Gas-magma interactions in nuclear cavities and their effects on the xenon isotopes

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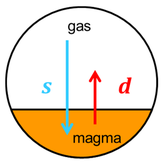
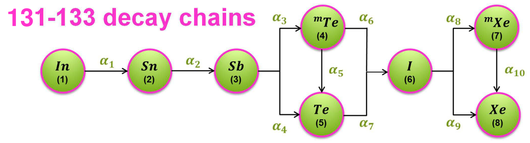
1. Rationale

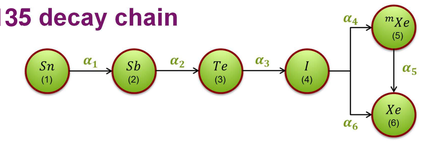


Along the **3 decay chains** leading to the 4 xenon isotopes of interest for the discrimination of underground nuclear explosions, the radionuclides can **dissolve (s)** in the **magma formed by melted rocks** in a nuclear cavity and **diffuse back (d)** to the gas phase. Numerical modeling and sensitivity analysis are used to quantify the effects of these gas-magma interactions on the source term activity and the xenon isotope ratios.

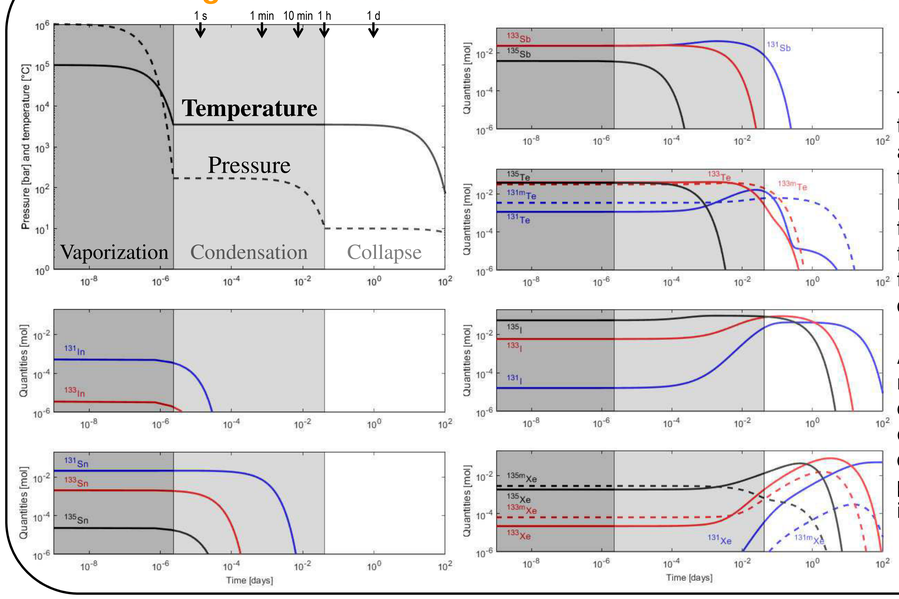
131-133 decay chains



135 decay chain



2. Gas-magma interactions must be considered from indium to xenon



Typical time evolution of the physical parameters of a nuclear cavity [1] and of the quantities of radionuclides, from indium to xenon, calculated as if they do not interact with the magma (see further down).

A magmatic phase (melted rocks formed after condensation) is present earlier than the complete decay of indium and is still present during the ingrowth of xenon.

3. Gas-magma interactions coupled to the Bateman [2] equations

$$\frac{dN}{dt} = AN$$

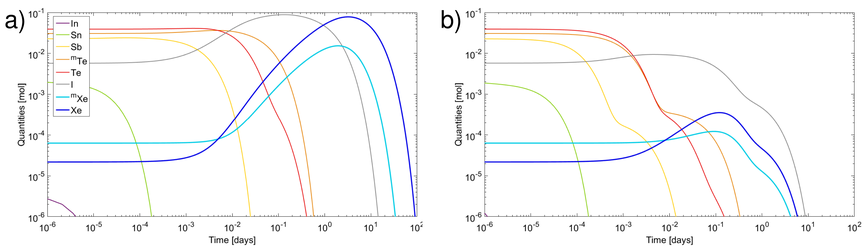
A =

-(k₁ + s₁)	d₁	0	0	0	0	0	0	0	0	0	0	0	0	0	0	In (gas)
s₁	-(k₁ + d₁)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	In (magma)
α₁ k₁	0	-(k₂ + s₂)	d₂	0	0	0	0	0	0	0	0	0	0	0	0	Sn (gas)
0	α₁ k₁	s₂	-(k₂ + d₂)	0	0	0	0	0	0	0	0	0	0	0	0	Sn (magma)
0	0	α₂ k₂	0	-(k₃ + s₃)	d₃	0	0	0	0	0	0	0	0	0	0	Sb (gas)
0	0	0	α₂ k₂	s₃	-(k₃ + d₃)	0	0	0	0	0	0	0	0	0	0	Sb (magma)
0	0	0	0	α₃ k₃	0	-(k₄ +s₄)	d₄	0	0	0	0	0	0	0	0	Te-m (gas)
0	0	0	0	0	α₃ k₃	s₄	-(k₄ + d₄)	0	0	0	0	0	0	0	0	Te-m (magma)
0	0	0	0	α₄ k₃	0	α₅ k₄	0	-(k₅ + s₅)	d₅	0	0	0	0	0	0	Te (gas)
0	0	0	0	0	α₄ k₃	0	α₅ k₃	s₅	-(k₅ + d₅)	0	0	0	0	0	0	Te (magma)
0	0	0	0	0	0	α₆ k₄	0	α₇ k₅	0	-(k₆ + s₆)	d₆	0	0	0	0	I (gas)
0	0	0	0	0	0	0	α₆ k₄	0	α₇ k₅	s₆	-(k₆ + d₆)	0	0	0	0	Xe-m (gas)
0	0	0	0	0	0	0	0	0	0	α₈ k₆	0	-(k₇ + s₇)	d₇	0	0	Xe-m (magma)
0	0	0	0	0	0	0	0	0	0	0	α₈ k₆	s₇	-(k₇ + d₇)	0	0	Xe (gas)
0	0	0	0	0	0	0	0	0	0	α₉ k₆	0	α₁₀ k₇	0	-(k₈ +s₈)	d₈	Xe (magma)
0	0	0	0	0	0	0	0	0	0	0	α₉ k₆	0	α₁₀ k₇	s₈	-(k₈ + d₈)	

$$\begin{cases} \dfrac{\partial N^{gas}}{\partial t} = -sN^{gas} + dN^{magma} \\ \dfrac{\partial N^{magma}}{\partial t} = -dN^{magma} + sN^{gas} \end{cases}$$

Reaction matrix A of 131 and 133 chains with row by row writings of the quantity N of each radionuclide i, respectively for gas and magma, from indium (In) to xenon (Xe), taking into account classical radioactive decay and ingrowth (with decay rate k_i and branching factor α_i), as well as sink-source effects due to solubility (s) from the gas phase to the magma and back-diffusion (d), as detailed in the set of the two coupled equations (left).

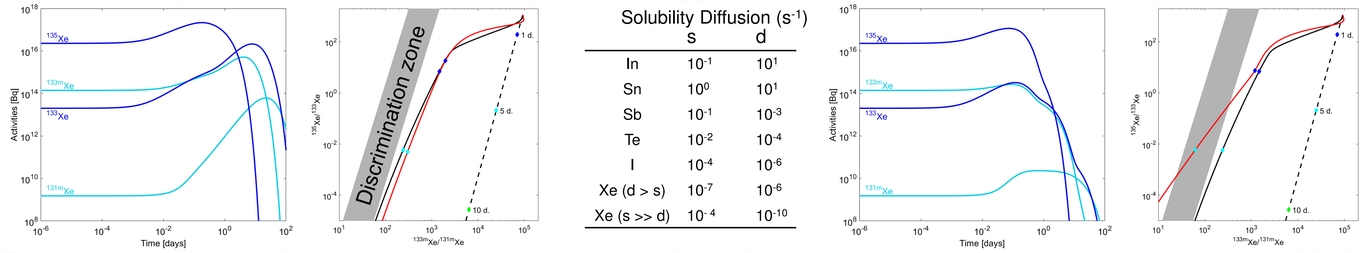
4. Solubility in magma controls radionuclides quantities available in gas phase



Time evolution of the quantity (mol) of the radionuclides indium to xenon in the gas phase of a nuclear cavity, calculated for the 133 chain using MATLAB ode23s for 10 kT ^{235f}U fission. a) Without gas-magma interaction, results are similar as those obtained by Sun et al. (2014). b) Gas-magma interactions may seriously lower the amount of xenon in the gas phase (s and d values are given in the table below, with s >> d for Xe).

a) Without gas-magma interaction, results are similar as those obtained by [3]. b) Gas-magma interactions may significantly lower the amount of xenon in the gas phase (s and d values are given in the table below, with s >> d for Xe).

5. Solubility and back-diffusion may lead to apparent fractionation of Xe isotopes

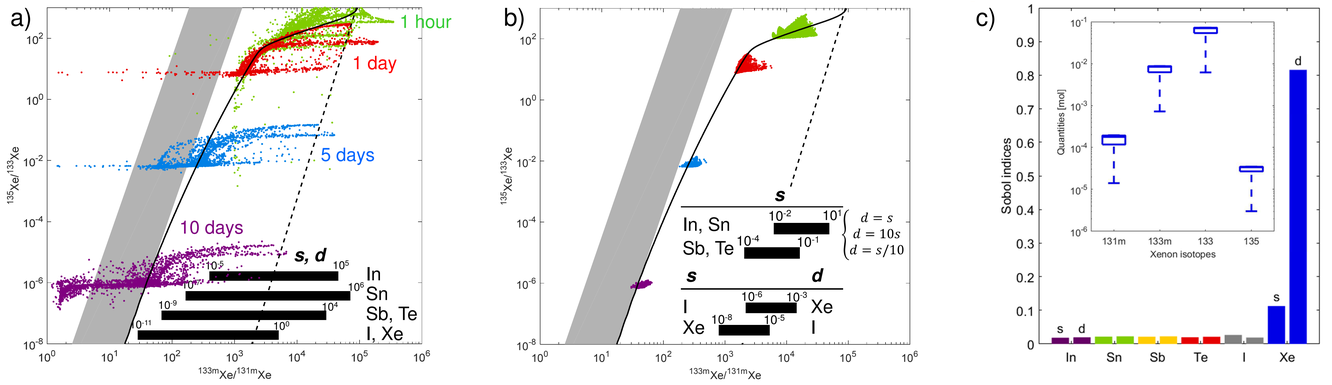


Solubility Diffusion (s^{-1})

	s	d
In	10^{-1}	10^{1}
Sn	10^{0}	10^{1}
Sb	10^{-1}	10^{-3}
Te	10^{-2}	10^{-4}
I	10^{-4}	10^{-6}
Xe (d > s)	10^{-7}	10^{-6}
Xe (s >> d)	10^{-4}	10^{-10}

Time evolution of **activities** (Bq) for the 4 xenon isotopes and their **activity ratios** compared to the case without gas-magma interaction (**black**): (left) Xe produced in the magma diffuses back to the gas phase while Xe produced in the gas phase has low affinity for the magma (d>s), (right) reverse situation where Xe is trapped in the magma (s >>d). Discrimination of military or civilian activity after [4].

6. Despite undetermined values, solubility-diffusion shall not hamper discrimination



10,000 simulations of activity ratios: a) without constraint on *s* and *d* randomly picked with PSUADE (Tong, 2012) ±5 orders of magnitude from decay rates (see inset); b) with limited ranges for *s* and *d* values, consistently with known affinity of elements. c) Sobol sensitivity index of Xe quantity to *s* and *d* parameters: constraints on back-diffusion of Xe from the magma to the gas phase are crucial.

7. Conclusion

- ✓ A simple formalism is proposed to account for gas-magma interactions with decay and ingrowth.
- ✓ Loss of Xe isotopes before migration to the atmosphere would be typically of 10-50%, up to factor of 10.
- ✓ Solubility of the parent radionuclides in the magma and low back-diffusion of Xe to the gas phase may lead to apparent fractionation of Xe isotopes. It is however associated with low activities of the 4 Xe isotopes.
- ✓ The Xe activity ratios are modified by gas-magma interactions but rarely to the point that military activities could not be discriminated from civilian ones.
- ✓ Better constraints are needed on the values of solubility and back-diffusion, especially for xenon out of the magma.

References: [1] Chapin, C. E., 1970, CONF-700101(vol1), 463-480, IAEA. Claassen, H. 1978, 712-D, USGS. Hudson, B. et al., 1987, UCRL-96354, LLNL. Olsen, C. W., 1967, J. Geophys. Res. 72, 5037-5041. U.S. Congress, 1989, OTA-ISC-414. [2] Bateman, H., 1910. Proc. Cambridge Phil. Soc. 15: 423–427. [3] Sun Y. et al., 2014. Pure Appl. Geophys.: 1-23. [4] Kalinowski, M.B. et al. 2010. Pure Appl. Geophys., 167(4-5): 517-539. [5] Tong, C. (2012). report 94551-0808, LLNL.