 1. Production process for ¹³⁵Xe through nuclear fission.

The radioxenon isotopic ratios provide signatures that may be utilized to distinguish between the production source. The goal of the work detailed in this proposal is to develop computational models that calculate the Xe fission product signatures from underground nuclear explosions, commercial nuclear reactors, and medical isotope production environments. These models will include proper treatment of the chemical fraction of Xe from its parent nuclides. This work will reduce the uncertainty associated in utilizing radioxenon ratios to differentiate between nuclear explosions and other anthropogenic sources.

RESEARCH ACCOMPLISHED

Underground Nuclear Explosions

Underground nuclear tests result in the release of xenon isotopes from direct fission yield as well as the release of isotopes that decay to xenon. In addition to isotope production, the rapid fission process releases a substantial amount of energy into the local environment that results in thermal and moisture gradients that will affect multiphase transport below ground. Modeling the movement of xenon in the subsurface is further complicated by the fact that it is mildly soluble in water (von Antropoff, 1910; Dresel and Waichler, 2004) and sorbs to the surfaces of many porous media (Maghusin et al, 1997; Filimonova et al., 2004). As a result, gaseous diffusion as well as transport of dissolved xenon, and its precursors, would need to be considered for a full treatment of xenon mobility after an underground nuclear test. In addition, the binary diffusion coefficients for xenon gas will be slightly different for each of the individual isotopes, owning to the differences in their masses. For the purpose of tracking isotope ratios, as would be important for monitoring under the Comprehensive Nuclear-Test-Ban Treaty, the decay of radioxenon would also need to be taken into consideration.

If gas phase diffusion were the only factor in the transport of xenon in the subsurface the movement of xenon could be modeled using Equation 1.

$$\frac{\partial C_i}{\partial t} = \nabla \cdot \left(D_i \nabla C_i + S_i - gC_i \right)$$
⁽¹⁾

Here C_i is the concentration of the i'th xenon isotope, D_i is its diffusion coefficient, S_i is the rate of xenon production from β^- decay of the respective iodine isotope and g is a function that takes into consideration the rate at which xenon isotopes are lost through sorption, dissolution in fixed subsurface moisture, or through decay. The concentration of iodine is itself time dependent and subject to transport.

Some data exists on the binary diffusivity of xenon and a value of $0.10 \text{ cm}^2 \text{ s}^{-1}$ has been reported (Dresel and Waichler, 2004). However, the diffusivity is a function of the temperature and pressure within a system and is generally given by Equation 2.

$$D_{T_{j}P_{j}} = D_{T_{j}P_{j}} \frac{p_{i}}{p_{j}} \left(\frac{T_{j}}{T_{i}}\right)^{3/2} \frac{\Omega_{i}}{\Omega_{j}}$$
(2)

Here T is temperature, p is pressure, Ω is the molecular collision integral and i, j are the respective states at which the diffusivity is desired [Bird *et al.*, 1960]. The diffusivity of two isotopes of mass $M_A > M_B$ is also predicted by classical theory to be proportional to:

$$(D_B/D_A) \sim (M_B/M_A)^{1/2}$$
 (3)

The solubility of xenon in water is small but potentially significant if the yield of a subsurface test is small and the device was deeply buried. A value of 0.10 at 293 K is reported in von Antropoff (1910) which is in keeping with the values available in the literature. This same paper also shows that the solubility of xenon is mildly temperature dependent, ranging from a low of 0.90 at 323 K to 0.23 at 273 K. The sorbtivity of xenon on subsurface media is less well established and additional experimental is required to establish saturation values.

If Equations 1, 2, and 3 where the only ones required to model xenon transport in the subsurface, the problem would already be complicated. Unfortunately, this is not the case. Because xenon and its precursors can dissolve in water, they can also be transported with it as it redistributes itself after a nuclear test, or as it moves through evaporative processes in the vadose zone. It is well established that the movement of a liquid phase in a porous medium that is itself subject to thermal gradients is given by Equation 4.

$$q_{\rm liq}/\rho = -D_{\theta \rm liq}\nabla\theta - D_{\rm T \, liq}\nabla{\rm T} - {\rm Ki}$$
⁽⁴⁾

Here q_{liq} is the liquid phase fluid flux, ρ is its density, $D_{\theta \ liq}$ is the Darcy diffusion coefficent, θ is the moisture content of the media, T is temperature, K is the hydraulic conductivity of the media and i is a unit vector. The parameter $D_{T \ liq}$ is the thermal liquid diffusivity which is equal to $K\sigma P_c$ where, σ is the surface tension of the water, and P_c is the capillary pressure (Philip, 1957).

Because thermal gradients are important in Equations 2 and 4 it is important to be able to model the subsurface temperature response to both the weapons test and other relevant heat sources (e.g., surface solar flux which can drive evaporation from the vadose zone). Energy transport within the subsurface can be well modeled using Equation 5.

$$C\frac{\partial T}{\partial t} = \nabla \cdot \lambda \nabla T + \rho L \nabla \cdot D_{\theta vap} \nabla \theta$$
(5).

Here C is the volumetric heat capacity of the soil, λ is the thermal conductivity of the soil, and L is the latent heat of evaporation, and $D_{\theta vap}$ is the water vapor diffusivity (Bird et al., 1960). Importantly, $D_{\theta liq}$, $D_{T liq}$, λ , and K are all strong functions of θ which makes Eqs (3–4) highly nonlinear and, in most cases, analytically untractable.

Models are being developed utilizing the above equations for the calculation of radioxenon transport resulting from an underground nuclear weapon test.

Commercial Nuclear Reactors

A collaboration has been established with Crystal River nuclear power station in Florida. Data mining has been initiated to obtain radioxenon release data from the nuclear power plant. Initial comparisons have been made between the reported release data and fuel cycle models run in ORIGEN. Figure 2 shows a comparison between the Crystal River 3 (CR3) radioxenon release data and the data resulting from ORIGEN runs. The ORIGEN runs represent a fuel burnups ranging from 0.1 MWd/MTU to 70,500 MWd/MTU. The CR3 release data is largely to the left of the radioxenon values modeled in ORIGEN due to the use of holding tanks at CR3. Noble gasses are accumulated and held for release. During this period, the radioxenon decays causing a decrease in both the ^{133m}Xe/^{131m}Xe and ¹³⁵Xe/¹³³Xe ratios.



Figure 2. Comparison of ^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe release data (CR3 Data) and results from ORIGEN-ARP calculations for plant operations . ORIGEN burnup values are in units of MWd/MTU.

Medical Isotope Production

An extensive modeling effort was initiated to calculate the source signatures resulting from medical isotope production. ORIGEN-ARP, ORIGEN 2.2, and Excel have been utilized. ORIGEN-ARP was utilized for commercial reactor modeling. ORIGEN 2.2 was utilized for cases where fast neutron fission is required to model. ORIGEN 2.2 also the capability to model the separation of daughter isotopes from their parents. Excel calculations were conducted from derived activation and decay equations. These equation are quite complex and are utilized for quality control checks on the ORIGEN calculations.

As a starting point the source signature of a nuclear weapon was modeled. Figure 3 illustrates this signature for both a ²³⁵U and a ²³⁹Pu device. There are two lines for each weapon signature. The line to the left represents the radioxenon signature resulting from complete separation of radioxenon from its parents immediately after the explosion. The line to the right represents the signature of radioxenon with the complete cumulative production from parent fission products. Since partial fractionation is a likely scenario for underground nuclear explosions, these two lines set the normal bounds for radioxenon source signatures from nuclear weapons tests. The results shown in Figure 3 are commensurate with those shown in Kalinowski and Tuma (2009).



Figure 3. Source signature of ²³⁵U and ²³⁹Pu weapons.

The next step was to model a default medical isotope production radioxenon signature. Production of medical isotopes through highly enriched uranium target activation was the method modeled since this is one of the most common production methods. An irradiation time of five days, a decay time of 12 hours, and subsequent separation and decay are modeled. The irradiation and decay values are reported to be standard times for such production (Nuclear and Radiation Studies Board, 2009). The radioxenon signature for the standard medical isotope irradiation is shown in Figure 4. The data show that the two source signatures are very close and likely not discernable in an environmental monitoring scenario.



Figure 4. Radioxenon source signature comparison between nuclear weapon and medical production.

A sensitivity study was conducted to determine the magnitude of the effect due to changes in the radioxenon ratios. Table 2 summarizes the results of the study. Of the six variables studied, only two showed a moderate impact on the radioxenon ratios: irradiation time and use of a holding tank. Figures 5 and 6 show the magnitude of changes in radioxenon signatures due to irradiation time and use of a holding tank, respectively. Note that the values do not change much due to the use of a holding tank for 30, 60, and 90 days due to the respective half-lives of the radioisotopes being examined.

Parameter	Range of Values	Effect on Radioxenon Ratios
Neutron Flux	10^{11} to 10^{15} n cm ⁻² s ⁻¹	Minimal
Cross Sections	BWR, PWR, and Pure Thermal	Minimal
Fuel Enrichment	19.9% to 95% ²³⁵ U	Minimal
Irradiation Time	5 to 21 days	Moderate
Decay Time	6 hours to 7 days	Minimal
Use of Holding Tank	0 to 90 days	Moderate

Table 2. Sensitivit	tv studv fo	r radioxenon	signature fro	om medical	isotope	production	facility.
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Figure 5. Radioxenon ratios resulting from changes in irradiation time.



Figure 6. Radioxenon signature resulting from use of holding tank.

CONCLUSIONS AND RECOMMENDATIONS

Initial studies have been conducted to examine the radioxenon signatures resulting from underground nuclear weapons tests, commercial nuclear reactor facilities, and medical isotope production facilities. Further modeling efforts will ensue to increase the fidelity of our understanding of these source signatures. Experiments will also be conducted to provide data that may improve the models.

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