Radiological Impacts in Life Cycle Assessment. Part II: Comparison of Methodologies

3 Abstract

4 In a complementary article, an overarching framework was proposed to include radiological impacts 5 in Life Cycle Impact Assessment (LCIA). Two methodologies were derived embodying the framework: 6 the Critical Group Methodology (CGM), adapted from the approach commonly used in Human and 7 Environmental Risk Assessment (HERA), and UCrad, based on the compartment modelling approach 8 commonly used in LCIA. In this paper, characterisation factors obtained by the two methodologies 9 are compared in detail to investigate the consequences of the different approaches to fate modelling 10 and the sensitivity of the characterisation factors to the radionuclides' half-life. Characterisation 11 factors from the CGM methodology are strongly affected by radioactive decay at low half-life and by 12 dilution at large distances. Conversely, UCrad factors are not affected by dilution and are affected less than CGM by radioactive decay. It is concluded that UCrad is more appropriate than CGM for LCA 13 14 because it is consistent with the general approach used in LCIA. However, CGM can be used alongside 15 UCrad to make recommendations on the location and scale of specific processes emitting 16 radionuclides.

17 Keywords

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

20 1 Introduction

Life Cycle Assessment (LCA) aspires to provide a complete analysis of the environmental impacts of 21 22 delivering products and services. The impacts are usually estimated as contributions to a recognised 23 set of impact categories. However, in the absence of a generally accepted approach to assessment, 24 radiological impacts have usually been omitted. To correct this omission, Paulillo (2018) and Paulillo 25 and colleagues (2019a) proposed a general framework for inclusion of radiological impacts in Life Cycle 26 Impact Assessment (LCIA), leading to two methodologies to model the fate of radioisotopes and the 27 resultant impacts: UCrad and Critical Group Methodology (CGM). The two methodologies respectively represent two different cultures: Life Cycle Assessment (LCA) and Human and Environmental Risk 28 29 Assessment (HERA). LCA assesses average potential impacts of a very large number of pollutants 30 released over the life cycle delivering a product or service, calculated per functional unit of the product 31 or service. As the life cycle may be spread over many locations around the world, LCIA is usually not 32 site-specific and is concerned with global rather than regional scales. UCrad adopts this approach. By 33 contrast, HERA quantifies the actual, absolute risks to humans and the environment associated with the release of pollutants from a specific process whose location is defined. The assessment is carried 34 35 out on a regional scale, usually with the focus on a few selected substances of relevance to the process 36 under study. The Critical Group Methodology (CGM) follows the general approach used in HERA. As 37 noted by a number of authors (Nishioka et al., 2002; Saouter and Feijtel., 2000; Sonnemann et al., 38 2004), LCA and HERA may lead to different results when applied to the same process. Udo de Haes et 39 al. (2006) concluded that, although the two tools differ in a number of respects, the fundamental 40 difference lies in the use of the functional unit, which differentiates the relative impacts of LCIA from 41 the absolute ones of HERA. Further differences, similarities and application of these tools have been 42 discussed by Cowell et al. (2002), Olsen et al. (2001), Owens (1997), Udo de Haes et al. (2006), Wegener Sleeswijk et al. (2003) and Wrisberg et al. (2002) amongst others. 43

The general framework and the two methodologies are presented in a complementary paper (Paulillo 44 et al., 2019a). The objective of the present paper is to provide a detailed and quantitative comparison 45 46 between the results of the UCrad and CGM approaches, to relate the differences between them to 47 the underlying assumptions in fate modelling. The consequences of implementing a risk assessmentbased methodology within the LCA framework are examined by comparing results from CGM 48 49 quantitatively with those from the approach commonly used in LCA, represented by UCrad. Finally an 50 approach combining both methodologies is proposed to meet the need, identified by some authors 51 (e.g. Flemström et al., 2004; Udo de Haes et al., 2006), for deeper integration between LCA and HERA. 52 The article begins with an overview of the main features and differences between the fate models 53 used in UCrad and CGM (Section 2). Section 3 presents a detailed comparison between the 54 characterisation factors obtained from the two methodologies. Section 4 discusses the significance of 55 the comparisons, in particular on the significance of radionuclide half-life, and proposes a way to combine the two approaches. Finally, the main findings of the article are summarised in Section 5. 56

57 2 Overview of fate module of CGM and UCrad

Paulillo (2018) and Paulillo et al. (2019a) set out the overarching framework within which the two methodologies for radiological impact assessment have been developed; they differ solely in the transport/dispersion models employed in the fate analysis. The focus here is on how these different approaches and the radionuclides' half-lives affect the resulting characterisation factors. This section gives an overview of the main features and differences between the fate modules of the two methodologies.

The fate module in UCrad follows the widely-used multimedia fugacity approach developed by Mackay
(2001) to predict the distribution of emitted species between different environmental media. Mackay
models are not new to LCIA: they are commonly used to assess the impact of toxic pollutants, USEtox
being a notable example (Rosenbaum et al., 2008). In Mackay models, each medium is represented as

a homogenously mixed compartment, while exchange between compartments occurs by advection
and diffusion. The fate module in UCrad uses a nested compartmental model comprising two spatial
scales: continental and global. The resulting environmental concentrations represent steady-state
conditions. In principle, UCrad is able to consider emissions to all compartments at both scales.
However, for the purpose of comparing the two methodologies, this article considers direct emissions
from routine operations into three compartments: air, fresh and seawater. Results for emissions from
a Geological Disposal Facility (GDF) into groundwater are included in the Supporting Information.

75 The Critical Group Methodology (CGM) is conceptually different: it sets out to estimate the impact of 76 emissions on a specific group of humans, referred to as the "Critical Group". The concept of "Critical 77 Group" was developed for Risk Assessment; it represents the group of people expected to receive "the 78 highest dose due to their lifestyle, location and habits" (ICRP, 1990; NRPB, 1993). The critical group 79 does not necessarily represent a worst-case scenario; rather, it allows selection of the most appropriate distance at which to assess radiological impacts. As explained by Paulillo and co-workers 80 81 (2019a), the fate module of CGM relies on two earlier studies with different purposes: models 82 developed by the International Atomic Energy Agency (IAEA, 2001) are used to describe routine direct 83 discharges from processing operations, whilst the Post-Closure Safety Assessment (PCSA) (NDA, 2010), 84 developed by Radioactive Waste Management Ltd. (RWM), is employed for long-term unplanned 85 emissions from a GDF. For direct discharges, characterisation factors were developed for emissions to air, river and seawater. The behaviour of species discharged is described by analytical models and the 86 resulting predicted concentrations depend on a number of site-dependent parameters such as height 87 88 of atmospheric emission, width of river, etc. With the purpose of making the methodology generally 89 applicable, Paulillo et al. (2019a) based the CGM model on generic values for all parameters except 90 one: the distance of the critical group from the emission source. The significance of this parameter is 91 explored in detail in the present paper. The transport model used by the PCSA is similar to those used 92 by the IAEA, but is applied to a different medium: groundwater. The model however is not configured

93 to produce distance-dependent characterisation factors; therefore, comparison of characterisation
94 factors for emissions from a GDF is not pursued in significant detail in this article.

95 **3 Results: Comparison of methodologies**

96 This Section presents a detailed comparison between characterisation factors calculated by the CGM 97 and UCrad methodologies presented by Paulillo and colleagues (2019a), for continuous emissions to 98 three receiving environmental media (air, freshwater and seawater) and for four distances from 99 source to receptor modelled using the CGM approach. The comparison for emissions from a GDF is 100 included in the Supporting Information, but not discussed in detail because the CGM is not set up to 101 produce distance-dependent characterisation factors for these emissions.

The differences between the two sets of characterisation factors are expressed in terms of logarithmic deviations, i.e. the logarithms to base 10 of the ratio between the factors, which indicates order-ofmagnitude discrepancies (see Paulillo et.al., 2019a). The Mean Log Deviation (MLD), quantifies the average order-of-magnitude discrepancy between factors: a nil MLD indicates general agreement between the methodologies; a negative MLD indicates that the reference values are generally larger than the set being compared against them; and a positive MLD indicates the converse relationship.

Figure 1 reports Mean Log Deviations for characterisation factors obtained from the CGM model compared to values given by UCrad, as functions of the half-lives of radionuclides. Numerical values of characterisation factors and log deviations are reported in Paulillo et al. (2019b). MLD values for each set of characterisation factors are reported in Table 1 for all values of half-life and for values higher than 7e+07 s (i.e. 2.2 years), approximately equal to a value of the decay constant of 1e-09 s-¹. This time is comparable to the slowest environmental transport covered by the model: transport over 10,000 km in seawater.

- Table 1 Mean log deviations of CGM characterisation factors relative to UCrad for different receiving media and distances of the critical group.

	MEAN LOG DEVIATION			
DISTANCE	All values of half-life	Half-life > 7e+07 s		
Air emissions				
1 km	5.7E+00	5.2E+00		
100 km	2.1E+00	2.0E+00		
1 000 km	-2.4E+00	1.8E-01		
10 000 km	-1.2E+01	-1.6E+00		
Freshwater emissions				
1 km	4.0E+00	2.9E+00		
100 km	3.4E+00	2.9E+00		
1 000 km	-1.7E+00	2.9E+00		
10 000 km	-9.1E+00	2.9E+00		
Seawater emissions				
1 km	5.2E+00	3.9E+00		
100 km	-2.8E+00	1.7E+00		
1 000 km	-9.0E+00	4.8E-01		
10 000 km	-2.5E+01	-7.5E-01		



Figure 1 - Log deviations of CGM characterisation factors relative to UCrad as a function of half-life, for air, fresh and seawater emissions and for four distances of the critical group.

Figure 1 reveals two clear regimes that are consistent across all receiving media and distances of the receptor greater than 1 km. For high half-life, log deviations are independent of half-life; at low halflife, log deviations sharply decrease with decreasing half-life. The boundary between these two regimes is given by values of half-life that increase with the distance and differ between environmental media. This is most evident for seawater, where MLD values decrease from ~5 to ~-25 (Table 1). The trend is weaker for emissions to air and freshwater, with MLD diminishing from ~6 to ~-12 for air and from -4 to ~-9 for freshwater. Thus, the best agreement between the methodologies is found for a distance between 100 and 1,000 km for emissions to air and freshwater, and between 1 and 100 km for emissions to seawater. The two regimes, low and high half-life, are analysed separately in Figure 2 and Figure 3. The results for emissions to seawater in Figure 1 also reveal some outliers; these are most evident in Figure 3 and are discussed below.

133 Figure 2 shows log deviations between the two models against the ratio of isotope half-life to transit 134 time from source to receptor for each distance from source to receptor and each environmental 135 medium. The transit times are calculated as the ratio of the distance between source and receptor to 136 the speed of the carrier, i.e. wind for emissions to air, river flow for emissions to freshwater and 137 marine current for emissions to seawater; they are reported in Table 2. The curves for low half-life coincide when shown as a function of the ratio of half-life to transit time. The sharp decrease of log 138 deviations starts when half-life is of comparable magnitude to transit time; i.e. the ratio is around 139 140 unity. This indicates that the distinction between the two regimes depends on whether a significant 141 portion of nuclides decay before reaching the receptor in the CGM model.



Figure 2 - Log deviations between characterisation factors from CGM and UCrad models as a function of the ratio of halflife to transit time, for air, fresh and seawater emissions and for four distances of the critical group.

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Table 2 – Transit times for emissions to air, freshwater and seawater in CGM

		Transit time (s)	
Distance (km)	Air	Freshwater	Seawater
1	5.00E+02	9.26E+02	1.00E+04
100	5.00E+04	9.26E+04	1.00E+06
1 000	5.00E+05	9.26E+05	1.00E+07
10 000	5.00E+06	9.26E+06	1.00E+08

Figure 3 reports log deviations for CGM factors relative to UCrad factors against the distance for values of half-life greater than 7e+07 s; i.e. where nuclide decay between source and receptor is negligible. Log deviations decrease considerably with distance for emissions to air, from an average value of ~5 at 1 km to ~-1 at 10 000 km (Table 1), and for emissions to seawater, from ~4 to ~-0.7, but remain constant for emissions to freshwater, at ~3. For high half-life, the best agreement between UCrad and CGM is found for a distance between 1 000 and 10 000 km for emissions to air and seawater, whilst for emissions to freshwater the methodologies do not converge.

Figure 3 uses colour-coding to rank nuclides from greatest log deviation (red) to least (blue). The ranking does not change with distance for emissions to air and freshwater. For emissions to seawater, the ranking changes between a distance of 1 km and distance greater than 100 km. This behaviour is exemplified by Eu155 and Co60, whose log deviations increase from 1 km (respectively about 3 and 2.4) to 100 km (~4), and by U234 for which the value remains roughly constant at about 0.4. From 100 to 10 000 km, the log deviations of these nuclides decrease in line with those of other nuclides.



Figure 3 - Log deviations for half-life higher than 7e-07s between characterisation factors of CGM and UCrad as a function of distance for air, fresh and seawater emissions. Colours identify different nuclides, and are ranked from red to blue

165 according to log deviations at 1 km.

166 **4 Discussion**

167 4.1 Comparison between CGM and UCrad

The comparison between the characterisation factors from the CGM and UCrad methodologies for different receptor distance and receiving media gives interesting insight into the two methodologies; notably, it highlights that the deviations between the factors from the two methodologies depend on half-life at low half-life and on the distance from the source of the emission to the receptor at all halflives.

173 Figure 1 and Figure 2 highlight the distinct regimes for low and high half-life; the boundary between 174 the regimes does not depend on half-life only (as shown in Figure 1), but rather on the value of half-175 life relative to transit time (Figure 2). When half-life is lower than the transit time, radioactive decay 176 is faster than dispersion of radionuclides between source and receptor (left side of Figure 2). By 177 contrast, dispersion is faster than decay when half-life is higher than transit time (right side of Figure 178 2). The transition from one regime to the other occurs when half-life is comparable to the transit time. 179 The ratio between half-life and transit time is analogous to the inverse of the Damköhler number 180 (Fogler, 2006) used in Chemical Reaction Engineering to relate the timescale of the transport 181 phenomena inside a reactor to that of the chemical reaction: when the Damköhler number is less than 182 one, the chemical reaction is slow compared to the transport processes, and thus conversion of 183 reactants into products is low; a higher conversion is achieved when it is greater than one, that is when 184 the chemical reaction is faster than the transport processes.

When half-life is lower than transit time, the CGM factors are strongly dependent on half-life, decreasing sharply with decreasing half-life down to negligible values compared the factors from UCrad. The fate models in CGM use time-dependent analytical models, such as the Gaussian plume for atmospheric emissions, to simulate dispersion from source to receptor. The model allows for the transit time required for nuclides to reach the receptor and therefore includes decay. By contrast,

190 UCrad employs compartment-type models at steady state conditions. Radioactive decay is 191 independent of time and is modelled as a removal process from each compartment so that the 192 dependence on half-life is less explicit and less strong.

193 The log deviations for half-life greater than transit time (Figure 3) show that CGM factors are still 194 dependent on the distance between source and receptor for emissions to air and seawater: the higher 195 the distance, the lower are the characterisation factors. This effect is also present in the results for 196 low half-life but is obscured by the stronger dependence on radioactive decay. The analytical models 197 for emissions to air and seawater in CGM account for dispersion and resultant dilution to estimate 198 how the separation distance affects the concentration to which the receptors are exposed; i.e. the 199 radiological doses to the critical group. In contrast to air and seawater, factors for emissions to 200 freshwater remain constant with distance because dilution is not considered for this environmental 201 medium: CGM models freshwater bodies as rivers, so that dispersion is constrained by the river banks.

202 Because in CGM dilution applies equally to all radionuclides, the ranking of radionuclides with respect 203 to their characterisation factors remains the same at different distances; this is shown in Figure 3 for 204 emissions to air. However, for emissions to seawater the ranking changes from a distance of 1 km to 205 distances greater than 100 km, as highlighted above. The fate model for emissions to seawater in CGM 206 calculates two concentrations: one relevant to predict accumulation of radionuclides in fish, and 207 another to estimate build-up on coastal shorelines. For distances of the receptor that are relatively 208 low compared to the distance between source of emissions and shoreline (e.g. 1 km), the radioactive 209 plume does not reach the shoreline. Changes in the ranking occurs for those nuclides for which the 210 effects of external exposure are more significant than the effects of ingesting fish. This is exemplified 211 by Eu 155, Co60 and U234, for which the characterisation factors increase or remain approximately 212 constant when the distance increases from 1 km to 100 km.

The effect of dilution on the characterisation factors obtained from CGM highlights the most important limitation of the methodology: it assumes that the population affected by emissions is

215 concentrated at a specific location. For a uniformly distributed population, such as that considered in 216 the Human Health Damages approach (Frischknecht et al., 2000) or in the UNSCEAR methodology 217 (UNSCEAR, 2017), spreading of the plume of pollutant does not affect total collective dose because 218 the reduction in concentration is offset by the increase in the total number of receptors exposed 219 (Dreicer et al., 1995). For a Linear No-threshold dose-response function, the impact is also unaffected: 220 as the plume disperses, the number of receptors affected increases but probability or severity of 221 damage for any individual is proportionately decreased (Spadaro and Rabl, 1999). Therefore, as is 222 routinely assumed in LCIA, the characterisation factors should not be dependent on distance from the 223 point of release. By contrast, it is implicit in UCrad that there is no effect of dilution because a 224 compartment-type fate model already assumes uniform dispersion of radionuclides in each 225 environmental compartment and thus leads to characterisation factors that are not dependent on the 226 location of the target group.

Because the concentrations predicted by CGM depend strongly on half-life and dilution, the distance between source and receptor that gives average agreement (MLD=0) between UCrad and CGM also depends on these parameters. When radioactive decay is negligible (i.e. when half-life is greater than transit time), the methodologies converge. For emissions to air and freshwater (Table 1), this occurs for distance between 100 km and 1 000 km. Because the transit times in seawater are greater than in air and freshwater (Table 2), this convergence is seen for lower distances, between 1 and 100 km. The models do not converge for emissions to freshwater because dilution is not considered (see above).

The comparison between UCrad and CGM for emissions to seawater also highlights three notable outliers - krypton- 85, radon-222, and xenon-133 - whose log deviations different considerably from those of radionuclides with similar half-life (see Figure 2 and Figure 3). Radioactive noble gases differ from other radionuclides because they impart negligible doses through internal pathways like ingestion and inhalation; their main impact pathways are external, for instance through exposure to a radioactive plume (IAEA, 2001; NCRP, 1995). The fate models in UCrad enable radionuclides to disperse to all environmental media irrespective of the medium to which they are released, whereas in CGM radionuclides primarily remain in the environmental medium to which they are released. By ignoring the transfer of noble gases from seawater to the atmosphere, CGM omits the most significant exposure pathway. This is why factors for CGM are considerably lower than those for UCrad for emissions to seawater.

245 The analysis described in Section 3 did not include factors for emissions from nuclear waste disposed 246 in a GDF although characterisation factors are available for both methodologies. This is because the 247 CGM methodology is not set up to produce distance-dependent factors for these emissions (Paulillo 248 et al., 2019a). Figure 1 in the Supporting Information compares the UCrad factors with the only set of 249 factors for CGM. MLD for different types of nuclear waste range from ~6 to ~8. They are higher than 250 those for emissions to air, freshwater and seawater at 1 km (Table 1) despite the distance between 251 source and receptor being set at ~3 km for these emissions. This is because the PCSA model, on which 252 CGM relies for calculating factors for emissions from GDF, assumes a worst-case scenario in which, for 253 instance, the critical group's potable water is obtained directly from contaminated groundwater.

4.2 A practical rule for the application of UCrad and CGM

255 To conclude, we discuss practical applications of these methodologies. We distinguish two distinct 256 types of problem in environmental assessment: selection of technology and selection of site and scale 257 for a plant using a particular technology. LCA is the appropriate tool for comparing technologies, whilst 258 HERA is appropriate for decisions over siting and scale of a plant. Although the two types of problem require different approaches, the methodologies should be compatible and methodologically 259 260 consistent so that their results can be compared to reveal, for example, the extent to which a specific 261 group receives impacts different from those on the general population, and whether reduction of 262 global impacts is achieved at the expense of damage to a specific group.

It is common practice in LCA to distinguish between the foreground system, comprising specific
 processes for which primary, site-specific data are available and whose selection or mode of operation

265 is affected by decisions based on the study, and the background system which exchanges materials 266 and energy with the foreground (Clift et al., 2000). UCrad represents the most appropriate 267 methodology for assessing the impacts of ionising radiations in LCA because it is consistent with the 268 general approach used in LCIA; i.e. assessment of global impacts of the whole system, foreground plus 269 background, on a uniformly disperse population. CGM, on the other hand, can be used to describe the 270 geographically specific impacts of the foreground processes. UCrad and CGM describe different 271 impact pathways – generalised and localised – but the comparison in section 4.1 shows that 272 differences between their predictions are explicable and represent real differences between the two 273 types of assessment. Given that the approaches are methodologically consistent and compatible even 274 when their specific predictions are different, they can be used alongside each other to assess the 275 impacts of ionising emissions for the different purposes and treated as equivalent.

Thus, combined application of UCrad and CGM can help to resolve one of the concerns in LCA: how to reconcile site-specific with generalised assessment (see Clift et al., 2000). However, it has notable implications for definition of the functional unit, which as noted in Section 1 is one of the fundamental differences between LCA and HERA. Comparison of technologies can be based on any functional unit, be it arbitrarily chosen by the LCA practitioner or representative of actual operational data. However, recommendations on the location and the scale are meaningful only if the functional unit is related to the actual or potential scale of operations of the plant.

To take a concrete example: imagine an LCA study that aims to compare two uranium mines, one underground and one superficial (known as open-pit) in different locations and at different distances from inhabited centres. An arbitrary functional unit, as used in conventional LCA, might be 1 kg of uranium mined. However, 7000 tonnes might be a realistic functional unit representing the potential mine¹. UCrad, used to assess which mine is environmentally preferable by considering emissions

¹ This figure actually represents the approximate annual output of the Cigar Lake underground mine in Canada, the largest uranium mine by volume of production in 2017 (WNA, 2019).

arising both from the foreground mine site and the background processes making up the entire life cycle, can be based on either arbitrary or actual functional units. By contrast, CGM must be based on the actual scale of the mine to estimate its radiological impacts on a chosen critical group, e.g. the closest centre of human population.

The combination of CGM and UCrad represents a first step towards a possible integration of HERA and
LCA, an idea that remains to be explored further.

294 **5** Conclusions

295 Based on the general framework and two specific methodologies for assessing radiological impacts in 296 LCA proposed by Paulillo et al. (2019) - UCrad and the Critical Group Methodology (CGM) - this article 297 presents a detailed quantitative comparison between the characterisation factors obtained from 298 these methodologies. The characterisation factors for CGM are strongly affected by radioactive decay 299 and dilution of the radioisotopes in the pollutant plume. By contrast, UCrad does not account for 300 dilution and is less affected by radioactive decay because it is based on compartment-type models at 301 steady-state conditions to predict the fate of nuclides. Therefore, characterisation factors obtained 302 from CGM are much lower than those from UCrad when a significant proportion of the radionuclide 303 decays during transit from emission source to receptor in a specific environmental medium; i.e. when 304 the half-life of radioactive decay is lower than the transit time. When the half-life is long, radioactive 305 decay is negligible and the factors for CGM are primarily affected by dispersion of the plume: dilution 306 becomes more significant at greater distances, resulting in characterisation factors that decrease with 307 distance for CGM. Factors for emissions to freshwater remain constant at high half-life because 308 transport is assumed to occur by riverine flow and dilution is not considered.

Finally, a practical rule for the applications of these methodologies has been proposed. UCrad represents the appropriate methodology for assessing the impacts of ionising radiations in LCA because it is consistent with the general approach of Life Cycle Impact Assessment (LCIA) and the

- 312 methodologies for assessing impacts of toxic pollutants. However, the CGM methodology can be
- 313 applied alongside UCrad to enable recommendations to be made on the site and scale of a plant using
- a particular technology, for example to comply with regulatory limits.

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