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NOVEL METHOD FOR RADIATION DOSE ESTIMATION APPLIED TO DISPERSION SIMULATIONS OF NUCLEAR DETONATIONS

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Abstract: Since recent geopolitical tensions have grown, the concern for nuclear detonations has increased. The last atmospheric nuclear test was in 1980 (Foo et al., 2017; Kimball, 2022), nuclear preparedness has been scaled down worldwide to the safety and security of nuclear power plants and medical isotope facilities. To be prepared, we need to understand the development of both the geometry and the nuclear characteristics of a detonation cloud, as well as have an efficient method to estimate the radiation dose. This is not yet the case, as nuclear detonations evoke different issues than releases from nuclear facilities.
So far, atmospheric dispersion calculations of nuclear fall-out or nuclear facility incidents follow each radionuclide separately, which takes huge resources and time. To boost the assessment, the problem is often simplified. E.g., using pseudo nuclides, or the few "most important" radionuclides are selected for a particular exposure path in a particular timeframe, outside of which the subset of radionuclides will underestimate the dose (Kraus & Foster, 2014). Generally, radioactive decay is modelled as clean decay. The ingrowth of all progeny radionuclides is hardly ever considered, because of the difficulty of tracking even more radionuclides in the process of dispersion. As a result, the assessment of the radiation dose of nuclear releases is prone to errors.
We have developed an approach solving these limitations: cocktail-DCC. For a given initial radionuclide vector, we calculate the decay chain in advance, from one minute to millions of years after the release, including all 3821 nuclides. For the dose calculations, we apply the ICRP dose conversion factors, covering about 1258 radionuclides. The resulting time-dependent cocktail-DCC accounts for the dose of all (ingrowing) radionuclides. Atmospheric dispersion modelling can be limited to one (or several) representative passive tracer(s). Additional information on the characteristics and geometry of a nuclear cloud is found through literature study.
We will demonstrate how we validate the cocktail-DCC concept. We combine simulations of a nuclear detonation and assessment of dose distributions.

Key words: *Radionuclides, decay, comprehensive method, dose calculation, transition matrices*

INTRODUCTION

The last atmospheric nuclear test was in 1980 (Foo et al., 2017; Kimball, 2022). Since then, nuclear preparedness has shifted worldwide to the safety and security of nuclear power plants and medical isotope facilities. Recent geopolitical changes have made clear that there is still a need for atmospheric dispersion modelling to also be prepared for atmospheric releases from nuclear weapons with large quantities and exotic radionuclides.

To facilitate calculation of the atmospheric dispersion and dose scenarios in a timely manner without losing too much precision, several attempts, e.g. by Axelsson et al. (2023) and Kraus & Foster (2014), are made to condense the source terms with 200+ radionuclides to lists that cover at least a certain percentage of the total dose in a certain pathway. However, these selections are only valid within a certain time range and are of dubious quality outside their given time frame. These two issues make that method error prone and often less accurate. In this presentation, we demonstrate a method that includes all nuclides from a source term, so no compromises need to be made on which nuclides are applicable and should therefore be selected at

certain time frames for a certain pathway. This method, called Cocktail-DCC (Van Dijk et al., 2024; accepted for publication in Health Physics Journal) is not limited to only nuclear detonations but can be applied to any source term. Through the use of the ENDF dataset (Brown et al., 2018) containing 3821 different nuclides, we develop and make a method available that is able to account for all decay phenomena in delays ranging from a minute to more than two million years, with the possibility to calculate the potential dose. We will demonstrate the use of our method in several applications, such as (1): the ingrowth of daughter nuclides from U-238, (2): the evaluation of dose rate development of a given source term and (3): combine the dose with dispersion of a nuclear detonation. Altogether we hope this method can be widely adopted for operational use. Code and auxiliary files are available for free download.

METHOD COCKTAIL-DCC, DECAY MATRICES

In the course of time *t*, the initial source term A(0), also called the "cocktail", will decay into A(t), following the Bateman equation (Bateman, 1910). We solve this equation with the method of Dillen et al. (2019):

$$A_i(t) = \sum_j H_{ij}(t) A_j(0), \quad (1)$$

with $H_{ij}(t)$ the result of the matrix exponential:

$$H(t) = e^{Mt}, \quad (2)$$

where the matrix M has all its components defined as:

$$M_{ij} = \nu_{ij}\lambda_i, \quad (3)$$

where $\lambda_i = ln(2)/T_{1/2,i}$ is the decay constant, directly linked to the half-life $T_{1/2}$ of nuclide i and ν_{ij} indicates the fraction of parent nuclide j that becomes progeny nuclide i upon decay. If the nuclides j are no parents of i, the fraction is 0. For the case of $i = j$, we define the branching ratio as -1. Note that components of matrix H are not simply e to the power of the components of M, see e.g. Moler & Van Loan (2003).

METHOD COCKTAIL-DCC, ADDING DISPERSION AND DOSE ESTIMATION

The initial cocktail will not only decay over time; in case of atmospheric release, it will also disperse in the air. A cocktail can be represented with a single passive tracer, meaning that only the dimensionless thinning factor at a certain location needs to be known and this thinning factor can be applied to the total dose of the cocktail at any moment t or location x. The thinning $T_{air/ground}$ factors are found by dividing air concentration $\rho(x,t)$ and ground deposition $\sigma(x,t)$ of this passive tracer by the initial total amount of passive tracer considered. Equation 4 depicts how we can calculate the (potential) dose contributions D [Sv] to people with breathing rate B [$m^3\ s^{-1}$] through different pathways between t_1 and t_2.

$$D_{\text{inh}}(x, t_1 \rightarrow t_2) = \int_{t_1}^{t_2} BT_{\text{air}}(x,\tau)\text{DCC}_{\text{inh,cocktail}}(\tau)\text{d}\tau \quad (4a)$$

$$D_{\text{ext,air}}(x, t_1 \rightarrow t_2) = \int_{t_1}^{t_2} T_{\text{ground}}(x,\tau)\text{DCC}_{\text{ext,air,cocktail}}(\tau)\text{d}\tau \quad (4b)$$

$$D_{\text{ext,ground}}(x, t_1 \rightarrow t_2) = \int_{t_1}^{t_2} T_{\text{ground}}(x,\tau)\text{DCC}_{\text{ext,ground,cocktail}}(\tau)\text{d}\tau, \quad (4c)$$

The cocktail-DCCs $\text{DCC}_{\text{cocktail}}$ are defined as the sum of the dose rates from all nuclides in the cocktail, taking account for their decay chains (which introduces another sum):

$$\text{DCC}_{\text{inh,cocktail}}(t) = \sum_i \text{DCC}_{\text{inh},i} \sum_j H_{ij}(t) A_j(0) \quad (5a)$$

$$\text{DCC}_{\text{ext,air,cocktail}}(t) = \sum_i \text{DCC}_{\text{ext,air},i} \sum_j H_{ij}(t) A_j(0) \quad (5b)$$

$$\text{DCC}_{\text{ext,ground,cocktail}}(t) = \sum_i \text{DCC}_{\text{ext,ground},i} \sum_j H_{ij}(t) A_j(0) \quad (5c)$$

For a more detailed description, see the paper of Van Dijk et al. (2024), which will be available shortly. This method needs a one-time calculation, saving time during a dispersion or dose calculation.

ASSUMPTIONS AND LIMITATIONS

For this method to work, we made the following assumptions:

- No activation or fissure takes place during the time frame to which the cocktail-DCC is applied.
- The source term has a common reference point in time when its composition is known.
- We assume all particles disperse and deposit in the same way. It is possible to split the source term into several tracers that all describe a group with similar characteristics. E.g., one tracer for noble gasses, a second for elements with high boiling points, and a third one for elements with low boiling points (aerosols).

RESULTS INGROWTH

Using equation 1, we can determine the activity in a cocktail of a given initial source at any given time. For a source of 1 Bq U-238 these activities are displayed in Figure 1, showing the ingrowth of 21 daughter nuclides within 1e14 seconds. Note that U-238 has a half-life of 1.4e17 seconds, thus this head of chain nuclide itself does not show a significant decline in activity on the time scale shown.

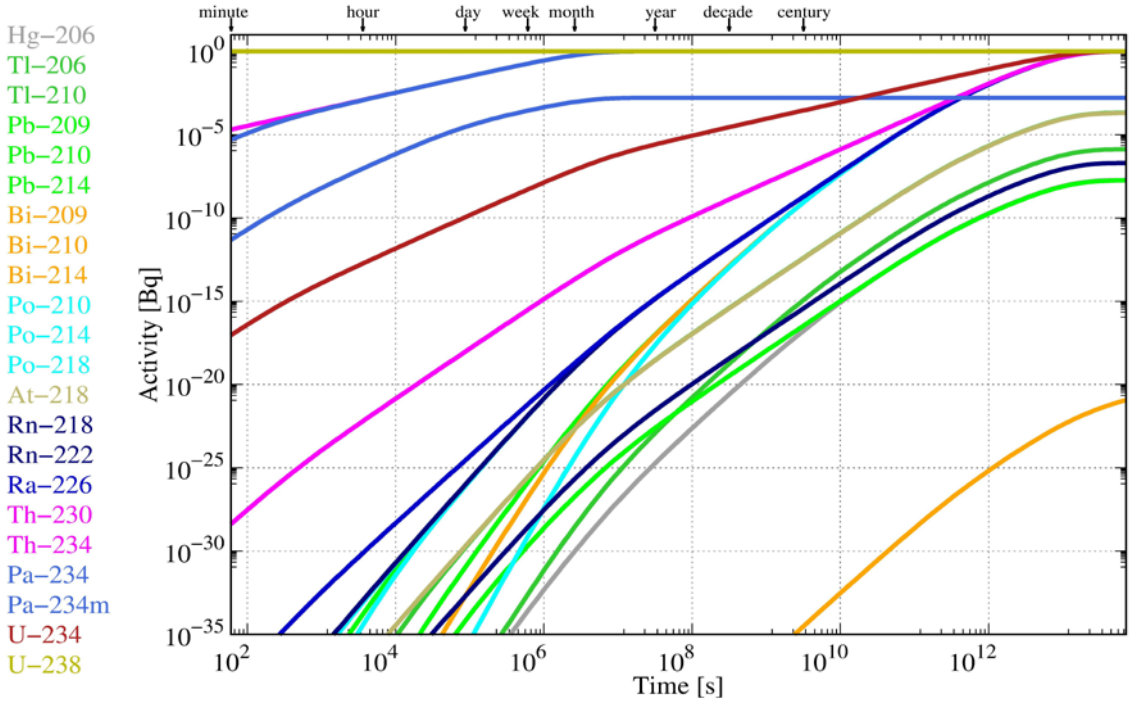

Figure 1: Ingrowth of progeny for 1 Bq U-238 during roughly 2.3 million years.

RESULTS COCKTAIL-DCC

With equation 5 we can determine the full potential dose of source terms after a nuclear detonation at any given moment. In figure 2, we compare our cocktail-DCC method (in black), which has all dose contributions, with the dose rate from the different optimized subsets of nuclides as proposed by Kraus & Foster (2014) and Axelsson et al. (2023) (shown as the thick lines in colour, with extrapolations outside of the time ranges of validity shown in thinner lines). It is clear that nearly all nuclide subset approximations, used in the classical approaches, lead to underestimations of the dose rate, even in their ranges of validity, whereas the cocktail-DCC method gives a universally valid dose rate estimate. This is most noticeable around the one-year mark.

RESULTS NUCLEAR DETONATION

The Cocktail-DCC method can be easily connected to an atmospheric dispersion model, since the (time dependent) cocktail-DCC can be calculated completely separately from a dispersion run. The cocktail-DCC can be either applied as a post-processing or integrated with the dispersion model.

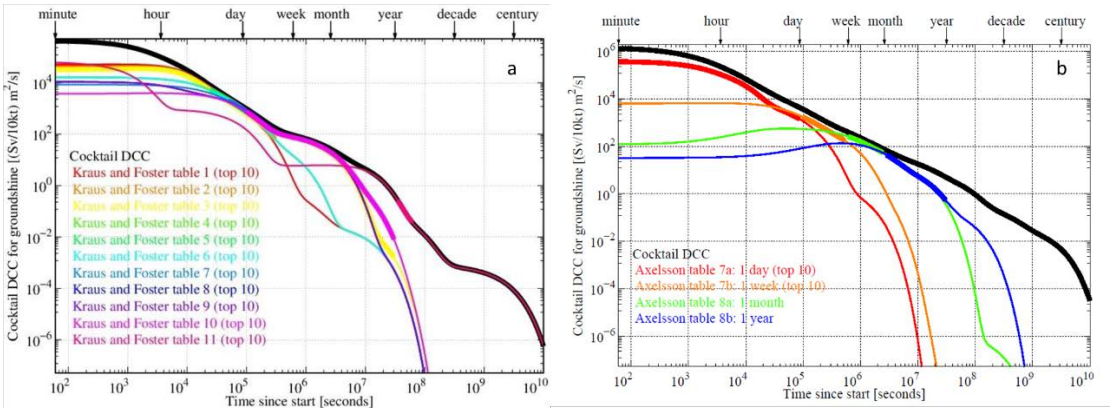

Figure 2: Comparison of our cocktail-DCC for groundshine (shown in black) and the dose rate associated with the first 10 radionuclides in tables of prioritized radionuclides of (a): the eleven tables in the appendix in Kraus and Foster (2014) and (b): the four tables from Axelsson et al. (2023). In thick lines each nuclide list has been applied only to the time range for which the respective table had been optimized, in thin lines the same curves have been extended to the full-time range shown.

Figure 3 displays the simulation with dispersion model NPK-PUFF (Tomas et al., 2021) of a fictional nuclear detonation. Through the use of a passive tracer, we calculate the thinning factors *T* used in equation 4. The top panels show the column integrated amounts of tracer on the left and the concentration at the surface level on the right. These two panels demonstrate that the patterns higher in the atmosphere do not necessarily match with those at the surface. The bottom panels show the total tracer deposition on the left and the corresponding dose of all pathways on the right. These two panels show that the highest depositions and dose from fall-out are not necessarily close to the location of detonation, which is shown by a red dot. While with a nuclear power plant the general patterns show more dose and deposition close to the source of release.

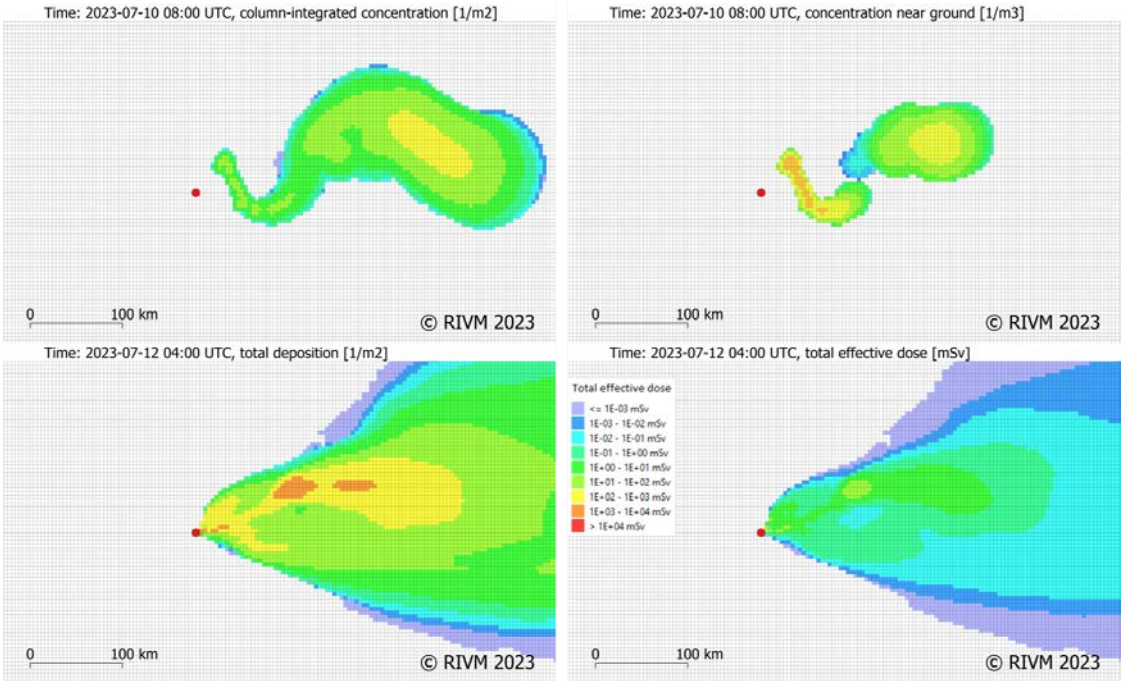

Figure 3: Example effects of a nuclear detonation. Top left: Column-integrated tracer concentration scaled with the amount released, 4 hours after detonation, logarithmic color scaling. Top right: Tracer concentration near ground scaled with the amount released, 4 hours after detonation, logarithmic color scaling. Bottom left: Total tracer deposition scaled with the amount released, 48 hours after detonation, logarithmic color scaling. Bottom right: Total effective dose 48 hours after detonation in mSv. The location of detonation is indicated by the red dot.

CONCLUSIONS

The cocktail-DCC method has the potential to improve, simplify, and speed up many radiological dose calculations, because the dose calculations are done in advance, all progeny is included, and it works with a minimum of an atmospheric dispersion model that can follow one passive tracer, meaning that in principle every dispersion model can be combined with the Cocktail-DCC. These characteristics, plus the fact that model and auxiliary files are freely available, make the cocktail-DCC fit for widespread integration and use. This new method can be applied to any chemical or physical process that is only time dependent,

DATA AVAILABILITY

The code for the cocktail-DCC method (including pre-calculated decay matrices in sparse format, source terms from literature for different nuclear weapons and the associated cocktail-DCCs) is available on: https://github.com/rivm-syso/Cocktail-DCC. Feel free to use our method. Links to relevant 3rd party material are given in the github readme.

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