# Radiological Impacts in Life Cycle Assessment. Part I: General framework and two practical methodologies


## Abstract

To date, impacts of ionising radiations have been largely disregarded in Life Cycle Assessment (LCA). This omission can be linked to the lack of a standard and comprehensive framework for including the effects of radionuclides alongside other emissions from industrial processes. Drawing on a recent review of Radiological Impact Assessment methodologies for LCA studies, this article proposes an overarching framework for integrating impacts of radionuclides in the Impact Assessment phase of LCA. From this framework, two alternative methodologies have been derived. They differ mainly in the way transport and dispersion of radionuclides in the environment are modelled: UCrad represents the first-of-its-kind compartment-type methodology for radionuclides, whereas the alternative Critical Group Methodology (CGM) has been adapted from standard Risk Assessment practices. Characterisation factors for a range of emitted species have been calculated using both methodologies and compared with those obtained from the Human Health Damages methodology, which is the only approach to radiological impacts yet implemented in LCA. For both UCrad and CGM the results are in general agreement with the Human Health Damages methodology, but UCrad gives factors closer to those obtained by the CGM approach. UCrad represents a major step towards incorporating ionising radiation impacts in LCIA. A subsequent paper will explore quantitatively the main differences between the UCrad and CGM methodologies.

## Keywords

Life Cycle Impact Assessment; Ionising radiations; Radiological Impacts; Risk assessment; Radionuclides; Nuclear Waste.

# 1 Introduction

Life Cycle Assessment (LCA) studies aim at assessing the potential impacts on human beings and the environment of the complete life cycle providing a product or service. The prime feature of LCA, and also its main advantage with respect to other environmental tools, lies in its holistic environmental perspective, which has made it a central concept for both environmental management in industry and public policy-making (Malcolm, 2019). To justify the claim to be holistic and balanced, Life Cycle Impact Assessment (LCIA) methodology must assess potential impacts of all main types of pollutants, including ionising radiations. However, to date impacts of ionising radiations have been largely disregarded. The nuclear industry is the main source, whether measured by the scale of the releases and waste or by the radioactivity of the materials used. Other industries (e.g. coal, oil and gas, fertiliser and construction) routinely release emissions containing radionuclides to air and water, whilst others (mainly nuclear energy, hospitals and weapons production) generate radioactive solid waste. At present, radioactive wastes are either disposed of in near-surface landfills or stored awaiting construction of long-term geological repositories (WNA, 2018). Over tens of thousands of years, waste canisters will deteriorate, releasing the stored radionuclides into the environment.

Paulillo et al. (2018) reviewed the methodologies proposed for including radiological impacts in LCIA. They have been either developed exclusively for LCA applications (Frischknecht et al., 2000; Heijungs et al., 1992; Solberg-Johansen, 1998; Solberg-Johansen et al., 1997) or adapted from standard assessment procedures used in other fields (e.g. Simmonds et al., 1995; Wareing, 2009). Paulillo et al. concluded that none of the methodologies currently available is sufficiently comprehensive for use as a standard procedure for radiological impact assessment in LCIA. The Human Health Damages (HHD) approach (Frischknecht et al., 2000) is the only methodology so far included in general LCIA methods – e.g. CML (Guinée et al., 2002), RECIPE (Goedkoop et al., 2013), Eco-indicator 99 (Goedkoop and Spriensma, 2001) and Impact 2002+ (Humbert et al., 2012) - although it has been classified as "recommended but in need of some improvements" (Hauschild et al., 2013). Paulillo et al. (2018)

concluded that a generally usable methodology must be site-independent, applicable to both direct discharges and emissions from a geological repository, and produce average (rather than worst case) estimates of impacts. This led to a general framework for assessing the human impacts of radioactive emissions, embodied in two alternative, and conceptually different, methodologies introduced in this paper. This work meets some of the needs identified by the EU Joint Research Centre (JRC, 2011), including extending the number of radionuclides and ensuring compatibility between impact models for radionuclides and toxic substances. One of the methodologies, UCrad (Paulillo, 2018), applies the multimedia compartment-type environmental modelling approach proposed by Mackay (2001), widely used in LCIA; e.g. in USEtox (Rosenbaum et al., 2008). UCrad is similar to the approach proposed by Joyce et al. (2016) and Goronovski et al. (2018) to predict human impacts from routine process emissions containing Naturally Occurring Radioactive Materials (NORM), but extends the assessment to other radionuclides and to emissions from disposal of radioactive wastes. The alternative approach, the Critical Group Methodology (CGM), has been adapted from Human and Environmental Risk Assessment (HERA) approaches to provide a comparative basis for assessing the results of UCrad (Paulillo, 2018). CGM uses Gaussian plume dispersion models to assess the exposure of a "critical group" of humans; this makes the methodology site-dependent, but does not necessarily mean that it produces worst case estimates (see Section 2.2.1).

Section 2 provides a detailed description of the general framework and the two derived methodologies. Section 3 presents characterisation factors from the two methodologies and compares them with those obtained from the HHD methodology. Section 4 analyses the main features, differences and limitations of the methodologies. The main findings are summarised in Section 5 with a glossary of acronyms provided in Section 6. A complementary paper will set out a more quantitative comparison of characterisation factors from the two methodologies to reveal the consequences of critical assumptions in fate modelling.

# 2 Methods and Materials

## 2.1 General framework

The main purpose of this work is to establish a standard framework for integrating radiological impacts into LCA as a new impact category. The approach is to generate characterisation factors to enable the inventory of radioactive emissions from a process or product system to be converted into potential impacts measured in the common unit of the environmental category (man-Sievert, Sievert or Risk).

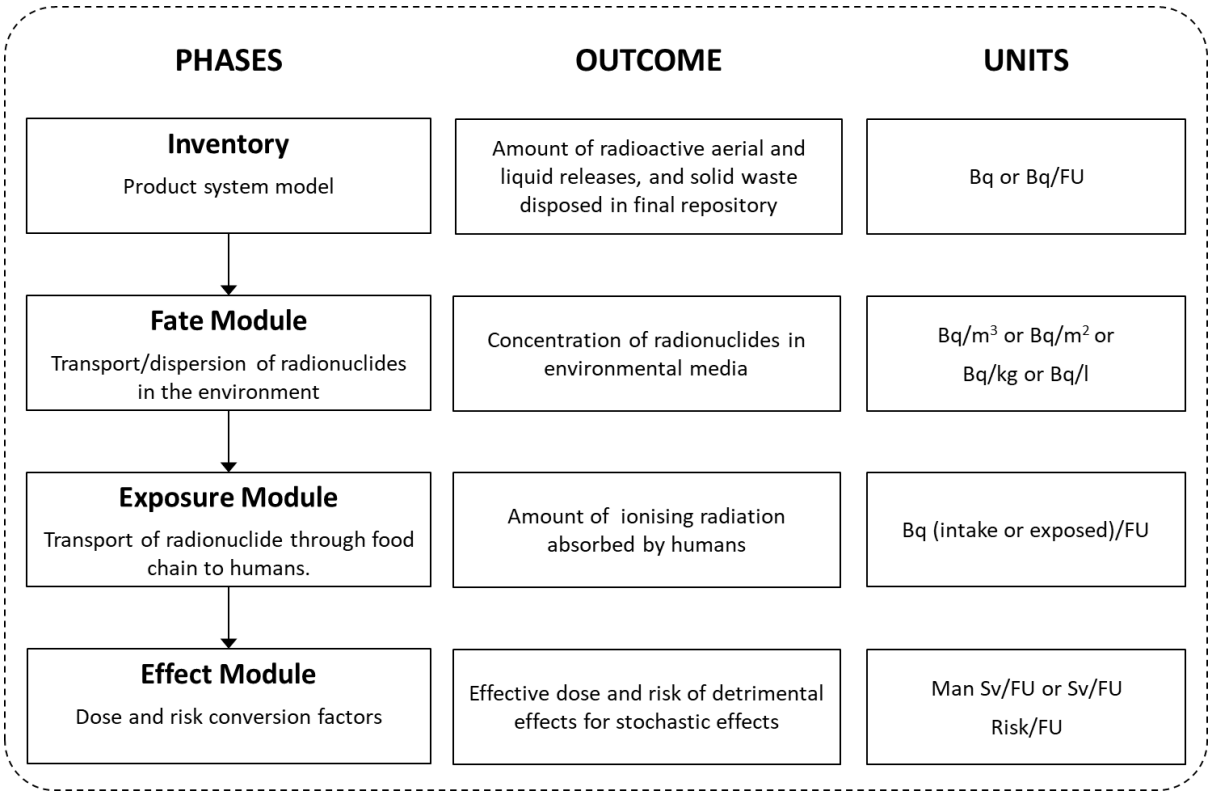


**Figure 1 – Overview of the impact assessment framework. Bq: Becquerels; Sv: Sieverts; FU: Functional Unit.**

The framework is outlined in Figure 1. The CGM and UCrad methodologies share the same Exposure and Effect Modules but differ in the Fate Module, which models transport and dispersion of radionuclides following release to estimate their concentrations in the different environmental media. Section 2.2.1 distinguishes between the approaches adopted in the two models. The Exposure Module (Section 2.3) uses the resulting environmental concentrations to estimate the amount of ionising

radiation absorbed by human beings, expressed in terms of Becquerels (Bq), according to specific habits and behaviours. Finally, the Effect Module (Section 2.4) converts the predicted exposures into an effective dose, measured in Sieverts (Sv), allowing for both the type of radiation and the human tissue affected. If desired, the dose may be converted into a risk metric for detrimental effects.

## 2.2 Fate Module

### 2.2.1 Critical Group Methodology

The CGM Fate Module uses analytical models describing dispersion and transport of releases to predict the exposure of a Critical Group defined as "the individual members of a population who can realistically be expected to receive the highest dose due to their lifestyle, location and habits" (ICRP, 1990; NRPB, 1993). This approach introduces two significant assumptions: i) the population comprising the Critical Group is concentrated in a specific location, and ii) the impacts depend on where that location is positioned relative to the point of release. Unlike the approach used in Risk Assessment, the critical group in CGM need not represent a worst case; rather, it enables selection of the most appropriate distance at which to assess radiological impacts.

The CGM methodology covers both direct discharges and releases from waste disposed in a final repository. These impacts, however, occur on very different time scales - days/weeks/months vs tens of thousands of years - so that any comparison between them must be interpreted with caution. The two approaches are discussed in Sections 2.2.1.1 and 2.2.1.2 respectively. Impact assessment of direct discharges is based on the IAEA (2001) framework for routine discharges from nuclear plants, whilst potential impacts of radionuclides in solid wastes in a generic Geological Disposal Facility (GDF) are assessed following the generic Post-Closure Performance Assessment (PCSA) developed by Radioactive Waste Management Ltd. for the UK Nuclear Decommissioning Authority (NDA, 2010a).

Recently, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) published an updated methodology for assessing impacts on the general public from direct discharges

of a limited number of radioisotopes (UNSCEAR, 2017). The methodology resembles IAEA (2001), but considers a uniformly distributed population rather than a Critical Group and so gives no guidance on the distance from the source at which exposure should be assessed. This is appropriate for routine direct emissions, considered by the UNSCEAR methodology, because total impacts are insensitive to distance when a uniform population is exposed to a pollutant whose dose-response function is linear with no threshold (Dreicer et al., 1995; Spadaro and Rabl, 1999). However, the approach explored here is intended to apply to both direct releases and discharges from waste disposal sites; it therefore uses the Critical Group concept to align it with the PCSA rather than the UNSCEAR methodology.

#### *2.2.1.1 Direct discharges*

Figure 2 shows the approach used in the Fate Module for direct discharges, using a simple model for dispersion in a generic environment. It represents a compromise between accuracy and data requirements, based on the second level of the series of simple screening procedures recommended by IAEA (2001) to demonstrate compliance with environmental standards. The IAEA framework includes three direct discharge pathways: atmospheric, to surface (both fresh and sea) water and to sewage. However, the CGM methodology developed here considers only the two pathways shown in Figure 2; discharge to sewage is disregarded as much less significant. Figure 2 shows the associated transport pathways - dispersion, deposition and advection including irrigation - and the environmental compartments where resulting concentrations are modelled.

For atmospheric discharges, the Gaussian plume model (Gifford, 1976; Pasquill, 1961), widely used in radiological assessment, is employed to describe dispersion of long-term releases of pollutants undergoing downwind transport (advection) and mixing (turbulent diffusion). Atmospheric concentrations predicted by this model for large distances are known to be uncertain, so the IAEA recommends that it only be used for distances up to 20 km. Limiting the assessment range to 20 km means that the travel time is short; consequently radionuclides decay is negligible, and is ignored in the IAEA model. However, the CGM methodology has been developed for distances well above 20 km;

it therefore incorporates radioactive decay. In addition to distance from the source, the atmospheric concentration depends on other factors, of which height of release, geometric mean wind speed at height of release, and presence of buildings near the release source are the most significant.

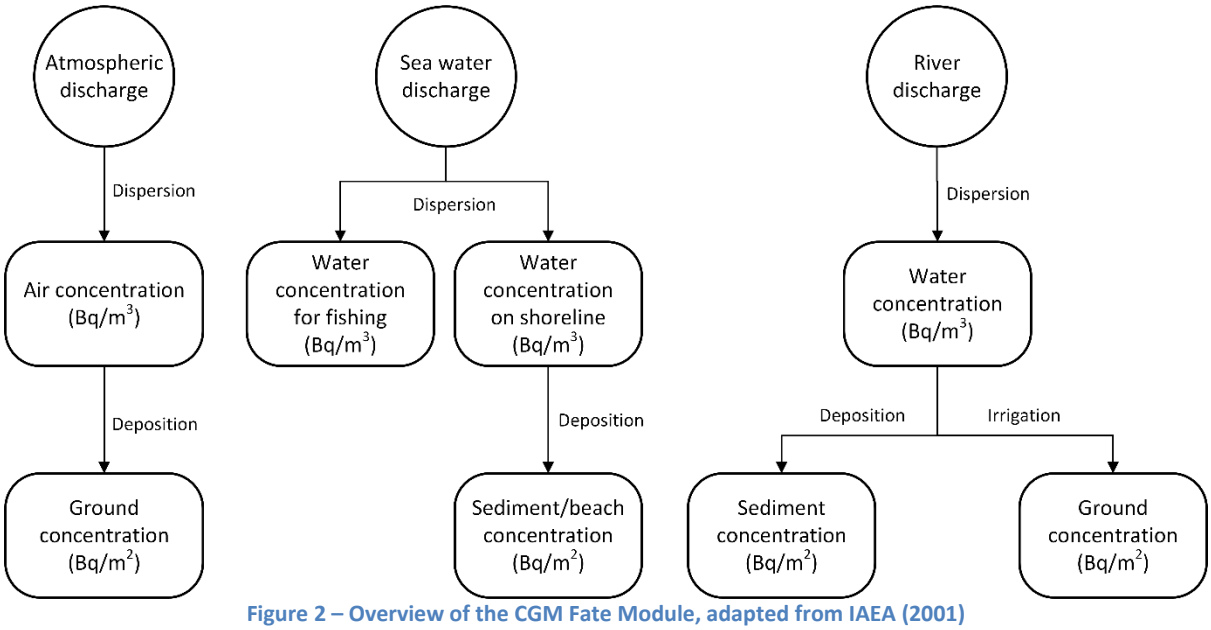


**Figure 2 – Overview of the CGM Fate Module, adapted from IAEA (2001)**

For liquid discharges, IAEA (2001) recommends an analytical solution to the advection-diffusion equations for steady uniform flow. Amongst the different water bodies considered by IAEA, CGM describes only discharges to rivers and coastal waters because these represent the majority of discharges by the nuclear industry. The concentration of radionuclides in water depends on distance from the release and on features of the water body such as: width, depth and net velocity for rivers; and depth, distance from release to shoreline and average coastal current for marine emissions. The coastal water model estimates two different concentrations of radionuclides, one of interest for fishing purposes and another related to activity along the shoreline. The river model estimates only one concentration, the total concentration of radionuclides in water, with the general assumption of perfect vertical and horizontal mixing.

153 The air and water concentrations are also used in the IAEA framework to calculate ground
154 concentration due to atmospheric deposition and irrigation, and sediment/beach concentration due
155 to build-up of deposited sediments from freshwater and seawater (Figure 2).

156 Table 1 reports the parameters used in the CGM fate models. The distance of the critical group from
157 the source is not included as it is an additional independent variable on which the CGM exposure
158 depends. Most of the parameters are taken from IAEA (2001) but some are assigned specific values
159 appropriate to the UK. For instance, data from Sellafield (the industrial complex whose main purpose
160 was reprocessing of spent nuclear fuels) have been used for the height of atmospheric discharges.

161 **Table 1 – Parameters used in CGM Fate Module**

| Emission | Parameter | Value | Comments |
|---|---|---|---|
| Air | Height of discharge (m) | 100.00 | *Based on Sellafield THORP[1] stack height* |
| | Fraction of time wind blows towards receptor (-) | 0.25 | *IAEA recommended value* |
| | Wind speed (geometric mean) (m/s) | 2.00 | *IAEA recommended value* |
| Sea water | Water depth at discharge (m) | 15.00 | *Based on Sellafield sea data[2]* |
| | Distance from shoreline (m) | 2100.00 | *Based on Sellafield liquid discharges[3]* |
| | Mean coastal current(m/s) | 0.10 | *IAEA recommended value* |
| Rivers | Width (m) | 21.00 | *Medium-sized river[4]* |
| | Depth (m) | 0.34 | *Calculated from IAEA table III* |
| | Net flow rate ($m^3/s$) | 0.70 | *Calculated from IAEA table III* |
| | Flow rate ($m^3/s$) | 5.00 | *Calculated from IAEA table III* |

[1] *The Thermal Oxide Reprocessing Plant (THORP) at Sellafield was the flagship plant for reprocessing UK and international spent nuclear fuels; it ceased operation in 2018.*

[2] *Obtained from FlyToMap website (http://flytomap.com)*

[3] *Obtained from Radioactivity in Food and the Environment report (Environment Agency et al., 2014)*

[4] *As Sellafield site has no significant liquid emissions to rivers, width of a medium-sized river has been chosen.*

#### 2.2.1.2 Solid waste

Radioactive solid wastes are classified according to their activities. The UK classification recognises four categories: High-Level Waste (HLW), Intermediate-Level Waste (ILW), Low-Level Waste (LLW) and Very Low-Level Waste (VLLW) (Wilson, 1996). HLW is the bulk of the fission product after vitrification; ILW mainly consists of sheared claddings from fuel rods and plutonium-contaminated materials; and V/LLW mostly comprises discarded equipment and materials from decommissioning activities. Other wastes, classified separately either because of their peculiar features or source or because they have not been declared as waste but could become waste in future, include Used Nuclear Fuel (UNF) assemblies to be disposed of without reprocessing; plutonium (Pu) retrieved from reprocessing of Spent Nuclear Fuel (SNF); Highly Enriched Uranium (HEU) containing 20% or more of uranium-235 (U235); and Depleted, Natural and Low Enriched Uranium (DNLEU) with U235 concentration below or only slightly above the naturally occurring concentration of ~0.71%. Depleted uranium includes enrichment tails, a by-product of the enrichment process. Low-enriched uranium arises from reprocessing of SNF, usually with concentration between 1 and 1.6%.

Management and disposal of nuclear solid waste continues to be much debated, without international agreement on a standard approach. Most countries with a civil nuclear industry (such as the UK, US, Sweden and Switzerland) have decided to treat and condition wastes and then consign HLW, SNF, Pu, HEU, DNLEU and most ILW to deep underground Geological Disposal Facilities (GDFs), with V/LLW and some short-lived ILW placed in near-surface repositories (WNA, 2016). However, there is no experience from which to assess the long-term potential impacts of nuclear waste stored in GDFs. For this reason, the solid waste pathway in CGM is based on the reference scenario of a preliminary study by Radioactive Waste Management Ltd. (NDA, 2010a) using a simplified analytical model for radionuclides migration (Carter et al., 2013; NDA, 2013). The methodology does not include V/LLW and short-lived ILWs in near-surface repositories because no performance assessment for such facilities appears to be available.

## Characterisation factors for radionuclides disposed in a Geological Disposal Facility

The methodology to calculate characterisation factors for radionuclides stored in a Geological Disposal Facility is based on the Post-Closure Safety Assessment (PCSA) that forms part of the safety case developed to support the design of a GDF in the UK. The potential impacts of leaks from the GDF are estimated by assessing the exposure doses and consequent risks incurred by a critical human population, termed the 'potentially exposed group' (IAEA, 2003; NDA, 2010b). The PCSA model comprises three sub-models: the engineered system, which includes the excavated vaults and their contents; the geosphere, i.e. the rocks surrounding the GDF and extending up to the surface; and the biosphere, defined as 10 $km^2$ of land surface representing the receptor for radioactive releases. The model is not specific to any particular site and therefore some of its parameters are generic estimates.

The main assumptions and limitations in the PCSA model are:

- The reference waste in the GDF is the 'derived inventory' estimated by Pöyry Energy Ltd. (2010a, 2010b, 2010c), based on the 2007 UK Radioactive Waste Inventory[5] (Pöyry Energy Ltd, 2008), with appropriate conditioning and packaging for each type of waste (NDA, 2010c).
- The only pathway for transport of radionuclides escaping from the GDF is through groundwater. A gas pathway is also considered in the qualitative analysis but is not included in the quantitative model because it is associated with high uncertainty.
- The GDF is constructed to the design for higher strength rock based on the phased geological repository concept (UK Nirex Ltd, 2003) and the Swedish SKB KBSD-3V concept (SKB, 2004). It includes separate areas for ILW, LLW and DNLEU and for HLW, SNF, HEU, and Pu.

The quantitative analysis predicts the mean annual radiological risk, showing the contribution of different wastes and radionuclides (see Figure 1 in the Supporting Information). Only those

[5] A more recent inventory was published in 2016, but the 'derived inventory" has yet to be updated.

radionuclides (usually no more than ten) showing the highest risk are included. The PCSA only presents predicted risks, with the fate and exposure calculations not reported explicitly. Therefore, the results are used here to calculate the characterization factors for emissions from solid waste, without using the Exposure and Effect modules (see Figure 1) introduced in subsequent Sections. Characterization factors for each waste type and radionuclide in the GDF are calculated as:

$$CF_{w,i} = \frac{Rm_{w,i}}{Q_{w,i}} \quad (2.1)$$

where:

$CF_{w,i}$ is the characterisation factor for radionuclide *i* in waste type *w* (risk/Bq y);

$Q_{w,i}$ is the amount of radionuclide *i* in waste type *w* (Bq);

$Rm_{w,i}$ is the average annual risk arising from radionuclide *i* in waste type *w*, calculated as:

$$Rm_{w,i} = \frac{\int_{t_{start\_w,i}}^{t_{max}} R_{w,i}(t)dt}{t_{max} - t_{start\_min}} \quad (2.2)$$

In Equation 2.2, the numerator represents the overall risk arising from radionuclide *i* contained in waste type w; whilst the denominator is the time range over which the risk for that waste type is not negligible. The individual terms are:

$R_{w,i}$ annual risk in year t arising from radionuclide *i* contained in waste type *w* (risk/y);

$t_{start\_w,i}$ time (in years) at which $R_{w,i}$ starts to rise; for example, this is expected to be about 173,000 years for Cs135 in HLW (NDA, 2010a);

$t_{start\_min}$ earliest time (in years) at which radiological risks from the whole mixed waste start to rise; for example, this is expected to be about 65,000 years for HLW (NDA, 2010a);

$t_{max}$ upper time boundary considered in the assessment, taken as a million years.

### 2.2.2 UCrad: a compartment-type methodology

Compartment-type models (also known as Mackay models, after their originator) are multimedia environmental models widely used to predict the fate of chemicals released into the environment. They were conceived to provide a basis for developing targeted strategies to reduce pollution from industrial processes (Mackay, 2001). Mackay's approach represents the environment in terms of distinct but interlinked media compartments (e.g. air, freshwater, seawater, agricultural soil, etc.) at different spatial scales (e.g. regional, continental and global). Substances are exchanged between different media at the same spatial scale and between compartments at different spatial scales within the same medium by advective/diffusive processes. Partition coefficients are used to represent how substances segregate at equilibrium between different environmental media and different phases within the same medium. Semi-empirical relations have been developed to predict substance-specific partition coefficients when they are not available from direct measurement or experiments. However, the available coefficients mainly refer to organic chemicals, and none are available for radionuclides.

Several multimedia compartment models have been operationalised for assessing the potential impacts of toxic substances within LCA, e.g. USES-LCA (Huijbregts et al., 2000), IMPACT 2002 (Jolliet et al., 2003), Eco-Indicator 99 (Goedkoop et al., 1998), CalTOX (Hertwich et al., 2001), USEtox (Rosenbaum et al., 2008). USEtox is the most widely used in LCIA; it focuses on organic substances and characterisation factors for inorganic substances are flagged as 'interim'. Even so, Joyce et al. (2016) used USEtox as the starting point for developing a model for assessing the impacts of Naturally

238 Occurring Radioactive Materials (NORM) (Goronovski et al., 2018). UCrad[6], the compartment-type
239 model developed in this work, was also developed by adapting USEtox to account for radionuclides
240 rather than organic chemicals. Like USEtox, UCrad is implemented in the Microsoft Excel environment.

241 #### *2.2.2.1 Features of UCrad*

242 UCrad uses a nested compartmental model comprising two scales: continental and global. The indoor
243 and urban scales that feature in USEtox have been removed as anthropogenic radioactive emissions
244 are highly unlikely to occur in densely inhabited areas. Figure 3 shows the eight environmental
245 compartments considered: air, fresh and seawater, natural and agricultural soil, freshwater and
246 marine sediments and groundwater. The features of all these compartments are based on the default
247 landscape used in USEtox version 2.0 (Rosenbaum et al., 2008; Shaked, 2012). However, only the first
248 five are recognised in USEtox. Freshwater and marine sediments have been added because, unlike
249 organic chemicals, radionuclides accumulating in these compartments can impact human health. The
250 sediment compartments are linked solely to the respective water compartment, and exchange
251 between these compartments occurs by sedimentation/re-suspension and adsorption/desorption
252 processes. The groundwater compartment has been added because most performance assessment
253 studies on Geological Disposal Facilities (GDFs) – e.g. those developed in Sweden (Kautsky et al., 2016),
254 Switzerland (Brennwald and van Dorp, 2009), UK (NDA, 2010a) and France (Andra, 2005) - recognize
255 groundwater as the main pathway by which radionuclides from stored wastes can reach the
256 biosphere. Other potential pathways, such as gas permeation through rocks or human intrusion
257 through wells, are either poorly studied or result from a probabilistic event and therefore fall outside
258 the scope of conventional LCA. As in the GLOBOX model (Wegener Sleeswijk and Heijungs, 2010), the
259 groundwater compartment is modelled as fed by water percolating from natural and agricultural soil

[6] Note that UC stands for University College (London), where the methodology was developed.

and linked to the ocean and freshwater through ground flows and to uncultivated and agricultural soils through irrigation.

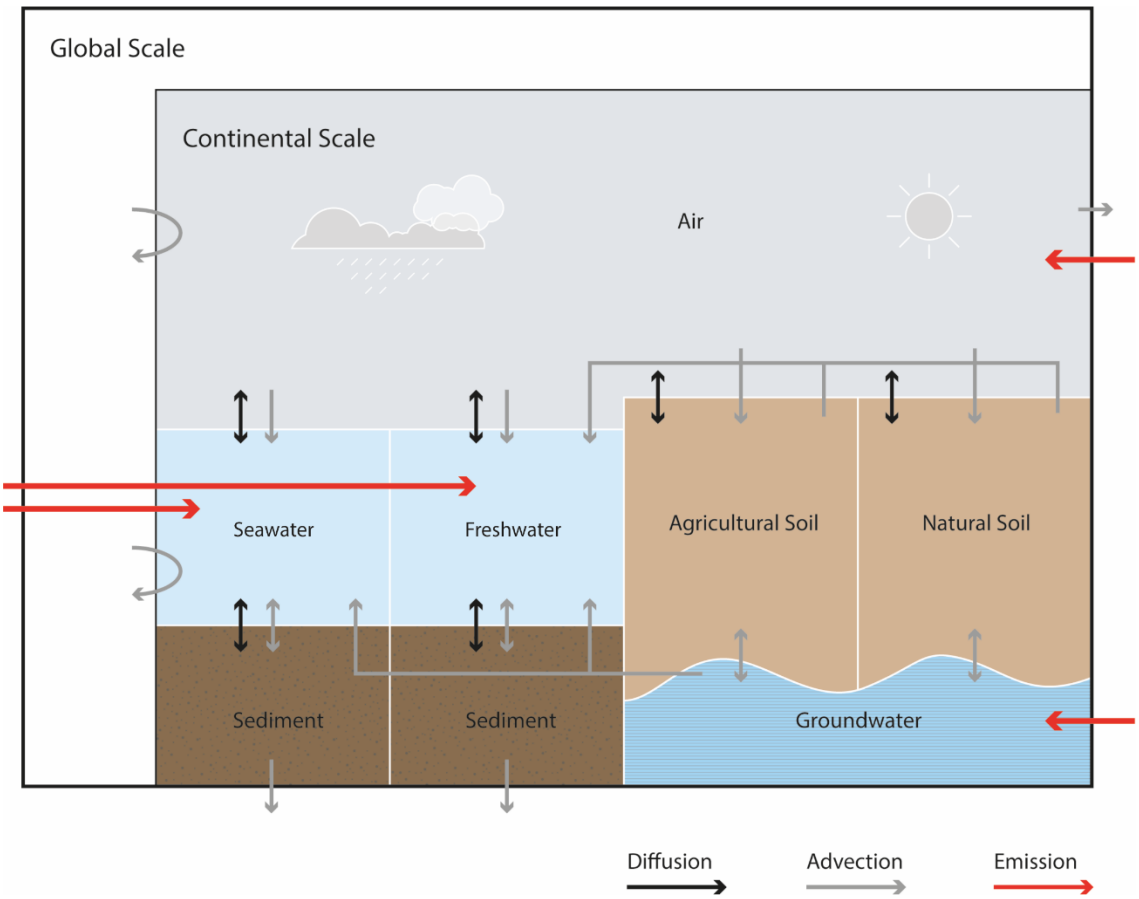


**Figure 3 – Compartments considered in UCrad fate model.**

Multimedia fate models incorporate two mechanisms of intermedium transport: partitioning and advection/diffusion. UCrad employs the same advective/diffusive equations as USEtox and uses substance-specific partition coefficients gathered from the literature; the chemical database supporting UCrad is discussed in Section 2.5. Unlike USetox, UCrad des not use predictive equations when partition factors are not available because those available are mostly suitable for organic chemicals. Air-water partition coefficients ($K_{aw}$) in UCrad are problematic as there are no values reported in literature for radionuclides. For this reason, based on the modelling of inorganic substances in USEtox, a default value of 1E-20 (Pa $m^3$ $mol^{-1}$) has been assigned to the majority of radionuclides, meaning that these nuclides behave as involatile solids. Only noble gases and a few

273 other radionuclides feature non-negligible $K_{aw}$ values; these have been obtained from the Henry
274 constants of the pure substance (Sander, 2015).

275 Carbon-14 (C14) and Tritium (H3) represent two special cases: following the approach used by IAEA
276 (20010), they are modelled as carbon dioxide and water vapour, the forms in which they are most
277 likely to be emitted.

278 UCrad treats radioactive decay of nuclides as a removal process, in the same way as USEtox treats
279 degradation of pollutants or migration of atmospheric pollutants to the stratosphere, and ignores
280 decay products (known as daughters). This means that UCrad characterisation factors represent only
281 the impacts of the radionuclides released and neglect the impacts of nuclides in the decay chain.
282 Improving treatment of the decay chain is one of the key points for further development of UCrad.

283 One of the crucial features of UCrad is its ability to evaluate the impact of solid nuclear waste disposed
284 in a Geological Disposal Facility (GDF). It is acknowledged that a failure in the engineered system will
285 eventually occur so that the GDF has two main purposes: to contain radionuclides for as long as
286 possible and, when a failure occurs, to delay the impacts of radionuclides by increasing their travel
287 time to the biosphere. For this reason, GDFs are constructed several hundred meters underground,
288 so that the layers of rocks that separate the GDF from the biosphere provide the retardation. The main
289 pathway by which radionuclides can pass from the engineered system to the biosphere is through
290 groundwater. UCrad and PCSA differ in the way transport through groundwater is modelled, as shown
291 in Figure 4. UCrad includes the groundwater compartment but cannot model the retardation explicitly.
292 In general, Mackay models are equilibrium models so that time is included implicitly as the time
293 needed to reach equilibrium. In the specific case of UCrad, retardation is included by treating
294 emissions from the GDF to groundwater as the “far-field flux” (i.e. the flux of radionuclides from the
295 geosphere into the biosphere after passing through several layers of rocks) rather than the “near-field
296 flux” of radionuclides leaving the GDF. The “far-field flux” is obtained from the generic Post-Closure
297 Safety Assessment report (NDA, 2010a) outlined in section 2.2.1.2. As a consequence, UCrad embodies

all the assumptions about the performance of the GDF made in the PCSA (e.g. type of waste containers, host rock, etc.) so that the two approaches are compatible and should be consistent.

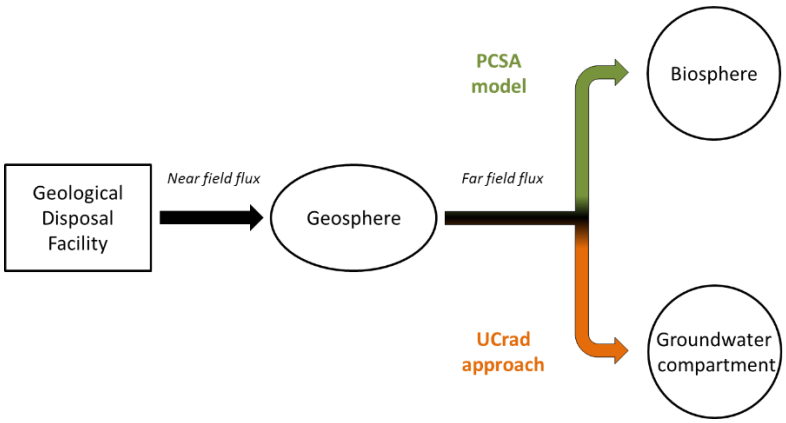


Figure 4 – Schematic representation of UCrad and PCSA approaches for GDF characterisation factors.

## 2.3 Exposure module

The Exposure Module represents the second step of the framework (see Figure 1). The outcome is the time-weighted concentration of radionuclides to which humans are exposed due to an increase in environmental radiological concentrations following a radioactive discharge, and the amount of radionuclides taken in as a result. The Exposure Module does not depend on the approach adopted to calculate the environmental concentrations and therefore it is common to both CGM and UCrad. The location of the pollutant in the environment and its physical state affect the pathway by which exposure occurs. Two main pathway categories are identified: external and internal. Airborne and deposited radionuclides contribute to the external pathway, chiefly through gamma radiation; alpha and beta radiations operate over very short distances, so that their contribution to the external pathways is negligible. Intake of radionuclides through ingestion and inhalation constitute the internal pathway. Ingestion of radionuclides is caused by transfer of radionuclides to crops and cattle, for instance due to wet and dry deposition. “Usage factors” are employed to establish consumption patterns of contaminated food and water, whilst “exposure factors” are used to estimate average time that individuals are exposed to a contaminated environment. Usage factors have been taken from USEtox (2015), whilst exposure factors have been adapted from IAEA (2001).

As in the UCrad fate module, H3 and C14 require special treatment; both H3 and C14 can be incorporated in many different chemical compounds within the human body so that assessment of their potential impact is too complex to be incorporated into generic methodologies. Exposure to H3 and C14 is modelled using Specific Activity Models (SAMs) (IAEA, 2001), based on the assumption that the ratio between a radioactive nuclide and its widespread stable form is fixed at equilibrium.

## 2.4 Effect module

The Effect Module (see Figure 1) assesses the consequences on human beings of exposure to and intake of radionuclides. This is achieved in a two-step process: first, the effective dose is calculated; second, this is translated into a risk metric for detrimental effects. The ICRP defines three types of doses: absorbed, equivalent and effective (ICRP, 2007). The absorbed dose refers to the amount of energy imparted by ionizing radiations per unit of body mass. From the absorbed dose, the equivalent dose is obtained by considering the biological effectiveness of the radiation, which depends on its type and energy. Finally, the effective dose is derived from the equivalent dose by accounting for the biological tissue involved in the process, measured in Sieverts (Sv) and calculated by means of established conversion factors (see Section 2.5). Ionising radiations can result into two different effects on human beings: deterministic and stochastic (ICRP, 2007). Deterministic effects result mainly from killing/malfunctioning of cells; they are nil below a specific threshold and increase linearly above it. Stochastic effects, on the other hand, accounts for modifications of cells, which may cause cancers and heritable effects. They occur for low doses (less than 100 mSv), have no threshold, and their likelihood increases linearly with the dose; this is the so-called (and much debated - e.g. see Allison, 2015) Linear No-Threshold model. CGM and UCrad consider only stochastic effects because they apply only to releases leading to low doses. The nominal risk coefficient, for both fatal and non-fatal cancer, is taken as 5.5 $10^{-2}$ $Sv^{-1}$ (ICRP, 2007). Hereditary effects with much lower probability are not considered, to achieve consistency with other LCIA methodologies.

## 2.5 Database of radionuclide properties

A database containing physico-chemical properties and dose/risk conversion factors for more than 100 radionuclides has been compiled to support both methodologies (Paulillo et al., 2019). The database covers radionuclides considered by IAEA (2001), augmented with some radionuclides of crucial importance in GDFs; i.e. radionuclides with very high half-lives not directly discharged from routine operations but so long-lived that they will still be present in the GDF when failure is assumed to occur. Experimental data have been preferred over estimates, with the exception of the water-sediment distribution factor that has been estimated as one tenth of the water-suspended sediment factor as suggested by the IAEA. As knowledge of the behaviour of radionuclides in the environment is still limited, data for a number of radionuclides are totally or partially missing. IAEA (2010) suggests the use of analogues when specific data are missing, recognising three types of analogues: isotopes, elements and species. Analogue isotopes are used in the main, as most data (e.g. bio-transfer factors) refer to elements rather than specific isotopes. In the few cases where data for specific elements are missing altogether, analogue elements (i.e. elements with similar chemical properties) provide the most reliable estimates. Furthermore, because several authoritative sources of radionuclide properties databases are available, a hierarchical approach for data selection has been applied; i.e. the most comprehensive source was used first, continuing to less comprehensive sources only for data not available in a preferred source. The main sources for parameters used in UCrad and CGM, ranked according to the hierarchical approach, are reported in the Supporting Information. Parameters not listed have been taken from USEtox.

# 3 Results

## 3.1 Characterisation factors

Characterisation factors calculated according to CGM and UCrad methodologies are reported in Paulillo et al. (2019). These include factors for direct discharges of 107 and 115[7] radionuclides obtained from CGM and UCrad respectively, and for emission from nuclear waste of around ten radionuclides. For CGM we have set a distance from the point of release of 1000 km, corresponding roughly to the scale of the European continent. The effect of varying the distance is explored in a complementary article.

Characterisation factors are reported both in absolute terms as yearly risk per Bq released, and in relative terms as Bq equivalent; these are obtained by dividing the impact factor for each nuclide by that for a reference substance emitted to a specific environmental compartment. Uranium-235 (U235) emitted to air is the reference radionuclide for air, seawater and freshwater categories; while uranium-238 (U238) in HLW is the reference for emissions from the GDF. Using two different reference substances prevents misleading comparisons between impacts of direct discharges and solid waste, which occur on considerably different time scales.

## 3.2 Comparison with Human Health Damages approach

Figure 5 compares characterisation factors from the Human Health Damages (HHD) approach with those obtained from UCrad and CGM, for emissions to air and to fresh and seawater. Two sets of HHD characterisation factors are reported; they represent two different cultural perspectives, namely Individualist (I) and Egalitarian/Hierarchist (E/H), as defined by cultural theory (Thompson et al., 1990). The factors obtained from the E/H version of the HHD model are used as the reference set against

[7] The additional eight radionuclides included in UCrad factors for direct discharges are actinium-227 (Ac227), beryllium-10 (Be10), chlorine-36 (Cl36), selenium-79 (Se79), tin-126 (Sn126, thorium-229 (Th229), uranium-233 (U233), uranium-236 (U236). They are not included in the IAEA framework on which CGM is based.

383 which the other factors are compared. In Figure 5, characterisation factors obtained from UCrad and
384 CGM and from the Individualist version of HHD are shown on the ordinate, with the factors from the
385 E/H HHD model as the parameter on the abscissa.

386 The Mean Log Deviation parameter, defined as the average value of the logarithm (to base 10) of the
387 ratio of the characterisation factors of each radionuclide, has been used to quantify the average
388 difference between sets of characterisation factors and the reference set. MLD values are reported in
389 Table 2, with values for individual radionuclides given in Paulillo et al. (2019). A nil MLD value indicates
390 that the two sets of characterisation factors compared are consistent on average across the whole
391 set, although individual factors may show high deviations. A negative MLD indicates that the reference
392 values are generally larger than the set being compared against them; a positive MLD indicates the
393 converse comparison.

394 Individualist (I) characterisation factors show the least deviation from the reference set for emissions
395 to each environmental medium, with MLD values in the order of 0.01 (Table 2). This is perhaps to be
396 expected, given that the two sets of factors result from two variants of the same model.

397 The UCrad set is the next closest, featuring MLD lower than unity, indicating order-of-magnitude
398 agreement, for all radionuclides and receiving media. For most emissions to air, log deviations are
399 around 0.1, indicating good general agreement over the range of species considered. The largest
400 deviation is given by C14 and a noble gas, radon-222 (Rn222), which feature log deviations of about -
401 4 and -6 respectively. Characterization factors for emissions to freshwater feature a MLD around +0.2,
402 whilst the MLD value for emissions to seawater is about +0.1. As for air emissions, the largest deviation
403 is given by C14 for emissions to seawater; emissions of C14 to freshwater are not considered by the
404 Human Health Damages methodology.

405 The CGM methodology also gives factors close to the E/H HHD model for air and seawater emissions,
406 with MLD values approximately -0.6 and -0.8 for air and seawater emissions. As for UCrad, the largest
407 deviations are given by air emissions of C14 and Rn222. However, the factors for CGM are considerably

higher than those for HHD for emissions to freshwater, with log deviations ranging from ~1.5 (caesium-137) to ~5 (silver-110m) and an average value of ~3. The significance of these comparisons is introduced in Section 4.2 below and investigated in detail in the complementary paper.

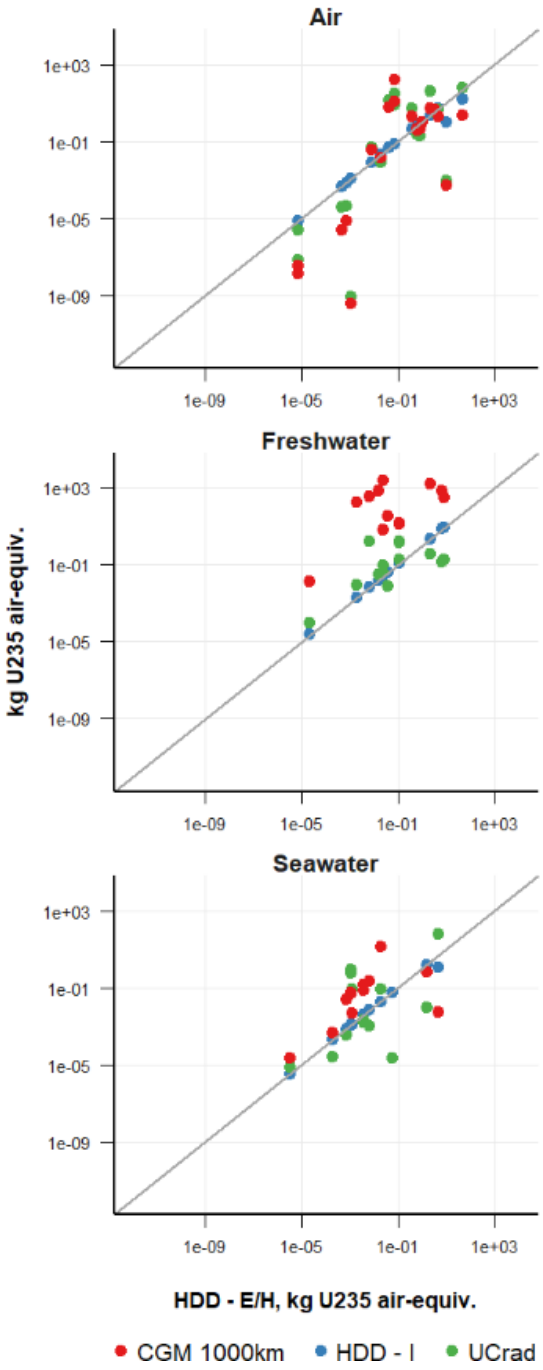


**Figure 5 – Comparison of characterisation factors obtained from HHD, CGM and UCrad models; E/H: Egalitarian/Hierarchist; I: Individualist.**

Table 2 - Values for Mean Log Deviation (MLD) of Human Health Damages (HHD-I), Critical Group Methodology and UCrad relative to Egalitarian/Hierarchist version of HHD model.

| | MEAN LOG DEVIATION | | |
|---|---|---|---|
| | Air | Freshwater | Seawater |
| Human Health Damages – Individualist | -0.058 | +0.017 | -0.035 |
| Critical Group Methodology | -0.63 | +3.1 | +0.88 |
| UCrad | -0.30 | +0.18 | +0.097 |

# 4 Discussion

In this Section we first analyse the main similarities and differences of CGM and UCrad, and, where appropriate, relate them to the essential requirements for radiological impact assessment methodology introduced in Section 1: the ability to account for both direct discharges and emissions from a geological repository, and to produce average site-independent estimates of impacts. Then, we discuss the quantitative comparison between characterisation factors obtained from CGM and UCrad and those from the Human Health Damages methodology.

## 4.1 Similarities and differences of CGM and UCrad

As outlined in Sections 2.1 and 2.2, the only methodological difference between CGM and UCrad lies in the approach to fate modelling: CGM employs analytical models based on simplifying assumptions to solve the basic radionuclide transport equations, whilst UCrad treats the environment as divided into homogeneously mixed compartments, each representing an environmental medium. The generic approaches behind the two models stem from developments to address two very different objectives. Analytical models are generally used in risk assessment studies to estimate the highest possible impacts that human beings may experience, whilst compartment-type models are widely used in LCIA

433 to provide estimates of the average impacts of pollutants. Because LCA quantifies average rather than
434 worst case impacts, we use the critical group in CGM to allow selection of an appropriate distance
435 from the source at which radiological impacts should be assessed. In this way CGM, like UCrad, is
436 intended to produce average estimates of impacts. The different approach to fate modelling has two
437 further implications. First CGM assumes that the population is located at a particular distance from
438 the source of emissions, enabling calculation of dilution due to dispersion of the radioactive plume
439 and the resulting reduction in radiological dose to the population. The effects of this assumption are
440 explored in more detail in a complementary paper. Second, this makes CGM site-dependent because
441 it relies on some specific environmental parameters; in this work, values appropriate to the UK have
442 been used.

443 The two methodologies also differ in their time scales: Level III Mackay models – i.e. the type of
444 compartment models generally used for LCIA purposes - assume steady-state conditions, whilst CGM
445 fate models employ time-dependent equations to estimate dispersion of radionuclides in the
446 environment. However, CGM assumes that the release of radionuclides has occurred for 30 years or
447 more. With this condition, it can be assumed that steady-state conditions are reached in the soil and
448 therefore bio-accumulation factors may be used to relate soil and food chain concentrations (e.g. in
449 vegetables and cattle). Thus both models actually describe steady state conditions following the
450 release of radionuclides, although steady state exposure is reached sooner in the CGM model. This
451 has implications for the modelling of radioactive decay, explored in the complementary paper. Both
452 UCrad and CGM ignore in the fate analysis the formation of “daughter” radionuclides through
453 radioactive decay, but this does not represent a limitation to CGM because of the lower time required
454 by radionuclides to travel from emission sources to critical groups compared to the time to reach
455 steady-state conditions in UCrad.

456 Both UCrad and CGM include impacts from radionuclides in nuclear waste; to do so they rely upon the
457 Post-Closure Safety Assessment (PCSA) of a generic repository developed by Radioactive Waste

458 Management Ltd. (NDA, 2010a) for characterising emissions from solid waste, as explained above. The
459 PCSA study uses a model, publicly available only in outline and thus not customizable, that estimates
460 the potential impacts of radionuclides that escape the GDF and expose a small village located just
461 above the GDF; the area impacted is in the order-of-magnitude of ten $km^2$. This means that for
462 emissions from solid waste the critical group in CGM is located up to about 3.3 km from the release
463 source, and cannot be amended. The same limitation does not apply to UCrad because the model only
464 uses the far-field flux of radionuclides from the repository. Relying on the PCSA model introduces
465 further limitations. First, it constraints both CGM and UCrad to the radionuclides considered in PCSA,
466 i.e. the ten (or fewer) radionuclides expected to have the greatest impact. Second, it also limits their
467 applicability to a GDF located in either high or low strength rock formation. PCSA focuses on high
468 strength rock formation through the assumption that groundwater is the most critical escape pathway
469 for radionuclides. Because groundwater is likely to be present in both high and low strength rock,
470 characterisation factors for emissions from solid waste are applicable to both cases. However they are
471 not applicable for a GDF located in a salt rock formation where the presence of water is highly unlikely.
472 This represents an important area for development of the models: to enable characterisation factors
473 to be obtained for different GDF designs and geologies, to support decisions over the siting of a GDF.
474 As GDFs are developed in many countries, it may be expected that further safety assessments will be
475 produced, thus providing data for such a development.

476 In addition to the assumptions in the PCSA, CGM and UCrad rely on a number of other assumptions,
477 whose consequences cannot be assessed due to lack of data in the literature. The most notable
478 concern partition factors, specifically for air-water, and bio-accumulation/transfer factors. A negligible
479 air-water partition coefficient has been attributed to the majority of radionuclides to represent
480 negligible volatility, whilst for a number of radionuclides the bio-accumulation factors have been
481 estimated by the analogue elements approach. Both parameters control the models for dispersion in
482 environmental media, and thus can have a strong effect on the characterisation factors. For instance,

483 a higher air-water partition coefficient leads to higher concentrations in the atmospheric
484 compartment and in turn leads to higher characterisation factors. This is explored in the
485 complementary article.

486 Finally, both methodologies are also deficient in accounting for the impacts of radionuclides contained
487 in Very Low or Low Level Waste (VLLW or LLW) disposed in near-surface repositories, because no
488 analysis like the PCSA study appears to have been published or to be in the public domain for these
489 repositories. This represents a serious limitation and must be a priority area for future model
490 development.

## 491 4.2 Comparison of CGM and UCrad with Human Health Damages approach

492 The characterisation factors for direct discharges estimated by the Human Health Damages
493 methodology have been sufficiently reviewed to serve as an appropriate basis for validating the
494 methodologies proposed here. The methodology however is not generally considered sufficiently
495 established for inclusion in LCIA, primarily because it fails to include emissions from solid waste
496 (Paulillo et al., 2018).

497 Despite the significant differences between CGM and UCrad discussed above, and also between both
498 methodologies and HHD, the quantitative comparison shows that there is general agreement between
499 their characterisation factors, indicated by the Mean Log Deviation parameter. The only substantial
500 deviation occurs for emissions to freshwater for which the CGM methodology consistently generates
501 higher characterisation factors than both UCrad and HHD. The explanation for this lies in how dilution
502 is modelled for emissions to freshwater compared to air and seawater; this is investigated in detail in
503 a complementary article. For all other emissions except atmospheric emissions of Rn222 and C14, the
504 methodologies agree within an order of magnitude. As expected the lowest deviation is found
505 between the Egalitarian/Hierarchist and Individualist characterisation factors of the HHD
506 methodology, with MLD values around 0.01.

Because UCrad shows lower average absolute deviations from HDD than CGM, especially for emissions to freshwater, the comparison suggests that UCrad should be preferred over CGM. The case for UCrad is also reinforced by the recommendations of the JRC to ensure a better compatibility between radiological impact assessment methodologies and USEtox (JRC, 2011). In a complementary article we assess quantitatively and in detail the differences between CGM and UCrad, and suggest a practical rule for their application.

# 5 Conclusions

Radiological impacts have generally been omitted from Life Cycle Impact Assessment, primarily due to the lack of an accepted methodology for including them alongside non-radiological impacts. An appropriate framework for including radiological impacts in the Impact Assessment phase of LCA (LCIA) can be based on three modules, covering Fate, Exposure and Effects of radionuclides, combined to yield characterisation factors. This framework can be developed in two ways, respectively using approaches applied in Human and Environmental Risk Assessment (HERA) and in Life Cycle Impact Assessment (LCIA). The former approach is the Critical Group Methodology (CGM) and the latter is UCrad, developed in the present work. These two methodologies differ only in the way radionuclides fate is modelled: CGM uses established analytical models for dispersion and transport of radionuclides in the environment, whilst UCrad employs a compartment-type model adapted from USEtox , widely used in LCIA. Both methodologies produce average estimates of impacts and can include impacts from nuclear waste disposed in a Geological Disposal Facility (GDF). The main limitation of both methodologies lies in their reliance on the Post-Closure Safety Assessment model for characterisation of emissions from solid waste. This limits the number of radionuclides considered and constrains application of the methodologies to a Geological Disposal Facility (GDF) in a specific geology.

Characterisation factors have been calculated by both methodologies for over 100 radionuclides for direct discharges to air, fresh and seawater, and for around ten radionuclides emitted from different

types of nuclear wastes disposed in a GDF. The validities of the methodologies are confirmed by comparing the resulting characterisation factors with the factors obtained from the only other approach that has been implemented in LCA: the Human Health Damages (HHD) methodology (Frischknecht et al., 2000). UCrad characterisation factors show better agreement than CGM with the Human Health Damages (HHD) methodology but, with the exception of factors from CGM for emissions to freshwater, the results from both methodologies differ by less than an order of magnitude from the factors obtained by the Human Health Damages approach.

Together, UCrad and CGM represent a fundamental step towards incorporating ionising radiation impacts in LCIA, especially because they include both direct discharges and emissions from a geological repository. It is suggested that UCrad should be preferred over CGM because it shows better agreement with HHD and ensures better compatibility with USEtox. A detailed comparison between these methodologies is the focus of a complementary article.

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# 7 Glossary

| | |
|---|---|
| CGM | Critical Group Methodology |
| GDF | Geological Disposal Facility |

| | |
|---|---|
| DNLEU | Depleted, Natural, Low Enriched Uranium |
| HHD | Human Health Damages |
| HLW | High Level Waste |
| IAEA | International Atomic Energy Agency |
| ILW | Intermediate Level Waste |
| LLW | Low Level Waste |
| MLD | Mean Log Deviation |
| PCSA | Post-Closure Safety Assessment |
| SNF | Spent Nuclear Fuel |
| VLLW | Very Low Level Waste |

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