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Radiological impact assessment approaches for life cycle assessment: a review and possible ways forward

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Abstract

Many industrial processes routinely release radionuclides into the environment. Such emissions may be recognised in the inventory phase of LCA, but are rarely carried forward to the Life Cycle Impact Assessment (LCIA) phase because a standard approach for assessing their impact is still lacking. The aim of this article is to collect and critically analyse radiological impact assessment methodologies to establish a basis for developing a standard approach. Seven methodologies have been reviewed. Amongst these, the Human Health Damages approach represents the only methodology to date included in LCA impact methodologies. Furthermore, five of the reviewed methodologies are concerned with impacts on human beings, whilst the remaining two address effects on the environment. The article concludes that even though a number of methodologies are currently available, none is suitable as the basis for a standard procedure in LCIA. Two main features have been identified as crucial: the ability to treat all types of waste forms by which radionuclides can be released and the use of a fate analysis which returns average (rather than worst case) estimates of impacts. In light of the findings of this review, a novel framework for radiological impact assessment on humans has been devised; its development is being pursued by the authors.

Keywords: Radiological Impact Assessment, Life Cycle Impact Assessment, Ionising Radiations, Radionuclides, Nuclear Waste, Risk Assessment.

1 Introduction

Many industrial processes during routine operations release radionuclides into the environment in the form of air- or water-borne materials or solid wastes (UNSCEAR 2008). The nuclear (Dreicer et al. 1995), coal (Qifan et al. 2015, Ruirui et al. 2015), oil and gas (Cowie et al. 2008), fertiliser (Othman and Al-Masri 2007, Aoun et al. 2010) and building industries (Aliyev 2005, Gehrcke et al. 2011) are among the major contributors (Kathren 1998, Ryan et al. 2005). To date such emissions have been recognised in the inventory phase of Life Cycle Assessment (LCA), usually aggregated in terms of a single inventory term measured in Becquerels (which is the SI unit of radioactivity). However, the inventory data have rarely been carried forward to the Life Cycle Impact Assessment (LCIA) phase, mainly due to the lack of a standardised framework for classification and characterisation (Cucurachi et al. 2014). Nonetheless, in some cases, the impact of radionuclides on human beings and ecosystems may be critical in the comparison between alternative technologies for providing a specific product or service; comparisons between energy sources are an obvious example. When comparing different electricity generation options, nuclear energy outperforms fossil fuels in almost all the conventional non-radiological impact categories and therefore emerges as one of the cleanest sources of energy, comparable with renewable energies (Gagnon et al. 2002, World Energy Council 2004, Poinssot et al. 2014). Where radiological impacts have been considered, notably in LCA studies in the nuclear sector, they have almost always been considered separately from non-radiological impacts. This has led to a disjointed approach to environmental management in which control and reduction of one impact is undertaken without considering the other impacts (Brown 1992, Tran et al. 2000, Chen and McKone 2001, Shiels et al. 2002). One result has been that minor reductions in radiological impacts have been implemented even though they lead to major increases in non-radiological impacts, usually unacknowledged, at other stages of an activity’s life cycle (Shiels 2002).

This inevitably raises the question: how will comparisons and approaches to environmental management be affected if the radiological impacts are included in the assessment on an appropriate and consistent basis? By “appropriate”, we mean an approach able to assess the environmental impacts of every type of radioactive waste arising anywhere in the life cycle. “Consistent” denotes a holistic approach able to consider and evaluate both radiological and non-radiological impacts on a consistent basis. The optimum environmental strategy should be defined as that delivering the minimum overall environmental impact resulting from both radiological and non-radiological impacts across the whole life cycle. The aim of this work is to review and critically analyse radiological impact assessment methodologies to establish a basis for developing an appropriate and consistent framework for assessing radiological impacts in LCA. We focus specifically on radiological impacts linked to releases of radioactive nuclides; other sources of ionising radiations, such as high energy electromagnetic waves or direct radiations from buildings without modern level of shielding, have minor impacts and are rarely considered in radiological impact assessments. The methodologies included in this review either have been proposed and developed exclusively for LCIA, or are currently part of standard assessment procedures in other fields and for other purposes (e.g. risk assessment for industry internal reviews) but may be adaptable for incorporation in LCIA.

The article is organised as follows: In Section 2 we provide an overview of the general approach to radiological impact assessment; Section 3 reviews current approaches to radiological impact assessment within LCA; in Section 4 we comment upon these methodologies, discuss their suitability for inclusion in LCIA and suggest possible approaches for further development. Section 5 provides a summary of the most significant conclusions of this review leading to future work to develop a sound approach to include radiological impacts in LCIA. Finally, a glossary of the most recurring acronyms is reported in Section 6.

2 Radiological Impact Assessment

The Impact Assessment phase of LCA (LCIA) aims to analyse and assess the environmental impacts of human interventions identified in the inventory (Saur 1997, Udo de Haes et al. 1999). For this reason, LCIA is probably one of the most debated stages in the LCA methodology. LCIA conventionally includes non-radiological impact assessment, i.e. the non-radiological toxic effects of emissions on humans and on non-human biota in the environment. However, the impacts from releases with radiological impacts are usually disregarded or, at best, considered as an optional category to be included at the discretion of the LCA practitioner (Cucurachi et al. 2014). For instance, in the method developed at CML (Guinée et al. 2002) they are defined as study-specific impacts, i.e. impacts that may merit inclusion depending on the goal and scope of the LCA study. The approaches currently used in LCIA methods are reported in Table 2 and discussed in Section 4.

2.1 Radionuclide Properties

A radioactive nuclide or radionuclide is an unstable atom in an excited state, i.e. its energy level is higher than the ground state (the state of lowest energy). An atom cannot remain in an excited state indefinitely: it decays to another state at lower energy, eventually returning to the ground state. During this process the atom releases the excess energy in the form of gamma rays, subatomic particles such as alpha or beta particles or conversion electrons; together, these are commonly termed "ionizing radiation" (Lamarsh and Baratta 1955). Radionuclides share a number of chemical and physical characteristics with heavy metals and organic chemical species; thus they pose similar difficulties with regard to the impact assessment stage. The most apparent characteristic is that many radionuclide species are extremely persistent: they typically have long half-lives so that they can survive in a specific environmental medium sufficiently long to have impacts over extended periods of time and to be transported over long distances. Secondly, they have the propensity to bio-accumulate, which refers to the ability to concentrate in living tissue. Thirdly, radionuclides are both toxic and radioactive. This means that, not only do they contribute to internal exposure through ingestion or inhalation (as do heavy metals and organic chemicals) but they can also cause external impacts from radiation (Shiels 2002).

2.2 Human Health Impacts

Radioactive nuclides cause several detrimental effects to human health. The conventional approach to human radiological impact assessment covers some or all of the following three steps: determination of the radionuclide environmental concentration as a result of a release; estimation of the exposure of human beings to ionising radiations; and, eventually, assessment of the dose that individuals will receive due to this exposure (Till and Meyer 1983, NCRP 1995, IAEA 2011).

The environmental concentration of radionuclides within various environmental media is obtained by modelling the transport and dispersion of radionuclides from the release source, using generic or site-specific environmental data (Scott 2003). On a general basis a number of approaches are available to estimate the environmental concentration of radionuclides. Numerical calculations, based on Lagrangian "puff" (e.g. Apsimon et al. 1985) or Eulerian grid (e.g. Lange 1978, MacCracken et al. 1978) models, transform the basic equations providing a detailed representation of the physical processes of dispersion into finite difference or finite element forms. However, the calculations are very demanding of computer resources and so are usually adopted only as a last resort when all other screening models show unacceptable results, e.g. above the legally permitted limits. Analytical models solve the basic radionuclide transport equations by using simplifying assumptions. The Gaussian plume model is one such model widely adopted for dispersion of pollutants into the atmosphere (Sutton 1932, Pasquill 1961, Gifford 1976); the basic assumption is that the concentration of a specific pollutant downstream of a point source has a normal distribution which widens out with increasing distance from the source. Finally, compartment or box type models (Mackay 2001) treat the environment as divided into spatial domains of different scales, each composed of several compartments. Each compartment represents an environmental medium (e.g. air, sea water, soil etc.) modelled as homogeneously mixed and able to exchange material with other connected compartments.

From the environmental concentration, the human exposure to radioactive materials may be estimated. The aim is to quantify the amount of radioactive material with which human beings come in contact. As noted above, humans can be exposed to radioactive materials via two main routes: external and internal irradiation. The external irradiation may be directly estimated from the concentration of radionuclides in air, water and soil whilst the internal route comprises two main pathways: inhalation and ingestion. The air concentration of radionuclides is the sole source in the inhalation pathway. The ingestion of radioactive materials, however, is estimated by coupling the concentration of radionuclides in each food category (e.g. vegetables, meat and dairy produce) with their specific consumption patterns (IAEA 2001). The concentration in food is obtained through specific models that establish how radionuclides move from each environmental medium to each food category (IAEA 2009). Consumption patterns, on the other hand, represent the eating behaviour of individuals: they define how much of each food category is consumed; so called “usage factors” express this quantity. They can be country- or region-specific, or global averages (see for instance FAO, 2013).

Finally, the last step involves determining the amount of energy received and its potential interactions with human beings due to exposure to radioactive materials. This is quantified and expressed by means of dosimetric quantities. The fundamental dosimetric quantity in radiation protection is the absorbed dose (D), defined as the mean energy (per unit mass) imparted to matter by ionizing radiations and measured in Grays (Gy) (ICRP 2007). The International Committee on Radiological Protection (ICRP), however, has developed other dosimetric indicators (ICRP 2007, Seltzer et al. 2011). As different types of radiation can cause different effects in biological tissue and as different organs may be more or less susceptible to irradiation, the absorbed dose is weighted twice to take in consideration those aspects: the resulting value is termed the Effective Dose and is measured in Sieverts (Sv) (ICRP 1977, 1991, 2007). Furthermore, in order to consider the prolonged effect of ingestion or inhalation, another quantity has been developed: the Committed Effective Dose, also measured in Sieverts, is the time integral of the effective dose rate over a specified period of time (ICRP 1977, 1991, 2007). The dose depends not only on the intensity of ionising radiations, but also on the physical characteristics of the receivers (e.g. weight, age, etc..,). For this reason, the procedure of dose calculation has been based on the concept of the “Reference man” (ICRP 1975, 2002): its purpose is to define a “standard” individual (“of reference”) with average characteristics, for which doses can be calculated. In this way the procedure is significantly simplified and the final dose depends solely on the amount of ionizing radiation received. Ultimately, the “Reference man” leads to a database of conversion factors (obtained for the standard individual) which allows estimation of the effective/committed dose as a result of the exposure to ionising radiations. Dose limits are defined for the “Reference Man”: the ICRP annual limit on the effective dose for the Reference Man has been set as 1 milli-Sievert (mSv) (ICRP 2007).

Exposure to ionising radiation can lead to two distinct types of effects: deterministic and stochastic (ICRP 1990, 2007). Deterministic effects result from the killing of cells which, if the dose is large enough, causes sufficient cell loss to impair the function of the tissue. They do not occur below a threshold (typically around 1 Sv), which depends on individuals’ radio-sensitivity; whilst above the threshold, the severity of the harm increases with the dose (lower part of Figure 1) (ICRP 1990, 2007). Because individuals show different sensitivity (curves a, b and c) to ionising radiations, the probability of the harm in a population follows a sigmoid function (upper part of Figure 1) that is zero when the dose is below the threshold for all individuals in the population, and is 100% when the dose exceed the threshold for the entire population (ICRP 1990, 2007). Stochastic effects, on the other hand, result when an irradiated cell is modified rather than killed; the modified cells may develop into a cancer. In this case, the probability of cancer but not its severity depends on the dose: the probability of cancer is considered to be roughly proportional to the dose for doses below the threshold for deterministic effects, and to follow a quadratic trend for doses above this threshold. It is also believed that there is no minimum threshold for stochastic effects (curve A in Figure 2) (ICRP 1990, 2007). This model, and in particular the proportional section at low doses, is referred to as Linear Non-Threshold (LNT); and has been criticized as not totally supported by experimental evidence (Allison 2015, Siegel et al. 2015). Furthermore, as most epidemiological information available for stochastic effects refers to high doses in the quadratic section, the dose-response relationship between received dose and probability of cancer at low doses (linear section) is estimated from data at high doses by means of the so-called "Dose and Dose-Rate Effectiveness Factor" (DDREF) (ICRP 1990, 1991). The DDREF is defined by the ICRP as the ratio of the slope of the linear fit to high dose data to the slope of linear fit at low dose data (i.e. α_L (Curve B) to α_1 (Curve D) in Figure 2). The ICRP has found that the DDREF ranges between 2 and 10 and recommends using a value of 2 as the best estimate for extrapolation to low doses (ICRP 1990, 2007), meaning that the increased probability of cancer per Sv observed at high doses is divided by 2 to estimate the response at low doses. Finally, in the approaches reviewed here, and more generally in all the approaches that deal with routine releases of radioactive materials, only stochastic effects and therefore low doses are taken into account; deterministic effects come into play only in the case of nuclear accidents.

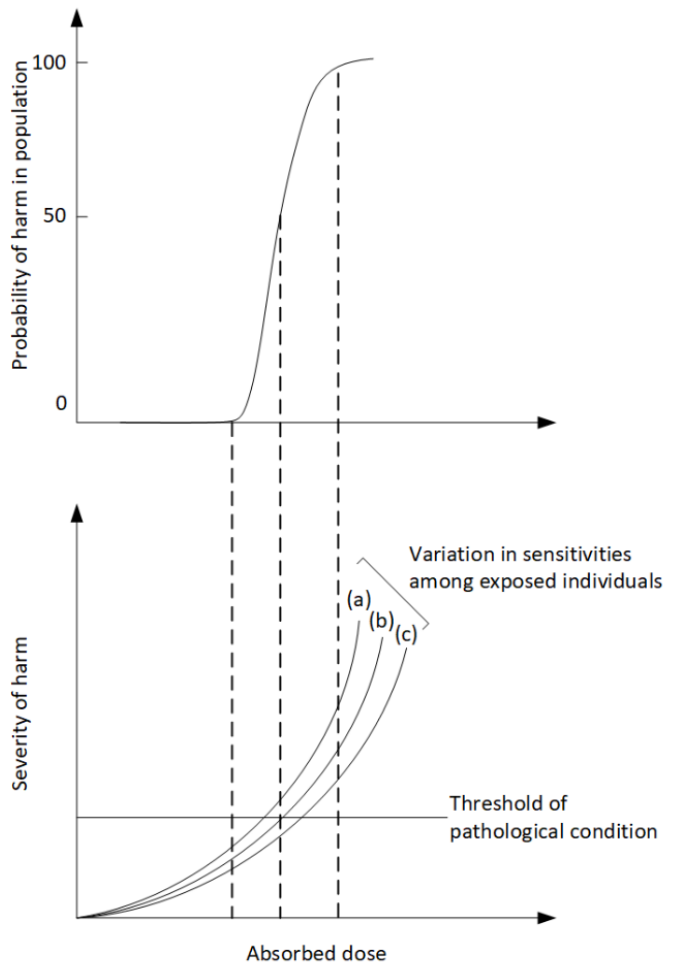

Figure 1 – Typical dose-effect relationships for deterministic effects expressed in a population (adapted from ICRP 1984).

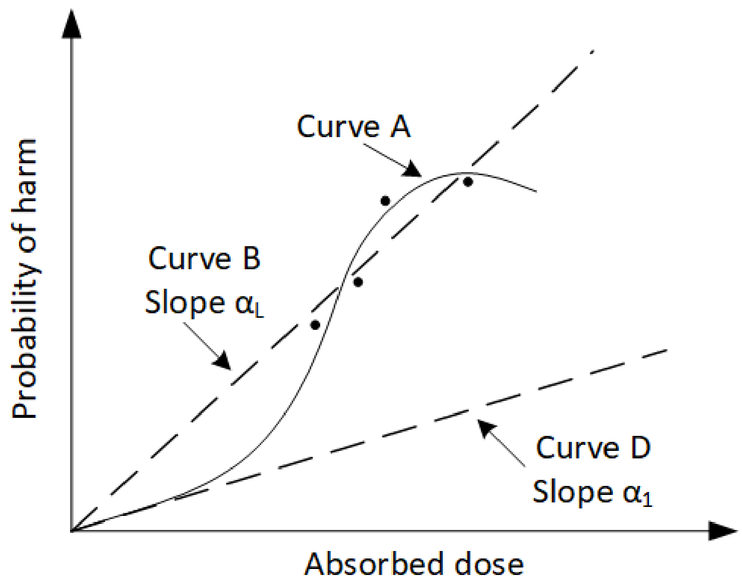

Figure 2 – Schematic curves of incidence vs adsorbed dose (adapted from NCRP, 1980).

2.3 Ecological Impacts

At present there are no internationally agreed criteria or policies that explicitly address protection of the environment from ionising radiation. So far, radiation protection has been focussed upon human impacts; on the assumption that the system in place for protection of human beings must afford an acceptable level of protection to non-human organisms, most environmental monitoring of ecosystems concentrates on only those species or materials which are part of the critical pathways to humans. This line of thought has been set out in the ICRP recommendations of 1977 (ICRP 1977), reiterated in the recommendations of 1990 (ICRP 1990) and supported by the International Atomic Energy Agency (IAEA 2006). However, interest in protection of the environment has greatly increased in recent years: society's concern for environmental risk has put pressure on policy makers and regulators to define protection strategies that specifically and explicitly include the environment. The assumption underlying the ICRP approach was firstly challenged by Thompson (Thompson 1988): he reasoned that this hypothesis is valid only when humans and other biota inhabit the same part of the environment; in different circumstances non-human organisms could be exposed to higher concentrations and hence there could be an impact on certain species without an associated impact on humans.

The need for a broader framework for environmental protection was accepted by the ICRP in 2000 when it set up a Task Group to advise on the development of a policy and to suggest an effective approach. The Task Group duly proposed a new framework for the protection of the environment from ionising radiation (ICRP 2003). The ICRP subsequently established "Committee 5: Protection of the Environment", and in 2007 incorporated environmental protection as one of the integral elements of the radiation protection system (ICRP 2007). The approach is similar to that developed for the assessment of human impact, based on a set of Reference Animals and Plants defined as hypothetical entities with assumed biological characteristics that are used to relate exposure to dose and dose to effects. Committee 5 published in 2008 a first set of Reference Animals and Plants along with their relevant databases (ICRP 2008), followed by two more reports covering approaches to model the transfer of radionuclides to non-human biota (ICRP 2009) and to extend the application of the radiological protection system to different exposure conditions, e.g. unplanned events) (ICRP 2014). The proposed framework, however, is not intended to set regulatory standards; rather, it is conceived as a practical tool to provide high-level advice and guidance. However, it does not preclude the derivation of standards; on the contrary, it provides a basis for such standards (Clarke and Holm 2008).

3 Review of Published Methodologies

Since the establishment of a standard framework for LCA, a number of impact categories addressing different kinds of impacts of human activities have been developed. Two organisations in particular have been involved in work on LCIA: ISO and SETAC. Whilst the former mainly deals with procedures rather than specific methodologies (e.g. ISO 2000), the latter, especially through the work of the SETAC –Europe and –US Working Group on Impact Assessment, focussed on establishing a “best available practical method” for each impact category (e.g. Potting et al. 2001). The Handbook on Life Cycle Assessment (Guinée et al. 2002), on the other hand, sets out all relevant methodologies and developments in LCIA. To select an appropriate approach, the most relevant criteria are:

- Impact methodologies should be based on scientifically and technically valid models;
- Impact indicators should be linear in relation to the magnitude of emissions;
- Impact methodologies should include modelling of fate, exposure/intake and effects, as relevant;
- Impact methodologies should be time and location independent.

With regards to the last criterion, in recent years a movement towards spatially differentiated models to account for differences in both populations’ habits and environmental parameters has started (e.g. see Wegener Sleeswijk and Heijungs 2010); nonetheless, this approach has yet to achieve widespread acceptance. Location-dependent models enable impacts but not inventory data to be aggregated across the life cycle, because different characterization factors apply to emissions occurring in different locations.

In addition, it must be recalled that LCA was born as a tool for assessing and comparing different product system under ‘normal conditions’; i.e. LCA deals with routine planned emissions, not with stochastic events or safety issues. This represents a further problem in applying LCA to the nuclear field: whether to include stochastic events, primarily possible future disturbance of nuclear waste repositories.

Finally, LCA studies are intended to produce estimates of average impacts to groups of people inhabiting specific regions, countries or continents. Hence impacts to subgroups of individuals particularly sensitive to specific emissions are not part of LCA studies; rather, they are the focus of risk assessment studies.

Table 1 lists the methodologies for radiological impact assessment considered here, with their main features and references. To highlight the key features of and main differences between the methodologies, Figure 3 and 4 summarise the emission sources included and the source-pathway-effect model behind each, for human and ecological impacts respectively.

Table 1 – Radiological impact assessment methodologies reviewed

Methodology	Emission type	Scope	Applicability	Impacts estimation	Indicator	Metric	Reference
Critical Volume	Direct routine discharges	Humans	Site-independent	Worst case	Mid-point	$kg_{body\ weight}$	(Heijungs et al. 1992b)
Site-specific	Direct routine discharges	Humans	Site-specific	Worst case	Mid-point	Sv or ManSv	(Simmonds et al. 1995)
Damage-based	Direct routine discharges	Humans	Site-independent (preferably applicable in Europe)	Average	End-point	DALY	(Frischknecht et al. 2000)
Human Irradiation	Direct routine discharges and emissions from solid waste	Humans	Site-dependent	Worst case	End-point	Risk	(Solberg-Johansen 1998)
NDA Value Framework	Direct routine discharges and emissions from solid waste	Humans	Site-dependent	Worst case	End-point	Sterling	(Wareing 2009)
Environmental Irradiation	Direct routine discharges	Ecosystems	Site-dependent	Worst case	Mid-point	-	(Solberg-Johansen 1998)
SLERA	Direct routine discharges to fresh water	Ecosystems	Site-independent	Average	Mid-point	CTUe	(Garnier-Laplace et al. 2009)

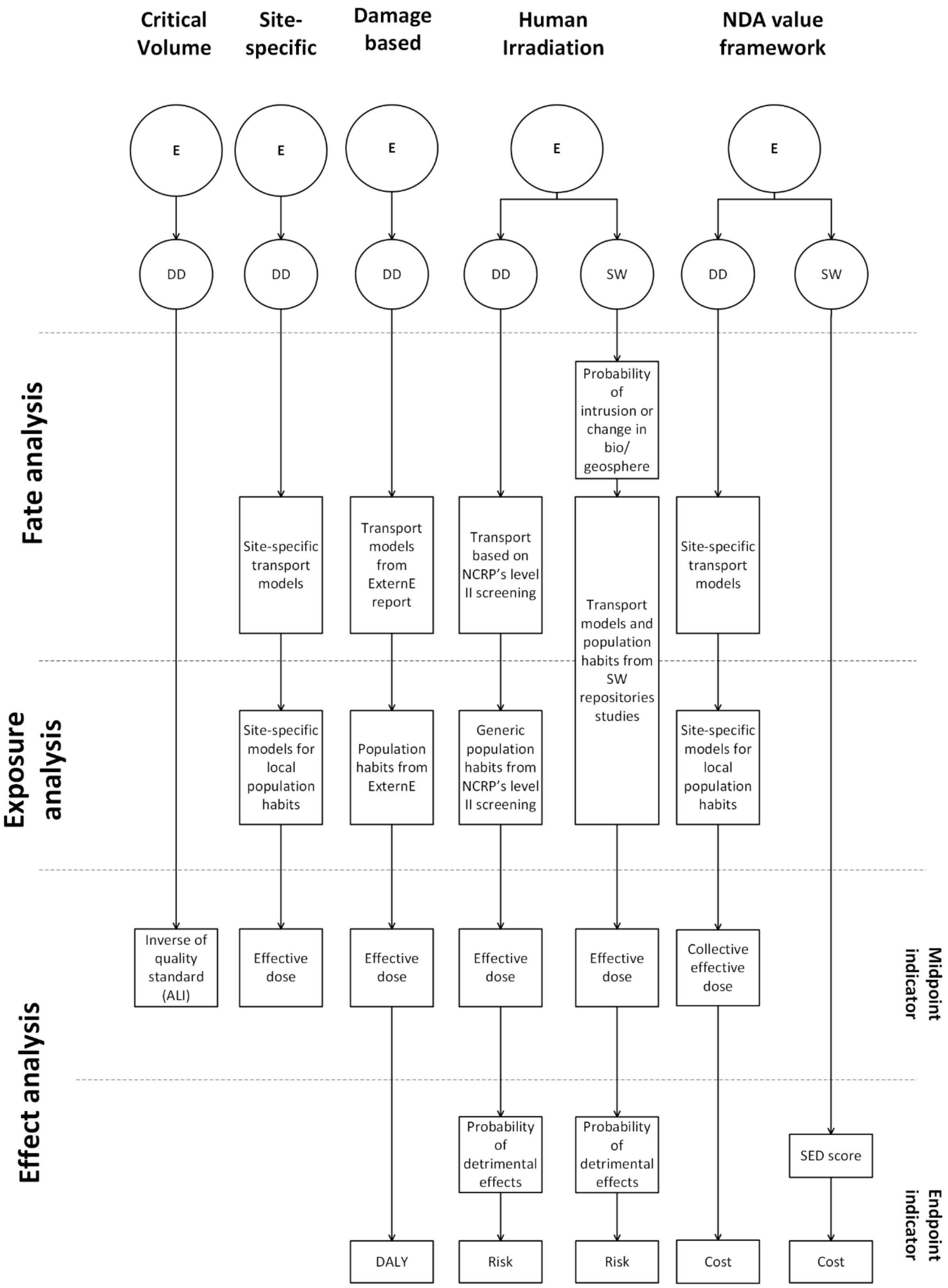

Figure 3 – Outline of methodologies for radiological impact assessment on humans. E= Emission; DD= Direct Discharges; SW= Solid Waste.

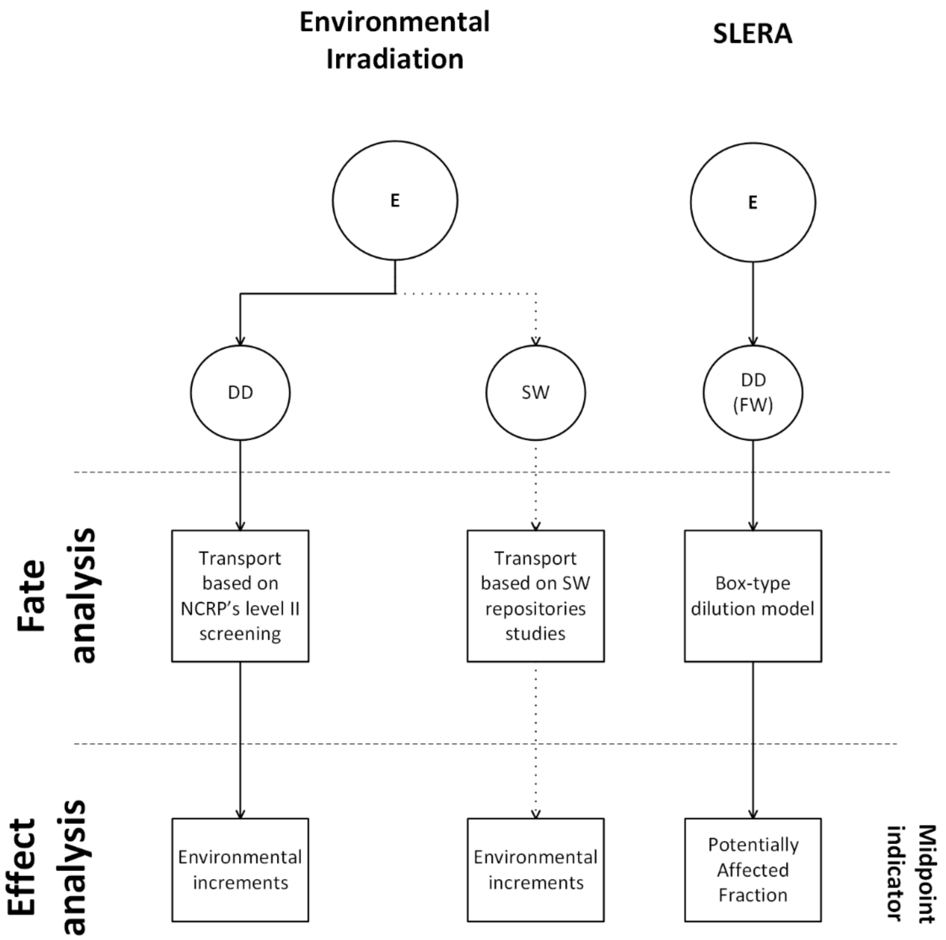

Figure 4 – Outline of methodologies for radiological impact assessment on ecosystems. E= Emission; DD= Direct Discharges; SW= Solid Waste; FW= Freshwater.

3.1 Human health impacts

3.1.1 Critical Volume approach

Heijungs and colleagues were the first to discuss how radionuclide release could be included in LCIA (Heijungs et al. 1992a, 1992b) and to propose a possible approach (Heijungs 1994). At that time, few models were available for determining the fate and exposure of radionuclides and the relation between absorbed and equivalent dose. The so-called "Critical Volume" approach represented the only method available to assess the potential effects of radionuclides on human beings from emission values, without fate or exposure analysis (see Figure 3). The authors, however, recognised that the lack of a fate analysis was a major flaw and proposed it only as an interim step towards development of a more comprehensive methodology. Without considering transport and dispersion processes, the methodology assumed that the receptor was in direct contact with the waste streams emitted and hence exposed to the maximum possible radioactivity, meaning that the methodology only produced the highest possible estimates of impacts. The lack of a fate analysis also implies that the methodology is independent of the source of emission. The total contribution to ionising radiation impact is obtained by summing the product of the activity of each radionuclide at the point of release and the radionuclide specific characterisation factor. This also implies that the impacts of individual radionuclides are additive.

The "Critical Volume" approach calculates the characterisation factor for each radionuclide as the inverse of the maximum permissible concentration or quality standard in the receiving medium. As radiation standards had only been defined for workplace exposure, the Annual

Limit of Intake (ALI), intended by the ICRP (ICRP 1979, 1991) to define the basic limit for occupational exposure to a given radionuclide, was chosen by Heijungs and coworkers as the quality standard. The ALI is defined as the activity (in Becquerels) which taken in on its own would commit a person, represented by the "Reference Man", to the annual limit on the effective dose, which was set at 20 mSv. The ALI considers exposures by ingestion and inhalation and also impacts of daughter products, and represents the largest annual intake that would satisfy limits for both stochastic and deterministic effects. In LCIA it is common to distinguish between mid-point and end-point indicators. The latter are defined at the level of the areas of protection (Human health, Environmental load and Use of resources), whilst the former are located along the cause-effect chain prior to the end point (Bare et al. 2000, Baumann and Tillman 2004, Clift 2013). In view of this, it can be said that the characterisation factors produced by the Critical Volume adopt the mid-point perspective.

3.1.2 Site-Specific Approach

Site-specific models can be used to predict the actual impact of radionuclide releases from a definite site by estimating the resulting individual or collective doses to humans within specific groups. Figure 3 shows the key steps of the site-specific approach: fate, exposure and effect analysis. The exposure and effect analysis models are the same in all methodologies; they were first developed by the National Council on Radiation Protection and Measurements (NCRP 1995) and further improved by the IAEA (2001) and now constitute a standard framework. A number of site-specific methodologies have been developed, differing in their basic assumptions and mathematical models for fate analysis.

In the UK, Sellafield Ltd. and the Environment Agency currently use the site-specific model CREAM (Consequences of Releases to the Environment: Assessment Methodology) (Simmonds et al. 1995), for assessing the radiological consequences to the "Critical Group" of routine releases of radionuclides into the atmosphere and aquatic environment. The "Critical Group" is defined as the member(s) of the public predicted to receive the highest dose due to their lifestyle, location and habits (ICRP 1990, NRPB 1993). The dose to members of the critical group is assessed as the mean of the sums of effective doses from external irradiation and their committed effective doses arising from all relevant pathways. CREAM was developed by the National Radiological Protection Board (NRPB) under contract to the Commission of the European Communities (CEC) (Simmonds et al. 1995) and translated into the computer code PC-CREAM (Mayall et al. 1997) which has since been continuously updated (Smith and Simmonds 2009). CREAM requires site-specific parameters such as meteorological conditions and individuals' habits, but provides accurate and reliable estimates of the resulting doses.

Atmospheric dispersion of radionuclides is represented in CREAM by a Gaussian plume dispersion model (Pasquill 1961, Gifford 1976). The model may be used for releases of both short and long duration, the difference being the variability of the wind rose. Removal processes, such as depletion of radionuclides by wet and dry deposition and radioactive decay, as well as reflection from the ground and from the top of the mixing layer are taken into account. A direct consequence of the concept of critical group and the use of a Gaussian plume model is that the resulting environmental concentrations of radionuclides are a function of space, i.e. distance from the release point. Once the receptor location is chosen, the transfer of radionuclides through the terrestrial environment to that location is modelled with the aim of assessing the irradiation dose via inhalation of re-suspended activity, ingestion of contaminated foodstuffs and external irradiation due to surface deposition.

The aquatic compartment is represented in CREAM by four compartments (termed "sectors" in CREAM): freshwater bodies (rivers), estuaries, local marine zones and regional marine zones. A discharge into a river may result in movement of radionuclides through all four compartments, whilst for a discharge into the sea only the local and regional marine zones may have to be considered. Although models have been developed for the estuary compartment they have not been yet included in the PC-CREAM 08 methodology (Smith and Simmonds 2009). The river section is modelled by means of two different theoretical approaches: simple dilution and semi-empirical (or dynamic) models. The latter retain some of the spatial and temporal resolution of detailed hydraulic models but use empirically-derived coefficients to describe the distribution between suspension and sediment (Schaeffer 1975). The marine section contains two multi-compartment models representing respectively northern European (Simmonds et al. 2002) and Mediterranean seas (Cigna et al. 1994). Dispersion on a local scale, up to a few kilometres from the discharge point, is modelled by a single well-mixed water compartment. The local marine zones are connected to regional zones which represent the dispersion of radionuclides in European coastal waters, in the Atlantic Ocean and other world oceans. Each of the water compartments has an associated suspended compartment, and water compartments in contact with the sea bed have underlying seabed sediment compartments. Different exposure pathways are considered for the river and marine sections. The main pathways for the former are ingestion of drinking water and fish, external exposure and application of river sediments as soil conditioner; the exposure due to the marine section is modelled as arising from sea-spray, ingestion of seafood and inhalation, ingestion and external exposure from beach material.

Finally, global circulation models are included for those radionuclides whose half-life and behaviour in the environment make them highly persistent and therefore globally dispersed. Four particular radionuclides are considered as globally dispersed: Krypton-85, Tritium, Iodine 129 and Carbon-14; they act as long term sources of irradiation impacting both regional and world populations.

Although being mainly used for assessment of critical groups, site-specific models can also be used to estimate doses to a wider group of people or extended to include regional, national and worldwide impacts. However, the wider the range of the study, the more data (site-specific) and calculation time will be required. The approach can provide an accurate approach to dose estimation; however, it is not readily applicable to LCIA, which favours approaches that are not dependent on location and geographically specific parameters. Furthermore, as noted in Section 3, site-specific approaches do not allow aggregation of inventory data, rather only of impacts.

3.1.3 Human Health Damages approach

Frischknecht et al. (2000) suggested a different approach to assess the human health effects of routine releases of radioactive substances to the environment, specifically devised to be integrated into LCA. As shown in Figure 3, it included fate, exposure and effect analysis using both midpoint (dose) and endpoint (DALY) indicators. The fate and exposure analysis are generalised from site-specific modelling of the French nuclear fuel cycle carried out by Dreicer et al. (1995) within the ExternE ("Externalities of Energy") project, covering routine atmospheric and liquid discharges from all steps of the cycle. The environmental dispersion models are very similar to those in CREAM (see Section 3.1.2). Aerial discharges are modelled by a Gaussian plume model using wind roses developed from past measurement of the meteorological conditions at specific French sites to represent average annual conditions. For discharges to rivers, a simple MacKay model (Mackay 2001) is used, with the watercourse represented as a number of homogeneously mixed compartments taking into account characteristics of the river and human utilisation; whilst for sea discharge, the methodology employs an early version of the European sea model (Charles et al. 1990) used in CREAM. Finally, the same models for globally dispersed radionuclides as in CREAM are used. The pathways considered in the exposure analysis include inhalation, external irradiation and ingestion of both terrestrial and seafood. Exposure factors were derived from the ExternE project complemented with data from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 1982, 1988, 1993). The estimated absorbed dose is converted into a whole body dose (Sv) by means of the ICRP factors (ICRP 1990).

The final step consists of the damage analysis which estimates the health effects to human beings in terms of DALYs (Disability Adjusted Life Years), a metric developed by Murray et al. (Murray and Lopez 1996a) for the World Bank and World Health Organization (WHO). The DALY is a defined as a “measure of overall disease burden expressed as the cumulative number of years lost due to ill-health, disability or early death” and calculated as the sum of the Years of Life Lost (YLL) due to premature death and Years Lost due to Disability (YLD). Both terms are estimated using a number of parameters such as the average severity of disability, the average age of onset, the average duration of disease, the lethality fractions and probability of occurrence of different cancers (ICRP 1990, Murray and Lopez 1996a, 1996b, Ron and Muirhead 1998).

Following the different perspectives identified in Cultural Theory (Thompson et al. 1990), the damage-based methodology considered three scenarios from which two sets of damage factors were derived. Cultural theory describes five ways of life that are viable combinations of cultural biases and social relations. Notably, three cultural perspectives are particularly meaningful for public decision making and LCA studies: Individualism, Hierarchism and Egalitarianism. The choice of cultural perspectives influences the time horizon and the consequent age-weighting applied. In an egalitarian perspective, future is considered at least as important as present: Egalitarians would prefer that society adjust its needs to limit the exposure of future generations. From an individualistic point of view, the present is much more valuable than the future: in the case of unacceptable future exposure, technical solutions to limit them will have to be conceived. In the hierarchic perspective, present and future are equally important.

These three cultural perspectives lead to two scenarios within which impact factors can be developed:

- The egalitarian and hierarchist scenarios are considered equivalent; they assume the longest time horizon (100,000 years) and make no use of age-weighting (or discounting).
- The individualist scenario extends assesses exposure over a period of 100 years and applies age-weighting for the calculation of DALYs.

Therefore, the methodology results in two set of impact factors applicable for discharges to different environmental media: air, fresh water and sea water. As these factors represent potential effects on human health of ionising radiation, they constitute end-point indicators (see definition in Section 3.1.1).

As Frischknecht’s methodology was explicitly developed for LCA purposes, it matches all the requirements that an LCA methodology should fulfil; notably it is intended to be location independent and produces average estimates. This explains why it is the only methodology that has actually been used in LCA studies; this is further discussed in Section 4.

3.1.4 Human Irradiation approach

A different approach, also specifically developed with LCIA in mind, was devised by Solberg-Johansen (1998), who proposed bringing elements of risk assessment within the LCIA framework to include impacts from solid waste disposed in final/long-term repository. This approach differs from all other LCA methodologies in extending the assessment to include stochastic events. The rationale was that disposal options for radioactive solid waste involve containment or isolation from the biosphere: natural radionuclide releases are very slow so accidental events rather than continuous emissions present the greatest long-term risk. Risk assessment can consider quantitatively not only the probability of an event but also the probability of exposure. The approach covers irradiation of both human populations and ecosystems; ecological impacts are discussed in Section 3.2.1. As shown in Figure 3, the Human Irradiation approach distinguishes between routine ‘direct’ discharges and emissions arising from solid waste disposed in final repository, and applies a fate, exposure and effect analysis for each. Notably, the solid waste pathway includes both the natural, gradual degradation by ground water of conditioned waste and its containers disposed in a repository and stochastic events, such as human intrusion or gross distortion of the geosphere. The ICRP (1985) "Extended Dose Limitation System" forms the basis for the proposed impact category methodology where the risk, defined as the probability of serious detrimental health effect occurring in a potentially exposed individual, is taken as indicator and calculated as the product of three terms:

- The probability of radioactive release leading to the individual incurring a dose (P);
- The Effective or Committed dose received (E); and
- The probability (per unit dose) of detrimental effects (F).

This definition of risk limits the approach to stochastic events leading to low dose exposures; at high doses, as noted in Section 2.2, deterministic effects come into play, requiring a different approach for risk calculation. The risk of detrimental health effect is taken to represent the contribution to the Human Irradiation category, from which specific Human Irradiation characterisation factors are calculated. These factors adopt an end-point perspective, like those obtained from the Human Health Damages methodology.

The ICRP identified three major detriments from receiving a radiological dose: fatal and non-fatal cancer, and hereditary effects. For each, the ICRP calculated their probability of occurrence per unit dose (F), which is a constant annual risk factor. The Human Irradiation category only includes fatal and non-fatal cancers, whose risk factors sum to an annual probability of 0.06/Sv (ICRP 1990).

The probability (P) of radioactive release depends on the nature of the discharge. In the case of doses arising from routine release processes, that is direct discharges (to atmosphere or water bodies) and gradual degradation of disposed radioactive waste, this probability is assumed to be unity. Other events, however, are not routine and have to be treated as probabilistic; for those, it is necessary to identify all possible stochastic events and estimate the probability of their occurrence.

The effective dose incurred (E) is obtained by modelling the transport of radionuclides in the environment. Notably, for direct discharges the National Council on Radiation Protection and Measurements (NCRP) Screening Level II models (NCRP 1995) are used to determine the radiological impact. These employ models for fate and exposure analysis similar to the site-specific approaches, but with generic rather than site-specific parameters. The estimated dose, and eventually the risk, resulting from routine releases of radionuclides are calculated for a period of 12 months following 30 years of operation of a nuclear facility. The time frame of 30 years is recommended by the National Radiological Protection Board (NRPB) to represent the period needed by discharged radionuclides to reach equilibrium (e.g. air and water pathways) or a stable concentration of long lived radionuclides in the environment (e.g. soil and sediment pathways) in the absence of a disturbance.

The effective dose calculation for solid waste differs from that for direct discharges in two respects. Firstly, both deterministic and probabilistic releases are considered, and only the pathway which gives rise to the highest individual dose in each case is used for the purpose of developing characterisation factors. Secondly, predictions of anticipated exposure dose are based on four different studies:

- The NRPB's assessment of the radiological impact from the disposal of solid Low Level Waste (LLW) at the Drigg facility, used to determine the impact of LLW in a near surface repository (Smith et al. 1987, 1988);

- The “Disposal of Radionuclides in Ground” figures in the NCRP Screening models, used to appraise the radiological impact arising from the disposal of mill tailings (NCRP 1995);
- The Performance Assessment of Geological Isolation Systems for radioactive waste (PAGIS), used to determine the impact of vitrified High Level Waste (HLW) (Marivoet and Bonne 1988, Storck et al. 1988);
- The Performance Assessment of Confinements for Medium level and α contaminated waste (PACOMA), used to calculate the impact of α-bearing waste and medium level radioactive waste (Storck et al. 1988, Mobbs et al. 1991).

Each of these studies models the migration of radionuclides and predicts dose and risk values arising from a specific inventory of radioactive material disposed in a specific design of repository. This means that results are given only in terms of anticipated dose, with the fate and exposure calculations not reported explicitly, and also that characterisation factors are specific not only to radionuclides but also to waste type and disposal facility. Furthermore, it is assumed that exposure may occur at different times as a result of different evolutionary scenarios, but the methodology does not include any time discounting. However, peak times are reported so that they can be used in the valuation phase.

Like the Site-specific approach (Section 3.1.2), the fate models for both direct discharges and disturbance of solid waste make use of the concept of the critical group. The results depend on the location of the critical group relative to the source. Therefore, the methodology is site-dependent, as reported in Table 1, produces worst case estimates of impacts and only allows aggregation at the level of impacts. The main feature of the Human Irradiation approach lies in considering both routine discharges and emissions from disposed nuclear waste. However, as noted in Section 3, LCA does not conventionally consider stochastic emissions and the inclusion of probabilistic emissions may inhibit general acceptance of the approach; this is further discussed in Section 4.1.

3.1.5 NDA’s Value Framework

The UK Nuclear Decommissioning Authority (NDA) – a non-departmental public body of the British Department of Energy and Climate Change (DECC) - developed an approach, termed Value Framework, for the purpose of demonstrating that it is delivering value for money across its entire estate (Wareing 2009). Like the Human Irradiation approach (Section 3.1.4), the NDA approach includes both “direct” discharges and stochastic emissions from solid waste (see Figure 3). The NDA adopted a Cost-Benefit Analysis (CBA) approach based on that mandated by UK Treasury (HM Treasury 2003). CBA relies on reducing environmental impacts to monetised damage costs, an approach that has been widely criticised, most significantly because it assumes that “value” is a single attribute that can always be reduced to monetary terms. This enables different attributes to be aggregated into a single figure for “benefit” (included avoided damages) that can simply be compared with the costs of the remedial action (e.g. Foster 1997, RCEP 1998). In LCA, this approach is usually termed “Valuation” and has generally been rejected (Baumann and Tillman 2004). Thus the NDA methodology itself is not likely to achieve the acceptance necessary for general adoption in LCIA. We therefore focus here how radiological impacts are modelled in the NDA approach and whether the approach might be adapted for LCA.

The value framework consists of a set of four criteria (attributes) that represent the key aims of NDA’s mission: Hazard Reduction, Safety and Security, Environment and Socio-economic. The NDA interprets the Environment attribute in terms of limiting radiological and non-radiological discharges separately. For radiological impact assessment, the relevant attributes are Environment and Hazard Reduction; the former deals with ‘direct’ discharges from routine processing operations, whilst the latter applies to facilities containing radioactive materials, including solid waste. As hazard reduction is the main benefit, in the NDA approach the ‘direct’ radioactive discharges refer to the emissions arising from hazard reduction activities. Also like the Human Irradiation approach, the NDA methodology combines a site-specific approach for routine discharges (including fate, exposure and effect analysis) and an approach based on risk assessment for emissions from solid waste. Radiological impacts on the human population are expressed, using the site-specific approach, in terms of collective dose to a critical group of people over a specified time period. The costs of different practices are evaluated against estimated reductions in the collective dose using a damage cost figure first proposed by NRPB in 1986 and revised in 1993 (NRPB 1993) to £20k/manSv for doses to the general public. Scaled to 2017 values, the equivalent figure is £25k/manSv.

Hazard Reduction, rather than the Environment attribute, is identified as being the main benefit delivered by the NDA. The metric describing hazards is termed the Safety and Environmental Detriment (SED) score, a measure of the hazards posed by different storage facilities, which assesses the potential impact of releases of stored material into the environment, taking into account facilities' conditions, typology and status of contents but not considering the probability of an event leading to release. The SED score is based on the assumption that all the facility contents are released in their most dispersible form. This is in marked contrast with the Human Irradiation approach (Section 3.1.4) which adopts a risk assessment perspective based on the probability of occurrence of stochastic events. In this perspective, the kind of worst case scenarios considered by the NDA approach would be ranked as improbable and considered nugatory, whereas the NDA argues that improbable events occur on a regular basis so that hazards evaluation must be based on worst case scenarios (Wareing 2009). Two SED scores can be calculated: one that applies to facilities and another to areas of contaminated land. In practice, the SED scores are calculated from several parameters, termed "descriptors", that describe various features of the waste stored and the nuclear facility or land to be remediated (such as the Radiological Hazard Potential, Facility Descriptor, Speed to Significant Risk, etc.). For each descriptor, the NDA has developed a table that includes sets of statements describing different states of the feature represented by the descriptor; each set of statements is associated with a numerical value to be used in calculating the SED score (NDA 2011). A notable example is the Facility Descriptor, i.e. a set of statements describing whether the building is within its original design life, whether it has any known defects, etc. The set of statements that best describes the current state of the facility is selected, possibly introducing an element of subjectivity into the methodology.

The crucial feature of radiological impact assessment in the NDA value framework is its ability to consider discharges from both routine operations and possible disturbance of solid waste (even though the former only refers to emissions from hazard reduction activities). However, the approach used for calculating a single SED score represents a considerable limitation for inclusion in LCIA. Furthermore, as noted for the Human Irradiation approach (Section 3.1.4), inclusion of stochastic events (discussed in Section 4.1) and use of site-dependent fate models (Section 4.2) may represent a barrier to acceptance.

3.2 Ecological impacts

3.2.1 Environmental Irradiation approach

As discussed above, radiation protection is chiefly focussed upon human impacts; however, there are movements to embrace impacts on both human and non-human entities. Along with the Human Irradiation Category discussed in Section 3.1.4, Solberg-Johansen (Solberg-Johansen et al. 1997, Solberg-Johansen 1998) also considered an Environmental Irradiation Category using the approach summarised in Figure 4. The methodologies share the same fate model (and thus are subject to the same limitations as site-dependent models), but differ in three aspects (see Figure 3 and Figure 4). Firstly, due to lack of knowledge about the effects of radionuclides upon non-human biota, the Environmental Irradiation approach does not adopt a risk metric. Secondly, it identifies as receptor the environment as a whole. Exposure analysis is therefore not needed: exposure is represented by the total concentration of radionuclides in each environmental medium. Finally, the approach includes only routine discharges; however, it could also be extended to include emissions from solid waste repositories (illustrated by the dotted line in Figure 4), provided that the exposure concentration in different media can be derived from the site-specific models.

The contribution of each radionuclide to Environmental Irradiation category in each environmental medium is calculated as the product of the radionuclide environmental concentration and an effect factor. The environmental concentrations are quantified using the same fate models as in the Human Irradiation Category; this implies that the methodology is site-dependent and produces worst case estimates of impacts (see Table 1). However, the Effect Factors are not related to exposure routes and risk-based dose relationships; instead, they rely on the "Environmental Increment" (EI) concept (Amiro 1993). This is based on the assumption that as organisms have always been exposed to some natural background concentration of radionuclides, they can tolerate a range of concentrations within the local natural variability – which represents one of the main arguments against the LNT model (see Section 2.2) (Allison 2015, Siegel et al. 2015). Consequently, Amiro arbitrarily assumed that an additional concentration of up to one standard deviation of the "background noise" is environmentally acceptable and represents one unit of Environmental Increment (EI) for each radionuclide. It must be noted that the EI factors are not necessarily related to toxic shock, and can only be used as screening tools to give an indication of the potential harmful concentration of radionuclides released to the environment. Adoption of the EI concept also means that the approach takes a mid-point perspective. The Effect Factors are calculated from the EI for a specific time period over which detrimental effects are considered. Since the total impact potency of a radionuclide is calculated over its life-time in the environment, the Effect Factor allows for the limited time period by weighting in inverse proportion to the "life-time" of each radionuclide, represented by the reciprocal of its decay constant. The Environmental Irradiation approach most resembles the ecotoxicity category in conventional LCIA categories: EI values play the same roles as the Maximum Tolerable Concentration factors in the ecotoxicity effect factors (Solberg-Johansen 1998).

The EI approach can readily be applied to radionuclides that occur naturally in significant quantities, as sufficient data regarding their environmental concentration and variability can be found. However, a problem is posed by anthropogenic radionuclides (i.e. radionuclides produced by humans, mainly arising from the nuclear industry, that would not otherwise be found in appreciable quantities in nature), for which no natural baseline concentration can be established. In this case, the approach proposes to base the Environmental Increments on other radionuclides with analogous chemical behaviour; for instance, Iodine 127 may be used as a proxy for Iodine 129.

3.2.2 SLERA

SLERA – Screening Level Ecological Risk Assessment – represents the first attempt to develop an ecological impact category for radionuclides in the same form as that used for non-radiological toxic substances, e.g. as in USEtox (Henderson et al. 2011). The approach has been tested by its authors in a case study for the Rhone river watershed (Garnier-Laplace et al. 2009). SLERA is a screening-type approach conceived to evaluate and compare potential effects of different emissions to receptor ecosystems. Screening-type approaches are usually recommended as first tier in Ecological Risk Assessment (ERA) (European Commission 2003, Beresford et al. 2007). Their purpose is to offer a simple and quick assessment with the lowest data requirement, by comparing estimated values with threshold levels, e.g. Predicted No-Effect Concentration (PNEC). The SLERA approach was born as a spin-off from the ERICA (Environmental Risks from Ionising Contaminants: Assessment and Management) project for LCA purposes. ERICA (Brown et al. 2008, Howard and Larsson 2008, Larsson 2008) is one of a number (e.g. Copplestone et al., 2001; Environment Canada, 2001; US DoE, 2003) of initiatives aimed at establishing a scientific, internationally accepted system for assessing the ecological impact of ionising radiation. The ILCD found SLERA to be the best available characterization method for assessing radiological impacts on ecosystems (Hauschild et al. 2013) but did not go so far as to recommend it for use; rather it is classified as an interim methodology, mainly because it still has to be reviewed fully.

SLERA addresses potential effects of both chemicals and radioactive substances entering the environment as routine emissions; here we focus on the radioactive substances. Although the methodology has the potential to cover emissions to all environmental media (i.e. freshwater, sea water, soil, etc.), it has so far been developed solely to address the freshwater impacts; i.e. effects on non-human biota of emissions to freshwater bodies. As shown in Figure 4, the methodology comprises the two familiar steps of fate and effect analysis.

The fate analysis uses the MacKay modelling approach and employs a single box-type dilution model to estimate the concentration of a given substance in fresh water. The methodology is thus site-independent, produces average estimates of impacts and also allows aggregation at the inventory level. Concentration in sediments is then calculated by means of partition coefficient (Kd) parameters, while concentration in organisms is obtained through Concentration Ratios (CR); these parameters represent the equilibrium concentration ratio between two environmental media (e.g. soil and water) and between organisms and environmental media (e.g. algae in lakes) respectively. Both Kd and CR values are taken from the ERICA project database (Beresford et al. 2007).

The effect analysis relies on the Potentially Affected Fraction (PAF), which is usually defined as a mid-point indicator, of species as a proxy of the potential damages to ecosystems. The PAF expresses the percentage of species that experience an exposure level above their EC50, i.e. the concentration at which 50% of the population experiences a deleterious effect such as inhibition of growth and mortality (Pennington et al. 2004). The effect factor expresses the increase of PAF per unit of concentration of a given radionuclide and is obtained directly from the HC50, another parameter frequently used in toxicology. The HC50 represents the environmental concentration for which 50% of the species in the ecosystem experience concentration values above their EC50, and is calculated as the geometric mean of the EC50 for all the species considered (see Pennington et al. 2004). For chemical substances, HC50 and EC50 values for different biotic species can be obtained directly from laboratory tests. For radionuclides, however, effects are related to the (absorbed) dose rather than the concentration (Garnier-Laplace et al. 2008). In this case, EC50 and HC50 are obtained from two other parameters: the Effective Dose Rate (EDR_{50}) and its associated Hazardous Dose Rate (HDR_{50}), which are the equivalents of EC50 and HC50 but refer to the dose rather than the concentration; for instance, the EDR_{50} represents the effective dose giving a 50% change in observed effect from chronic exposure (Garnier-Laplace et al. 2008). The scarcity of these data represents one of the major limitations of this methodology.

4 Discussion: Incorporating impacts of radionuclides in LCIA

In Section 3, seven methodologies for assessing the impact of ionising radiations have been reviewed; their principal features and differences are summarised in Table 1. Amongst these, only two methodologies - Environmental Irradiation (Section 3.2.1) and SLERA (Section 3.2.2) – deal with radiological impacts on non-human biota; all the others focus on potential human health effects. The site-specific approach (Section 3.1.2) and the NDA's value framework (Section 3.1.5) represent the only methodologies developed as assessment procedures in other fields, not specifically for LCA.

As noted at the beginning of Section 3, LCA impact methodologies must meet a number of criteria to find broad acceptance; notably, four main criteria have been mentioned. These deal with the scientific basis of the methodology, the principle of linearity between emissions and impacts, the inclusion of fate, exposure and effect analysis as relevant, and the geographic and time coverage. Besides these generic criteria, which apply regardless of the scope of the methodology, other criteria may also be considered, specific to each impact. In the case of radiological impacts, a specific criterion is the ability to include both continuous direct discharges from operations and future emissions from radioactive waste disposed in final repository.

4.1 Inventory issues: inclusion of emissions from solid waste

The incorporation of future emissions from disposed radioactive waste represents the most critical inventory challenge for radiological impact assessment in LCA. Amongst the approaches reviewed, Human Irradiation and the NDA value framework (discussed in Sections 3.1.3 and 3.1.5 respectively) are the only ones considering emissions from nuclear waste; in principle, also the Environmental Irradiation approach could include such releases, but this has never been operationalised. The remaining methodologies simply avoid the issue by neglecting the potential radiological impacts associated with nuclear waste (see Table 1).
The most accepted solution for disposal of nuclear wastes envisages their disposal in either near-surface or deep underground repositories, according to their level of radioactivity. Although repositories are projected to last indefinitely, it is expected that, at some point in the distant future, the containment system will fail due to either corrosion by groundwater or due to stochastic disturbance of the repository. The former may be regarded as the "natural evolution" of the system and can be predicted, although with much uncertainty due to the long time frame involved. The latter, on the other hand, includes that probabilistic element which, as noted in Section 2, falls outside the scope of established LCA.

The need to incorporate impacts from nuclear waste prompted Solberg-Johansen to develop a novel approach for radiological impact assessment. The distinguishing feature of Solberg-Johansen's Human Irradiation methodology (Section 3.1.4) is indeed the inclusion of both routine direct releases from processing operations and future emissions from disposed solid waste; the methodology, however, considers both deterministic and probabilistic events as in common approaches to Risk Assessment. Solberg-Johansen argues that this is the only way to take into account both types of emission, as a dose-based system would ignore possible releases that can result in massive radiological impacts. Her Environmental Irradiation approach could be operationalized to apply the same approach to environmental rather than human impacts from solid waste. The NDA approach (Section 3.1.5) also includes both direct discharges and emissions from solid waste; however, as opposed to the Human Irradiation approach, it includes only the most hazardous and thus least likely events, without regard to their probability of occurrence. This means that the kind of events considered by the NDA would be deemed impossible and excluded in a probabilistic assessment perspective.

The inclusion of stochastic emissions for assessing impacts from radioactive waste is likely to be contentious because, as noted above, LCA does not conventionally deal with this type of event. If stochastic events are considered for radiological impacts, should they also not be included for non-radiological impacts? A notable example may be found in mining operations, where numerous environmental disasters have been caused by failures of tailing dams (e.g. see Hatje et al. 2017) constructed to impound the fine tailing materials left from milling of mined ores (Kossoff et al. 2014). Recent studies have shown that tailings dams failures occur more than once a month worldwide (Rico et al. 2008, Azam and Li 2010) – although the number has considerably decreased from the 1970s and 80s – and that at least one major failure occurs each year (ICOLD 2001). However, databases such as Ecoinvent (Wernet et al. 2016) include short and long-term emissions to atmosphere and to groundwater (leachate) from uranium (Doka 2009, Dones et al. 2009) and generic sulfidic tailings (Doka 2008) but do not include tailings dam failures. This highlights the general methodological question: if stochastic emissions from solid nuclear waste are included, should failures of tailings dams not be considered given their frequent occurrence and potential to cause environmental damage? This question applies equally to many other operations commonly included in LCA, such as landfilling where routine operation is normally included but containment failure is not.

The general issue of the treatment of stochastic events in LCA is not addressed here. Rather, we focus on the specific question of how to include emissions from radioactive solid waste. Given the significance of the production of solid waste in the nuclear fuel cycle, excluding the disposal route for solid waste could lead to incorrect conclusions. The comparison between reprocessing and direct disposal of used nuclear fuels is a relevant example. Effectively reprocessing separates long-lived nuclear materials with long half-lives, such as uranium and plutonium that can be reused as fuel in nuclear reactors, from fission products, which represent the by-products of the fission reaction. If it is assumed that the vitrified waste containing the fission products is segregated from the uranium and plutonium, reprocessing generates a final nuclear waste that has a shorter half-life and is less radioactive than the spent nuclear fuels. Therefore, if radiological impacts from nuclear waste are neglected, the advantages of reprocessing in terms of reduced radioactivity of the waste will be lost in the assessment. Because of its ability to consider emissions from solid waste, the approach proposed by Solberg-Johansen appears very promising, provided that only the "natural evolution" scenario is considered and probabilistic releases are not. By contrast, the NDA framework appears to be unsuitable for inclusion in LCIA since it only considers stochastic events, and more specifically only the most hazardeous, and thus least likely one. Neglecting emissions from solid waste represents a crucial flaw of the remaining methodologies, and the main barrier towards their accceptance for general adoption in LCIA.

The inclusion of future emissions from radioactive solid waste is also accompanied by another issue: how should impacts caused by current and future emissions be compared? Radiological impacts linked with radioactive solid waste occurs on very long time scales (tens to hundreds of thousands of years) compared to other kinds of impacts. The ability to compare future and present emissions is particularly important if radiological impacts from solid waste are to be compared with those associated with direct gaseous and liquid discharges, for instance in choosing between reprocessing and direct disposal approaches. At present, impacts from future emissions are poorly handled in LCA: they are either time discounted or cut-off by limiting the time span of the assessment. Both approaches are unsuitable for radiological impacts because they would inevitably overlook impacts arising from disposed solid waste. Hence, development of an approach for incorporating future emissions in LCA is of extreme importance for radiological impacts.

4.2 Human Health impacts

Table 2 lists approaches currently used to incorporate human impacts of ionising radiations in LCIA, along with the metric adopted and characteristic features of each methodology. Only the Human Health Damages approach (discussed in Section 3.1.3) is currently ever included in LCIA; other LCIA methods not listed in the table - e.g. TRACI (Bare 2011) and EDIP (Hauschild and Potting 2006) - omit radiological impacts completely. Although only the Human Health Damages approach is incorporated, characterisation factors differ between the methods. This is due to differences in the metrics used, the scenario adopted within the cultural perspective, and also whether hereditary effects are included in the effect analysis.

Table 2 – Radiological impact categories in LCA impact methods

Method	Version	Approach	Metric	Features	Source
CML	2001, non baseline	Human Health Damages	DALY	Egalitarian/Hierarchist	(Guinée et al. 2002)
RECIPE	1.08; 1.07	Human Health Damages	KgU_{235} eq.; DALY	Egalitarian/Hierarchist and Individualist; Hereditary effects.	(Goedkoop et al. 2013)
Eco-Indicator	1999	Human Health Damages	DALY	Egalitarian/Hierarchist and Individualist.	(Goedkoop and Spriensma 2001)
Impact2002+	2002	Human Health Damages	DALY	Egalitarian/Hierarchist and Individualist.	(Humbert et al. 2012)
ILCD	2011	Human Health Damages	KgU_{235} eq.;	Egalitarian/Hierarchist	(JRC 2011, 2012)

The Human Health Damages approach was developed specifically for use in LCIA and embodies two features specific to this use (see Table 1). First, by adopting an end-point perspective, expressed in terms of DALYs, it allows comparison of radiological and non-radiological impacts. The methodology is therefore suitable for those LCA methods, such as RECIPE, that aim to aggregate different end-point impacts into a smaller number of parameters but it is less appropriate for those methods, such as CML, that adopt the mid-point perspective. However, as others have pointed out (e.g. Bare et al. 2000), although end-point analysis may be more accessible to a non-expert audience, it has the significant disadvantages of increasing uncertainty and reducing transparency; adopting the mid-point perspective enables radiological and non-radiological impacts to be kept distinct. Second, the Human Health Damages approach produces characterisation factors that represent average estimates of impacts and are intended to be location-independent, thus allowing aggregation of inventory data across the life cycle. Notably, the methodology is applied regardless of the source of emissions; however, as noted in Section 3.1.3, the underlying model was developed for French installations so that there are major uncertainties in applying it elsewhere. A further limitation of the approach is that it includes only a small proportion of the radionuclides that are or could conceivably be discharged, thus neglecting some with potentiallly significant impacts such as atmospheric discharges of Strontium 90, Americium 241, and Curium 242.

Amongst the other approaches for human health reviewed here, Human Irradiation (Section 3.1.4) appears to be of particular interest, mainly because, as discussed in Section 4.1, it considers impacts of both direct discharges and solid waste, but also because it includes a significantly higher number of radionuclides than the Human Health Damages approach. More than one hundred radionuclides are included for direct discharges, but only a limited number is considered for emissions arising from solid waste, on the basis that only a few radionuclides last long enough to be present in escapes from a final repository. Like the Human Health Damages approach, Human Irradiation adopts an end-point perspective; however, risk rather than DALY is used as metric. Adhering to this metric would limit comparison of radiological and non-radiological impacts (usually quantified in DALYs) but the approach could be easily reformulated in terms of DALY. The main limitation of the Human Irradiation approach is that, unlike the Human Health Damages approach, it is site-dependent (see Table 1), linked with identification of the critical group. Therefore, although the models for estimating the environmental concentrations are geographically generalised, some specific information is still needed, such as the location of the critical group: characterisation factors are dependent on the distance between the critical group and the source of emissions, meaning that more than one set of factors is needed and that inventory data cannot be aggregated across the life cycle. Furthermore, both the generic nature of the fate models and the use of the critical group mean that characterisation factors represent worst case estimates of impacts, whereas LCA studies are concerned with relative comparisons between product systems and therefore need average rather than worst case values (Guinée et al. 2002). The Human Irradiation approach has two further limitations. First, the models used for estimating the environmental concentrations of radionuclides have been developed for use within a limited area (usually a regional scale), and their application on bigger scales may lead to major uncertainties. Second, characterisation factors for solid waste incorporate a number of assumptions regarding the final repository including the geology at the potential site, the size and layout of the repository and the type and contents of the canisters for the disposed wastes that may not be representative of future operating repositories.

Like Human Irradiation, the NDA approach (Section 3.1.5) for radiological impact assessment has the merit of including both direct discharges and emissions from solid waste. In addition to the aspects discussed in Section 4.1, the approach has two major limitations. First, the approach devised for calculating a SED score is based on expert judgments and is therefore open to the criticism that it lacks a rigorous scientific basis. Second, impacts of direct radioactive emissions are estimated by means of a site-specific methodology (e.g. similar to CREAM, discussed in Section 3.1.2). This is the most accurate and reliable way of assessing the actual rather than potential impacts of radionuclides but it is difficult to reconcile with the geographically generalised approach conventionally used in LCIA. A site-specific model requires a considerable body of specific data and its output describes the effects of emissions within a very limited regional scale on a specific group of individuals realistically considered to be the most exposed (critical group), meaning that it only produces worst case estimates. The geographical coverage may in principle be extended to wider scales - regions, countries and continents - but this would greatly increase both the amount of site-specific data required and the time and complexity of calculations.

The remaining approaches are either inherently unsuitable for LCA or are insufficiently mature. The Critical Volume approach (Section 3.1.1) is the simplest approach with the least data requirements; it needs as input the amount of radioactive emissions released and the only further parameter used is the Annual Limit of Intake (ALI). However, it is also the most limited approach as the fate of radionuclides is not accounted for and all exposure pathways other than ingestion are neglected. It must be noted that this approach was proposed when no others were available; as such it played a crucial role by acting as a stimulus for the development of more representative models. Site-specific approaches (Section 3.1.5), such as CREAM, have mainly been developed for use by nuclear site operators with the aim of proving compliance with legal regulatory limits, and are therefore subject to requirements that make them inappropriate for LCIA. Limitations of site-specific methodologies have been already discussed above.

In view of this analysis, it is clear why the Human Health Damages is the only approach currently used: it holds a number of features that makes it easily applicable and consistent with conventional LCA. However, as noted above and in Section 4.1, the approach has significant limitations in terms of type of emissions and number of radionuclides covered, fate modelling and approach to modelling the cause-effect chain. To achieve acceptance for adoption in LCIA a radiological impact assessment approach should employ site-independent fate models and combine the extended radionuclides inventory of Human Irradiation with the kind of average estimates considered in Human Health Damages, expressed primarily through mid- point and possibly also through end-point indicators for comparison with non-radiological impacts.

4.3 Ecological impacts

As opposed to Human Health, approaches for assessing ecological impacts are yet to be included in any LCIA method. Amongst the approaches reviewed, SLERA (Section 3.2.2) appears to be the most promising; notably, it was found to be the best available characterisation method by the ILCD, although it was not recommended as ready for implementation (Hauschild et al. 2013). SLERA has the merit of representing an important step towards complete harmonisation of toxic and radiological impacts. The methodology is in fact closely related to the approach used for assessing ecological toxic impacts in USEtox (Henderson et al. 2011); notably, like USetox, SLERA adopts the Mackay modelling approach and mid-point indicators expressed in terms of the Potentially Affected Fraction (PAF) of species. As noted above, the latter is a favourable feature as it avoids the criticisms directed at end-point indicators; however, it is possible to develop the approach to an end-point perspective by means of the Potentially Disappeared or Vanished Fraction (PDF or PVF) indicator (Larsen and Hauschild 2007). In addition to inventory issues discussed in Section 4.1, SLERA has two major limitations. At present, it only considers emissions to fresh water (Table 1); it needs to be extended to cover all relevant types of emissions. Second, the lack or shortage of data on the bio-availability of radionuclides and the effects of ionising radiations to non-human biota represent considerable obstacles for calculation of the effect factors. This is, however, not specific to SLERA; rather it is a broad issue affecting every methodology dealing with radiological impacts on ecosystems.

Environmental Irradiation, discussed in Section 3.2.1, is the only other approach for ecological impacts. The main differences between the two approaches lie in the inclusion of emissions from solid waste (discussed in Section 4.1) and in the effect analysis, which uses site-dependent models (whose limitations have been discussed in Section 4.2) and Environmental Increments (EI) instead of PAF to quantify potential impacts on ecosystems. However, since the EI values are based on strong assumptions on the range of environmental radioactivity non-human biota can tolerate, the use of PAF is favoured. The Environmental Irradiation approach is also subject to a limitation similar to that discussed for SLERA regarding the shortage or lack of data for calculating effect factors, in this case primarily on the concentration and natural variability of radionuclides in environmental media used to produce the Environmental Increments factors.

In light of this analysis, the main challenge of ecological impact approaches lies in the availability of ecotoxicological data without which detrimental effects of neither direct discharges nor emissions from solid waste on ecosystems can be properly quantified. Furthermore, the amount of ecotoxicological data available also affects the number of radionuclides that can be included in any ecological impacts approach. A suitable LCIA approach for impacts on ecosystems should be based on SLERA, and include all relevant types of emissions including those arising from disposed solid waste; notably, the SLERA approach has already been designed with comparison of radiological and non-radiological impacts in mind.

5 Conclusions and future work

This article has presented a comprehensive review of approaches for radiological impact assessment to prepare the way for the development of a general approach to incorporate the impacts of radionuclides in Life Cycle Assessment. Although a number of approaches have been proposed either specifically for LCA purposes or for use in other types of assessment, none is sufficiently comprehensive, mature or consistent with life cycle thinking for general adoption in LCIA.

The main limitation common to both human and ecological impact assessment lies in the inclusion in the inventory of emissions from radioactive waste disposed in a final repository alongside direct routine discharges. Although the inclusion of stochastic emissions represents a possible approach to account for potential impacts associated with stored nuclear waste, it would be inconsistent and outside the current scope of LCA, which only deals with routine, planned discharges. The Human Irradiation approach for radioactive waste represents the most promising approach provided that stochastic emissions are left out and only the "natural evolution" of the repository is considered.

For human impacts, the Human Health Damages is the only approach currently included in LCIA methods; however, it has serious limitations and the Human Irradiation approach devised by Solberg-Johansen appears to overcome some of these. To achieve broad acceptance in LCA, a radiological impact assessment approach should employ site-independent fate models and combine the extended radionuclides inventory of Human Irradiation with the kind of average estimates considered in the Human Health Damages approach, expressed primarily through mid- point and possibly also through end-point indicators for comparison with non-radiological impacts.

The main challenge for estimating potential impacts on ecosystems consists in gathering more information on both the bio-availability of radionuclides and their effects on non-human biota. The SLERA methodology appears to represent a promising approach, especially in harmonising impact assessment for toxic and radioactive substances, but the methodology needs to be extended to cover all relevant types of emissions.
In light of this review and its conclusions, a novel, overarching framework for radiological impact assessment on humans has been devised, potentially suitable as a basis for including radiological effects in Life Cycle Impact Assessment. The framework would combine elements of the Human Irradiation approach proposed by Solberg-Johansen (Sections 3.1.4) with the approach embodied in SLERA to assess impacts on ecosystems. The methodology is now being developed in detail to derive impact factors for use alongside non-radiological impacts in proceeding from Inventory Analysis to Impact Assessment.

6 Glossary

ALI	Annual Limit of Intake
CREAM	Consequences of Releases to the Environment: Assessment Methodology
DALY	Disability Adjusted Life Years
DDREF	Dose and Dose-Rate Effectiveness Factor
EI	Environmental Increments
IAEA	International Atomic Energy Agency
ICRP	International Committee on Radiological Protection
LNT	Linear Non-Threshold model
NDA	Nuclear Decommissioning Authority
NCRP	National Council on Radiation Protection and Measurements
NRPB	National Radiological Protection Board
PAF	Potentially Affected Fraction of species
SED	Safety and Detriment Score
SLERA	Screening Level Ecological Risk Assessment

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