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Uncertainty quantification for discrimination of nuclear events as violations of the comprehensive nuclear-test-ban treaty

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ABSTRACT

Enforcement of the Comprehensive Nuclear Test Ban Treaty (CTBT) will involve monitoring for radiologic indicators of underground nuclear explosions (UNEs). A UNE produces a variety of radioisotopes which then decay through connected radionuclide chains. A particular species of interest is xenon, namely the four isotopes ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe. Due to their half lives, some of these isotopes can exist in the subsurface for more than 100 days. This convenient timescale, combined with modern detection capabilities, makes the xenon family a desirable candidate for UNE detection. Ratios of these isotopes as a function of time have been studied in the past for distinguishing nuclear explosions from civilian nuclear applications. However, the initial yields from UNEs have been treated as fixed values. In reality, these independent yields are uncertain to a large degree. This study quantifies the uncertainty in xenon ratios as a result of these uncertain initial conditions to better bound the values that xenon ratios can assume. We have successfully used a combination of analytical and sampling based statistical methods to reliably bound xenon isotopic ratios. We have also conducted a sensitivity analysis and found that xenon isotopic ratios are primarily sensitive to only a few of many uncertain initial conditions.

1. Introduction

Radioxenon isotopes, ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe, are considered as possible indicators for detecting and discriminating underground nuclear explosions. Because of their short half-lives, their background concentrations are extremely low. Xenon concentrations and isotopic ratios have been studied for distinguishing nuclear explosions from civilian nuclear application sources (Carrigan et al., 1996; Kalinowski et al., 2010; Kalinowski, 2011; Carrigan and Sun, 2014; Sun and Carrigan, 2014; Sun et al., 2015). Radioactive decay and ingrowth have been modeled numerically (Kalinowski et al., 2010) and analytically (Sun et al., 2015) by solving systems of first-order ordinary-differential equations (ODEs). The resulting solution of the first-order reaction networks (Fig. 1) has been further used to calculate the Multi-Isotope Ratio Chart (MIRC, Kalinowski et al., 2010). An arbitrary line is drawn on the log–log MIRC to separate the ratio correlations between an underground nuclear explosion (UNE) and a civilian application. Although numerical methods are available for integrating coupled ODEs, the stiffness of ordinary differential equations due to greatly differing decay rates may require extra computational effort to obtain solutions.

Bateman (1910) first derived an analytical solution of the first-order sequential reactions without considering branching and converging connections. Since then, many papers have been published in analytical solution development for sequential decay-ingrowth in batch-reactor systems (e.g., Cetnar, 2006; Yuan and Kernan, 2007; Zhou et al., 2015) and in studying radionuclide transport in the subsurface (e.g., van Genuchten, 1985; Sun et al., 1999; Slodička and Balážová, 2008; Clement et al., 1998; Dai et al., 2012). However, the uncertainty propagation from parent to daughter species has not been studied in the literature.

The distribution of isotopes characterizing the independent yield depends on the direct probability of forming a nuclide after prompt neutron emission and before its decay. Measuring the independent yield is difficult, especially for nuclides with short half-life. Although measurement uncertainties of independent yields have been reported by England and Rider (1994), the impact of those uncertainties on the MIRC remains unknown. Uncertainty quantification (UQ) of xenon isotopic ratios has not yet been done

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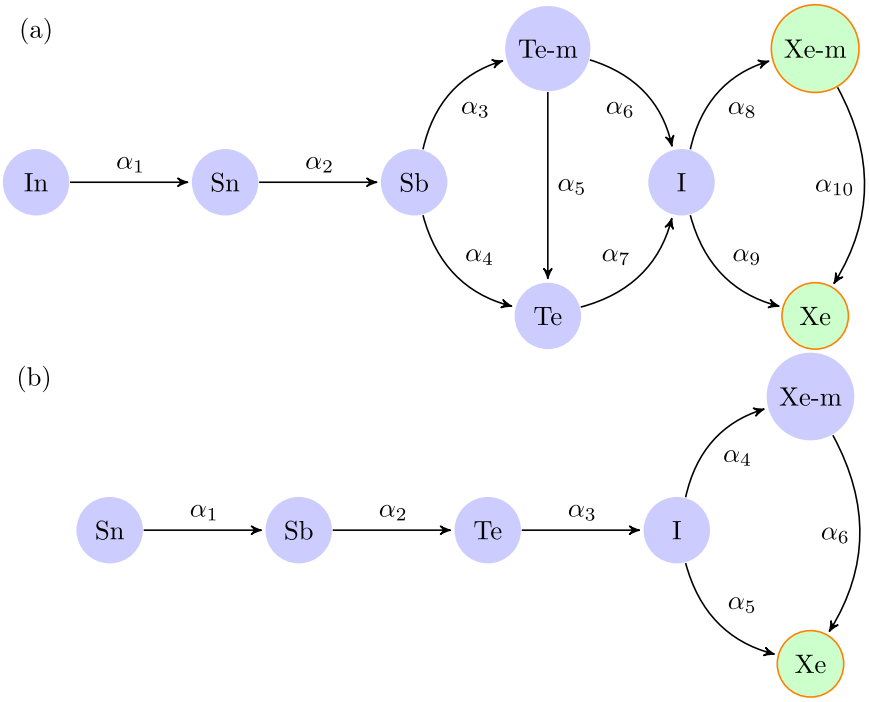

Fig. 1. Radioactive decay chains for xenon 131/133 and xenon 135 isotopes. (a) 131 and 133 decay chains. (b) 135 decay chain. α denotes branching factor. Half lives and independent yields are given in Table 1.

for confidence building on the MIRC. In this paper, we derive an analytical UQ form of xenon isotopic ratios and create a Multi-Isotope Ratio Chart (Kalinowski et al., 2010), which takes into account the uncertainties in both xenon isotopic ratios resulting from substantial uncertainties in the independent yields of a UNE. We compare the analytical UQ form with sampling-based UQ methods for studying the uncertainty propagation from independent yields (at t=0) to xenon ratios at some later time. Using one-standard deviation of xenon isotopic ratios, that is expressed analytically as a function of uncertainties of independent yields, we quantitatively define the discrimination line on the MIRC.

2. Materials and methods

2.1. Coupled first-order reactions

A typical system of coupled first-order reactions is described using coupled ordinary differential equations. For example, the rate equations of 135 decay chain (Fig. 1b) can be expressed as

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ c_6 \end{bmatrix} = \begin{bmatrix} -k_1 & 0 & 0 & 0 & 0 & \\ \alpha_1 k_1 & -k_2 & 0 & 0 & 0 & 0 \\ 0 & \alpha_2 k_2 & -k_3 & 0 & 0 & 0 \\ 0 & 0 & \alpha_3 k_3 & -k_4 & 0 & 0 \\ 0 & 0 & 0 & \alpha_4 k_4 & -k_5 & 0 \\ 0 & 0 & 0 & \alpha_5 k_4 & \alpha_6 k_5 & -k_6 \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ c_6 \end{bmatrix} = A\mathbf{c} \tag{1}$$

where c_i, i=1,⋯,6, denote concentrations of Sn, Sb, Te, I, Xe-m, and Xe in 135 decay chain, k_i, i=1,⋯,6, are their decay rates, and α_i,i=1,⋯,6, are branching factors. The reaction matrix A in Eq. (1) can be decomposed analytically as A=$S\Lambda S^{-}$ (see Appendix B of Sun et al., 2015). The analytical solution of Eq. (1) is derived by Sun et al. (2015). Using the same method of matrix decomposition, generalized closed-form solutions of radionuclide concentrations with user-defined reaction networks and unlimited numbers of isotopes and decay chains are derived in Sec. 2.2 and analytical statistics of concentrations and isotopic ratios are derived in Sec. 2.3.

2.2. General reaction solver

In order to address the problem of ratios of radioisotopes in xenon 131, 133, and 135 decay chains, a generalized computer code was developed in Python for analytical solutions to irreversible and first-order reaction systems. Consider a set of M independent nuclear decay chains consisting of a total of N species. Each of these reaction chains may have both branching points, where one parent species splits into two or more daughter species, and converging points, where multiple parents converge to the same daughter species. Each independent reaction can be characterized by a reaction matrix A_k. We then define the joint reaction matrix as the block diagonal matrix A, which carries each independent reaction matrix A_k as a block,

$$A = A_1 \oplus A_2 \oplus \cdots \oplus A_M = \begin{pmatrix} A_1 & 0 & \cdots \\ 0 & \ddots & 0 \\ \vdots & 0 & A_M \end{pmatrix}. \tag{2}$$

Writing the set of reactions in terms of a single block matrix will allow a more unified treatment of an arbitrary number of reaction chains which escapes the notational complexity of indexing reactions in addition to reaction rates and branching factors. The concentrations of each of the N total species as a function of time are then given in the vector $\mathbf{c}(t)$, which satisfies the differential equation

$$\frac{\mathrm{d}\mathbf{c}}{\mathrm{d}t} = A\mathbf{c}, \tag{3}$$