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Cocktail-DCC method to determine radioisotopes quantities in source terms

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INTRODUCTION AND MAIN RESULTS

The novel Cocktail-DCC method, enables fast and robust calculation of radiological effects of a given source term, such as a nuclear detonation, including all nuclides and their dynamics over all time-scales. Allowing to assess source terms with high accuracy and speed.

This poster shows the new method and an example how it can be used to determine all different progeny activity over time of one Bq Uranium-238. To illustrate its use for identifying sources, we show how isotope ratios change over time for several source terms.

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Cocktail-DCC method

The cocktail-DCC (Van Dijk et al.,2024) method aims to simplify and speed-up the calculation of the decay of a given source term. Allowing researchers and policymakers easier access to information on isotopes.

Cocktail-DCC is a continuation of the work of Van Dillen et al. (2019). It will calculate a transition of a source term, $A(0)$, into a nuclide vector at a given time, $A(t)$, per nuclide A_i, to do so, it sums the ingrowth of that nuclide of all parents within the $A(0)$, as shown in eq. 1. Where the decay from parent to child is defined with the decay matrix $H_{ij}(\mathrm{t})$ and is calculated with eq. 2, note that $H_{ij} \neq e^{M_{ij}(t)}$, as it is a matrix exponential and should be calculated as such. Eq 3., shows how the matrix component of the exponential should be constructed, with lambda the decay constant and nu yield, being negative one for decay (at $i = j$). Since they are matrix exponents, the decay chains can continue all the way to the lowest stable isotope, if the timeframe allows it..

To add dose to activities of a source term, they are multiplied with the dose conversion coefficient for the appropriate pathway, eq 4. shows the example for air submersion, given the dose rate for the complete source term.

Decay calculations

Eq.1 $$A_i(t) = \sum_j H_{ij}(t) A_j(0)$$

Eq.2 $$H(t) = e^{Mt},$$

Eq.3 $$M_{ij} = \nu_{ij}\lambda_i,$$

ν_{ij}=yield if i≠*j* (production)
ν_{ij}=-1 for *i*=*j* (decay)

Dose calculations

Eq.4 $$DCC_{\mathrm{air,cocktail}}(t) = \sum_i DCC_{\mathrm{air},i} \sum_j H_{ij}(t) A_j(0)$$

Dose conversion factors (DCCs) from:

Inhalation:

- Eckerman et al. (2013) (ICRP 119)
- Kawai et al. (2002)

External radiation:

- EDC-Viewer, conform ICRP Publication 144 (Petoussi-Henss et al., 2020)

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Example with U-238

Figure 1 shows both up- and downstream progeny tree, the downstream part is shown in **Figure 2**. Where a time scale of a one-minute up to 2 million years is shown. The activity of all the progeny is shown to increase over time and several of them reach an equilibrium. Note that the half-life time of U-238 is 4 billion years and would not give a noticeable difference on this graph.

A graph like this would be very calculation intensive with equal time steps, seeing that decay goes with exponents, it makes sense to use a logarithmic timestep. In our simulation every pinpoint is for a moment in time 15% larger than the previous. E.g., for t0 at 60s, t1 = 69s, t2=79.35s ….. t200 =7.19x10^{13}s. Allowing a smooth overview over long periods without loss of detail or an extensive use of calculation powers.

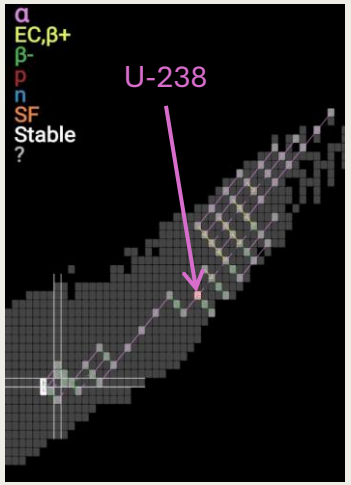

Figure 1: progeny tree, focused on U-238

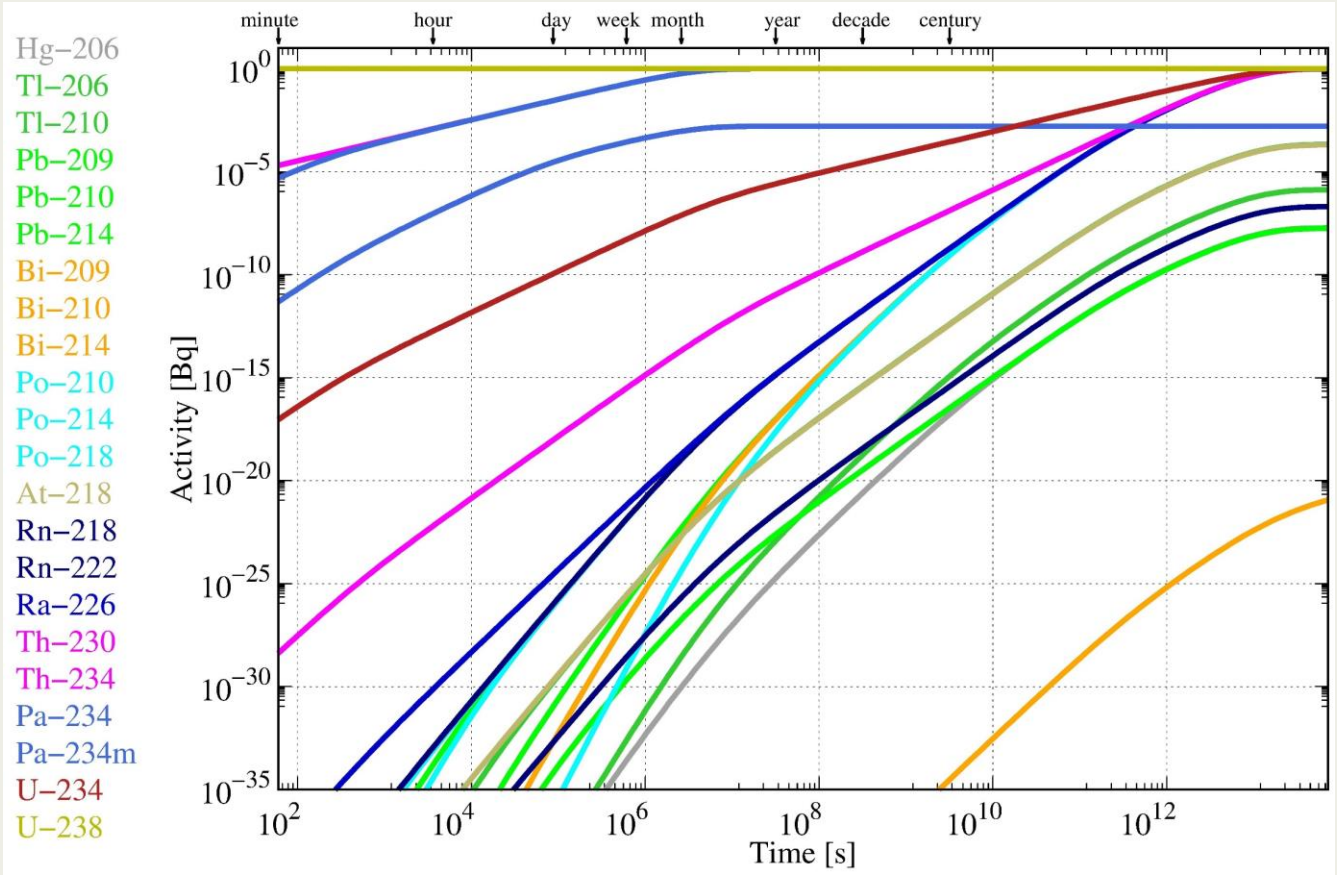

Figure 2: Activities of U-238 and its progeny

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Sources terms for calculations

The work of Kalinowski et al. (2021) shows that using four different Xe isotopes (Xe-131m, Xe-133, Xe-133m, and Xe-135) it is possible to distinguish between the nuclear power plant emission and possible nuclear detonations. For this research, we use the source terms as generated for U-235, U-238, and Pu-239. We have not generated the source terms themselves but obtained them from PREDICT done in way as explained by Axelsson et al. (2023).

All these three sourceterms include 500+ radionuclides, excluding leftover fuel and activation products. For these source terms, we calculate the decay over time, both with ingrowth and without ingrowth. Such that we can more accurately determine within what regions the isotope ratios are expected to be. **Table 1** shows the initial activities for the different fuels if 1 kT of TNT equivalent material would fissure. While **Table 2 & 3** show for how long it would be detectable if no dispersion would occur.

Concentration and detections

Table 1: Inital activities of the four different Xe-isotopes for different fuels in Bq per kT TNT equivalent.

Fuel	Xe-131m	Xe-133	Xe-133m	Xe-135
U-235	1.05E9	1.56E13	1.14E13	7.80E15
U-238	3.01E8	8.51E12	2.73E12	4.38E15
Pu-239	3.49E10	4.16E13	1.22E14	2.24E16

Table 2: Detection limits per radioisotope for 1 kT TNT equivalent and how long that would take per fuel, with ingrowth.

Isotope	Detection limit [Bq]	U-235 time [d]	U-238 time [d]	Pu-239 time [d]
Xe-131m	1.06E-4	709	710	712
Xe-133	1.66E-4	348	347	349
Xe-133m	1.06E-4	139	139	140
Xe-135	8.30E-4	26.4	26.3	26.5

Table 3: Detection limits per radioisotope for 1 kT TNT equivalent and how long that would take per fuel, without ingrowth.

Isotope	U-235 time [d]	U-238 time [d]	Pu-239 time [d]
Xe-131m	511	490	571
Xe-133	296	291	303
Xe-133m	124	119	131
Xe-135	24.0	23.7	24.5

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Validation and plotting the ratios

Figure 3 shows how the ratios develop with our method and source terms. It reflects the results from Kalinowski et al. (2021) , with slightly difference. Which were to be expected as we were not using the same source terms. The number in black indicate the age of the source term and the dashed line connects that moments where a source would have the same age.

A good thing to note is that the ingrowth of Xenon isotopes happens within the first few hours. Therefore, it is not very sensible to include those lines as serious options for borders of the possible observable ratios.

With the Xe-135 being the first isotope that is no longer measurable it is important to be close to the source as the dispersion will still reduce the amount per volume greatly. A dilution factor of 1e6 will reduce the time Xe-135 is detectable by roughly 8 days, and a 11-day-reduction occurs with a 1e9 reduction.

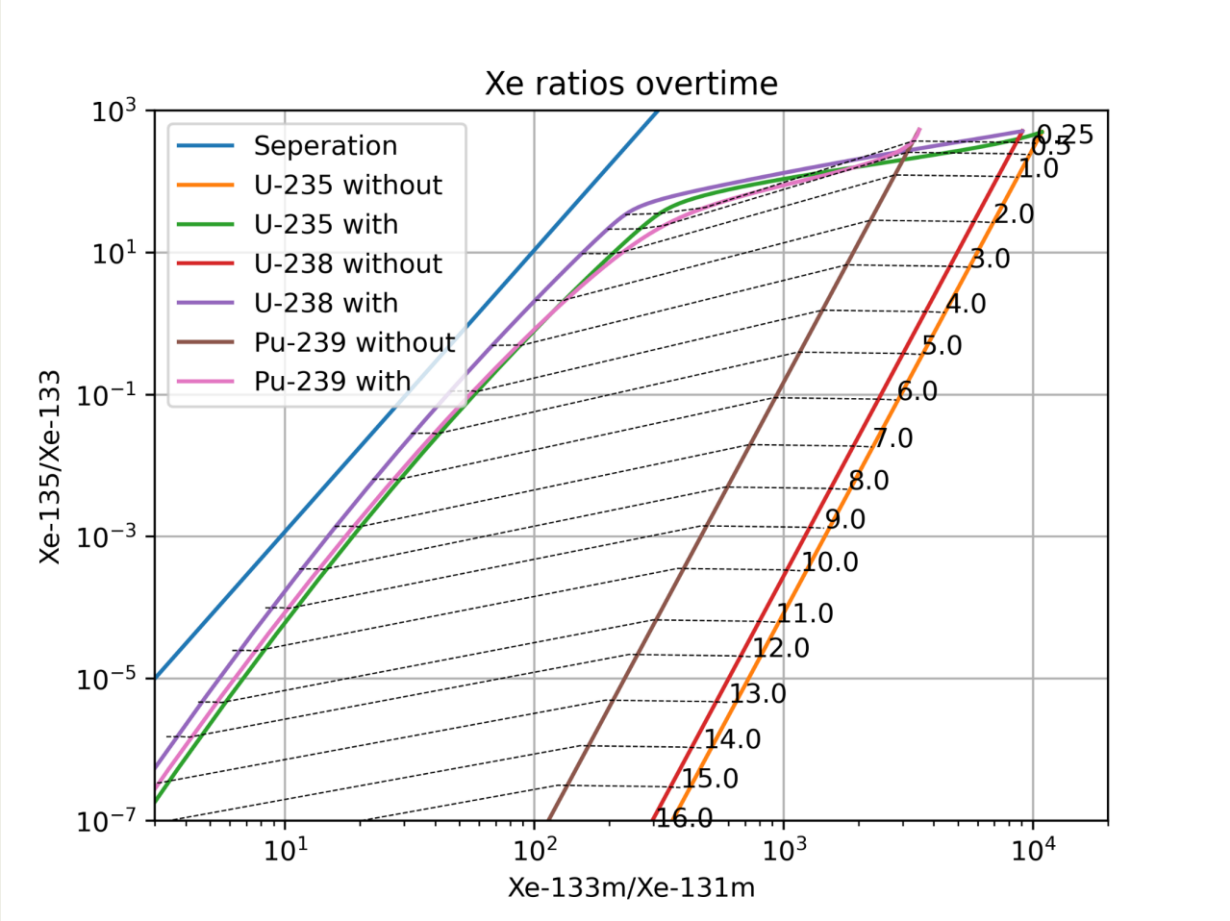

Figure 3: Ratios of Xenon isotopes for 4 different types of fuel, both with and without progeny. The numbers with the dotted line indicate the age of the ratio and the Seperation lines is for possible CTBT event.

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Discussion

The results agrees with the results obtained in other research. Showing similar patterns and development over time.

Currently, no (spontaneous) fission processes are included and only the simpler decay methods are accounted for. Nonetheless, this is an extension that could be included if needed.

The Cocktail-DCC method itself does not include for contamination or dispersion of the source term. This has to be accounted for through other means. Although, cocktail-DCC can be included into a dispersion model.

Much of the uncertainty lies within the composition of the given source term, as that is an important component of the uncertainty of radionuclide compositions over time.

Conclusions & recommendations

The Cocktail-DCC method allows for a fast and precise method to determine the age and activity of all progeny of a given source term.

It is applicable in both source term ageing, dose-rate, and dose determination. Where it will reduce the amount of time needed to calculate different components of the

With adjustments it could also be used for fission reactions. Other options are to adjust the default settings and work on the very short time-scales just after detonations.

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