

## DEVELOPMENT OF A DATABASE: DACTARI FOR A RADIOTOXIC ELEMENT RANKING METHODOLOGY

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Dosimetric impact studies aim at evaluating potential radiological effects of chronic or acute releases from nuclear facilities. A methodology for ranking radionuclides (RN) in terms of their health-related impact on the human population was first developed at CEA with specific criteria for each RN that could be applied to a variety of situations. It is based, in particular, on applying physico-chemical criteria to the complete RN inventory (present in the release or in the source term) and on applying norms related to radiation protection and chemical toxicology. The initial step consisted in identifying and collecting data necessary to apply the methodology, with reference to a previous database of long-lived radionuclides (LLRN, with half-lives ranging from 30 to  $10^{14}$  y) containing 95 radionuclides. The initial results have allowed us to identify missing data and revealed the need to complete the study for both toxic and radiotoxic aspects. This led us to the next step, developing a specific database, DAtabase for Chemical Toxicity and Radiotoxicity Assessment of RadIonuclides (DACTARI), to collect data on chemical toxicity and radiotoxicity, including acute or chronic toxicity, the chemical form of the compounds, the contamination route (ingestion, inhalation), lethal doses, target organs, intestinal and maternal-foetal transfer, drinking water guidelines and the mutagenic and carcinogenic properties.

### INTRODUCTION

Dosimetric impact studies aim at evaluating potential radiological effects of chronic or acute releases from nuclear facilities. Only atmospheric or liquid releases were studied. In order to perform them, different tools were needed with the following objectives: (a) describe release modes, dispersion processes as well as environmental impact from biosphere up to man<sup>(1)</sup> (Figure 1), (b) integrate the whole processes and (c) be able to perform impact calculations relative to type cases. These releases constitute the source term, which will be the basis for the calculation of radiological impact and they represent a fraction of the inventory present in the considered facility. This source could be:

- the fuel inventory at ‘reactor exit’ which could be then dealt with, either through disposal or storage, or treated in the appropriate facility;
- the waste package inventory which will be then dealt with, either through disposal or storage in the appropriate facility, and which is a fraction of the fuel inventory;

- the inventory of any facility containing or manipulating some radionuclides (in the case of a dosimetric impact) or other compounds (in the case of chemical impact), including the above mentioned facilities.

Consequently, the development of such calculation of radiological impact will lead to consideration of an important number of isotopes. This justifies the fact of having a methodology allowing to rank the isotopes for many reasons: (a) to rapidly identify the most toxic group of isotopes (or even elements) that ought to be taken into account in the calculations of impact, on the basis of radiological or radiotoxicological criteria, or (b) to perform a comparison between different types of inventories in order to appreciate the appearance or loss of toxic elements or isotopes.

Hence, a methodology for ranking radionuclides in terms of health-related effects on the human has been developed in order to be applied in various cases: different types of fuel or waste packages, waste management options, nominal or altered situations, general public versus nuclear worker, ingestion versus inhalation, etc. This methodology relies, in particular, on the application of different

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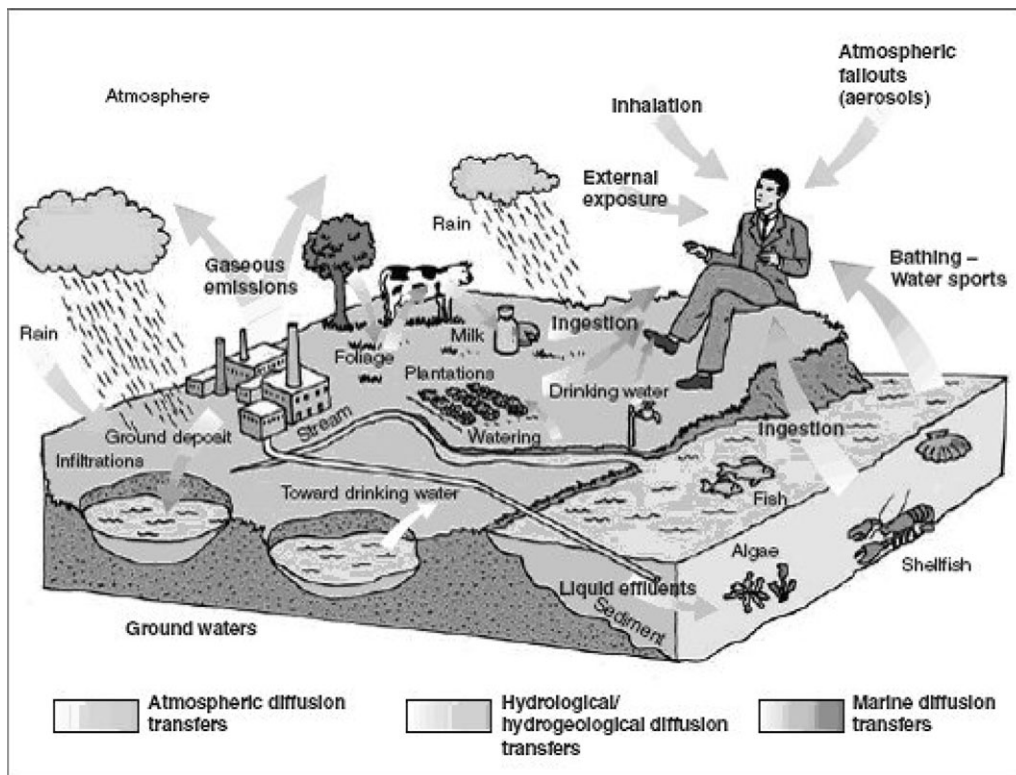


Figure 1. Transfer of radioactive contaminant in biosphere (soils, lakes, rivers, atmosphere, fauna, flora and human).

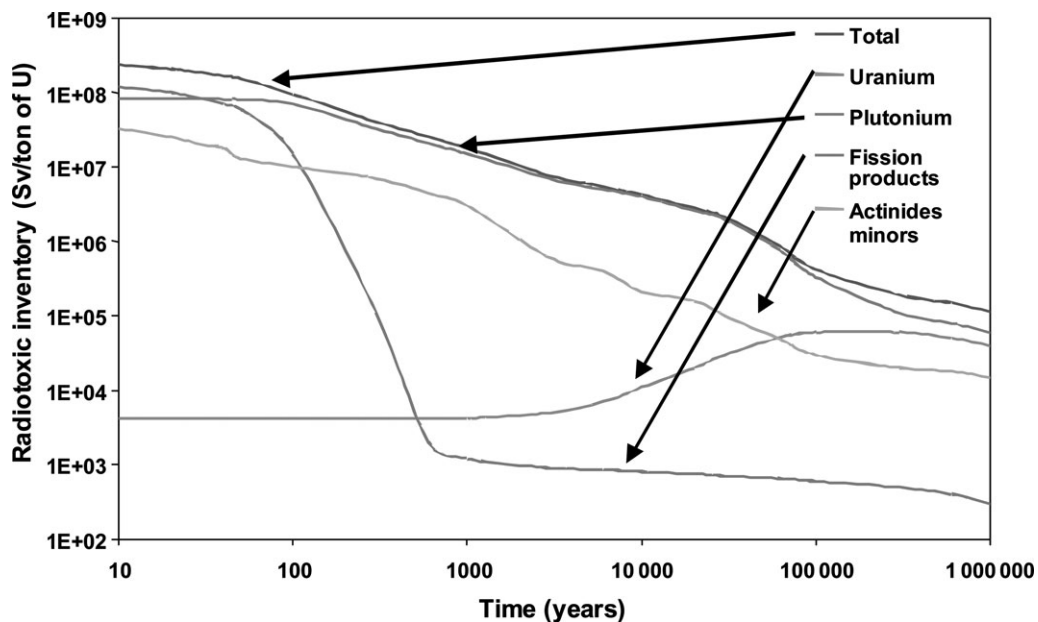


Figure 2. Evolution of radiotoxic inventory (in Sv/ton of U) for a UOX spent fuel (60 GWd/t)<sup>(2)</sup>.

physico-chemical criteria to the whole inventory. The order of application of these criteria (activity, period, physical form, solubility, potential dose, drinking water limit, radiological limit, etc.) has been studied in order to appreciate its influence on the ranking result.

Figure 2 illustrates, for example, the use of one criterion, namely the potential radiotoxicity (calculated to be the product of a ranking radionuclides (RN) activity by its dose factor and which corresponds to the dose in case of the RN ingestion). This allows comparing the contribution of each nuclide to the radiotoxic inventory present in a spent fuel and permits to apply the relevant fuel management strategies<sup>(2)</sup>.

The aim of this study is to present both databases, which have been developed in the framework of the ranking methodology. The first step consists of the collection and inventory of all data necessary for the application of the methodology. In fact it relies on the database developed on long-lived radionuclides (LLRN database)<sup>(3)</sup>, which will be presented below. These results have allowed us to identify some missing data and the need to take into account of chemical elements which is potentially toxic and radiotoxic. In particular, a more specific database DAtabase for Chemical Toxicity and radiotoxicity Assessment of RadIonuclides (DACTARI), has been enlarged in order to collect data on both toxicity and radiotoxicity. This base relies on data coming from various databases and is presented below.

#### LONG-LIVED RADIONUCLIDES (LLRN) DATABASE

This database<sup>(3)</sup> has been created at the CEA between 1998 and 2002 in order to support all ranking studies and has allowed gathering of information about long-lived radionuclides (with half-lives ranging from 30 to  $10^{14}$  y) on seven different topics.

- 'Radioactive data' gathers fundamental data characterising radioactive properties of radionuclides considered (half-life, disintegration mode, radiation energy, etc.).
- 'Formation and inventory' collects data on radionuclide formation ways as well as quantities formed during fuel irradiation (for PWR fuels—UOX and MOX—as well as for SFR fuel).
- 'Biological effects' gives data characterising both radiotoxicity for each radionuclide and some characteristics of chemical toxicity for the considered element.
- 'Partitioning' specifies element repartition according to the different ways of Purex reprocessing and complementary partitioning

Table 1. Example of acute toxicity data implemented in DACTARI database for few elements.

Element	Chemical form	LD 50 <sup>a</sup> (mg/kg)	Animal species (route)	Reference	Classification LD50 (EPA) <sup>b</sup>	f <sub>A</sub> : fractional absorption in the gastrointestinal tract (ICRP, 88) <sup>(7)</sup> 100) <sup>(6)</sup>	Materno-fetal transfer (ICRP, 88) <sup>(7)</sup>
Sr	Sr	1.20E + 03 - 3.12E + 03	Rat (oral)	INTOX <sup>b</sup>	3-NC	0.25	0.3-8
Tc	Pertechnetate (TcO <sub>4</sub> )	1.30E + 01	Mouse (IV) <sup>c</sup>	ATSDR <sup>b</sup>		0.8	1
I	I	7.60E + 01 - 1.40E + 04	Rat (oral)	HSDB <sup>b</sup>	2-NC	1	1
U	U	1.14E + 02 - 2.04E + 02	Rat (oral)	ATSDR <sup>b</sup>	2	0.02	0.1-1
Pu	Pu 238	1.89E + 01	Neonatal rat (gavage)	ATSDR <sup>b</sup>	1		
	Pu 239	5.72E + 00 - 9.34E + 00	Rat (IV) <sup>c</sup>	ATSDR <sup>b</sup>			
		7.58E + 01 - 1.56E + 02	Rat (IV) <sup>c</sup>	ATSDR <sup>b</sup>		5.0E - 04	0.03-1

<sup>a</sup>LD50, Lethal dose.

<sup>b</sup>EPA, ATSDR, HSDB defined in Table 2.

<sup>c</sup>IV, intravenous.

- processes possible for isolating chemical elements corresponding to the considered radionuclides.
- 'Transmutation' provides data allowing to evaluate in part the feasibility and destruction performances of radionuclides by transmutation with neutrons.
  - 'Behaviour in waste packages' gathers some data relative to the properties of waste packages confinement. Only data on vitrified waste packages are given there.
  - 'Behaviour in diluted solution' provides data used for element migration calculations.

In each of these topics, each dataset is accompanied by a comment giving the reason for their choice, their origin (for traceability) as well as a robustness index (traducing the confidence given to the value if well-known, known or lack of information) and the way to use it. Ninety-five radionuclides as well as 85 daughter radionuclides have been listed.

#### DACTARI DATABASE

Although 'biological effects' had been taken into account in the LLRN database, some data were missing, which leads to the creation of another database, DACTARI, more specifically targeting the chemical and radiological toxicity of certain elements. This database was initiated in 2004 and now contains only 24 elements (Be, B, Cr, Co, Ni, Zn, Se, Sr, Zr, Nb, Mo, Tc, Cd, Sn, Sb, I, Cs, Pb, Th, U, Np, Pu, Am, Cm) identified during the initial ranking and dose impact studies carried out by the CEA on the basis of a UOX fuel source term.

Initially, the database was developed under the most likely scenario of contamination by ingestion, based on concepts such as the physico-chemical

form or speciation of these compounds and cases of acute or chronic toxicity.

Several parameters were selected to increase the three fundamental aspects of the database.

#### Physical and chemical properties

Parameters were collected, including the half-life, specific activity, type of radioactive emission, main oxidation states in solution, solubility products in three reference media (hydroxide, carbonate and phosphate), redox potentials, radiotoxicity group<sup>(4)</sup>, use in the fuel cycle and main bibliographical references.

#### Acute toxicity

The selected parameters are the 50% lethal dose for which four categories are defined for oral ingestion (very toxic (<25 mg kg<sup>-1</sup>), toxic (>25–200 mg kg<sup>-1</sup>), harmful (>200–2000 mg kg<sup>-1</sup>) and non-classified (>2000 mg kg<sup>-1</sup>))<sup>(5)</sup>, the intestinal transfer factor  $f_i$  based on various information sources (ICRP<sup>(6)</sup>, EPA, Table 2) and materno-foetal transfer (ICRP Publication 88<sup>(7)</sup>). Table 1 is an example of the database contents for five of the 24 elements.

#### Chronic toxicity

The selected parameters are the WHO guidelines for radionuclides<sup>(8)</sup> and for chemical elements<sup>(9)</sup>, the classification in terms of carcinogenicity (EPA and EEC references) and reproduction (EEC). Only the EEC reference in the form of a directive contains statutory values<sup>(5)</sup>. A 'non-carcinogen' column contains values or remarks concerning critical effects, the reference dose (RfD in mg kg<sup>-1</sup> d<sup>-1</sup>).

**Table 2. Website addresses of the main institutes or organisations providing toxicological profiles.**

Organism	Web address
ATSDR (USA)	Agency for Toxic Substances and Disease Registry Centers for Disease and Control; 268 profiles were published as final Toxicological Profiles: <a href="http://www.atsdr.cdc.gov/toxpro2.html">http://www.atsdr.cdc.gov/toxpro2.html</a>
EPA (USA)	Environmental Protection Agency; 45 files <a href="http://www.epa.gov/iris/toxreviews/">http://www.epa.gov/iris/toxreviews/</a>
INERIS (France)	Institut National de l'environnement Industriel et des Risques; 64 files <a href="http://www.ineris.fr/index.php?module=cms&amp;action=getContent&amp;idl_heading_object=3">http://www.ineris.fr/index.php?module=cms&amp;action=getContent&amp;idl_heading_object=3</a>
INRS (France)	Institut National de Recherche et de Sécurité; 259 files <a href="http://www.inrs.fr/dossiers/fichtox/somft.htm">http://www.inrs.fr/dossiers/fichtox/somft.htm</a>
IRSN (France)	Institut de Radioprotection et de Sûreté nucléaire; 13 files (environment and health) <a href="http://www.irsno.org">http://www.irsno.org</a>
IARC	International Agency for Research on Cancer, World Health Organisation; <a href="http://www.iarc.fr/">http://www.iarc.fr/</a>
HSDB (USA)	Hazardous Substances Data Bank: belongs to Toxicology Data Network (TOXNET) <a href="http://toxnet.nlm.nih.gov/cgi-bin/sis/htmlgen?HSDB">http://toxnet.nlm.nih.gov/cgi-bin/sis/htmlgen?HSDB</a>
INTOX (USA)	IPCSINTOX Databank: Inchem. Chemical Information from Intergovernmental Organisations <a href="http://www.intox.org/databank/index.htm">http://www.intox.org/databank/index.htm</a>
NIOSH (USA)	National Institute for Occupational Safety and Health <a href="http://www.cdc.gov/niosh/chem-inx.html">http://www.cdc.gov/niosh/chem-inx.html</a>

All the data collected and selected for this work were taken from national and international databases and are listed in Table 2, with the organisation name and website to provide direct access.

## CONCLUSION

Health-related impact calculations rely on tools integrating both transfer models and databases. From a given source term, the principle is to apply different criteria, among them the physico-chemical properties as well as chemical toxicity and radiotoxicity.

The study performed at CEA has conducted for the elaboration of two databases, LLRN and DACTARI, which would allow to have a better evaluation of doses to humans, taking into account of chemical, but also toxicological and radiotoxicological parameters.

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