Operationalising the use of Life Cycle Assessment to nuclear waste management

Andrea Paulillo

Department of Chemical Engineering

University College London

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I, Andrea Paulillo, confirm that the work presented in this thesis is my own. Where information has been derived from other sources, I confirm that this has been indicated in the thesis.

Abstract

After decades of declining interest, nuclear energy is poised for a comeback in the UK, driven primarily by pledges and binding agreements on limiting greenhouse gas emissions, but also by increasing energy security concerns. However, the industry has yet to tackle some of its most crucial challenges regarding management of used nuclear fuels, and especially of highly radioactive wastes. Life Cycle Assessment (LCA) – indeed the most mature and also the only standardised life-cycle methodology – represents a widely accepted tool for quantifying the environmental impacts associated with goods or services and supporting decision-making processes. This Thesis aims at operationalising the use of LCA to nuclear waste management.

After introducing the LCA standard methodology, the Thesis proceeds with a comprehensive review of methodologies for assessing radiological impacts – the lack of an appropriate approach for radiological impacts in LCA is in fact identified as the crucial barrier for its application to the industry, especially with respect to waste management. Building upon the main findings of the review, the Thesis presents an overarching framework and two practical methodologies – namely UCrad and the Critical Group Methodology (CGM) – for assessing radiological impacts of direct discharges, and crucially, of nuclear waste disposed of in a geological repository. The LCA and the methodologies for radiological impacts are then applied to two case studies. The first is a prospective attributional study that examines the current procedure for managing used nuclear fuels and the UK Government policy for disposal of nuclear waste in the UK. The objective is to identify hot-spots and suggest potential improvements. The study shows that the highest impacts are due to the production of chemicals required by the reprocessing process and the materials used for High Level Waste canisters rather than the construction and decommissioning of a final repository for nuclear waste. The second study focuses on future scenarios for managing used nuclear fuels in the UK, including direct disposal and four reprocessing options, and clearly demonstrates how LCA can be used to support decisions. Reprocessing of uranium, but especially of plutonium, is shown to be of crucial importance from an environmental perspective.

Impact

Life Cycle Assessment (LCA) is the oldest, the most developed and widely used, and the only ISO standardised life-cycle methodology. The European Commission (EC) has concluded that LCA provides the best available framework for assessing the potential environmental impacts of products [1] and selected LCA as the reference model for the impact assessment of policies in the European Union [2]. However, the use of LCA in the nuclear industry is, yet, quite limited, especially in supporting decision-making processes. Consideration of environmental impacts from a life cycle perspective are imperative if nuclear energy is to play a key role to global and national decarbonisation policies.

Within this context, this Thesis aims to play a crucial role for a wider adoption of LCA as a decision-support tool in the nuclear industry. This objective is achieved through:

- development of a novel methodology for assessing radiological impacts in LCA that allows comparison between direct discharges and nuclear waste impacts, and dissemination of its characterisation factors for inclusion in widely adopted LCA methods;
- application of the LCA methodology to two illustrative case studies, both extremely relevant to the UK nuclear industry. These evaluated the environmental impacts of current and future approaches for managing used nuclear fuels, thus addressing the question of whether the UK should continue reprocessing used fuels (current) or it should move towards their direct disposal.

At the time of writing, this project has already attracted considerable interest in the UK; this is demonstrated by two consulting projects being pursued by the Author and some of the project Supervisors with the UK Environment Agency and the National Nuclear Laboratory.

Acknowledgments

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Chapter 1.
Introduction

Sustainable development can be regarded as one of the most important concepts of the 21st Century, developed in the mid-90s at a time when human kind began to realize the adverse effects of human activities on the planet and its ecosystems. In recent years, the Planetary Boundaries approach attempted to quantify the boundaries for those processes that are key to earth functioning, thus identifying an effective "safe operating space for humanity". Climate change driven by increase in greenhouse gases concentrations represents only one of the boundaries that have been exceeded, but indeed it is the one gaining the most attention globally. Because it is still heavily reliant on fossil fuels, the energy sector, and especially electricity generation, has the highest potential for reducing greenhouse gas emissions. The Paris Agreement (signed in 2015) marked an historical milestone in the path of human kind to reconcile its activities with planet Earth with a global action plan to curb greenhouse gas emissions and restrain global warming to well below 2 °C. The United Kingdom, as member of the European Union, committed to the goals of the Paris Agreement. Although it outperformed the first carbon budgets, current policies are not sufficient for achieving long-term goals. The UK Government regards nuclear energy as a key contributor to its decarbonisation path, and in future scenarios nuclear energy could contribute up to 40-50% to the UK electricity mix. The nuclear fuel cycle includes all the stages and activities associated with the production of electricity, starting from mining of uranium, through enrichment and fabrication of nuclear fuel, and operation of power reactors, to management and disposal of nuclear waste. The nuclear industry is facing many challenges including finding a sustainable way of disposing of highly radioactive waste; in this context, life-cycle thinking methodologies can be used to assist decision-making processes. This Thesis focuses on operationalising the use of Life Cycle Assessment (LCA) to the nuclear industry to demonstrate how it can be used as a decision-support tool.

1.1 A safe operating space for humanity

It was not until the 20th Century that human kind began to realize the increasing pressure it was putting on the planet and its ecological systems. Rapid development in science and technology allowed the symptoms of this extreme pressure to be monitored, investigated and linked to different human activities. In 1987, the concept of sustainable development was developed to provide a framework for the integration of environment policies and development strategies. It was defined as the "development that meets the needs of the present without compromising the ability of future generations to meet their own needs" [3]. The definition of sustainable development contains two key concepts: needs, in particular the essential ones of the world's poor, to which higher priority should be given and limitations imposed by the state of technology and social organisation on the environment's ability to meet present and future needs. The concept of sustainable development revolutionised the idea of growth – previously intended solely as economic growth – to incorporate also environmental and social aspects. For this reason, the idea is often depicted as three circles, each referring to one of these aspects: sustainable development is found in the overlap of these circles (Figure 1.1 A).

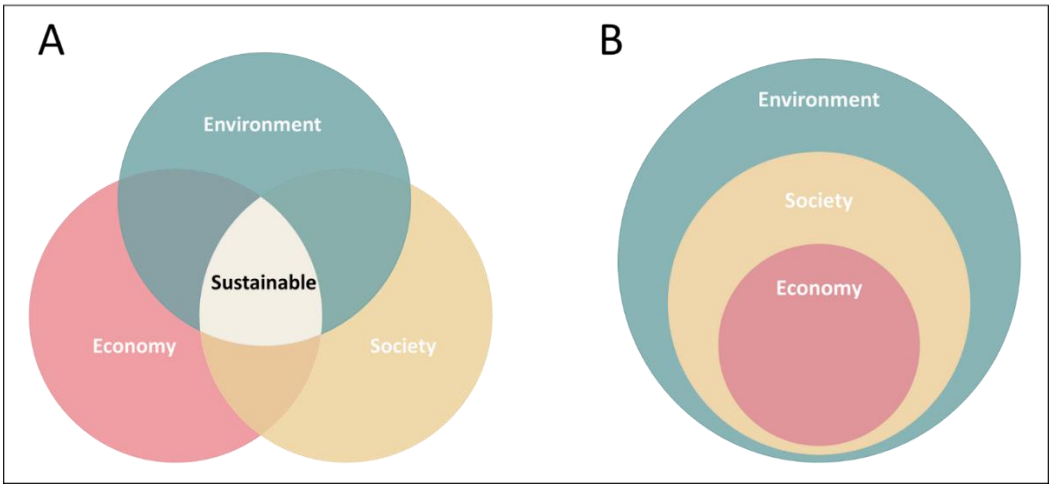

Figure 1.1 – The overlapping and enclosed circles of Sustainability

Although the effects of soaring environmental pressure were perceived in the 20th century, overwhelming scientific evidence arrived only in the 21st century. The knowledge build-up eventually lead to the formulation of the "Planetary Boundaries" concept [4], [5]: an attempt to quantify the boundaries for those processes considered to be crucial for the functioning of the Earth system that should not be transgressed. Effectively the

Planetary Boundaries define a safe operating space for humanity, within which we should aim to stay to avoid destabilizing the planet. The Earth has been in a relatively stable geological epoch, the Holocene, for over 10.000 years; this is the period that allowed agricultural and complex societies, including ours, to develop and flourish, and therefore it is the only one we know for certain can support contemporary human societies [6]. However, the Holocene is being destabilised by the dramatic growth of the human enterprise [7]–[9]; and it has been proposed that the Earth has entered a new epoch, the Anthropocene, where humans constitute the main driver of change to the Earth [10], [11]. The Anthropocene concept raised the question "what are the non-negotiable planetary preconditions that humanity needs to respect in order to avoid the risk of deleterious of even catastrophic environmental change at continental to global scales?" [4]. The Planetary Boundaries concept is based on the assumption that we want to keep a Holocene-like state: the range within which the Earth system processes varied in the Holocene is taken as a scientific reference point for a desirable planetary state. In accordance with the Planetary Boundaries Authors' view, the concept of sustainable development is best depicted by three enclosed circles (Figure 1.1 B), with environment the outermost and economy the innermost circle. They reason that environmental boundaries set the physical limits within which societal and economic developments can occur: the Planetary Boundaries represent such limits; they are depicted in Figure 1.2.

The Planetary Boundaries are not equivalent to a global threshold or tipping point: a precautionary principle has been used whereby boundaries delimit a safe zone (green) beyond which an uncertain one with increasing risk is identified (yellow). Finally, the uncertainty zone is also delimited and marks the entrance to a zone of high risk (red).

Nine Earth-system processes have been identified as key to Earth functioning, and eleven planetary boundaries based on the same number of control variables have been proposed. Notably, three planetary boundaries (novel entities, atmospheric aerosol loading and functional diversity part of biosphere integrity) are yet to be quantified. Of the remaining eight boundaries, three have already exceeded the zone of uncertainty: these represent loss of biodiversity (i.e. genetic diversity part of biosphere integrity) and interferences to nitrogen and phosphorus cycles (i.e. biochemical flows of phosphorus and nitrogen); and two boundaries are outside the safe operating space but within the zone of uncertainty, these represent loss of land covered by forest biomes (i.e. land-

system change) and change of atmosphere radiative forcing due to greenhouse gases (i.e. climate change).

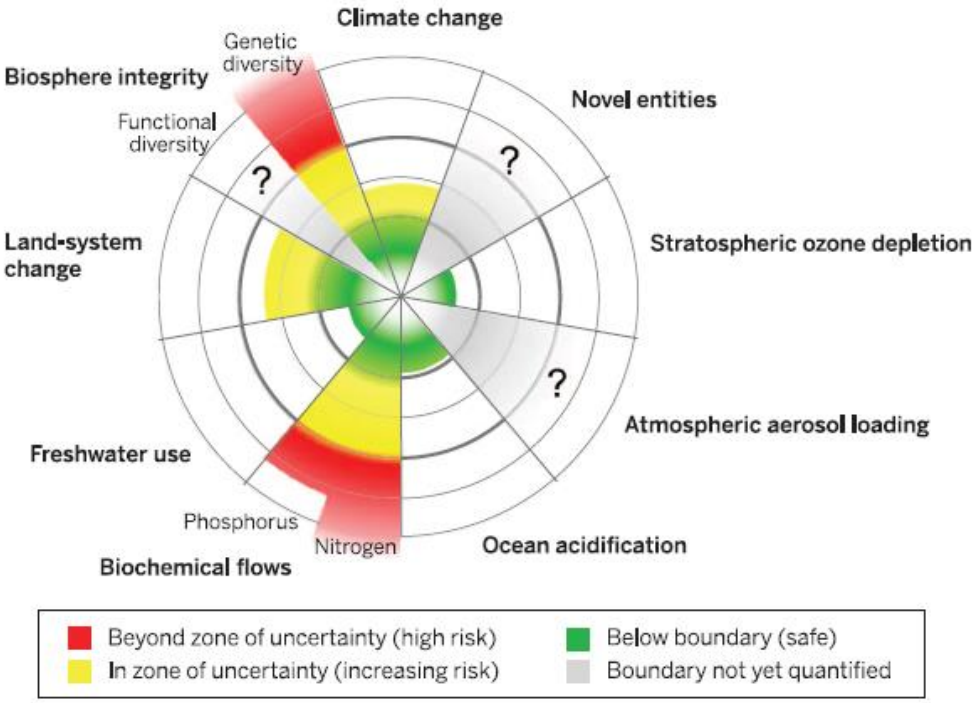

Figure 1.2 – Status of the control variables of the Planetary Boundaries [5]

It is not possible to single out a process, product, service or industrial sector that is the primary cause of the exceedance of planetary boundaries – that would simplify quite significantly our efforts to live within the safe operating space: it is easier to tackle one rather than a complex, interlinked net of causes. Agriculture, energy production, mining, transportation, manufactured products contribute to all planetary boundaries in different manners and magnitudes. For instance, the energy sector is the primary source of greenhouse gases; manufacture of fertilizers for the food industry and cultivation of leguminous crops are the main causes to distortion of the phosphorus and nitrogen cycles; the mining and agriculture sectors, and urbanization are the primary drivers for conversion of natural systems, and the consequent loss of habitat is the main cause to the decline of genetic biodiversity. Although a single cause cannot be identified, the rising environmental pressure on the Earth system can be linked to the rapid increase of human population driven by the great acceleration of the human enterprise in the 20th century [11]. At present, the rate of growth has slowed down, but according to the UN projections, the absolute number of people added each year will not begin to fall until mid-21th century [12]. The surge in the number of human beings leads to a rising demand for essential and

luxury goods: every human process is polluting, but it is their increasing magnitude that it is able to affect the Earth system as a whole. Human population is not one of the nine boundaries – although it has been proposed by some [13] – because it does not represent an environmental physical boundary; rather, it is a social limit that lies within the environmental safe operating space. Achieving population stabilisation represents one of the available options to restrict human development to within the nine planetary boundaries.

Although several boundaries are outside the safe operating space, climate change is certainly the one gaining much of the global attention – perhaps because global warming is easier to be monitored and its effects are being witnessed at an increasing rate in recent years. The Paris Agreement marked an unprecedented and historic event: it set out a global action plan to curb greenhouse gas emissions and restrain global warming to well below 2 °C [14]. Countries around the world are developing policies to contribute to a fundamental transformation in the energy sector, where reliance on fossil fuels is shifted towards alternative and renewable sources. This Thesis focuses on nuclear energy as an alternative, low-carbon source of electricity, with a focus on its environmental impacts.

1.2 World energy outlook

Massive energy production and consumption are the basis on which the modern society has developed and continue to do so. For this reason, the advocates of the Anthropocene concept assign its inception to the industrial revolution, regarded as "one of the three or four most decisive transitions in the history of human kind" [11]. As a matter of fact, industrial societies use four or five times as much energy as agrarian ones do which, in turn, used three or four times as much as did hunting and gathering societies [15]. Without the transition to an energy-intensive society, it is unconceivable that global population would have risen from about a billion in 1920 to more than six billion today or that we – or rather a billion of us amongst the more fortunate ones – could maintain high living standards.

Carbon dioxide (CO_2) accumulation in the atmosphere is regarded as the signature of the human imprint on the Earth system, and it can be used to track the progression of the Anthropocene. CO_2 concentrations have risen steadily from pre-industrial level – in the Holocene CO_2 concentration fluctuated between 260 and 285 ppm [16] – to over 300 ppm

by 1950 [17], after which it accelerated exponentially exceeding the 400 ppm threshold in 2016 [18]. For this reason, the period from the Second World War onwards is referred to as the "great acceleration" of the human enterprise, and is associated with massive increase in the human imprint [11]: three quarters of the anthropogenic driven rise in the CO_2 concentration has occurred since 1950 and about half of the total rise has occurred in the last 30 years.

Nowadays the energy market looks very different to that it used to in the past, and is required to change dramatically in the future if the Paris Agreement goals are to be met. According to the International Energy Outlook (IEO) 2016 published annually by the US Energy Information Administration (US-EIA), world consumption of marketed energy is predicted to increase by 48% from 2012 to 2040. The most marked growth in energy demand occurs in developing nations outside the Organization for the Economic Cooperation and Development (non-OECD) with a projected increase of 71%, compared with slower-growing OECD economies that are expected to grow by only 18%. Notably, economic growth – measured in Gross Domestic Product (GDP) – is a key factor in energy demand growth: non-OECD countries' GDP are projected to grow twice as fast as OECD countries. Figure 1.3 shows past and future projections of world energy consumption from different fuel sources over the 1990–2040 period. According to the US–EIA projections, demand increases for all energy sources other than coal. Fossil fuels continue to provide most of the world's energy, accounting for 78% of total consumption in 2040: liquid fuels remain the largest source of energy – although their share declines from 2012 to 2040 – whilst natural gas experiences the most marked growth, overtaking coal as the second largest source of energy. This is mainly driven by increased supply from shale formations, principally in the US and Canada. The slowing growth of coal is linked with climate policies aimed at reducing CO_2 emissions from the energy sectors: coal is thus being replaced by other fossil fuels, which have lower carbon footprints, or by other alternative energy sources. With government policies and incentives promoting the use of non-fossil energy sources in many countries, renewable energy and nuclear are currently the world's fastest-growing source of energy, at a pace of 2.6% and 2.3% per year respectively.

Electricity generation from nuclear power is forecast to increase from 2.3 in 2012 to 4.5 trillion kWh in 2040. Development of new nuclear capacity is stimulated by energy

security concerns and limits on greenhouse gas emissions, and occurs primarily in non-OECD Asian countries, with strongest growth projected for China and India, followed by Middle-East countries. The projections are also positively affected by world average capacity utilization rates, which have risen steadily in the past 3 decades from 68% in 1980 to 80% in 2012. By contrast, the Fukushima Daiichi disaster in March 2011 and planned retirements of nuclear power plants in OECD Europe countries have contributed to curbing nuclear energy growth.

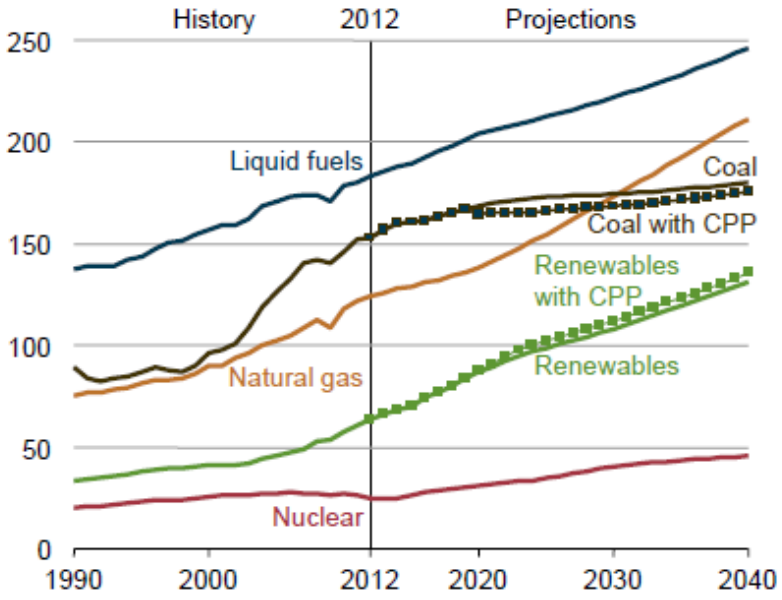

Figure 1.3 – Total world energy consumption by energy source, 1990–2040 (quadrillion Btu). CPP refers to the US Clean Power Plan issued in August 2015 [19] [1].

The electric power sector remains one of the most dynamic areas of growth amongst energy markets. Electricity is the world's fastest-growing form of end-use energy consumption, as it has been for many decades. In line with projection for other energy sources, the strongest growth in electricity is expected to occur in non-OECD nations, driven by rising living standards. Figure 1.4 shows world net electricity generation from different energy sources over the 2012–2040 period. According to IEO2016, electricity generation increases by 69% from 21.6 in 2012 to 36.5 trillion kWh in 2040. As opposed to the entire energy sector, in the electric power one coal continues to be the most widely used fuel. However, renewables are the fastest-growing source of electricity generation – with hydropower being the predominant source – with an annual increase averaging

[1] It must be noted that the IEO2016 reference case assumes current trends and reflects the effects of current policies, and does not anticipate new policies that have not been announced or implemented yet.

2.9% from 2012 to 2040. After renewable energy sources, natural gas and nuclear power are the next fastest-growing sources of electricity generation at a pace of 2.7% and 2.4% respectively.

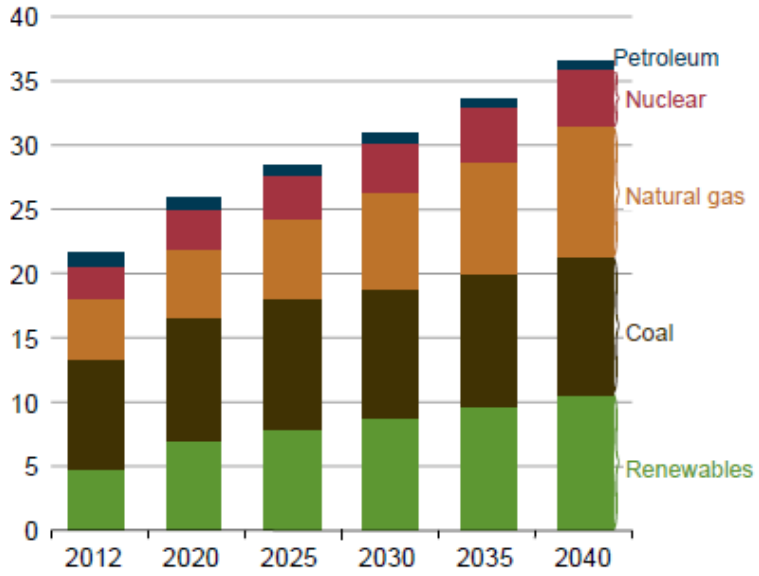

Figure 1.4 – World net electricity generation by energy source, 2012–2040 (trillion kWh) [19].

The projections of the US-EIA, along with those of other institutions such as the International Energy Agency (IEA), indicate that although fossil fuels demand in the energy and electric power sector is slowly but steadily decreasing, the energy market in 2040 will still be heavily reliant on conventional, carbon-intensive energy sources. The sector needs a rapid transition to alternative sources if the Paris Agreement goals are to be met.

1.3 Implementing the Paris Agreement

The Paris Agreement set a milestone in the path of humankind to reconcile its activities with planet Earth. The global consensus on limiting global warming to 2 °C and the aspiration to achieve 1.5 °C by 2100 is fully aligned with climate research [20], [21] and the scientific assessment for a safe operating space for humanity (discussed in Section 1.1). However, while the goal of the Paris Agreement is relatively clear on reaching net-zero emissions “by balancing anthropogenic emissions by sources and removal by sinks” [14], a quantitative emission pathway to achieve this goal is missing – and also, bizarrely, the words “fossil fuels” is never used. According to scientific assessments, nations will devour their remaining carbon budget rather soon [22]. A global roadmap for a rapid transition to a carbon-free society is thus imperative.

Figure 1.5 presents a global roadmap in line with the Paris Agreement, for a 50% chance of limiting warming by 1.5 °C and probability higher than 66% of meeting the 2 °C target, developed as an illustrative scenario by Rogelj and co-workers [23], [24]. The roadmap envisages that global CO_2 emissions peak no later than 2020 at ~40 metric gigatons, and then reduce to ~24 in 2030, ~14 in 2040 and ~5 in 2050. Rockstrom and co-workers went even further by proposing a simple, heuristic "carbon law" – in analogy with Moore's law for shrinking chip trend [25] – of halving gross anthropogenic carbon-dioxide emissions every decade [22]. They argue this would allow for a small, but crucial carbon budget contingency that could be used for risks of biosphere carbon feedback or delay in the deployment of CO_2 removal technologies. Notably, a CO_2 reduction trajectory applies to all sectors and countries at all scales; for instance, in the case of Rockstrom and colleagues' carbon law, it means doubling of zero-carbon shares in the energy systems every five to seven years - a rate consistent with the trajectory of the past decade[2].

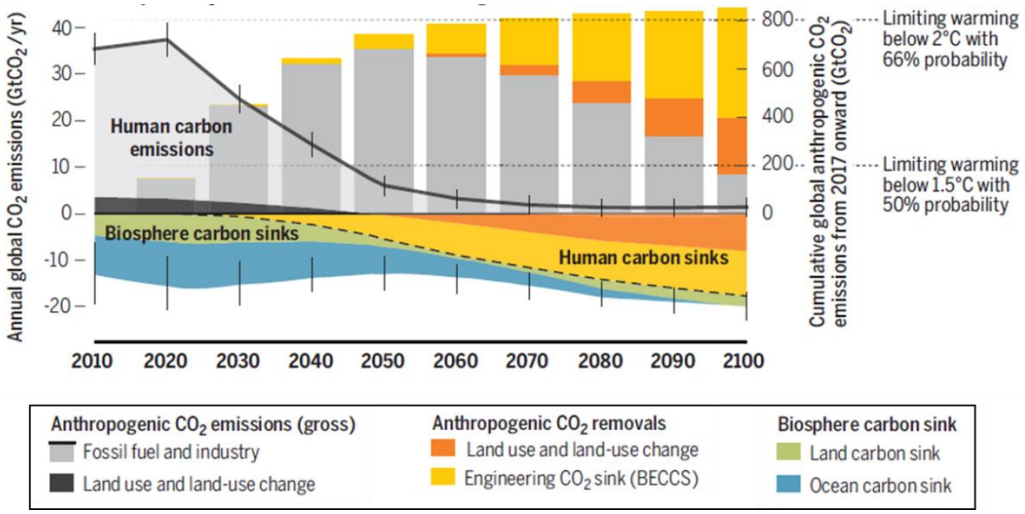

Figure 1.5 – Decarbonisation pathway consistent with the Paris Agreement [23], [24].

However, efforts to reduce CO_2 emissions are not enough on their own and should be complemented with scalable carbon removal and efforts to ramp down land-use CO_2 emissions. According to the global roadmap sketched by Rockstrom and co-workers [22], public and private investment in research and development for climate solutions should increase by an order of magnitude by 2030 and Bioenergy with Carbon Capture and Storage (BECCS) and Direct Air Carbon Capture and Storage (DACCS) should roll out by

[2] It must be noted that the IEO, Section 1.2, assumes a different, more conservative, future trend for renewable energies growth

2040 and exponentially increase until the end of the century. In parallel, research to ascertain the resilience of remaining biosphere carbon sinks should also be carried out.

The human enterprise is on a good track, but Herculean efforts must be taken until the end of the 21st century to limit planet warming and avoid irreversible effects that would put the earth on an unstable trajectory far from the stable Holocene-like state, within which our society was born. The following Section will briefly examine the climate policies of the European Union (EU) and the United Kingdom (UK), and how these compare with the Paris Agreement goal.

1.3.1 European Union

The European Union (EU) 2020 package, proposed in 2007 and enacted into legislation in 2009, set three ambitious climate and energy targets to be achieved by 2020; these are 20% reduction in Greenhouse Gas (GHG) emissions (from 1990 levels), 20% share of EU energy from renewables and 20% improvement in energy efficiency [26]. To ensure the Union would meet its goals, the European Commission (EC) took actions in several areas by implementing policies and initiatives and developing nation-specific guidance. For instance, the Emission Trading System (ETS) set a legal limit for greenhouse gas emissions from large scale facilities in the power, manufacturing and aviation sectors; the system covers all industries emitting approximately 45% of the EU's total greenhouse gas emissions. For those sectors not covered by the ETS (e.g. housing, agriculture, waste and transport excluding aviation), the EU established national emission target reductions based on national wealth, which were taken as binding annual targets by EU countries. In addition to this, the EU countries set binding annual targets to increase the share of renewables in their national energy consumption by 2020. Finally, the EU supports and finances research and development of low carbon technologies through several funding programmes, such as the well-known Horizon 2020 [26]. The EU has already met its first target and is heading towards 25% reduction in GHG emissions by 2020 [27], and is well on track to meet its second target of 20% renewables (in 2015 share of renewable energy stood at ~17% [28]); but it will not meet its energy efficiency target unless further efforts are made [29].

In 2011 the EU reconfirmed the 2020 targets and developed a roadmap that extended well above 2020 to 2050: the 2050 low-carbon economy roadmap set out the objective of a reduction of domestic GHG emissions by at least 80% (below 1990 levels) by 2050

and a cost-efficient pathway envisaging 40% and 60% intermediate targets to be achieved in 2030 and 2040. All industrial sectors, according to their technological and economic potential, are required to contribute if a transition to a low-carbon economy is to be achieved. Figure 1.6 shows an emission reduction pathway consistent with the 2050 roadmap, according to which the transport sector should achieve 60% reductions, energy intensive industries 80% and buildings 90%. However, the power generation and distribution sector has the biggest potential for cutting emissions: the sector must be virtually decarbonized (93-99%) by 2050 [27]. The chart also indicates that the 2050 target (of 80% GHG emission reduction) would be missed by nearly 40% with policies implemented up until 2011.

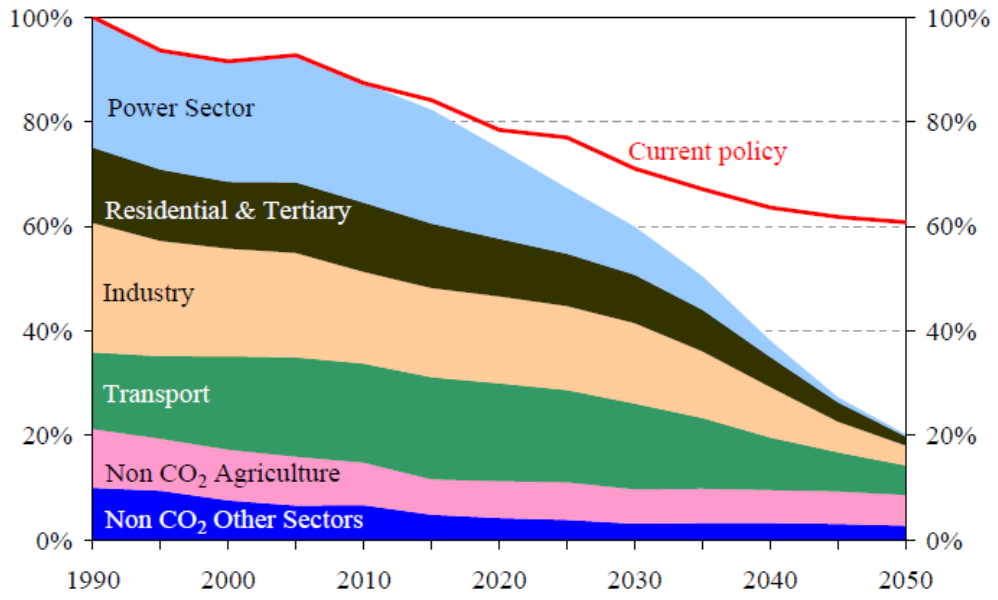

Figure 1.6 – GHG emission reductions by sector required to achieve the 80% target in 2050, and trend of current policies [27].

The transition of the energy system is further explored in the Energy Roadmap 2050 [30] where four routes towards a more sustainable, competitive and secure energy system are investigated: energy efficiency, renewable energy, nuclear energy, and carbon capture and storage; these are combined in different ways to create seven possible scenarios for 2050. Notably, the EU has acknowledged that nuclear energy will provide an important contribution to the energy transition, especially in those states where it is pursued, as it remains a key source of low carbon electricity generation. The highest contribution of nuclear would be required in the case of delayed deployment of Carbon Capture and

Storage (CCS) and of diversified supply technologies scenarios (18 and 15% respectively); interestingly, these are also the scenarios with the lowest total energy cost.

It must be noted that the EU's current climate targets are calibrated to limit global warming to 2°C, rather than 1.5°C: the 2050 target is based on the 2007 IPCC's 4th Assessment Report, which provided a range of 80 to 95% of emission reductions for developed countries by 2050 [31]. However, as the EU has agreed to pursue efforts to stay within 1.5 °C, the EU 2050 strategy and its intermediate targets must be revised; for instance, reductions far below 40% by 2030 would be required to meet the 1.5 °C target [32].

1.3.2 The United Kingdom

The UK policy on reducing GHG emissions was set by the Climate Change Act in 2008, which established a legally binding target of 80% (below 1990-1995[3] levels) to be achieved by 2050 [33]. As with the EU 2050 roadmap, the UK target is based on the IPCC's 4th Assessment Report to restrain global temperature increase to 2°C, which, as noted above, is not aligned with the Paris Agreement ambition to pursue efforts to remain within 1.5 °C. The Climate Change Act introduced a system of carbon budgets that provide legally binding limits on the amount of emissions that may be produced in successive five-year periods. These are designed to prepare the UK to meet its 2050 target in the most cost-effective way, as well as to track progress. There are five carbon budgets currently set in legislation: the first (23% reduction), covering the period 2008-12, was met successfully; the fifth carbon budget covers 2028-32 and is equivalent to a 57% reduction. The last Carbon Plan, published by the Government in 2011, sets the strategy for delivering the fourth carbon budget [34]. The UK is currently on track to outperform the second (2013-2017) and third (2018-2022) budgets, with emissions in 2016 being 38% below reference levels [35]. However, current policies are not enough to deliver the existing carbon budgets that Parliament has set: according to the UK Committee on Climate Change's (CCC) 2016 Progress Report, these would at best deliver around half of the emissions reductions required by 2030 [36]. Figure 1.7 shows the gap (in red) between current low-risk (i.e. with sufficient funding and addressing known barriers) and at-risk (that either

[3] The 1990 level includes carbon dioxide, nitrous oxide and methane emissions, whilst the 1995 one for hydrofluorocarbons, perfluorocarbons and sulphur hexafluoride.

lack sufficient funding or address unknown barriers) policies and the cost-effective path to meeting the legislated carbon budgets.

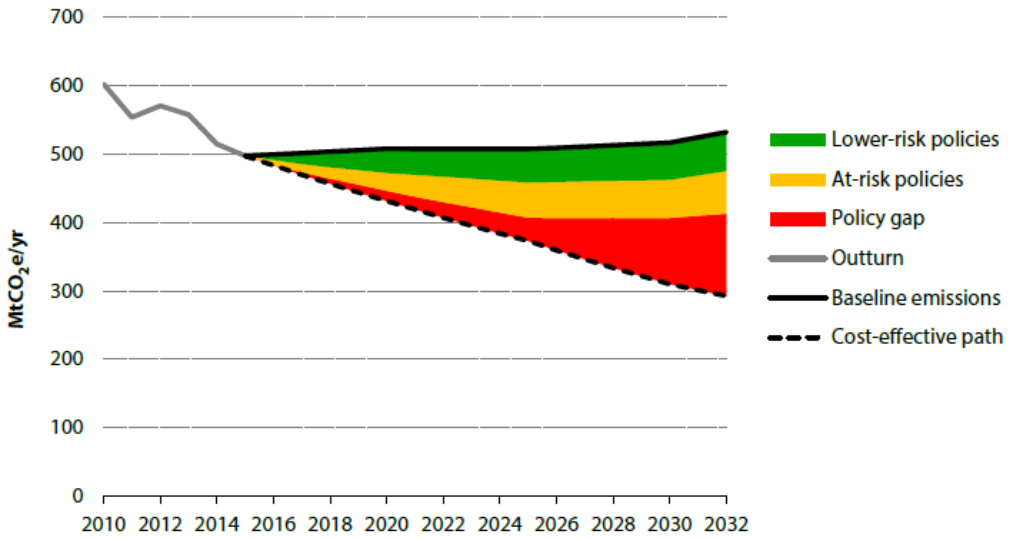

Figure 1.7 – Assessment of current policies against the cost-effective path to meet carbon budgets [36].

By considering a global carbon budget required to limit global warming to 2 °C, the UK target is representative of an allocation based on the inertia principle, that is the emission reduction are based on national emissions in a reference year. Another approach that has been proposed (amongst many others) applies effort sharing principles of equity [37], that is emission reductions are based on a per capita basis. For a developed country such as the UK, equity leads to significantly more stringent targets according to which the UK would fall even shorter of its 2050 target [38]. (Notably, the same could be said for the EU targets.) Although the UK target should be revised in line with the Paris Agreement and possibly for fairer allocation of global carbon budgets, the Committee on Climate Change (CCC) recommends that the current priority should be to take robust near-term action to close the gap to existing budgets; there will be several opportunities to revisit the UK's targets in future as low-carbon technologies and options for greenhouse gas removals are developed [39].

As already noted above, all industrial sectors are required to contribute if national targets are to be met, with the power and energy sector having the highest potential of becoming fully decarbonized by 2050. In the UK, this is projected to occur through exploitation of three main sources: renewable energy, particularly onshore and offshore wind farms; a new generation of nuclear power stations; and gas and coal-fired power stations fitted

with CCS technology. Fossil fuels without CCS are expected to be used as back-up electricity capacity at times of very high demand. The UK reference scenario for the 2050 power production mix developed by the Department of Energy and Climate Change (DECC) sees nuclear power having ~33% share of electricity production, with renewables sitting at ~45% and fossil fuel with CCS at ~22%. However, the DECC has developed several other scenarios: the most favourable for the nuclear industry sees its contribution up to 40-50% of the total UK electricity supply. This represents the most cautious scenario in the event that there are major hurdles in deployment of CCS and renewable energy technologies [34], [40].

1.4 Nuclear power in the UK

1.4.1 The energy trilemma and challenges of nuclear power

As noted above, the UK Government regards nuclear energy as a key contribute to a future low-carbon energy system. However, besides decarbonisation, the Government has two other priorities: keeping energy bills as low as possible for households and businesses, and having a secure and resilient energy system. These three objectives represent the so-called energy trilemma, because they are in tension one against the other (see Figure 1.8). Nuclear power contributes to all three objectives of the trilemma not only because it is a low-carbon technology, but because it also provides baseload electricity, irrespective of climatic conditions (security of supply); and it is comparable with other low-carbon technologies over the life time (affordability) [41].

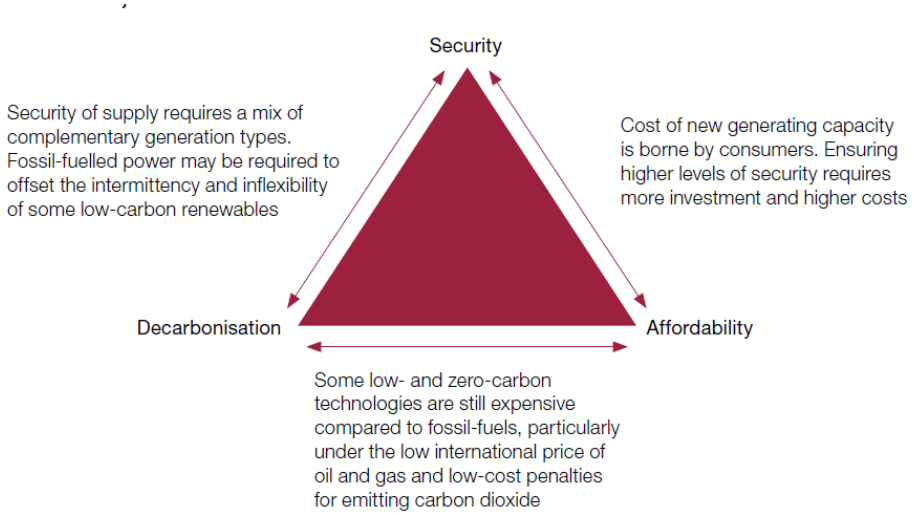

Figure 1.8 – The Department of Energy and Climate Change (DECC) trilemma [41]

At present, however, the development of nuclear power programmes presents a number of specific challenges; the most notable being the disposal of nuclear waste and the decommissioning of old nuclear plants:

- Some types of nuclear waste are very radioactive, and remain so for millennia. Currently, there is not an established technology for reducing their hazard and geological disposal represents the most favoured solution by the majority of nuclear countries.
- There is very little experience on decommissioning nuclear power stations, therefore related uncertainties and costs are significant, especially when compared to other low-carbon technologies.

Other challenges related to nuclear power are:

- Nuclear power plants have very high upfront costs, which have increased considerably in recent years given the required extra safety considerations after the Fukushima disaster, and take a very long time to become operational. Therefore, investments in nuclear power are exposed to external risks, including changes in Government policy and market fluctuations, such as falls in the wholesale price of electricity. However, the high upfront costs are offset to some extent by low running costs that are not linked to the price of the fuel in the way that fossil fuel-based power plants are.
- Nuclear power plants are ideal for providing baseload electricity, but are rather inflexible, i.e. they cannot change their power output as easily and cost effectively as renewable and fossil-based plants.

Finally, there are also some challenges that are specific to the UK:

- Because no new nuclear power plants have been built in the UK in the past 20 years, the country lacks a proven and skilled supply chain to support construction thus making the construction of first-of-a-kind power stations considerably more expensive.
- The fact that the UK energy sector has been fully privatised in the UK means that private companies have to take the whole upfront costs of building nuclear plants including decommissioning and waste management, and there are very few of those able to risk such large upfront investments.

1.4.2 Development of the nuclear industry

The UK was pre-eminent in the development of nuclear energy during the 1940s and 50s. Following the Second World War, nuclear research in the UK – and elsewhere – was solely focused on defence, military-related applications: the country's first large nuclear reactors – the Windscale piles commissioned in 1947 at the Sellafield site – had the purpose of breeding plutonium, the key component of nuclear weapons. However, soon it became obvious that nuclear reactors could also be used to generate electricity: in 1953 the construction of the first commercial nuclear power reactors at Calder Hall on the Windscale site (currently known as Sellafield site) commenced following the announcement by the Government that the country would begin a civil nuclear power program ("A Programme of nuclear power" white paper published in 1955), and continued in 1956 with the opening by Queen Elizabeth II of the world's first commercial nuclear power station, based on the British-designed Magnox reactors. These reactors were initially dual-purpose, combining power generation with plutonium production; however, the latter function was confined to other facilities from mid-1960s. The success of the first reactor brought enthusiasm to the industry, and Magnox units were scaled up tenfold and optimised for electricity production. The Suez crisis in 1956 raised questions about the reliability of fossil fuel supplies from the Middle East and contributed to further expansion of nuclear power in the UK. A total of 26 Magnox reactors were built from 1956 to 1971, with a total capacity of approximately 4200 MWe net. The design was also exported to Japan and Italy, and very similar reactors were built in France. "The Second Nuclear Power Programme" in the UK was announced in 1964, with new nuclear plants expected to be commissioned in 1970s. Following much debate over the choice of the design, the Advanced Gas-Cooled Reactor (AGR), also designed in the UK, was adopted as the standard. The construction of AGR plants unexpectedly presented numerous technical and financial problems, but eventually seven twin-unit stations were built around the country, the first of which started operation in 1976 at Hinkley Point B. The total capacity of the AGR units was double that of Magnox, totalling approximately 8800 MWe net. The problems related with the construction of AGR units led to a third nuclear power programme and a renewed debate over the choice of reactor design: in 1987 the construction of a Pressurised Water Reactor (PWR, the first UK reactor to be built that was not designed in the UK) started and became operational in 1994. Further PWRs were planned, but a review of nuclear policy by the Government in 1995 concluded that nuclear

plants were not economically viable and they would thus not receive public sector support.

1.4.3 Present and future of the nuclear industry

In 1990s nuclear power contributed around 25% of total annual electricity generation in the UK, but this has gradually declined with retirement of life expired Magnox plants: in 2016 nuclear energy contribution totalled approximately 21% [42] provided by 15 operating reactors (see Table 1.1). The last operating Magnox reactor – Wylfa 1 – shut down in December 2015, leaving the UK fleet made up by the seven twin-unit AGR stations and one PWR reactor, all of which are owned and operated by EDF Energy. Furthermore, the operating life of most of these reactors has already been extended: half of the UK capacity is expected to be retired by 2025 and all the AGR reactors by 2030, leaving the sole PWR to operate until 2035 [43]. This poses a significant problem for the UK to meet its carbon budgets and to guarantee the security of supply.

Table 1.1 – Power reactors currently in operation [43]

Plant	Type	Present capacity (MWe net)	First power	Expected shutdown
Dungeness B 1&2	AGR	2 x 520	1983 & 1985	2028
Hartlepool 1&2	AGR	595, 585	1983 & 1984	2024
Heysham I 1&2	AGR	580, 575	1983 & 1984	2024
Heysham II 1&2	AGR	2 x 610	1988	2030
Hinkley Point B 1&2	AGR	475, 470	1976	2023
Hunterston B 1&2	AGR	475, 485	1976 & 1977	2023
Torness 1&2	AGR	590, 595	1988 & 1989	2030
Sizewell B	PWR	1198	1995	2035
Total: 15 units		**8883 MWe**		

The decision to halt the expansion of nuclear agreed in the 1990s was effectively reversed in 2006 following another review of energy policy carried out by the Government. Since then, the Government has been supportive of new nuclear plants that, in line with the policy of market liberalisation, should be financed and built by the private sector with internalised costs of waste management and disposal and plants decommissioning [44]. There has been substantial international interest in the UK's 21st century nuclear programme; Table 1.2 includes nuclear stations that are planned or have been proposed to be built in the UK. Amongst these, EDF Energy's Hinkley Point C is currently the most

advanced project, after the final decision in the 2016 by EDF to go forward with the £18 billion project. EDF Energy also plan to build two further reactors at Sizewell. For both Hinkley Point C and Sizewell EDF will use the European Pressurised Reactor (EPR) design. Further projects include Horizon (a 50:50 joint venture of RWE power and E.ON UK) to build two twin-stations at Wylfa and Oldbury respectively, using the Advanced Boiling Water Reactor (ABWR) design; and NuGeneration, owned by Toshiba and expected to be fully acquired by Korea Electric Power Corporation (Kepco), to build three twin-stations at Moorside using the Kepco AP1000 design. These projects amount to a planned new generating capacity of 15 GWe gross. Finally, two further plants have been proposed by General Nuclear Systems, a joint venture by China General Nuclear and EDF, to be built at Bradwell B, using the Chinese Hualong One reactor design with a gross capacity of 2.3 GWe. The planned and proposed nuclear plants together represent an additional total gross capacity of 18 GWe, which would replace the retiring AGR and PWR plants and increase the contribution of nuclear energy to the total UK electricity production.

Table 1.2 – Nuclear plants planned and proposed to be built in the UK [43]

Proponent	Site	Locality	Type	Capacity (MWe gross)
EDF Energy	Hinkley Point C1&2	Somerset	EPR	2x 1670
	Sizewell C1&2	Suffolk	EPR	2x 1670
Horizon	Wylfa Newydd 1&2	Wales	ABWR	2x 1380
	Oldbury B1&2	Gloucestershire	ABWR	2x 1380
NuGeneration	Moorside 1&2&3	Cumbria	AP1000	3x 1135
Total planned				**15605 MWe**
General Nuclear Systems	Bradwell B1&2	Essex	Hualong One	2x 1150
Total proposed				**2300 MWe**

Besides conventional power reactors, the UK in its 2015 program to “revive the UK’s nuclear expertise” expressed its interest towards the development of small modular reactors (SMRs) – which are essentially smaller reactors that can be manufactured centrally at a plant and brought on site fully constructed as completed modules. Several companies have shown interests in deploying SMRs, including Nuscale, Westinghouse, Rolls-Royce and Ge-Hitachi. Notably, GE-Hitachi’s PRISM reactor appears particularly appealing to the UK: it is a fast reactor designed with the purpose of burning reactor-

grade plutonium and the UK has one of the largest stockpiles of such plutonium in the world.

1.5 The nuclear fuel cycle

This Section describes the various stages associated with production of electricity from nuclear fuel – collectively known as the nuclear fuel cycle. This is not intended to be an exhaustive introduction to the topic; on the contrary, it aims to provide just the basic information required to grasp the research undertaken in this Thesis. The nuclear fuel cycle starts with mining of uranium ore – or any other ore containing fissionable material, such as thorium – and ends with disposal of Used Nuclear Fuels (UNFs) or High Level Waste (HLW), depending on whether UNFs are reprocessed.

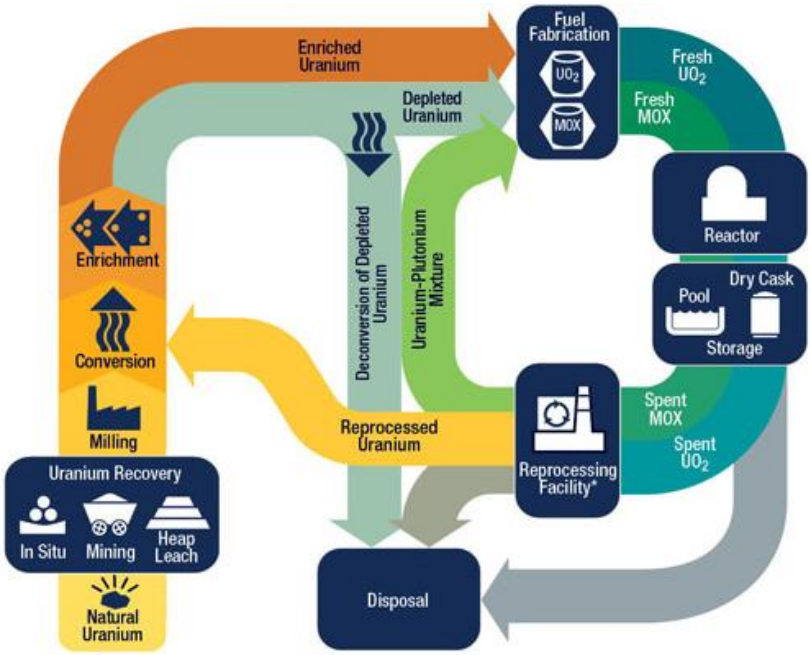

Figure 1.9 – Stages of the nuclear fuel cycle (adapted from [45]).

The front-end of the cycle includes all the stages that aim to prepare uranium and manufacture fuel assemblies to be loaded into nuclear power reactors; whereas the back-end consists of all the stages required to safely manage, contain and either reprocess or dispose of the nuclear fuels after being removed from the nuclear power reactors. A schematic illustration of the nuclear fuel cycle is reported in Figure 1.9. The next Sections include a brief introduction to the element uranium and a description of each of the main steps of the nuclear fuel cycle.

1.5.1 Uranium

Uranium is a slightly radioactive metal (most of its radioactivity is in fact associated with its decay products) that occurs throughout the earth's crust in six isotopes. uranium-238 (U238) is the most abundant (~99.7%) isotope, but uranium-235 (U235), which is significantly less abundant (~0.7%), is at present the most valuable because it is the only isotope that undergoes fission reaction in current commercial power reactors (i.e. thermal reactors, discussed in Section 1.5.6).

Uranium is as common as tin and zinc in the Earth's crust, and about 500 times more abundant than gold; and can be found in the majority of rocks and soils, but also dissolved in fresh and sea water. However, for the extraction process to be economically feasible, uranium has to be found in relatively high concentrations – from 100 ppm found in very low grade deposits (e.g. Namibia) to 200,000 ppm in very high grade ones (e.g. Canada); a mineral deposit with such characteristics is often defined as the orebody. At present, neither uranium dissolved in water nor granites are considered orebodies due to the low concentration of uranium present, but they could become so if uranium price were to rise sufficiently.

Around 51% of known resources (i.e. those that are reasonably assured plus inferred sources) of uranium are concentrated in just three countries, which in 2016 produced more than two thirds (~71%) of total world supply: Australia has the largest world reserves (~29%) but produces only a tenth of total supply; Kazakhstan on the other hand is the largest producer (~33%) despite having smaller reserves (~13%); while Canada is the second largest producer (~22%) and has the third largest reserves [46], [47].

1.5.2 Uranium mining and milling

Mining is the process of extracting valuable ores from the ground; generally speaking, uranium mining is no different from other kinds of mining unless the ore is very high grade. There are four main methods of extraction whose choice depends on the depth of mineralisation and the ore grade:

- Open-pit mines are used for near-surface deposits and involve a large excavation hole and the removal of much overburden (overlying rock) as well as a lot of waste rock. The Ranger Mine in Australia, Rossing in Namibia and most of the mines in Canada's Northern Saskatchewan are open pit mines.

- Underground mines are used for deep deposits that cannot be accessed through an open pit. They require construction of access shafts and tunnels, but produce much less waste rock and usually have lower environmental impacts than open-pit mines. The Olympic Dam in Australia and some of the most productive mines in Canada, such as the McArthur River, Rabbit Lake and Cigar Lake in Canada are underground mines.
- In-Situ Leaching (ISL) mines are used when the orebodies lie in a confined groundwater environment in porous unconsolidated material. The uranium is extracted by dissolution in acidified or alkaline groundwater enriched with oxygen which is pumped and extracted by means of wells. The leaching solution is then treated in a surface plant where the uranium is recovered as a precipitate. This technique causes no major ground disturbance and has the least environmental impacts; however, it cannot be applied in the case it may threaten potable water supplies. All Kazakhstan mines use the ISL technique.
- Heap Leaching mines are used for very low-grade (below 0.1%) orebodies. The broken ore is stacked about 5-30 metres high on an impermeable pad and irrigated with acid solution over many weeks; the pregnant liquor is collected and treated to recover uranium as done for ISL.

Conventional mining methods, that is open pit and underground, require a mill plant to crush and grind the ore to free the mineral particles, which are then leached with sulphuric acid (as done for ISL and heap leaching mines) or with a mixture of sodium carbonate and bicarbonate to dissolve the uranium oxide. Sometimes a physical beneficiation process is used to concentrate the ore before the chemical treatment. The uranium in solution (either from the mill plant or from heap leaching/ISL) is recovered using either resin/polymer or liquid ion exchange, stripped and then precipitated, in the form of ammonium diuranate. This is dewatered and roasted to produce U_3O_8, the form in which uranium is marketed and is known as yellowcake.

1.5.2.1 Tailings management

Uranium tailings are waste by-products from conventional mining methods and milling. As noted above, both open pit and underground mines generate a significant amount of overlying and waste rock. However, milling operations also produce considerable amount of tailings: approximately 99% of the unprocessed ore mass is discarded as waste; this

contains about 85% of the radioactivity originally present in the ore (due to the presence of other radioactive materials such as thorium-230 and radium-226) as well as numerous biotoxic substances [48]. Tailings from milling represent a substantial hazard to public health and the environment mainly due to potential contamination of groundwater, soil and sediments, gaseous emissions of radium and radon (which are produced from decay of uranium) and failures of the storage system. Earth-fill impoundments known as tailings dams are used to store waste by-products from milling, which are usually covered by water to reduce radioactive gaseous emissions. On completion of mining operations, tailings are usually covered by clay or topsoil to resist erosion and allow the growth of vegetation; sometimes they may be returned to the open-pit or underground mine.

1.5.3 Conversion

Regardless of the type of fuel to be manufactured and the design of the nuclear reactor, the yellowcake has to be converted into volatile uranium hexafluoride – a gaseous form needed for enrichment; usually, at the same facility the yellowcake is also purified from some remaining impurities that are detrimental to its final use. These are neutron absorbers such as boron and cadmium; elements forming volatile fluorides, such as molybdenum, chromium and vanadium, that could contaminate gaseous uranium hexafluoride produced at the end of the conversion process; and other elements with similar chemical properties to uranium, e.g. thorium. Although both a dry and a wet process have been developed, the majority of commercial refiners today – including the one at Springfields, UK [48] – employ the latter, which involves the yellowcake being dissolved in nitric acid: insoluble elements are removed by filtration and soluble ones by solvent extraction (usually tri-butyl phosphate in kerosene). To produce uranium in a gaseous form, the purified uranyl nitrate solution is first evaporated and denitrated to produce uranium trioxide (UO_3) powder, and reduced to uranium dioxide (UO_2) in a kiln. Then, UO_2 is reacted with hydrogen fluoride (HF) in another kiln reactor to produce uranium tetrafluoride (UF_4), which by reaction with elemental fluorine in a fluidised bed reactor is converted into uranium hexafluoride (UF_6).

1.5.4 Enrichment

Uranium enrichment is the process in which the proportion of the fissile isotope U235 is increased from its natural content by physical separation of the U235 from the U238. Today the majority of nuclear reactors require concentration of U235 in the fuel to be

around 3-5%; the British Magnox and the Canadian CANDU are amongst the few reactor designs using non-enriched fuel. The enrichment process requires uranium in the form of gaseous UF_6 (above 60 °C) and exploit the difference in mass between U235 and U238 (the former is about one percent lighter than the latter) to produce two streams, each containing higher concentration (than that can be found in nature) of one of the two radionuclides; the by-product stream containing higher concentration of U238 is usually referred to as “enrichment tails” or simply “tails”. Uranium enrichment is a very energy-intensive process: plant capacity is measured in Separative Work Units (SWU or kgSW), a complex unit that indicates the energy input relative to the amount of uranium processed, the degree to which it is enriched and the level of depletion of the tails.

Several enrichment processes have been developed and demonstrated historically, but only two have been operated on a commercial scale. The diffusion process is known as first generation enrichment technology and it is no longer in use. The centrifuge process – referred to as second generation – is the current standard in the enrichment industry because it is considerably more efficient than diffusion. (In the centrifuge process the separation effect is proportional to the absolute mass difference rather than the square root of the relative difference.) The process involves a series of many hundreds of centrifuges under vacuum each containing a rotor spinning at 50000-70000 rpm; the heavier molecules of UF_6 (mainly U238) concentrates towards the cylinder’s outer edge and lighter molecules of UF_6 (U235) towards the centre; usually several hundred stages are needed to obtain the desired enrichment level. A third generation of enrichment technology is represented by laser technology. It promises lower energy inputs, lower capital costs and lower tails assays, but it is yet to be tested on a commercial scale.

1.5.5 Nuclear fuel fabrication

Fabrication of nuclear fuel structures represents the last stage of the front-end of the nuclear cycle (Figure 1.9). Essentially fuel assemblies consist of two components: fissile material – generally uranium, sometimes complemented with plutonium – and a metal cladding to protect the fissile material from the external environment and to prevent fission products from escaping. Because during the fission process a large amount of energy is released, the cladding is fundamental in holding the fissile material in a robust physical form capable of withstanding high operating temperature and pressures, as well as an intense neutron radiation environment.

1.5.5.1 Fuel manufacturing

Nuclear fuels can be distinguished depending on the chemical compound in which uranium has been produced (metal uranium or uranium oxide) and on the level of enrichment. The majority of nuclear reactors today require enriched uranium in an oxide state.

Metal fuels have been used in the first generation of British power reactors, known as Magnox, and in few other reactor designs. Magnox fuel comprises an alloy of uranium metal of natural content with traces of iron and aluminium, contained in a magnesium-based cladding alloy (from which the name Magnox), which was developed specifically to resist the oxidising effects of carbon dioxide coolant in the reactor. The Magnox design has two major limitations in terms of operating conditions that substantially reduced its thermodynamic efficiency. First, the cladding must be kept at temperature below 450°C to avoid excessive oxidation; and second, at ~660 °C uranium undergoes a phase change from α to β accompanied by a significant expansion that could cause excessive stress to the fuel rods. As noted in Section 1.4.3, there are currently no operating Magnox reactors after the shutdown of Wylfa 1 in 2015.

The use of oxide fuels and zirconium alloy or stainless steel cladding allows higher temperature (and thus higher efficiency) to be reached inside the reactors. Enriched oxide fuels are used in the majority of current nuclear reactor designs with exception of the Canadian design (CANDU) that uses oxide fuel with natural or slightly enriched content of U235. Fabrication of oxide fuel starts from UF_6 or UO_3 depending on whether uranium has been enriched or not; in both cases uranium is converted to uranium dioxide (UO_2) prior to pellet fabrication. Deconversion of UF_6 to UO_2 is usually achieved in a two-step process that involves UF_6 to react with steam in autoclaves (to produce uranyl fluoride, UO_2F_2) and then with hydrogen at high temperatures. To obtain UO_2 from UO_3, water is added to uranium trioxide to form a hydrate which is then fed to a kiln operating in a reducing atmosphere. All commercial fuel fabricators use uniaxial dry pressing to form UO_2 powder into the required pellet shape; the pellet is then sintered, converting the UO_2 from green compressed powder to a true ceramic one capable of enduring the high operating conditions of nuclear reactors; and finally, the pellet is finished: the diameter is checked and the sides ground to produce parallel-sided pellet with a tolerance of few microns.

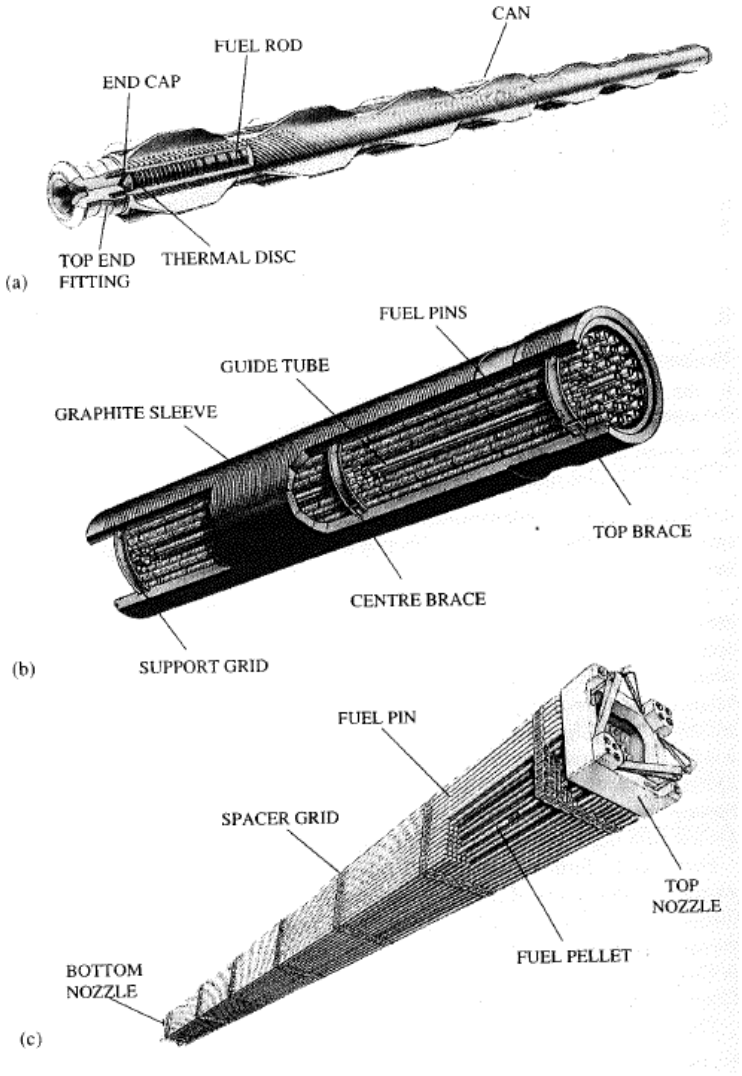

Figure 1.10 – Fuel assembly designs. a) Magnox; b) AGR; c) BWR. [48]

The UO_2 pellets are then assembled into pins, inserted into a tubular metal cladding, and hold in groups by a strong framework made of steel and zirconium, known as fuel assembly. The tubular metal cladding is made of stainless steel in Advanced Gas-cooled Reactors (AGR) and Zircaloy in Pressurised Water Reactor (PWR), Boiling Water Reactors (BWRs), Pressurised Heavy Water Reactors (PWHR, known as CANDU) and water-cooled graphite moderated reactors (RMBK); the fuel assemblies are cylindrical for AGR and CANDU, and have a square cross-section for PWR, BWR, and RBMK (see Figure 1.10 for a schematic representation of some fuel assembly designs). They support 36 pins in the AGR design up to 289 (arranged in a 17 x 17 array) in the BWR one. Fuel assemblies are made of support and central bracing grids to allow easy insertion of single fuel pins and depending on the reactor design also control (PWR) and water (BWR) rods.

1.5.6 Power reactors

The central element of the nuclear fuel cycle is the nuclear reactor; this is where thermal energy is produced and converted into electricity. Nuclear energy can be produced by two different reactions: nuclear fission involves the nucleus of an atom to split into smaller parts, whereas nuclear fusion occurs when two or more atomic nuclei come close enough to form one or more different atomic nuclei and subatomic particles. In both cases the energy released is proportional to the difference in mass between reactants and products. Thus far, only fission has been used for energy production; fusion is significantly more advantageous – especially with respect to nuclear waste produced – but we are far from being able to produce meaningful quantities of energy from it.

Nuclear reactors are classified into thermal or fast reactors according to the kinetic energy of the neutrons sustaining the fission reaction. Atoms undergoing fission release fast neutrons (with energies around 5 MeV); in thermal reactors, these are slowed down to reach thermal equilibrium with the medium by means of a neutron moderator. The most commonly used moderator in modern power reactors is ordinary water; other used moderators are graphite as in the British gas-cooled reactors and heavy water (D_2O) as in the Canadian CANDU reactors. Both graphite and heavy water are superior as moderators to the extent of absorbing fewer neutrons for every one slowed down neutrons, and are therefore preferred for use with low-enriched or natural uranium fuels.

The number of nuclear reactor designs that have been conceived since the first experimental pile in Chicago in 1942 is extraordinary but only few of them have been operated on a commercial scale. Four generations of reactors are commonly distinguished. Generation I reactors were developed in the 1950-60s and are no longer in operation after the shutdown of Wylfa 1 in 2015; they mostly used natural uranium fuel and graphite as moderator. Most of existing reactors belong to Generation II; these use enriched uranium fuel, and are mostly cooled and moderated by water. Generation III includes advanced reactors that are the development of Generation II with enhanced safety, the first of which are in operation in Japan, and from early 2018 in China and the UAE, with many others under construction or about to be ordered. Finally, Generation IV includes reactors whose design is still under development and will not be operational before the mid-2020s. Some of these will be capable of burning long-lived actinides (which are now part of the nuclear waste), breeding fissile material and producing hydrogen

(alongside electricity). Of seven designs under development, four or five will be fast neutron reactors.

There are currently over 400 commercial power reactors in the world [49], using five reactor designs: PWR, BWR, gas reactors including Magnox and AGR, PWHR and RBMK. They are primarily distinguished by the geometry of the reactor and the type of moderator used.

Gas reactors have been largely developed by the UK; they are graphite moderated reactors cooled by pressurised carbon dioxide. The first generation, Magnox, uses non-enriched metal uranium, whereas the second generation, the Advanced Gas-cooled Reactor (AGR), uses uranium oxide fuel approximately 3% enriched. The most distinctive characteristic of gas-graphite reactors is the bulk of the core: the fuel sits in individual channels through the moderator (which in some cases allow the reactor to be refuelled on power) while carbon dioxide is driven by large blowers through the core from the bottom to top and into heat exchanger to generate steam. The large size of the core makes the capital cost very high, but does have some advantages in safety as any fault heating up the core tends to proceed very slowly. Both Magnox and AGR reactors rely on control rods for fine tuning during operations and for shutdown; they are designed to fall into the core under gravity in case of drive mechanism power loss. AGR reactors also incorporate a system for emergency shutdown by injecting nitrogen – a very strong neutron absorber.

PWRs are water-moderated reactors consisting of a compact core in a pressure vessel. The heat absorbed by the pressurised water is used in a steam generator for electricity production. The neutron flux is monitored by neutron detectors centred-placed in the core and is controlled by a neutron absorbing materials, such as boron or gadolinium incorporated in the fuel assemblies or in free-standing rods, and boric acid (H_3BO_3) dissolved in the water. BWRs have much in common with PWRs: they also are water-moderated and water-cooled and have approximately the same reactor size; however, they feature a lower pressure in the vessel that allows the water to boil, thus eliminating the need for steam generators as the steam is directly used to produce electricity. The neutron flux is controlled by neutron-absorbing rods inserted from the bottom and gadolinium oxide (Gd_2O_3) incorporated into the fuel pellets; notably, boric acid (H_3BO_3) is impracticable to use as the moderator changes phase from water to steam. The Canadian

reactors design, PHWR, represent the only commercialised power reactors using heavy water (D_2O), which, as noted above, is a superior moderator than normal water, thus allowing use of non-enriched uranium. Furthermore, because of heavy water, PHWR require steam generators to produce electricity, in common with PWR. The neutron flux is controlled by neutron-absorbing rods and gadolinium nitrate [$Gd(NO_3)_3$] dissolved in the heavy water. The RBMK is a Soviet Union design very similar to the British AGR design, but cooled by light water instead of carbon dioxide; its main feature is the modular design that can be tailored to requirements by extending the size of the core and the number of pressure tubes as necessary.

1.5.7 Storage and transport of irradiated fuel

After a time span that depends on the reactor design and fuel burnup (usually around three years), the nuclear fuel becomes unable to sustain a nuclear reaction; it is thus removed and replaced with fresh fuel. The irradiated nuclear fuel – also known as Used Nuclear Fuel, UNF, or less appropriately as Spent Nuclear Fuel, SNF – represents a significant liability for nuclear operators that requires to be temporarily stored regardless of whether the fuel is to be directly disposed of or reprocessed. Interim storage allows to reduce radioactivity and heat generated by the irradiation fuel while protecting both operators and the public. This is required before transportation of irradiated fuels whether that is to a storage facility, to a final repository, or for fuel reprocessing purposes. Nuclear power stations are usually designed with limited internal storage sufficient only for their short-term requirements; larger, usually centralised, facilities are required for long term storage. An example of a pond at Sellafield site can be seen in Figure 1.11.

Interim storage, either at plant or in centralised stores, can occur either in water ponds (wet storage) or in dry casks (dry storage). The former are commonly known as cooling-ponds because they allow the fuel to cool ready for easier transportation, and are the preferred storage solution of nuclear operators. Water at least 2.5 metres deep is both effective shield and a good heat-transfer medium, besides allowing easy manipulation and clear visibility of the fuel. The water, usually demineralised, must be of high quality, and controlled to the desired pH. Corrosion inhibitors may be added, and for external ponds also a biocide to prevent the growth of algae that would impair visibility. Heat dispersion is obtained by natural convection (with fuel of relatively low radioactivity) or incorporated heat-exchangers (for larger stocks or more highly-irradiated fuel). Radiation

from a cooling pond may be due not only to the fuel contained within it, but (particularly in older facilities) also to radioactive contaminants dissolved or suspended in the shielding water: notable examples are cobalt-60 (Co60) from steel cladding and caesium-137 (Cs137) from the fuel in case of failure of the cladding. For this reason, the water must be continuously purified by filtration and ion exchange, or purged with a clean supply. Although wet storage is a well proven technology, water is a corrosive medium and can corrode some fuel cladding; this is particularly true for the magnesium alloy (Magnox) cladding. Furthermore, the hydrostatic pressure of the water places considerable stresses on the fuel assemblies. Both problems are avoided or mitigated by dry storage, which in turn requires more elaborate arrangements to shield, cool, and handle UNFs.

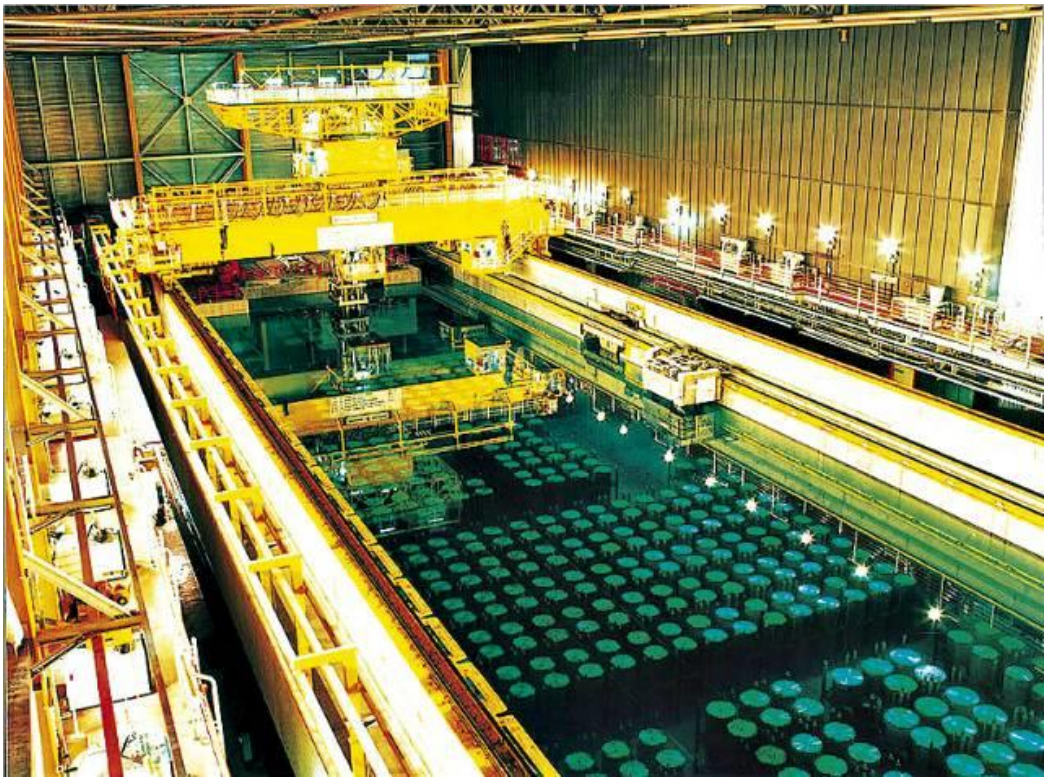

Figure 1.11 – Cooling pond of the Thermal Oxide Reprocessing Plant (THORP) at Sellafield site [50].

Transportation of irradiated fuel must satisfy stringent safety precautions, particularly as the transport infrastructure is designed to link centres of population and may make passage through them unavoidable. Carriage may be by road, rail or sea (often in combination), within or across national boundaries; the former is usually limited to the distance between reactor and railhead or port. Flasks (or casks) are the typical transport system; these consist of heavily-shielded containers and, like interim stores, can be

divided into wet or dry categories depending on whether the internal heat-transfer medium is water or a gas.

1.5.8 Reprocessing of irradiated fuel

A distinctive aspect of nuclear energy is that used fuels can be reprocessed to recover unused, fissile material from which new fuel can be manufactured. Typically, about 96% of initial uranium (U) is contained in UNFs, whilst 1% is converted to plutonium (Pu) and 3% to Fission Products (FPs). Reprocessing recovers the residual U and Pu and produces a highly radioactive waste form containing FPs that is suitable for disposal. This not only obtains some 30% more energy from the original uranium source, but also reduces the amount of radioactivity and volume of wastes to be disposed of. However, moving to fourth generation fast neutron reactors would change the outlook dramatically: firstly, because not only used fuel but also Depleted Uranium (DepU) from enrichment plants becomes a fuel source; and secondly, because also long-lived actinides could be burnt in fast neutron reactors, therefore increasing the interest in their recovery alongside uranium and plutonium. Generation IV reactors drastically reduce the demand for uranium mining thus achieving an almost complete closure of the fuel cycle, from which the name “closed cycle” is derived. When only uranium and plutonium are recovered and MOX fuel is produced, the fuel cycle is referred to as “partially closed cycle” or “Twice Through Cycle”, because usually MOX cannot be reprocessed and thus the fuel effectively passes two times through the reactor. By contrast, if used fuels are considered a waste and directly disposed the nuclear fuel cycle is referred to as an “Open Cycle” or “Once Through Cycle”.

Conceptually, reprocessing used fuels is the same as processing the concentrate of any metal mineral: here the ore is hard ceramic uranium oxide containing an array of other elements, including FPs and actinides. PUREX (Plutonium Uranium Redox EXtraction) is the *de facto* standard process for recovery of U and Pu from UNFs. The process involves dissolving fuel elements in concentrated nitric acid, so that they are separated from the cladding, followed by chemical separation of U, Pu and FPs in a two-step solvent extraction process. The first separation is achieved by a countercurrent solvent extraction process used in pulsed columns using tributyl phosphate dissolved in kerosene or dodecane; U and Pu enter the organic phase whilst FPs and other elements remain in the aqueous solution. In the second step, plutonium and uranium are separated by reduction

of Pu to a less extractable form: Pu is transferred to the aqueous phase with U remaining in the organic one. Further steps in the process involve purification and finishing of the Pu and U streams. The former aims at removing radioactive contaminants from the streams: for instance, in the case of uranium the objective is the removal of alpha emitters (Pu and Neptunium, Np) and FPs such as ruthenium-106 (Ru106) and technetium-99 (Tc99). In the latter, the separated streams are converted by means of either thermal denitration or precipitation into oxide forms, namely uranium trioxide (UO_3) and plutonium dioxide (PuO_2), which represent the feed products for manufacture of new fuel. Further to PUREX, other processes have been developed, but not yet operationalised. These aim primarily at separating long-lived actinides (for the reasons noted above) and increasing the proliferation resistance of the fuel cycle by avoiding production of a pure plutonium stream.

There are two main approaches for reusing the recovered uranium (RepU) and Pu from UNFs in existing commercial power reactors. Plutonium cannot be used alone as fuel in thermal reactors due to its high reactivity, and it is thus diluted with RepU or DepU to simulate as far as possible the nuclear properties of enriched uranium fuel. The blended product is named Mixed Oxide fuel (MOx) and can be used in all thermal reactors in perfect substitution of enriched uranium. The production process for fuel assemblies manufacturing is very similar to that of natural uranium. Concentrations of Pu range from about 1% up to 10% depending on plutonium properties and reactor design. Reprocessed uranium, by contrast, can also be used alone: it can either be re-enriched for use in water or gas-cooled reactors or diluted with DepU for use in PHWRs reactors. Notably, the concentration of U235 in RepU is lower than that of the starting enriched fuel but higher than that of natural uranium, with an usual residual level in the range of 0.4–1 1% [48]. This also implies that less work is required to bring RepU back to the starting enrichment level. However, it must be noted that RepU also contains other isotopes of uranium, which have been produced during irradiation in nuclear reactors; these are uranium-232 (U232), which decays with a 70-year half-life to yield strongly gamma-emitting daughter products; uranium-234 (U234), which gives rise to similar undesirable levels of activity; and finally uranium-236 (U236), which is not radiologically troublesome but is a strong neutron absorber (which has to be balanced by a higher concentration of U235). Like Pu, further processes required to manufacture the final fuel assembly are similar to those of natural uranium, except for further shielding required by operators due to the presence of U232.

1.5.9 Waste management and disposal

In common with any other industry, the nuclear industry generates several waste streams that are to be managed and disposed of according to the hazards they pose. Interestingly, nuclear is a very concentrated source of energy (i.e. large amount of energy from a small amount of fuel) that compared with other power generating industries generates very small amount of waste. However, nuclear wastes are radioactive and, like hazardous materials, require specific treatment and disposal; but unlike typical hazardous materials, their radioactivity (i.e. their main hazard) diminishes over time. Radioactive wastes are not peculiar to the nuclear fuel cycle; they are also a by-product of medicine, agriculture, research, and minerals exploration

The term nuclear waste usually refers to the solid part; however, also liquid and gaseous discharges may be regarded as wastes. Strict regulations exist on direct discharge of these radioactive streams – which in most cases are also treated to reduce their radioactivity, generating additional solid waste – with usually a maximum allowed level of radioactivity or concentration of radionuclides set by the regulatory organisations. Nuclear facilities are required to evaluate the radiological impacts of their discharges by performing site-specific assessment (discussed in Chapter 3): a notable example is the annual report published by Sellafield Ltd. [51], the UK industrial site where reprocessing of UNFs is carried out.

Disposal of nuclear wastes represents the biggest challenge the nuclear industry is facing. Wastes are essentially produced throughout the whole nuclear fuel cycle; however, mining and milling of uranium ores (so-called tailings, discussed in Section 1.5.2.1), electricity production or reprocessing of irradiated fuels (depending on the type of fuel cycle implemented), and decommissioning of plants represent the predominant sources. The conventional approach envisage the following classification [48]:

- Very Low Level Waste (VLLW) contains radioactive materials that can be safely disposed of with ordinary refuse.
- Low Level Waste (LLW) includes waste with a low level of radioactivity that cannot be disposed with ordinary refuse. They have a radioactive content not exceeding 4 GBq/t of alpha or 12 GBq/t of beta/gamma activity.

- Intermediate Level Waste (ILW) represents wastes exceeding the upper boundaries for LLW, but the heat they generate, which is lower than 2 kW/m^3, is not sufficient to pose any hazard for their handling, storage or disposal.
- High Level Waste (HLW) comprises wastes that also exceed the upper boundaries for LLW and in which the temperature may significantly rise as a result of their radioactivity (heat output higher than 2 kW/m^3), therefore requiring special considerations for handling and in the storage and disposal designs.

In practice, HLW includes wastes from reprocessing of UNFs, that is the bulk of the fission products after being treated and immobilised; UNFs that have been declared as wastes are either included in HLW or regarded as a different category; ILW typically comprises stripped or leached remains of the cladding, chemical sludges or Plutonium Contaminated Materials (PCM) such as filters or process residues carrying significant amounts of plutonium; LLW comprises discarded equipment, tools or protective clothing. Notably, HLW makes around 3% in volume but 95% of total radioactivity of nuclear waste; ILW makes up some 7% in volume and has 4% of total radioactivity, whereas LLW comprises some 90% of the total volume but only 1 % of radioactivity [48].

The above classification of nuclear waste is recognised to be less than ideal, in that it only takes into account the current radioactivity without regard to half-life; for this reason, sometimes short and long-lived ILWs are distinguished, with the former being disposed with LLW and the latter with HLW. A different classification of nuclear waste is implemented in the US; this is based effectively on the source of the material rather than its content and has the advantage of avoiding any question about the categorisation of a given package, but may lead to unnecessarily expensive disposal routes for relatively innocuous consignments.

Most nuclear wastes cannot be disposed of in the form they are produced; rather they need to be further processed with the aim of converting them into a stable form that is suitable for disposal. The treatment process depends on the waste classification: HLW are immobilised into a matrix of borosilicate glass, to provide an insoluble waste form that is anticipated to remain stable for many thousands of years, and packaged into stainless steel canisters; UNFs, by contrast, are not immobilised, but simply dismantled and packaged in stainless steel containers with specific shapes for each fuel type; ILW are encapsulated in specially formulated grouts, to immobilise the radioactive material that

is usually in various forms of sludges or precipitates, and packaged in 500 litre containers; finally, two different processes are typically used for LLW: they are either compacted or incinerated in specially engineered kilns, with emissions being treated to conform to national or international standards – both approaches aim at reducing the final volume of waste.

As with waste treatment, disposal of solid waste depends on the waste classification. As noted above, VLLW is the least hazardous waste and can be disposed of along with ordinary refuse. LLW and sometimes short-lived ILW are disposed of in near-surface repositories; these are facilities on or below (but close to) the surface, where the protective covering is of the order of a few metres thick. Waste containers are placed in constructed vaults that when at full capacity are backfilled and covered with impermeable materials. The LLW repository at Drigg in Cumbria (UK) is an example of a near-surface repository. Because of their higher level of radioactivity, HLW, UNFs, long-lived and sometimes also short-lived ILW require a higher level of isolation from humans and the environment. From 1946 to 1993 these wastes were dumped in oceans as a means of disposal; in 1993 this practice was banned by international treaties and since then no other disposal concept has been implemented. At present, the favoured option by the majority of countries worldwide consists of disposal in a repository located several hundred metres (usually between 600 and 1000 metres) underground in a geologically stable environment – from which the name Geological Disposal Facility (GDF). Not a single commercial scale GDF has become fully operational anywhere in the world: the Onkalo repository at Olkiluoto, Finland is likely to be the first in operation by 2020s [52], with the UK national GDF not expected to be operational before the beginning of the 22nd century. The Waste Isolation Pilot Plant (WIPP) in the US, built to accommodate only transuranic radioactive waste, is the only pilot scale GDF currently in operation. Unlike the near-surface repository, disposal in a GDF requires that wastes are packaged in a further container designed to last for millennia. A number of designs have been developed, notable examples are the Swedish 3KBS concept, based on an outer layer of copper [53], and the Swiss full-stainless steel design [54].

1.6 Life Cycle Thinking methodologies

With diffusion of the concept of sustainability and increasing awareness of its associated challenges, the need has arisen for quantitative, science-based tools to evaluate each of the three aspects of sustainability with a holistic perspective covering a complete product system. Life Cycle Thinking implies the understanding that materials are extracted from the earth, converted into process materials, combined with other materials to make parts, assembled into a finished product, shipped to customers who use the products and finally, that the products are disposed of in some fashion (Figure 1.12).

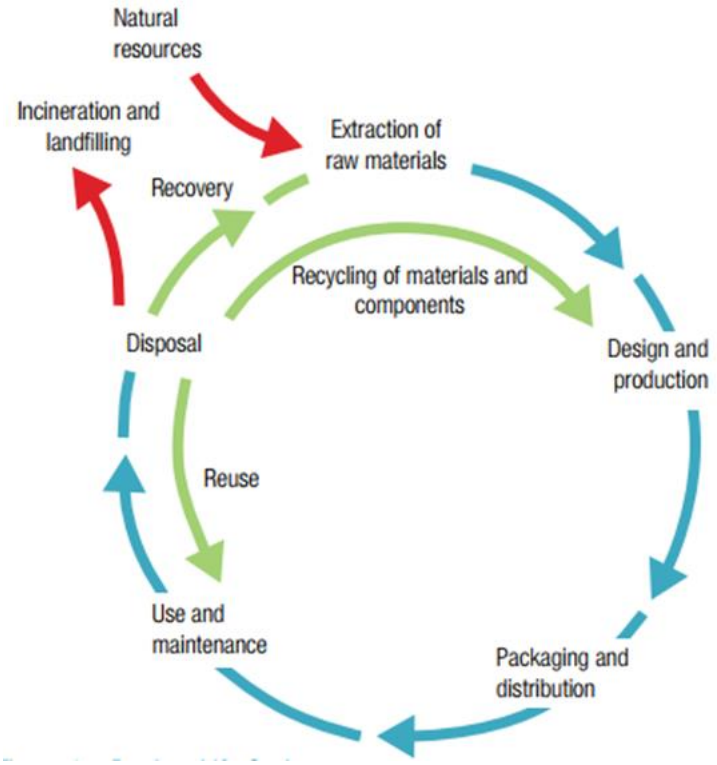

Figure 1.12- Generic product's life cycle [55]

Along the value chain, energy and other natural, social and economic resources are used, waste generated, and the related impacts, both positive and negative, are distributed across societies to varying degrees around the globe. With specific focus on the industrial sector, life cycle thinking means going beyond the narrow traditional focus on the manufacturing step, to take into account the entire life cycle of a product, thus from extraction of raw materials (cradle) to disposal of waste (grave). The key aim of Life Cycle Thinking, which sets it apart from other approaches such as those for environmental analysis, is to avoid shifting problems from one step of the life cycle to another; when a

narrow perspective is adopted, e.g. focusing on only one process or step, efforts to reduce the associated impacts may lead to increase impacts elsewhere in the life cycle.

Under the umbrella of Life Cycle Thinking, three life-cycle methodologies have been developed each focusing on one of the three aspects of sustainability:

- Life Cycle Assessment (LCA) is concerned with the environmental aspect and is the only life-cycle methodology that has been put in a standard framework (ISO 14040-14044) [56], [57]. LCA quantifies the amount of materials and energy used, and emissions and waste produced over the life-cycle, and translates them into different environmental impacts, each addressing a specific problem.
- The economic aspects are accounted in the Life Cycle Costing (LCC) methodology. This quantifies the costs associated with the complete supply chain, but goes beyond the purely economic analysis to includes the costs that are expected to be internalised in the decision-relevant future [58]. Notably, LCC is based on the LCA framework, and therefore both tools can be easily integrated.
- Social Life Cycle Assessment (S-LCA) is the most recent and least known life-cycle methodology. It is used to assess and identify the positive and negative impacts that a product has over its life-cycle on the society; this is usually expressed in terms of how the well-being of humans is affected [59], [60].

Integration of these three methodologies allow a comprehensive assessment on the sustainability of a product or service to be carried out, this is known as Life Cycle Sustainability Assessment (LCSA).

This Thesis focuses on the environmental aspect of sustainability that are linked with nuclear power, in particular with management of nuclear wastes.

1.7 Aims of the Thesis

As noted in Section 1.4, there has been a revival of the nuclear power industry in the UK after many decades of under investment, and nuclear power is expected to play a critical role in the future UK energy system. One of the biggest challenges associated with development of a new nuclear power programme is linked with finding sustainable approaches to dispose of nuclear wastes, especially of those being highly radioactive, that is HLW and UNF. Geological disposal is the favoured option by the majority of nuclear

countries, but its environmental impacts have never been quantified from a life cycle perspective; this is due to a lack of both operational data and methodologies to assess impacts arising from nuclear wastes. Furthermore, a big debate remains on whether it is more environmentally sustainable to perform a Once or Twice Through Cycle with existing nuclear reactors. This Thesis aims to address these points in the context of demonstrating how the LCA tool can be used to support the decision-making process in the nuclear industry.

The objectives of this Thesis are as follows:

1. Identify the main barriers that prevented application of LCA to the nuclear field, and develop a novel framework for incorporating radiological impacts of direct radioactive discharges and especially of future emissions arising from disposed solid waste, to be incorporated in the Impact Assessment phase of LCA;
2. Quantify and evaluate the environmental impacts of nuclear waste management practices to identify hot-spots and possible improvement options;
3. Demonstrate with practical applications how LCA can be used to support present and future decisions in the nuclear industry.

This project is part of DISTINCTIVE (Decommissioning, Immobilisation and Storage soluTIons for NuClear wasTe InVEntories) [61], a multi-disciplinary collaboration of ten universities and three key industry partners from across the UK's civil nuclear sector, that aims at tackling the challenges of decommissioning and nuclear waste management to establish safe and cost-effective solutions and promote the UK as a global leader in this area. Collaboration with industrial partners has proven crucial for completion of the project.

1.8 Outline of the Thesis

The Thesis comprises an introduction (this Chapter), six core chapters and a final Chapter that discusses and summarise the main findings, organized as follows:

Chapter 2 introduces the Life Cycle Assessment (LCA) technique from its inception in the late 1960s to the current standardized methodology, discussing its advantages and limitations, possible applications, and future developments. This Chapter is intended to

identify the main barriers that limit LCA application to the nuclear field, and provide the basic knowledge to perform an LCA study in accord with the most current research.

Chapter 3 includes a detailed and comprehensive review of Radiological Impact Assessment methodologies that have been either proposed to be included in the Impact Assessment phase of LCA or that are part of standard assessment procedures in either fields, but that could be suitable for incorporation. The review identifies the main limitations of methodologies for assessing the impacts of radioactive emissions in LCA and establishes the basis for the development of a novel approach.

Chapter 4 presents a novel, overarching framework for Radiological Impact Assessment in LCA, from which two, conceptually very different, methodologies have been derived. The methodologies are here qualitatively and quantitatively compared between each other and with a reference methodology introduced in Chapter 3, with the aim of investigating the effects of the two different approaches. The comparison is also extended to toxic substances for further validation of the methodologies.

Chapter 5 includes a LCA study on the current policy for managing AGR used fuels in the UK. This envisages their reprocessing in the Thermal Oxide Reprocessing Plant (THORP) at Sellafield site, and the disposal of ILW and HLW in the future, national Geological Disposal Facility. The study presents the first application of the methodologies developed in Chapter 4; identifies critical stages and recommends improvements.

Chapter 6 presents a further LCA study that focuses on alternative approaches for managing AGR used fuels that could eventually be implemented in the UK. The study builds upon Chapter 5 and compares the Once Through Cycle with several reprocessing approaches that envisage different ways of managing and reusing uranium and plutonium recovered from the irradiated fuels, including the conventional Twice Through Cycle with production of MOX fuel. The analysis identifies trade-offs amongst alternative approaches and concludes with recommendations on the most favourable approach (from an environmental perspective) that could be implemented in the short-term.

Chapter 7 discusses, summarises and contextualise the main findings of the Thesis, identifies open questions that have not been addressed and proposes further research that could stem from this Thesis.

Chapter 2.
The Life Cycle Assessment methodology

In recent years the need to integrate environmental concerns into decision-making processes has arisen, mainly due to increasing awareness by the humankind of the adverse effects of anthropogenic activities on the planets and its ecosystems. Amongst the myriad of environmental methodologies developed, Life Cycle Assessment (LCA) has attracted great interest chiefly for its capability to support decision-makers by evaluating the environmental impacts of products or technologies over their life cycle, from the extraction of raw materials (cradle) to disposal of waste (grave). This Chapter starts with a brief discussion on the history and development of LCA from its conception in the late 60s and early 70s, through diffusion and standardisation in the 90s, until present day. The Chapter proceeds by demonstrating different approaches for conducting LCA studies, with a particular focus on Attributional and Consequential LCA, which indeed represent the most common and well-known modes of LCA. Finally, the Chapter introduces the LCA methodology as it is applied in this Thesis, following the ISO standardised framework that envisages an iterative procedure consisting of four distinct phases, namely Goal and Scope Definition, Life Cycle Inventory, Life Cycle Impact Assessment and Life Cycle Interpretation. This Section represents a comprehensive, high-level introduction to the LCA methodology, but that it is not intended to be a substitute for the many pedagogical texts available today.

2.1 Introducing Life Cycle Assessment: History and Development

As concerns about the environment have become key matters of public debate and drivers for policy makers, public administrators, businesses and individuals, the approach to integrating environmental considerations into challenging decisions about our society have been increasingly valued in recent years [62]. For this purpose many methodologies for assessing and benchmarking environmental impacts have been developed [63], [64]; examples include Life Cycle Assessment (LCA), Strategic Environmental Assessment (SEA), Environmental Impact Assessment (EIA), Environmental Risk Assessment (ERA), Cost-Benefit Analysis (CBA), Material Flow Analysis (MFA) and Ecological Footprint. Amongst those, LCA and the Life Cycle perspective are gaining increasing interests, as demonstrated, for instance, by their inclusion into EU legislation through the Integrated Product Policy [1], [65]. LCA can in fact serve as a supportive tool for decision makers and decision-making processes by evaluating the environmental burdens associated with a product or technology[4] over its life cycle [66]–[68]. In addition to the above, many other applications may be identified, including market communication and product development [66]. As opposed to other environmental tools, LCA is a comprehensive assessment that embraces all types of impacts to both human beings and the environment, which are commonly represented under the three areas of protection, namely Human Health, Environment and Natural Resources. The unique feature of LCA, that is its main difference and advantage over other environmental tools, is the system approach that focuses on products and technologies over their life cycle. The comprehensive scope of LCA is useful to avoid problem-shifting from one phase of the life-cycle to another, from one region to another, and even from one environmental problem to another [56].

The first studies that we now recognise as Life Cycle Assessments date back to the late sixties and early seventies, a time remembered by many for the oil crisis and the energy debate. The main driver behind the first LCA studies, however, was an earlier and less remembered environmental debate associated with wasteful resource use [69]. As a

[4] The term product is used to refer to both goods and services, while technology encompasses a range of different processes delivering the same product [355].

matter of fact, the first LCA studies were all focussed on packaging and waste management, which were known as resource analysis, resource and environmental profile analysis, ecobalance and ecoprofile [70]. The oil crisis was also instrumental in developing and spreading the LCA methodology. The combination of the debate on wasteful resource use and energy is probably the main reason as to why LCA came to be such a comprehensive methodology.

The first well-known LCA study was conducted in 1969 by the US Midwest Research Institute for Coca-Cola [66], [71]–[73]. The company was looking at a number of issues related to packaging, including alternative beverage containers (plastic bottles, refillable glass bottles and disposable container) and environmental consequences of package manufacturing. One of the most interesting outcomes of the study was that the company switched from glass to plastic bottle: a radical idea because at the time plastic had the reputation of an "environmental villain" [72]. Besides the Coca-Cola study, other independent initiatives can be identified around the same time: in the UK Ian Boustead constructed a simple LCA case study around milk packaging when writing a teaching text [71]; in Germany the Federal Ministry of Education and Science commissioned a study to elucidate the role of plastics in the growing problem of packaging [74]; in Sweden LCA studies were inspired by Tetra Pak intention to introduce a PVC bottle, which was strongly criticised not only because it was disposable, but also because it was an important source of acidifying substances when burnt in incinerators [66].

In the wake of the first studies, the LCA approach gained momentum and the concept spread quickly within the packaging industry and amongst policy makers on waste management; and the energy crisis added further interest in the energy part of the analysis [75]. In industry, new LCA studies came about for reasons related to competition between industries and marketing. In the early eighties and onwards, the LCA approach expanded from primarily internal corporate decision-making towards the domain of public debate as environmental issues became more than ever of public interest; this was driven by massive environmental disasters such as the chemical accident in Bhopal, India (1984), the nuclear reactor explosion in Chernobyl (1986) and the oil spill from the tanker Exxon Valdez (1989). The early methodology was a rather distant relative of today's LCA: it was not standardised; environmental impacts were quantified in terms of energy and material resource consumption and the amount of waste produced; and only a limited

amount of emissions was reported. The first form of impact assessment was only introduced in 1984: the Critical Volume approach reported the volume of air or water needed to dilute emissions to harmless level [76].

It is in the 90s that LCA was recognised "as one of the most important tools for decision-making in the field of environmental management" [77] when the idea that environmental protection should go beyond end-of-pipe strategies and emissions control to include optimisation of product systems gained support. LCA was then an appealing tool: it was product-oriented, quantitative (and thus seemingly objective) and somehow structured. As the interest and use of LCA spread, also criticism towards the methodology increased. Critics claimed that LCA studies were often biased and used by product manufacturers who sponsored the study to promote their own product [78], [79]. The key issue was that LCA methodology was not yet standardised; on the contrary, its application was quite subjective. This concern was the driver behind the exceptional efforts directed at developing and improving the LCA methodology. In the 90s, the first scientific conferences on LCA were organized by the Society of Environmental Toxicology and Chemistry (SETAC), which was also instrumental to the development of the LCA methodology with the first guidelines published in the Code of Practice [80]. The work on methodology development culminated with the standardisation of LCA by the International Organization for Standardization (ISO), which started in 1993 and was completed in 2002 with the publication of the first 14040 series [81]–[85]. Furthermore, the work undertaken to develop the methodology has led to the establishment of LCA as an academic subject. A sign of increasing research activity on LCA is the number of articles published in academic journals, which as shown in Figure 1 increased tenfold between 1990-1999 and 2000-2009; in 2013 more than a thousand articles were published [86]. The field also developed its own journals, most notably the International Journal of Life Cycle Assessment.

Besides universities, several national and international organisations are involved at present in the continuous development of LCA, these include SETAC, the ISO and the United Nations Environment Programme (UNEP). Research undertaken in academia and organisations has also led to the development of several LCA methods, each trying to adapt the standard LCA methodology to specific applications.

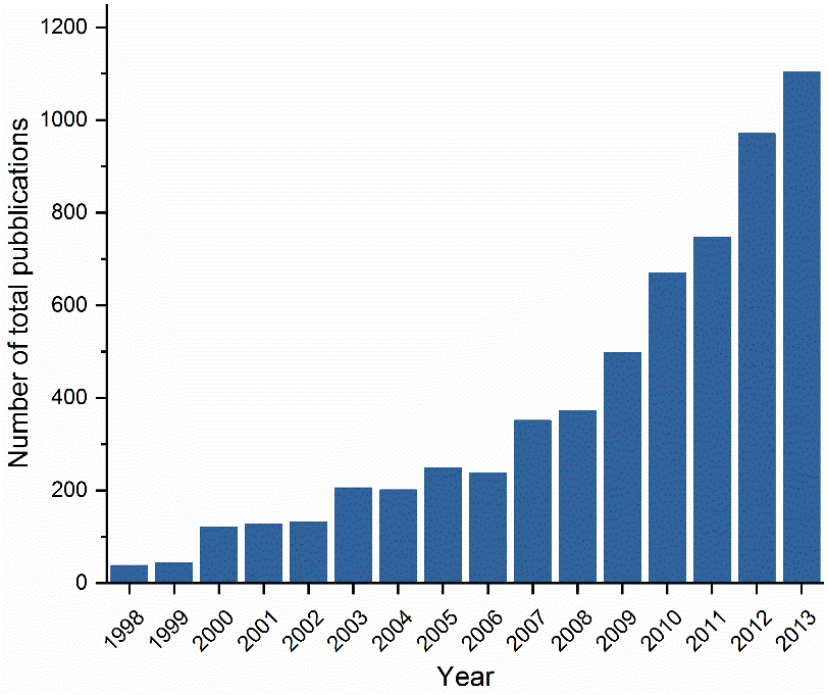

Figure 2.1 – Number of LCA articles per year from 1998 to 2013 [86]

2.2 Modes of LCA

Since the beginning of the LCA methodology development, it has been recognised that depending on the purpose of the individual study different modes of LCA can be applied [80]. Their development has been carried out almost in parallel with the development of the general methodology (mentioned in Section 2.1); however, it gained momentum in what Guinee and colleagues [87] defined as the "the decade of elaboration of LCA" (2000-2010) and still continues today. The first distinction was introduced in the 90s by Weidema [88] first and then by Tillman [89] but it wasn't properly addressed until 2001 at an international workshop on electricity data where the terms Attributional (ALCA) and Consequential (CLCA) LCA were coined [90]. ALCA was defined by its focus on the environmental burdens due to relevant physical flows to and from the studied system; CLCA by its aim to describe the environmental burdens due to changes in environmentally relevant flows as a response to possible decisions taken. Effectively, ALCA aims to answer the question "what environmental impact can be associated with this product?" whereas CLCA aims to address the generic question "What would be the environmental consequences of the implementation of these decisions?". Very similar distinctions have been proposed in several other publications, but often using different terms such as retrospective/prospective [91] or descriptive/change-oriented [89]. Besides ALCA and

CLCA, other modes of LCA have also been proposed. Frischknecht and Stucki introduced Decisional (DLCA) LCA, which uses actual or anticipated financial and contractual relations as the main basis of information [92]. The authors also propose to use the economic size of the product under study as a criterion for choosing amongst different modes of LCA. Guinee and Heijungs suggested another potentially interesting mode of LCA: Back-casting (BLCA) LCA is a scenario-based way to model specific product systems to normative future targets [93].

The emergence of different modes of LCA and the fact that their distinctions have yet to be clearly defined has created confusions for LCA practitioners. To further complicate things, the ISO standards do not recognize (yet) different modes of LCA and a unique framework is proposed to be applied regardless the purpose of the study [56], [57].

Leaving aside DLCA and BLCA, which are rather niche LCA modes yet to achieve broad consensus, let us focus on ALCA and CLCA, indeed the most common and best known modes of LCA. This topic has been the subject of numerous reviews and articles [91], [94], [95] looking at applicability and methodological differences; also, some attempts have been made in comparing the results obtained from the two modes [96]–[98]. The remainder of this Section aims at discussing the differences between these two modes of LCA in terms of their purpose and methodological principles.

With respect to their purpose, many authors [89], [99], [100] suggests that CLCA should be used for decision-making and ALCA should be used for learning purposes when no decision is at hand. However, Lundie also argues that if the difference between ALCA and CLCA results is small or if the uncertainties in the consequential modelling assumptions outweighs the insights gained from it, then attributional mode should be preferred over consequential [99]. Conversely, others maintain that both ALCA and CLCA can be used for decision-making and for learning purposes [91], [101]. Notably, according to Ekvall and co-authors [91] Consequential LCA is relevant in teleological ethics when the consequences of a situation or a rule is assessed; whereas Attributional LCA is relevant for assessing the rightfulness of a rule from either a teleological or deontological perspective. Furthermore, many authors have associated ALCA with retrospective or backward-looking studies and CLCA with prospective or forward-looking studies – although this was not mentioned when these modes were first proposed [90]. These time perspectives refer to the temporal position of the object of the study with respect to the position of the analyst,

that is the present. However, both Ekvall and co-authors [91] and Sandén and Karlström [101] argue that both CLCA and ALCA can be used for modelling past, future and current systems.

In this context, the framework proposed by Sanden appears very useful in distinguishing between different modes [102]. He reasons that there are two components of time that matter in LCA: one refers to the retrospective/prospective perspective discussed above, and the other refers to the temporal direction of the cause-effect mechanisms modelled, that is whether the study looks back at the causes of environmental impacts (attributional) or forward at the environmental effects of decisions (consequential). From these, four modes of LCA can be distinguished, each used in a different context: for instance, retrospective attributional studies are used in Environmental Product Declarations (EPDs); prospective attributional studies in assessment of the environmental impacts of future product systems; and consequential study in ex-post project evaluation (retrospective) or ex-ante project assessment (prospective). As proposed by Sanden, the framework seems to imply that the choice between consequential/attributional and retrospective/prospective studies is binary. Suh and Yang, however, argue that ALCA and CLCA represent the two poles of a continuous spectrum of possibilities, that is the definitions coined for ALCA of CLCA identify idealised approaches that cannot be achieved by real-life LCAs [103]. For instance, they reason that no CLCA is able to include all the relevant consequences of a potential decision, rather only a subset of them; and most importantly, there is no assurance that the modelled consequence will represent reality. Although this has not been pointed out by Suh and Yang, a spectrum of possibilities also exists between retrospective and prospective studies, i.e. some studies may be retrospective in some aspects and prospective in others. In view of this analysis, it is possible to conclude that the purpose of the LCA does not differentiate between different modes of LCA: it is the question that the study aims to answer that identifies the mode.

With respect to methodological differences, CLCA and ALCA share the same modelling principles. Weidema and colleagues define CLCA as “a steady-state, linear, homogenous model, with each unit fixed at a specific point in time” [104]; the same definition also applies to ALCA. The methodological differences between the two modes of LCA reside in how system boundaries are drawn, how environmental impacts of multi-functional systems are allocated and what type of data is used. These concepts are explained in

details in Sections 2.3.1.2, 2.3.2.2 and 2.3.2.3, here the discussion will focus on the methodological differences.

In ALCA the system boundaries include all the unit processes required to fulfil the function of the system under study. Conversely, as the aim of CLCA is to assess the environmental effects of decisions, the system boundaries also have to include all relevant processes affected by the decisions. Sandén and Karlström [101] identified three types of mechanisms through which effects of decisions can be modelled, and Weidema and co-workers [104], [105] developed a procedure to guide LCA practitioners in the choice of the specific processes (usually named "marginal technologies") to be included (see Section 2.3.2.1). CLCA and ALCA are also distinguished in how environmental burdens of multifunctional systems (e.g. co-product systems) are taken into account. Typically, ALCA studies use allocation rules based on physical or other (e.g price) relationships between the co-products to partition the environmental burdens amongst different functions; conversely in CLCA, the system boundaries are expanded to include the additional functions (namely the "system expansion" approach). However, it must be noted that according to the ISO standards, partitioning of environmental burdens should be avoided whenever possible, therefore many ALCA studies use the system expansion approach too. This further reinforces the existence of a continuous spectrum of possibilities between ALCA and CLCA. Indeed, the most obvious difference between CLCA and ALCA is represented by the type of data used. "Average data" represent the average environmental burdens for obtaining a unit of goods or services; whereas "marginal data" represent the effects of a small change in the system on its environmental burdens. Average data are typically used in ALCA, whilst marginal data are used in CLCA to represent the processes affected by the induced change to the system [106].

In this Thesis, the framework proposed by Sanden is used as a guide in the choice between different modes of LCA; however, it is also acknowledged that those different modes of LCA represent the two poles of a continuous spectrum. This means that while some studies can be associated with a specific mode, other cannot be strictly categorized: the LCA case study introduced in Chapter 5 is a prospective attributional LCA, whereas Chapter 6 presents a consequential LCA study (neither retrospective nor prospective) that under some specific assumptions is in effect identical to a retrospective attributional study.

2.3 The LCA methodology

The ISO international standards establish the LCA as a rigorous approach for the analysis of the environmental burdens of a product consisting of four phases as shown in Figure 2.2, these are:

1. Goal and Scope Definition;
2. Life Cycle Inventory analysis (LCI);
3. Life Cycle Impact Assessment (LCIA);
4. Life Cycle Interpretation

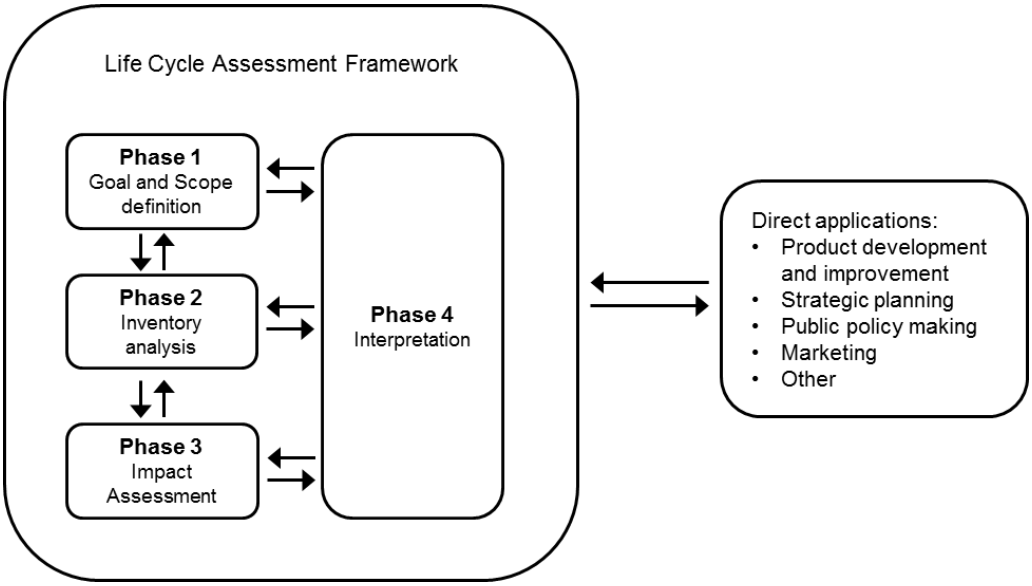

Figure 2.2 – Phases of Life cycle Assessment, adapted from ISO 14040 [56]

It must be noted that an LCA study is an iterative process because earlier phases may be revisited in light of the results of later phases. Changes in the material input to a manufacturing process or changes in the process itself may trigger the need to update the inventory component; whilst new information about the impact of substances on the environment will require the Impact Assessment to be updated. Thus, the continuous interaction between the components of an LCA is essential for a successful study. It is, however, important to recognise that phases such as Impact Assessment are continuously developing and further work is required in several areas. The four phases of LCA are explained in the following Sections.

2.3.1 Goal and scope definition

At the outset, it is essential that the purpose of the study is defined unambiguously; this includes the reason for carrying out the study (why?), its intended application (what?), the intended audience (which affects the technical level of reporting and the interpretation of results) and the commissioner of the study (to highlight potential conflicts of interests). If decision support is the primary reason, what decision the study is intended to inform also needs to be included.

The scope of the study has to include a clear statement on the specifications of the functions of the product, quantified by the functional unit, and on the processes and operations which are to be considered, expressed in terms of the system boundary; these concepts are explained in the following sub-Sections.

Finally, in the goal and scope definition also the principles for allocation (discussed in Section 2.3.2.2), data quality requirements (Section 2.3.2.3) and choice of impact categories and method for impact assessment (Section 2.3.3.4) have to be specified.

2.3.1.1 Functional unit

The Functional Unit (FU) is a quantified description of the function to be satisfied by the product under study; while the reference flow is a quantified amount of the product necessary to deliver the function; its primary purpose is to provide a reference to which input and output data are normalized. It is thus essential that the functional unit is clearly defined and measurable. As LCA studies are commonly performed to compare alternative ways of delivering some function, the functional unit also serves as a basis for the comparison. It must be noted that the results of an LCA study are strongly related to the choice of the functional unit, and thus functional units and results must never be separated.

2.3.1.2 System boundary

The system boundary separates the technical system, which includes all the activities part or affected by the life cycle of the product, from the surrounding environment (see Figure 2.3). Material or energy flows between processes are named technical flows, whereas flows between processes and the surrounding environment, that is those flows that cross the system boundaries, are named elementary flows. An elementary flow is otherwise defined as a “material or energy entering in the system which has been drawn directly

from the environment without previous human transformation", or as a "material or energy leaving the system, or discarded into the environment without subsequent human alteration" [56]. Ideally, the technical system should be modelled in a way that all inputs and outputs are elementary flows. However, this is not practical due to time and other constraints, and often not feasible due to lack of data; therefore, decisions should be taken as to which processes are to be included. These considerations have to be included in the goal and scope definition.

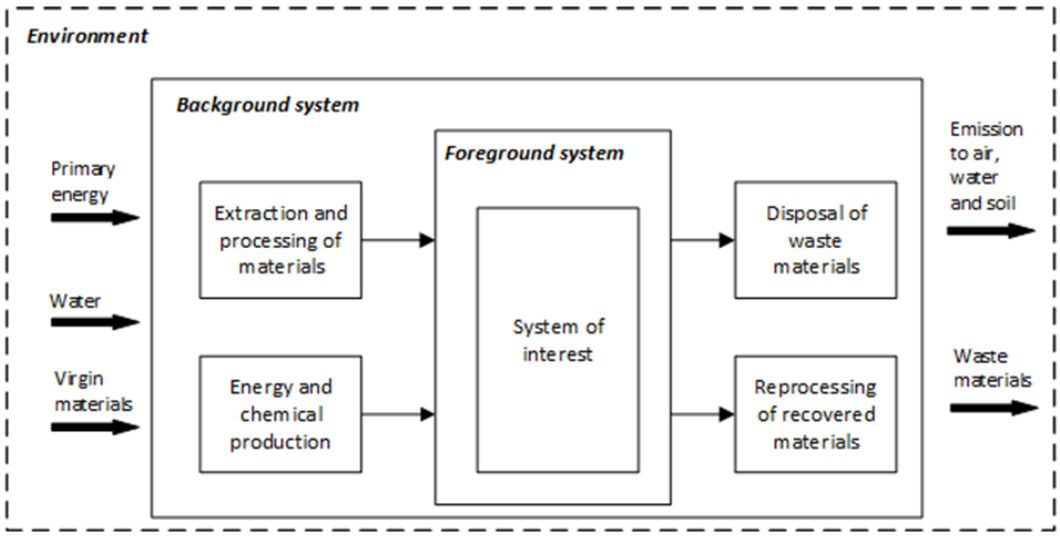

Figure 2.3 – Generic diagram of a product system illustrating the system boundary, the division between Foreground and Background system and the elementary flows

Many authors [89], [107], [108] suggest dividing the technical system into a Foreground and a Background, as shown in Figure 2.3. These concepts were developed in 1999 by the SETAC working group on enhancement of the inventory methodology [109]: the Foreground system comprises "the set of processes whose selection or mode of operation is affected directly by decisions based on the study"; the Background system comprises "all other processes which interact with the Foreground, usually by supplying or receiving material or energy". It was also reported that a sufficient but not necessary condition for a process to be in the Background is that the exchange with the Foreground takes place through a homogeneous market [108]. The distinction between Foreground and Background does not imply any distinction due to the importance of the burden related to those systems – the environmental loads of any of the two can be the largest. The division of the technical system between Foreground and Background can guide the choice of the type of data to be used. Clift and co-authors advise to use preferably primary

data for the Foreground and secondary data for the Background [108]; these are further discussed in Section 2.3.2.3.

As noted in Section 2.2, a distinguishing feature of attributional and consequential approaches lies in how system boundaries are drawn: typically, CLCA studies use enlarged system boundary to include the affected processes by the decisions under investigation. Sandén and Karlström [101] identified three typologies of consequences – that is three types of cause-effect mechanisms – of use in consequential studies:

1. First order effects represent the linear systemic response to marginal changes in the product system. For instance, a positive marginal increase in production of a good (e.g. electricity from wind turbines) may result due to market equilibrium in the decrease in production of a substitute (e.g. electricity from fossil sources); the system is credited for the "avoided" impacts of the substitute good.
2. Second order effects include indirect consequences governed by negative feedback. These take into account the economic flows related to the physical flows, and are propagated by price mechanism controlling the supply of different goods and services. For instance, a marginal change can affect the price of the product itself and of competitors' products, which may lead to a shift to a new equilibrium between demand and supply: the system is credited according to the new equilibrium. Notably, these effects are included by using partial or general equilibrium models [96], [110]–[114].
3. Third order effects include indirect consequences governed by positive feedback. These consider the effects of the cumulative build-up of stocks, structures and knowledge in producers, users and institutions. They are quantified in terms of cost reductions by using an experience (or learning) curve, and are especially relevant for emerging technologies. Third order effects are used to allocate future potential avoided emissions to a current investment into an emerging technology. For instance, the economies of scale may lead to increased performance to cost ratio when more units are produced or to an adaption of regulations and the educations system; in this way early investment in radically new technology may set in motion a self-re-enforcing process.

Notably, second order effects are typical of neo-classical economics, whilst third order effects are drawn by theories of technical change [115]. First and second order effects are often incorporated in consequential modelling; however, this is not the case for third order effects, perhaps due to difficulties in their integration in the LCA framework.

The types of consequences to be included in an LCA study depend on its goal, on the technical readiness levels (i.e. mature vs emerging) and on the time frame (i.e. short vs long term effects). For each consequence, the specific processes to be included in the system boundary – that is those that are actually affected by changes in demand or supply, namely marginal technologies (introduced in Chapter 2.2) – need to be identified. For this purpose, Weidema and co-workers [105] developed a five-step procedure to support the identification of the most relevant marginal technologies for each specific case. However, Mathiesen and co-workers [116] argued that, because LCA results are very sensitive to the choice of the marginal technology, a range of marginal technologies – rather than a single one – should be used. From an historical analysis of the Danish energy sector they demonstrated that when applying the theoretical recommendations of consequential LCA to the identification of the marginal technology, the actual marginal technology is never identical to the one predicted by CLCA.

2.3.2 Life Cycle Inventory analysis (LCI)

The second phase of LCA, Life Cycle Inventory analysis (LCI), involves collection of data for all the process units included in the system boundaries, and calculation procedures to quantify relevant inputs and outputs of the product system [56]. The LCI result is a list of quantified elementary flows (defined in Section 2.3.1.2) and represents the input for the Impact Assessment phase of LCA, discussed in Section 2.3.3. Collection of relevant, and especially high-quality data is typically the most time-consuming and resource-demanding phase of LCA. Fortunately, it is rarely required to collect high quality data for all the processes included in the system boundary to meet the goal and support the intended applications of the LCA; and sometimes it is also possible to neglect some processes or flows – i.e. cut-offs, discussed in Section 2.3.2.1 – that are deemed to have little contribution to the overall results of the study. The biggest challenge of the LCI phase is indeed represented by handling of multi-functional processes; the ISO developed a hierarchical approach to guide LCA practitioners [56], which has been subject to numerous debates and critiques.

The LCI phase starts with identification of the processes to be included in the product system, continues with data collection and construction of the LCI model, and end with calculation of LCI results. The approach presented in this Section requires knowledge about the industrial processes taking part in the life cycle, and it is usually referred to as process-based or bottom-up approach. The opposite of process-based LCI, namely Environmentally Extended Input Output Analysis (EEIO), adopts a top-down, macroscale perspective where data is collected from national statistics on the trade of products and services between sectors and information on elementary flows associated with economic sectors [117].

Nowadays, thanks to LCA softwares such as SimaPro and Gabi, and LCA databases, like Ecoinvent and Gabi, the LCI phase is significantly more rapid and immediate.

2.3.2.1 Identification of processes

A good approach to identification of the processes to be included in the product system consists in starting from the Foreground system and in particular from the unit process delivering the reference flow [57]. From this process the Foreground system should be constructed by identification of all processes that are upstream, i.e. those that deliver flows that will be physically embodied in the reference flows, and downstream, i.e. those that lead from the reference flow to waste management and disposal. Once the Foreground system is complete, the Background system should be constructed to include all the processes that supply the Foreground system with materials or energy, usually through a homogenous market [108].

As a general rule, the product system should include every process and every flow that contributes to the Functional Unit. However, not all these processes or flows may be quantitatively relevant; those that are deemed to be irrelevant may be entirely cut-off, i.e. excluded from the product system. The ILCD recommends that only those processes that do not contribute to more than 5% of the total impact may be excluded. Because if the overall impact was known there would be no reason to apply cut-offs, these are usually applied by defining a minimum required completeness of the system, that is the LCI model represent a set percentage of the real product system for each impact category [118].

Consequential LCI modelling

As noted in Section 2.3.1.2, CLCA usually uses enlarged system boundaries to include the affected processes by the decisions under investigation. Consequential LCI is a very challenging task that includes knowledge about market trend and response to changes in demand and supply, identification of those technologies likely to be affected by the change in demand and supply, and of those products that may substitute others.

Bjorn and co-authors developed a 4-step procedure (reported in Figure 2.4) for identification of those processes to be included in LCI [119]. The first step regards consideration of whether the question at hand addresses changes in demand or supply for a given product. If the decision relates to changes in demand, the procedure envisages identifying possible constraints in the market (step two), defined as the market in which changes in demand will not lead to changes in supply. If the market is constrained, a substitute that delivers the same function of the product affected by the decision and the production technology affected by the change should be identified (step three and four). However, if the market is not constrained, the production technology likely to respond to the change in demand should be identified. On the other hand, if the decision at hand relates to changes in supply, the procedure envisages first identification of product substitutes and then the production technology affected by the change in demand (step three and four).

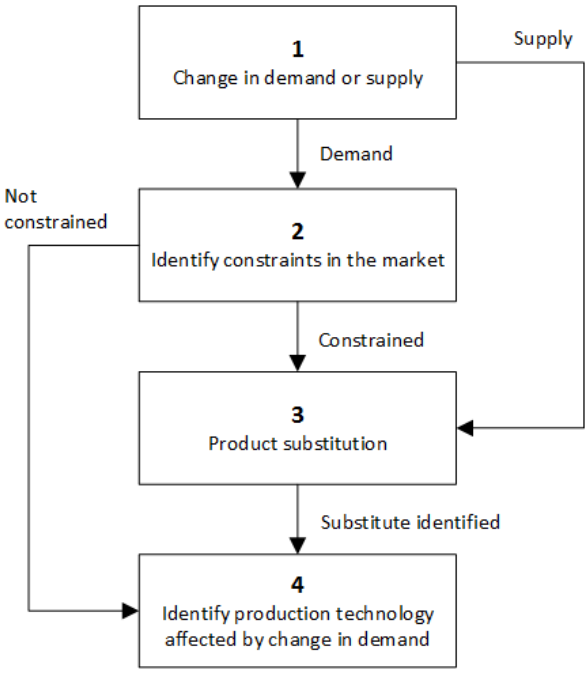

Figure 2.4 – Four-step procedure for identification of processes to be included in consequential LCI [119]

Identification of the marginal technology represents another challenging task in LCI modelling. In 1999, Weidema and co-workers proposed a 5-step procedure for identification of marginal technologies for both short and long-term studies, which is reported in Figure 2.5 [105].

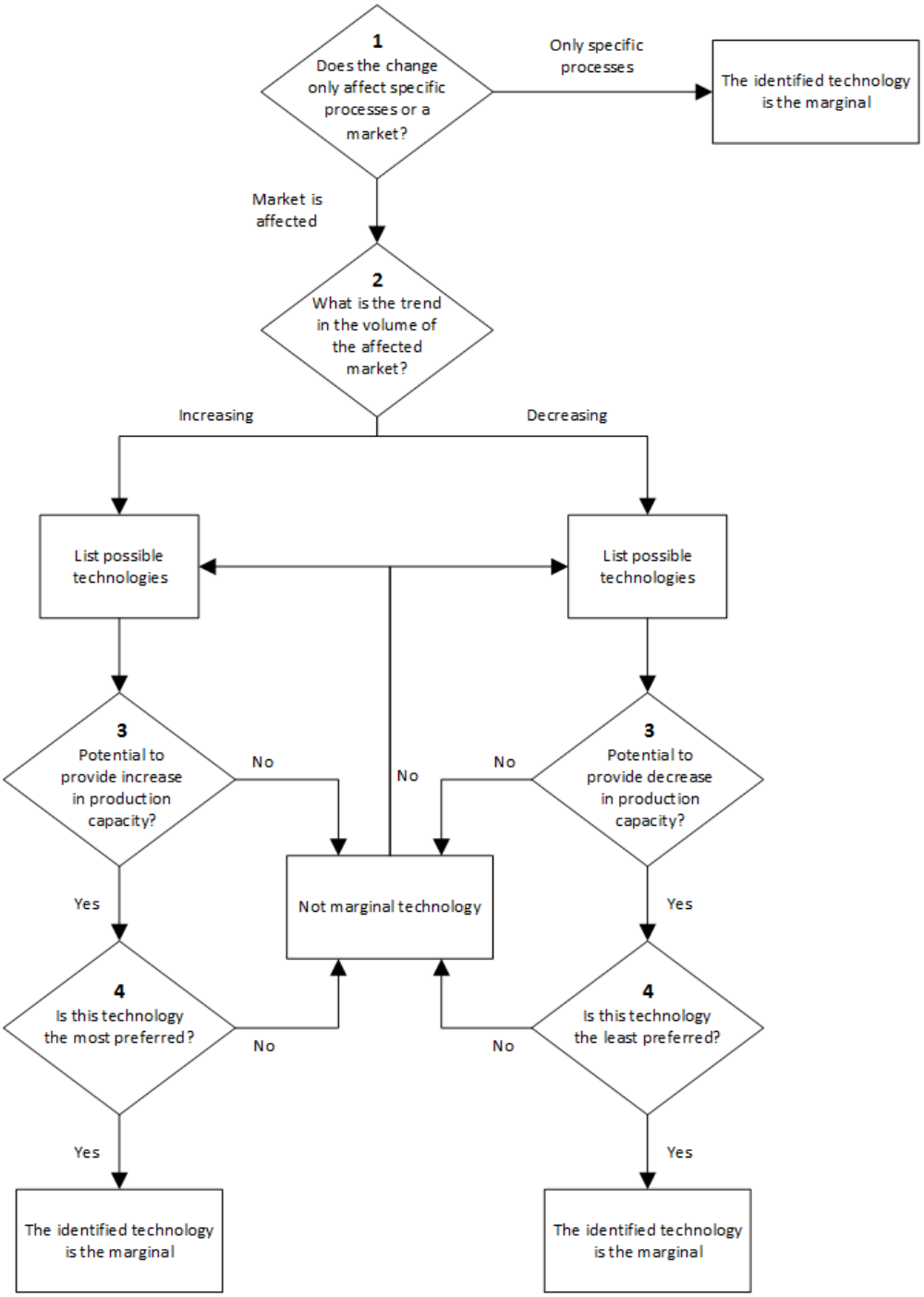

Figure 2.5- Approach for identifying marginal technologies [105]

The first step of the procedure envisages understanding whether the changes at hand only affect a specific process or the whole market. If only a specific process is affected, the technologies of these processes are by definition the marginal technologies; however, if the market is affected, the procedure continues to the following step that consists in analysing the trend of the market, that is whether it is a decreasing or increasing market (step two). If the market is generally decreasing, the least competitive technology is the marginal technology. By contrast, if the market is generally increasing, then new capacity need to be installed and it will be the most competitive technology. Depending on the market trend, the third step envisages identification of those technologies that have the potential to provide an increase or reduction in production capacity: only those processes that are capable to respond to changes in the market can be marginal technologies. Finally, amongst the potential marginal technologies identified, depending on the market trend the most or least preferred is selected as the marginal technology (step four).

2.3.2.2 Multi-functional processes

Multi-functional processes are those processes that deliver additional functions to the one identified by the Functional Unit. A typical example is a municipal solid waste incinerator that provides the multiple services of waste disposal and source of heat and electricity. Multifunctional processes represent a methodological challenge in LCA because as soon as secondary products or services are generated, the multifunctional process becomes part of another product system as well, and therefore the environmental impacts from such process can no longer be fully ascribed to the product system studied. Approaches to "solve multi-functionality" aim at attributing to multi-functional process only the environmental burdens associated with the primary function.

For this purpose, the ISO developed a hierarchical approach for handling multifunctional processes [56]:

1. Subdivision should always be the first choice when approaching multi-functional processes. Because unit processes can be defined at many levels of details (and usually a high level of detail is not required for LCA studies), the multi-functionality may be revealed as artificial by simply increasing the resolution of the model; in this case, the process can effectively be divided into two separate processes delivering separate functions. Applying subdivision effectively means cutting-off those parts of the process providing secondary functions.

2. If multi-functionality is not solved by subdivision, then system expansion should be used. In comparative assessments this means expanding the product system that does not feature the multi-functionality with the most likely alternative way of providing the secondary function provided by the multi-functional product system. For instance, in the comparison shown in Figure 2.6 between power plant 1, which co-generates electricity and heat (the second function) and power plant 2, which only produces electricity two power plants producing electricity, the system boundary of power plant 2 is expanded to include the most likely alternative way of providing district heat in that region (part a of Figure 2.6). Expansion of system 2 is mathematically equivalent to subtracting to system 1 the alternative way of providing district heat (part b of Figure 2.6); this approach is known as "crediting" the system for the inputs and outputs (and thus environmental impacts) of the avoided secondary function. It must be noted that unlike system expansion, crediting can also be used for studies focusing only one product system [120]. Although system expansion and crediting are mathematically equivalent, the ISO only mentions the former.

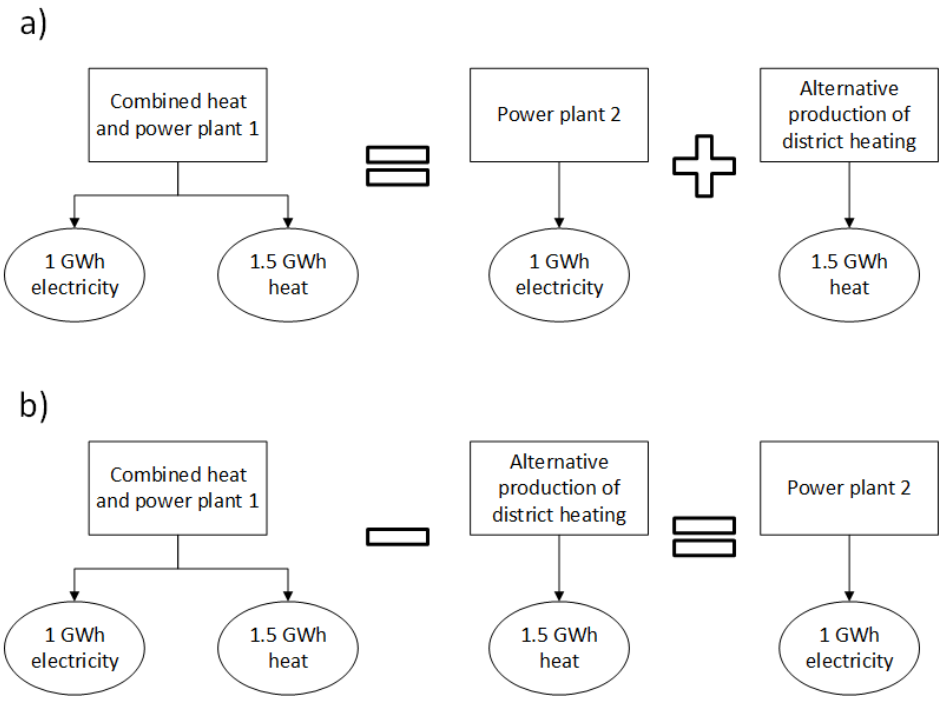

Figure 2.6 – Example of system expansion (a) and crediting (b) approaches for handling multifunctional systems in a comparative analysis of two electricity production technologies

3. Finally, if multi-functionality cannot be solved by both subdivision and system expansion/crediting, the ISO standards recommend partitioning the inputs and outputs amongst the different functions; this approach is referred to as partitioning, or less correctly as allocation. (By allocation the ISO standards refer to the whole procedure of handling multi-functional processes, which includes steps 1, 2 and 3). Partitioning of inputs and outputs can be performed according to different principles. The ISO recommends using underlying physical relationships between products or functions that should reflect the way in which inputs and outputs are changed by quantitative changes in the products or functions. A physical relationship can be based for instance on mass or energy content. If no physical relationship can be identified, partitioning should be based on other relationships, such as the economic value of the products.

As noted in Section 2.2, the ISO standards do not acknowledge the existence of different modes of LCA such as consequential and attributional. Therefore, if the ISO standards are to be followed, the same hierarchical approach should be applied to both LCA modes. For this reason, the ISO procedure has been the subject of strong criticism and numerous debates on whether the current hierarchy of solutions works for both attributional and consequential approaches [121]. Subdivision is widely accepted to be the favoured approach in both cases. However, with respect to steps two and three, some argue that partitioning is the correct and only method to be used in attributional studies [98]–[100]; whereas system expansion is already an inherent part of consequential LCA because the system boundaries are already defined to include all the activities that may be affected by the decisions investigated [106], [121]. Others maintain that the system expansion should be preferred over partitioning (hence, in accordance with the ISO hierarchy) in both attributional and consequential studies [94].

2.3.2.3 Data collection

For each process unit identified in the previous Section (2.3.2.2) and included in the system boundary, data representative of routine operations of the same processes should be collected. In this phase, planning is of crucial importance and has the purpose of balancing the effort of data collection by the relevance of the respective data and information. This is essential in order to avoid wasting time on collecting high-quality data that have low relevance for the LCA results or spend too little time on collecting data that

may be highly relevant for the results. According to the ISO standards, four classes of data can be distinguished [56], these are:

- Energy inputs, raw materials, ancillary inputs, other physical inputs;
- Products, co-products and waste;
- Emissions to air, discharges to water and soil; and
- Other environmental aspects.

Furthermore, data can also be distinguished as primary and secondary data according to their quality. Primary data represent high-quality data either measured directly at the plant site, or derived from measurements, process flowsheets or questionnaire compiled by people with technical knowledge of the process. Collection of primary data is very time and resource-consuming, especially for LCA consultants (as opposed to companies' in-house LCA practitioners) because it requires liaising with companies, obtaining permissions to go on site, signing non-disclosure agreements and preparing questionnaires that are clear and easily understandable by people with little or no knowledge of LCA. Secondary data, on the other hand, either represents the average performance of an industry sector according to its market distribution or describes the same process but occurring at a different site (thus being of lower quality than primary data). Secondary data sources are scientific literature, reports or LCI databases, such as Ecoinvent and Gabi. Clift and co-workers advise to use primary data for the Foreground system, and secondary data for the Background systems and for those processes for which primary data could not be collected [108].

A further classification of data envisages distinction between average and marginal. The former represent the average performance of a process unit, whilst the latter describe the effect of small changes of its output [94]. Attributional LCA excludes the use of marginal data, whilst consequential LCA focuses on marginal data, especially when relevant for the purpose of assessing the consequences of a decision.

2.3.2.4 Constructing the LCI model and calculating LCI result

Once all the processes of the product system have been identified and data collected for each of them, the LCI model is constructed by linking all the process units, which represent building blocks, so that the Functional Unit defined in the goal and scope definition phase is satisfied. The LCI result represents all the flows that are either obtained from the

environment or released to the environment, namely elementary flows; these are the basis for the subsequent phase in LCA, namely Life Cycle Impact Assessment (LCIA).

2.3.3 Life Cycle Impact Assessment (LCIA)

The third phase of LCA, Life Cycle Impact Assessment (LCIA), aims at describing the environmental consequences of the elementary flows quantified in the Life Cycle Inventory (LCI), with the objective of producing results that are more intelligible and easier to communicate. Results of the inventory analysis are represented by extensive tables of elementary flows that are not easy to grasp for LCA practitioners, let alone for people without a scientific or LCA-specific background. LCIA allows to reduce this complexity by grouping elementary flows that contribute to the same kind of environmental consequence (e.g. acidification of water ecosystems, depletion of ozone layer), and to compare different elementary flows in terms of the importance of their environmental consequences (e.g. is it worse to release 1 kg of methane or 10 kg of carbon dioxide in terms of global warming?). Effectively, the Impact Assessment phase "translates" elementary flows into environmental impacts representing the consequences of human interventions (emissions or resources consumption) in a specific point along the cause-effect chain. It is important to keep in mind that the results of LCIA should be interpreted as potential impacts, and not as actual impacts nor as exceeding of thresholds or safety margins or risks; this is because: i) they are referred to the functional unit, and do not necessarily represent the real magnitude of emissions or resources consumption; ii) the inventory data is integrated over space and time, and thus LCIA results represent impacts occurring at different locations and over different time horizons; iii) impacts are assessed using generic, rather than site-specific models.

In practice, the LCIA phase is largely automated and requires the practitioner to choose an LCIA method, i.e. a collection of LCIA models developed or recommended by an institution (see Section 2.3.3.2 for a more detailed definition), and few other settings via menus and buttons in the LCA software. However, without an adequate understanding of underlying principles of LCIA, neither an informed decision of LCIA method nor a meaningful interpretation of results is possible.

Figure 2.7 reports the three mandatory and three optional steps that compose the LCIA phase according to the ISO standards [56], [57].

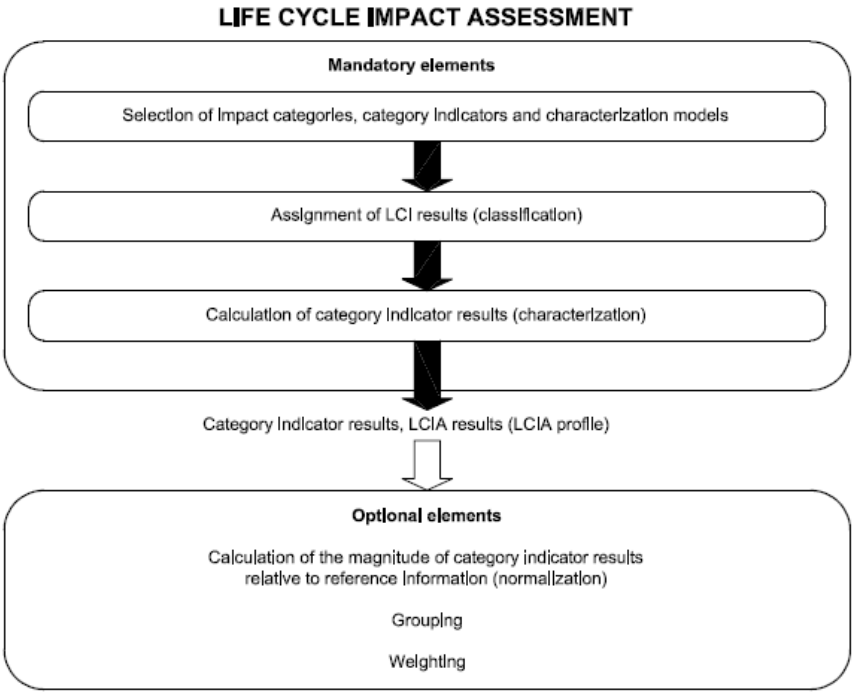

Figure 2.7 – Optional and mandatory steps of LCIA [56]

2.3.3.1 Terminology

Before commencing description of the LCIA mandatory and optional steps, it is worth introducing some essential terminology and definitions. Table 2.1 provides practical examples for each term introduced.

Table 2.1 – Examples of LCIA terms, adapted from ISO 14040 [56]

Term	Example
Impact category	Climate change
LCI results	Amount of a greenhouse gas per functional unit
Characterization model	Baseline model of 100 years of the Intergovernmental Panel on Climate Change`
Category Indicator	Infrared radiative force (W/m^2)
Characterization factor	Global warming potential (GWP_{100}) for each greenhouse gas (kg CO_2-equivalent/kg gas)
Category Indicator results	Kilograms of CO_2-equivalents per functional unit

An impact category is defined as a class of environmental issues to which Life Cycle Inventory (LCI) results may be assigned; and category indicators are a quantifiable representation of impact categories. The environmental mechanism represents the combination of physical, chemical and biological processes that through cause-effect chains (or impact pathways) link LCI results to category indicators. Characterisation

models reflect the environmental mechanism by describing the mathematical relationship between the LCI results and category indicators; they are used to derive characterisation factors, which effectively translate LCI results to the common unit of the category indicator.

2.3.3.2 Mandatory steps

Selection of impact categories, category indicators and characterisation models

The first step of LCIA entails selection of impact categories, category indicators and characterisation models, whose definitions are included in Section 2.3.3.1. These should include those most suitable amongst those available that are consistent with the declared goal of the study; and should be accompanied by a comprehensive documentation, with all information and sources being referenced. Notably, the selection of impact categories should reflect as closely as possible those environmental issues that are related to the product system being studied. The ISO 14044 provides a detailed list of requirements and recommendations on how to perform the selection [57].

Generally, several impact categories, category indicators and characterisation models are combined into predefined sets, usually referred to as LCIA methods; notable examples are ILCD [122], [123], ReCiPe [124], CML [125], TRACI [126], EDIP [127], Impact 2002+ [128]. The ISO standards by principle do not provide any recommendations about which LCIA method should be used, but some organisations do recommend the use of a specific LCIA method or parts of it. For instance, the Joint Research Centre (JRC) of the European Commission has carried out a systematic review that has led to recommendation of best available approaches for impact assessment [122], [129]; the LCIA method based on such recommendations is known as ILCD (International reference Life Cycle Data system) [123]. Furthermore, some methods with a stronger national focus are recommended by national governmental bodies for use in their respective country, such as LIME in Japan, or TRACI in the US.

Because of the increasing numbers of LCIA methods or impact categories, category indicators and characterisation models, the choice is not trivial and requires a significant and deep knowledge of their main features and differences.

Classification

Classification involves sorting and assigning the LCI results to the impact categories to which they contribute. This step, thus, requires knowledge of what the implications of emissions or resource use are on the environment. For example, to perform the classification step LCA practitioners need to know that NO_x is both an acidic substance and a nutrient, thus contributing to both acidification and eutrophication categories, but also that it can promote creation of secondary pollutants, thus contributing to the photochemical oxidant formation category. Notably, this example also illustrates that emitted substances can contribute to multiple impact categories through either parallel mechanisms, e.g. NO_x impacting on acidification and eutrophication category, or through a chain of effects (i.e. in series) that eventually lead to other impact categories, e.g. NO_x can take part in chemical reactions leading to photo-oxidant formation.

Characterisation

Characterisation is the quantitative step that calculates the category indicator results by conversion of LCI results into common units and aggregation of the converted results within the same impact category according to the following formula:

$$IR_i = \sum_j \left(CF_j * E_j\right) \qquad \text{eq. 2.1}$$

The category Indicator Result (IR) for impact category i is thus the summation of the product of all the LCI results assigned to a specific impact category (E_j) and their respective Characterisation Factor (CF_j).

Characterisation factors represent the impact contribution per unit of elementary flow to a specific impact category. They are obtained from characterisation models, which reflect the environmental mechanisms leading to an adverse effect. The starting point of the environmental mechanism is set by an environmental intervention in the form of an elementary flow in the LCI, and the contribution from the LCI flow is measured by the ability to affect an indicator for the impact category, which is selected along the cause-effect chain of the impact category.

Typically, the further down the cause-effect chain an indicator is chosen, the more environmental relevance it has; at the same time, however, the level of uncertainty of the model may increase and measurability decrease. It must be noted that, contrary to a frequent misconception, this does not mean that the total uncertainty of an indicator

increases when going further down the cause-effect chain, because the increase in model uncertainty is compensated by an increase in environmental relevance. If the latter is low (as is the case for indicators placed early in the cause-effect chain) the relationship of an indicator to an environmental issue is assumed but not modelled and thus hypothetical and therefore uncertain [130]. Two different types of impact categories indicators have been established: midpoint indicators are located early in the cause-effect chain and are the most commonly used; while endpoint indicators are located at the end of the cause-effect chain to represent impacts on one of the three Areas of Protection (AoP), namely Human Health, Ecosystems Quality or Nature Environment, and Natural Resources and Ecosystems Services. All endpoint indicators related to a given AoP share the same unit and their contribution can be summed up to give an overall impact score per AoP (with or without weighting). When using midpoint indicators, aggregation and contribution analysis of different impact categories can only be performed after normalisation and weighting (discussed in Section 2.3.3.3). For illustrative purposes, Figure 2.8 reports the ILCD framework for midpoint and endpoint indicators.

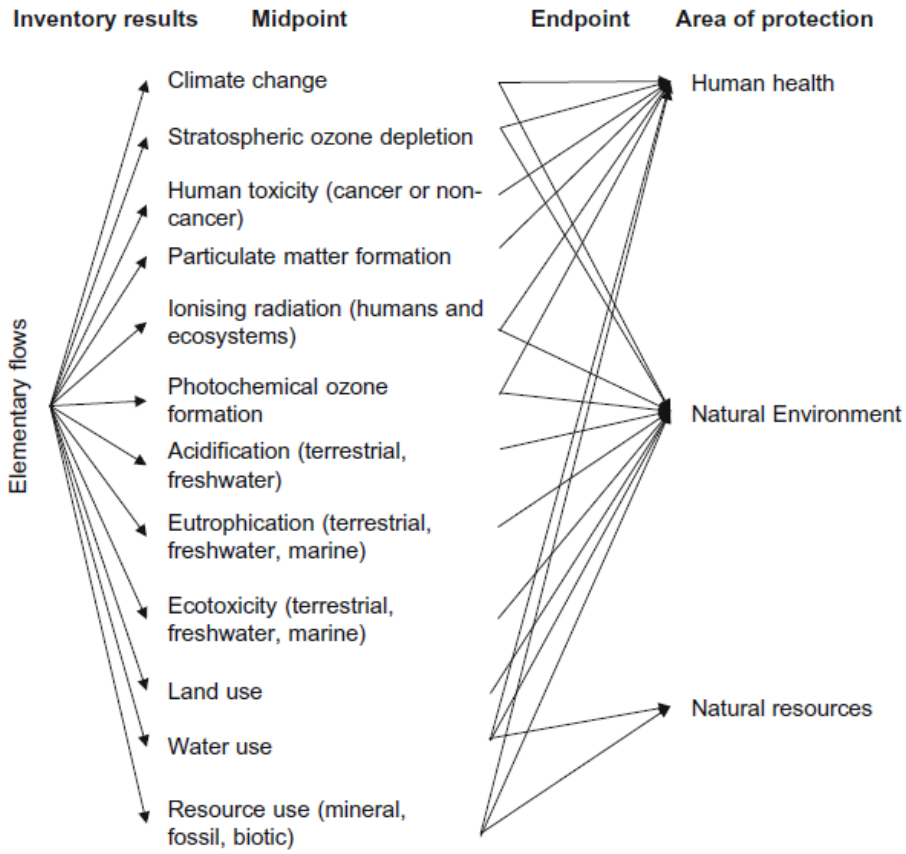

Figure 2.8 – ILCD framework for midpoint and endpoint levels of impact assessment [130]

Midpoint and endpoint indicators shouldn't be considered as two distinct approaches. Performing an LCIA on both midpoint and endpoint level is a viable option when they are functional to complement each other output respectively (i.e. in ILCD method) and it is in some cases necessary for supporting the interpretation of the results obtained.

2.3.3.3 Optional steps

Normalisation

Normalisation is the first, and in the majority of cases the only, optional step performed in LCIA. Its primary purpose is to allow comparison amongst indicator results of different impact categories that otherwise cannot be performed due to the different units involved. This is achieved by relating the indicator results to a common scale representing the total impact of a reference system. In this way, normalisation also provides information on which impacts are large or small. Typical reference systems are:

- Geographical areas, e.g. global, continental, national, regional or local;
- Inhabitants of a geographical area;
- Industrial sector of a geographical area;
- Baseline reference scenario, such as another product system.

If the reference system is one of the compared alternatives in an LCA study, the normalisation is defined as internal.

Normalisation can in theory be performed on both midpoint and endpoint indicator results. However, it is much more common to normalise midpoint rather than endpoint results, which are already aggregated in fewer categories and thus comparison can be performed at the level of the areas of protection.

Normalised Scores (NI) or impacts are obtained by multiplying the Indicator Results (IR) for each impact category i by the respective Normalisation Factor (NF_i):

$$NI_i = IR_i * NF_i \qquad \text{eq. 2.2}$$

Normalisation factors are calculated per impact category by conducting an LCI and LCIA on the reference system. For instance, in the case the reference system represents the potential impact per inhabitant of a geographical zone, the Normalisation Factors (NF) per impact category i are obtained as follows:

$$NF_i = \left(\frac{\sum_j (CF_j * E_j)}{P}\right)^{-1} \quad \text{eq. 2.3}$$

Where E are the environmental interventions quantified in LCI, CF the characterisation factors for each intervention j and P the population of the reference region.

Typically, two different approaches exist for collection of the inventory data required for calculation of NFs, these are production- and consumption-based. They differ whether the inventory represents the interventions taking place in the reference system or those that are caused somewhere else as a consequence of the consumption taking place in the reference system.

It must be noted that normalised scores must be interpreted with caution because the procedure essentially changes the results and consequently may change the conclusions drawn from these. Notably, the normalisation step may introduce significant biases that depend on the choice of the reference system and the extent of its inventory data. To reduce biases it is recommended to choose large reference systems and make sure that all the interventions of the product system take place in the same geographical area as those of the reference system [130]. Furthermore, although normalisation helps identifying impacts that are large compared to the chosen reference, this does not always coincide with those impacts being more important or representing the highest priorities [130].

In practice, normalisation factors are included in LCIA methods alongside characterisation factors. Because calculation of normalisation factors requires use of characterisation factors, LCA practitioners should use the same LCIA method for characterisation and normalisation.

Weighting

The weighting step is performed by application of equal or different weights to each category indicator results after normalisation. The weighting scheme used needs to be in accordance with the goal and scope definition, which implies that both the target group and the decisions to be supported are to be considered.

Weighting is chiefly used to prioritise impact categories according to an agreed weighting scheme. The weighting step also allows aggregating impact scores into several or one

single indicator, comparison across impact categories and communication of results representing an underlying prioritisation of ethical values. It is essential to note that because criteria underpinning the weighting method have no scientific bases, this step is based on the subjective choices of one person or a group of individuals.

Grouping

This step involves aggregating impact categories in one or more groups as predefined in the goal and scope definition. The purpose is to increase effectiveness of communication and readability of results. Grouping may also involve sorting or ranking, applying one of the two following methods:

- Sorting and clustering midpoints impact categories on a geographical basis.
- Ranking the impact categories according to a defined hierarchy of priority.

Examples of groups or cluster can be global/regional/local impacts, or impacts with high/medium/low priority.

2.3.3.4 Impact categories

In this Thesis, the ILCD midpoint method for impact assessment developed by the European Commission Joint Research Centre (EC-JRC) to promote sustainable consumption and production patterns in the Union has been adopted. Table 2.2 lists the impact categories recommended within the ILCD, along with the assessment model and indicator results; notably, no model has been found to be mature enough for recommendation in the Ionising Radiation (ecosystems) and ecotoxicity (terrestrial and marine) impact categories. A generic introduction to these impact categories is presented in the following Sections.

Acidification

The Acidification category quantifies the impact of pollutants with the potential of causing acidifications of soil or aquatic ecosystems. Acidifying pollutants are mainly released by combustion processes occurring in thermal power plants, combustion engines, waste incinerators, e.g. sulphur and nitrogen oxides and hydrochloric acid, and agriculture, which is the main contributor to emissions of ammonia. Following the release, acidifying compounds are trapped by water in the form of rain, fog and snow, and then deposited onto different receptors. Examples of impacts are fish mortality in lakes, such as in Scandinavian lakes in the 1990s, leaching of toxic metals from soil and rocks, and damage

to coral reefs, forests, and even buildings and monuments [130]. Because of their high water solubility, the atmospheric residence time of acidifying pollutants is limited to a few days, and therefore acidification represents a regional effect.

Table 2.2 – Midpoint impact categories of the ILCD method

Impact category	Assessment Model	Indicator result	Source
Acidification	Accumulated Exceedance	mol H^+ equivalent	[131], [132]
Climate change	Bern model – Global Warming Potential over a 100 years' time horizon	kg CO_2 equivalent	[133]
Ecotoxicity, freshwater	USEtox	CTUe[5]	[134]
Eutrophication, aquatic	EUTREND	kg P (freshwater) and kg N (sea water) equivalent	[135]
Eutrophication, terrestrial	Accumulated Exceedance	mol N equivalent	[131], [132]
Human toxicity, cancer effects	USEtox	CTUh[6]	[134]
Human toxicity, non-cancer effects	USEtox	CTUh	[134]
Ionising radiation, human health	Human Health Damages	kg U^{235} equivalent (to air)	[136]
Land use	Soil Organic Matter (SOM) model	kg SOM (deficit)	[137]
Ozone depletion	World Meteorological Organization (WMO) model over an infinite time horizon	kg CFC-11 equivalent	[138]
Particulate matter/Respiratory inorganics	RiskPoll	kg PM2.5 equivalent/kg (intake fraction for fine particles)	[139]
Photochemical ozone formation	LOTOS-EUROS	kg NMVOC[7] equivalent	[140]
Resource depletion, mineral, fossil	CML 2002 model	kg antimony (Sb) equivalent	[125]
Resource depletion, water	Swiss Ecoscarcity model	m^3 water use related to local scarcity of water	[141]

[5] Comparative Toxic Units for Ecosystems
[6] Comparative Toxic Units for Humans
[7] Non-Methane Volatile Organic Compounds

The first acidification models were based solely on the potency of the emitted substance in terms of its ability to release protons. Latest models, including Accumulated Exceedance (included in ILCD, see Table 2.2), take also into account regional parameters such as the sensitivity of the receiving environment in terms of buffering capacity of the soils and sensitivity of the ecosystems to acidification as expressed by their critical load.

Climate Change

The Climate Change category expresses the impact of Greenhouse Gas (GHG) emissions based on the extent to which they increase the radiative forcing in the atmosphere. Out of the total sunlight reaching the Earth's atmosphere, around 28% is directly reflected back into space by air molecules, clouds and the surface of the earth such as oceans and icy regions (albedo effect); the remaining portion is absorbed by GHG (around 21%) and the Earth's surface (around 50%) [130]. The latter heats up the planetary surface and is released back into the atmosphere as infrared radiation (black body radiation) with a longer wave-length than the absorbed radiation. This infrared radiation is partially absorbed by GHGs and kept in the atmosphere instead of being expelled into space, explaining why the temperature of the atmosphere increases with its content of GHGs. The major anthropogenic contributions to the greenhouse effect are represented by emissions of carbon dioxide, methane and nitrogen oxides mainly from burning fossil fuels and deforestation.

Until now, Global Warming Potential (GWP) developed by the Intergovernmental Panel on Climate Change (IPCC) has been unanimously adopted as climate change indicator in LCIA methods, and is defined as the ratio of the cumulative radiative forcing over a time span T of a given GHG and that of CO_2:

$$GWP_{T,i} = \frac{\int_0^T a_i c_i(t)dt}{\int_0^T a_{CO_2} c_{CO_2}(t)dt} \qquad \text{eq. 2.4}$$

where a_i is the radiative forcing per unit concentration increase of GHG i (W/m2), c_i (t) is the concentration of GHG i at time t, and T is the time over which the integration is performed (year). 100 years is the most widely used time horizon.

Freshwater Ecotoxicity and Human Toxicity

Today's toxicology science adheres to the principle stated some 500 years ago by Paracelus according to which the dose is what differentiates a poison from a remedy. As

a consequence any substance emitted may lead to toxic impacts depending on a number of factors including emitted quantity (calculated in the LCI phase), mobility, persistence, exposure patterns and bioavailability, and toxicity. The latter four are accounted in the characterisation factors.

For both damages to ecosystems and human beings, the environmental mechanism is modelled in three steps (four for endpoint indicators):

1. Fate modelling estimates the increase in concentration in a given environmental medium as a consequence of an emission of a toxic pollutant in a specific environmental medium.
2. The exposure model quantifies the amount of toxic pollutant with which ecosystems or humans can get in contact. For the human category, this step relates the amount of chemical in a given environmental medium to the chemical intake by humans, known as exposure rate. With respect to ecosystems impacts, this step estimates the fraction of the toxic pollutant that is accessible to an organism for uptake or adsorption (bioavailability).
3. The effect model relates the amount taken in by humans or the amount bioavailable in ecosystems to toxic effects. For humans, toxic effects are usually expressed as the increase of cases of carcinogenic and non-carcinogenic diseases; whilst for ecosystems toxic effects are quantified by the fraction of species that will experience a detrimental effect.

Characterisation factors for both humans and ecosystems are emission compartment-specific, and are obtained by multiplication of a fate (FF), exposure (XF) and effect (EF) factor, as follows:

$$FF * XF * EF = CF \qquad \text{eq. 2.5}$$

USEtox (recommended in ILCD, Table 2.2) is a scientific consensus model, born under the auspices of UNEP and SETAC and based on an extensive comparison with other existing approaches. It is the most widely used model for assessing impacts of toxic substances on both humans and ecosystems.

Eutrophication, aquatic and terrestrial

The two Eutrophication categories describe the impact of macro nutrients, the most important of which are nitrogen (N), and phosphorus (P), on aquatic and terrestrial ecosystems respectively. Excessive levels of nutrients in the aquatic ecosystem trigger a cause-effect chain that causes growth and blooming of algae and other aquatic plants, and reduction of oxygen availability, leading to degradation of water quality, altered species composition and loss of biodiversity. For terrestrial systems, eutrophication primarily causes changes in the function and species composition of nitrogen-poor ecosystems and also damages to crops and forests leading to reduced yields. Because of these environmental mechanisms, eutrophication is a regional impact category, highly dependent on local conditions.

Like the Acidification category, early eutrophication characterisation factors quantified exclusively express the number of moles of nitrogen (for aquatic ecosystems) and phosphorus (for terrestrial ecosystems) that can be released into the environment from one mole of the substance emitted; whereas recent models such as the Accumulated Exceedance (included in ILCD, Table 2.2) also take into account regional parameters.

Ionising radiations, human health

The Ionising Radiations category covers impacts of radioactive emissions on human beings. Radionuclides are routinely emitted by anthropogenic activities such as the nuclear, coal and building industries, in the form of air and water-borne materials. Exposure of humans to radioactive materials can lead to both stochastic and deterministic effects in terms of fatal and non-fatal cancers and hereditary effects.

Because radionuclides share a number of characteristics with toxic substances such as heavy metals, their impact on humans tends to be modelled in a similar way. The model developed by Frischknecht and co-workers, included in ILCD (Table 2.2) is one example.

The Ionising Radiations category is fully addressed in Chapter 3.

Land use

Land use refers to anthropogenic activities in a given soil area – such as agriculture, urban settlement, mineral extraction – that cause reduction of soil availability and quality. Land use is typically distinguished in transformation and occupation: the former refers to the conversion from one state to another of a given area, whilst occupation includes the use

of a certain area for a particular purpose. Both types of use through different environmental mechanisms can lead to the same effects, which include decrease or loss of biodiversity, changes in water cycle (caused for instance by river diversion), change in local and regional climate regulation due to changes in land cover and albedo (e.g. desertification and tropical deforestation), decline in food production and rise in flood and drought risks.

Land use is a relatively new impact category, with the first operation models available from 2010 [130]. Land use models focus either on impacts on biodiversity or on ecosystem services. The model developed by Milà i Canals and co-workers and included in ILCD (Table 2.2) belongs to this latter category and uses Soil Organic Matter (SOM) as a proxy for soil quality and thus life support functions.

Ozone Depletion

The Ozone Depletion category quantifies the effect of bromated and chlorinated substances on the depletion of the ozone layer. Ozone (O_3) is a harmful pollutant in the lower atmospheric layers, i.e. tropospheric and ground-level (See Photochemical Ozone Formation category), but it is an essential substance in the upper atmosphere (stratosphere) as it screens out more than 99% of the energy-rich ultraviolet (UV) radiation from the sun, preventing it from reaching the Earth's surface. The impact of UV on living organisms depends on its wavelength: short-wavelength UV (type C) is the most dangerous wavelength but it is almost completely filtered by the ozone layer; UV-B (medium wavelength) is of the greatest concern due to the ozone layer depletion; UV-A (long wavelength) is not absorbed by ozone. Impacts are also dependent on duration and intensity of the exposure, and include skin cancer, cataracts, immune system disease to humans, epidermal damage to animals, and radiation damage to the photosynthetic organs of plants. Emissions of substances related to ozone depletion have been successfully addressed with the enforcement of the Montreal Protocol signed in 1987.

Ozone is formed by reaction of oxygen with UV radiation and is destroyed by UV radiation, visible light and brominated and chlorinated substances acting as catalyst. The Ozone Depletion Potential developed by the World Meteorological Organisation (WMO) is the category indicator used in all existing LCIA methods; it reflects the change in the stratospheric ozone column in the steady-state due to the emissions of that substance relative to those of R-11 (trichlorofluoromethane), according to the following equation:

$$ODP_i = \frac{\delta[O_3]_i}{\delta[O_3]_{R-11}} \quad \text{eq. 2.6}$$

Where $\delta[O_3]_{i,R-11}$ represent the change in the ozone column for a given substance *i* or R-11.

Particulate Matter/Respiratory Inorganics

The category of Particulate Matter/Respiratory Inorganics quantifies toxicity-related effects on human health caused by Particulate Matter (PM). Exposure to PM leads to numerous detrimental effects including chronic and acute respiratory diseases, cardiovascular diseases, chronic and acute mortality and lung cancer. In 2013 outdoor and household PM pollution contributed alone to 71% of premature deaths attributable to environmental factors and 19% to all factors [130].

PM can be distinguished according to formation type (primary and secondary) and aerodynamic diameter (respirable, coarse, fine and ultrafine). Primary PM includes particles that are directly emitted (e.g. from road transport or power plants), whilst secondary PM refers to particles formed by reactions with precursor substances such as nitrogen oxides, sulphur oxides, ammonia and Volatile Organic Compounds (VOCs).

Generally, calculation of characterisation factors for the Particulate Matter/Respiratory Inorganics category is based on a framework very similar to that developed for toxic substances – thus consisting of three or four step depending on whether the midpoint or endpoint approach is adopted. The need to separate impacts from PM and other toxic substances originated from significant characteristics of PM modelling including the complex atmospheric chemistry involved in the formation of secondary PM, the effect of different emission heights and the effect assessment based on exposure-response functions derived from epidemiological studies – which is not possible for toxic chemicals due to poor data availability. The RiskPoll model included in ILCD (Table 2.2) is a comprehensive and flexible method capable of accounting for a number of factors and parameters.

Photochemical Ozone formation

The Photochemical Ozone formation category addresses the impacts caused by ozone and other reactive oxygen compounds; these are formed as secondary contaminants in the troposphere by the oxidation of the primary contaminants, mainly volatile organic

compounds (VOC) and carbon monoxide, in the presence of nitrogen oxides and under the influence of light. The most important source of emissions of VOC derives from road traffic and use of organic solvents; whilst carbon monoxide is mainly emitted from combustion processes with insufficient supply of oxygen, including road traffic and other forms of incomplete combustion of fossil fuels and biomass.

The negative impacts are associated with their reactive nature that enables them to oxidise organic molecules: when inhaled they can cause damages to the respiratory tract tissue and trigger respiratory diseases in humans; or they can attack surfaces of plants or even enter plant leaves damaging the photosynthetic organs.

The LOTOS-EUROS as adapted by Van Zelm and co-workers and included in ILCD consists of a detailed fate and exposure model designed for calculating continent-specific characterisation factors, and spatially differentiated factors for the European continent [140].

Resource depletion, mineral, fossil and renewable

Natural resources can be classified according to their origin into biotic and abiotic, that is whether resources are or are not living at the moment of extraction, or according to their availability into stock (resources with a finite and fixed reserve), fund (resources that are regenerated but can be depleted if the extraction rate exceeds regeneration) and flows (resources that are provided as flows, e.g. solar radiation and wind). There is still much debate about what the issue of concern of natural resources is and about how this should be addressed in LCIA [129]. There are three main approaches for quantifying impacts of resource use:

1. Aggregate natural resource consumption based on an inherent property;
2. Relate natural resource consumption to resource stocks or availability;
3. Relate current natural resource consumption to consequences of future extraction of natural resources (e.g. increased energy use or costs).

The most widely accepted method focuses on depletion of abiotic resources (stocks), using either the total estimated reserves of the resource (ultimate reserves approach) or only that part that has reasonable potential to become economically and technically feasible to exploit (reserve base approach). The 2002 CML model included in ILCD (Table 2.2) is based on the former approach.

Resource depletion, water

With respect to the distinction of natural resources made above, water is a resource provided as flow that cannot be depleted. There is sufficient water on our planet to meet current needs of ecosystems and humans: of the total water deposited every year on land, about 62% is returned directly to atmosphere while only about 3% is used by humans and human activities. However, despite the small fraction, there are still important issues associated with water use; for instance, many rivers are running dry from overuse, leading to significant damages to local ecosystems. The issue is not about having too little water; rather it is about mismanagement of a resource that is required by both humans and ecosystems. Excessive consumption of water may lead to poor availability for humans, which may lead to deployment of backup technologies such as desalinisation of water if socio-economic resources are available, or otherwise cause deprivation and therefore water-associated diseases if socio-economics means are not sufficient. Excessive consumption of water also leads to damages to ecosystems such as loss of biodiversity due to reduction of available habitat.

The majority of impact categories including water impacts are based on a Demand-To-Availability (DTA) or Distance-To-Target (DTT) approach. One of such latter, the Swiss Ecoscarcity model, is included in the ILCD (Table 2.2).

2.3.4 Life Cycle Interpretation

The last phase of LCA, Life Cycle Interpretation, proceeds through three steps: identification of significant issues, evaluation, and conclusions and recommendations.

2.3.4.1 Identification of significant issues

This step aims at identifying those elements that have the potential to change significantly the final results of the LCA study. These elements are referred to as significant issues and may include inventory data, impact categories and individual process units or groups of processes that have significant contributions to LCI or LCIA results. Other significant issues are methodological choices and assumptions, characterisation factors, normalisation or weighting factors used in the Impact Assessment phase. In comparative studies, this step should also focus on the analysis of how the environmental profiles of the compared systems differ.

2.3.4.2 Evaluation

The objectives of the Evaluation step are to establish the basis for the conclusions and recommendations that represent the final step of Life Cycle Interpretation. For this purpose, the significant issues identified in the previous step are evaluated with respect to their influence on the overall results. The evaluation phase involves:

- Completeness check, performed on the inventory and impact assessment results to determine the degree to which available data is complete for the significant issues.
- Sensitivity check, which has the purpose of assessing the reliability of the final results. When possible, it is recommended to perform both sensitivity and uncertainty analysis. The former aims at assessing how sensitive are the results with respect to changes in inputs, such as inventory data; whilst the purpose of the latter is to evaluate how much each input parameters contributes to the output variance [142].
- Consistency check, performed to determine whether the assumptions, methods and data are consistent with the goal and scope of the study. In the case of comparative studies, consistency check also investigates whether these elements are in accordance between the systems compared.

2.3.4.3 Conclusions and recommendations

The final step of Life Cycle Interpretation, and thus of LCA, has the purpose of drawing conclusions based on the outcomes of the other phases of LCA. In this step it is essential to verify that conclusions are in accordance with the goal and scope of the study, and in particular with data quality requirements, assumptions, limitations of the study and application-oriented requirements. Based on final conclusions, recommendations related with the intended application of the study should be developed.

Chapter 3.
Radiological impact assessment approaches for LCA: a review

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 Chapter is to collect and critically analyse radiological impact assessment methodologies to establish a basis for developing a standard approach. The review prepares the ground for the development of a general approach and practical methodologies to incorporate the impacts of radionuclides in LCA; these are presented in Chapter 4. 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 Chapter 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.

3.1 Introduction

Many industrial processes during routine operations release radionuclides into the environment in the form of air- or water-borne materials or solid wastes. The nuclear [144], coal [145], [146], oil and gas [147], fertiliser [148], [149] and building industries [150], [151] are among the major contributors [152], [153]. 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. 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 [154]–[156]. 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 [157]–[160]. 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 [161].

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. "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 Chapter 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. This Chapter focuses 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 levels 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 suitable for incorporation in LCIA.

The Chapter is organised as follows: an overview of the general approach to radiological impact assessment is presented in Section 3.2; Section 3.3 reviews current approaches to radiological impact assessment within LCA, which are commented upon in Section 3.4: it is discussed their suitability for inclusion in LCIA and suggested possible approaches for further development. Section 3.5 provides a summary of the most significant conclusions of this review leading to future work (Chapter 4) to develop a sound approach to include radiological impacts in LCIA.

3.2 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 [162], [163]. 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. For instance, in the method developed at the Institute of Environmental Sciences (CML) at Leiden University [125] 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 3.2 and discussed in Section 4.

3.2.1 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" [164]. 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 [161].

3.2.2 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 [165]–[167].

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 [168]. On a general basis a number of approaches are available to estimate the environmental concentration of radionuclides. Numerical calculations, based on Lagrangian "puff" [169] or Eulerian grid [170], [171] 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 [172]–[174]; 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 [175] 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 [176]. The concentration in food is obtained through specific models that establish how radionuclides move from each environmental medium to each food category [177]. 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, e.g. see [178].

Finally, the last step involves determining the amount of radioactive 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) [179]. The International Committee on Radiological Protection (ICRP), however, has

developed other dosimetric indicators [179], [180]. 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 into consideration those aspects: the resulting value is termed the Effective Dose and is measured in Sieverts (Sv) [179], [181], [182]. 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 [179], [181], [182]. 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" [183], [184]: 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) [179].

Exposure to ionising radiation can lead to two distinct types of effects: deterministic and stochastic [179], [185]. 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 3.1) [179], [185]. 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 3.1) 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 [179], [185]. 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 3.2) [179], [185]. 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 [186], [187].

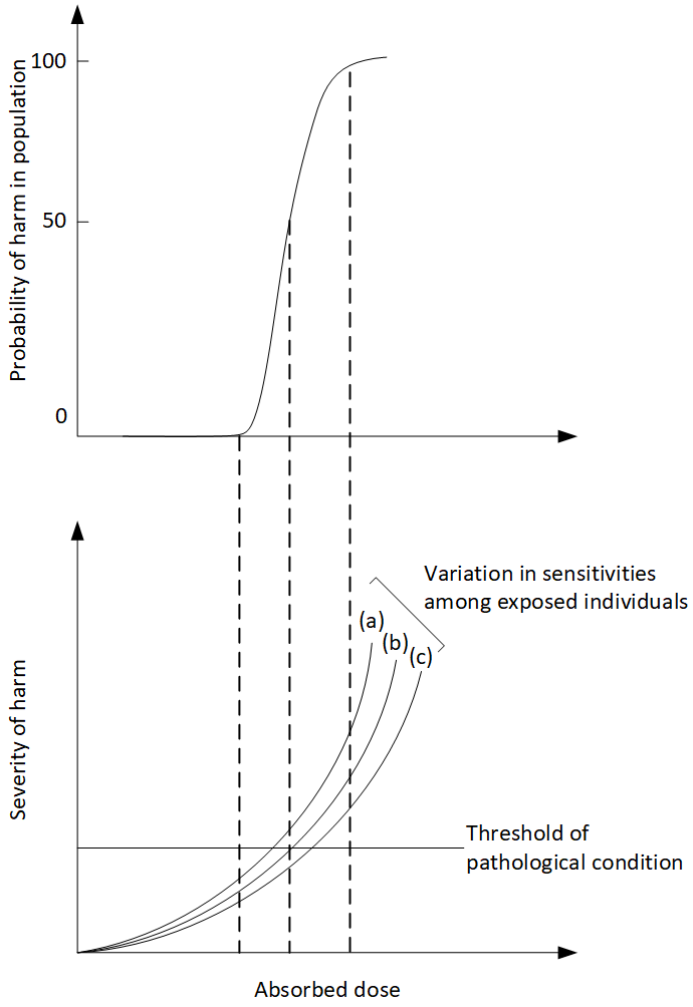

Figure 3.1 – Typical dose-effect relationships for deterministic effects expressed in a population (adapted from [188])

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) [182], [185]. 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 3.2). 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 [179], [185], 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.

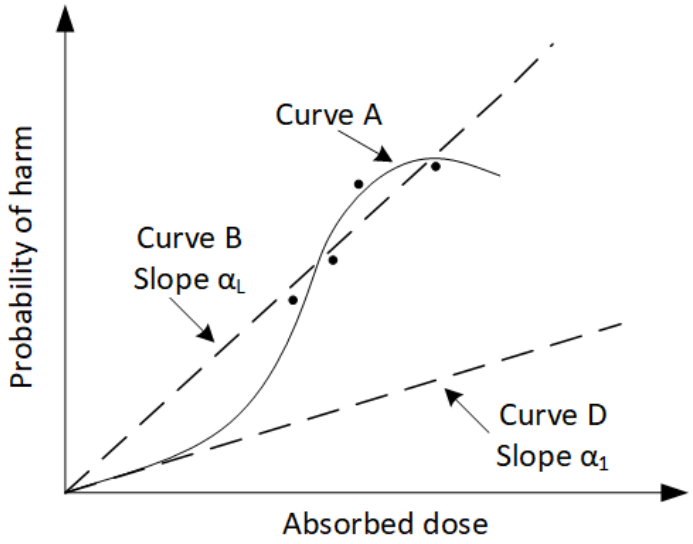

Figure 3.2 – Schematic curves of incidence vs adsorbed dose (adapted from [189])

3.2.3 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 [181], reiterated in the recommendations of 1990 [185] and supported by the International Atomic Energy Agency [190]. 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 [191]: 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 [192]. 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 [179]. 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 [193], followed by two more reports covering approaches to model the transfer of radionuclides to non-human biota [194] and to extend the application of the radiological protection system to different exposure conditions, e.g. unplanned events) [195]. 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 [196].

3.3 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 [197]. The Handbook on Life Cycle Assessment [125], 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 [198]). 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 3.1 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 3.3 and Figure 3.4 summarise the emission sources included and the source-pathway-effect model behind each, for human and environmental impacts respectively.

Table 3.1 – 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	[199]
Site-specific	Direct routine discharges	Humans	Site-specific	Worst case	Mid-point	Sv or ManSv	[204]
Damage-based	Direct routine discharges	Humans	Site-independent (preferably applicable in Europe)	Average	End-point	DALY	[211]
Human Irradiation	Direct routine discharges and emissions from solid waste	Humans	Site-dependent	Worst case	End-point	Risk	[220]
NDA Value Framework	Direct routine discharges and emissions from solid waste	Humans	Site-dependent	Worst case	End-point	Sterling	[227]
Environmental Irradiation	Direct routine discharges	Ecosystems	Site-dependent	Worst case	Mid-point	-	[220]
SLERA	Direct routine discharges to fresh water	Ecosystems	Site-independent	Average	Mid-point	CTUe	[235]

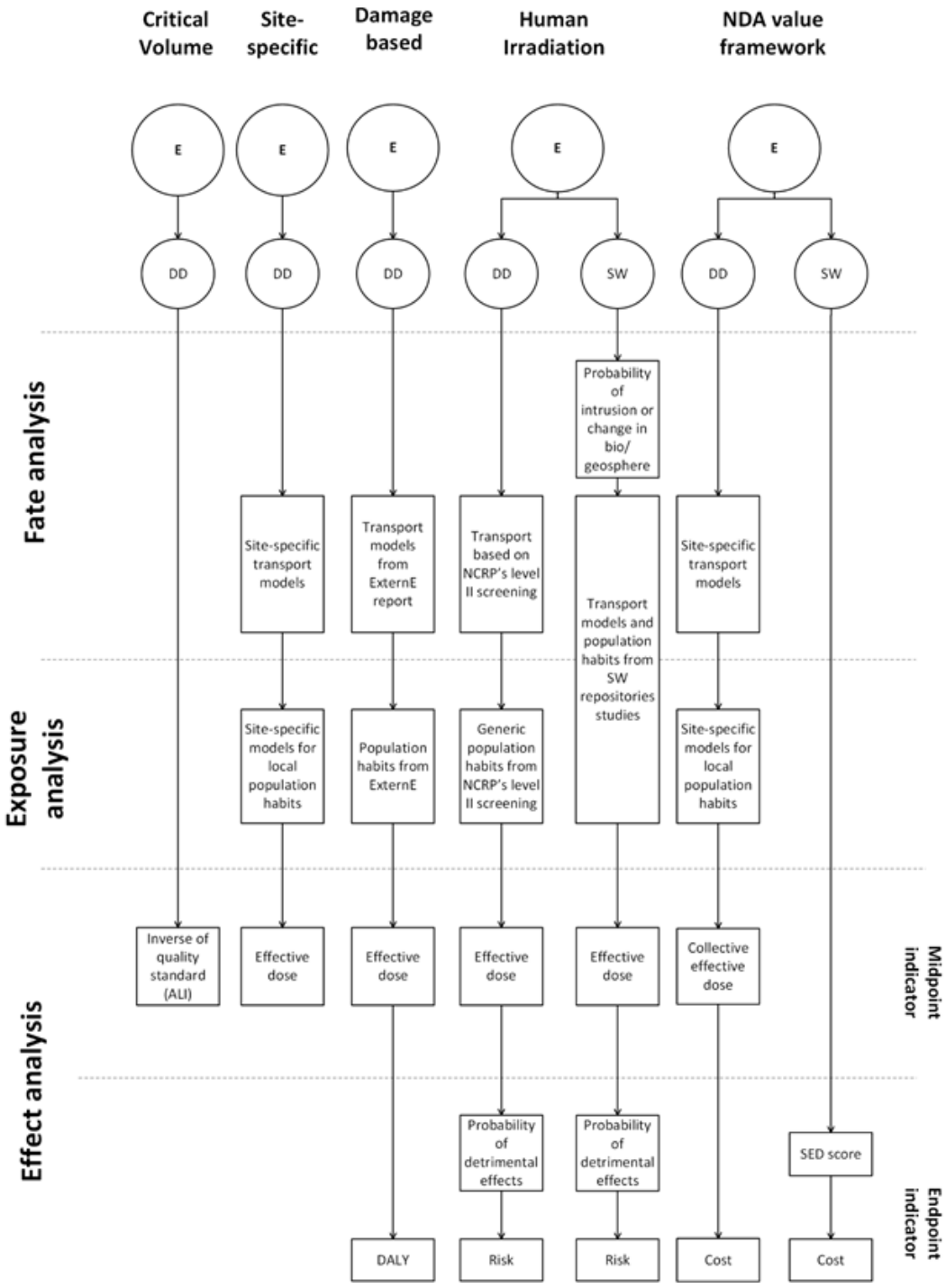

Figure 3.3 – Outline of methodologies for radiological impact assessment on humans. E= Emission; DD= Direct Discharges; SW= Solid Waste.

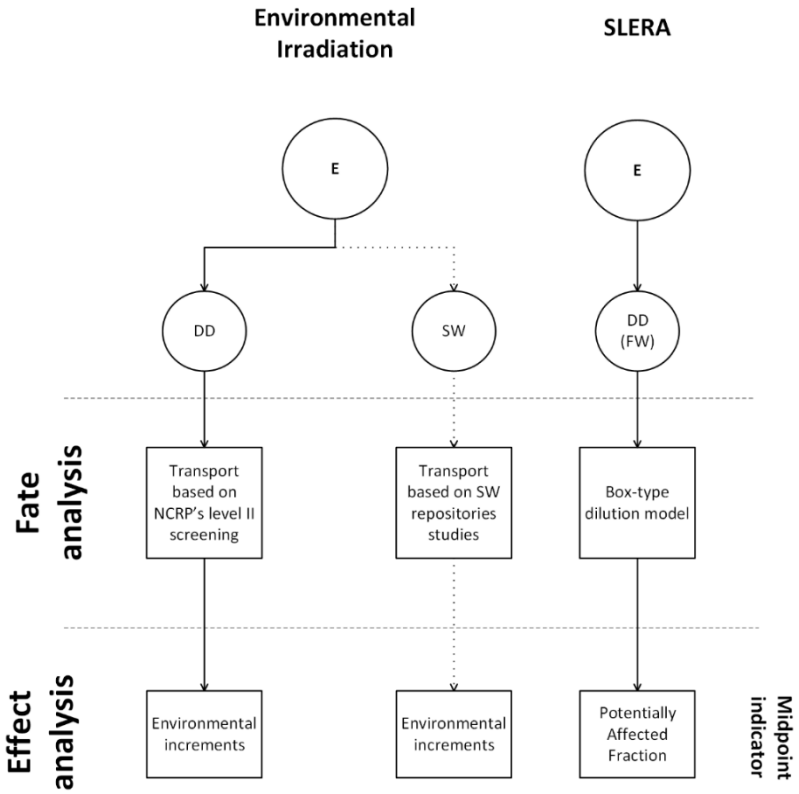

Figure 3.4 – Outline of methodologies for radiological impact assessment on ecosystems. E= Emission; DD= Direct Discharges; SW= Solid Waste; FW= Freshwater.

3.3.1 Human health impacts

3.3.1.1 Critical Volume approach

Heijungs and colleagues were the first to discuss how radionuclide release could be included in LCIA [199] and to propose a possible approach [200]. 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 3.3). 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 [182], [201] to define the basic limit for occupational exposure to a given radionuclide, was chosen by Heijungs and co-workers 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 (see Section 2.3.3.2). 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 [66], [202], [203]. In view of this, it can be said that the characterisation factors produced by the Critical Volume adopt the mid-point perspective.

3.3.1.2 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 3.3 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 [166] 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) [204], 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 [185], [205]. 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) [204] and translated into the computer code PC-CREAM [206] which has since been continuously updated [207]. 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 [173], [174]. 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"): 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 yet been included in the PC-CREAM 08 methodology [207]. 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 [208]. The marine section contains two multi-compartment models representing respectively northern European [209] and Mediterranean seas [210]. 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 (site-specific) data 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.

3.3.1.3 Human Health Damages approach

Frischknecht and colleagues [211] 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 3.3, 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 and co-workers [136] 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 is used [175], 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 [212] 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 [213]–[215]. The estimated absorbed dose is converted into a whole body dose (Sv) by means of the ICRP factors [185].

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 and co-workers [216] 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 [185], [216]–[218].

Following the different perspectives identified in Cultural Theory [219], the damage-based methodology considered three scenarios from which two sets of damage factors were derived. Cultural theory describes five ways of living 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 3.3.1.1).

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 3.4.

3.3.1.4 Human Irradiation approach

A different approach, also specifically developed with LCIA in mind, was devised by Solberg-Johansen [220], 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 3.3.2.1. As shown in Figure 3.3, 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 3.2.2, 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 [185].

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 [166] 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 [222], [223];
- 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 [166];
- The Performance Assessment of Geological Isolation Systems for radioactive waste (PAGIS), used to determine the impact of vitrified High Level Waste (HLW) [224], [225];
- 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 [225], [226].

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 3.3.1.2), the fate models for both direct discharges and disturbance of solid waste make use of the concept of the critical group. Thus, the methodology is site-dependent, as reported in Table 3.1, produces worst case estimates of impacts (i.e. the highest possible values) 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 3.3, 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 3.4.1.

3.3.1.5 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 [227]. 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 [228]. 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 [66]. Thus, the NDA methodology itself is not likely to achieve the acceptance necessary for general adoption in LCIA. This Section focuses 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 [205] 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 3.3.1.4) 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 [227]. 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 [231]. 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 3.3.1.4), inclusion of stochastic events (discusses in Section 3.4.1) and use of site-dependent fate models (Section 3.4.2) may represent a barrier to acceptance.

3.3.2 Ecological impacts

3.3.2.1 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 [220], [232] also considered an Environmental Irradiation Category using the approach summarised in Figure 3.4. 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 3.3 and Figure 3.4). 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 3.1). However, the Effect Factors are not related to exposure routes and risk-based dose relationships; instead, they rely on the "Environmental Increment" (EI) concept [233]. 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 3.2.2) [186], [187]. 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 [233]. 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 [220].

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.

3.3.2.2 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 [234]. The approach has been tested by its authors in a case study for the Rhone river watershed [235]. 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) [236], [237]. 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 [238]–[240] is one of a number [241]–[243] 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 the ecosystems [129] 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 3.4, the methodology comprises the two familiar steps of fate and effect analysis.

The fate analysis uses the MacKay modelling approach [175] 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 [237].

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 [244]. 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 [244]. 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 [245]. 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 [245]. The scarcity of these data represents one of the major limitations of this methodology.

3.4 Discussion: Incorporating impacts of radionuclides in LCIA

In Section 3.3, 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 3.3.2.1) and SLERA (Section 3.3.2.2) – deal with radiological impacts on non-human biota; all the others focus on potential human health effects. The site-specific approach (Section 3.3.1.2) and the NDA's value framework (Section 3.3.1.5) represent the only methodologies developed as assessment procedures in other fields, not specifically for LCA.

As noted at the beginning of Section 3.3, 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.

3.4.1 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 3.3.1.3 and 3.3.1.5 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 3.1).

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 3.2, 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 3.3.1.4) 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 3.3.1.5) 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 [246] constructed to impound the fine tailing materials left from milling of mined ores [247]. Recent studies have shown that tailings dams failures occur more than once a month worldwide [248], [249] – although the number has considerably decreased from the 1970s and 80s – and that at least one major failure occurs each year [250]. However, databases such as Ecoinvent [251] include short and long-term emissions to atmosphere and to groundwater (leachate) from uranium [252], [253] and generic sulfidic tailings [254] 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 acceptance 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.

3.4.2 Human health impacts

Table 3.2 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 [126] and EDIP – 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 3.2 – Radiological impact categories in LCA impact methods

Method	Version	Approach	Metric	Features	Source
CML	2001, non baseline	Human Health Damages	DALY	Egalitarian/Hierarchist	[125]
RECIPE	1.08; 1.07	Human Health Damages	kg U_{235} eq.; DALY	Egalitarian/Hierarchist and Individualist; Hereditary effects.	[124]
Eco-Indicator	1999	Human Health Damages	DALY	Egalitarian/Hierarchist and Individualist.	[255]
Impact2002+	2002	Human Health Damages	DALY	Egalitarian/Hierarchist and Individualist.	[128]
ILCD	2011	Human Health Damages	kg U_{235} eq.;	Egalitarian/Hierarchist	[122], [123]

The Human Health Damages approach was developed specifically for use in LCIA and embodies two features specific to this use (see Table 3.1). 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 [202], 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 3.3.1.3, 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 potentially 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 3.3.1.4) appears to be of particular interest, mainly because, as discussed in Section 3.4.1, 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 3.1), 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 [125]. 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 larger 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 3.3.1.5) 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 3.4.1, 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 3.3.1.2). 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 3.3.1.1) 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 3.3.1.5), 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 3.4.1, 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.

3.4.3 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 3.3.2.2) 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 [129]. 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 [234]; notably, like USEtox, SLERA adopts mid-point indicators expressed in terms of the Potentially Affected Fraction (PAF) of species. As noted above, this 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 [256]. In addition to inventory issues discussed in Section 3.4.1, SLERA has two major limitations. At present, it only considers emissions to fresh water (Table 3.1); 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 the ecosystems.

Environmental Irradiation, discussed in Section 3.3.2.1, 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 3.4.1) and in the effect analysis, which uses site-dependent models (whose limitations have been discussed in Section 3.4.2) and

Environmental Increments (EI) instead of PAF to quantify potential impacts on the 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 the 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.

3.5 Conclusions

This Chapter 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; this is discussed in Chapter 4. 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 omission in the inventory of emissions from radioactive waste disposed of in a final repository. 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 general 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.

Chapter 4.
A framework and two practical methodologies for assessing radiological impacts in LCA

Radioactive materials are routinely released by several industrial processes, but their impacts are rarely included in LCA studies because a standard approach is still lacking. Based on the recommendations of Chapter 3, an overarching framework for incorporating radiological impacts in the impact assessment phase of LCA has been developed in this Thesis, from which two practical methodologies have been derived. UCrad is the first-of-its-kind multimedia compartment model for radionuclides, based on that developed for toxic substances in LCA (USEtox). The Critical Group Methodology (CGM) is inspired by typical Human and Environmental Risk Assessment (HERA) practices, notably by adopting the concept of critical group and Gaussian-type dispersion models. Characterisation factors have been computed for both methodologies, while normalisation factors only for UCrad; they are both included in Appendix A. A detailed analysis of the methodologies has been carried out with the purpose of validating them and investigating the effects of different assumptions and modelling approaches. The analysis includes three types of comparisons: i) UCrad and CGM with a peer-reviewed methodology, namely Human Health Damages, ii) UCrad and CGM for various distances of the critical group, iii) USEtox and CGM applied to toxic substances. Results show that i) UCrad is in better agreement than CGM with Human Health Damages, although both methodologies feature low average deviations; ii) the half-life is a critical parameter resulting in large deviations between CGM and UCrad; iii) the same conclusions for radionuclides also apply to toxic substances. Finally, practical applications of the developed methodologies are discussed: one is immediately apparent and envisages that either is used; the other responds to the need identified by some authors of combining HERA and LCA (which are respectively represented by CGM and UCrad) by proposing their combined use.

4.1 Introduction

As discussed in Chapter 2 the key feature of LCA lies in its holistic perspective, which has made it a central concept for both environmental management in industry and environmental policy-making in public government. For this reason, the LCA methodology must be able to consider and evaluate potential impacts of all main types of pollutants; only in this way is its holistic feature maintained. Nonetheless, as discussed in Chapter 3, to date impacts of ionising radiations have been largely disregarded in LCA studies. Several industrial processes (e.g. nuclear, coal, oil and gas, fertiliser and building industries) routinely release radionuclides in the form of air and waterborne streams; whilst others (mainly nuclear industry, hospitals and defence departments) generate radioactive solid waste, which are either disposed of in near-surface landfills (after inertisation), or stored awaiting construction of special repositories. Radioactive solid waste will eventually (in the order of tens of thousands of years) deteriorate releasing the stored radioactivity into the environment. Amongst the many industries, indeed the nuclear is one of the main sources, both for its scale and the materials it consumes and produces. From this reasoning arises the need to develop a framework which allows LCA practitioners to integrate in the impact assessment phase the effects of radioactive emissions.

A small number of methodologies for radiological impact assessment are currently available for use in the Life Cycle Impact Assessment (LCIA) phase. These have been either developed exclusively for LCA applications [199], [211], [220], [232], or can be adapted from standard assessment procedures used in other fields [204], [227]. Amongst those, the Human Health Damages (HHD) developed by Frischknecht and co-workers [211] is to date the only methodology included in LCA impact methods, e.g. in CML [125], Recipe [124], Eco-indicator 99 [255], Impact 2002+ [128]. A detailed review of these methodologies has been presented in Chapter 3; it concluded that none of the methodologies currently available is mature and holistic enough to be included as standard procedure within the LCIA. Besides analysing advantages and disadvantages, similarities and differences of a number of methodologies, the review also identified the essential features of a methodology for radiological impact assessment, these are the ability to include both direct discharges and emissions from a geological repository, being site-independent and producing average estimates.

In recognition of the review findings, a general framework for assessing the impact of radioactive emissions on human beings has been conceived. This general framework lays the foundations on which two conceptually very different methodologies have been developed. UCrad is a multimedia compartment-type model; it is based on the approach first proposed by Donald Mackay and widely used for toxicity characterisation in LCA [175], e.g. USEtox [134]. To date, no compartment-type model has ever been implemented for radionuclides; thus UCrad constitutes the first-of-its-kind multimedia model. The Critical Group Methodology (CGM), on the other hand, has been developed with the purpose of providing a basis for analysing the performance of a compartment-type model on radionuclides. It has been adapted from typical Human and Environmental Risk Assessment (HERA) practices and makes use of the concept of the "critical group" and Gaussian plume-type dispersion models; this makes the methodology site-dependent, but does not necessarily mean that it produces worst case estimates (this is discussed in Section 4.2.1.1). It must be noted that the practice of adapting a risk assessment methodology to the Life Cycle Impact Assessment (LCIA) phase does not constitute an element of innovation: Human and Environmental Toxicity Potential categories such as USEtox represent a notable example.

CGM and UCrad are thus representative of two different cultures: HERA and LCA. HERA quantifies the actual, absolute risks to humans and the environment associated with release of pollutants from a specific process whose location is defined. The assessment is carried out on a regional scale, usually with the focus on few selected substances of relevance to the process under study. Conversely, LCA assesses average impacts, linked with the choice of a Functional Unit, of a very large number of pollutants (all those for which characterisation factors have been developed) released over the life cycle of a product (i.e. good or service). As different steps or processes of a product's life cycle take place, usually, over many locations around the world, LCA focuses on global (rather than regional) scales. For these reasons – and as noted by some authors [257]–[259] – HERA and LCA may lead to contrasting results when applied to the same process. De Haes and colleagues [260] concluded that, although the two tools feature a number of differences, the crucial and insurmountable one is represented by the use of the Functional Unit, which differentiate relative impacts of LCA from absolute ones of HERA. This Chapter does not mean to discuss further differences and similarities of these tools – this is the topic of several articles including [260]–[265]; rather the primary aim is to evaluate the effects of

implementing a risk assessment-type methodology within the LCA framework, by comparing it with a typical approach largely used in LCIA. The basis of the discussion lies in the LCA environment as both methodologies are meant to be used along with the concept of the Functional Unit. The study will also serve to validate the compartment-type methodology UCrad through comparison of its characterisation factors with a proven methodology largely used in the nuclear industry. Finally, at the end of the Chapter a brief proposal for a combined use of both methodologies – and in a broader sense of HERA and LCA – is provided.

This Chapter is structured as follows: Section 4.2 introduces in details the general framework and the two practical methodologies for radiological impact assessment; Section 4.3 includes three types of comparisons: first, CGM and UCrad are compared with the Human Health Damages (HHD) methodology; second, CGM and UCrad are compared for several distances of the critical Group, but also as a function of radionuclides' half-life; third, the same comparison is performed for three key toxic substances with the aim of validating the results obtained for radionuclides. Section 4.4 follows with a detailed discussion of the main findings, including qualitative and quantitative analyses of the results as well as a proposal for practical applications of the methodologies. Finally, Section 4.5 concludes the Chapter by summarising the key points.

4.2 The general framework and two practical methodologies

The framework for radiological impact assessment has been conceived in this Thesis with two main purposes in mind: establish a standard, widely agreed approach for integrating radiological impacts in LCA, and generate characterisation factors for all type of radioactive emissions to be included as a new impact category.

A simple outline of the framework is shown in Figure 4.1, which reports the four phases that make up the framework, along with their respective output and metric.

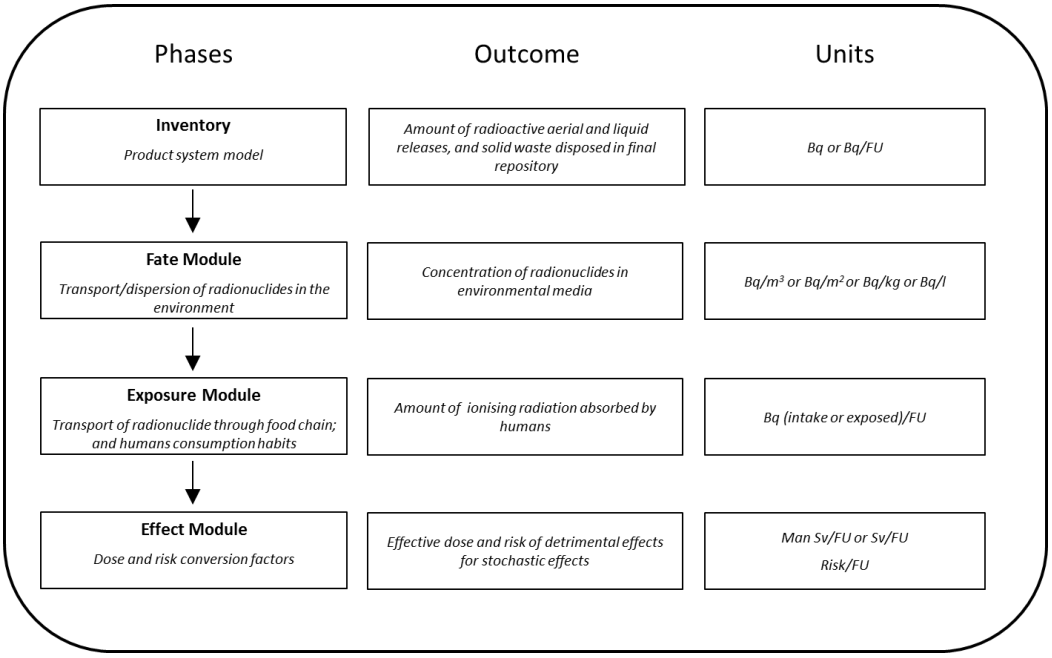

Figure 4.1 – Overview of the framework for radiological impact assessment. FU: Functional Unit.

The four phases of the framework allow estimating the impact of a product as a function of its emissions, included in the inventory. However, in the context of impact factors development, the inventory phase must be excluded as impact factors are obtained per unit of emission. Therefore, like most general models for human impact assessment, this framework consists of three main modules: a fate module, an exposure (or intake) module and an effect module (dose and risk calculation). The purpose of the fate module (discussed in Section 4.2.1) is to model transport and dispersion of radionuclides from source of release to estimate their concentrations in environmental media. For this purpose, a number of approaches may be used (e.g. numerical, analytical, box-type) depending on the level of accuracy required and the amount of information available; Section 4.2.1 distinguishes between the approach adopted by CGM (Section 4.2.1.1) and UCrad (Section 4.2.1.2). The exposure module (Section 4.2.2) directly follows the fate module and uses environmental concentrations to estimate the amount of ionising radiation absorbed by human beings according to specific habits and behaviours. Finally, the effect module (Section 4.2.3) consists of two steps, one of which being optional: firstly, the amount of radioactive radiations to which individuals are exposed (expressed in terms of Becquerels, Bq) is converted into an effective dose measured in Sieverts (Sv), which is the SI unit that considers both the type of radiation and the human tissue involved in the process; and then, the dose may be converted into a risk metric for detrimental effects (the optional step).

As noted in Section 4.1, from this general framework two different methodologies have been derived. They share the same exposure and effect modules, but differentiate in the fate module; specifically, in the type of model used to simulate transport and dispersion mechanisms of radionuclides in the environment. The exposure and effect module, by contrast, constitute standard approaches relying on sets of parameters and assumptions widely accepted in the radiological protection community.

4.2.1 Fate module

4.2.1.1 Critical Group Methodology

The Critical Group Methodology (CGM) adopts the concept of "critical group", developed for risk assessment purposes and defined as "the individual members of a population who can realistically be expected to receive the highest dose due to their lifestyle, location and habits" [185], [205]. This concept is also equivalent to the representative person, defined by the International Commission on Radiological Protection (ICRP) [179], [266]. Adoption of the critical group concept has one considerable consequence: results of the assessment are location-dependent, that is they depend on where (i.e. how far) the critical group is located with respect to the source of release. It must be noted that, unlike in Risk Assessment studies, the critical group location used in CGM does not have to represent a worst-case scenario; rather it allows selecting a distance at which radiological impacts are assessed. With respect to the fate analysis, CGM makes use of analytical models – such as the Gaussian plume model for atmospheric emissions – to simulate the dispersion and transport of radionuclides in the environment. Furthermore, the methodology includes both radioactive direct discharges (i.e. gases to atmosphere or liquids to freshwater bodies or sea), and radioactive solid waste disposed in a final repository. Their impacts, however, occur on very different time scales (days/weeks/months vs tens of thousands of years), hence any comparison between them must be drawn with caution.

Two reports constitute the foundations of CGM's fate module: the IAEA framework for assessing the impact of routine discharges from nuclear plants is used for direct discharges [176]; whilst the generic Post-Closure Performance Assessment (PCSA) [267] developed by the Radioactive Waste Management Ltd. (RWM) under contract to the UK Nuclear Decommissioning Authority (NDA) is used for assessing potential impacts of radionuclides contained in nuclear solid wastes disposed in a generic design of a Geological Disposal Facility (GDF).

Direct discharges

The IAEA framework provides a series of simple screening techniques conceived to demonstrate compliance with environmental standards or other reference levels [176]. These models are aimed at estimating not only environmental concentrations (fate module), but also doses received by human beings (exposure and effect modules) as a consequence of routine releases of radioactive materials. As accurately assessing doses can be a complex and time-consuming process, the IAEA conceived two levels of screening. They make part of a structured, iterative approach for increasing the complexity of modelling as predicted doses approach or exceed a reference level, which is related to dose limiting criteria specified by the regulatory authority.

The first level of screening is a very simple assessment based on the very conservative assumption that members of the public are exposed to radioactive material at the point of discharge – this is referred to as "no dilution model". The second stage envisages using a simple generic environmental model that accounts for the dispersion of radioactive materials in a generic environment whose features are based on a set of assumptions. If estimated doses exceed reference levels, the IAEA recommends to firstly verify the applicability of those assumptions to the specific environment in question, and eventually to carry out a full site-specific assessment. Notably the screening level II offers a good compromise between accuracy and data requirements; and for this reason, it constitutes the basis of CGM's fate module for direct discharges. Figure 4.2 reports an outline of the fate module approach (adapted from the overall IAEA framework).

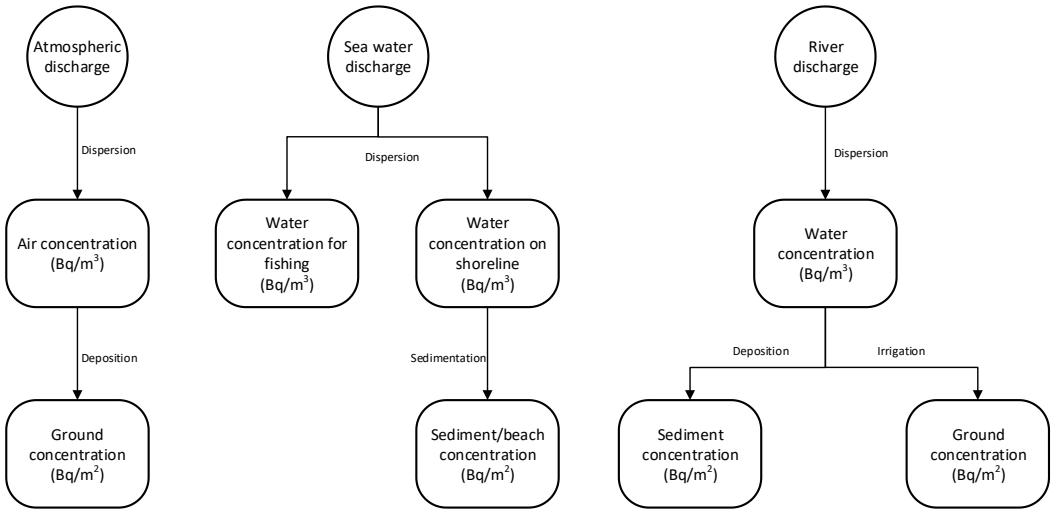

Figure 4.2 – Overview of the CGM's fate analysis approach, adapted from IAEA [176]

The IAEA framework includes three types of direct discharge pathways: atmospheric, to surface (both fresh and sea) water and to sewage. Nevertheless, CGM only considers two of those; discharge to sewage is currently disregarded as it is a much less common discharge pathway.

For atmospheric discharges, the Gaussian plume model [173], [174] is employed to assess the dispersion of long-term releases. Such model has been extensively used and is widely accepted in radiological assessment practices. The Gaussian plume model estimates air concentration of pollutants undergoing downwind transport (advection) and mixing processes (turbulence diffusion) at a specific distance from the release source. The IAEA states that such model should not be used for distances longer than 20 km because of high uncertainties linked with transport over long distances. For this reason, the IAEA implementation of the Gaussian plume model does not consider radioactive decay of radionuclides: limiting the assessment range to 20 km means that radionuclides travel time is very short and consequently radionuclides decay is practically negligible. However, impacts of atmospheric discharges are quantified for distances well over 20 km in the CGM methodology to allow comparison with UCrad (Section 4.3.3); for this purpose, radionuclides decay has been incorporated in the Gaussian plume model. Alongside distance, the air concentration depends on a number of other factors; amongst the most important there are: geometric mean of wind speed at height of release, height of release and presence of buildings near the release source.

With respect to liquid discharges, an analytical solution to advection-diffusion equations in steady-state uniform-flow conditions is provided by the IAEA. Amongst several different water bodies available in the IAEA report, CGM considers discharges only to two water bodies: rivers (as representative of freshwater bodies) and coastal waters. Most discharges in the nuclear industry, in fact, happen to fall in one of those categories. The concentration of radionuclides in water is dependent on the distance from the release source and some features of the water body such as: width, depth and net freshwater velocity for rivers; and depth, distance from release point to shoreline and average coastal current for sea emissions. The coastal water model estimates two different concentrations of radionuclides in water: one of interest for fishing purposes, and another related to activity along the shoreline. The river model, in turn, estimates only one

concentration, the total concentration of radionuclides in water, with the general assumption of perfect vertical and horizontal mixing.

The air and water concentrations constitute the two pillars of the IAEA framework; from them all other concentrations can be derived. Notably, from the air concentration ground superficial activity due to wet and dry deposition processes is estimated; whilst from the water concentration superficial activity in shore/beach sediment due to sedimentation and ground concentration due to irrigation are obtained. Concentration of radionuclides in plant, animals and fish is herewith not considered part of the fate module; rather it constitutes the starting point of the exposure module.

Table 1 reports the default parameters used in CGM fate models. The distance of the critical group from the release source is not reported in the table as CGM impact factors have been conceived to be dependent upon it. Most of the parameters reported are recommended in the IAEA report; for those not recommended, specific values based on some arbitrary assumptions have been set. For instance, data from Sellafield site (the UK industrial complex whose main purpose involves reprocessing of spent nuclear fuels) have been used for estimating the height of atmospheric discharges.

Table 4.1 – Parameters used in CGM fate module

Emission	Parameter	Value	Comments
Air	Height of discharge (m)	100.00	*Based on Sellafield THORP*[8] *stack height*
	Fraction of time wind blowing (-)	0.25	*IAEA recommended value* [176]
	Wind speed (geometric mean) (m/s)	2.00	*IAEA recommended value* [176]
Sea water	Water depth at discharge (m)	15.00	*Based on Sellafield sea data*[9]
	Distance from shoreline (m)	2100.00	*Based on Sellafield liquid discharges*[10]
	Mean coastal current (m/s)	0.10	*IAEA recommended value*
Rivers	Width (m)	21.00	*Medium-sized river*[11]
	Depth (m)	0.34	*Calculated from IAEA table III* [176]
	Net flow rate (m^3/s)	0.70	*Calculated from IAEA table III* [176]
	Flow rate (m^3/s)	5.00	*Calculated from IAEA table III* [176]

[8] *The Thermal Oxide Reprocessing Plant (THORP) is Sellafield flagship plant for reprocessing UK and international spent nuclear fuels*.

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

[10] *Obtained from Radioactivity in Food and the Environment report* [356].

[11] *As Sellafield site does not have significant liquid emissions to rivers, width of a medium-sized river has been chosen.*

Solid waste

Radioactive solid wastes are classified in a number of categories depending on their characteristics (see Chapter 1). The generally accepted classification in the UK envisages that radioactive wastes are grouped according to their content into four categories: High-Level Waste (HLW), Intermediate-Level Waste (ILW), Low-Level Waste (LLW) and Very Low-Level Waste (VLLW) [48]. There are also other wastes that are not usually included in those categories, rather they are treated separately either because of their peculiar features or source, or because they haven't yet – but may be in the future – been declared as waste. These include Used Nuclear Fuel (UNF) assemblies that are to be disposed of without reprocessing; plutonium (Pu) retrieved from reprocessing of SNF; Highly Enriched Uranium (HEU), with 20% or higher concentration of uranium-235 (U235); and Depleted, Natural and Low Enriched Uranium (DNLEU), that is uranium with concentration of U235 lower, equal or slightly above the natural occurring concentration equal to ~0.71%. Notably, depleted uranium includes enrichment tails (the by-product of the enrichment process, see Chapter 1), whilst low-enriched corresponds to uranium retrieved from reprocessing of SNF, usually with concentrations between 1 and 1.6%.

Nowadays management and disposal of nuclear solid waste constitutes a much debated topic. A standardised, and internationally agreed approach, has not yet been developed and at present many countries haven't yet taken or are re-considering their final decision. The most favoured solution by nuclear countries (such as the UK, US, Sweden, Switzerland) is to dispose of – after required treatment and conditioning processes – HLW, SF, Pu (depending on the fuel cycle), ILW (in some cases only long-lived ones), HEU and DNLEU in Geological Disposal Facilities (GDFs) and V/LLW (and in some cases also short-lived ILWs) in a near surface repository [268].

The lack of an operational GDF in the world and the little – if not absence of – knowledge regarding the potential future behaviour of GDFs means that there are no operational frameworks for assessing the potential impacts of nuclear waste stored in deep underground repositories. For this reason, the solid waste pathway in CGM is based on the reference scenario of a preliminary study of the Radioactive Waste Management Ltd. [267]. VLW and those LLW that are suitable for disposal in near surface repository, however, are not (yet) taken into account in the methodology; the main reason being that no suitable performance assessment could be retrieved for near-surface repositories.

UK Generic Post-Closure Safety Assessment

The generic Post-Closure Safety Assessment (PCSA) [267] forms part of the environmental section of the generic Disposal System Safety Case (DSSC) developed by the RWM as part of its programme to support the implementation of the future UK Geological Disposal Facility (GDF). The PCSA is a generic study of the potential impacts of a GDF after its closure onto a critical group, which in the PCSA report is referred to as "Potentially Exposed Group", is best described in the Biosphere status report [269] and defined on the basis of the recommendations of the IAEA [270]. The main purpose of the PCSA is to illustrate, by example, how a post-closure safety assessment would be carried out at a site to be identified in the future. However, there are some quantitative components in the assessment, which inevitably depend on some fairly arbitrary assumptions as no potential site has yet been identified in the UK.

The main assumptions regarding the PCSA base scenario are reported below:

- The reference or baseline inventory of nuclear waste to be disposed of is based on the Derived Inventory [271]–[273], which reviewed and enhanced the 2007 UK Radioactive Waste Inventory (UKRWI) [274]. A specific conditioning and packaging approach for disposal has been considered for each type of waste; further information are reported in the Derived Inventory and in the RWM's disposability assessment [275].
- Groundwater is the only pathway considered in the quantitative analysis for escape and transport of radionuclides from the GDF. A gas pathway is also considered in the qualitative analysis, but its inclusion in the calculations has been restricted due to high uncertainties associated with its behaviour.
- The GDF layout corresponds to the design for higher strength rock based on the Nirex phased geological repository concept [276] and the Swedish SKB KBSD-3V concept for spent fuel [53]. The GDF design is the result of the combination of two disposal concepts engineered in two separate areas, which would host ILW, LLW and DNLEU in one area, and HLW, SF, HEU, and Pu in another area.
- The total system modelled comprises three subsystems:
    a) The engineered system comprises the excavated vaults and their contents. Its behaviour is modelled through only one parameter which relates to the time taken for failure of the waste container to happen.

b) The geosphere, i.e. the rocks surrounding the GDF and extending up to the surface, is represented by a set of four key parameters: specific discharge through the undisturbed host rock; groundwater travel time from GDF to surface; groundwater mixing flux in overlying rocks; discharge area into which contaminant plume is released at the surface.
c) The biosphere is defined as the surface environment, which behaves as a receptor for radioactive releases. It is used to estimate doses, and consequently risks, incurred by human beings as a consequence of the release. A "Reference Biosphere" is used to model the behaviour of the Potentially Exposed Group, as described in the NDA's Biosphere status report [269]. An order-of-magnitude area of 10 km^2 is considered for the assessment.

RWM Ltd. team recognised that even after an extensive site investigation, values for the key parameters for the engineered system and the geosphere will not be known precisely. Therefore, a log-triangular probability density function is given to each of them with different lower and upper bound depending on the specific parameters. A sensitivity analysis has then been carried out to understand how these parameters affect the overall results of the study. With respect to the base scenario, central value of each parameter is considered.

Results of the quantitative analysis display total mean radiological risk against time, showing contribution of different waste type and different radionuclide within a single waste type (Figure 4.3). Notably, only the radionuclides (usually no more than ten) showing the highest risk are included. It must be noted that results are only given in terms of risks, with the fate and exposure calculations not reported explicitly, but aggregated together with the effect module. Because the fate model could not be retrieved, for the solid waste category the PCSA model replaces all three phases of the assessment framework (as reported in Figure 4.1).

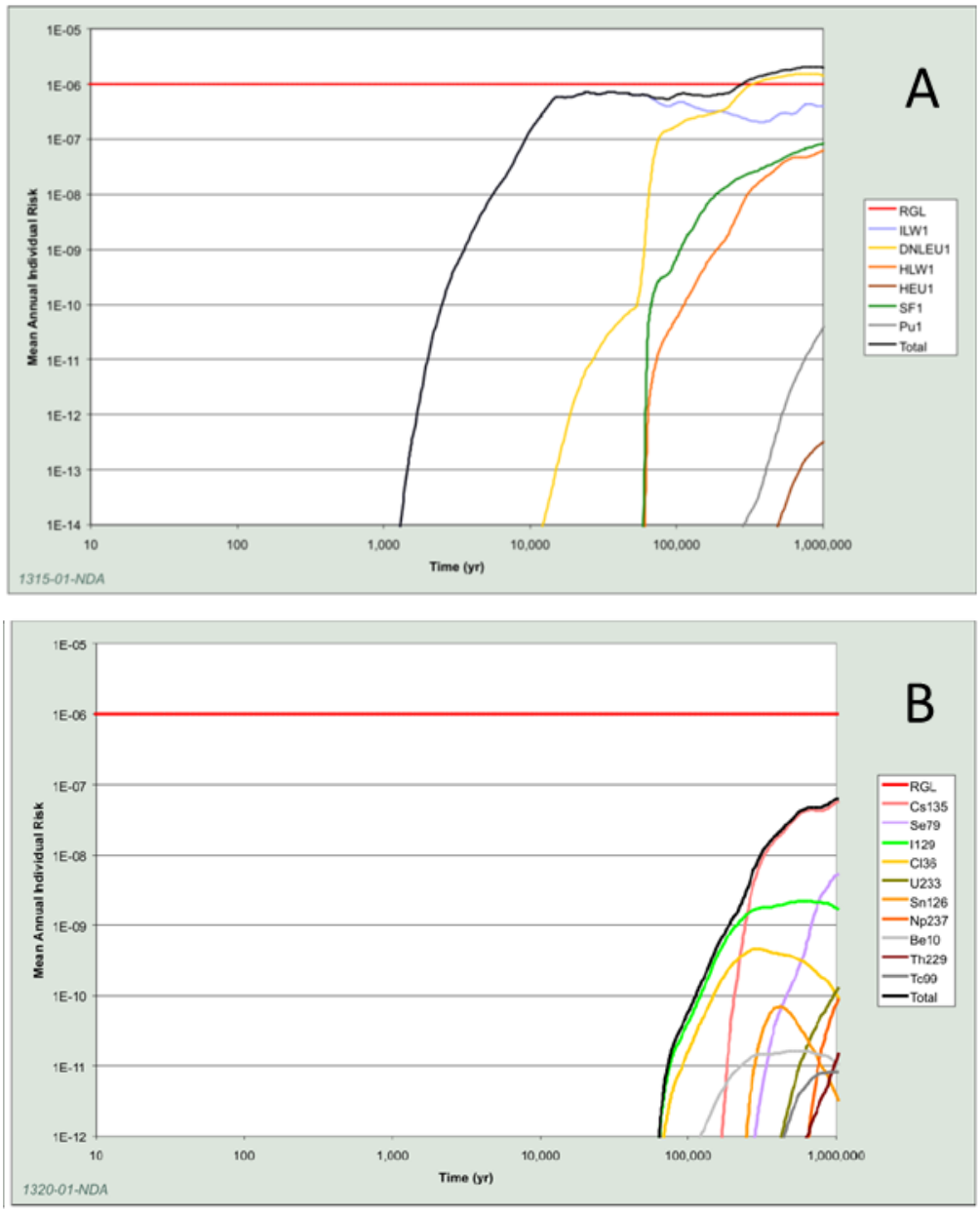

Figure 4.3 – Reference case radiological risk against time: for different waste type (A) and for key radionuclides for ILW and LLW (B) [267]. RGL: Risk Guidance Level.[12]

From the PCSA results, a set of characterisation factor for nuclear solid waste in terms of risk per Bq of each radionuclide disposed in the GDF has been calculated according to the following equation (4.1):

$$CF_{w,i} = \frac{Rm_i}{Q_{w,i}} \qquad \text{eq. 4.1}$$

[12] The Risk Guidance Level is defined in the UK by the Guidance on Requirements for Authorisation [357] which encompass management, radiological and technical aspects of the safety case for a Geological Disposal Facility.

Where:

- $CF_{w,i}$ is the characterisation factor for radionuclide *i* in waste type w stored in a GDF (Risk/Bq);
- Rm_i is the risk time integral over a period of approximately one million years[13] for radionuclide *i* (risk);
- $Q_{w,i}$ is the amount of radionuclide *i* in waste type *w* stored in a GDF (Bq).

The $Q_{w,I}$ values have been obtained from the Derived Inventory. The timeframe for which the integral has been computed has been fixed for each waste type and ranges from the time at which the total risk starts to rise (e.g. around 80.000 years for I-LLW in Figure 4.3) to the end time of the assessment, that is one million years.

4.2.1.2 UCrad: a compartment-type methodology

Compartment-type models (also known as MacKay models) are multimedia environmental models largely used in environmental chemistry to simulate the behaviour and predict the fate of organic chemicals released in the environment. They were conceived by Donald MacKay with the aim of laying the groundwork for the development of remedial and proactive strategies, and sustainable approaches to industrial processes [175].

MacKay's approach consists in dividing the environment in a number of interlinked compartments (e.g. air, fresh water, sea water, agricultural soil, etc.) and spatial scales (e.g. regional, continental and global). Exchange of substances, and pollutants, occurs among different compartments within the same spatial scale and among same compartments belonging to different spatial scales. Partitioning and advective/diffusive mechanisms govern exchange processes. Notably, partition coefficients are indeed the key parameters in MacKay-type models; they represent how substances segregate at equilibrium among different environmental media, and different phases within the same medium. Many numerical relations have been developed to estimate substance-specific

[13] The time over which the integral is resolved corresponds to the difference between the maximum time at which risks are estimated in the PCSA (1 million years) and the earliest time for all radionuclides at which radiological risks starts to rise. This corresponds approximately to 1 million years.

partition coefficients, if these are not available in the literature; the majority, however, has been specifically conceived for organic chemicals, and none exists for radionuclides.

To date, several multimedia models have been developed and operationalised for assessing the potential impact of toxic substances within LCA, e.g. USES-LCA [277], IMPACT 2002 [278], Eco-Indicator 99 [279], CalTOX [280], USEtox [134]. All these methodologies employ a MacKay-type approach in their fate analysis; specifically, a level III model, which is defined as an open system in steady state with continuous compartment-specific emissions, transformation processes and active transport [175]. Amongst the many models developed, USEtox is the most widely used in LCA studies. In a recent article focusing on Natural Occurring Radioactive Materials (NORM) impacts, USEtox was found to be the most appropriate for fate modelling of radioactive materials [281]. USEtox was developed under the United Nations Environment Program (UNEP) – Society for Environmental Toxicology and Chemistry (SETAC) Life Cycle Initiative with the aim of carrying out a comprehensive comparison of life cycle impact assessment toxicity characterisation models. The result of such comparison was the development of a scientific consensus model that provided recommended LCIA characterisation factors for more than 1,000 chemicals. The USEtox model mainly focuses on organic substances; inorganic substances characterisation factors are flagged as interim, i.e. to be used with caution.

UCrad is a novel compartment-type methodology for radionuclides that has been developed with this Thesis. USEtox represents the foundations of UCrad. Essentially, the development of UCrad consisted in modifying the USEtox consensus model to include key features (e.g. bioaccumulation factors) and mechanisms (e.g. radioactive decay) of radionuclides, and to account for differences in the behaviour of organic chemicals and radionuclides. Like USEtox, UCrad is implemented in the Microsoft Excel environment and consists of a database and a calculation package. An outline of UCrad compartment model is shown in Figure 4.4.

The main features of UCrad along with them main differences with respect to USEtox are reported in the following sub-Sections.

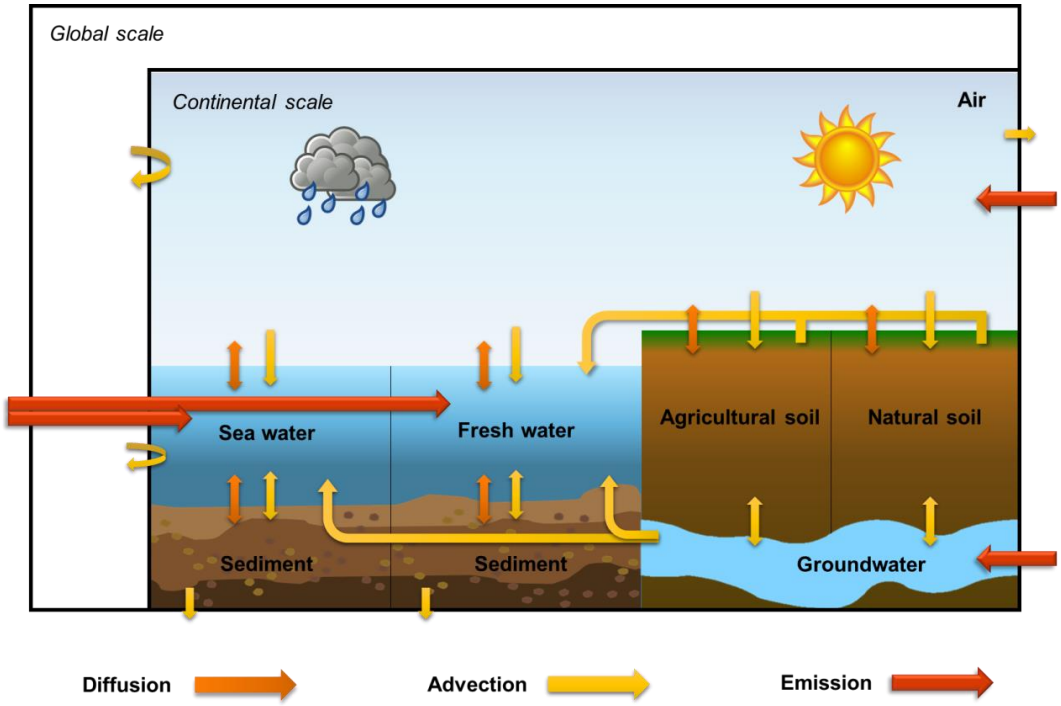

Figure 4.4 – Compartment setup of UCrad fate module.

Compartments

Eight environmental compartments are distinguished in UCrad: air, fresh and sea water, natural and agricultural soil, fresh water and marine Sediments and groundwater. Notably, USEtox only includes the first five: freshwater and marine sediments have been added with the consideration that radionuclides accumulating in those compartments, as opposed to organic chemicals, can affect human health. The sediment compartments are solely linked to the respective water compartment, and the exchange process between those is governed by sedimentation/re-suspension and adsorption/desorption mechanisms. The groundwater compartment, on the other hand, has been added with the aim of considering the impacts of radioactive emissions arising from wastes stored in a Geological Disposal Facility (GDF). As a matter of fact, most of the performance assessment studies on GDFs – e.g. those developed in Sweden [282], Switzerland [283], UK [267], France [284] – recognize groundwater as the main pathway through which radionuclides can reach the biosphere. Other potential pathways such as gas permeating through the rocks or human intrusion through wells are either poorly studied or are consequence of probabilistic events, which fall outside the scope of conventional LCA (see Chapter 3). The groundwater compartment has been adapted from the GLOBOX model [198]: it is fed by water leaching from natural and agricultural soil and is linked to the

ocean and freshwater (through ground flows) and to natural and agricultural soils (through irrigation).

Scales

UCrad uses a nested compartmental model comprising two scales: continental and global. The indoor and urban scales that feature in USEtox have been removed for the sake of simplicity as radioactive emissions are highly unlikely to occur in densely inhabited areas.

Intramedium transport

USEtox – and generally multimedia fate models – rely on two main mechanisms for intramedium transport: partitioning and advection/diffusion. UCrad employs the same advective/diffusive equations used in USEtox. However, with respect to partitioning coefficients USEtox has been conceived to use firstly substance specific factors and then, when those are not available, predictive equations. Because predictive equations have been primarily developed for organic chemicals, their usage for inorganics – let alone radionuclides – is not recommended. For this reason, UCrad only uses substance-specific partition coefficients gathered from a number of reliable sources. The chemical database supporting UCrad is discussed in Section 4.2.4.

Gases and particles

The air-water partition coefficient (K_{aw}) represents a tricky parameter in UCrad as there are no available values reported in literature for radionuclides. For this reason – and on the basis of how inorganic substances have been modelled in USEtox – a representative value of 1E -20 (Pa m^3 mol^{-1}) has been given to the majority of radionuclides. This value represents a negligible volatility, meaning that those nuclides mainly behave as particles. Few radionuclides, e.g. noble gases and few others, feature non-negligible K_{aw}s; these have been obtained on the basis of the Henry constants of the pure substance as reported by Sander [285].

Carbon-14 and tritium

Carbon-14 (C14) and tritium (H3) represent two special cases. As most discharges involving those radionuclides happen in the form of carbon dioxide and water vapour, their fate has been modelled on the basis of these species – the same approach is used by the IAEA [176].

Radioactive decay chain

UCrad treats the radioactive decay of nuclides as a removal process from the environmental compartments, in the same way as USEtox treats degradation processes by micro-organisms or the escape of atmospheric pollutants to the stratosphere. The products of the decay (known as daughters) are not accounted for in the model; this means that UCrad characterisation factors represent exclusively the impacts of the specific radionuclides that are released, and that impacts of nuclides in the decay chain are neglected. The way in which UCrad handles the decay chain represents a significant limitation and one of the key points for its future development.

Geological Disposal Facility

One of the crucial features of UCrad is its ability to evaluate the impact of solid nuclear waste disposed of in a deep geological repository. As it is acknowledged that eventually a failure in the engineered system will occur, the GDF's main purpose is twofold: contain radionuclides for as long as possible and, when a failure occurs, delay the escape of radionuclides by increasing their travel time to the biosphere. For this reason GDFs are constructed several hundred meters underground: the layers of rocks that separate the GDF from the biosphere act as retardation factor. Because groundwater is the main pathway through which radionuclide can escape the engineered system, GDF's impact in UCrad has been implemented by inclusion of the groundwater compartment (see Figure 4.4), which however does not take into account the above mentioned retardation. Mackay-type models are equilibrium models; this means that a time factor is indeed taken into account, but it is not something that can be set arbitrarily: the time considered by Mackay-type models is the time needed to reach equilibrium. For this reason, in UCrad the retardation factor is not directly applied to the model, rather it is to the emissions. GDF emission values to groundwater used in UCrad do not correspond to the radioactive flux leaving the GDF, namely "near-field flux", rather they correspond to the so-called "far-field flux", or the flux leaving the geosphere and entering the biosphere system. This represents the flux of radionuclides that have escaped the repository, have travelled through several layers of rocks and have finally reached the biosphere; therefore, it incorporates the retardation factor. The far-field flux is obtained from the generic Post-Closure Safety Assessment report [267], where it is assumed to feed into the biosphere, a complex mathematical model simulating dispersion of radionuclides in the surface environment [269]. Figure 4.5 provides a schematic representation of the approach used

for modelling emissions from GDF in UCrad compared with the PCSA model (discussed in Section 4.2.1.1).

The fact that UCrad approach for emissions from a Geological Disposal Facility is based on the PCSA has one important consequence, that is that it relies (and is dependent) upon all the assumptions that have been made on the GDF in the PCSA (e.g. type of waste containers, host rock, etc.).

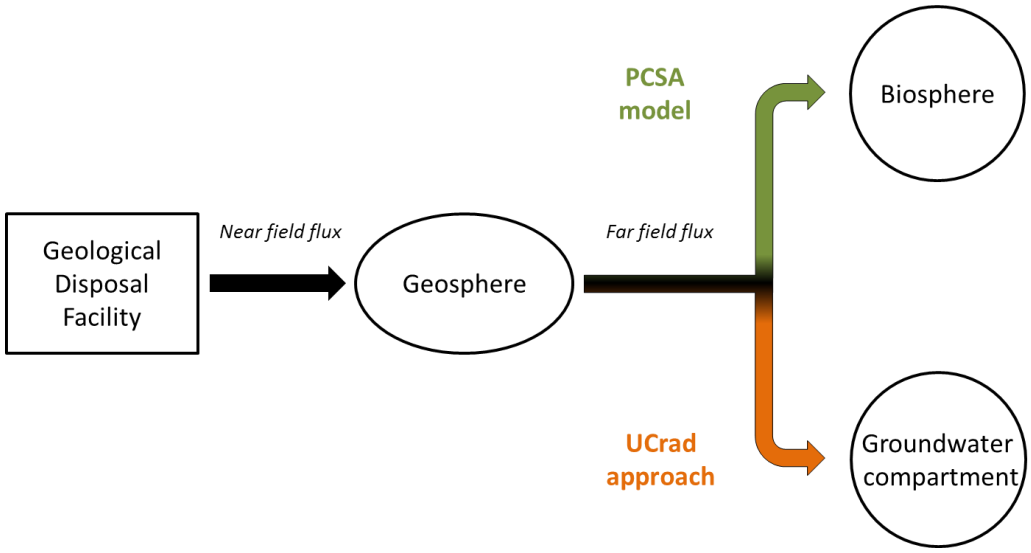

Figure 4.5 – Schematic representation of UCrad approach for GDF characterisation factors compared with the PCSA base model

4.2.2 Exposure module

The exposure module represents the second step of the framework (see Figure 4.1). It quantifies the amount of ionising radiations absorbed by humans as a consequence of an increase in the environmental concentrations of radionuclides following a radioactive discharge (estimated by the fate module). The exposure module does not depend on the approach adopted to calculate the environmental concentrations and thus it is applied identically to CGM and UCrad. The location of the pollutant in the environment and its physical state affect the pathway through which the exposure may occur. Two main categories of pathways may be identified: external and internal. The intake of radionuclides through ingestion and inhalation contributes to the internal pathway; whilst airborne and deposited radionuclides contribute to the external pathway, chiefly by means of gamma radiation. Alpha and beta radiations decay on very short distances, thus their contribution to the external pathways is negligible. The ingestion of radionuclides occurs because, once emitted and dispersed into the environment, radioactive substances

enter the food chain. For instance, wet and dry depositions, and soil contamination contribute to radionuclides transfer to crops and cattle, which in turn are consumed by humans. This means that the exposure module is also equipped to estimate radionuclides concentrations in human food, e.g. meat, dairy products and below and above ground crops, by means of bio-accumulation and bio-transfer factors. Finally, usage factors are employed to establish consumption patterns of contaminated food and water; whilst exposure factors are used to estimate the length of time that an individual may be exposed to a contaminated environment. Usage factors have been taken from USEtox [286], whilst exposure factors have been adapted from the IAEA report [176]. Furthermore, two nuclides, namely H3 and C14, represent special cases requiring a specific treatment: the IAEA's Specific Activity Models (SAMs) are used for this purpose [176]. The underlying reason is that both H3 and C14 can be incorporated into a great variety of different chemical compounds within the human body; hence assessment of their potential impact would be too complex to be incorporated in generic risk assessment studies, let alone LCIA methodologies. A specific activity approach is based on the assumption that a steady state equilibrium has been reached, and thus the ratio between a radioactive nuclide and its stable form is fixed; from this, the radionuclide's concentration in the food chain and human body may be estimated. The final outcome of the exposure module is the amount of radionuclides taken in through ingestion and inhalation, and the time-weighted concentration of radionuclides to which human beings are exposed.

4.2.3 Effect module

The effect module (see Figure 4.1) assesses the consequences onto human beings of radionuclides intake and exposure, and as the exposure module it is applied identically in CGM and UCrad. This is achieved in a two-step process: first, the effective dose is calculated; and then, this is translated into a risk metric for detrimental effects. As discussed in Chapter 3, the ICRP defines three types of doses: absorbed, equivalent and effective [179]. The absorbed dose refers to the amount of energy imparted to matter per unit mass by ionizing radiations. From the absorbed dose, the equivalent dose is obtained by considering the biological effectiveness of radiation, which depends on the radiation type and energy. Eventually, the effective dose is derived from the equivalent dose by accounting for the biological tissue involved in the process; its metric unit is Sieverts (Sv). In the effect module, the effective dose is calculated by means of established conversion

factors, which have been obtained from two reliable sources as explained in Section 4.2.4. With respect to the risk metric, as noted in Section 3.1 ionising radiations can result into two different effects on human beings: deterministic and stochastic [179]. Deterministic effects are mainly due to killing/malfunctions of cells; they are nil below a specific threshold, and increase linearly above it. Stochastic effects, on the other hand, accounts for modifications of cells, which may cause cancers and heritable effects. They occur for low doses (less than 100 mSv), have no threshold and their likelihood increases linearly with the dose – this is the so-called – and much debated [186] – Linear Non-Threshold (LNT) model. For the purpose of this framework, only stochastic effects are considered, the main reason being that only routine releases, and thus relatively small doses, are taken into account. The nominal risk coefficient for stochastic effects, obtained from the 2007 recommendations of the ICRP, is equal to 5.5 10^{-2} Sv^{-1} , with cancer (both fatal and non-fatal) being the only effect considered [179]. Hereditary effects – which have little probability of occurring – have not been considered to achieve consistency with other LCIA methodologies.

4.2.4 Database of radionuclide properties

A database containing radionuclides' properties has been built on a standalone spreadsheet to support both methodologies. The database reports physico-chemical properties and dose/risk conversion factors for more than 100 radionuclides. Specifically, the list of radionuclides included has been taken from the IAEA report [176] and integrated with a number of radionuclides of crucial importance for impact assessment of GDF, i.e. radionuclides with very high half-lives usually not directly discharged by human activities. Experimental have been preferred over estimated data, with the sole exception of the water-sediment distribution factor that has been estimated as being one tenth of the water-suspended sediment factor as suggested by the IAEA [176]. As there is still incomplete knowledge of radionuclides behaviour in the environment, data for a number of radionuclides is still totally or partially missing. For this purpose, the IAEA suggests the use of analogues to provide relevant information when data are missing. The IAEA states that there are three types of analogues that may be used for derivation of values: isotopes, elements and species. Except for molecular weights and half-lives, isotopes analogues are much used as most data (e.g. bio-transfer factors) refer to elements rather than specific isotopes. In few cases, though, data for specific elements is missing altogether; when this occurs, elements analogues represent a good estimate [176].

Furthermore, as several authoritative sources containing radionuclide properties databases have been published, a hierarchical approach for data selection has been applied. The main sources for parameters used in UCrad and CGM ranked according to the hierarchical approach are reported in Table 4.2. Parameters not listed in Table 4.2 have been taken from USEtox.

Table 4.2 – Hierarchical approach and parameters sources used in CGM and UCrad

Radionuclides half-life
Two main sources that complement each other have been used: • Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment [176]; and • The UK Radioactive Waste Inventory main report [287]. As data were missing for three noble gas isotopes (Kr85, Rn222 and Xe133), the IAEA live chart for nuclides has been used [288].
Henry's constant
For most radionuclides a negligible Henry's constant (1E-20) in Pa m^3 mol^{-1}, representing an imperceptible volatility, has been set. However, for noble gases and few other radionuclides the Sander's compilation of Henry's constant has been used [285].
Deposition velocity
The deposition velocity reference value has been taken from the IAEA report [176]. It has been set to 1000 m/d for all substances except for non-reactive gases (i.e. noble gases) where a nil value has been set to indicate a negligible tendency to deposition.
Dose conversion factors
Dose conversion factors have been obtained from two main sources dealing respectively with internal and external exposure: • The IAEA "Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards"[167]) is used for internal irradiation conversion factors. • The U.S. Environment Protection Agency (EPA) federal guidance report [289] is adopted for the external irradiation pathway.
Partition coefficients – Soil
1. IAEA's Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment [288] 2. US-EPA's Partition coefficients for metals in surface water, soil, and waste [290] 3. User's Manual for RESRAD Version 6 [291] 4. Development of Comparative Toxicity Potentials of 14 cationic metals in freshwater [292] 5. Elements analogues
Partition coefficients – Fresh water sediments
1. IAEA's Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment [288] 2. The Methodology for Assessing the Radiological Consequences of Routine Releases of Radionuclides to the Environment Used in PC-CREAM 08 [207] 3. US-EPA's Partition coefficients for metals in surface water, soil, and waste [290] 4. Development of Comparative Toxicity Potentials of 14 cationic metals in freshwater [292] 5. Elements analogues

Partition coefficients – Sea water sediments

1. IAEA's Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment [293]
2. Element analogues

Bio-accumulation factors – Above and below ground produce

1. IAEA's Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment [288]
2. US-EPA's Constituent screening for coal combustion wastes constituent screening for coal combustion wastes [294]
3. Accumulation of metals in plants, a contribution to the evaluation of the intervention values and the location-specific risk assessment of contaminated soils [295]
4. Elements analogues

Bio-transfer factors – Milk, meat and fresh water fish

1. IAEA's Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment [288]
2. NCRP's Screening models for releases of radionuclides to atmosphere, surface, water, and ground [166]
3. User's Manual for RESRAD Version 6 [291]
4. Elements analogues

Bio-transfer factors – Sea water fish

1. IAEA's Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment [293]
2. NCRP's Screening models for releases of radionuclides to atmosphere, surface, water, and ground [166]
3. Elements analogues

4.2.5 Characterisation Factors

Characterisation factors obtained from both methodologies are included in Section A.1 of Appendix A. Although the UCrad methodology is in principle able to calculate characterisation factors for discharges in all environmental compartments, only three direct emission categories have been included. These represent the most recurring types of radioactive emissions in the environment, but they are also the emission categories in common to both CGM and UCrad methodologies; these categories are air, fresh water and sea water. River emissions in CGM are taken as representative of the broader freshwater compartment employed in UCrad. The IAEA model, in fact, distinguishes between different freshwater bodies (i.e. river, small and big lakes), whilst UCrad – and generally Mackay models – merges them in one sole emission category. With respect to emissions from a Geological Disposal Facility (GDF), characterisation factors for six waste categories (HLW, I-LLW, SF, HEU, DNLEU and Pu) have been calculated. Direct discharges categories include characterisation factors for 106 and 112 radionuclides in CGM and

UCrad respectively (the six additional radionuclides included in UCrad correspond to those that have been added specifically for emissions from GDF), whilst a maximum of ten characterisation factors for each waste type have been calculated for emissions from GDF.

Characterisation factors have been calculated for two specific cases. UCrad set has been obtained for the default landscape as defined in USEtox version 2.0 [286], [296] and supplemented with additional average data for parameters unique to UCrad (e.g. for the groundwater compartment). By contrast, since CGM methodology characterisation factors depend on the distance between the receptor and the point of release, different sets of characterisation factors can be obtained. Characterisation factors have been expressed by two different units. The two methodologies have been designed to generate characterisation factors expressed in terms of yearly risk per Bq released; in addition, factors have also been expressed in terms of Bq equivalent, obtained by dividing each nuclide by the impact factor of a reference substance emitted to a specific environmental compartment. For air, fresh water and sea water categories, uranium-235 (U235) emitted to air has been chosen as reference; while uranium-238 (U238) in HLW has been chosen for GDF emissions categories. Two reasons led to choosing two different reference substances. Firstly, the use of U235 allowed comparison with the Human Health Damages (HHD) factors; this radionuclide, however, features no characterisation factors for HLW and I-LLW emission categories. Secondly, by using two different reference substances we are also preventing comparisons between direct discharges and solid waste impact factors. Because impacts of direct discharges and solid waste occurs on much different time scales, their comparison must be carefully considered before being operationalised.

4.2.6 Normalisation Factors

Unlike characterisation factors, normalisation factors have only been calculated for UCrad methodology. Distributed sources of emissions do not represent an ideal “terrain” for CGM, and the fact that these are usually aggregated over the whole reference systems or parts of it, complicates things further. Because CGM evaluates radiological impacts based on the distance of the critical group from the source of emissions, each source should be assessed individually – taking in consideration that the relative distance of the critical group changes from one source to another – and results aggregated in a successive phase. However, when inventory data reports distributed emissions in an aggregated form, the application of CGM becomes meaningless: because information on the location of the

emissions source is not available, the methodology requires the assumption that all emissions take place at the same location – something that would produce illogical results.

Normalisation factors have been calculated in terms of the overall emissions and emissions per person of a geographical area based on two different sources, namely the CML EU25+3 [297] and the ILCD EU27 [298], [299]. CML EU25+3 includes radioactive emissions of the 25 countries that were part of the European Union in 2006 complemented by those of Iceland, Norway and Switzerland. The ILCD EU27, on the other hand, combines the countries included in the CML EU25 with Bulgaria and Romania, which entered the Union in 2007. Calculation of the magnitude of emissions per person for both reference systems has been performed according to the European population as reported in the ILCD [298]. Inventories for both sources are reported in Section A.2 of Appendix A.

4.3 Methodologies comparison

This Section presents a detailed analysis of the methodologies developed within this Thesis to highlight how different modelling approaches and assumptions affect the methodologies output. The analysis method is presented in 4.3.1. Section 4.3.2 reports a comparison between CGM, UCrad and Human Health Damages (HHD) with respect to their characterisation and normalisation (only for UCrad and HHD) factors. Section 4.3.3 includes a comprehensive comparison between CGM and UCrad characterisation factors and an analysis of how radionuclides' half-lives affect the methodologies' characterisation factors. Finally, Section 4.3.4 presents a comparison analogous to that performed in Section 4.3.3, but applied to toxic substances, between CGM and USEtox – the methodology on which UCrad is based; the aim is to demonstrate that results of the analysis are not limited to radioactive substances.

4.3.1 Comparison method: log-deviation and distance

Quantitative analysis between methodologies has been performed by comparison of characterisation factors illustrated by means of parity plot graphs, with the set of characterisation factors plotted on the x-axis referred to as reference set. The comparison has been performed for the three emission categories that are common to the methodologies, namely air, fresh and sea water. As noted in Section 4.2, the river category

in CGM is taken as representative of the broader freshwater category in UCrad and HHD methodologies.

Mean Log Deviation (MLD) has been used to quantify the average difference amongst different sets of characterisation factors compared to the reference set. A nil MLD value identifies two sets of characterisation factors featuring on average (i.e. across the whole set) negligible deviation (although single factors may feature high deviations).

For Sections 4.3.3 and 4.3.4 and only for direct discharges characterisation factors, the comparison between methodologies has been performed across multiple distances of the critical group. Because MLDs can be obtained for different distances of the critical group, a further parameter has been calculated; this is the virtual distance that returns an MLD value equal to zero, and hence the best agreement between the methodologies. The virtual distance has been calculated as the root of the function fitting MLD values plotted as a function of the distance of different sets of characterisation factors. Calculation of such distance allows quantitative comparisons to be performed over different emission categories and classes of substances.

4.3.2 CGM/UCrad vs Human Health Damages

4.3.2.1 Characterisation Factors

Figure 4.6 illustrates a comparative analysis between Human Health Damages (HHD), UCrad and CGM characterisation factors. The distance of the critical group in CGM has been set to 1 000 km. This value approximates in terms of equivalent circular radius the extension of the European continent and it is very close to that used in UCrad (but also it is the value that returns the best agreement amongst those investigated in Section 4.3.3). Two sets of HHD characterisation factors are reported; they represent two different cultural perspective, namely Individualist (I) and Egalitarian/Hierarchist (E/H), as defined by the cultural theory [219]. They are expressed either in terms of damages to human health (Disability Adjusted Life Years, DALY) or in terms of equivalency factors (Bq U235 air-equiv.). To allow comparison across different methodologies, equivalency factors have been used for all methodologies. HHD E/H factors are used as reference whilst HHD-I, CGM and UCrad are plotted on the y-axis.

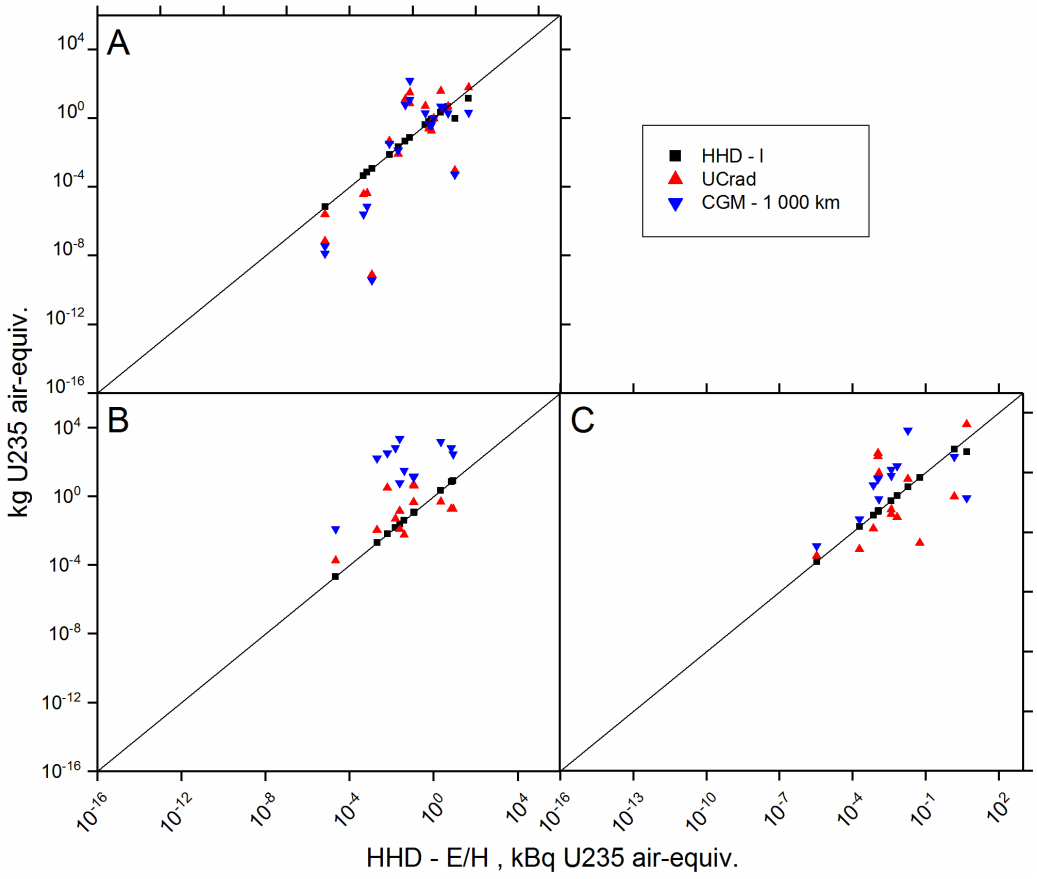

Figure 4.6 – CGM – UCrad-Frischknecht characterisation factors comparison: A) air emissions; B) fresh water emissions; C) sea emissions. E/H: Egalitarian/Hierarchist; I: Individual. HHD= Human Health Damages.

Individualist (I) characterisation factors show minimal deviation from the reference set for each emission category, with MLD values in the order of 0.01. UCrad set follows featuring MLD values lower than one. In the air emission category (Figure 4.6 A), UCrad features an MLD of -0.50; here, the largest deviation is given by two noble gases (both located on the left-hand side), namely Xe133 and Rn222, which feature MLDs of -4.7 and -6.2 respectively. SAM nuclides (C14 and H3) also show very large deviations, respectively equal to -4.0 and -1.2. By contrast, freshwater and sea water categories do not include any radionuclide behaving significantly differently from the average. The freshwater category features MLD equal to -0.40, whilst sea water to 0.03. CGM methodology, on the other hand, features for air and sea water emissions MLDs equal to -0.83 and 0.81; notably, the biggest difference can be noticed for the freshwater category where CGM characterisation factors are located well above the bisecting line featuring log deviations ranging from 0.4 for uranium-238 to 4.2 for silver-110m, with an average MLD equal to 2.7.

4.3.2.2 Normalisation Factors

Table 4.3 reports UCrad and HHD normalisation factors according to CML and ILCD inventories in terms of the overall emissions (namely domestic) and emissions per persons (namely person equivalent).

Table 4.3 – Domestic and person equivalent normalisation factors calculated according to HHD and UCrad methodologies and based on CML and ILCD inventories.

	Normalisation factors			
	CML inventory		ILCD inventory	
	HHD	UCrad	HHD	UCrad
Domestic (Bq U235 air-equiv)	7.97E+12	4.23E+11	5.64E+11	1.08E+11
Person equivalent (Bq U234 air-equiv/person)	1.60E+04	8.48E+02	1.13E+03	2.16E+02

With respect to the CML inventory, UCrad normalisation factors are approximately twelve times lower than HHD ones; this difference is halved to approximately five times when the ILCD inventory is used. The discrepancy between UCrad and HHD is due to the differences in the characterisation factors generated by the two methodologies as discussed in Section 4.3.2.1, most notably with respect to atmospheric emissions of carbon-14 (C14) and freshwater emissions of iodine-129 (I129). (C14 has a higher characterisation factors in HHD than in UCrad, while the opposite occurs for I129.) Figure 4.7 and Figure 4.8 illustrate contributions of each radionuclide to normalisation factors of HHD and UCrad obtained from CML and ILCD respectively.

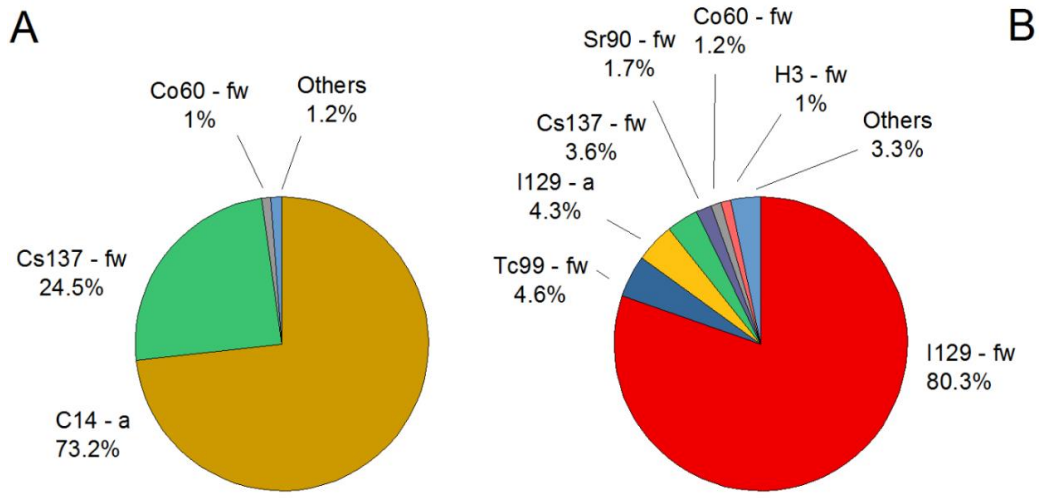

Figure 4.7- Contribution to EU25+3 impact based on the CML inventory and calculated according to HHD (A) and UCrad (B) methodologies.

The graphs show that C14 atmospheric emissions contribute to approximately 73% and 83% of HHD normalisation factors calculated by ILCD and CML, which complemented with water emissions of caesium-137 (Cs137) amount to over 90%. The situation is utterly different for UCrad: I129 emissions to water represent the largest source of impact, with contributions approximately equal to 80% and 91% of CML and ILCD respectively. The remaining portion is scattered amongst various radionuclides which, interestingly, do not include C14.

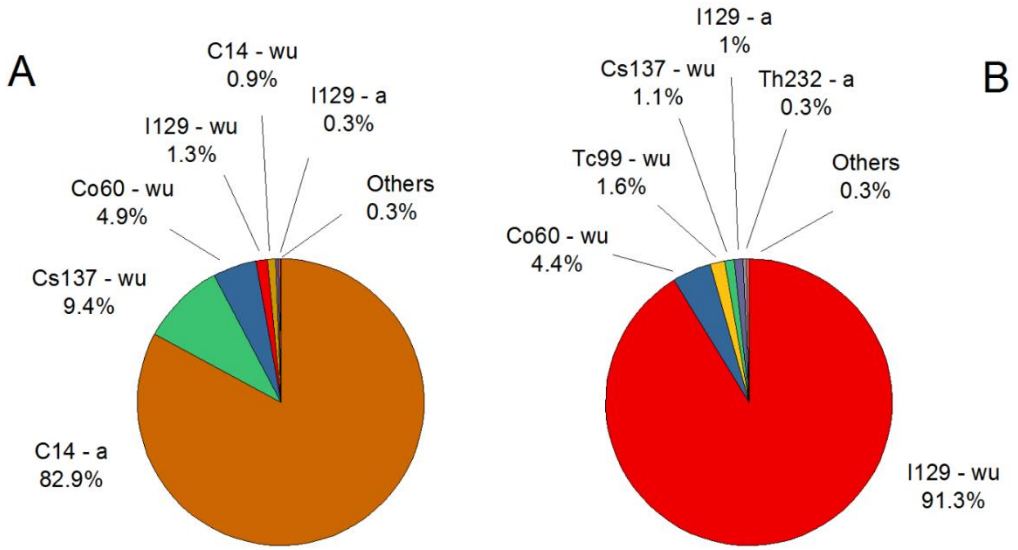

Figure 4.8 – Contribution to EU27 impact based on the ILCD inventory and calculated according to HHD (A) and UCrad (B) methodologies.

Unlike differences between UCrad and HHD for the same inventory (which are due to discrepancies in the characterisation factors), those associated with ILCD and CML inventories for the same impact methodology are due to differences in the magnitude of the radioactive emissions. Most notably, the fact that the discrepancy between CML and ILCD inventories for HHD is higher than that exhibited by UCrad (Table 4.3 - which leads to the fact that the discrepancy between HHD and UCrad is lower when ILCD is used instead of CML) leads to the conclusion that differences between C14 atmospheric emissions in CML and ILCD inventories are higher than those for water emissions of I129.

4.3.3 CGM vs UCrad

4.3.3.1 Characterisation factors

Figure 4.9 includes three charts where characterisation factors for air, fresh water and sea water are compared; Figure 4.10 reports comparison for nuclear waste disposed in a GDF;

and finally, Figure 4.11 shows relevant comparisons for Specific Activity Models (SAM) nuclides, that is C14 and H3. In Figure 4.9, Figure 4.10 and Figure 4.11 charts, UCrad characterisation factors are located on the x-axis and used as reference set, while CGM sets are plotted on the y-axis. Notably, four sets of CGM characterisation factors are reported for air, fresh water and sea water emission categories; these correspond to four distances of the critical group from the release source, ranging from 1 to 10 000 km. Conversely, only one set is reported for emissions from GDF (see Section 4.2.5). It is worth reminding that accuracy and significance of analytical models for radionuclides dispersion is generally not assured for long distances (see Section 4.2.1.1); nonetheless, in this analysis the range of the assessment has been extended up to 10 000 km with the purpose of showing the scale for which the two methodologies exhibited an acceptable level of agreement. Finally, distances returning an MLD equal to zero for both radionuclides and toxic substances (analysed in Section 4.3.4) characterisation factors are shown in Table 4.4.

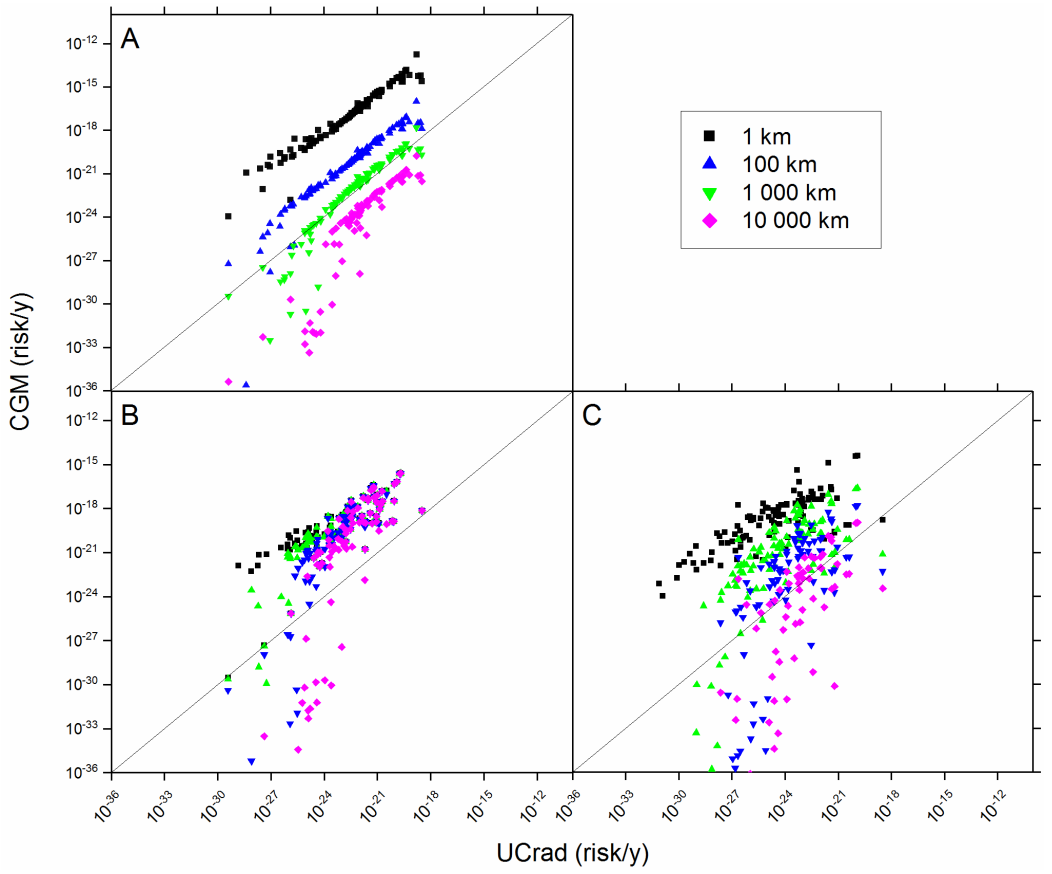

Figure 4.9 – CGM – UCrad characterisation factors comparison: A) Air emission; B) Fresh water emission; C) Sea water emission.

The chart for air emissions (Figure 4.9 A) shows that by increasing the receptor distance CGM factors decrease, and approach – or even go below – UCrad reference set. The best agreement between the two methodologies (i.e. MLD = 0) is found for a distance equal to 601 km, as shown in Table 4.4. This means that for distances below 601 km, CGM methodology returns on average higher characterisation factors than UCrad, while the opposite occurs for distances above 601 km. Characterisation factors for air emissions show very little scattering with nuclides almost being placed on straight lines (in a log-log graph). By contrast, fresh and sea water factors (Figure 4.9 B and C) appear to be considerably more scattered. For the fresh water emission category, an MLD value equal to zero is obtained for a distance of 722 km; whilst a much lower value, 175 km, is found for sea water emissions (Table 4.4). This means that, given the same distance, the CGM methodology returns higher factors (relative to UCrad) for sea water emissions, followed by air and river.

Table 4.4 – Distances resulting in an MLD=0 for air, fresh water and sea water emissions of both radionuclides and toxic substances

	Air	Fresh water	Sea water
Radionuclides	601 km	722 km	175 km
C14 & H3	218 km	393071 km	233 km
Toxic substances	440 km	106139 km	1389 km

The chart comparing characterisation factors for wastes stored in a GDF (Figure 4.10) gives a different picture: the radionuclides considered in each category feature considerable deviation between the methodologies, with MLDs ranging from 6.7 (HEU) to 7.8 (Pu).

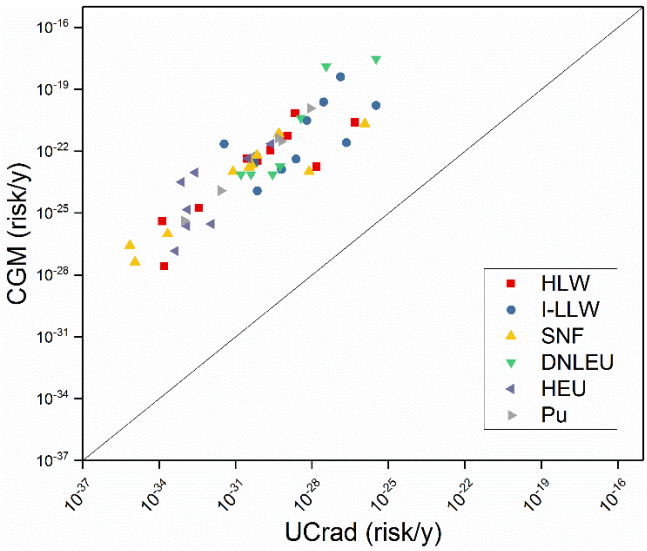

Figure 4.10 – CGM – UCrad characterisation factors comparison for wastes disposed in GDF. HLW = High Level Waste; I-LLW = Intermediate/Low Level Waste; SNF = Spent Nuclear Fuel; DNLEU = Depleted, Natural, Low Enriched Uranium; HEU = Highly Enriched Uranium; Pu = plutonium

Finally, Figure 4.11 focuses on two specific nuclides, C14 and H3, and consists of three charts where characterisation factors for air, river and sea emissions are compared. Emissions from GDF are not included as both C14 and H3 feature too low half-lives – compared to the radionuclides included in this emission category – to result in meaningful impacts after transportation from GDF to biosphere. Furthermore, only H3 appears in fresh water and sea water categories. Potential impacts of C14 through those pathways are, in fact, neglected by the IAEA's SAMs. As reported in Table 4.4, for air and sea water emissions MLD values of 0 are found for 218 and 233 km. For fresh water emissions, however, this distance is much bigger and well above 10 000 km. The calculated distance is approximately equal to 393 000 km, meaning practically no convergence of the two methodologies.

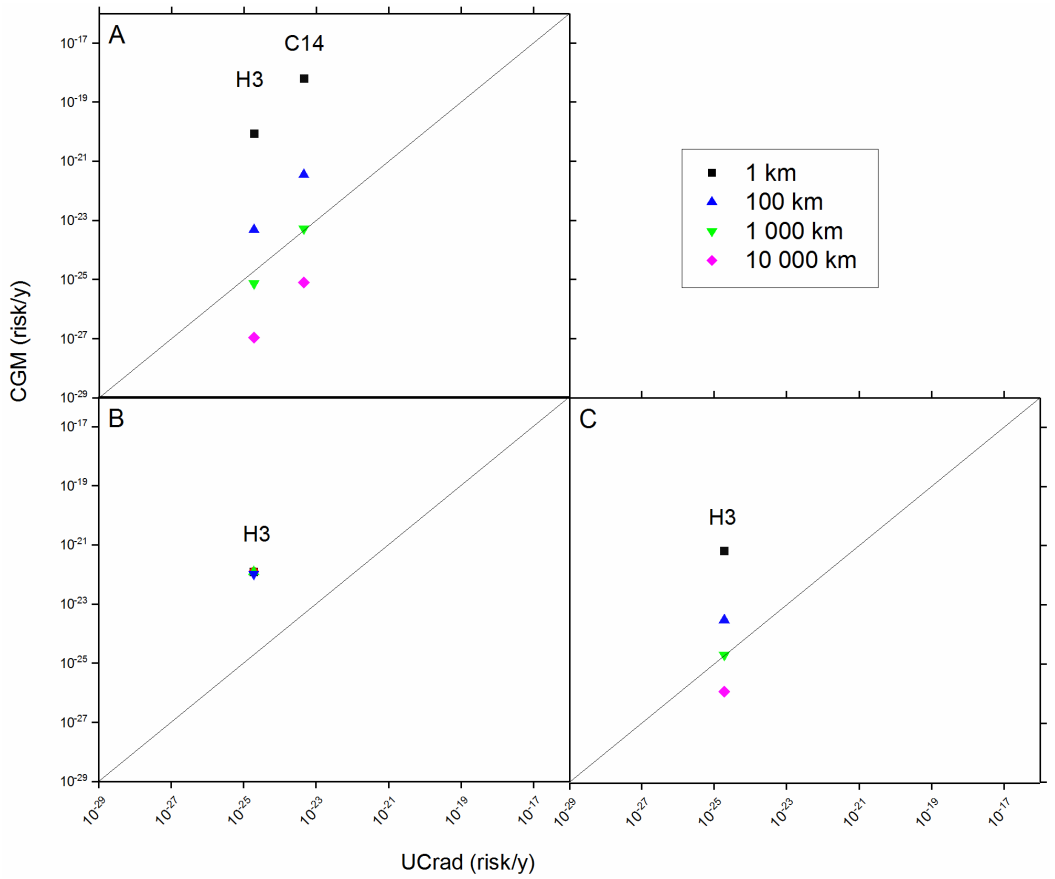

Figure 4.11 – CGM – UCrad characterisation factors comparison for H3 and C14. A) Air emission; B) Fresh water emission; C) Sea water emission.

4.3.3.2 Radionuclides' half-life

Figure 4.12 reports log deviations of four CGM sets of characterisation factors compared to UCrad reference set for three emission categories: air, fresh and sea water. It must be noticed that the charts do not report all values of log deviations. Because log deviations for very low half-lives may decrease over several orders of magnitude, their inclusion would compromise the overall readability of the plots: For this reason, y-axes have been limited to -90.

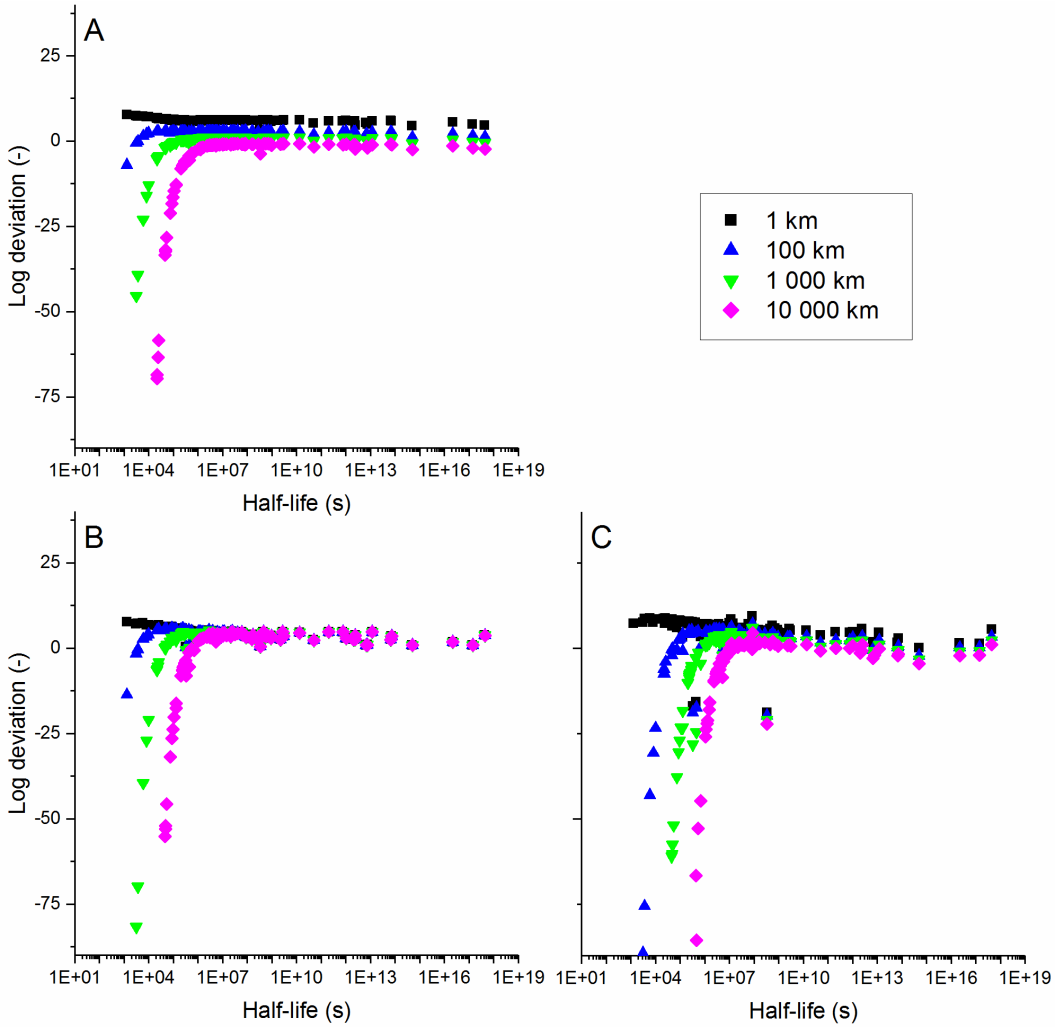

Figure 4.12- Log deviation of CGM vs UCrad characterisation factors for four distances of the critical group as a function of radionuclides' half-lives. A) Air emissions; B) Fresh water emissions; C) Sea water emissions.

The graphs show quite clear trends that are consistent across all emission categories. As half-life decreases, log deviations at first are approximately constant and then, for values of half-life specific to each distance and emission category, they sharply decrease to negligible values. The number of radionuclides showing significant deviations from the

horizontal asymptote at high half-lives depends both on the receptor distance and the emission category. It increases with the receptor distance (as it is clearly visible from the graphs); but also from air (Figure 4.12 A), to fresh water (Figure 4.12 B), to sea water (Figure 4.12 C) as demonstrated by calculating the half-life values at which the decrease starts. For instance, at 100 km this is approximately equal to 3.5E+3 s for air, 5E+3 s for fresh water and 9.5E+4 s for sea water emissions. However, at low distances (e.g. 1 km), log deviations feature no significant decrease; rather for very small half-lives they show an increasing trend. This is particularly evident for air (Figure 4.12 A) and fresh water (Figure 4.12 B) emissions. For sea water emission, an initial increasing trend may be spotted, but this however shows to peak and then decrease.

4.3.4 CGM vs USEtox

Figure 4.13 reports a comparison between CGM and USEtox characterisation factors for air (Figure 4.11 A), fresh water (Figure 4.11 B) and sea water (Figure 4.11 C) emissions; and for three reference chemicals: benzene (B), mercury (Hg) and pentachlorobenzene (PeCB). These chemicals are meant to represent a wide variety of cases: mercury is an inorganic element; whilst benzene and pentachlorobenzene are organic compounds featuring very different behaviours in the environment, which have been extensively studied for impact assessment purposes (e.g. see [300]). For achieving a meaningful comparison, CGM methodology has been amended to use the same exposure and effect module of USEtox. Both methodologies characterisation factors have been expressed in terms of cases (of cancer) per year, according to conversion factors used in the USEtox model [286].

Figure 4.13 shows that toxic substances follow a similar trend to radionuclides, although absolute values of deviations differ. With respect to the distance returning an MLD equal to zero, the air emission category (Figure 4.11 A) feature the smallest value, at 440 km, followed by sea water, at 1 389 km; this is in contrast with what occurs for radionuclides. Fresh water, however, keeps returning the highest value, which in this case is well above the upper value of the distance range (~105 000 km).

4.4 Discussion

This Section aims at discussing from both qualitative and quantitative perspectives the main findings of this Chapter, starting with examining the main advantages, disadvantages and differences of the radiological impact assessment methodologies developed within this Thesis (Section 4.4.1), moving to discussion of quantitative analyses presented in Section 4.3, and concluding with recommendations on practical applications of the methodologies developed.

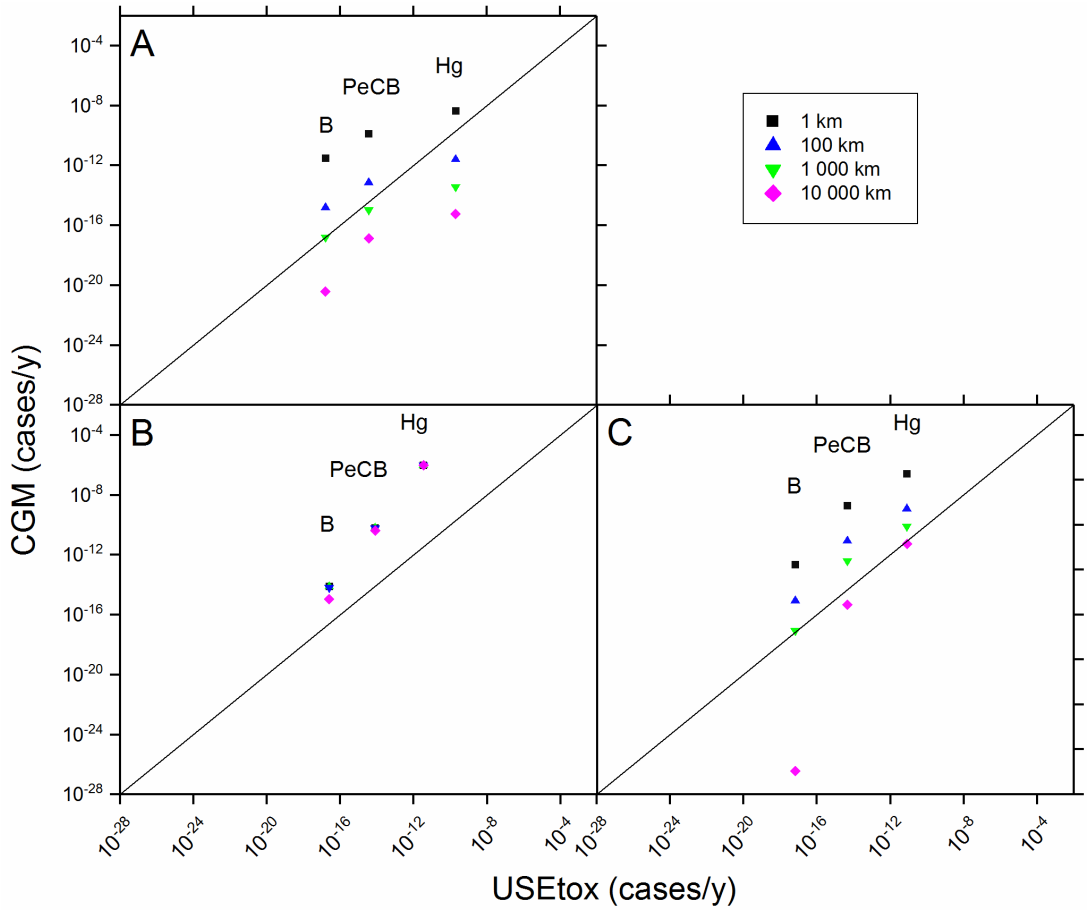

Figure 4.13 – CGM – USEtox characterisation factors comparison: A) Air emissions; B) Fresh water emissions; C) Sea water emissions.

It is worth keeping in mind during the discussion of the results that graphs presented in each Section use different units. Specifically, CGM/UCrad vs HHD comparison has been performed by means of equivalent factors (e.g. Bq U235 air-equiv.Bq), CGM vs UCrad on a risk per year basis, and CGM vs USEtox on cases (of cancer) per year basis. This implies that direct comparisons across different charts must be restricted to qualitative considerations, as absolute values may differ by several orders of magnitude. In principle,

the use of equivalency factors would have allowed comparisons across different graphs; however, when possible (that is for comparison between UCrad/USEtox with CGM) equivalence factors have been avoided. Because equivalency factors are obtained by dividing each characterisation factor by a reference value (specific to each methodology), their comparison carry a double, interconnected effect that is representative not only of the substance-specific characterisation factor, but also of the reference one. This would in turn compromise both readability and ease of understanding of the results.

4.4.1 Advantages and disadvantages, similarities and differences between CGM and UCrad

As discussed in Section 4.2.1, CGM and UCrad only difference with respect to the general framework lies in the approach adopted in the fate module. The former employs analytical models to solve the basic radionuclide transport equations by using simplifying assumptions; whilst the latter treat the environment as divided into homogeneously mixed compartments, each representing an environmental medium. The two approaches stem from two very different needs. Analytical models are generally used in risk assessment studies to estimate the highest possible impacts that human beings may experience. Usually a number of tiers (or levels) are available with increasing accuracy and data requirements; first tiers are used as a screening tool and represent worst case scenarios. Compartment-type models, on the other hand, are widely used in Life Cycle Impact Assessment (LCIA) as a good estimate of average impacts of pollutants. While analytical models have been largely used to simulate dispersion of radionuclides, to date no compartment model has been developed for radionuclides. Radioactive nuclides behave in the environment much alike heavy metals; and these – and more in general inorganic compounds – represent a known issue in compartment models leading to the prediction of unrealistic accumulation in environmental media. For this reason, the CGM methodology constitutes a good reference point against which UCrad characterisation factors may be compared and validated (Sections 4.3.3 and 4.3.4).

Another important difference between the two methodologies lies in their time boundaries. Level III Mackay models – i.e. the type of compartment models generally used for LCIA purposes – assume steady-state conditions; whilst CGM fate models employ time-dependent equations to estimate dispersion of radionuclides in the environment. However, CGM also uses one crucial assumption that is that the release of radionuclides

has occurred for 30 years or more; this allows assuming that steady-state conditions are reached in the soil and therefore partitioning factors may be used to relate soil and food chain concentrations (e.g. vegetables, cattle).

A further consideration worth discussing is that the solid waste category of CGM relies upon the Post-Closure Safety Assessment (PCSA) of a generic repository developed by RWM Ltd. This study relies on a model – not entirely publicly available and thus not customizable – that estimates the potential impacts of radionuclides escaping the GDF onto a receptor represented by a small village located just above the GDF. The area of the assessment is assumed to be in the order-of-magnitude of ten km^2 (meaning that the critical group is located approximately 3.3 km from the release source), and cannot be amended, which in turn prevents a similar analysis to that of direct discharges to be performed. Like CGM, also UCrad relies on the PCSA report due to the lack of an integrated model for simulating escape of radionuclides from the engineered system of the GDF and transportation through the geosphere to the biosphere. Notably, UCrad fate model requires as input the flow of radionuclides leaving the geosphere and entering the biosphere – the far-field flux (Section 4.2.1.1) – and upon this it applies characterisation factors for emissions to groundwater. For both CGM and UCrad, the adoption of the PCSA model limits the number of radionuclides to that included in the report, usually representing the ten (or less) radionuclides having the greatest impact.

Furthermore, both CGM and UCrad characterisation factors are strictly linked to a number of assumptions made in the PCSA. One of crucial importance is that the GDF is located in higher strength rock formations. This assumption brings a number of implications, for instance that groundwater is the most critical escape pathway for radionuclides. Groundwater is likely to be present in both higher and lower strength rock, but it can be safely assumed not to be in salt rock; therefore, for a GDF located in salt rock formation, the obtained characterisation factors would not be suitable. This represents a further area for future improvements where characterisation factors may be diversified according to different GDF designs. As construction of GDFs in many countries commences, an increasing number of GDF safety assessments will be produced in the next years, thus providing data for such improvements.

Besides assumptions in the PCSA, the methodologies rely on a number of other assumptions, whose effects have not and cannot be quantified due to lack of data in the

literature. The most notable concern the attribution of air-water partition coefficients and bio-accumulation factors, with the former being specific to UCrad. A negligible air-water partition coefficient has been attributed to the majority of radionuclides (to represent negligible volatility), whilst for a number of radionuclides the bio-accumulation factors have been attributed via the analogues approach. Both parameters control how radionuclides disperse in different environmental media, and thus can strongly affect the characterisation factors; for instance, a higher concentration in the air compartment for a specific radionuclide (that is associated with a higher partition coefficient) is usually linked with higher characterisation factors.

On the downside, both methodologies don't account for the impact of radionuclides contained in Very Low or Low Level Waste (VLLW or LLW) suitable to be disposed in near-surface repositories: this represents a crucial limitation of the methodologies and an area for future improvements. The reason is that is that not a single report similar to PCSA but for near-surface repositories was found in the literature. A further limitation specific to UCrad is that it does not take into account daughter radionuclides in the fate and exposure analysis: decayed radionuclides are assumed to have left the system in the same way as organic compounds are removed due to degradation. Notably, this does not represent a limitation to CGM because of the lower time required by radionuclides to travel from emission sources to critical groups compared to the time to reach steady-state conditions in UCrad. Furthermore, neither of the methodologies accounts for the impacts of the products of the radioactive decay chain. This assumption is more significant for UCrad than for CGM. Because UCrad is a steady-state model, the radioactive decay of radionuclides may lead to significant accumulation of daughter radionuclides in the environmental media, and thus to potentially higher radiological impacts. On the other hand, because CGM is based on analytical models that simulate transport and dispersion of radionuclides from the source of emission to the receptor, the production of daughter radionuclides is considerably less significant: notably, the shorter the distance of the critical group, the less significant the radioactive decay is.

Finally, it is important to stress that both methodologies represent a significant step forward for assessment of radiological impacts in LCA: impact factors for more than 100 radionuclides are calculated, compared to approximately 25 radionuclides (not in all

emission categories) considered by Frischknecht and co-workers' Human Health Damages (HHD) methodology.

It is evident that UCrad represents a better candidate than CGM for incorporation in the Impact Assessment phase of LCA, primarily because it is in line with the main findings of the review of radiological impact assessment approaches for use in LCA, presented in Chapter 3, but also with the research needs identified by the JRC within the recommendations of methods for LCIA in the European context [122]. Notably, the approach includes impacts from nuclear solid waste disposed in an underground disposal facility; unlike CGM, it is site independent and produces average estimates of impact; and it ensures better compatibility with toxic impact models (USEtox). Because the CGM methodology is adapted from established models used for risk assessment in the nuclear industry, it represents a suitable methodology for comparing and validating UCrad characterisation factors. However, this does not necessarily imply that CGM has no practical applications; these are discussed in Section 4.4.4.

4.4.2 CGM/UCrad vs Human Health Damages

The comparative analysis between CGM, UCrad and HHD characterisation factors presented in Section 4.3.2 shows that on average there is a good agreement between the methodologies. This is particularly true for HHD characterisation factors obtained according to Egalitarian/Hierarchist and Individualist perspective of the cultural theory, but also for both air and sea water emission categories of CGM and UCrad; these feature MLD values below unity, indicating that there is a mean deviation of less than an order of magnitude with HHD reference set, i.e. E/H. By contrast, while for UCrad the freshwater category shows consistent results in terms of MLDs; the same cannot be said for CGM, which features a significant positive deviation, signalling that CGM methodology produces higher characterisation factors compared to HHD, but also to UCrad. The discrepancy for freshwater emissions characterisation factors is further investigated in Section 4.4.3 when UCrad and CGM are compared in detail, but essentially it is due to the relative difference between radionuclides' half-life and the characteristic time of the dispersion process in fresh water.

Results of this comparative analysis can be seen from a twofold perspective. On the one hand, they clearly show that there is a better agreement between HHD and UCrad methodology, rather than CGM (this is particularly true for the fresh water emission

category); if HHD were to be taken as the gold standard, this would thus reinforce the assertion that UCrad represents a better candidate than CGM. On the other hand, the analysis also contributes to validating the proposed methodologies by showing that the characterisation factors are in the same range of magnitude of those published in a peer-reviewed article.

In view of what discussed above, normalisation factors present a rare example of how UCrad and HHD can lead to different results. The large discrepancy is largely due to how differently the two methodologies evaluate radiological impacts caused by emissions of C14 and I129. Radiological impacts per unit emission of C14 are considerably higher in HHD than UCrad (10 vs 0.00093 Bq U235 air-equiv/Bq), which is strictly linked to the use of Gaussian Plume models (i.e. space dependent models) in HHD and Specific Activity Models (i.e. steady state models) in UCrad. The opposite occurs for I129 (4.8 vs 63.7 Bq U235 air-equiv/Bq) which can be traced back to its significant half-life value (the effect of radionuclide's half-lives is discussed in Section 4.4.3).

Finally, differences between normalisation factors obtained according to CML and ILCD inventories are also quite significant (around one order of magnitude). These are mainly due to differences in emissions of C14 reported in the inventories; but cannot be attributed to their different geographic coverages (EU25+3 vs EU27). The countries that are included in one methodology and not in the other and vice versa have in fact low installed nuclear capacities and are thus unlikely to be major emitters of radionuclides. Therefore, the primary cause lies in the methodology applied for building the inventories: CML used data obtained from the UK Environment Agency to extrapolate the emission profile of the EU 25+3 countries, whereas the JRC employed accurate, country-specific data, mainly obtained from UNSCEAR inventory [301].

4.4.3 CGM vs UCrad/USEtox

The comparison between CGM and UCrad methodologies (Section 4.3.3) performed for multiple receptor distances and emission categories gives interesting insights into the two methodologies and enables to highlight how (differently) sensitive the methodologies are with respect to the radionuclides' half-lives . At the same time, the application of CGM methodology to toxic substances and comparison with USEtox (Section 4.3.4), the compartment-type model on which UCrad is based, contributes to further validate the main outcomes of the analysis by extending its validity to toxic substances.

Comparative analyses for both radionuclides and toxic substances show that by increasing the receptor distance CGM characterisation factors decrease substantially, from being significantly higher to similar or even significantly lower than UCrad/USEtox factors. The distances for which MLD is equal to zero, reported in Table 4.4, vary substantially amongst different emission categories, but also between same emission categories of different substance classes (radioactive and toxic). Further to this, the charts reporting characterisation factors for direct discharges of radionuclides (Figure 4.9) show how crucial the half-life parameter is, and especially how sensitive the methodologies are to it. On a general basis, the lower the half-life, the lower the characterisation factor is; for this reason, radionuclides with low half-lives are located on the bottom-left section of the charts. CGM, however, appears to be significantly more affected than UCrad, which can be noticed by the peculiar 'tail' that CGM sets feature when compared to UCrad reference set. As an example let us focus on the graph for air emissions (Figure 4.9 A). I134 features very low half-life ($t_{1/2}$ = 3.2E+3 s) and is located on the far left side of the graph (UCrad characterisation factor = 6.5E-28 risk/y); its CGM characterisation factor decreases from 1.6E-20 risk/y at 1 km to a negligible value at 10 000 km. U238 (UCrad factor = 3.3E-20 risk/y), which has higher half-life ($t_{1/2}$=1.4E+17 s), on the other hand, shows a reduction of only seven orders of magnitude, from 2.3E-15 risk/y at 1 km to 2.96E-22 risk/y at 10 000 km. The methodologies' sensitivity to the half-life parameter is clearly visible in Figure 4.12, which reports logarithmic deviations of CGM characterisation factors compared to UCrad ones for different distances and emission categories as a function of the radionuclides' half-life. Logarithmic deviations appear to be partially dependent on the half-life: at high values the log deviation is approximately constant, meaning that the two methodologies feature a constant difference and hence are equally sensitive to the half-life; the opposite occurs at low values, where log deviations appear to sharply decrease with decreasing values of half-life. This means that the difference between the two methodologies factors reduces, and thus that CGM factors decrease relatively more than UCrad ones. The number of radionuclides affected depends both on the distance of the receptor and the emission category. As shown in Section 4.3.3, this increases with the distance and also from air to fresh and to sea water emission categories. The former is clearly visible in Figure 4.12, but also in Figure 4.9 where it is represented by an enlarging 'tail'; the latter, on the other hand, can be noticed by identifying the half-live values at which the decrease commences (Figure 4.12). Both trends can be explained according to

the following reasoning: the driving cause is the relative – rather than absolute – value of the radionuclides' half-life compared to the characteristic time of the dispersion processes in the CGM methodology. Characteristic times can be obtained by dividing the distance to be travelled (e.g. 1, 100, 1 000 km) by the speed of the carrier (e.g. wind speed = 2 m/s, etc.). Table 4.5 reports characteristic times for different distance and emission categories. They increase with distance, and from air to fresh and sea water at the same distance. As the characteristic time increases, dispersion processes take longer to complete, hence allowing for a larger percentage of radionuclides to decay.

Table 4.5 – Characteristic time of dispersion process in air, fresh water and sea water

	Characteristic time (s)		
Distance (km)	Air	Fresh water	Sea water
1	5.00E+02	9.26E+02	1.00E+04
100	5.00E+04	9.26E+04	1.00E+06
1000	5.00E+05	9.26E+05	1.00E+07
10000	5.00E+06	9.26E+06	1.00E+08

The different sensitivity of CGM and UCrad to the half-life parameter is essentially linked to the models setup. As discussed in Section 4.4.1, UCrad is a steady-state model whilst CGM is time-dependent; effectively UCrad represents a Continuous Stirred Tank Reactor (CSTR) whilst CGM a Plug Flow Reactor (PFR). These two models are based on very different assumptions, i.e. perfect mixing in all directions vs absence of axial mixing, which are the cause to how CGM and UCrad respond to varying half-life values.

With respect to emissions from nuclear wastes disposed in a Geological Disposal Facility (GDF), only one set of CGM has been reported for each waste type. For this reason, it has not been possible to calculate the distance resulting in a negligible deviation between the methodologies, which however does not prevent further discussion of the results. The considerable deviation in terms of MLD (in the range of 6.7–7 8) between CGM and UCrad methodologies can be traced back to the scale of the specific model employed for this emission category. The PCSA, in fact, considers an area of approximately 10 km^2, which coincides with an equivalent circular radius of about 3.33 km^2 [269]: the significant deviation is thus due to the very small scale of the assessment. This can be further explained by considering the average MLD value for air, fresh water and sea water emissions at 1 km, which is equal to 5.5 and somewhat close to what found for emissions from a GDF. Due to irretrievability of the specific models used in PCSA, no further analysis

or consideration on this emission category has been carried out. Finally, results of the comparison between C14 and H3 characterisation factors appear to be in line with those shown by other radionuclides. Air and sea water categories feature the lowest distance returning an MLD equal to zero, while for fresh water such distance becomes considerably high. This is due to the fact that both radionuclides feature half-life values much bigger than the characteristic time of dispersion in fresh water, hence diminishing very little with increasing distance.

It is interesting to notice how characterisation factors for radionuclides are located on the charts. For air emissions they are almost placed on straight lines (except for the final 'tail'), while they appear much more scattered for fresh and sea water emissions. This is due to inherent features of the methodologies. CGM relies on the use of advective/diffusive models for species dispersion in the environment; nonetheless, the exchange between different environmental media is poorly addressed. (This is due to the fact that this model is used mainly for impact assessment on low distances where equilibrium amongst environmental media is neglected.) The main consequence is that nuclides will tend to stay in the media where they are released (rather than dispersing across the whole environment), thus resulting in higher concentration in those media which are potentially more dangerous. For instance, atmospheric releases will result in radionuclides either remaining in the air or entering the food chain (through deposition), both of which are very impactful pathways for the majority of radionuclides. By contrast, in UCrad radionuclides would also disperse in soils, sediments and fresh and sea bodies, hence accessing compartments with lower exposure factors (i.e. soils and sediments). This line of reasoning explains why river and sea water emissions impact factors appear to be much more scattered than air ones: from an emission to fresh or sea water, UCrad allows radionuclides to disperse in ideally all compartments, whilst in CGM most radionuclides will remain in the water compartment or disperse poorly in sediments. These compartments have access to fewer and somewhat less impactful pathways, which coupled with the variability of radionuclide parameters (i.e. dose conversion factors for different pathways or partitioning factors) result in some radionuclides having low impact factors in CGM and relatively higher impact factors in UCrad and vice versa. The opposite occurs for the air emission category (i.e. less scattered impact factors), which for this reason gives the easiest results to read. Furthermore, the graphs in Figure 4.12 how that

the higher level of scattering featured by fresh and sea water emission categories cannot be linked with half-life as it occurs across the whole half-life spectrum.

Characterisation factors for toxic substances also give interesting insights. Both absolute values of the virtual distances calculated and their ranking for emission categories are somewhat different with respect to radionuclides. However, by following the same line of thought as above, it is possible to explain those differences, which ultimately depend on the half-life parameter. As mentioned above, low half-life radionuclides strongly affect the MLD value, the more the larger the distance. Radionuclides for which characterisation factors have been obtained feature half-life values in the range of 1E-18 – 1E-4 s. Toxic substances also feature degradation processes and are characterised by their own degradation factors, which, like radionuclides, differ according to the media in which it occurs. For the selected toxic substances, degradation factors range from 1E-7 for benzene to 1E-8 for PeCB and 1E-20 (i.e. negligible) for Hg. Therefore, differences in the distances computed are a direct consequence of the different ranges of degradation/radioactive decay factors considered; whilst differences in their ranking is caused by degradation factors varying from one environmental medium to another.

A fair comparison between toxic and radionuclides characterisation factors would have to consider the same range of degradation/radioactive decay parameters. For instance, fresh water MLD is strongly affected by radionuclides featuring low-half-lives; however, if only those radionuclides with half-lives higher than 1E-7 (the lowest degradation factor of the selected toxic substances) are considered, the resulting MLD value would be almost constant with distance and equal to 3.4, as shown in Figure 4.12 B. In the case of toxic substance, this would range from 3.63 to 3.16.

The comparison between the methodologies, beyond highlighting specific features of each methodology, also allows to draw some important conclusions. Firstly, amongst the four CGM sets it is possible to identify one (1 000 km) which on average gives closer results to UCrad. Such distance is also in accordance with the size of the continental scale used in both USEtox and UCrad. Secondly, the good agreement between the methodologies further validates the characterisation factors produced by UCrad.

4.4.4 Practical applications

To conclude, the practical applications of the methodologies developed within this Thesis are discussed. Two alternative approaches are possible. The first one is immediately apparent – either of these two methodologies can be used; the choice may be based on the goal and scope of the study and data availability. Nonetheless, as already discussed in Sections 4.1 and 4.3.2, it is evident that UCrad is a more appropriate methodology for Life Cycle Impact Assessment (LCIA). The other approach links to the Introduction (Section 4.1) and responds to the need identified by some authors of combining HERA and LCA. This approach envisages the combined use of CGM and UCrad. CGM would be exclusively used for the Foreground system – as defined by Clift and co-authors [108] – where usually more information is available; whilst UCrad may be used for both or only the Background system. As impacts of ionising radiation represent a much-debated topic and stringent limits have been put in place to limit radioactive emissions, the application of CGM methodology to the Foreground system would give a very different perspective from UCrad. The first case would allow comparison of average and worst case impacts and enable to calculate what proportion of the overall radiological impact affects the critical group. The second case would have the advantage of producing only one overall radiological impact figure, but it may be argued that summing worst case (for the Foreground) and average (for the Background) impacts would make little sense.

Nevertheless, the combined use of CGM and UCrad would allow recommendations not only on which product is more environmentally sustainable, but also on the location (e.g. a similar analysis has been performed by Spadaro and Rabl (1999)) or the scale of the processes in the Foreground system. However, whilst recommendations on the location may be given for every functional – be it fictitious or representative of actual operational data -, advises on the scale would be meaningful only if a realistic (i.e. that describes the actual operations of a plant) functional unit is employed. Let us make an example on this difference and focus on two processes that produce the same product and are placed in different locations. For each functional unit, the CGM methodology would be able to tell which location is preferable by identifying for each process the relative position of the critical group and its specific impacts. Conversely, the use of a realistic functional unit would also enable to evaluate how radiological impacts on the critical group compare with regulatory limits; this would allow not only to assess whether a specific scale of a plant complies with the limits, but also which is the maximum scale that could be

operationalized. If required, the assessment could also be made location dependent by using specific environmental parameters. The fact of using functional units representing real operations, in place of fictitious ones, represents a grey area where HERA and LCA overlap. A study carried out in such way would maintain typical LCA features (e.g. number of pollutants and impacts considered, assessment over the life cycle, etc.), but at the same time would acquire a regional perspective typical of risk assessment.

4.5 Conclusions

This Chapter has presented an overarching framework for Radiological Impact Assessment for use in the Impact Assessment phase of LCA (LCIA), from which two alternative methodologies for calculation of impact factors have been developed, namely Critical Group Methodology (CGM) and UCrad. These are expression of two somewhat distant cultures: Risk Assessment (HERA) and Life Cycle Assessment (LCA), and differentiate only for the approach used to modelling radionuclides' fate. The Critical Group Methodology has been extensively used in risk assessment studies in the nuclear field, thus representing a proven approach and a basis for comparison. By contrast, UCrad represents the first-of-its-kind compartment model for radionuclides. For both methodologies, characterisation factors have been calculated for several emissions categories including direct discharges to air, fresh and sea water, and emissions from different types of nuclear wastes disposed in a Geological Disposal Facility (GDF); whilst normalisation factors have only been calculated for UCrad. A quantitative analysis of methodologies has then been carried out. First, UCrad and CGM have been compared with Human Health Damages – the only methodology for radiological impacts included in present LCIA methods – for those characterisation factors in common to the methodologies; and then UCrad normalisation factors have been compared with HHD ones according to two inventories developed by CML and ILCD. The comparison shows that UCrad characterisation factors feature lower deviations from HHD than CGM, especially with respect to the freshwater category; nonetheless, UCrad and HHD normalisation factors show considerable deviation. This is due to significant differences in the magnitude of the characterisation factors for carbon-14 (C14) and iodine-129 (I129) computed by the methodologies. The analysis has then focussed specifically on CGM and UCrad, with the objective of investigating in detail how different approaches to fate modelling affect characterisation factors. For this reason, first CGM and UCrad

characterisation factors have been compared, and then an analogous comparison has been performed between CGM and USEtox for three reference toxic substances. Notably, four sets of CGM characterisation factors with varying distances of the critical group from the source of the release has been compared and the distance returning on average a negligible deviation (i.e. MLD=0) has been calculated. With respect to the comparison between CGM and UCrad, the analysis shows that such distance varies substantially amongst different categories, but ultimately it depends on the characteristic time of the dispersion process of each emission category in CGM. The effect of radionuclides' half-life has also been investigated and appears to be more significant in CGM than in UCrad. Results of the comparison between CGM and USEtox contribute to further validate the methodologies: although values of the critical group distance returning a nil MLD differ from the ones obtained for radionuclides, the difference can be traced back to the influence of radionuclides' half-life. Finally, practical applications of these methodologies have been discussed. It is evident that UCrad represents a better candidate than CGM for incorporation in the Impact Assessment phase of LCA: in line with conclusions of the review of radiological impact assessment approaches presented in Chapter 3, it includes impacts from solid waste disposed in a Geological Disposal Facility, and, unlike CGM, it is site independent and produces average estimates of impact; however, although CGM has been mainly developed for comparative purposes, this does not mean that it has no practical applications. Notably, two alternative applications of those methodologies have been proposed. One envisages that either of those is used; whilst the other their combined use, which would enable LCA studies to give recommendations also on the location and the scale of the process. Such proposal responds to some authors' plea of an integration of LCA and HERA tools.

Chapter 5.
Assessing the impacts of reprocessing Used Nuclear Fuels: a UK case study

This Chapter is divided in two main Sections, each with a different objective. First, a comprehensive LCA study on the current approach for managing Used Nuclear Fuels (UNFs) and the UK Government policy for disposal of higher activity wastes is presented. In the UK, a "nominal" Twice-Through Cycle is implemented whereby Used Nuclear Fuels (UNFs) are reprocessed, but uranium and plutonium are not recycled – they are stored pending a future decision by the Government. By contrast, the Government is clear on the policy for managing higher activity wastes that envisages their disposal in a Geological Disposal Facility (GDF). After failure in 2013, consultations for siting a repository have recently restarted, but the repository will not be available, at the earliest, for several decades. The underpinning purpose of the study is to inform policy and decision-makers concerned with decisions on the future of the UK Nuclear Fuel Cycle. The study relies on a combination of operational data collected during a short secondment to the Sellafield site and literature data on the GDFs, and on a number of assumptions regarding the GDF design and disposal of higher activity wastes. Results include normalised impact scores and detailed hot-spot and sensitivity analyses; and revealed that a great proportion of impacts can be linked to two specific causes represented by indirect burdens from production of uranyl nitrate, which is used to separate plutonium from uranium, and copper, used as the outer layer of the disposal canister for High Level Waste (HLW). The second part of the Chapter builds upon the analysis of radiological impact methodologies presented in the previous two Chapters. Thus, focusing on radiological impact, this part presents a comparison between the methodologies developed within this Thesis, namely UCrad and the Critical Group Methodology (CGM), and with the site-specific methodology employed by Sellafield Ltd. in the annual report "Monitoring Our Environment". The comparison shows how conceptually different approaches lead to significant discrepancies

in both absolute values of radiological impacts and radionuclides' contribution. UCrad and CGM produce very different radiological impacts; however, these represent upper and lower estimates of radiological impacts within which results of more accurate (i.e. site-specific) methodologies, such as the Sellafield site-specific methodology, are anticipated to be found.

5.1 Introduction

As noted in the introductory Chapter, the UK has been a pioneer in the development of nuclear energy for many decades after the Second World War. It established the world's first civil nuclear programme with the opening in 1956 of Calder Hall, the first nuclear power station to deliver electricity in commercial quantities (although its primary purpose was the production of weapon-grade plutonium), and it fostered development of two gas-cooled nuclear reactor designs, namely Magnox and its successor AGR (Advanced Gas-cooled Reactor). With closure of the last Magnox reactor (Wylfa 1) 2015, at present the UK has a fleet of 15 operating nuclear reactors including 14 AGR and 1 PWR (Pressurised Water Reactor) [303]. At its peak, in 1997, nuclear energy was contributing 26% to UK domestic electricity production, but since then several reactors have reached their end-of-life, and nuclear contribution has declined to 21% in 2017 [42]. Further to this, almost 90% of this capacity, that is all the AGR reactors, it is anticipated will be retired by 2030. The structure of the nuclear industry has been complicated by various divestments and changes in ownerships in recent years. Apart from uranium mining and milling, the UK has full fuel cycle facilities and is self-sufficient in both the front- (conversion, enrichment and fuel fabrication) and the back-end (reprocessing and waste treatment) [43].

With current available technologies two nuclear fuel cycles are possible (see Chapter 1). The "Once-Through Cycle" (OTC or "open cycle") envisages that nuclear fuel passes only once through power reactors; Used Nuclear Fuel (UNF) is classified as waste (and is usually referred to as Spent Nuclear Fuel, SNF) and sent for disposal. The other option, namely "Twice-Through Cycle" (TTC or "closed cycle"), aims at recycling the considerable amount of unused fissile material in UNF, which is reprocessed, and uranium (RepU[14]) and plutonium (Pu) separated from fission products and reused for electricity generation purposes. The most common approach envisages mixing uranium (U) and plutonium (Pu) to produce Mixed Oxide Fuel (MOx). Today, the majority of countries that have a nuclear generation programme either follow an open cycle or have yet to take a decision. France is one of few examples of countries operating a full TTC , along with China and Japan that are currently building their own reprocessing plants [304]–[306]. Nominally, the UK is also

[14] Note that RepU specifically refers to Reprocessed Uranium, which has specific features that differ from other forms of uranium such as uranium from mining (referred to as NatU), Low Enriched Uranium (LeU) and so forth.

pursuing the same route, with UNFs currently being reprocessed at the Sellafield site in two reprocessing plants – Magnox and THORP (Thermal Oxide Reprocessing Plant) – dealing respectively with metal and oxide uranium-based fuel. However, at present RepU and Pu are not reintroduced into the fuel cycle, rather they are stored at Sellafield pending a future decision by the Government on their fate.

Radioactive solid wastes are produced throughout the whole fuel cycle, but especially from the back-end activities. They are classified according to radioactivity and heat generation level in High Level Waste (HLW), Intermediate Level Waste (ILW), Low Level Waste (LLW) and Very Low-Level Waste (VLLW) [48]; notably, HLW and ILW are together known as higher Active Wastes (HAWs). In practice, HLW contains the bulk of the fission product following vitrification; ILW is largely made up of sheared claddings and Plutonium Contaminated Materials (PCM); and V/LLW mostly comprises discarded equipment, tools, protective clothing or materials from decommissioning activities. Nowadays, management and disposal of solid nuclear waste constitutes a much debated topic. While V-LLW and some ILW (typically short-lived ones) can be disposed in near-surface repository (e.g. the Low Level Waste Repository at Drigg, UK), HLW and ILW require a different treatment. Many options have been investigated – including disposal in space and deep sea, incineration and direct injection into rock (e.g. see the recommendations of the Committee on Radioactive Waste Management to the UK government [307]) – and it is the intention of the UK [308], and most other countries [309], to dispose of higher activity wastes in deep repositories built several hundred meters underground in a geologically stable environment, usually known under the name of Geological Disposal Facilities (GDFs). However, not a single GDF has yet been built in the world; Finland and Sweden closely followed by France are expected to be the first countries to complete construction of GDFs. After failure of the 2013 consultation exercise in the UK, the process to decide on siting a repository was reviewed to be then restarted in 2018 [310]. According to the timeline set up by the revised process, construction of the GDF is not expected to start for at least 25 years; with construction and operation of the facility projected to last for approximately 100 years [308].

In view of the current situation, the nuclear industry finds itself at a turning point where crucial decisions need to be taken. Life Cycle Thinking (LCT) approaches may be used to

support the decision-making process by assessing and comparing alternative options according to one of the three pillars of sustainability (see Chapter 1).

This Chapter presents a comprehensive Life Cycle Assessment (LCA) study on the current approach for managing Used Nuclear Fuels (UNFs) in the UK, with the aim of informing policy and decision-makers concerned with the back-end of the nuclear fuel cycle. The study focuses on management of AGR used fuels; it includes reprocessing in THORP at Sellafield site and assumes that higher activity wastes are sent for disposal in a national Geological Disposal Facility (GDF). In accordance with current UK policy, uranium and plutonium recovered from used fuels are not considered wastes nor valuable products, thus falling outside the scope of this study. (Uranium and plutonium management strategies are analysed in Chapter 6). Besides assisting the decision-making process, results of an LCA study on the nuclear industry may also be used to improve public attitudes towards the nuclear energy, provided that they are utilized in an open and transparent way.

Until now, a small number of LCA studies on the nuclear industry have been performed [154], [220], [311]–[313]; as highlighted in Chapter 3, the main obstacle is represented by inclusion of ionising radiation within the LCA framework for impact assessment (LCIA). In fact, not one reported LCA study has considered the impacts of radionuclide releases from operation of nuclear reactors, let alone from nuclear waste.

This Chapter also aims at demonstrating, by practical application, the methodologies for radiological impact assessment developed within this Thesis. Because UCrad represents a better candidate for incorporation of radiological impact in Life Cycle Impact Assessment (LCIA), the LCA study includes UCrad impact categories for direct discharges and nuclear waste alongside non-radiological impact methodologies. However, in the second part of the Chapter a detailed comparison is performed between radiological impacts obtained through UCrad, the Critical Group Methodology (CGM) and also the site-specific methodology developed by Sellafield Ltd (SL) and used in the annual report “Monitoring Our Environment” [51].

This Chapter is divided into two main Sections, focusing respectively on the LCA study (Section 5.2) and on radiological impact assessment methodologies (Section 5.3). Section 5.2 is organised as follows: the LCA methods including definition of the goal and scope of the study, system boundary, allocation methodology and impact categories are presented

in Section 5.2.1; Section 5.2.2 includes results of normalisation, hot-spot and sensitivity analyses, which lead to a discussion presented in Section 5.2.3. With respect to Section 5.3, first results of CGM are analysed and compared with those obtained by UCrad (Section 5.3.1), and then the comparison is extended to include SL site-specific methodology (Section 5.3.2); results of the analysis are discussed in Section 5.3.3. The Chapter ends with Section (5.4) that discusses and summarises the main findings.

5.2 Life Cycle Assessment

5.2.1 Methods

5.2.1.1 Goal and Scope

The goal of this study is to quantify and evaluate the environmental impacts associated with reprocessing of Used Nuclear Fuels (UNFs), by means of a prospective attributional (see Chapter 2) LCA focusing on the UK ongoing procedure, and the agreed, but not yet implemented, policy for disposal of nuclear waste. (Notably, the prospective feature is linked with the fact that the study is looking forward to a time when disposal of nuclear waste is implemented.) Of the two types of UNFs (Advance Gas-cooled Reactor, AGR, and Light Water Reactor, LWR) treated at Sellafield, this study focuses on the AGR, as it is currently powering all but one reactor of the UK fleet. As the objective is to describe the *status quo*, neither effects of future decisions nor potential procedures which are outside current UK Government policy are contemplated. In this study, commissioning and decommissioning are not taken into account for existing facilities, but they are for future facilities which are part of the current policy. A notable example for which commissioning and decommissioning have been considered is the Geological Disposal Facility (GDF): this is the preferred option for long-term disposal of nuclear waste, but its construction is not expected to start for at least a couple of decades and to be completed within the century [308], [314]. This line of reasoning also explains why the fate of RepU and Pu is not part of the scope of this study: the UK Government has not reached a final decision, and current UK Government policy acknowledges various options for their use or disposal.

The Functional Unit corresponds to the reprocessing of UNFs assemblies containing 1 tonne of uranium pre-irradiation. In practice, this refers to the amount of uranium in fuel assemblies before being loaded into nuclear reactors. The uranium content post-

irradiation is indeed lower due to fission and other transmutation reactions converting uranium into fission products and other actinides.

5.2.1.2 System boundary

Following the methodological approach developed by Clift and co-workers for integrated solid waste management [108] and introduced in Chapter 2, the product system is divided into a Foreground and Background system. The distinction is primarily used for inventory purposes, and it is not pursued in the impact assessment phase, where the focus is on streams rather than systems.

The Foreground system includes all activities from receipt of UNFs at Sellafield to disposal of solid nuclear waste in a GDF. Notably, interim storage prior to disposal of heat-generating High Level Waste (HLW) is not included in the system. This is because the facility designed to interim-store this type of waste has negligible environmental impacts: it releases no routine emissions, and its electricity consumption is minimal. All activities included in the Foreground system take place on-site at Sellafield, except for the GDF, which is assumed to be located elsewhere in England (see Section 5.2.1.3). The Background system includes all processes that supply materials, chemical and energy to support operations activities in the Foreground. The methodological approach and the system boundary are shown in Figure 5.1. The Foreground system consists of the following subsystems: Thermal Oxide Reprocessing Plant (THORP), Waste Treatment Plants (WTPs) and the Geological Disposal Facility (GDF).

THORP

THORP combines all the facilities necessary for reprocessing used uranium oxide fuel under one roof. Its construction was at the time one of the world's most complex civil engineering projects; it began in 1984 and 10 years later the first fuel was sheared. In the system boundary (Figure 5.1) THORP is divided into six subsystems. The Fuel Handling Plant (FHP) is not technically part of THORP; it is a separate building whose functions serve both THORP and Magnox plants. Here, however, the FHP plant has been included in THORP subsystem for practical purposes.

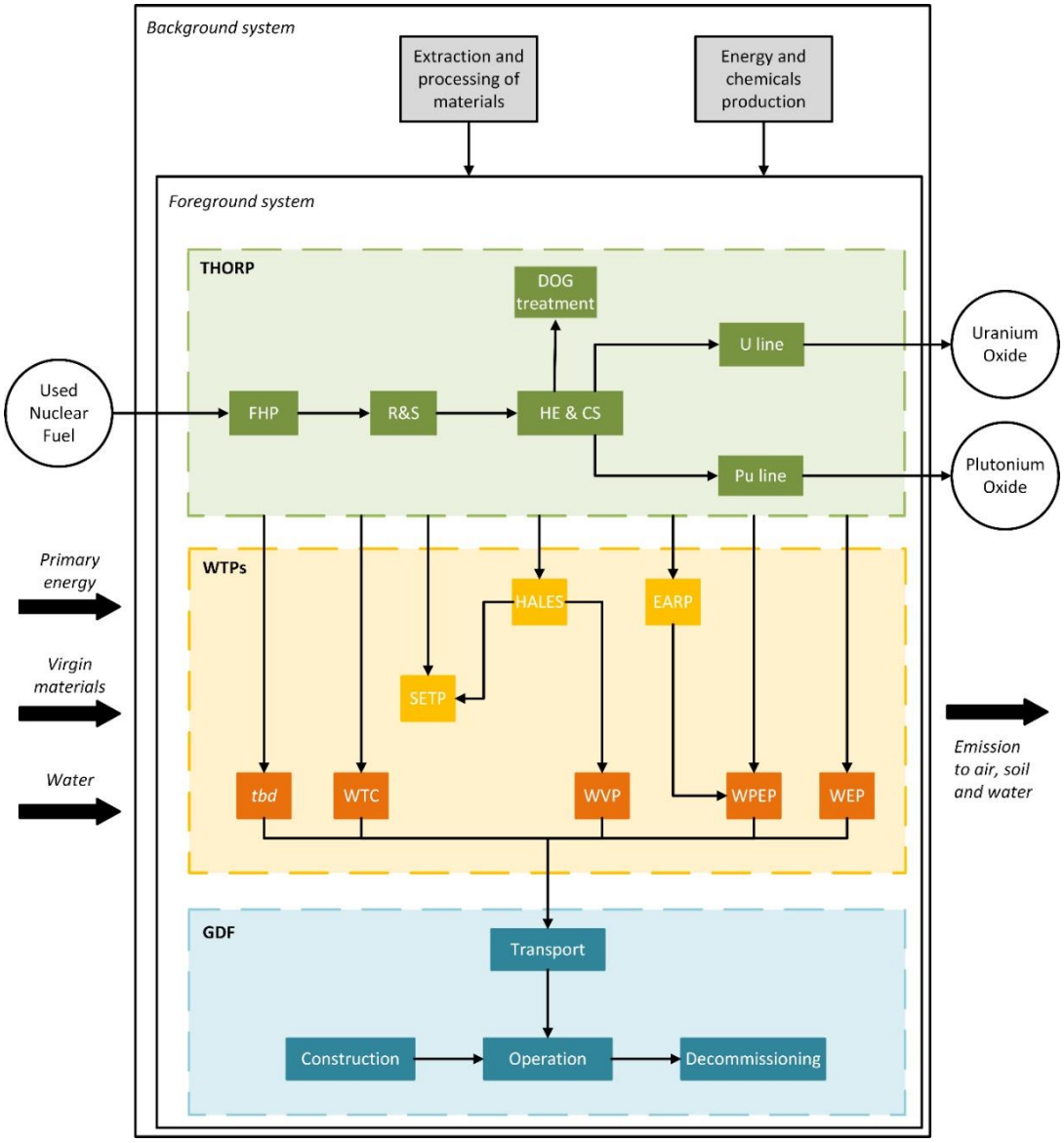

Figure 5.1 – System boundary. A list of acronyms is provided at the end of the Chapter. Note that tbd identifies a non-existing plant, hence to-be-defined (*tbd*). FHP: Fuel Handling Plant; R&S: Receipt and Storage; HE & CS: Head End and Chemical Separation. DOG: Dissolved Off-Gas; WTC: Waste Treatment Complex; SETP: Segregated Effluent Treatment Plant; HALES: Highly Activity Liquid Evaporation and Storage; EARP: Enhanced Actinide Removal Plant; WVP: Waste Vitrification Plant; WPEP: Waste Packaging and Encapsulation Plant; WEP: Waste Encapsulation Plant.

UNF assemblies are transported by rail from nuclear power plants to Sellafield site in transport flasks. The FHP plant is responsible for their receipt, temporary storage and mechanical processing. Fuel assemblies are removed from the transport flasks, wet-stored to allow for complete or partial decay of short-lived products (cooling), and then dismantled. Single pins are removed from assemblies, transferred to stainless steel containers and finally send to THORP Receipt and Storage (R&S), where, if needed, they are further wet-stored to allow for additional radioactive decay. Overall, a minimum of

five years of storage is needed to allow the short-lived fission products to decay further and thus facilitate the handling of UNFs. In the Head End (HE), fuel element assemblies are sheared into nominally five cm lengths via a vertically operated shear blade and collected into baskets. These are moved into dissolvers where nitric acid is used to dissolve uranium (RepU), plutonium (Pu) and Fission Products (FPs). Gas produced by the dissolution process (mainly NOx), namely Dissolver Off-Gas (DOG), is treated by a dedicated system (DOG treatment) to remove radioactive and chemical contaminants prior to atmospheric discharge through the THORP stack. From dissolution, stainless steel claddings, namely "hulls", are removed from the basket and sent to the Waste Encapsulation Plant (WEP). The solution, on the other hand, is clarified and then routed to the Chemical Separation (CS) unit, where the PUREX solvent extraction process, by means of tributyl phosphate in kerosene as the solvent, is employed to separate FPs, RepU and Pu. Firstly, RepU and Pu are removed from the solution by transfer to the solvent; then plutonium is separated from uranium by chemical reduction (of plutonium) to a non-dissoluble form. The solution containing FPs, termed Highly Active Liquor (HAL), is routed to HALES (Highly Active Liquor Evaporation and Storage) for further processing. Pu and U, on the other hand, are further processed via two dedicated lines, where they are purified and then converted into oxide forms representing the final valuable products produced by THORP.

Waste Treatment Plants

In addition to gaseous streams, which are directly discharged through a central stack, THORP routinely generates several liquid and solid waste streams. These are treated by a number of interconnected plants, whose functions are mostly shared with other facilities at the Sellafield site. HALES, SETP (Segregated Effluent Treatment Plant) and EARP (Enhanced Actinide Removal Plant) are some of the plants that deal with liquid effluents, and the only ones considered in this study. HALES concentrates by evaporation and then stores the HAL received from THORP, which is then routed to the Waste Vitrification Plant (WVP). EARP treats low and medium active effluents to reduce their activity level prior to discharge to sea to "as near zero as possible" [315]. It carries out two discrete processes treating Bulks (low active) and Concentrates (medium active) effluents; the latter is the only one considered hereafter, as THORP contribution to Bulks is in practice negligible. Two waste streams leave EARP: the majority of the activity is concentrated in the floc and sent to the Waste Packaging and Encapsulation Plant (WPEP), whilst the purified liquid

stream is directly discharged to sea. Finally, SETP treats low active liquid effluents (acidic, alkaline and suspect active steam condensate streams), not requiring treatment in EARP or SIXEP prior to sea discharge, by removing solids materials and adjusting PH. Other plants which are not considered in the study, either because not being directly involved with THORP or because THORP contribution may be considered negligible, are the Site Ion Exchange Plant (SIXEP), the Solvent Treatment Plant (STP) and the Separation Area Lagoon.

With respect to solid radioactive wastes, these are addressed by a number of other plants. The Waste Vitrification Plant (WVP) converts the concentrated HAL received from HALES into borosilicate glass yielding a stable and durable waste form suitable for long term disposal. The vitrified product is then encapsulated into stainless steel canisters [316], and is currently stored in the Vitrified Product Store (VPS) awaiting construction of the national Geological Disposal Facility. This study assumes that HLW are then packaged into high-integrity disposal canister based on the Swedish KBS-3V concept [53] – the illustrative design is reported in Figure 5.2. Plutonium Contaminated Materials (PCM) – which arise daily from operations in THORP and include PVC gloves, filter, process residues and plant items – are treated by the Waste Treatment Complex (WTC). This provides for their super-compaction into 200l drums, which are then stacked in larger 500 litres stainless steel drums such that there is a cement annulus between the basket and the drum skin. Finally, the Waste Packaging and Encapsulation Plant (WPEP) and the Waste Encapsulation Plant (WEP) immobilize arisings of effluent treatment flocs from EARP and multiple waste streams respectively, within a cement grout (whose composition depends on the form and type of waste) and then provide for their encapsulation into 500 litres stainless steel drums. Waste streams treated by WEP include solid fuel hulls (i.e. cladding), Barium Carbonate (BC) slurry (produced by the C14 removal plant), Multi-Element Bottle (MEB) crud, centrifuge cake (produced by decanting of hulls) and maintenance scrap (from shear cave, basket handling cave crane and other miscellaneous items). Two further waste streams are included in the system boundary, though not being currently treated; these contain respectively AGR Graphite and stainless steel fuel assembly components. Current procedures allow these waste streams to be packaged without encapsulation in grout and stored at the Encapsulated Product Stores (EPS). However, it is the intention of Sellafield Ltd. (SL) to proceed to immobilisation in the near future. In this study, these

waste streams are assumed to be treated in a plant to-be-defined (*tbd*), immobilised in a cement matrix and encapsulated in 500 litres stainless steel drums [271].

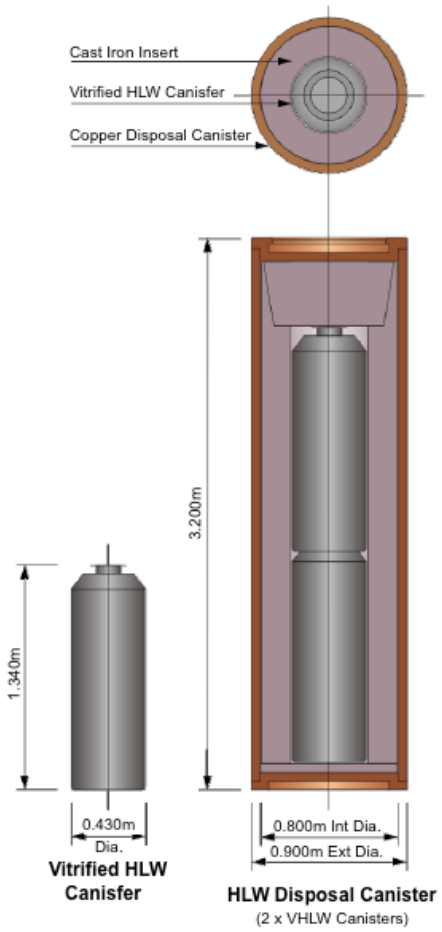

Figure 5.2 – The KBS-3V concept for disposal of HLW, adapted from [317]**.**

Geological Disposal Facility

As shown in Figure 5.1, the Geological Disposal Facility (GDF) subsystem consists of four units. Transportation of packaged waste from waste producer, i.e. Sellafield site, to the repository site is based on the NDA Generic Transport Safety Case [318]. This report is a preliminary assessment of the future transportation system that will be needed when the national GDF will be in operation; it includes considerations on transportation modes and transportation canisters. Three modes of transportation are considered: rail, road and sea; and multiple combinations of them are taken into account. In this study, it is arbitrarily assumed that transportation of waste occurs via rail, and that the distance between Sellafield site (the waste producer) and the GDF is 350 km. This figure represents the average distance between Sellafield site and the centroid of seven zones covering England and Wales, in which the GDF is assumed to be located [318]. Transportation

canisters, in which packaged wastes are envisaged to be transported, are not further considered in this study, due to lack of design data in publicly available literature.

The construction, operation and decommissioning units are based on the NDA's generic GDF design for higher strength rock [319], which is dimensioned to accommodate the UK baseline inventory of radioactive waste [271]–[273]. The design for higher strength rock is taken as an example for use in this case study, and it does not correspond to the preferred option of the NDA. Ultimately, the design will depend on the results of the consultation process for siting the repository. The construction of a GDF includes excavation of a repository at a depth of 650 metres and building of above- and below-ground facilities. The operational phase consists of emplacement of waste and backfilling of its surroundings. The NDA design envisages that ILW/LLW are placed in disposal vaults and HLW in vertical deposition holes drilled along a series of disposal tunnels. Backfilling is carried out each time a disposal vault, a single deposition hole and a tunnel has reached full capacity. The Nirex Reference Vault Backfill (NRVB) is used for ILW/LLW [320], bentonite for deposition holes and a mixture of crushed rocks and bentonite at 70:30 ratio for HLW tunnels [319]. Finally, decommissioning of GDF involves progressive backfilling of remaining tunnels, facilities, shafts and drifts to seal the repository after end of its operational life; the same backfilling material as for HLW is assumed to be used [319].

5.2.1.3 Life Cycle Inventory

According to the distinction made in Section 5.2.1.2 between Foreground and Background systems, different types of data are used. Operational data gathered by the Author on site at Sellafield are used for most of the units in the Foreground system. Unit-specific operational flowsheets supplemented by personal communications with Sellafield Ltd. (SL) technical teams constitute the basis upon which chemicals consumption, RepU oxide production and all streams linking units within THORP subsystem and between THORP, SETP, HALES and EARP have been estimated. No specific data was released by Sellafield Ltd. on the plutonium line; thus, production of plutonium oxide has been estimated from industry average data, according to which UNFs post-irradiation contains around 1% plutonium (and ~3% FPs – 96% U) [321]. With respect to environmental discharges, data for both liquid and air emissions for the year 2017 – which is made publicly available by the Environment Agency – have been obtained from SL Environmental team. Finally, data on electricity consumption has been estimated from Sellafield site annual consumption

for each plant. Notably, electricity and process steam are supplied to Sellafield by a dedicated 188 MW CHP plant (Fellside), which is located just outside the Sellafield site perimeter. This plant is part of the Background system and has been modelled by average data from the Ecoinvent database [251].

For solid waste, a different approach has been used. Streams linking THORP to WEP, WPEP, WVP, WTC and *tbd* (defined in Section 5.2.1.2) have been estimated from the 2016 UK Radioactive Waste Inventory [322]. This represents a coarse, but comprehensive report of the total UK Inventory for radioactive waste with a breakdown on type and source, from which it has been possible to estimate the production of specific types of waste per amount of fuel reprocessed. Data on solid waste conditioning and encapsulation, disposal canister and the specific activity of each waste type have been obtained from the 2007 Derived Inventories [272], [273]. Notably, the specific radioactivity of wastes does not correspond to the time at which wastes are produced, rather to that at which they are disposed of. Immediate disposal is assumed for both ILW and LLW, whilst storage time of 50 years prior to disposal is assumed for HLW.

Due to lack of an operational GDF in the world, data for construction, operation and decommissioning have been retrieved from the NDA's generic GDF design for higher strength rock [319] and supplemented with the Ecoinvent database for nuclear energy [253], e.g. for operations of construction equipment.

Finally, according to the approach generally adopted in attributional studies, the Background system is modelled with average market data, obtained from the Ecoinvent database version 3.3. However, because several chemicals used in THORP, including barium nitrate, gadolinium nitrate and uranyl nitrate, are not covered in the database, they have only been accounted for the burdens of the reagents required for their production, according to stoichiometric ratios. An example is uranyl nitrate, which is produced from yellowcake (i.e. uranium ore) and nitric acid (50% wt/wt) in a ~3.3:1 moles ratio.

Data used for the Foreground system is included in Section B.1 of Appendix B.

Allocation

Sellafield is one of the most complex industrial sites in Europe, with numerous inter-related plants and many activities taking place. UNFs reprocessing at the THORP and

Magnox plants is the most significant, but not the only activity. This implies that Waste Treatment Plants (WTPs) do not function exclusively to support a specific plant, such as THORP, rather they support multiple activities by treating a specific type of effluent or waste. WTPs may thus be described as multi-input processes, for which the problem of allocating environmental burdens to one specific activity arises. Allocation is indeed one of the most debated methodological issues in LCA [121], [323], [324]. As discussed in Chapter 2, the International Organisation for Standardisation (ISO) recommends that whenever possible allocation should be avoided by using system expansion or subdivision [84]. Nonetheless, given the peculiarity of the processes investigated and the consequent lack of data for system expansion purposes, the allocation approach based on physical partitioning of environmental burdens has been used. Three criteria have been employed according to data availability and type of information (e.g. material consumption, emission, etc.). Radioactivity has been the preferred criterion to allocate radioactive emissions (either liquid or gaseous) to a specific unit, when radioactive compositions of all feed streams to a multi-input activity are known. An allocation factor has been calculated for each radionuclide based on the contribution of a process unit to the multi-input process (as percentage of the total input for each radionuclide). For instance, this approach has been used for allocating sea water discharges from SETP to THORP. Allocation based on mass flow of feed streams has been applied when information on the radioactive compositions of waste streams is not complete or when the objective is to estimate consumption of reagents and electricity. For instance, this approach has been used for allocating electricity consumption, but also atmospheric discharges from WEP, which treats several solid waste streams. Finally, in case very little information on the feed streams is known, the allocation criterion has been based on the number of streams feeding to the multi-input process, e.g. for estimating chemicals consumption of EARP.

5.2.1.4 Impact categories

This study adopts the mid-point approach based on the ILCD (International Life Cycle Data System) recommendations [122], [123] (see Chapter 2) with all impact categories, but Land Use and Ionising Radiations, included. Notably, the Land Use category has been considered irrelevant due to lack of data for the Foreground system, whilst the Ionising Radiations category has been replaced by categories based on the UCrad methodology presented in Chapter 3. Table 5.1 reports the impact categories considered, along with their metrics and acronyms used in charts included in the results Sections.

Table 5.1 – Impact categories analysed

Impact category	Metric	Acronym
Acidification	[Mole of H+ eq.]	A
Climate Change	[kg CO_2 eq.]	CC
Ecotoxicity freshwater	[CTUe]	ECf
Eutrophication freshwater	[kg P eq.]	Ef
Eutrophication marine	[kg N eq.]	Em
Eutrophication terrestrial	[Mole of N eq.]	Et
Human Toxicity, Cancer effects	[CTUh]	HT-c
Human Toxicity, Non-Cancer effects	[CTUh]	HT-nc
Ionizing Radiations	[Bq U235 air eq.]	IR
Ionizing Radiations, waste	[Bq U238 ILW eq.]	IRw
Ozone Depletion	[kg CFC-11 eq]	OD
Particulate Matter/Respiratory Inorganics, human health	[kg PM2.5 eq.]	PM/RI
Photochemical Ozone Formation, human health	[kg NMVOC]	POF
Resource Depletion, mineral, fossils and renewables	[kg Sb eq..]	RDm
Resource Depletion, water	[m^3 eq.]	RDw

5.2.1.5 Normalisation

Results of the impact assessment phase have been normalised according to factors developed by the JRC for the ILCD method [298], and supplemented by those developed for UCrad in Chapter 4. It must be noted that normalisation factors for the Ionising Radiation category for nuclear waste have not been developed due to lack of an existing operational GDF.

5.2.1.6 Sensitivity analysis

Finally, sensitivity analyses have been performed on a subset of the model parameters with the aim of assessing reliability and enhancing quality of the LCA results. The focus has been on those parameters considered to be the most uncertain; these include: amounts of both liquid and solid waste streams generated by THORP and treated by WTPs; plants' electricity consumption and direct radioactive emissions by plant and per type of waste treated in case of multi-input processes. The amounts of all waste streams have been estimated from the UK derived radioactive inventory [271]–[273] with the exception of Highly Active Liquor (HAL), obtained through communications with THORP technical team (Section 5.2.1.3). Electricity consumption and direct radioactive emissions have been obtained by different allocation methods, described in Section 5.2.1.3.

A one-at-a-time approach has been adopted whereby each parameter is changed by an arbitrary value equal to a 1% positive variation, while all others being kept constant. The percentual change of the results, named multiplier after Heijungs and Suh [325], represents the magnitude and the direction of changes in impact categories due to variations in the model parameters. For linear systems, multipliers are within 0-1 range (for positive variation); values higher than unity are distinctive of non-linear systems where small perturbations in input parameters are amplified in the results. An uncertainty analysis could not be carried out due to the unavailability of data regarding the level of uncertainty of the collected data.

5.2.2 Results

The results Section is divided in three sub-Sections: Section 5.2.2.1 presents normalised scores, Section 5.2.2.2 reports comprehensive hot-spot analyses for the product system and the most relevant sub-systems, and finally Section 5.2.2.3 includes sensitivity analyses for amount of waste streams generated by THORP, electricity consumption and radioactive emissions. Unless otherwise stated, the LCA results refer to both the foreground and background system. They have been calculated by means of Gabi sustainability software version 8 [326]. Absolute impact scores for the product system and each main sub-system are only included in Section B.2 of Appendix B.

5.2.2.1 Normalised impacts

Figure 5.3 shows normalised impacts expressed in terms of person equivalent (pe). Ionising Radiations from direct discharges (IR) features the highest normalised impact at 1.4E4 pe, around one order of magnitude higher than toxicity (i.e. Ecotoxicity freshwater, ECf, and Human Toxicity, cancer (HT-c) and non-cancer, HT-nc, effects) and resource depletion of minerals, fossils and renewables (RDm) categories that follow in second place. Freshwater (Ef) has the highest value (approximately equal to 2.5E2 pe) amongst eutrophication categories, followed by marine (Em) and terrestrial (Et). Acidification (A), Climate Change (CC), Photochemical Ozone Formation (POF) and Resource Depletion water (RDw) feature comparable values, at around 3-6E1 pe, with Particulate Matter/Respiratory Inorganics (PM/RI) slightly higher, at 1.1E2 pe. The lowest normalised impact is shown by Ozone Depletion (OD) at 1.9 pe.

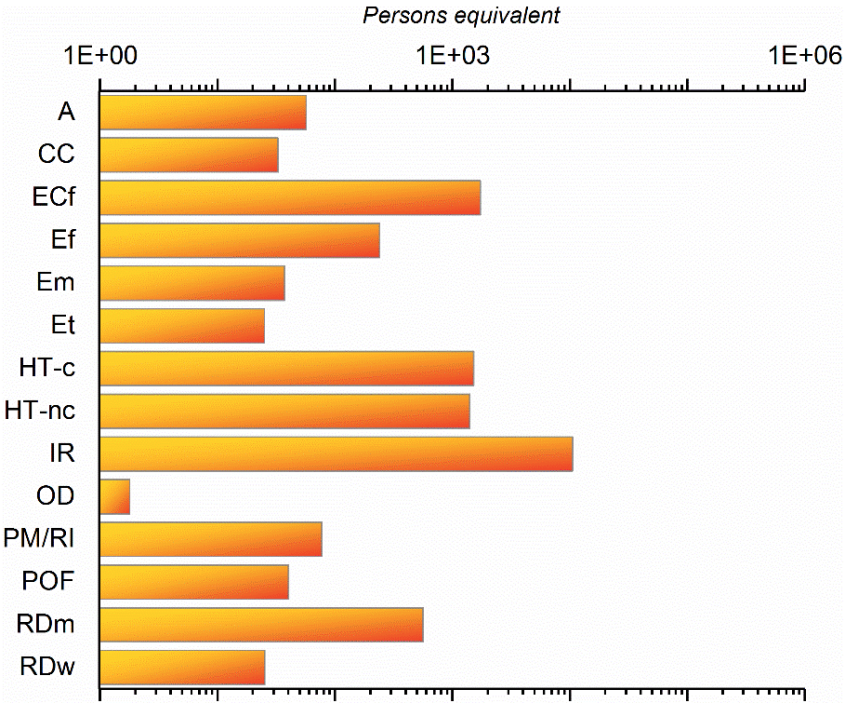

Figure 5.3 – Normalised impacts for the whole product system.

5.2.2.2 Hot-spot analysis

Main results of the hot-spot analysis are reported in Figure 5.4 to Figure 5.12. Figure 5.4 shows contributions of each main subsystem, that is THORP, WTP and GDF (see Figure 5.1), to the overall impact for each category including both Foreground and Background activities. The bar chart shows that THORP has the largest share of impacts in CC, Em, Et, IR, OD, PM/RI, POF and RDm. Notably, it is responsible for ~90% of impacts from ionising radiations (IR), for 36-40% of Et, OD and PM/R, and for over half of the remaining impact categories.

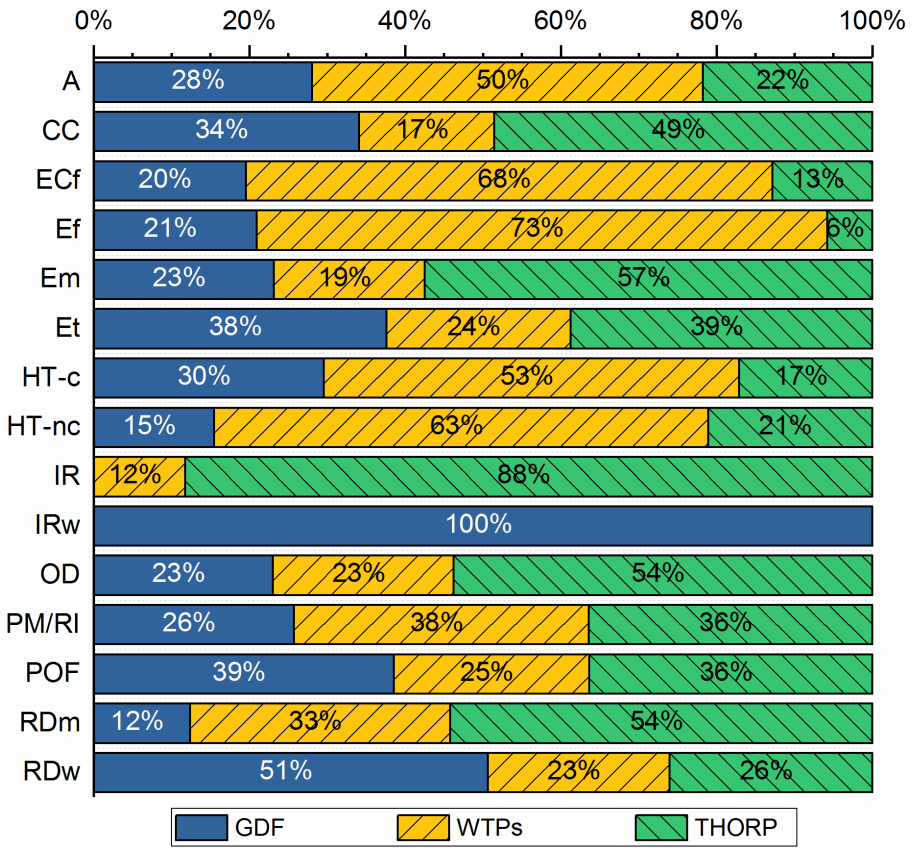

Figure 5.4 – Hot-spot analysis of the three main subsystems

Figure 5.5 shows a detailed hot-spot analysis, including both impacts from direct and indirect burdens due to production of fuel, electricity and chemicals, of THORP subsystems excluding the FHP plant whose impacts are negligible; these include R&S, HE, CS, and U and Pu lines (see Figure 5.1). Indirect burdens associated with consumption of uranyl nitrate emerge to be the major cause of impacts. They are, in fact, alone responsible for over 70% HT-c and -nc, ECf, Em, PM/RI and RDm impacts, and for more than half of all other impacts except RDw (at 32%) and CC and OD (at 20%). For those (CC and OD), indirect burdens from electricity consumption represent the main source of impacts, at 52-56%. Direct emissions seem not to have significant impacts in any category but Ionising Radiation (IR), where they along with indirect emission from uranyl nitrate production are the sole sources responsible for the category score, at 79 and 21% respectively. Figure 5.6 reports a detailed breakdown of the main radionuclides contributing to radiological impacts from atmospheric emissions; discharges to coastal waters are negligible. Iodine-129 (I129) features the largest contribution (~78%), followed by Kr85 at 21.6%; remaining radionuclides have negligible impacts (0.1%).

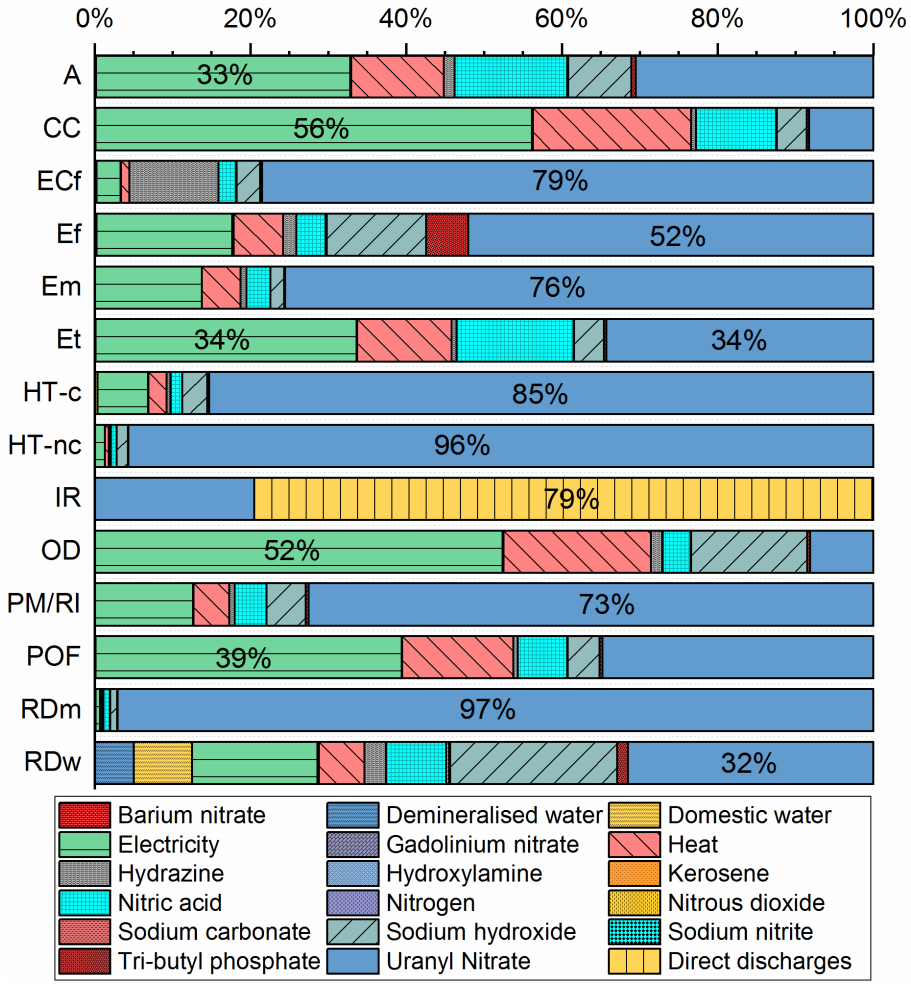

Figure 5.5 – Detailed hot-spot analysis for THORP subsystem, including indirect and direct burdens

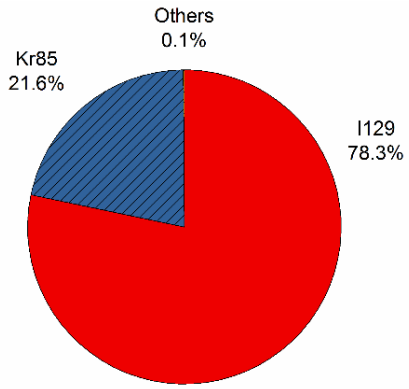

Figure 5.6 – Radionuclides breakdown for Ionising Radiation impacts from THORP atmospheric discharges

As illustrated in Figure 5.4, WTPs subsystem is the largest contributor to A (50%), ECf (68%), Ef (73%), HT-c (53%) and HT-nc (63%) impact categories, has minor (~10%) contributions to IR, and none to IRw; and is responsible for approximately 20-30% of the remaining categories' score.

The hot-spot analysis for WTPs (Figure 5.7) shows that the majority of these impacts are caused by two specific plants. The Waste Vitrification Plant (WVP) may be regarded as the sole cause (i.e. with shares above 95%) to impacts in ECf, Ef, HT-c and -nc, and RDm categories; it contributes with 88 and 89% to PM/RI and A respectively, and with more than 70% to Em, Et and POF. The Segregated Effluent Treatment Plant (SETP), on the other hand, is the largest contributor to IR (72%) and OD (50%) categories. Finally, SETP and WTP (the Waste Treatment Plant) together are responsible for more than 90% of water consumption (RDw).

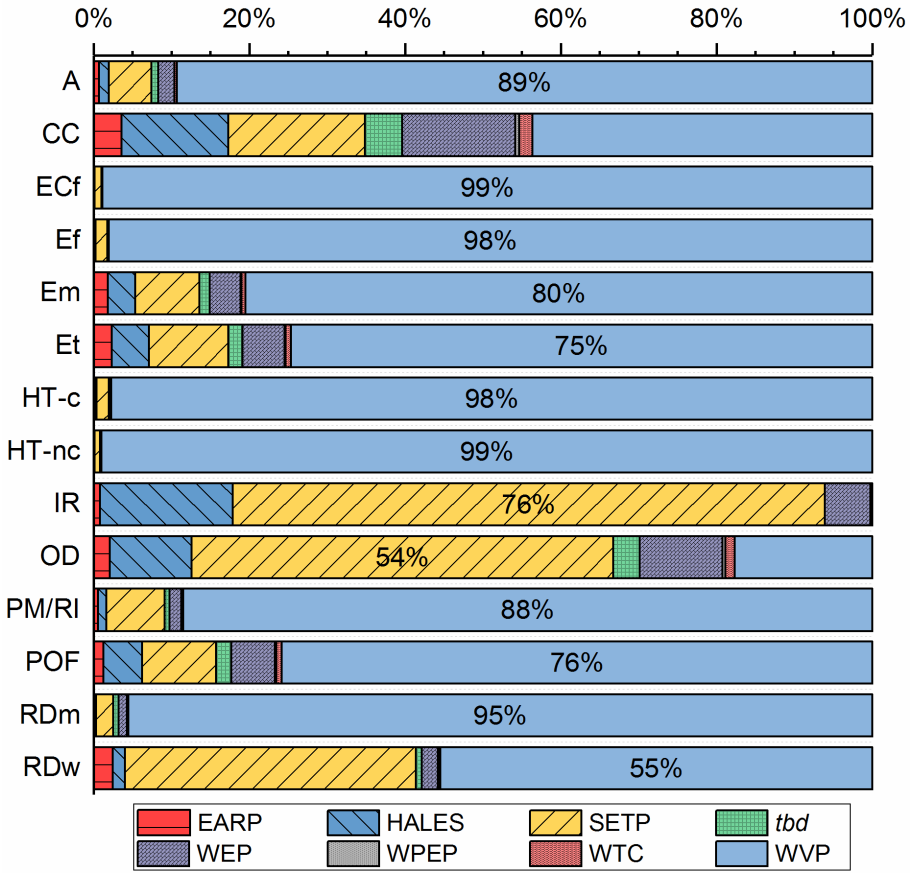

Figure 5.7 – Detailed hot-spot analysis of the Waste Treatment Plants (WTPs) subsystem

Figure 5.8 and Figure 5.9 illustrates hot-spot analyses for the Waste Vitrification Plant (WVP) and the Segregated Effluent Treatment Plant (SETP).

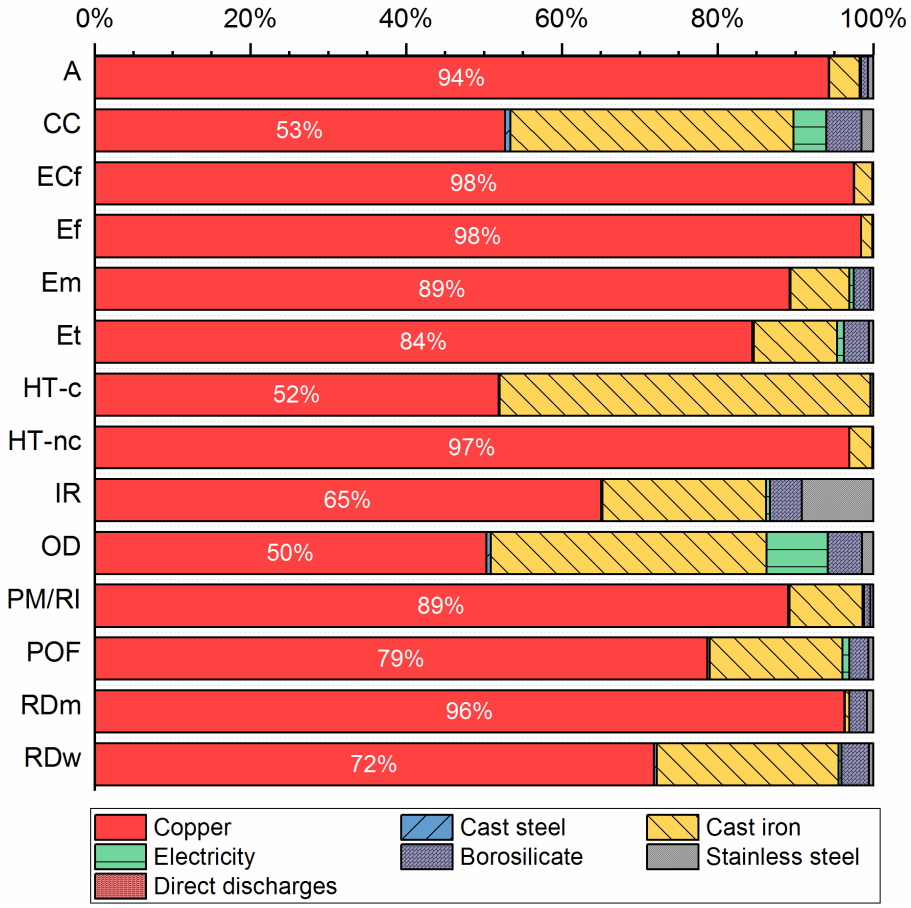

Figure 5.8 – Detailed Hot-spot analysis of the Waste Vitrification Plant (WVP)

Indirect burdens from production of copper and cast iron used in the disposal canisters for HLW are jointly responsible for the majority of WVP impacts, from 85% up to around 99%. Notably, copper contribution ranges from as low as ~50% in CC, HT-c and OD, up to 96-98% in ECf, Ef, HT-nc and RDm; whilst cast iron has substantial contributions (34-47%) to CC, HT-c and OD, and negligible (below 5%) to A, ECf, Ef, HT-nc and RDm. Remaining sources of impacts include production of borosilicate glass, cast steel, stainless steel and electricity in the Background system, and direct discharges in the Foreground system. Amongst these, only stainless steel has significant contribution, equalling 15% to IR category.

Indirect burdens associated with production of sodium hydroxide is the primary source to non-radiological impacts in SETP, with contributions ranging from 95% up to ~100%; radiological impacts are entirely attributable to direct discharges to sea.

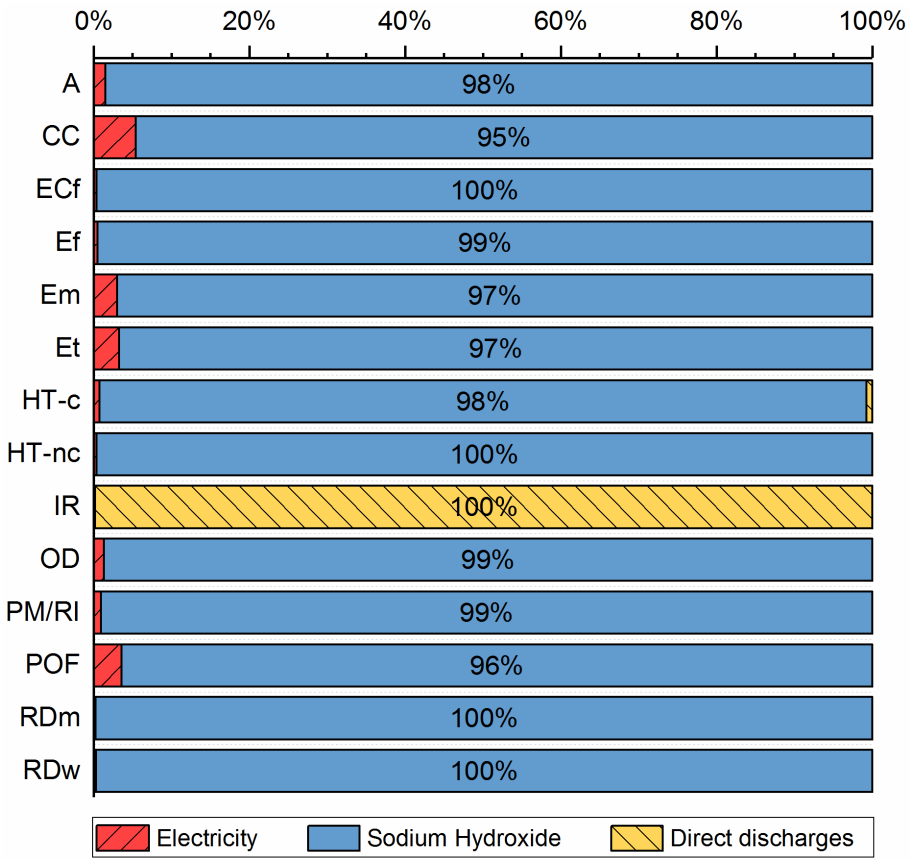

Figure 5.9 – Detailed hot-spot analysis of the Segregated Effluent Treatment Plant (SETP)

With respect to the remaining subsystem, Figure 5.4 shows that the GDF represents a minor contributor (~12%) to RDm and the largest (51%) to RDw category; in the remaining categories it features contributions ranging from 15% to 39%. Figure 5.4 also shows that the GDF is the only cause to radiological impacts from solid waste disposal (IRw), however this should not be of any surprise as it goes along with the definition of the category itself.

A detailed hot-spot analysis of the IRw category is reported Figure 5.10 with a breakdown by waste stream (part A) and radionuclide (part B). The analysis shows that four solid waste streams contribute to over 96% of the overall impact: MEB crud (35%), Vitrified HAL (VHAL, at 32%), AGR cladding (11%) and Centrifuge Cake (CC, at 17%); the remaining wastes contribute with just over 3%. It must be noticed that radioactive inventories for two waste streams namely Plutonium Contaminated Materials (PCM) and maintenance scrap were not available (see Table B.8 in Appendix B). Iodine-129 (I129) and chlorine-36 (Cl36) are the two main sources of impacts at approximately 65% and 29% respectively, with uranium-234 (U234) standing as low as 3.7% and all other radionuclides combined at 2.6%.

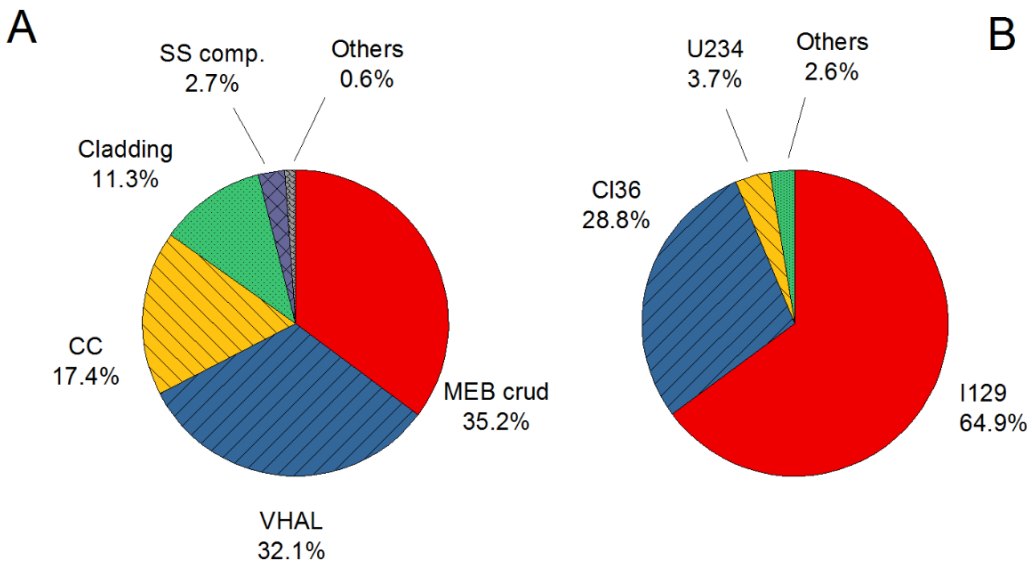

Figure 5.10 – Detailed hot-spot analysis for Ionising Radiation (waste) impact from GDF. A: Breakdown by solid waste streams; B: Breakdown by radionuclides. A glossary of acronyms is included at the end of the Thesis.

Hot-spot analysis of the remaining impact categories (Figure 5.11) shows that the main source of impacts is represented by the Vitrified Highly Active Liquor (VHAL) stream, which contributes with 66% to RDw and with over 95% to the remaining categories. Amongst the other waste streams considered, only AGR Stainless Steel (SS) components and cladding have significant (i.e. over 5%) contributions, at approximately 7 and 15% in RDw category.

Given the significance of the VHAL stream, a detailed hot-spot analysis for construction, transportation, operation and decommissioning phases of the GDF subsystem associated with HLW waste is reported in Figure 5.12. The bar chart shows that both transportation and operational phases have negligible impacts. The GDF decommissioning phase contributes to over half of EM, Et, OD and POF impacts, between 30 and 41% in A, CC, PM/RI and RDm and below 15% in the remaining categories. Impacts are entirely attributable to indirect burdens due to production of bentonite used as backfilling. With respect to the construction phase, major sources of impacts are related to construction of above and below ground facilities (especially ECf, Ef and HT-nc), use of industrial machines (HT-c) and consumption of electricity (IR and RDw).

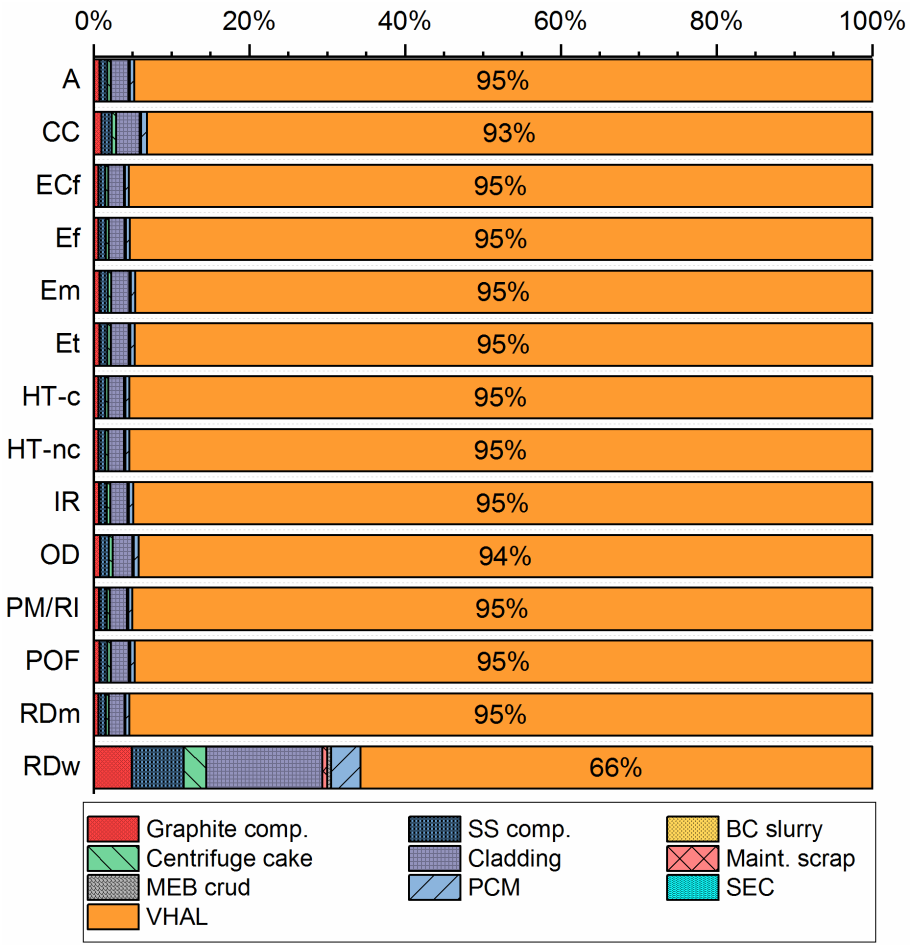

Figure 5.11 – Detailed hot-spot analysis by waste stream of the Geological Disposal Facility (GDF). SS: Stainless Steel; BC: Barium Carbonate

5.2.2.3 Sensitivity analysis

Results of the sensitivity analysis are reported in Table 5.2 to Table 5.4, and expressed in terms of multipliers for each model parameter and impact category. The first and second most influential parameters for each category are highlighted in red and yellow respectively.

Table 5.2 shows that the amount of Highly Active Liquor (HAL) generated by THORP is the most influential parameter for eleven impact categories and the second most for the two Ionising Radiation categories, namely IR and IRw, which are most sensitive to Low Active Effluent (LAE) and MEB crud. Salt Evaporate Concentrate (SEC) and AGR cladding are the first and second most influential streams, respectively, for Resource depletion of mineral and fossil (RDm), with HAL coming in third place, whilst none of the parameters considered affects the RDw category score. Importantly, all multipliers are below unity to

indicate absence of non-linearities in the model. Multipliers of the first and second most influential stream range from as low as 0.22 (HAL to OD) to as high as 0.68 (HAL to Ef).

LCA results appear to be less sensitive to plants' consumption of electricity and amount of radioactive emission. Amongst the plants considered, THORP and HALES result being the first and second most influential respectively for both cases. The highest multiplier for electricity consumption is equal to 0.22, whilst for radioactive emissions is as low as 3.84E-4.

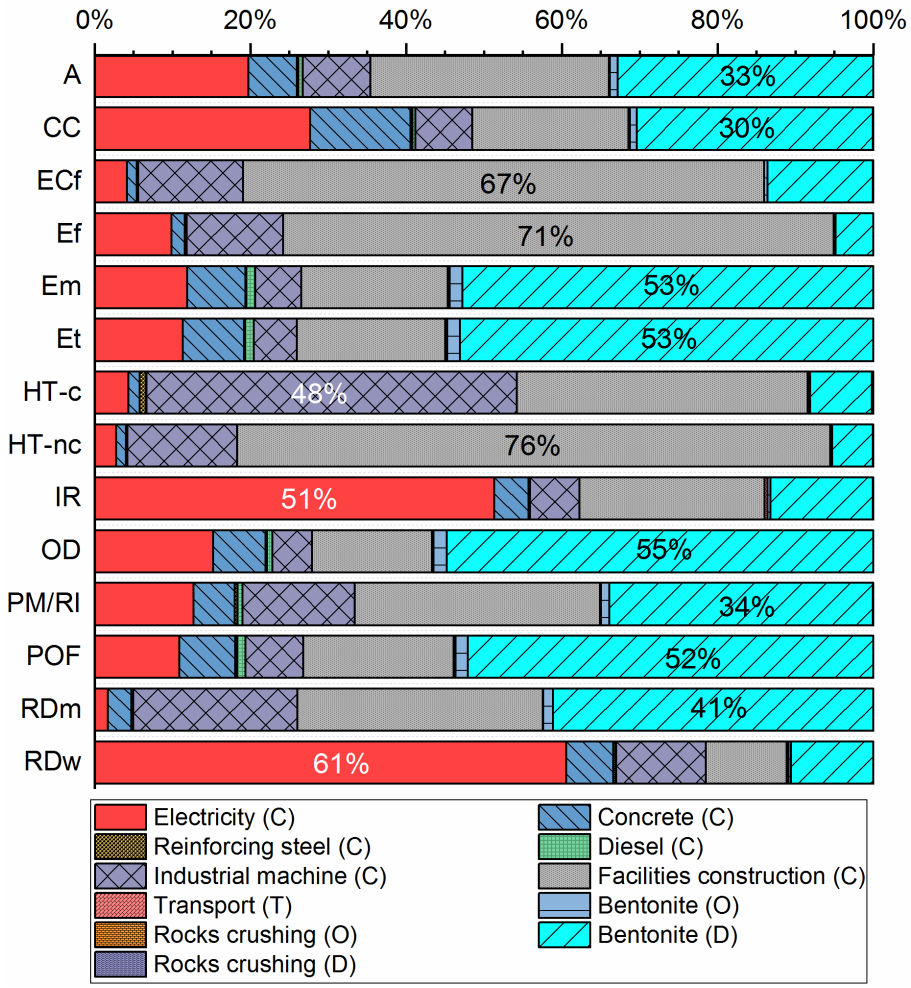

Figure 5.12 – Detailed hot-spot analysis for construction, transportation, operation and decommissioning of GDF for Vitrified HAL. C: Construction; O: Operation; T: Transportation; D: Decommissioning.

Table 5.2 – Sensitivity analysis on THORP waste streams. Red and orange represent first and second most influential parameters for each impact category. (Data is expressed in terms of of multipliers for each model parameter and impact category.)

	AP	CC	Ect	Ef	Em	Et	HT-c	HT-nc	IR	IRw	OD	PM/RI	POF	RDm	RDw
Cladding	9.96E-03	2.20E-02	2.60E-03	3.70E-03	1.14E-02	1.26E-02	3.85E-03	1.91E-03	3.31E-03	1.07E-01	1.97E-02	4.94E-03	1.34E-02	7.06E-02	0
Graphite comp.	2.99E-03	6.13E-03	8.39E-04	1.17E-03	3.29E-03	3.65E-03	1.23E-03	6.18E-04	9.43E-06	5.33E-03	5.32E-03	1.49E-03	3.86E-03	2.34E-02	0
SS Comp.	3.39E-03	5.67E-03	1.10E-03	1.47E-03	3.40E-03	3.76E-03	1.57E-03	8.15E-04	1.18E-05	2.57E-02	4.26E-03	1.72E-03	3.93E-03	3.15E-02	0
Centrifuge cake	1.84E-03	4.00E-03	4.88E-04	6.89E-04	2.08E-03	2.31E-03	7.21E-04	3.59E-04	5.95E-04	1.64E-01	3.58E-03	9.13E-04	2.45E-03	1.34E-02	0
HAL	4.94E-01	3.16E-01	5.27E-01	6.79E-01	3.76E-01	3.65E-01	4.57E-01	4.04E-01	2.82E-02	3.02E-01	2.20E-01	2.84E-01	3.78E-01	6.67E-02	0
LAE	1.87E-02	2.11E-02	3.74E-03	8.46E-03	1.52E-02	1.56E-02	4.80E-03	2.42E-03	3.85E-02	0	9.60E-02	1.37E-02	1.53E-02	0.00E+00	0
MEB crud	3.27E-04	7.26E-04	8.50E-05	1.21E-04	3.74E-04	4.16E-04	1.26E-04	6.24E-05	1.12E-04	3.32E-01	6.57E-04	1.62E-04	4.41E-04	2.32E-03	0
Maint. scrap	4.33E-04	9.56E-04	1.13E-04	1.61E-04	4.93E-04	5.49E-04	1.67E-04	8.27E-05	1.43E-04	0	8.56E-04	2.14E-04	5.80E-04	3.06E-03	0
PCM	2.12E-03	4.05E-03	6.31E-04	8.63E-04	2.26E-03	2.50E-03	9.13E-04	4.66E-04	6.95E-06	4.05E-03	3.34E-03	1.07E-03	2.63E-03	1.78E-02	0
SEC	0	0	0	0	0	0	0	0	0	3.86E-02	0.00E+00	0.00E+00	1.31E-01	6.74E-01	0

Table **5.3** – Sensitivity analysis on plants electricity consumption, per type of waste. Red and orange represent first and second most influential parameters for each impact category. Data is expressed in terms of of multipliers for each model parameter and impact category.)

	A	CC	Ect	Ef	Em	Et	HT-c	HT-nc	IR	IRw	OD	PM/RI	POF	RDm	RDw
THORP	4.87E-02	1.94E-01	2.43E-03	7.44E-03	7.75E-02	8.70E-02	6.30E-03	1.42E-03	6.61E-05	0	2.17E-01	2.23E-02	9.50E-02	0	0
HALES	4.24E-03	1.69E-02	2.12E-04	6.47E-04	6.74E-03	7.58E-03	5.49E-04	1.23E-04	5.75E-06	0	1.89E-02	1.94E-03	8.27E-03	0	0
EARP	6.28E-07	2.50E-06	3.14E-08	9.60E-08	1.00E-06	1.12E-06	8.14E-08	1.83E-08	8.53E-10	0	2.80E-06	2.88E-07	1.23E-06	0	0
SETP, from HALES	2.90E-04	1.15E-03	1.45E-05	4.43E-05	4.61E-04	5.18E-04	3.75E-05	8.43E-06	3.93E-07	0	1.29E-03	1.33E-04	5.65E-04	0	0
SETP, from THORP	4.58E-06	1.82E-05	2.29E-07	7.01E-07	7.30E-06	8.20E-06	5.94E-07	1.33E-07	6.22E-09	0	2.04E-05	2.10E-06	8.95E-06	0	0
WVP	5.65E-04	2.25E-03	2.82E-05	8.63E-05	8.99E-04	1.01E-03	7.31E-05	1.64E-05	7.66E-07	0	2.51E-03	2.59E-04	1.10E-03	0	0
WEP – SEC	1.49E-04	5.93E-04	7.44E-06	2.28E-05	2.37E-04	2.66E-04	1.93E-05	4.34E-06	2.02E-07	0	6.64E-04	6.82E-05	2.91E-04	0	0
WEP – Cladding	3.26E-03	1.30E-02	1.63E-04	4.98E-04	5.19E-03	5.83E-03	4.22E-04	9.49E-05	4.43E-06	0	1.45E-02	1.49E-03	6.36E-03	0	0
WEP – BC slurry	4.54E-07	1.81E-06	2.27E-08	6.94E-08	7.23E-07	8.13E-07	5.89E-08	1.32E-08	6.17E-10	0	2.02E-06	2.08E-07	8.87E-07	0	0
WEP – Centrifuge cake	5.86E-04	2.33E-03	2.93E-05	8.96E-05	9.34E-04	1.05E-03	7.60E-05	1.71E-05	7.96E-07	0	2.61E-03	2.69E-04	1.15E-03	0	0
WEP – Maint. scrap	4.87E-02	1.94E-01	2.43E-03	7.44E-03	7.75E-02	8.70E-02	6.30E-03	1.42E-03	6.61E-05	0	2.17E-01	2.23E-02	9.50E-02	0	0
WEP – MEB crud	4.24E-03	1.69E-02	2.12E-04	6.47E-04	6.74E-03	7.58E-03	5.49E-04	1.23E-04	5.75E-06	0	1.89E-02	1.94E-03	8.27E-03	0	0

Table 5.4 – Sensitivity analysis on radioactive emissions allocated to waste streams treated. Red and orange represent first and second most influential parameters for each impact category. (Data is expressed in terms of multipliers for each model parameter and impact category.)

Source	IR
SETP, from THORP	3.84E-04
SETP, from HALES	1.55E-04
EARP, from SEC treatment	5.98E-06
WEP – Cladding	3.28E-05
WEP – BC slurry	4.57E-09
WEP – Centrifuge Cake	5.89E-06
WEP – Maint. scrap	1.42E-06
WEP – MEB crud	1.11E-06

5.2.3 Discussion

This LCA study presents a comprehensive picture of the environmental performance of the UK approach for managing Used Nuclear Fuels (UNFs), which envisages their reprocessing at the Sellafield site followed by disposal of nuclear wastes into a Geological Disposal Facility (GDF). The combination of impact scores normalisation and contribution analysis allowed identifying the most critical impact categories and the units/flows/pollutants that are their main cause. The sensitivity analysis, on the other hand, contributed to reinforce those results, and to shed light on the influence of critical model parameters on the final results. This study also presents the first application of the UCrad methodology represented by two impact categories concerned with direct discharges and radioactive wastes disposed in a GDF (Chapter 4).

5.2.3.1 Normalisation and hot-spot analysis

From the normalisation step, the Ionising Radiation (IR) category for direct discharges results having the highest relative contribution, by over an order of magnitude, to the overall impact attributed to Europe. This may be surprising given the stringent limitations on radioactive discharges that are put in place both at the country (UK) and continent (Europe) level. It is, in fact, a common approach to regulate radioactive emissions under the principles of “As Low As Reasonably Practicable” (ALARP) and “Best Practicable Means” (BPM); the former forms the cornerstone of nuclear plant operational emissions and the latter represent the means to achieve it. The high normalisation impact of IR may rapidly prompt debate whether radioactive emissions should be the primary target of

mitigation strategies. This may well be the case; however, let us further analyse normalisation scores.

As discussed by Heijungs and colleagues [327], the procedure of normalization may introduce biases "in any direction and with any magnitude", especially in not well established and widely recognized categories dealing with a large number of substances; IR is thus a notable example. The primary cause of biases is incompleteness in either or both inventories for the product/service and the reference system. As shown in Chapter 4, discharges to water of two radionuclides, namely iodine-129 (I129) and cobalt-60 (Co60), dominate the category, by accounting for more than 96% of the total impact. Both these radionuclides, and also the other three having contributions higher than 1% (technetium-99 and caesium-137 discharged to water and iodine-129 discharged in the atmosphere), are included in the inventory data for the product system. The same can be said for the radionuclides contributing the most to the product system; these are direct atmospheric emissions of I129 and Kr85 (krypton-85) from THORP (Figure 5.6), and indirect emissions of Ra226 (radium-222) and Rn222 (radon-222) associated with uranyl nitrate production (Figure 5.5). Therefore, there are no significant inconsistencies in the inventories. Another factor, which was not highlighted by Heijungs and co-workers but that may lead to biases, is inconsistency in the geographical coverage of the product and reference system. The ILCD inventory is an updated version of the "Life Cycle Indicators for Resources" dataset [328], but it takes into account only domestic emissions and resources consumption in spite of the "apparent consumption" approach adopted in the latter, which also considers environmental impacts associated with traded goods. However, over half of the Ionising Radiations impacts are due to indirect burdens for production of Uranyl Nitrate, most of which can be attributed to mining of uranium. As reported by the World Nuclear Association (WNA), most of uranium mines, and all the largest-producing, are located outside of the European Union [47], and thus their impact not covered by normalisation factors based on the ILCD inventory. Therefore, the fact that the product system includes activities occurring outside the European Union causes the overestimation of the IR normalised impact.

Besides normalization biases, normalised impacts also need to be contextualised. Radioactive emissions are primarily, if not exclusively, associated with the nuclear industry, notably with front and back-end activities of the nuclear fuel cycle for power

production; this is in contrast with other classes of pollutants whose emission can be usually attributed to several industries. This peculiarity leads to relative contribution of IR category being higher than, for instance, climate change (CC). This is particularly true if we consider that I129 – a long-lived, natural occurring radionuclide primarily formed from fission of uranium and plutonium in nuclear reactors and dominant radionuclide to IR category – is primarily discharged during reprocessing operations (specifically from the fuel dissolution step [329]) and that in the EU, only France and UK carry out reprocessing on a commercial scale.

Looking at the contribution analysis, IR impacts are in practice equally attributable to direct atmospheric emissions, and indirect emissions associated with production of uranyl nitrate representing the other half. It must be noted that the approach used to modelling production of uranyl nitrate is conservative because only the burdens associated with uranium and nitric acid are considered; if operational data were available, the contributions of uranyl nitrate would be even larger. Uranyl nitrate is used in the Chemical Separation (CS) phase in THORP to separate RepU from Pu by reduction of the latter to the inextractable Pu(III) [48]. It is obtained from uranium and nitric acid, but its impacts, both radiological and non-radiological, are primarily attributable to mining of uranium. More specifically, radiological and toxic impacts are largely caused by management of uranium tailings, whilst remaining impacts by electricity and heat production. Uranium tailings are by-products of the mining process after uranium is removed from the uranium-bearings mineral, and are a source of radioactive emissions, including radium-226 (whose decay product, radon-222, is the main health hazard in uranium mines) various uranium isotopes (U234, U235 and U238) and Thorium-230; and also of metals – e.g. chromium, arsenic, vanadium, zinc and copper – leaching into soil and groundwater. With respect to the other Ionising Radiations category dealing with nuclear waste (IRw), a normalised impact could not be calculated due to the lack of operational GDFs and thus of inventory data for the reference system. Obviously, IRw is exclusively caused by the GDF subsystem (Figure 5.4) due to inherent definition of the category. Four waste streams account for over 95% of the impact, and around two thirds is attributable to I129 that along with Cl36 amounts to ~95%. Notably, only one quarter of iodine-129 is included in HLW, whilst chlorine is a specific feature of I-LLW. It is interesting to compare these results with radiological impacts obtained by the RWM [267]. In their Post-Closure Safety Assessment (PCSA), 100% of impact during 50 thousand years after closure of GDF is

caused by I-LLW; subsequently Depleted/Natural/Low Enriched Uranium (DNLEU) – which are not included in this study – becomes the dominant source of impact. Notably, I129 and Cl36 are the primary causes of impacts for I-LLW wastes. HLW, on the other hand, has very little impact, and this is dominated by caesium-135 (Cs135), which in this study accounts for less than 1%. This is not linked with the amounts of radionuclides disposed of, rather with the methodologies' characterisations factors; as noted in Chapter 4 and as it will be discussed in Section 5.3.3, I129 features a very high characterisation factor in UCrad due to its very long half-life.

Furthermore, it is worth noticing that the two radiological categories discussed above deal with impacts occurring on very different time scales. Whilst radionuclides emitted through liquid and air discharges are readily accessible in the environment, the ones contained in nuclear waste are not. According to RWM's simulations [267], risk arising from GDF is negligible before 2.000 years, steeply increases up to 10.000 years, and then continue raising at lower pace until 1.000.000 years. Therefore, comparison across those categories must be restricted at present, at least until a widely accepted framework for handling long-term emissions in LCA is conceived and accepted. A review on different approaches focused on long-term emissions from landfills [330] has already set the ground for future developments.

With respect to non-radiological impacts, the toxicity categories (i.e. ECf, HT-c and HT-nc) feature the highest normalised score, followed by depletion of renewable and non-renewable resources (RDm), Particulate Matter/Respiratory Inorganics (PM/RI) and Acidification (A). It is interesting to notice that those impacts can be traced back to indirect burdens associated with production of uranyl nitrate and copper. The latter is central to the SKB disposal concept and forms the external layer of the disposal canisters for HLW. Copper is, in fact, predicted to resist corrosion so that disposal canisters remain intact over a 100.000-year time span [53]. Notably, its corrosion rate is remarkably slow in anoxic conditions, which is well understood are the conditions of groundwater at 500m or plus depth [331]. Uranyl nitrate and copper are used in two specific units; the former in the Chemical Separation area of THORP and the latter in the Waste Vitrification Plant (WTP), part of the WTPs subsystem. This is why these subsystems (THORP and WTPs) in Figure 5.5, the Waste Vitrification Plant (WVP) in Figure 5.7, and the vitrified HAL stream in Figure 5.11 are the dominant sources of impact. An element of discussion regards the

reliability of data on uranium mining. This, in fact, has been modelled using the Ecoinvent database that as market activity includes relatively old data, mainly gathered in the 80s, although justified by the claim that mining procedures have not changed much in the last decades.

Impact categories featuring lower normalised values may still be associated with production of uranyl nitrate and copper, however those are not the prevalent sources. This is the case of Eutrophication (marine and terrestrial), Climate Change (CC), Photochemical Ozone Formation and Depletion of water, for which GDF overcome WTPs as being the main contributor along with THORP. GDF's impacts range from 12% (marine eutrophication) to 44% for water depletion (RDw). Potential impacts associated of the GDF are in practice entirely attributable to disposal of HLW (i.e. Vitrified HAL stream), contributing with no less than 90% (Figure 5.11). The Ionising Radiation (waste) category (IRw) and resource depletion of water (RDw) are two exceptions; the former has been discussed above, whilst only two thirds of the latter is attributable to HLW. It is also interesting to notice that impacts due to transportation and operational phase of GDF are in practice negligible (Figure 5.12). This means that use of bentonite for HLW and crushed rocks for ILW as buffers are a negligible fraction of the total amount needed to seal the GDF, and that the location of the GDF (and thus transportation distance) is not a game-changing factor in the overall environmental performance. However, this could constitute ground for discussions regarding safety and hazard of transporting high radioactive material through the country. The construction phase, through electricity consumption, is the main cause for the high contribution to water depletion (44%), whilst the decommissioning phase is attributable to use of bentonite as backfill for sealing the GDF. The fact that the construction and decommissioning phase (Figure 5.12), and vitrified HAL stream (Figure 5.11) are the dominant factors in the environmental performance of the GDF are strictly related. The design of the GDF envisages that HLW canisters are disposed in single deposition holes 5 metres apart aligned in tunnels, whilst I-LLW are stacked in deposition vaults. This implies that amount of HLWs is a major factor in determining the footprint of the GDF, and the higher the footprint the more the construction and decommissioning phases will impact [319].

5.2.3.2 Sensitivity analysis

From the sensitivity analysis (Section 5.2.2.3), two important conclusions can be drawn. Firstly, the LCA model and results are relatively stable with respect to the analysed parameters, meaning that the system does not feature strong non-linearities with the potential of amplifying potential errors or uncertainties from model parameters to results. This is demonstrated by the fact that the highest multiplier, i.e. HAL to Ef, is equal to 0.68 meaning that a 1% change in the HAL flow from THORP would induce a change in the Eutrophication (freshwater) score of 0.68%. In addition to this, the analysis allowed identification of two influential parameters, these are the Highly Active Liquor (HAL) and Low Active Effluent (LAE) waste streams leaving THORP. Notably – and reassuringly – the Highly Active Liquor flow is deemed to be the most accurate parameter because it has been provided directly by the THORP technical team. Electricity consumption and radioactive emissions, on the other hand, results having little influence on the LCA results. It is important to point out here that although a parameter may be very influential (i.e. it has a very high multiplier), its uncertainty range may be so little that its potential impact on the final results is almost negligible. For this reason, it is usually recommended to perform the uncertainty analysis as well, so that the so-called essential parameters [332], i.e. very influential and uncertain parameters, are identified. These are the parameters that need to be verified and, if possible, their valued refined [332], [333]. In this study, however, no uncertainty analysis could be performed due to unavailability of data regarding uncertainty ranges of the model parameters.

5.2.3.3 Assumptions

Before concluding, let us discuss some choices and assumptions made in this study. While liquid discharges from THORP fuel Receipt and Storage (R&S) have been included, those from the Fuel Handling Plant (FHP) plant and AGR Storage Plant (AGRSP) have been not. The latter, which is not part of the system boundary altogether, acts as an intermediate buffer storage for UNF between FHP and THORP R&S. Both FHP and AGRSP liquid purges are routed to a further facility known as SIXEP (Site Ion Exchange Effluent Plant) to reduce activity (principally caesium and strontium) prior to discharge to sea. SIXEP has not been considered in this study; the claim, in fact, is that there should be no radioactive or non-radioactive pollutants discharged through liquid streams from storage ponds, as waste containers are assumed to remain intact during storage period. However, at the Sellafield site there is also historical (or legacy) waste stored underwater that date back to the first

decades of nuclear power in the UK. These wastes have corroded producing contaminated liquor and liquid effluents, which need to be treated in SIXEP. Another plant that has been omitted from this study is the Solvent Treatment Plant (STP). This treats Tributyl Phosphate (TBP) and Odourless Kerosene (OK) solvent arising from fuel reprocessing. Two effluents leave STP: a low activity effluent routed to SETP, and a high activity to EARP before discharge to sea. STP contribution to both SETP and EARP is very low, and thus it may be neglected without committing significant errors. As discussed in section 5.2.1.2, the AGR (stainless steel and graphite) components streams are in this study assumed to be treated in plants *to-be-defined* (tbd). Although it is intention of Sellafield Ltd. (SL) to proceed with their immobilisation in the near future, location of this activity has not yet been defined; it may either take place in a new facility or in one of the existing ones. These plants are assumed to be already existing (meaning that their construction has not been considered); however, should new facilities need to be built, their construction is unlikely to significantly affect the results as these streams have very little, if not negligible, contribution to the impacts. A further assumption concerns the consumption of electricity. Collected data refer to overall consumption per plant in financial year 2015-16 and have been scaled down in the LCA model to the chosen Functional Unit. Because the majority of this consumption is fixed and independent of the throughput – for instance ventilation of buildings – the assumption of a linear correlation between throughput and electricity consumption is incorrect; however, as demonstrated by the sensitivity analysis, the errors introduced are minor and acceptable. Furthermore, data for electricity consumption of WTC plant was not available, and both WTC and the fictitious *tbd* plants consumptions have been assumed to be equal to WEP.

A number of assumptions have also been made on the GDF subsystem. The operation phase does not take into account the electricity and fuel needed to power the facility for the time it will be in operation; this, however, is likely to be negligible. The GDF considered in this study is based on the conceptual design for high strength rock developed by RWM [319], which is one of the three designs considered by the RWM for preliminary radiological assessments. However, other countries may choose in the future to implement different designs in response to the nature and quality of the host rock available. Thus, extending results of this study to other countries must be done with caution. For instance, it is known that evaporite rocks have generally very low rate (or perhaps even the absence) of groundwater flow [334], which is the major pathway for

radionuclides escape. Therefore, repositories built in such environment are likely to have lower, or maybe negligible, radiological impacts. The U.S. Waste Isolation Pilot Plant (WIPP) is the only such operational repository, excavated in a Permian layer in New Mexico. On this topic, a life cycle analysis of alternative GDF designs and their potential impacts would indeed not only be very interesting, but also crucial to support nuclear waste management policies.

5.2.3.4 Contextualising LCA results

The LCA results presented in this Chapter are specific to the back-end of the nuclear fuel cycle; however, it would be interesting to assess how they compare with the whole nuclear fuel cycle. Unfortunately, as noted in Section 5.1, very few LCA studies have been carried out on the nuclear industry, and even fewer consider impact categories other than global warming [312]. This practice is probably due to the lack of knowledge regarding other kinds of emissions, but it is certainly linked to the fact that the primary objective of numerous studies is to compare energy sources only with respect to their potential to tackle global warming and climate change by curbing greenhouse gas emissions.

Considering only the climate change category, the majority of studies reports emissions lower than 30 g CO_2 eq. per kWh for the entire nuclear fuel cycle – although there is still a significant amount of publications indicating higher emissions of up to 100 g CO_2/kWh [312]. These variations are caused primarily by differing system boundaries; the majority of studies in fact do not include the back-end of the fuel cycle. Notably, figures for decommissioning also vary quite considerably, from as low as 0.01 to as high as 35 g CO_2/kWh [312] – even higher than total emissions from the entire nuclear fuel cycle calculated by the majority of studies.

If it is assumed an average burnup of 40 GWd/teU for the nuclear fuel in the reactor and a conversion efficiency (thermal to electric) of one third, it is possible to estimate the consumption of uranium per kWh generated being equal to 3.13 E-10 teU/kWh. The global warming impact score for the management of 1 teU of UNFs assessed in this study is equal to 2.4 kg CO_2/teU, which equals 0.075 g CO_2 per kWh that has been generated by the fuel being managed (according to the figure for uranium consumption per kWh estimated above). Therefore, the greenhouse gas intensity of the back-end of the fuel cycle as considered in this study is considerably lower (and almost insignificant) compared

to that of the entire fuel cycle. Greenhouse gas emissions of nuclear energy are mainly caused by energy intensive phases like mining and milling, conversion and enrichment.

5.3 Radiological impacts

This Section reports radiological impacts estimated by the Critical Group Methodology (CGM, developed within this Thesis and presented in Chapter 4) in Section 5.3.1 and by the site-specific methodology employed by Sellafield Ltd. in producing their annual report "Monitoring Our Environment" in Section 5.3.2. The objective is to perform a detailed comparison of these methodologies with UCrad, whose results have been presented in Section 5.2.2.

5.3.1 Critical Group Methodology

The Critical Group Methodology is applied to the system boundary described in Section 5.2.1.2 and is only concerned with emissions occurring in the Foreground, so that a consistent comparison with UCrad results can be performed.

5.3.1.1 Identification of the critical group

Because of the inherent features of its fate model, CGM does not allow selection of a critical group for radiological impacts arising from nuclear waste disposed in a GDF. The methodology adopts that same critical group as the RWM's generic Post-Closure Safety Assessment [267], which represents a small village of about 300 people with an extension of about 10 km^2, located above the GDF [269]. A critical group, however, needs to be identified for direct discharges, which for this study is represented by inhabitants of Seascale, a small village located 2.5 km south of the Sellafield site. Characterisation factors for both direct discharges for a distance of 2.5 km of the critical group, and for nuclear waste are reported in Section A.1 of Appendix A.

5.3.1.2 Results

Table 5.5 and Table 5.6 report CGM radiological impacts associated respectively with AGR reprocessing operations carried out at the Sellafield site and with nuclear waste disposed of in a GDF, expressed in terms of risks per year and percentage contribution from each source. Besides CGM, each table also reports impacts obtained from the UCrad methodology. Figure 5.13 illustrates radionuclides contributions to the plants and waste streams with the greatest impact.

Table 5.5 – Radiological impacts for direct discharges from the units as indicated and calculated according to CGM methodology at 2.5 km and UCrad

Unit	CGM (2.5 km)		UCrad	
	Risk/y	*Share*	Risk/y	*Share*
THORP	2.56E-08	*89.4%*	8.16E-12	*86%*
EARP, from SEC treatment	6.17E-10	*2.2%*	1.14E-14	*0%*
HALES	6.74E-10	*2.4%*	2.29E-13	*2%*
SETP, from HALES	2.78E-10	*1.0%*	2.95E-13	*3%*
SETP, from THORP	5.63E-10	*2.0%*	7.30E-13	*8%*
WEP – Cladding	1.81E-10	*0.6%*	6.23E-14	*1%*
WEP – BC slurry	2.53E-14	*0.0%*	8.69E-18	*0%*
WEP – Centrifuge cake	3.26E-11	*0.1%*	1.12E-14	*0%*
WEP – Maint. scrap	7.85E-12	*0.0%*	2.70E-15	*0%*
WEP – MEB crud	6.12E-12	*0.0%*	2.10E-15	*0%*
WVP	1.06E-15	*0.0%*	3.08E-19	*0%*
FHP	6.73E-10	*2.4%*	3.76E-15	*0%*
Total	**2.86E-08**		**9.51E-12**	

Nearly 90% of radiological impacts from direct discharges are due to THORP operations; the remaining 10% is equally distributed to EARP, HALES, SETP (THORP) and FHP at ~2%, and SETP (HALES) at 1%, whilst the solid waste treatment plants, namely WEP and WVP, appear to have negligible impacts (Table 5.5). Figure 5.13 shows that discharge of iodine-129 (I129) to atmosphere and cobalt-60 (Co60) to coastal waters are responsible for over 90% of THORP impacts, at ~69.1% and 24.5% respectively. The remainder is shared by krypton-85 (Kr85, ~3%) and caesium-137 (Cs137, ~1.5%), with aggregated contributions from remaining radionuclides equal to 1.8%.

Like direct discharges, impacts from nuclear waste are attributable primarily to one source: HLW contributes to over 90% of radiological impacts arising from GDF. Centrifuge cake (4%), MEB crud (4%) and Cladding (1%) make up the remaining 9%, with other waste streams having negligible impacts (Table 5.6). The primary cause of this impact is represented by I129 emissions to water (95.9%) and air (3.2%); the remaining radionuclides contributes to less than 1% (Figure 5.13).

Similar to the results from CGM, the dominant source of impacts identified by UCrad is THORP, at 86%; however, the remainder is distributed differently. Impacts of SETP allocated to THORP and HALES amount to respectively 8% and 3% in UCrad, compared to

2% and 1% in CGM, whilst impacts of EARP allocated to treatment of SEC is nugatory in UCrad, instead of 2.2% calculated according to CGM. Differences in terms of radionuclides contributions are more considerable (for UCrad, they are illustrated in Figure 5.6). Firstly, whilst coastal water discharges contribute to approximately a quarter of CGM impacts, over 99.5% of impacts calculated by UCrad are due to atmospheric emissions. Interestingly, according to both methodologies atmospheric discharges of I129 represent the primary cause of radiological impacts (78% in UCrad and 65% in CGM); though, the remainder is attributed to atmospheric emissions of Kr85 in UCrad. Co60 still represent the highest impact from coastal water emissions, but its contribution to THORP radiological impacts in UCrad is lower than 1%.

Table 5.6 – Radiological impacts for emissions from GDF calculated according to CGM methodology at 2.5 km and UCrad

Waste stream	CGM		UCrad	
	Risk/y	*Share*	Risk/y	*Share*
Graphite comp.	6.25E-19	*0%*	5.97E-24	*1%*
SS comp.	3.02E-18	*0%*	2.88E-23	*3%*
BC slurry	2.90E-19	*0%*	1.54E-24	*0%*
Centrifuge cake	6.59E-17	*4%*	1.83E-22	*17%*
Cladding	2.20E-17	*1%*	1.20E-22	*11%*
HLW	1.68E-15	*91%*	3.39E-22	*32%*
Maint. scrap	-	*0%*	-	*0%*
MEB crud	7.01E-17	*4%*	3.73E-22	*35%*
PCM	4.76E-19	*0%*	4.54E-24	*0%*
SEC floc	5.27E-20	*0%*	2.02E-25	*0%*
Total	1.85E-15		1.06E-21	

Even more pronounced differences can be observed for the solid waste streams. According to UCrad, HLW is cause of only one third of the total radiological impacts (compared to 91% in CGM), with another third due to MEB crud (4% in CGM). Significant impacts are also due to centrifuge cake (17%, compared to 4% in CGM) and cladding (11%, compared to 1% in CGM). Finally, whilst in CGM disposal of graphite and stainless steel components have negligible impacts (lower than 1%), in UCrad they contribute to 1% and 3% respectively. The considerable discrepancy in waste streams' contributions is also reflected in radionuclides' contributions: UCrad impacts are dominated by I129 and Cl36, compared to 2.5% and 0.6% in CGM; and, vice versa, Cs135 and Se79, which dominate CGM impacts, make negligible contributions to UCrad impacts.

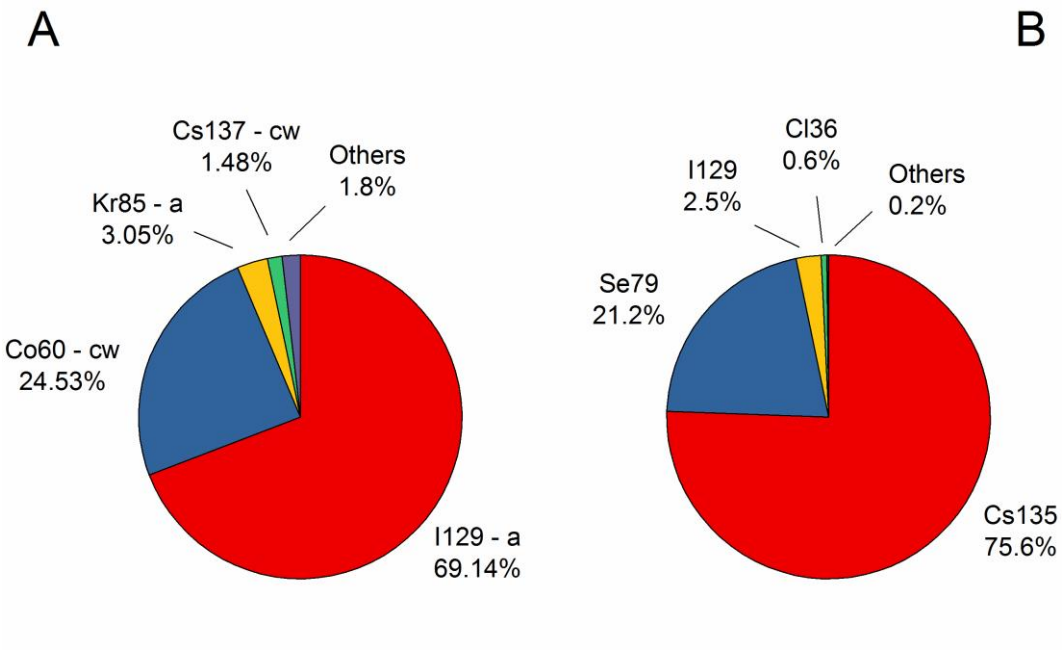

Figure 5.13 – Radionuclides contribution to direct discharges from THORP (A) and GDF emissions from HLW (B) calculated according to CGM (2.5 km). a: atmospheric; cw: coastal water.

5.3.1.3 A practical application of the Critical Group Methodology

In Chapter 4, two alternative approaches for applications of the methodologies developed within this Thesis (UCrad and CGM) have been conceived. One approach envisages that either of the methodologies is used, whilst the other approach envisages the combined use of CGM and UCrad. While the former is adopted in the LCA study presented in Section 5.2 where only UCrad has been included, the latter is discussed in this Section. Thus, the focus is on practical applications of CGM that is used to:

1. Calculate the maximum reprocessing throughput which would comply with the maximum dose to the critical group;
2. Compare the dose received by the critical group with the average one calculated with UCrad.

It is worth recalling that until now the radiological impacts discussed refers to the Functional Unit represented by the reprocessing of the amount of UNFs containing 1 tonne of uranium pre-irradiation. From these impacts it is possible to back-calculate the theoretical amount of AGR fuels that is required to be reprocessed for the critical group to receive the maximum allowed dose in the UK. This has been established by the Radioactive Substances Direction 2000 (based on the 1990 recommendations of the ICRP [185]) and is equal to 1 mSv [51]. The maximum AGR fuel throughput is approximately

equal to 2000 teU – a figure well above the normal throughput of THORP, and also the current combined throughputs of THORP and Magnox.

Finally, the annual risk due to radioactive discharges incurred by the critical group according to CGM is compared with the radiological impacts for both the Background and Foreground system calculated according to UCrad to assess how large the critical group dose is compared with the average person around the world (that is how the results from CGM compare with UCrad). Table 5.7 shows that the critical group is receiving a dose that is a thousand times higher than the average person around the world – but still well below the maximum allowed dose.

Table 5.7 – Radiological impacts of the Foreground system calculated by CGM, for both Foreground and Background systems calculated by UCrad, and the excess risk incurred by the critical group with respect to average person around the world.

	CGM (2.5 km)	UCrad	Critical group excess risk
Risk/year	2.86E-08	1.90E-11	1.51E+03

5.3.2 The Sellafield Ltd. site-specific methodology

"Monitoring our Environment" is an annual report published by Sellafield Ltd. (SL) – the company responsible for managing the the Sellafield site on behalf of the NDA including delivering reprocessing of fuel, waste management operations and the ultimate decommissioning of the site – that provides detailed information on radioactive and non-radioactive discharges and disposals, monitoring of the environment and radiological impacts. For radioactive impacts, the report presents annual discharges and disposal data over five years for all radionuclides specified in the environmental permit for Sellafield and Windscale sites. Non-radioactive discharges and disposals data refer only to the year of the report because it is argued that it would be impracticable to report discharges of all chemical species and performance against every condition in all permits and consents, even more so for a five-year period.

For assessment of radiological impacts, SL adopts a site-specific methodology based on the CREAM model (described in detail in Chapter 3) which, like CGM, focuses on doses received by members of the critical group. However, collective doses to larger groups of people, such as the UK, are also provided. The methodology uses a combination of analytical measurements (predominantly), and literature data and predictive models to quantify environmental concentrations of radionuclides.

5.3.2.1 Critical group

In determining the critical group, SL recognises that the relative doses from different pathways will depend on the habits of particular groups of individuals: a consumer of large quantities of seafood may receive only a minor exposure via pathways such as milk consumption or proximity to the site perimeter, whilst for another group, consumption of locally produced meat and milk may combine to result in an elevated exposure. Therefore, SL identifies two critical groups according to their dominant pathway, namely marine and terrestrial, described by data obtained by the Food Standards Agency, Environment Agency and the Centre for Environment, Fisheries and Aquaculture Science.

The marine critical group embodies a small number of people in the Cumbrian coastal community who are high-rate consumers of fish and shellfish, obtained from the Sellafield area between St. Bees and Selker. The terrestrial critical group identifies high consumers of terrestrial produce, primarily milk and root vegetables, living in the area around the Sellafield site.

5.3.2.2 Doses to the critical group

Table 5.8 reports results of the dose assessment to the critical group, whilst Figure 5.14 shows doses contributions by radionuclide for consumption of seafood and foodstuffs produce by the marine and terrestrial groups respectively. Breakdown for the other pathways was not provided in the report.

Table 5.8- Summary of critical group doses from operations at Sellafield (µSv)

Pathway	Dose (µSv)
Marine critical group (adults)	
Seafood consumption	58
Aerial pathways	2
External radiation from beach occupancy	44
Total dose to the marine critical group (adults)	100
Terrestrial critical group (adults)	
Terrestrial foodstuff consumption	6.7
inhalation	0.8
immersion	0.5
External radiation from beach occupancy (terrestrial)	2.6
marine foodstuff consumption	0.9
direct radiation	5.4
Total dose to the terrestrial critical group (adults)	16.9

The marine critical group receives a total dose of 100 µSv: 58 µSv from consumption of seafood and 44 µSv from external radiation due to beach occupancy, with only minor contribution (2 µSv) from aerial pathways (which includes both inhalation and consumption of agricultural produce). As shown in Figure 5.14 A, a significant part of the overall dose received from consumption of seafood is caused by americium-241 (Am241) and plutonium alpha (Pu alpha) at approximately 81%, while carbon-14 (C14), Cs137, Np237, technetium-99 (Tc99) and I129 together make up ~16%. The sum of the doses of the remaining radionuclides corresponds to around 3%.

The terrestrial critical group receives a significantly lower dose (~17 µSv) than the marine critical group, with consumption of foodstuffs (6.7 µSv) and direct radiation (5.4 µSv) contributing to over 70% of the total dose. The SL report only includes contributions from aerial pathways that include inhalation and foodstuff consumption (Figure 5.14 B); the highest dose is delivered by Sr90, at ~22%, followed by caesium-137 (~18%), Am241 (~17%) and I129 (~15%). Remaining radionuclides have individual contributions no higher than 10%. The report argues that doses from strontium-90 and caesium-137 are dominated by pre-1980 discharges, the testing of nuclear weapons in the 1960s and, for Cs137, the Chernobyl accident in 1986. In previous years the overall dose was dominated by milk consumption; however, in 2014, root vegetables and drinking water represented the dominant pathways.

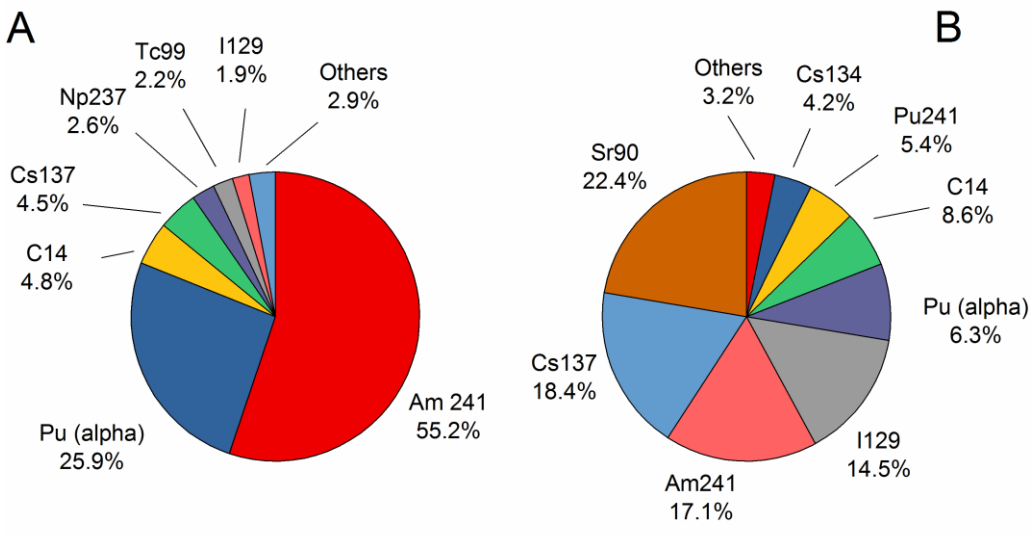

Figure 5.14 – Radionuclide breakdown for marine critical group from seafood consumption (A) and terrestrial critical group from inhalation and foodstuff consumption (B)

5.3.2.3 Comparison with UCrad and CGM

Figure 5.15 illustrates comparison between the methodologies developed within this Thesis, namely UCrad and CGM, with the site-specific methodology adopted by Sellafield Ltd, expressed in terms of radiological doses. Because doses reported in the SL report Monitoring Our Environment relate to emissions occurring throughout a year, a Functional Unit of 430 teU corresponding to a representative annual throughput of THORP is used. Thus, the bar chart includes doses calculated according to CGM and UCrad for a functional unit of 430 teU; doses received by the marine and terrestrial critical group as reported by the SL report (Table 5.8); and finally, dose calculated with CGM and UCrad methodologies according to emission data found in the SL report (Table B.15 and Table B.16 in Appendix B).

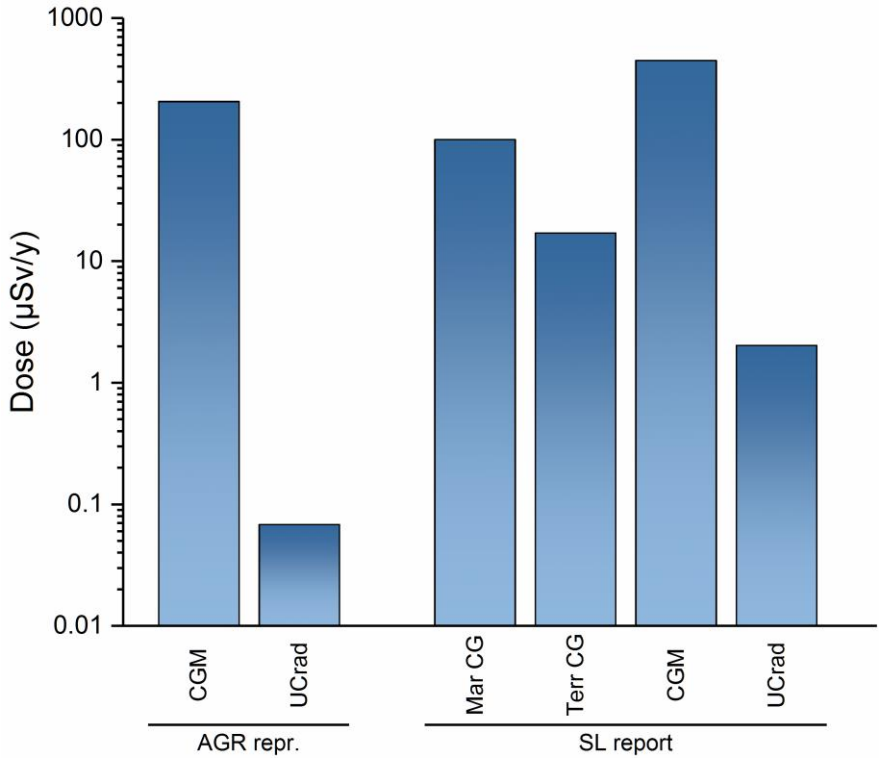

Figure 5.15 – Comparison of dose according to AGR reprocessing scenario and SL report, and CGM, UCrad and SL methodologies

The chart shows that doses estimated by CGM for the critical group identified Section 5.3.1.1 and for a Functional Unit equal to 430 teU of AGR reprocessed approximately 12 and 20 times higher than that received by the terrestrial and marine critical groups, respectively, in the SL report. Whilst, for the same Functional Unit the dose computed by UCrad is approximately three and four orders of magnitude lower. It must be noted that the reprocessing scenario includes only discharges related (i.e. allocated) to reprocessing

operations, whilst doses reported in the SL report includes all radioactive discharges from Sellafield site. When the SL report data are used, CGM impacts roughly double whilst UCrad scores increase by two orders of magnitude; this leads to an increase in the disparity between CGM and the site-specific methodology results (with CGM impacts being 24 and 40 times higher), and a reduction of the discrepancy associated with UCrad (with impacts being approximately one and two orders of magnitude lower).

Figure 5.16 reports radionuclides contributing to CGM and UCrad doses calculated according to the SL dataset. CGM dose is dominated by atmospheric discharges of I129 (~42%), followed by discharges to coastal waters of Co60, Ce144, Ru106, Cm243-244 and Cs137, which are jointly responsible for over half of the total dose. The situation is quite different for UCrad, where the dose is primarily caused by coastal water emissions of I129 (over 95%). Therefore, not only the absolute values of the doses, but also radionuclides' contribution show significant discrepancies.

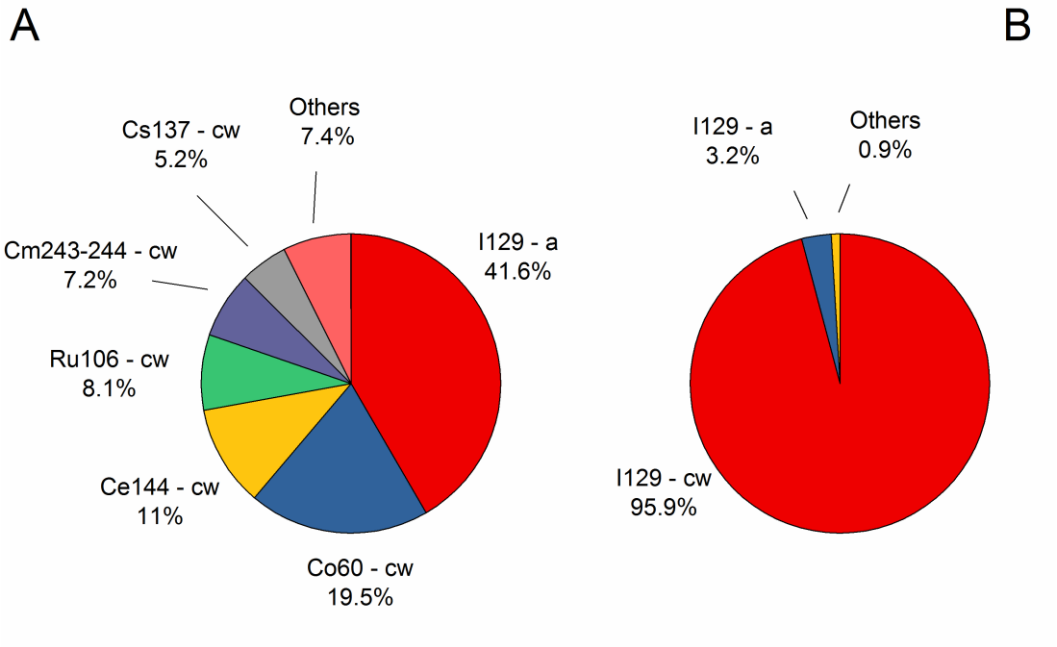

Figure 5.16 – CGM (A) and UCrad (B) applied to SL report discharges data

5.3.3 Discussion

The comparison between the Critical Group Methodology (CGM) and UCrad shows that for the same case study the methodologies produce results that are considerably different both in terms of absolute values and contributions by radionuclides. The cause of the differences in absolute values of impacts, which equal several orders of magnitudes, is obvious: it is related to the fact that CGM focuses on impacts on a group of people living

very close to the source of emissions, instead of average impacts quantified by UCrad. On the other hand, differences in the characterisation factors computed by the methodologies lead to differing contributions of radionuclides. Atmospheric emissions of iodine-129 represent a notable example; for the SL report dataset, they contribute to over 95% of UCrad impacts compared to approximately 41% in CGM (Figure 5.16). As noted in Chapter 4, the long half-life of I129 leads to it being more relevant in UCrad compared to CGM.

Although UCrad represents a better candidate for incorporation into LCA, the CGM methodology can still have some useful applications within the LCA methodology. Chapter 4 discussed how UCrad and CGM can be combined to give interesting insights, whilst this Chapter illustrated two practical applications. First, CGM has been used to calculate the maximum throughput of THORP (in terms of AGR fuel reprocess) that would not exceed the maximum radiological dose allowed in the UK. Results of such application should be used as a first approximation; they, by no means, aim to replace more accurate methodology like site-specific risk assessment studies. A further interesting application presented relates to comparing the dose received by the critical group with respect to that received by an average person around the world, to calculate the excess risk incurred by the group of people considered to be the most exposed to the specific release.

The second part of the analysis illustrates how CGM and UCrad compare with results of a site-specific methodology such as that employed by Sellafield Ltd (SL). Firstly, it is interesting to note that radiological impacts calculated by CGM are at least an order of magnitude higher than those estimated by SL methodology, even though they are only referred to those radioactive emissions attributed to AGR reprocessing. The opposite is observed for UCrad, which estimates impact to be considerable lower. Therefore, while is to be expected that when CGM and UCrad are applied to the emission data reported in the SL report, their estimates increase considerably, it is interesting to note that results of the site-specific methodology fall between those of CGM and UCrad for both critical groups considered. Effectively, UCrad and CGM yield the upper and lower estimates for radiological impacts within which more accurate estimates provided by site-specific results should expected to be found.

A final point worth discussing is how sources of radiological impacts change between different assessment approaches. The SL site-methodology reports that the most

significant sources of radiological impacts are generally represented by five radionuclides: Am241, Pu (alpha), Sr90, C14 and I129. Apart from I129, none of these radionuclides have major contributions to UCrad or CGM impacts. The dose received by the marine critical group is dominated by Am241 and Pu through consumption of mussels and winkles, both of which are not accounted for by CGM. Because of its generic features, CGM only includes a generic fish for the marine ingestion pathway. Notably, both Am241 and Pu have considerably higher tendencies for bioaccumulating in mussels and winkles, rather than in fishes. A similar example is provided by the terrestrial critical group dose. The four radionuclides that dominate doses from the terrestrial pathway as estimated by SL are Am241, I129, Sr90 and Cs137. Their atmospheric concentrations measured by Sellafield Ltd. on the perimeter of Sellafield site are significantly different from those estimated by CGM for a similar distance (see Table 5.9): for instance, iodine-129 is determined by SL in having a negligible concentration with respect to ~0.15 mBq/m^3 estimated by CGM.

Table 5.9 – Atmospheric discharges and concentrations as estimated by CGM and measured by SL

	Air concentration (mBq/m^3)	
	CGM	SL report
Am241	1.24E-04	2.00E-03
Cs137	1.87E-03	<4.00E-3
I129	1.49E-01	-
Sr90	3.73E-04	<5.00E-4

A similar kind of discrepancy can be observed for concentrations in above and below ground produce as shown in Table 5.10, where CGM overestimates I129 concentrations by around two order of magnitudes, whilst at the same time underestimating the others.

Table 5.10 – Radioactivity in above ground produce and milk as estimated by CGM and reported by SL

	CGM		**SL**	
	Crops	Milk	Highest veg produce	Milk
	Bq/kg	*Bq/l*	*Bq/kg*	*Bq/l*
Am241	7.09E-03	3.90E-08	1.30E-01	-
Cs137	1.06E-01	6.43E-03	2.10E-01	1.10E-01
I129	8.58E+00	6.13E-01	<9.00E-02	<7.00E-3
Sr90	2.18E-02	3.84E-04	2.10E-01	1.20E-01

It is clear that site-specific methodologies based on analytical data are more accurate than any generic methodology; however, increasing accuracy adds layers of complexity to the approach. For this reason, structured approaches are conceived so that more accurate (and thus complex) approaches are used as predicted doses approach or exceed a reference level. Impact assessment methodologies for LCA, however, are designed to be simple and to require as little data as possible on the source of emissions. In this sense, primarily UCrad, but also CGM, is indeed more consistent with the LCA philosophy than any site-specific methodology.

5.4 Conclusion

This Chapter presented a comprehensive assessment of the impacts associated with management of Used Nuclear Fuels (UNFs), but also demonstrated with a practical application the use of the methodologies for radiological impact assessment developed within this Thesis.

First, a Life Cycle Assessment (LCA) study has been performed on a product system corresponding to the current approach for managing UNFs that envisages their reprocessing, and the agreed policy for disposal of higher activity nuclear wastes in a Geological Disposal Facility (GDF). The valuable products obtained by reprocessing, i.e. plutonium and uranium oxides, are not included in the analysis, because their fate is still to be established by the UK Government. The Foreground system inventory is based on a combination of operational data gathered at Sellafield site and preliminary design of a GDF developed for the UK [319]; whilst average market data is used for the Background system. The impact assessment phase is based on the ILCD recommendations and supplemented with the UCrad categories for direct discharges and nuclear waste. The analysis shows that radiological and toxicity impact categories have the highest normalised impact; though the former may be due to specific features of radioactive emissions and biases introduced by the normalisation step. The majority of impacts can be attributed to the use of uranyl nitrate for separating uranium (RepU) from plutonium (Pu) in the Thermal Oxide Reprocessing Plant (THORP), and copper in the disposal canister for High Level Waste (HLW). Impacts associated with the GDF are generally minor (especially for categories with high normalised score) and mainly attributable to construction (electricity consumption and building of facilities) and decommissioning

phase (use of bentonite to seal the repository). The sensitivity analysis carried out on a subset of model parameters shows that the results are relatively stable and that the amount of Highly Active Liquor produced by THORP is the most influential parameter.

The second part of the Chapter focused specifically on radiological impacts, with the objective of comparing UCrad with CGM, but also with the site-specific methodology employed by Sellafield Ltd. Differences between UCrad and CGM are significant; they are due to inherent features of the methodologies, such as adoption of the critical group concept, and have been explained in detail in Chapter 4. UCrad and CGM impacts are also considerably different from those estimated by the Sellafield Ltd. site-specific methodology, but provide upper and lower estimates within which results of more accurate models are expected to be found. Finally, a practical application of a combined use of CGM and UCrad have been demonstrated to calculate the maximum THORP throughput related to the highest allowed dose allowed in the UK, and to estimate the excess risks to which the critical group is exposed compared to an average person around the world.

Chapter 6.
Reprocessing vs Direct Disposal: assessing the impacts of future scenarios for the back-end of the UK nuclear fuel cycle

The UK and its nuclear industry are entering a critical phase when crucial decisions are expected to be taken. The Nuclear Decommissioning Authority (NDA) has announced that reprocessing operations of Used Nuclear Fuels (UNFs) will cease by 2020 with the closure of both the Thermal Oxide Reprocessing Plant (THORP) and the Magnox reprocessing plant. However, the future of the UK nuclear fuel cycle is yet to be decided, with options ranging from a Once-Through Cycle with direct disposal of UNFs to a Twice-Through Cycle with reprocessing of UNFs and a fully integrated strategy for management of reprocessed uranium and plutonium. This Chapter presents a comprehensive Life Cycle Assessment (LCA) study of future scenarios for the back-end of the UK nuclear fuel cycle that aims at informing policy and decision-makers. The study considers the direct disposal approach and four reprocessing scenarios envisaging different strategies for disposal and/or reuse of reprocessed uranium and plutonium, and adopts a consequential approach including only short-term effects. These primarily represent reductions in demand for uranium mining due to recycling of uranium and plutonium, and are modelled upon identification of a marginal technology. Results of the study show that recycling of uranium, but especially plutonium is of paramount importance because of the avoided burdens associated with production of nuclear fuel from mined uranium. The reprocessing scenario envisaging reprocessing of UNFs with recycling of both plutonium and uranium primarily in MOX fuel is overall found to be the most favourable option, featuring "negative" impacts in all non-radiological impact categories. Depending on the marginal technology chosen, the direct disposal approach may be advantageous in terms of radiological impacts.

6.1 Introduction

The 2008 Climate Change Act established a legally binding target to reduce UK's greenhouse gases emissions by at least 80% below base year levels (1990-1995) by 2050 [33], [34]. This is in line with the EU low-carbon economy roadmap [27], other extra-EU countries' policies, e.g. Japan, Sweden [335], and falls within a global effort for restraining the increase in global average temperature to well below 2 °C above pre-industrial levels, ratified by the Paris agreement (COP21) in December 2015 [14]. Amongst the various sectors targeted by the Climate Change Act, the power generation industry represents a critical one. At present, the UK is home-producing around 95% of the total electricity supply. Natural gas represents the largest source of fuel at ~42%, followed by renewables at ~24% and nuclear at ~21%. Usage of coal, the largest source of fuel for electricity generation since the industrial revolution, has fallen steeply in 2016, and currently accounts for 9% [42]. The UK government in its Carbon Plan [34] outlined a number of future scenarios for achieving reduction targets, and reiterated its support for nuclear energy as a clean, secure and reliable source of energy. Nuclear could contribute up to 40-50% to the energy mix under the best possible scenario for the industry [34], [40].

As noted in Chapter 5, at present the UK operates a "nominal" Twice-Through Cycle or closed cycle whereby Used Nuclear Fuels (UNFs) are reprocessed, plutonium (Pu) and uranium (RepU[15]) are separated from fission products that are processed by utilising a Vitrification process into a final, manageable form suitable for disposal. The Twice-Through Cycle is just "nominal" because, as opposed to France for instance, RepU and Pu are not currently reused in the cycle, rather they are stored in an oxide form at Sellafield site pending a future decision by the Government on their fate.

The UK and its nuclear industry are entering a critical phase where crucial decisions are expected to be taken. The NDA has announced that reprocessing of UNFs will cease. Both THORP (Thermal Oxide Reprocessing Plant) and the Magnox reprocessing plant located at Sellafield are due to be closed in the two-year period from 2018-2020 [336]; the former when all current contracts are completed, the latter when all Magnox fuel has been reprocessed. Remaining and future arisings of UNFs, which come exclusively from the

[15] As in Chapter 5, RepU only refers to reprocessed uranium.

existing Advanced-Gas Cooled Reactors (AGRs), are planned to be temporarily wet-stored at the Sellafield site. The UK Government and the NDA have yet to take a decision on the future of the UK nuclear fuel cycle, and thus, at present, several options are possible. These range from implementation of a direct disposal (or Once-Through Cycle) to a Twice-Through Cycle approach with a full integrated management of nuclear waste and pre-determined fate of RepU and Pu. The latter could be implemented with construction of "THORP 2" based on a different chemical separation process from conventional PUREX – e.g. UREX, COEX [337] – for proliferation resistance purposes; or through a life-extension of the existing THORP plant. It, in fact, represents a considerable (~US$4 billion), and especially recent (1990s) investment [338]; its sister plant, the Magnox reprocessing plant, has in fact been operating since the 1960s (but is life-expired).

It must be noted that although the future of the UK nuclear fuel cycle is still uncertain, its approach to disposal of final, solid nuclear waste has already been established, and envisages disposal of higher activity wastes in a national Geological Disposal Facility (GDF) expected to be operational by the end of this century (see Chapter 1 and Chapter 5).

This Chapter presents a comprehensive Life Cycle Assessment (LCA) study aimed primarily at informing policy and decision-makers concerned with the future of the national nuclear industry, and in particular with the approach to management of Used Nuclear Fuels (UNFs). The LCA study considers both the direct disposal approach and four different scenarios for a Twice-Through Cycle approach that envisage continuing operations at the existing THORP plant.

The Chapter is organised as follows: Section 6.2 reports the study's goal and scope, the system boundaries, life cycle inventory and impact categories analysed; Section 6.3 presents the results of the LCA study in terms of hot-spot and comparative analyses for reprocessing and direct disposal approaches, leading to a discussion Section (6.4). Finally, the main findings of the study are summarised in a concluding Section (6.5).

6.2 Methods

6.2.1 Goal and scope

The goal of this study is to quantify, evaluate and compare the environmental impacts associated with two alternative approaches for managing UNFs in the UK: one envisaging

their direct disposal, and the other their reprocessing. The focus is on future environmental impacts that are a consequence of strategic choices made today; hence, the consequential approach is adopted, but linked with neither retrospective nor prospective perspectives, because decisions on which this study focuses are assumed to be taken in the present. However, because changes in the nuclear industry typically occur over large time scales – for instance decommissioning of power plants may take over a century – and thus they may difficult to predict, only short-term effects are included. These include first order direct physical effects, but not second and third order effects (known as negative and positive feedback effects, see Chapter 2), which are also typically included in consequential studies. A consequential study with such features is in effect identical to a prospective attributional using the "crediting" approach, in which the analyst assumes to be located in a future time when decisions have not only been already taken, but also implemented.

The functional unit corresponds to management of AGR UNFs containing 1 tonne of uranium pre-irradiation, which refer to the quantity of uranium before being irradiated in nuclear reactors, resulting in part of the uranium being converted either by fission or transmutation into other elements.

6.2.2 System boundary

This LCA study (like the one presented in Chapter 5) follows the methodological approach developed for integrated solid waste management by Clift and colleagues [108] that envisages distinction between Foreground and Background systems. The distinction, however, affects exclusively the inventory data collection, and it is not carried forward to the impact assessment phase, where the attention is on streams rather than systems. Figure 6.1 reports the system boundaries for the four reprocessing scenarios and direct disposal approach.

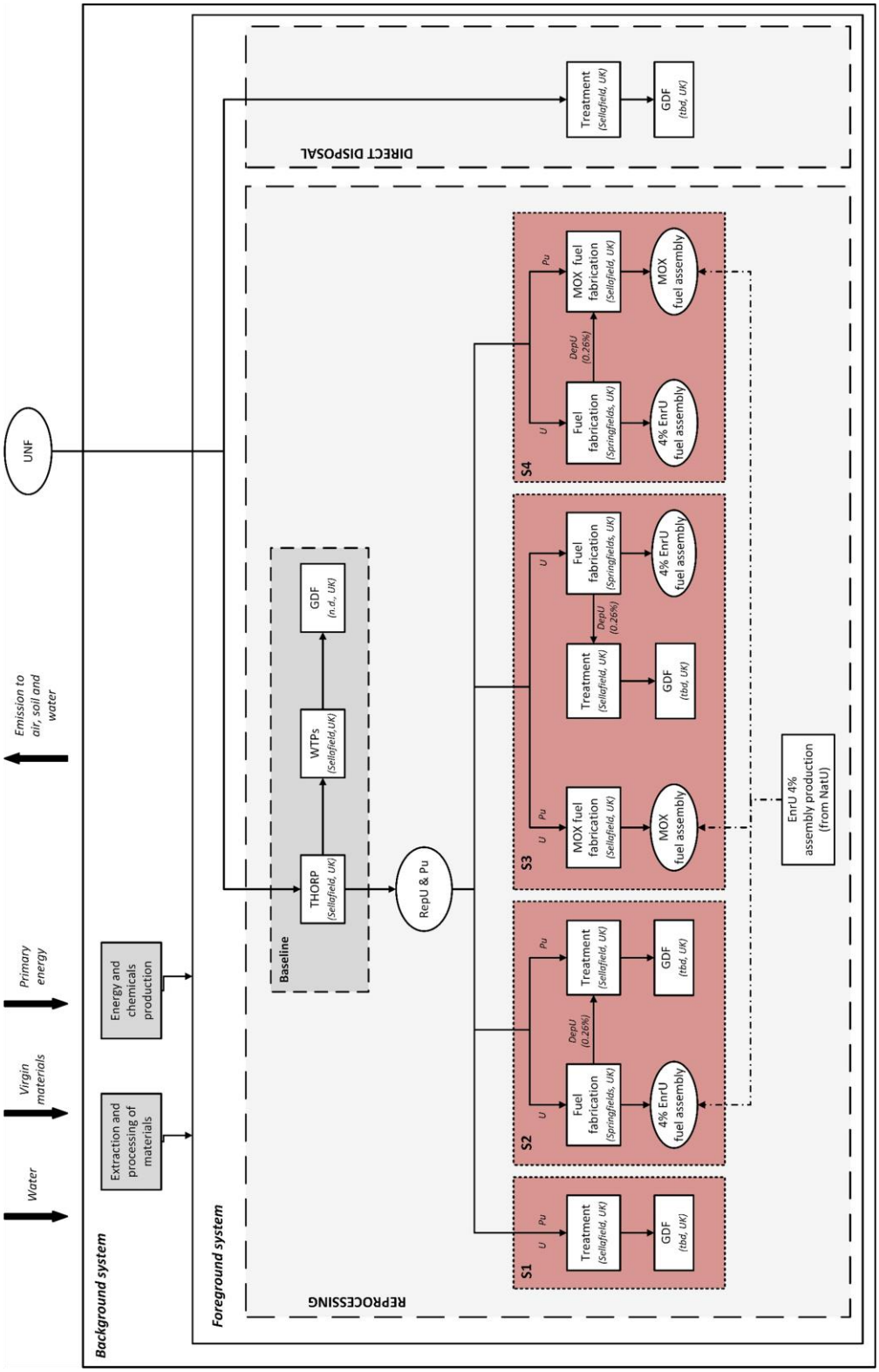

Figure 6.1 - System boundary for direct disposal and four different reprocessing scenarios. For clarity, transportation has not been included.

6.2.2.1 Reprocessing

The Foreground system for the reprocessing approach includes the current UNFs management practice in the UK (that is the "nominal" Twice-Through Cycle mentioned in Section 6.1 and investigated in Chapter 5), hereafter termed "baseline", and four scenarios for disposal and/or reuse of RepU and Pu. Notably, the baseline includes all the processes considered in the LCA study presented in Chapter 5, that is UNFs reprocessing, waste treatment and disposal in a GDF. Therefore, all the assumption made in Chapter 5 also apply here; for instance, that commissioning and decommissioning phases have only been considered for the GDF. The four reprocessing scenarios are introduced below:

Scenario 1

Both RepU and Pu are declared as waste and specifically treated to be prepared for disposal. Pu is assumed to be encapsulated according to the can-in-canister approach [339], developed in the USA for disposal of Pu alongside vitrified High Level Waste (HLW). The approach, illustrated in Figure 6.2, envisages plutonium oxide to be immobilised in a titanium-based matrix (Table C.1 in Appendix C), to form a puck 6.9 cm wide and 2.5 cm thick. One quarter of the matrix is made by uranium oxide to promote formation of cubic crystal structure; therefore, part of the RepU inventory is assumed to be used for this purpose and disposed of alongside Pu. Pucks are loaded into stainless steel cans, which themselves are encapsulated in borosilicate glass into a large steel canister – from which the name "can-in-canister". Each can has the capacity to contain 20 pucks, and 28 cans are loaded into each canister. Finally, it is assumed that each steel canister is packaged in a single disposal canister based on the Swedish KBS-3V concept also used for HLW (see Chapter 5). It must be noted that the disposal concept for Pu is still at a very early stage, even earlier than that for HLW. RepU, on the other hand, is assumed to be encapsulated in grout and packaged in 500 litre stainless steel drums; this is in line with the packaging approach used for other Intermediate Level Wastes (ILWs) [273]. RepU has a lower concentration of uranium 235 (U235) than Enriched Uranium (EnrU), but higher than Natural Uranium (NatU) and is usually classified as Low Enriched Uranium (LEU). Finally, both Pu and RepU packaged wastes are disposed in a GDF based on the NDA's generic design for higher strength rock discussed in Chapter 5. Pu is assumed to be disposed of in disposition tunnels alongside HLW and/or UNFs, whilst LEU is disposed in vaults with other ILWs.

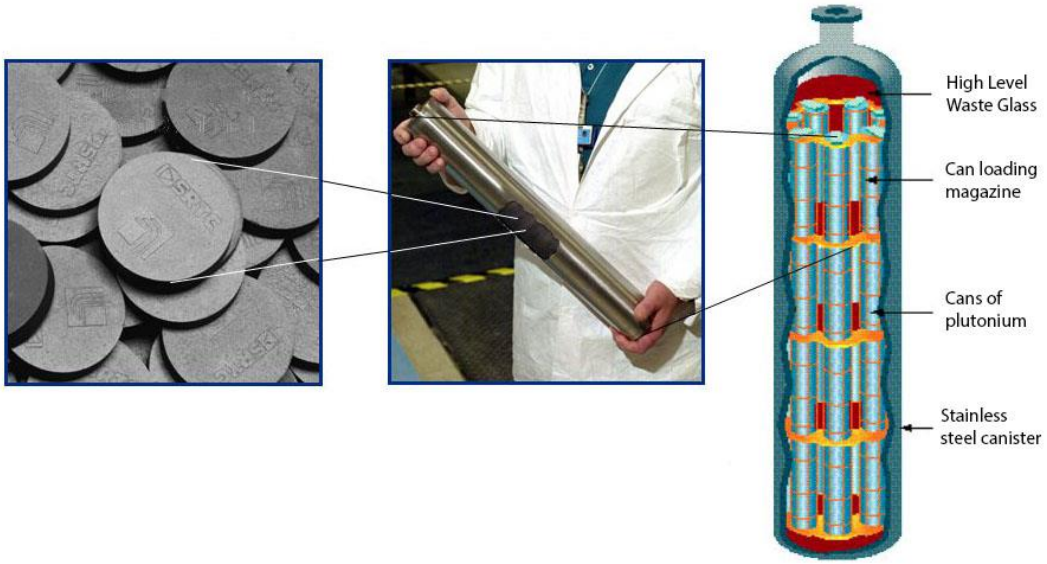

Figure 6.2 – Can-In-Canister disposal concept for plutonium, adapted from [339]**.**

Scenario 2

RepU is recognised as being a valuable product and recycled, whilst Pu is considered as waste. Treatment and disposal of Pu follow the same approach outlined in Scenario 1. With respect to RepU, this is re-enriched to be equivalent to 4% EnrU from NatU and used for fabrication of new fuel assemblies to be loaded into nuclear reactors for electricity generation purposes. Figure 6.3 reports the production process of fuel assemblies from RepU. Notably, RepU has to be enriched slightly in excess (4.1%) of the target assay to account for poison radionuclides, i.e. uranium 234 (U234), but mainly uranium 236 (U236) [340]. As the enrichment process requires uranium to be in a gaseous state as uranium hexafluoride at a relatively low temperature, RepU is first transported to the fuel manufacturing and conversion plant in Springfields, where it is converted into uranium hexafluoride (UF_6), and then to the Urenco centrifuge enrichment plant in Capenhurst, where it is enriched to 4.1%. Finally, the Enriched Uranium (EnrU) is transported back to Springfields, where it is reconverted into an oxide form, manufactured into pellets and loaded into fuel assemblies. Both fuel manufacturing/conversion and enrichment plants are currently operational; however, they have been designed to manufacture oxide fuel from NatU rather than RepU. Handling of RepU, in fact, requires alterations to the existing design, such as thicker barriers to protect against additional radiations due to U234, a strong alpha emitter with a moderate half-life [340]. Notably, this has not been taken into account because of lack of data in the literature; but it is deemed to have nugatory effects

on the overall environmental performance. The enrichment process generates two separate streams: the enriched product and the so-called "enrichment tails" (also referred to as Depleted Uranium, DepU), containing a very low concentration of U235, usually around 0.2–0 3%. The enrichment tails are considered waste and disposed of following the same approach as RepU (Scenario 1); the working assumption (as this is not currently carried out) is that they are encapsulated in grout, packaged in 500 litre drums and disposed of in a GDF.

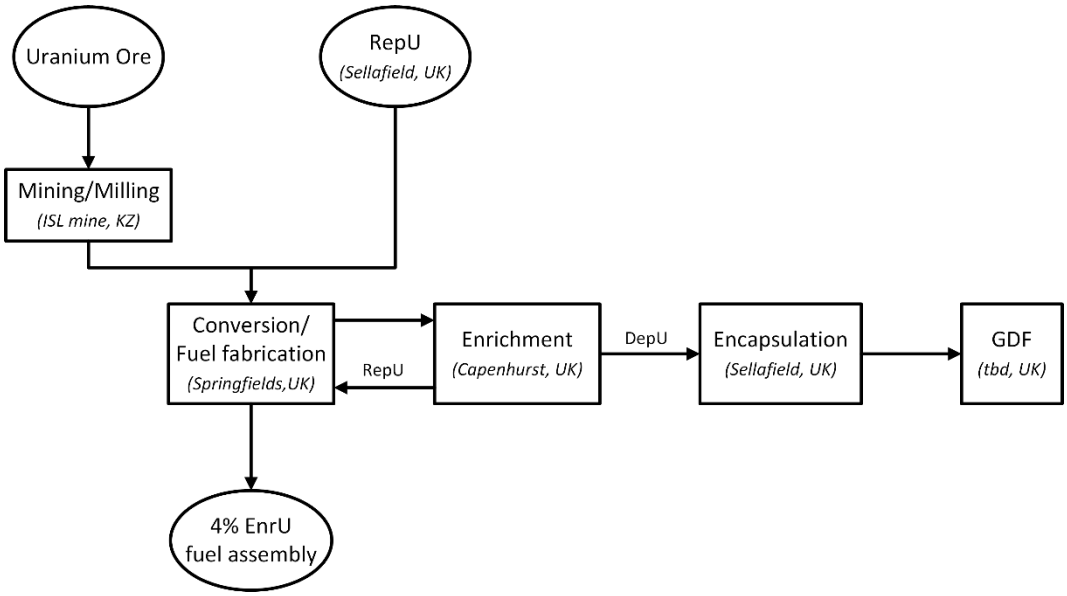

Figure 6.3 – Schematic outline for production of 4% Enriched Uranium (EnrU) from Reprocessed Uranium (RepU); and from uranium ore (i.e. Natural Uranium, NatU) assuming the marginal technology corresponds to a generic ISL mine located in Kazakhstan. *tbd*: to-be-defined

Scenario 3

Both uranium and plutonium in their oxide states are mixed to produce a new fuel termed Mixed Oxide fuel (MOX). The mixing proportion of uranium and plutonium depends on the fuel target assay, and is determined to have reactivity worth equivalent to enriched uranium, i.e. the potential of the fuel to produce the same amount of energy from fission as from a specific level of enriched uranium [341]. Other parameters that affect the mixing proportion are the concentration of fissile plutonium (239 and 249) and uranium (235) in both oxides, and the concentration of the isotope 236 of uranium in the uranium oxide. U236 is produced in nuclear reactors from capture of a neutron from U235 and emission of gamma radiation; it is a poison for the fuel since it is neither fissile nor fertile, but just a neutron absorber. This Scenario envisages production of MOX equivalent to 4% EnrU; details regarding calculation of the MOX mixing ratio are reported in Appendix C.2.

Concentration of U235 and U236 in RepU and the mixing ratio used for MOX are reported in Table 6.1 and Table 6.2. The uranium in excess of what required to produce the desired assay for MOX is re-enriched and used to produce uranium fuel assemblies according to the procedure outlined in Scenario 2 and reported in Figure 6.3.

Table 6.1 – Concentration (wt%) of uranium-235 and -236 in reprocessed and depleted uranium.

	RepU*		**DepU**	
	Concentration	Source	Concentration	Source
U235	1.80%	Figure 4 in IAEA tecdoc 1529**	0.26%	Ecoinvent database***
U236	0.39%	Figure 5 in IAEA tecdoc 1529**	0.00%****	

Notes:
*Data refers to 25GWd/t and 4% enrichment level
**[340]
***[253]
****U236 is found in traces in nature, and only in used nuclear fuels and reprocessed uranium its concentration is appreciable.

Table 6.2 – Proportion by weight of U and Pu in MOX fuel equivalent to 4% Enriched Uranium

	PuO_2	*(Pu)*	UO_3	*(U)*
RepU	5.1%	*(5.4%)*	94.9%	*(94.6%)*
DepU	8.1%	*(8.6%)*	91.9%	*(91.4%)*

Scenario 4

As in Scenario 3, both RepU and Pu are recycled. However, in this Scenario plutonium is mixed with depleted uranium (DepU) produced by the enrichment process (see Scenario 2), rather than with RepU. Table 6.2 shows that DepU-based MOX requires a higher proportion of plutonium to achieve the same target assay as in scenario 3 (i.e. equivalent to 4% EnrU); this is due to lower content of U235 than NatU (and thus also than RepU), although slightly counter-balanced by the fact that DepU contains only traces of U236, a stronger neutron absorber. The entire inventory of RepU is re-enriched and used to produce uranium fuel assemblies as done in Scenarios 2 and 3.

Avoided Burdens

The consequential perspective with inclusion of only short-term effects represented by first order direct effects is implemented by accounting for the additional or avoided burdens associated with changes in demand or production. Unlike Scenario 1, Scenarios 2,3 and 4 represent multifunctional product systems: they deliver the twofold function of providing a means for managing UNFs, and producing MOX and RepU fuels, that is nuclear

fuels that can be used in nuclear reactors for electricity generation purposes. First order direct effects thus represent reduction in demand for enriched fuel obtained from NatU, and the credits correspond to its avoided production according to the marginal technology that is identified in Section 6.2.3.

6.2.2.2 Direct disposal

The direct disposal approach (Figure 6.1) envisages Used Nuclear Fuels (UNFs) being stored for a number of years (around 50) either at power plants or at a centralised storage or a combination of the two, to allow nuclear fuel to cool and short-lived fission products to decay. In the UK, wet ponds at Sellafield site are likely to be used for this purpose. Since wet ponds do not routinely release discharges and operational consumptions are minor, interim storage of UNFs has not been included in the system boundaries. (Notably, the same has been assumed in the baseline of the reprocessing approach.) Following interim storage, UNFs are assumed to be encapsulated, packaged into disposal canisters and disposed of in a GDF. Encapsulation and packaging of UNFs is based on one of the approaches considered by RWM Ltd. [272], according to which AGR fuel assemblies are first dismantled, with graphite sleeves and other stainless steel components (e.g. support grids, braces) to be removed and processed separately as ILW (notably, this also occurs when they are reprocessed). Individual pins are then consolidated into bundles in specially designed containers, named slotted cans, which are themselves packaged into disposal canisters. Each disposal canister contains eight slotted cans, corresponding to approximately twenty-four AGR fuel elements [267], [272]. The design is inspired to the Swedish KBS-3V concept for LWR fuel [53] – as it is the design of HLW and Pu disposal canisters – but takes into account the different shape of AGR as opposed to LWR fuel assemblies (see Figure 6.4[16]). Encapsulation and packaging of UNFs is assumed to take place in a plant yet to-be-defined (hereby referred to as *tbd*) at Sellafield site, whose construction and decommissioning has not been considered in accordance with the assumptions made for the reprocessing approach.

[16] It should be noted that no decision on fuel disposal has been made by RWM. The concept shown is just one concept investigated by RWM, and other concepts are also under consideration.

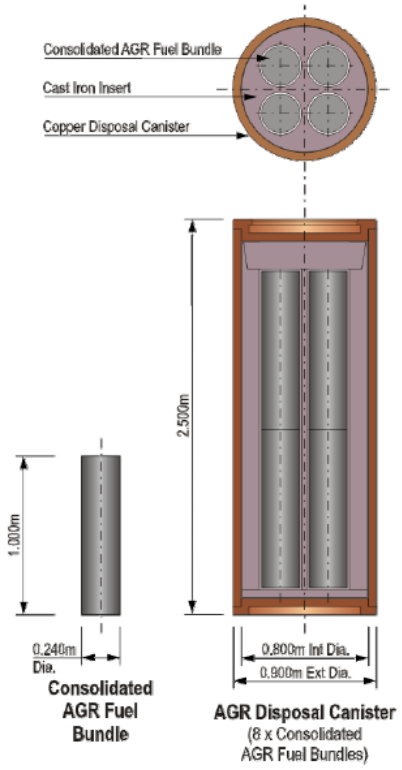

Figure 6.4 – The RWM's modified version of the KBS-3V concept for disposal of AGR UNF (adapted from [317])

6.2.3 Life Cycle Inventory

Following the recommendations of Clift and co-authors [108], two different classes of data are used for the Foreground and Background system. The former makes use of primary data, preferably process-specific operational or design data; whilst for the latter, secondary data, usually available from commercial database, is used. Inventory data for the reprocessing approach baseline, which includes UNFs reprocessing and disposal of higher activity wastes in GDF, has been described in Chapter 5 and is reported in Appendix B; whilst data for all other processes are reported in Section C.1 of Appendix C.

Mass balances related to the Functional Unit and detailing amounts of key intermediate and final products as well as waste generated for the direct disposal approach and each reprocessing scenario are reported in Table 6.3.

Data for encapsulation and packaging of RepU/Pu and UNFs have been obtained from the Derived Inventories for U and Pu [273], and HLW and SNF (note that UNF are referred to as Spent Nuclear Fuel, SNF, in the report) [272] respectively, prepared for the Nuclear Decommissioning Authority (NDA) by Pöyry Energy Ltd and RWM Ltd. Operational or design data for the processes of enrichment of RepU and NatU, production of MOX and

fabrication of fuel assemblies were not available; notably, with the exception of NatU enrichment, none of these activities is currently implemented in the UK. The Ecoinvent database v3.3. has been used instead; but some processes have been amended to reflect specific features of modelled activities. For instance, the Ecoinvent database does not include the process to enrich uranium to 4.1%, but only up to 4%, the main difference being the mass outputs and electricity consumption (quantified in Separative Work Unit, SWU). Furthermore, it is worth noting that the processes of MOX production and MOX fuel assembly fabrication in Ecoinvent are an approximation, due to lack of specific information, because they describe the operation of a uranium dioxide (UO_2) fuel fabrication plant [251]. However, this specific step is known to have little impact, as demonstrated by both Ecoinvent [311], [342] and this study.

Table 6.3 – Mass balances for management of 1 teU of AGR fuel.

Approach		RepU	Pu	Pack'd Pu	Pack'd U	Pack'd DepU	Pack'd UNF	MOX	EnrU
		(kgHM)	*(kgHM)*	*(m^3)*	*(m^3)*	*(m^3)*	*(m^3)*	*(kgHM)*	*(kgHM)*
Reprocessing	S1	970	30	3.5	0.57	-	-	-	-
	S2			3.5	-	0.34	-	-	384
	S3			-	-	0.31	-	96	351
	S4			-	-	0.34	-	64	388
Direct disposal		-	-	-	-	-	1.54	-	-

Data for construction, operation and decommissioning of the GDF is based on that used in Chapter 5 (reported in Appendix B). Radionuclide contents for disposed wastes containing Pu, RepU and UNFs have been obtained from the 2007 Derived Inventories [272], [273]. Notably, the radioactivity reported does not refer to the time at which wastes are generated, rather to that at which disposal occurs; as done in Chapter 5, immediate disposal is assumed for all wastes except HLW and UNFs, for which a storage time of 50 years is assumed.

Transportation between different plants and facilities have also been considered. Distances and mode of transportation have been obtained from Solberg-Johansen's Thesis (1998).

Marginal technologies

As noted in Section 6.2.2.1, production of MOX and RepU fuels for use in nuclear power plants is assumed to cause a decline in demand for enriched fuel from NatU. Because

production of uranium does not represent a constrained market, the procedure developed Bjorn and co-workers (see Chapter 2 Section 2.3.2.1) envisages identification of the marginal technology capable to respond to such changes, which is to be the least competitive technology according to the five-step procedure conceived by Weidema and colleagues and discussed in Section 2.3.2.1 of Chapter 2. However, because the choice of the technology to extract uranium chiefly depends on the depth of mineralisation and the grade of the ore, the least preferred technology cannot be determined objectively. Thus, in this study, the technology that is deemed to contribute to the most to the supply of uranium in the UK is taken to represent the marginal technology.

Apart for uranium mining, the UK has full nuclear fuel cycle facilities in both the front- and back-end of the nuclear fuel cycle. Therefore, the marginal technologies for enrichment and fuel manufacturing are represented by those processes currently carried out in the UK, described in Section 6.2.2.1 and illustrated in Figure 6.3. Uranium in the form of yellowcake, on the other hand, is purchased on the international market. According to figures published by the World Nuclear Association (WNA), uranium production increased by over 50% in the period from 2007 to 2016, from ~41 to ~62 thousand tonnes of U per annum [343]. For an increasing market, the marginal technology is represented by the one most preferred amongst those that are capable of responding to changes, i.e. those that are not constrained. It must be noted that uranium is typically purchased through contracts that may last from two up to ten years; this implies that a reduction of NatU imports to the UK may not occur immediately, but it is assumed that it will as soon as contracts expire. These are still short-term effects if compared to the scale of other changes in the nuclear industry.

No specific information could be retrieved as to which countries or mines currently supply uranium to the UK. According to the report “Governing Uranium in the UK” published by the Danish Institute for International Studies [344], the UK through the years has sourced uranium from about all uranium-producing countries. Although during the first decades of nuclear power in the UK Australia was the main source of uranium, present indications are that an increasing amount of uranium is obtained from Kazakhstan, which from 2009 became the World’s top producer (at the expense of Canada, 2nd, and Australia, 3rd) and has since steadily increased its production [47]. All major mines in Kazakhstan use the In-Situ Leaching (ISL) technology [345], which involves leaving the ore in the ground,

dissolving minerals and pumping the pregnant solution to the surface [346]. The marginal technology is thus represented by a generic ISL mine in Kazakhstan (see Figure 6.3) and the yellowcake is assumed to be transported to the UK via rail and sea freight according to transportation data reported in in Section C.1 of Appendix C. The ISL mine in Kazakhstan is modelled based on generic data included in the Ecoinvent database v3.3.

Furthermore, because of great uncertainty regarding the actual response of the market to marginal changes in uranium demand, it is crucial not to limit the analysis to one, but to consider a range of marginal technologies [116]. As noted above, Canada and Australia are the two major producers of uranium after Kazakhstan. Canada has two major uranium mines, McArthur River and Cigar Lake, both built several hundred meters underground in the Saskatchewan province [347]. In Australia there are three major uranium mines: Ranger is an open pit mine located in the Northern Territory, whilst Four Mile and Olympic Dam both located in the state of South Australia are respectively ISL and underground mines [348]. Both Ranger and Four Mile mines are exclusively devoted to production of uranium, whilst Olympic Dam is primarily a copper mine. Because uranium is effectively a secondary-product, the Olympic Dam mine represents a constrained technology with respect to the uranium market, and thus, it has not been considered in this study. McArthur River and Cigar Lake, Ranger and Four Mile mines have been modelled based on generic datasets for underground, open-pit and ISL mines included in the Ecoinvent database v3.3, but supplemented with country specific data regarding sources of electricity and heat. Additionally, a site-specific dataset gathered on site by Solberg-Johansen has been used for the Ranger open-pit mine in Australia [220]. Transportation data for each of the marginal technologies are reported in Section C.1.

6.2.4 Impact Assessment

This study uses the same impact categories as Chapter 5, and includes all impact categories included in the ILCD recommendations [122], [123] with the exception of Land Use and Ionising Radiations. The former has been excluded due to lack of data, whilst the latter have been replaced by the categories based on the UCrad methodology (Chapter 4). Table 5.1 reports the impact categories considered, along with their metrics and acronyms used in charts included in results Sections.

Table 6.4 – Impact categories analysed

Impact category	Metric	Acronym
Acidification	[Mole of H+ eq.]	A
Climate change	[kg CO_2 eq.]	CC
Ecotoxicity freshwater	[CTUe]	ECf
Eutrophication freshwater	[kg P eq.]	Ef
Eutrophication marine	[kg N eq.]	Em
Eutrophication terrestrial	[Mole of N eq.]	Et
Human toxicity, cancer effects	[CTUh]	HT-c
Human toxicity, non-cancer effects	[CTUh]	HT-nc
Ionizing radiations	[Bq U235 air eq.]	IR
Ionizing radiations, GDF	[Bq U238 ILW eq.]	IRw
Ozone depletion	[kg CFC-11 eq]	OD
Particulate matter/Respiratory inorganics, human health	[kg PM2.5 eq.]	PM/RI
Photochemical ozone formation, human health	[kg NMVOC]	POF
Resource depletion, mineral, fossils and renewables	[kg Sb eq..]	RDm
Resource depletion water	[m^3 eq.]	RDw

6.3 Results

The results Section is divided into four sub-Sections. First, Sections 6.3.1 and 6.3.2 report hot-spot analyses for the reprocessing scenarios and the direct disposal approach; then these options are compared in Section 6.3.3; and finally, Section 6.3.4 presents a comparison of several marginal technologies for uranium mining. LCA results have been calculated by means of Gabi sustainability software version 8 [326].

6.3.1 Reprocessing scenarios

Figure 6.5 shows results of the impact assessment phase for reprocessing Scenarios 1,2,3 and 4. Impacts are expressed as percentage additions to the impact of the baseline (see Section 6.2.2) of each process within a scenario (reported in full colour) and of each scenario, as net summation of each process (reported as a black and white sparse filling). The chart includes six processes, reported in the legend at the bottom of the figure. RepU and MOX fuel fabrication refer to all the activities required to produce the final fuel assembly (see Figure 6.3); the former includes mining and milling, transportation, enrichment and fuel fabrication, whilst the latter only transportation, enrichment and fuel fabrication. Disposal of RepU, Pu and DepU include encapsulation and packaging of waste streams, transportation and disposal in the GDF.

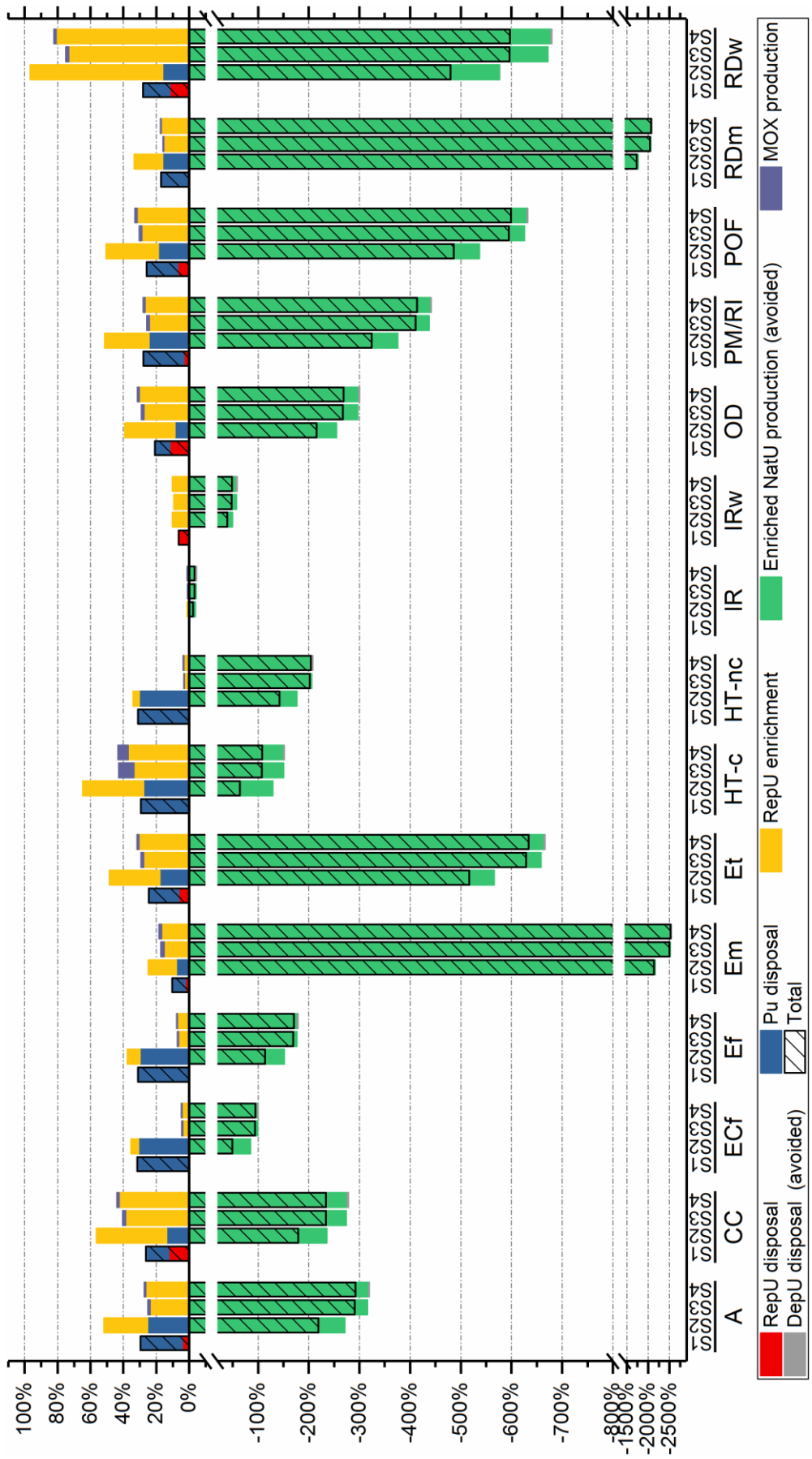

Figure 6.5 - Environmental impacts of Scenarios 1,2,3 and 4 as percentual additions to the impact of the baseline

In Scenario 1 disposal of Pu dominates all but a few impact categories. These include Climate Change (CC), Ozone Depletion (OD) and Resource Depletion of water (RDw), which are approximately equally caused by both disposal of Pu and RepU; and Ionising Radiations from nuclear waste (IRw), which by contrast is entirely attributable to disposal of RepU. More specifically, as reported in Figure 6.6, IRw impact is chiefly caused by two radionuclides, namely uranium-235 (U234) and -238 (U238), contributing to over ~99%. The other ionising radiation category, which is concerned with direct discharges (IR), results in having negligible impact, i.e. lower than 1%, compared to the baseline. Scenario 1 does not include any avoided burdens, thus the net impacts are positive, and range from 10-12% in Resource Depletion (minerals, fossils and renewables – RDm) and Particulate Matter/ Respiratory Inorganics (PM/RI) respectively, to ~20% for the majority of impact categories, and up to 30% in Ecotoxicity (freshwater – ECf). For instance, the value of 10% for RDm means that disposal of Pu and RepU cause an additional impact (to that of the baseline) equal to a tenth of the impact of the baseline.

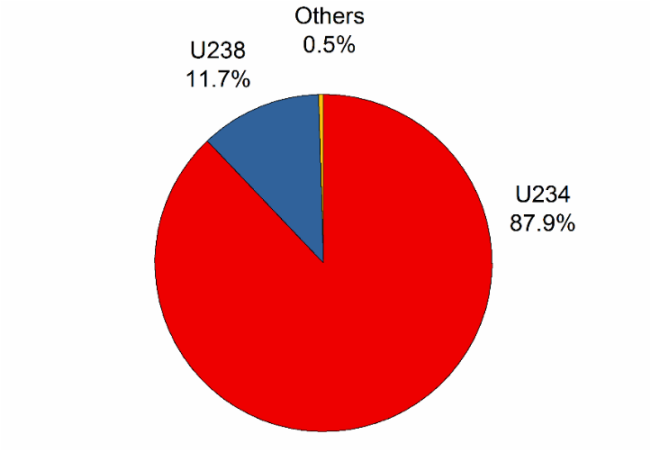

Figure 6.6 – Radionuclides contained in RepU contributing to the Ionising Radiation (waste) category. U234: uranium-234; U238: uranium-238.

In Scenario 2 RepU is recycled (rather than disposed of), meaning that the positive impacts are caused by RepU fuel fabrication and disposal of RepU, and that the system is credited for avoiding production of EnrU fuel based on NatU. The avoided burdens vary considerably in magnitude but are consistently higher than positive impacts and in some cases also higher than the baseline. IR represents the only exception, with both avoided burdens and additional impacts being negligible. ECf and IRw feature the lowest (in absolute terms) avoided burdens, which represent approximately 50-85% of the baseline. Acidification (A,) CC, freshwater Eutrophication (Ef), Human Toxicity (cancer, HT-c and

non-cancer HT-nc effects) and OD feature avoided burdens higher in magnitude than the baseline, from 1 up to 2.5 times; Terrestrial Eutrophication (Et), PM/RI, Photochemical Ozone Formation (POF) and RDw around 4-6 times; Marine Eutrophication (Em) and RDm as high as 17-20 times. Notably, the high score of Em is linked to mining practices, more specifically to significant long-term emissions to fresh water of nitrates contained in the leaching solvent used in uranium mining. With respect to positive impacts: ECf, Ef and HT-nc are dominated by disposal of Pu, while manufacturing of RepU fuel has significant contribution in the remainder. Notably, it dominates Em, CC, OD, IR, IRw and RDw categories, and contributes equally with Pu disposal to A, Et, HT-c, PM/RI, POF and RDm. Where disposal of Pu is the main contributor, positive impacts of Scenario 2 are similar to Scenario 1, otherwise they may be significantly higher. RDw features impacts as high as ~100%, whilst other categories are included in the 25-65% range. Interestingly, net impacts (i.e. sum of positive and negative) of all categories results in being negative, meaning that Scenario 2 contributes to reducing the environmental impacts of the whole process including the baseline; in addition, for the majority of the categories – that is, all categories excluding ECf, HT-c,IR and IRw – net impacts are even lower than -100%, indicating that avoided impacts of Scenario 2 are in absolute terms higher than impacts from the baseline, essentially making the whole process including the baseline "impact-free".

Scenario 3 envisages recycling of Pu alongside RepU, with production of MOX and RepU fuels. The recycling of Pu means that more NatU than in Scenario 2 is displaced, thus leading to higher credits (in absolute terms). Furthermore, since Pu is not disposed of and fabrication of MOX fuel has negligible impacts, also the positive impacts are lower than Scenario 2. Notably, the higher reductions are shown by those categories that are dominated by disposal of Pu in Scenario 2, i.e. ECf, Ef and HT-nc. The reduction of positive impacts coupled with increased avoided burdens leads to net impacts being considerably lower than Scenario 2, with ECf, HT-c and IRw showing the lowest (absolute) values, at minus 50-75% and RDm and Em the highest at approximately minus 2000% and 2500%.

Finally, in Scenario 4 DepU (rather than RepU) is mixed with Pu to produce MOX. The increase in fuel to be enriched and manufactured leads to a slight increase (up to 7%) of positive impacts compared to Scenario 3. However, because more NatU fuel is displaced, avoided burdens are marginally higher (in absolute terms) than Scenario 3. Overall, also

the net impacts of Scenario 4 results in being marginally lower than in Scenario 3. The credits due to avoided disposal of DepU appears to be negligible compared to the savings from producing NatU fuel.

6.3.2 Direct disposal

Impact assessment results for the direct disposal approach are shown in Figure 6.7 and Figure 6.8. The former reports a hot-spot analysis, whilst the latter focuses on the IRw category, showing contributions from individual radionuclides.

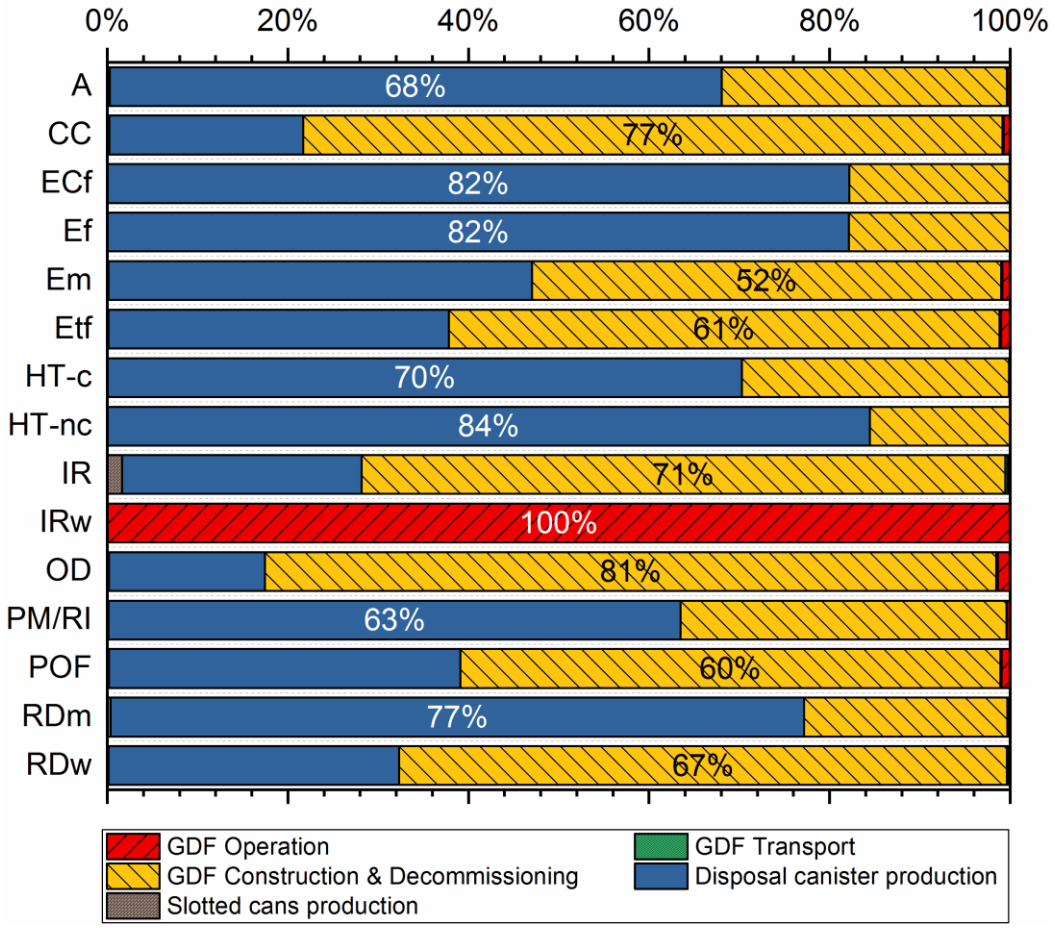

Figure 6.7 – Hot-spot analysis of the direct disposal approach

The contribution analysis shows that all impact categories are dominated by production of disposal canisters, and construction and decommissioning of the GDF, with the exception of the IRw category that is entirely attributed to the operational phase of the GDF. (Because the purpose of the GDF is to isolate for as long as possible nuclear wastes from the biosphere, it is assumed the radioactive emissions arising from the GDF are part of the operational phase.) Impacts related to transportation of UNFs, operation of GDF

and slotted can production (see Section 6.2.2) are in practice negligible. Notably, CC, Em, Et, IR, OD, POF and RDw are dominated by the decommissioning and construction phases, whilst the remaining categories are dominated by manufacturing of the disposal canister. Figure 6.8 shows that more than 95% of the IRw impacts is attributable to two specific radionuclides, namely caesium-135 (Cs135) and tin-126 (Sn126), with remaining 5% being shared by iodine-129 (I129), selenium-79 (Se79), chlorine-36 (Cl36) and nickel-59 (Ni59).

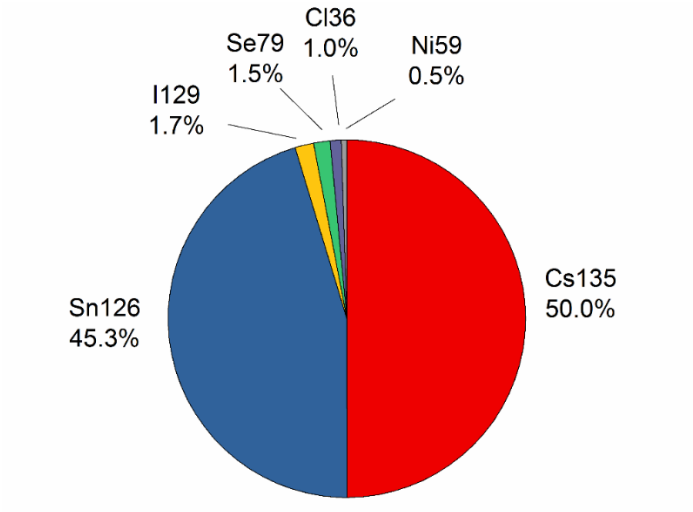

Figure 6.8 – Radionuclides contributing to the Ionising Radiation (waste) category. Cs135: caesium-135; Sn126: tin-126; I129: iodine-129; Se79: selenium-79; Cl36: chlorine-36; Ni59: nickel-59.

6.3.3 Comparison of reprocessing and direct disposal

Table 6.5 presents comparison of the environmental performances of the approaches for managing UNFs considered in this study. The table reports both absolute impacts and the difference between reprocessing scenarios and direct disposal impacts expressed as percentage of direct disposal. Figures refer to all activities required to manage UNFs, meaning that the reprocessing scenarios also include impacts of the baseline. For each category, impacts are ranked from lowest (green) to highest (red) by means of a colour-based system.

The table shows that reprocessing Scenario 1 is the worst performing alternative amongst the ones considered in 10 out of 15 impact categories, with impacts ranging from 8% up to ~560 times higher than direct disposal. Scenario 1 also features the second highest values in ECf, Ef, HT-c, HT-nc and IRw after direct disposal (from 10% to 19% lower). The direct disposal approach results in having the highest or the second highest environmental impacts in all categories with the exception of IR, for which it features the lowest figure (by around 540 times) amongst the options considered. On the other end of

the spectrum, Scenarios 3 and 4 result in being the best environmental options for all impact categories but IR, with negative differences with the direct disposal option ranging from 56% to as high as ~67 times. Scenario 2 shows slightly higher impacts than Scenarios 3 and 4, but considerably lower than the direct disposal approach. It must be noted that Scenarios 2, 3 and 4 results in having negative impacts in all categories concerning non-radiological impacts. No option, in fact, appears to have negative impacts in radiological categories.

Table 6.5 – Environmental impacts of reprocessing scenarios and direct disposal. The colour scale goes from red: highest, to green: lowest.

		Reprocessing				Direct disposal
		S1	S2	S3	S4	
A	[Mole of H+ eq.]	2.68E+03 *(8%)*	-2.47E+03 *(-199%)*	-3.94E+03 *(-259%)*	-3.97E+03 *(-260%)*	2.48E+03
CC	[kg CO2 eq.]	3.01E+05 *(116%)*	-1.88E+05 *(-235%)*	-3.18E+05 *(-328%)*	-3.18E+05 *(-329%)*	1.39E+05
ECf	[CTUe]	1.42E+07 *(-19%)*	5.46E+06 *(-69%)*	6.40E+05 *(-96%)*	5.49E+05 *(-97%)*	1.75E+07
Ef	[kg P eq.]	3.58E+02 *(-18%)*	-3.86E+01 *(-109%)*	-1.90E+02 *(-144%)*	-1.94E+02 *(-145%)*	4.34E+02
Em	[kg N eq.]	9.63E+02 *(202%)*	-1.77E+04 *(-5657%)*	-2.09E+04 *(-6649%)*	-2.11E+04 *(-6719%)*	3.19E+02
Et	[Mole of N eq.]	4.49E+03 *(55%)*	-1.50E+04 *(-618%)*	-1.91E+04 *(-758%)*	-1.93E+04 *(-764%)*	2.90E+03
HT-c	[CTUh]	5.01E-02 *(-18%)*	1.40E-02 *(-77%)*	-2.75E-03 *(-105%)*	-3.18E-03 *(-105%)*	6.08E-02
HT-nc	[CTUh]	6.56E-01 *(-19%)*	-2.12E-01 *(-126%)*	-5.11E-01 *(-163%)*	-5.20E-01 *(-164%)*	8.12E-01
IR	[Bq U235 air eq.]	1.88E+09 *(56008%)*	1.83E+09 *(54624%)*	1.82E+09 *(54214%)*	1.82E+09 *(54247%)*	3.35E+06
IRw	[Bq U238 ILLW eq.]	7.12E+10 *(-10%)*	4.09E+10 *(-48%)*	3.49E+10 *(-56%)*	3.45E+10 *(-56%)*	7.88E+10
OD	[kg CFC-11 eq.]	3.88E-02 *(221%)*	-3.70E-02 *(-406%)*	-5.37E-02 *(-544%)*	-5.41E-02 *(-547%)*	1.21E-02
PM/RI	[kg PM2.5 eq.]	2.50E+02 *(8%)*	-4.37E+02 *(-289%)*	-6.07E+02 *(-363%)*	-6.12E+02 *(-365%)*	2.31E+02
POF	[kg NMVOC]	1.29E+03 *(48%)*	-3.97E+03 *(-554%)*	-5.09E+03 *(-682%)*	-5.13E+03 *(-687%)*	8.74E+02
RDm	[kg Sb eq.]	5.69E+01 *(57%)*	-7.94E+02 *(-2298%)*	-9.44E+02 *(-2714%)*	-9.53E+02 *(-2741%)*	3.61E+01
RDw	[m^3 eq.]	2.11E+03 *(87%)*	-6.26E+03 *(-654%)*	-8.18E+03 *(-824%)*	-8.18E+03 *(-825%)*	1.13E+03

6.3.4 Comparison of marginal technologies

Table 6.6 presents a comparative analysis of the environmental impacts associated with four marginal technologies for uranium mining, namely an underground mine in Canada, an ISL mine in Australia and an open pit mine in Australia (described by generic and the Ranger mine site-specific datasets). The IRw category is not reported in the table because uranium mines do not generate wastes requiring deep geological disposal. The table uses a colour-based legend to rank impacts in each category from lowest (green) to highest (red).

Table 6.6 – Environmental impacts of five marginal technologies for uranium mining. The color scale goes from red: highest, to green: lowest.

		Kazakhstan	Canada	Australia		
		ISL	Underground	ISL	Open cast	Ranger mine
A	[Mole of H+ eq.]	1.31E+00	1.28E+00	1.31E+00	1.21E+00	2.76E-01
CC	[kg CO_2 eq.]	8.09E+01	7.71E+01	8.09E+01	8.57E+01	3.25E+01
Ef	[kg P eq.]	1.13E-01	2.47E-02	1.13E-01	3.23E-02	2.78E-03
Em	[kg N eq.]	5.82E+00	5.05E-01	5.82E+00	3.30E-01	2.76E-02
ET	[CTUe]	2.30E+03	1.58E+03	2.30E+03	1.82E+03	2.08E+02
Et	[Mole of N eq.]	5.46E+00	4.43E+00	5.46E+00	3.11E+00	3.02E-01
HT-c	[CTUh]	5.85E-06	1.11E-05	5.85E-06	1.09E-05	2.53E-06
HT-nc	[CTUh]	2.61E-04	5.00E-05	2.61E-04	5.25E-05	9.26E-06
IR	[Bq U235 air eq.]	3.58E+02	4.69E+06	3.57E+02	1.06E+07	1.29E+03
OD	[kg CFC-11 eq,]	1.57E-05	1.26E-05	1.57E-05	8.02E-06	4.24E-07
PM/RI	[kg PM2.5 eq.]	1.87E-01	1.63E-01	1.87E-01	1.32E-01	1.94E-02
POF	[kg NMVOC]	1.44E+00	1.24E+00	1.44E+00	9.08E-01	8.80E-02
RDm	[kg Sb eq.]	2.62E-01	2.51E-01	2.62E-01	2.51E-01	2.20E-01
RDw	[m^3 eq.]	1.81E+00	8.21E-01	1.81E+00	1.83E+00	1.67E-01

The analysis shows that, with the exception of the IR category and the Australian Ranger mine described by a site-specific dataset, impact scores do not differ by more than one order of magnitude. Some categories, like A or CC, feature the highest variance between options being as little as 8-10%. The IR category, on the other hand, features variations as high as five orders of magnitude between the ISL mines and the generic open pit mine in Australia. Overall, the generic ISL mines in Kazakhstan and Australia feature the highest environmental impacts in 10 out of 14 categories, and the lowest IR impact amongst the technologies considered, whilst the Australian Ranger results in having the lowest environmental impacts in all categories with the exception of IR. The difference between the Ranger mine and the remainders is considerable and up to one order of magnitude in

categories such as A, Ef, HT-c and HT-nc. The generic underground mine in Canada and the generic open cast mine in Australia have intermediate performance, with the former having the highest impact in HT-c, and the latter in CC, HT-c, IR and RDw.

The analysis demonstrates that if a different marginal technology is chosen between a generic underground mine in Canada or a generic open pit mine in Australia, the comparison amongst reprocessing scenarios and the direct disposal approach would show significant changes only in the IR category. Notably, because both the generic underground and open pit mines feature the highest IR impact scores, Scenarios 2, 3 and 4 would feature higher avoided burdens associated with uranium mining. The complete LCA results for the reprocessing scenarios employing the generic underground and open-pit mines are reported in Section C.4 of Appendix C. They show that reprocessing Scenarios 2, 3 and 4 have lower IR impact scores and that Scenarios 3 and 4 are the most preferred environmental options for all impact categories considered. On the other hand, the analysis also demonstrates that the choice of the Ranger mine as marginal technology would lead to changes in favour of the direct disposal approach in all impact categories. However, the LCA results reported in the Section C.4 show that Scenarios 3 and 4 are still the most preferred options for all impact categories but IR. Finally, it should also be noted that for marginal technologies other than ISL, Scenario 4 does not always result in being (although marginally) favourable than Scenario 3. For instance, when the marginal technology is represented by the Ranger mine, Scenario 3 has net environmental impacts lower than Scenario 4 in ten impact categories.

6.4 Discussion

This Chapter presented a comprehensive LCA study of five alternative approaches for management of Used Nuclear Fuels (UNFs), including four reprocessing scenarios and direct disposal. As noted in the introduction, the UK is currently pursuing a so-called “nominal” Twice-Through Cycle, with Pu and RepU lying in a limbo where they are neither considered waste nor valuable products. The reprocessing scenarios represent four alternative options for the UK should the Government decide to continue reprocessing of UNFs; they differ in whether and how RepU and Pu are disposed or recycled. The fifth is the preferred approach by the majority of countries nowadays (e.g. the US, Sweden, Finland): UNFs are declared waste (and perhaps more appropriately termed Spent

Nuclear Fuels, SNFs), packaged in final disposal canisters and disposed of in a GDF. It should be noted that the final destination for higher activity wastes, regardless how UNFs are managed, is the Geological Disposal Facility (GDF). Currently, deep geological disposal represents the safest, but also the only feasible approach for disposing nuclear waste, supported by the majority of nuclear countries. In 2016, the South Australia's Royal Commission on the Nuclear Fuel Cycle has recommended the government to pursue establishment of disposal facilities for national and international nuclear waste [349]. The recommendations represent a fundamental change: the establishment of a central repository, possibly in remote and isolated lands, may foster its social acceptance as well as bring down capital costs. Those countries that do not support the GDF have yet to identify an alternative solution to their nuclear waste problem, and intend to implement interim superficial wet or dry storage as a temporary solution.

Results of the comparative analysis (Section 6.3.1 and Table 6.5) show that, with the exception of radiological impacts due to direct discharges, the reprocessing approach is more environmentally beneficial than direct disposal only when either uranium or both uranium and plutonium are recycled (Scenarios 2,3 and 4). The production of nuclear fuel from recycled materials avoids mining and enrichment of uranium ore – the two steps of the nuclear fuel cycle with the greatest impact [220], [253]. The avoided production of NatU fuel, displaced by either only RepU fuel or RepU fuel and MOX, offset the additional environmental impacts and generates significant environmental gains, even higher than impacts linked with UNFs reprocessing and disposal of fission products, namely the baseline; Scenarios 2, 3 and 4, in fact, feature several impact categories with negative scores (Figure 6.6 and Table 6.5). The analysis also demonstrates that recycling of Pu is of paramount importance for two main reasons. First, disposal of plutonium is a relatively high impact activity (as shown in Figure 6.5, it dominates the majority of positive impacts), and second, plutonium generates 50% more energy than uranium on a mass basis. This means that Pu can be mixed with low enriched uranium (i.e. RepU and DepU) to produce nuclear fuel with a higher equivalent enrichment, thus avoiding the need for further enrichment – a high-impact, energy-intensive process. For these reasons, Scenario 3 and 4 results in being the most favourable options. The importance of recycling of Pu is also highlighted by the footprint of the GDF, in terms of above and below area, required to host the amount of nuclear waste generated by each approach. Figure 6.7 shows GDF footprints of each option relative to that of Scenario 3, that have been estimated from

the volume of packaged wastes generated by each option and the area per volume required by each type of waste according to the GDF generic design for higher strength rock developed by the NDA (details are reported in Section C.3 of Appendix C). According to Table 6.7, scenarios not envisaging recycling of Pu, i.e. Scenarios 1 and 2, require a GDF with a footprint a thousand times higher than those for Scenario 3 and 4.

Table 6.7 – GDF footprint relative to reprocessing Scenario 3

Reprocessing				Direct Disposal
S1	S2	S3	S4	
1106.2	1105.4	1.0	1.1	48.9

It is also obvious why Scenario 1 results in being the worst available option: the separation of fission products, uranium and plutonium for disposal purposes does not yield any actual environmental benefits – rather it makes little sense, especially when compared to direct disposal. Both reprocessing Scenario 1 and direct disposal, in fact, do not recognize the value of unused resources contained in UNFs; however, whilst the direct disposal approach envisages UNFs to be disposed of altogether, the reprocessing Scenario 1 separates individual waste streams with the final aim of disposing of them in the same way as the direct disposal approach. In addition to this, Scenario 1 increases proliferation risks, amount of wastes and size of the GDF required for their disposal. Besides these many flaws, Scenario 1 has one main benefit: vitrification of fission products (the most troublesome waste stream) in glass generates a homogenous, compact and secure waste form that, because it predominantly contains fission products (which have significantly shorter half-lives than uranium and plutonium), will decay much faster than SNFs.

Although recycling of valuable materials is an environmentally beneficial practice in all industries, in the nuclear field it assumes a different tone. Uranium is, in fact, a relatively concentrated resource; reprocessing and recycling of uranium and plutonium can thus guarantee the security of supply to those countries with no available uranium resources and protect against market volatility. Interestingly, the case for reprocessing may not be strictly linked with efficient use of resources, but rather with avoiding construction and operation of uranium mines. It is estimated that known and expected resources could last up to 300-400 years with current Once- and Twice Through Cycles, and that uranium could become nearly inexhaustible if unconventional resources such as uranium in sea water

are used [350]. The deployment of fast reactor technologies could also extend the lifetime of conventional and expected resources up to infinite.

The practice of reprocessing, however, has two main arguments against it. First, reprocessing is often linked with an increase in risk of nuclear proliferation due to the potential spread of nuclear weapons and fissionable material to non-"Nuclear Weapon States"; the plutonium stream obtained from UNFs reprocessing may in fact be diverted and used to make nuclear weapons. This concern has motivated development of other separation techniques designed to prevent the separation of a pure plutonium stream. Second, it has been found by two independent studies carried out by the Massachusetts Institute of Technology (MIT) on the future of the nuclear power [351] and Harvard on the economics of spent nuclear fuel management [352], that the cost of reprocessing is far higher than that of direct disposal. According to these studies, with uranium price equal to US$40/lb (today's price is ~US$20/lb), a reprocessing-based fuel cycle would increase the cost of nuclear electricity by US$1-2/MWh; otherwise, to break even the price of uranium would need to go up to US$360/lb – a price never reached and not likely to be seen in the coming years (even more so if price rebound effects are considered). This is based on the assumption that the price of the current primary source of energy, i.e. fossil fuels, remains stable – another highly unlikely scenario. Development of emerging countries, especially China and India, will substantially increase demand of fossil fuels, that coupled with reduction of fossil reserves and higher costs required for their exploitation, will indeed cause fossil fuels price to rocket. By then, economic feasibility of nuclear energy will only be based on the cost of renewable energies; only if their cost will be comparable, conventional[17] nuclear energy survives.

The direct disposal approach, on the other hand, represents a straightforward, low-cost and proliferation-resistant approach for managing UNFs that does not require construction and operation of complex reprocessing plants and associated waste treatment facilities. The comparative analysis has shown that it represents the option that delivers the lowest radiological impacts from direct discharges (IR) amongst the options investigated. Notably, this is valid only when the marginal technology corresponds to a generic In-Situ Leaching (ISL) located in Kazakhstan or Australia, or an open pit mine in

[17] This discussion does not include nuclear fusion, which, because it is yet to achieve a positive net production of energy, does not currently have a market.

Australia described by the Ranger site-specific dataset (Section 6.3.4). If the marginal technology is assumed to be represented by an underground mine in Canada or an open pit mine in Australia described by generic datasets, the reprocessing Scenarios envisaging recycling of RepU (Scenarios 2, 3 and 4) will result in having lower IR scores than the direct disposal approach. This is because the generic datasets for underground and open pit mining technologies report radioactive discharges arising from mining sites being considerably higher than the ISL technologies and the Ranger mine, thus increasing the significance of avoided burdens. The contribution analysis on the direct disposal approach has revealed that impacts are chiefly linked with use of copper as corrosion resistant material in disposal canisters (notably, the same result has been obtained for the baseline, see Chapter 5), and with construction and decommissioning of GDF.

Finally, the analysis of environmental impacts associated with different marginal technologies for uranium mining demonstrates that the ranking of the options is very stable. The choice of the marginal technology chiefly affects the IR category. This is because technologies like In-Situ Leaching release much less radioactive materials than underground and open pit. Besides IR, the options ranking for the other impact categories is only marginally affected.

To conclude, it must be pointed out that the approaches compared serve the purpose of managing nuclear waste. A one-to-one substitution has been assumed between MOX/RepU and NatU fuel. However, MOX and RepU fuels cannot be recycled indefinitely as the continuous building-up of radionuclides poisons reduce their efficiency. This implies that the results obtained here do apply for waste management systems, but cannot be immediately translated to assessment of nuclear fuel cycles, i.e. Once-Through Cycle vs Twice-Through Cycle.

This study includes valuable information for policy and decision-makers and clear indications for the British nuclear industry in view of crucial decision that are expected to be taken in future years. The study identifies reprocessing with recycle of both Pu and U as the most environmentally sustainable approach for the back-end of the UK nuclear fuel cycle. However, this must not be confused with the most sustainable. A truly sustainable approach is the one that meets the “three pillars” of sustainability, namely economics and social alongside the environment. Life-Cycle Costing (LCC) and Social LCA (S-LCA) studies

(see Chapter 1) on nuclear waste management systems are thus required to complement the present one.

6.5 Conclusion

Crucial decisions lie ahead for the UK in terms of the future of its nuclear fuel cycle. A notable example is reprocessing vs direct disposal. Is it worth continuing reprocessing Used Nuclear Fuels (UNFs)? And if so, what is to be done with reprocessed uranium and plutonium? Or should UNFs be directly sent for disposal? This Chapter aimed at addressing these questions by tackling their environmental aspects by means of a comprehensive Life Cycle Assessment (LCA) study. Five approaches for management of UNFs have been considered, including a direct disposal approach and four reprocessing scenarios envisaging different strategies for disposal and/or reuse of uranium and plutonium. The comparative analysis shows that recycling of Pu and RepU is of paramount importance: the environmental gains linked with avoided production of nuclear fuels from mined uranium offset the environmental impacts of additional activities such as separation of fission products from uranium and plutonium, resulting in environmental savings in numerous impact categories. Notably, the approaches envisaging production of MOX fuel from reprocessed or depleted uranium and plutonium (namely Scenarios 3 and 4) overall result in being the most preferred options from an environmental perspective, showing negative impacts in all categories concerned with non-radiological impacts. On the other hand, reprocessing of UNFs with disposal of reprocessed uranium and plutonium, and the direct disposal approach represents globally the worst option amongst those considered. The direct disposal approach may appear favourable only with respect to radiological impacts from direct discharges depending on the marginal technology chosen. Besides this category, the ranking of options is very stable with respect to the choice of the marginal technology. The case for reprocessing is even stronger if other factors, such as security of supply and protection against market volatility of uranium, are taken into account. By contrast, the high cost compared with direct disposal and the potential risks for proliferation of nuclear weapons are the biggest arguments against.

Chapter 7.
Conclusions and future work

7.1 Conclusions

The 21st century is likely to be remembered as the century of sustainability and sustainable transformations of industrial sectors, when the human race has finally acknowledged that its development solely aimed at economic growth is causing major adverse effects to the planet and its ecosystems. The Earth has been in a stable geological era, the Holocene, for millennia, that has allowed our society to grow and flourish; we must safeguard this state to avoid indefinite destabilization of the planet. The second decade of the 21st Century was marked by an historic and unprecedented event: the Paris Agreement, signed by the majority of nations, that brings all nations into a common cause to undertake efforts to combat climate change by curbing greenhouse gas emissions to restrain global warming to well below 2 °C and possibly even lower to 1.5 °C. Because the agreement lacks a quantitative path, nations around the world are developing and implementing their own policies to meet the Paris Agreement goals, but it is generally recognized that the power generation and distribution sector has the biggest potential for emissions reductions. Decarbonisation of this sector cannot be achieved through deployment of a single technology, rather only by diversification of energy sources and technologies: both the European Union (EU) and the United Kingdom (UK) rely on a combination of renewable energy, Carbon Capture and Storage (CCS) technologies and nuclear energy for their future energy systems' scenarios. The term nuclear renaissance has been used to refer to the case for an increase in the use of nuclear power after decades of shrinking capacity; the UK, for instance, has seen commencement of construction of its first nuclear reactors at Hinkley Point C after more than three decades, with other projects either planned or proposed. Nuclear energy contribution to global electricity supply is projected to be one of the fastest growing energy sources after renewables in the next decades. However, the nuclear industry needs to address a number of challenges that can undermine its credibility as a sustainable source of energy; notably, the biggest challenges refer to the

back-end of the nuclear fuel cycle, in particular to how Used Nuclear Fuels (UNFs) are managed and how the final, radioactive solid waste is disposed of. This Thesis aimed at using a Life Cycle Thinking approach to address these questions, with a specific focus on the environmental aspect. Life Cycle Assessment (LCA) methodology is the most used and the only ISO standardised amongst life-cycle methodologies, but thus far it has very seldom been applied to the nuclear industry.

Within this context, this Thesis sought to address the following objectives:

1. Identify the main barriers that prevented application of LCA to the nuclear field, and develop a novel framework for incorporating radiological impacts of direct radioactive discharges and especially of future emissions arising from disposed solid waste, to be incorporated in the Impact Assessment phase of LCA;
2. Quantify and evaluate the environmental impacts of nuclear waste management practices to identify hot-spots and possible improvement options. Two UK-based case studies have been performed, these addressed:
    a. Current practice for managing Used Nuclear Fuels (UNFs) and disposal of nuclear waste;
    b. Comparison of future approaches that the industry could implement to manage UNFs;
3. Demonstrate with practical applications how LCA can be used to support decisions in the Nuclear Industry.

The key findings of this Thesis can be summarized in the following points:

1. The lack of a standard framework and methodology for assessing the impacts of radioactive emissions, especially those arising from disposed nuclear waste, represents the major barrier towards the application of LCA to the nuclear field (Chapter 3). This Thesis aimed at tackling this issue by proposing a novel framework and two practical, and conceptually very different, methodologies (Chapter 4). The Critical Group Methodology (CGM) is inspired to risk assessment practices and mainly serves as a reference against which UCrad, the multimedia environmental model based on the UNEP-SETAC consensus model USEtox, is validated. Characterisation factors for both methodologies are calculated, and included for dissemination in this Thesis and in peer-reviewed articles.

2. This Thesis presented two LCA studies (Chapter 5 and Chapter 6) that i) provided a comprehensive analysis of the environmental profile of current and future nuclear waste management practices in the UK and ii) illustrated the first application of the radiological impact methodologies developed within this Thesis.
    a. The LCA study presented in Chapter 5 focused on the current practice for managing UNFs and the UK Government policy for disposal of nuclear waste. Analysis of the results showed that the most significant impacts are caused primarily by consumption of uranyl nitrate (used in the PUREX process to separate uranium from plutonium) and copper (used in the Swedish disposal canister design adopted as a reference in this Thesis), whilst construction, operation and decommissioning of the Geological Disposal Facility (GDF) only have minor contributions.
    b. An analysis of radiological impacts has also been performed in Chapter 5 through a comparison of results obtained from UCrad, CGM and the site-specific methodology used by Sellafield Ltd. The analysis showed that UCrad and CGM provides very different results, but these define a range of impacts within which more accurate estimates such as those from site-specific methodologies can be anticipated to be found.
    c. The LCA study presented in Chapter 6 extended the scope of the study presented in Chapter 5 by looking at future scenarios for the back-end of the UK nuclear fuel cycle, which include the Once Through Cycle (i.e. direct disposal of nuclear waste) and four alternative reprocessing scenarios (that differ according to how plutonium and uranium are used and disposed). The study demonstrated the significance of uranium mining with respect to the impacts of the whole nuclear fuel cycle; and concluded that reprocessing with production of Mixed Oxide (MOX) fuel is the most environmentally favourable option in terms of non-radiological impacts, whereas direct disposal may be more beneficial (depending on the choice of the marginal technology) with regard to direct radiological impacts.
3. The LCA study in Chapter 6 also illustrated how LCA can be used to support decision-making in the nuclear industry by comparing alternative options for

managing Used Nuclear Fuels (UNFs) in the UK. Results of the study seem to support the case for reprocessing UNFs with recycling of both Pu and U primarily for production of MOX.

Further details on the key findings are included in the following Sections.

7.1.1 Radiological Impact Assessment in LCA

The review on Radiological Impact Assessment methodologies presented in Chapter 3 has highlighted a significant methodological gap in the Impact Assessment phase of LCA. The review focused on seven methodologies – five of which focused on potential impacts on humans and two on the environment – that have been purposely developed for incorporation in Life Cycle Impact Assessment (LCIA) or that are standard procedures in other fields and could be integrated in LCIA. From the analysis it is concluded that none of the methodologies reviewed is sufficiently comprehensive, mature or consistent with life cycle thinking for general adoption in LCIA. Notably, the inclusion of future emissions arising from radioactive waste disposed in a final repository represents the most critical limitation. In this Thesis it is argued that only deterministic future emissions from disposed radioactive waste should be included in the Inventory phase; inclusion of stochastic emissions would be both inconsistent with other LCIA methodologies and outside the current scope of LCA.

For human impacts, two methodologies stand out. The Human Health Damages approach developed by Frischknecht and co-workers specifically for LCIA, is the only radiological impact approach ever included in LCIA methods thus far, but it is not equipped to assess impacts of solid waste disposed of in a final repository. The Human Irradiation approach developed by Solberg-Johansen, on the other hand, has been purposely developed to incorporate impacts of radioactive waste, but both stochastic and deterministic emissions are included. It is concluded that to achieve acceptance a radiological impact methodology should combine elements of both the Human Health Damages and Human Irradiation, i.e. the methodology should be consistent with other LCIA methodologies, potentially comparable with non-radiological impacts and include future deterministic emissions from solid waste disposed in a final repository. With respect to impacts on the environment, the main limitation is represented by the lack of data on bio-availability of radionuclides and their effects on non-human biota. Both the ecological impacts-focused methodologies appeared to be immature for general adoption: SLERA is a very promising

approach, mainly because it is based on USEtox, but it needs to be extended to cover all relevant types of emissions.

7.1.2 A novel framework and two impact methodologies

In light of the conclusions and recommendations of the review, a novel, overarching framework for assessing impacts of radionuclides within Life Cycle Impact Assessment (LCIA) has been developed, and is discussed in Chapter 4. The framework consists of three modules – namely fate, exposure and effect – and establishes the ground upon which practical methodologies can be developed. Within this Thesis, two methodologies have been developed and differ exclusively for the fate module, with the other two modules being in common. UCrad is the first-of-its-kind compartment-type methodology developed to specifically address radionuclides' fate; it is largely based on the USEtox model for toxic substances. The Critical Group Methodology (CGM) adopts the concept of critical group developed for risk assessment purposes, and uses analytical models such as the Gaussian plume for atmospheric emissions to estimate fate of radionuclides. Notably, adoption of the critical group concept entails that several set of characterisation factors can be calculated depending on the distance of the critical group from the source of the release. The exposure and effect modules, on the other hand, are based on established practices in the nuclear protection field. A comprehensive database based on a hierarchical approach for sources selection has also been developed to support both methodologies. An extensive analysis have been carried out to validate the methodologies and investigate the effects of the different approaches for modelling the fate of radionuclides; the analysis consists of three levels of comparison performed between i) UCrad, CGM for a distance of 1000 km of the critical group and the Human Health Damages; ii) UCrad and CGM for various distances of the critical group; and finally, iii) USEtox and an adapted version for toxic substances of CGM with varying distances of the critical group. The analysis demonstrates i) that the developed methodologies compare well with the Human Health Damages (discussed above and in Chapter 3), the only methodology included in LCIA methods thus far; ii) that UCrad, but especially CGM, are strongly affected by radionuclide's half-lives; and iii) that the same kind of results can be obtained for toxic substances. With respect to applications, UCrad indeed represents a more suitable candidate than CGM for general adoption in LCIA, primarily because it is consistent with impact methodologies developed for toxic substances; the Critical Group Methodology combines elements of LCA and Risk Assessment and its incorporation in

LCIA may be debated. Nonetheless, there could be some specific cases where CGM and UCrad may be used in combination, for instance with the purpose of determining the location or the maximum scale of a plant.

7.1.3 Assessing the impacts of Used Nuclear Fuels reprocessing: a UK case study

Management of Used Nuclear Fuels (UNFs) is indeed the most debated phase of the nuclear fuel cycle, especially in relation to its potential impacts on humans and the environment. Chapter 5 assesses the impacts of reprocessing UNFs in the UK. The study represents the first application of the radiological impact assessment methodologies developed within this Thesis and shows by practical application how LCA can be used in the nuclear industry to identify the processes, plants or streams with the highest impacts. From a methodological perspective, the study adopts an attributional approach with a prospective perspective: it focuses on the current practice for reprocessing UNFs at Sellafield site and the UK Government policy for disposing of higher activity wastes in the national Geological Disposal Facility (GDF). Because construction of the national GDF, and therefore disposal of higher activity wastes, has yet to commence, this study makes several crucial assumptions on both the GDF (e.g. location, geology and layout) and design of disposal canisters (e.g. for High Level Wastes the Swedish concept is used as a reference example; but it is only one of the concepts studied in the UK). The inventory consists of a mix of operational flowsheets obtained during a short secondment at Sellafield site and publicly available documents such as those published by the NDA and RWM Ltd. Analysis of the results shows that toxicity and radiological are the most critical impacts after normalization, though the former may be due to biases introduced in the normalisation procedure. The majority of impacts, especially those most critical (i.e. with high normalised scores), can be attributed to consumption of uranyl nitrate used in THORP to separate uranium from plutonium, and copper used in the Swedish-designed disposal canister for HLW. Impacts associated with construction, operation and decommissions of the GDF are generally minor, especially for those categories with the highest normalised impacts, and are mainly attributable to consumption of electricity in the construction phase and use of bentonite in the decommissioning phase. With respect to radiological impacts arising from disposed solid waste, over two thirds are caused by disposal of Multi

Element bottle (MEB) and Vitrified High Level Waste (VHAL) as streams and iodine-129 as radionuclide.

Besides the LCA study, Chapter 5 also presented an in-depth analysis of radiological impacts which included comparisons between UCrad and CGM, and with the site-specific methodology used by Sellafield Ltd. The comparisons show that differences between UCrad and CGM are consistent both in terms of absolute impact scores and of contributions by radionuclides; the result is not unexpected (as discussed in Chapter 4) and can be linked to adoption of the critical group concept and different approaches to fate modelling. Radiological impacts estimated by means of the site-specific methodology are also quite different, but they fall within the upper and lower figures estimated respectively by CGM and UCrad.

7.1.4 Direct disposal vs Reprocessing: Assessing the impacts of future scenarios for the UK back-end nuclear fuel cycle

With planned shutdown of reprocessing operations at Sellafield site and no final decision on how UNFs will be managed in the future, the UK and its nuclear industry are entering a critical time when decisions are expected to be taken. Chapter 6 presents a consequential LCA study that extends the scope of the study presented in Chapter 5 to consider possible approaches for managing UNFs in the UK. The study represents a clear example of how LCA can be used as a decision-support tool in the nuclear industry. Scenarios investigated include Once Through Cycle with direct disposal of UNFs and four reprocessing scenarios envisaging different combinations of recycling and disposal of reprocessed uranium and plutonium. The inventory is based on that produced for the attributional study (Chapter 5) and supplemented with data from public sources for those processes not included in its system boundaries. The study relies on a number of assumptions including those made in the attributional study (e.g. on the GDF, or the disposal canister design for HLW), and others such as disposal canister designs for plutonium or uranium. The comparative analysis between the five investigated scenarios showed that reprocessing of UNFs with production of MOX from plutonium and either reprocessed uranium or depleted uranium (Scenarios 3 and 4) are the most favourable approach for non-radiological impacts. With respect to radiological impacts, a clear trade-off exists between direct disposal and reprocessing with production of MOX: the former has the lowest impact from direct discharges whilst the latter from solid wastes disposed

in a GDF. The benefits of UNFs reprocessing with MOX production are linked to the avoided production of nuclear fuel from "fresh" uranium: the avoided burdens, mainly those from uranium mining, offset the environmental impacts of additional activities, including those carried out at THORP. Because the majority of avoided burdens are associated with uranium mining, the choice of the mining technology is of crucial importance. From a market analysis the study assumed that uranium imported in the UK comes from a generic mine in Kazakhstan using the In-Situ Leaching (ISL) technology; however, recognizing the uncertainty regarding the actual response of the market to marginal changes, several potential marginal technologies for uranium mining have been considered. The analysis showed that the choice of the marginal technology only affects radiological impacts from direct discharges, while the ranking for non-radiological impacts remains the same. For some marginal technologies such as a generic underground mine in Canada or a generic open-pit mine in Australia, reprocessing results in being the preferred option also for radiological impacts. With respect to the direct disposal approach, the majority of impacts are associated with manufacturing of the disposal canister (notably, with consumption of copper), and construction and decommissioning of the GDF. Results of this study support the case for continuing reprocessing of UNFs in the UK. The argument for reprocessing is even stronger if other factors, such as security of supply and protection against market volatility of uranium, are taken into account. The high cost compared with direct disposal and the potential risks for proliferation of nuclear weapons are, however, the biggest arguments against.

7.2 Future work

In light of the main findings, this Thesis concludes by recommending the following areas for future research works:

- UCrad represents a very promising methodology for general adoption in LCIA, and for this reason efforts should be directed into its further development. Notably, these should focus on three aspects: first, the methodology should be integrated with a model for estimating flow and travel times of radionuclides escaping from the GDF; second, the methodology should consider the full decay chain of radionuclides in fate modelling, instead of using the simplifying assumptions that decay represents a removal from compartments; and third, the methodology

should be integrated with USEtox so that toxicological and radiological impacts can be evaluated and comparted within a single framework.

- The LCA studies and their results are strictly linked to crucial assumptions related to GDF and disposal canisters designs. Further studies should be performed with the aim investigating the effects of such assumptions; and their results could be used to support decisions such as on the choice of the design for disposal canisters, or on the location of the GDF.
- Furthermore, because the LCA studies performed within this Thesis focused exclusively on the nuclear waste management phase of the nuclear fuel cycle, further works are recommended to: i) extend the scope to consider the nuclear fuel cycle in its entirety and ii) analyse different phases such as decommissioning of existing plants and land remediation. An LCA study of such type would aim at addressing questions such as: should plants decommissioning be deferred to allow decay of short-lived radionuclides or performed as soon as practicable? What are the benefits and trade-offs of both approaches? And how do they relate to the envisaged 'End State' for the site, the potential value of the land or its final possible future use following completion of the nuclear mission?
- Finally, this Thesis only looked at the environmental aspect of sustainability; further studies should be performed on the economic and social aspects by means of Life Cycle Costing (LCC) and social Life Cycle Assessment (S-LCA) (introduced in Chapter 1) to produce a comprehensive sustainability assessment. This may be of particular importance to support decisions on management of Used Nuclear Fuels (UNFs) that until now have solely been driven by economic considerations.

Glossary

Streams/Materials/Products/Wastes	
BC	Barium Carbonate
CC	Centrifuge Cake
DepU	Depleted Uranium (Enrichment tails)
DNLEU	Depleted/Natural/Low Enriched Uranium
DNLEU	Depleted/Natural/Low Enriched Uranium
EnrU	Enriched Uranium
FPs	Fission Product
HLW	High Level Waste
I-LLW	Intermediate-Low Level Waste
LAE	Low Active Effluents
LEU	Low Enriched Uranium
LLW	Low Level Waste
LWR	Light Water Reactor
MEB	Multi Element Bottle
MOX	Mixed Oxide fuel
NatU	Natural Uranium
Pu	plutonium
RepU	Reprocessed Uranium
SEC	Salt Evaporate Concentrate
SNF	Spent Nuclear Fuel
SS	Stainless Steel
U	Uranium
UNF	Used Nuclear Fuel
VHAL	Vitrified Highly Active Liquor

Units/Plants/Reactors	
AGR	Advance Gas-Cooled Reactor
CS	Chemical Separation
DOG	Dissolver-Off Gas
EARP	Enhanced Actinide Removal Plant
EPS	Encapsulated Product Store
FHP	Fuel Handling Plant

GDF	Geological Disposal Facility
HALES	Highly Active Liquor Evaporation and Storage
HE	Head End
LWR	Light Water Reactor
R&S	Receipt and Storage
SETP	Segregated Effluent Treatment Plant
SIXEP	Site Ion Exchange Plant
STP	Solvent Treatment Plant
tbd	to-be-defined
THORP	Thermal Oxide Reprocessing Plant
WEP	Waste Encapsulation Plant
WPEP	Waste Packaging and Encapsulation Plant
WTC	Waste Treatment Complex
WTPs	Waste Treatment Plants
WVP	Waste Vitrification Plant

Radiological impacts assessment

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
LNT	Linear Non-Threshold model
PAF	Potentially Affected Fraction of species
SED	Safety and Detriment Score
SLERA	Screening Level Ecological Risk Assessment

Organisations

IAEA	International Atomic Energy Agency
ICRP	International Committee on Radiological Protection
NCRP	National Council on Radiation Protection and Measurements
NDA	Nuclear Decommissioning Authority
NNL	National Nuclear Laboratory
NRPB	National Radiological Protection Board
RWM	Radioactive Waste Management Ltd.
SL	Sellafield Ltd.
US-EIA	U.S. Energy Information Administration
IEA	International Energy Agency

Others	
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LCT	Life Cycle Thinking
IEO	International Energy Outlook

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Appendix A

A.1 Characterisation factors

Table A.1 – UCrad characterisation factors for emissions into air, fresh water and sea water obtained for the default landscape

Nuclide	Emission to					
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Risk/Bq			*Bq U235 air-eq./Bq*		
Ac227	6.49E-20	2.97E-22	3.33E-24	1.40E+01	6.40E-02	7.17E-04
Ac228	8.71E-26	1.11E-26	1.37E-28	1.88E-05	2.38E-06	2.96E-08
Ag110m	2.42E-22	3.91E-22	2.07E-23	5.22E-02	8.42E-02	4.46E-03
Am241	1.13E-20	2.60E-22	4.15E-23	2.43E+00	5.59E-02	8.95E-03
As76	4.83E-26	1.33E-25	7.17E-27	1.04E-05	2.87E-05	1.54E-06
At211	4.29E-25	2.56E-26	8.70E-30	9.24E-05	5.52E-06	1.87E-09
Au198	3.26E-25	3.53E-25	1.38E-27	7.02E-05	7.61E-05	2.97E-07
Be10	1.17E-22	1.02E-23	1.01E-23	2.51E-02	2.19E-03	2.17E-03
Bi206	9.42E-24	2.18E-24	4.32E-27	2.03E-03	4.69E-04	9.30E-07
Bi210	4.11E-24	8.70E-26	5.45E-28	8.85E-04	1.87E-05	1.17E-07
Bi212	1.97E-26	1.94E-28	9.60E-31	4.25E-06	4.18E-08	2.07E-10
Br82	2.07E-24	9.89E-25	5.01E-26	4.46E-04	2.13E-04	1.08E-05
C14	4.33E-24	1.32E-25	1.03E-25	9.33E-04	2.85E-05	2.22E-05
Cd109	9.99E-23	6.21E-24	5.27E-24	2.15E-02	1.34E-03	1.13E-03
Ce141	2.09E-23	1.48E-24	1.15E-25	4.51E-03	3.20E-04	2.48E-05
Ce144	2.55E-22	3.01E-23	5.53E-24	5.49E-02	6.49E-03	1.19E-03
Cl36	1.01E-20	9.73E-21	9.53E-21	2.18E+00	2.10E+00	2.05E+00
Cm242	6.30E-22	1.62E-23	3.87E-26	1.36E-01	3.49E-03	8.32E-06
Cm244	6.52E-21	3.61E-22	1.15E-24	1.40E+00	7.78E-02	2.47E-04
Co58	3.90E-23	3.89E-23	3.59E-24	8.39E-03	8.37E-03	7.74E-04
Co60	8.62E-22	1.65E-21	3.93E-22	1.86E-01	3.54E-01	8.46E-02
Cr51	1.14E-24	4.00E-25	9.74E-27	2.46E-04	8.62E-05	2.10E-06
Cs134	1.18E-21	6.15E-22	7.38E-24	2.54E-01	1.33E-01	1.59E-03
Cs135	1.47E-22	2.73E-23	3.58E-23	3.17E-02	5.88E-03	7.71E-03
Cs136	4.67E-23	1.63E-23	3.21E-26	1.01E-02	3.51E-03	6.92E-06
Cs137	1.55E-21	7.96E-22	9.69E-24	3.35E-01	1.71E-01	2.09E-03
Cu64	5.29E-27	8.33E-27	1.80E-28	1.14E-06	1.79E-06	3.88E-08
Eu154	6.58E-22	4.53E-22	3.66E-22	1.42E-01	9.76E-02	7.87E-02
Eu155	3.21E-23	1.50E-23	1.16E-23	6.91E-03	3.23E-03	2.50E-03

Fe55	1.66E-23	4.04E-25	2.09E-27	3.57E-03	8.71E-05	4.50E-07
Fe59	6.81E-23	1.72E-23	3.00E-24	1.47E-02	3.70E-03	6.46E-04
Ga67	1.48E-25	3.09E-25	1.48E-26	3.19E-05	6.65E-05	3.19E-06
H3	2.03E-25	4.39E-25	3.25E-26	4.38E-05	9.47E-05	6.99E-06
Hg197	7.59E-26	1.16E-25	7.37E-26	1.63E-05	2.50E-05	1.59E-05
Hg197m	1.38E-26	8.07E-26	5.56E-26	2.97E-06	1.74E-05	1.20E-05
Hg203	7.39E-23	2.81E-23	3.39E-23	1.59E-02	6.05E-03	7.31E-03
I123	5.64E-27	1.17E-26	6.10E-29	1.21E-06	2.51E-06	1.31E-08
I125	1.18E-21	1.09E-21	5.57E-22	2.53E-01	2.36E-01	1.20E-01
I129	2.98E-19	2.96E-19	2.96E-19	6.42E+01	6.37E+01	6.37E+01
I131	2.22E-22	1.78E-22	2.75E-23	4.77E-02	3.84E-02	5.91E-03
I132	2.98E-27	3.46E-27	6.33E-30	6.42E-07	7.46E-07	1.36E-09
I133	1.77E-25	3.35E-25	2.86E-27	3.82E-05	7.21E-05	6.16E-07
I134	8.63E-28	5.22E-28	7.40E-31	1.86E-07	1.12E-07	1.59E-10
I135	1.17E-26	2.79E-26	6.78E-29	2.52E-06	6.01E-06	1.46E-08
In111	2.14E-25	2.28E-24	2.71E-27	4.61E-05	4.90E-04	5.83E-07
In113m	2.36E-28	9.20E-27	3.78E-30	5.08E-08	1.98E-06	8.13E-10
Kr85	1.20E-26	1.21E-26	1.17E-26	2.58E-06	2.61E-06	2.53E-06
Mn54	7.38E-23	1.47E-22	2.77E-23	1.59E-02	3.16E-02	5.96E-03
Mo99	2.56E-25	1.48E-25	9.98E-28	5.51E-05	3.19E-05	2.15E-07
Na22	5.25E-22	8.77E-22	2.13E-26	1.13E-01	1.89E-01	4.59E-06
Na24	1.28E-25	1.32E-25	9.40E-30	2.77E-05	2.84E-05	2.02E-09
Nb95	2.11E-23	1.79E-24	1.15E-24	4.55E-03	3.86E-04	2.48E-04
Ni59	4.22E-24	6.93E-25	1.35E-24	9.09E-04	1.49E-04	2.91E-04
Ni63	7.82E-24	2.38E-25	2.39E-25	1.68E-03	5.13E-05	5.14E-05
Np237	6.86E-21	8.01E-22	5.74E-22	1.48E+00	1.73E-01	1.24E-01
Np239	1.32E-25	3.11E-26	2.33E-29	2.85E-05	6.69E-06	5.01E-09
P32	1.01E-22	2.95E-21	4.02E-25	2.18E-02	6.36E-01	8.65E-05
Pa231	4.04E-20	1.38E-21	1.22E-22	8.71E+00	2.97E-01	2.62E-02
Pa233	2.33E-23	3.66E-25	2.22E-25	5.01E-03	7.88E-05	4.78E-05
Pb210	3.50E-20	1.74E-21	9.32E-23	7.54E+00	3.74E-01	2.01E-02
Pd103	3.42E-24	1.02E-25	3.85E-27	7.37E-04	2.20E-05	8.30E-07
Pd107	2.59E-24	5.08E-25	1.00E-24	5.57E-04	1.09E-04	2.15E-04
Pd109	3.25E-27	7.82E-27	3.69E-28	6.99E-07	1.68E-06	7.95E-08
Pm147	1.30E-23	5.64E-25	6.12E-27	2.80E-03	1.21E-04	1.32E-06
Po210	1.47E-19	5.44E-22	4.43E-24	3.17E+01	1.17E-01	9.55E-04
Pu238	1.48E-20	1.62E-20	1.90E-23	3.19E+00	3.50E+00	4.09E-03
Pu239	2.48E-20	1.89E-20	1.01E-22	5.34E+00	4.07E+00	2.18E-02
Pu240	2.45E-20	1.89E-20	8.90E-23	5.27E+00	4.06E+00	1.92E-02
Pu241	2.70E-22	2.72E-22	4.67E-25	5.81E-02	5.87E-02	1.01E-04
Pu242	2.39E-20	1.81E-20	1.03E-22	5.15E+00	3.91E+00	2.23E-02
Ra224	9.56E-23	8.83E-24	9.62E-26	2.06E-02	1.90E-03	2.07E-05
Ra225	1.65E-21	3.51E-23	5.65E-25	3.56E-01	7.57E-03	1.22E-04
Ra226	5.86E-20	7.82E-21	8.08E-22	1.26E+01	1.68E+00	1.74E-01

Rb86	6.28E-23	1.19E-23	2.92E-27	1.35E-02	2.57E-03	6.29E-07
Rh105	1.73E-26	2.08E-26	2.42E-28	3.72E-06	4.49E-06	5.21E-08
Rh107	3.70E-29	1.35E-29	1.16E-31	7.97E-09	2.91E-09	2.50E-11
Rn222	3.75E-30	3.49E-30	2.34E-31	8.07E-10	7.51E-10	5.04E-11
Ru103	2.60E-23	1.01E-23	2.23E-25	5.59E-03	2.18E-03	4.80E-05
Ru106	3.58E-22	7.21E-23	4.91E-24	7.70E-02	1.55E-02	1.06E-03
S35	7.08E-23	1.62E-24	2.06E-28	1.53E-02	3.50E-04	4.45E-08
Sb124	1.08E-22	3.71E-23	4.28E-25	2.33E-02	7.98E-03	9.22E-05
Sb125	1.17E-22	1.25E-22	1.49E-24	2.53E-02	2.68E-02	3.21E-04
Se75	1.34E-22	8.67E-23	8.61E-24	2.89E-02	1.87E-02	1.85E-03
Se79	5.21E-21	4.52E-21	6.10E-21	1.12E+00	9.73E-01	1.31E+00
Sn113	3.71E-23	1.94E-23	5.90E-24	8.00E-03	4.17E-03	1.27E-03
Sn126	1.60E-21	1.89E-22	4.19E-22	3.44E-01	4.07E-02	9.02E-02
Sr85	2.62E-23	5.58E-24	1.76E-27	5.64E-03	1.20E-03	3.79E-07
Sr87m	6.10E-28	1.73E-28	6.96E-32	1.31E-07	3.73E-08	1.50E-11
Sr89	9.34E-23	3.32E-24	1.50E-27	2.01E-02	7.15E-04	3.24E-07
Sr90	1.60E-21	1.47E-22	1.24E-25	3.44E-01	3.16E-02	2.66E-05
Tc99	2.71E-22	1.82E-22	1.47E-22	5.83E-02	3.92E-02	3.16E-02
Tc99m	8.18E-28	7.40E-29	1.87E-30	1.76E-07	1.59E-08	4.03E-10
Te125m	3.26E-23	2.51E-24	1.69E-25	7.03E-03	5.40E-04	3.65E-05
Te127m	9.95E-23	9.26E-24	7.18E-25	2.14E-02	2.00E-03	1.55E-04
Te129m	8.93E-23	5.90E-24	3.65E-25	1.92E-02	1.27E-03	7.86E-05
Te131m	1.56E-25	1.91E-25	9.73E-27	3.36E-05	4.11E-05	2.10E-06
Te132	2.73E-24	9.86E-25	5.06E-26	5.87E-04	2.12E-04	1.09E-05
Th228	4.26E-21	5.46E-22	2.55E-22	9.18E-01	1.18E-01	5.48E-02
Th229	3.75E-20	1.18E-21	7.81E-21	8.09E+00	2.54E-01	1.68E+00
Th230	1.80E-19	8.14E-21	8.83E-21	3.87E+01	1.75E+00	1.90E+00
Th232	2.48E-19	1.12E-20	1.12E-20	5.34E+01	2.41E+00	2.42E+00
Tl201	7.63E-26	5.32E-26	1.09E-26	1.64E-05	1.15E-05	2.34E-06
Tl202	6.52E-24	1.94E-24	2.08E-25	1.41E-03	4.18E-04	4.49E-05
U232	2.16E-20	1.21E-21	4.88E-24	4.66E+00	2.60E-01	1.05E-03
U233	2.98E-21	1.30E-22	4.63E-23	6.42E-01	2.79E-02	9.96E-03
U234	2.21E-20	6.25E-21	2.58E-21	4.77E+00	1.35E+00	5.55E-01
U235	4.64E-21	8.03E-22	3.93E-22	1.00E+00	1.73E-01	8.46E-02
U236	2.76E-21	1.42E-22	6.64E-23	5.95E-01	3.05E-02	1.43E-02
U238	2.40E-20	7.62E-21	3.76E-21	5.16E+00	1.64E+00	8.10E-01
Xe133	3.25E-28	3.82E-28	3.18E-29	7.00E-08	8.22E-08	6.85E-09
Y87	5.59E-25	7.14E-26	1.56E-26	1.20E-04	1.54E-05	3.36E-06
Y90	5.91E-25	1.17E-25	1.98E-27	1.27E-04	2.51E-05	4.27E-07
Y91	9.23E-23	1.82E-24	2.71E-25	1.99E-02	3.92E-04	5.84E-05
Zn65	2.35E-22	1.91E-22	8.96E-24	5.06E-02	4.11E-02	1.93E-03
Zr95	4.97E-23	1.51E-23	6.40E-24	1.07E-02	3.25E-03	1.38E-03

Table A.2 – CGM characterisation factors for emissions into air, fresh water and sea water obtained for a distance of 1 km

Nuclide	Emission to, at 1 km distance					
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Risk/Bq			*Bq U235 air-eq./Bq*		
Ac228	2.37E-19	9.35E-21	1.85E-20	2.24E-04	8.87E-06	1.76E-05
Ag110m	1.08E-16	2.18E-17	4.88E-18	1.03E-01	2.07E-02	4.63E-03
Am241	3.95E-15	3.25E-18	3.71E-18	3.75E+00	3.09E-03	3.52E-03
As76	5.93E-20	9.26E-21	2.30E-19	5.63E-05	8.78E-06	2.18E-04
At211	1.03E-18	2.51E-20	2.48E-21	9.74E-04	2.38E-05	2.36E-06
Au198	1.84E-19	6.00E-20	1.64E-20	1.75E-04	5.70E-05	1.56E-05
Bi206	4.17E-18	6.21E-20	1.09E-20	3.96E-03	5.89E-05	1.03E-05
Bi210	1.86E-18	3.58E-21	4.00E-21	1.77E-03	3.40E-06	3.79E-06
Bi212	2.52E-19	6.37E-22	1.21E-22	2.39E-04	6.04E-07	1.15E-07
Br82	2.64E-18	1.64E-20	2.70E-22	2.50E-03	1.56E-05	2.56E-07
C14	5.68E-19	-	-	5.39E-04	-	-
Cd109	3.64E-17	2.03E-19	1.56E-18	3.45E-02	1.92E-04	1.48E-03
Ce141	7.07E-18	2.63E-19	2.48E-19	6.70E-03	2.50E-04	2.36E-04
Ce144	9.08E-17	3.22E-18	3.02E-18	8.61E-02	3.05E-03	2.86E-03
Cm242	2.33E-16	4.44E-20	1.97E-19	2.21E-01	4.21E-05	1.87E-04
Cm244	2.37E-15	4.40E-19	1.87E-18	2.24E+00	4.17E-04	1.77E-03
Co58	1.55E-17	1.63E-18	2.06E-18	1.47E-02	1.55E-03	1.96E-03
Co60	4.34E-16	1.39E-17	1.74E-17	4.11E-01	1.32E-02	1.65E-02
Cr51	4.33E-19	9.99E-21	6.53E-21	4.10E-04	9.48E-06	6.19E-06
Cs134	4.88E-16	5.85E-18	4.68E-19	4.63E-01	5.55E-03	4.43E-04
Cs135	4.25E-17	3.45E-20	3.09E-20	4.03E-02	3.27E-05	2.93E-05
Cs136	1.82E-17	5.12E-19	6.05E-20	1.72E-02	4.85E-04	5.74E-05
Cs137	4.20E-16	2.59E-18	2.75E-19	3.98E-01	2.45E-03	2.61E-04
Cu64	1.07E-20	3.20E-21	1.13E-20	1.02E-05	3.03E-06	1.07E-05
Eu154	3.27E-16	9.17E-20	2.58E-17	3.10E-01	8.70E-05	2.44E-02
Eu155	1.47E-17	5.12E-21	1.26E-18	1.39E-02	4.86E-06	1.19E-03
Fe55	6.08E-18	1.01E-21	1.53E-18	5.77E-03	9.57E-07	1.45E-03
Fe59	2.54E-17	1.54E-19	1.49E-17	2.41E-02	1.46E-04	1.42E-02
Ga67	7.84E-20	1.16E-20	2.04E-19	7.43E-05	1.10E-05	1.93E-04
H3	7.92E-21	1.14E-22	6.08E-22	7.51E-06	1.08E-07	5.77E-07
Hg197	4.10E-20	2.28E-20	1.04E-18	3.89E-05	2.17E-05	9.83E-04
Hg197m	1.49E-20	1.96E-20	2.01E-18	1.42E-05	1.86E-05	1.90E-03
Hg203	2.37E-17	1.11E-18	8.79E-18	2.24E-02	1.05E-03	8.34E-03
I123	1.30E-20	1.37E-21	2.53E-22	1.23E-05	1.30E-06	2.40E-07
I125	2.16E-16	9.35E-20	2.08E-20	2.05E-01	8.87E-05	1.97E-05
I129	2.28E-15	6.57E-19	1.53E-19	2.17E+00	6.23E-04	1.45E-04
I131	6.02E-17	1.34E-19	3.03E-20	5.71E-02	1.27E-04	2.87E-05
I132	2.47E-20	1.95E-21	1.75E-22	2.34E-05	1.85E-06	1.66E-07
I133	2.79E-19	2.49E-20	5.45E-21	2.64E-04	2.37E-05	5.17E-06

I134	1.44E-20	6.79E-22	1.71E-23	1.37E-05	6.44E-07	1.62E-08
I135	4.46E-20	5.90E-21	9.72E-22	4.23E-05	5.60E-06	9.22E-07
In111	1.25E-19	2.39E-19	2.83E-20	1.18E-04	2.27E-04	2.69E-05
In113m	2.23E-21	1.36E-20	7.12E-22	2.11E-06	1.29E-05	6.76E-07
Kr85	1.47E-23	6.38E-26	3.41E-45	1.39E-08	6.05E-11	3.23E-30
Mn54	3.33E-17	6.15E-18	1.26E-17	3.16E-02	5.83E-03	1.19E-02
Mo99	1.60E-19	1.29E-20	4.96E-21	1.52E-04	1.23E-05	4.70E-06
Na22	2.43E-16	7.82E-18	5.01E-22	2.30E-01	7.42E-03	4.75E-07
Na24	2.40E-19	4.04E-20	5.85E-23	2.28E-04	3.83E-05	5.55E-08
Nb95	8.05E-18	3.69E-21	1.55E-18	7.63E-03	3.50E-06	1.47E-03
Ni59	1.22E-18	1.71E-22	9.72E-21	1.16E-03	1.62E-07	9.22E-06
Ni63	2.90E-18	4.05E-22	2.32E-20	2.75E-03	3.84E-07	2.20E-05
Np237	2.31E-15	2.62E-19	2.46E-20	2.19E+00	2.49E-04	2.33E-05
Np239	7.67E-20	1.90E-21	1.69E-22	7.28E-05	1.80E-06	1.60E-07
P32	2.12E-17	1.46E-17	1.11E-18	2.01E-02	1.38E-02	1.05E-03
Pa231	1.40E-14	1.61E-18	6.62E-18	1.33E+01	1.53E-03	6.28E-03
Pa233	7.92E-18	2.14E-21	5.86E-19	7.51E-03	2.03E-06	5.56E-04
Pb210	1.24E-14	2.82E-18	2.15E-17	1.17E+01	2.68E-03	2.03E-02
Pd103	1.13E-18	1.77E-21	8.93E-21	1.07E-03	1.68E-06	8.47E-06
Pd107	7.64E-19	1.01E-22	1.71E-21	7.24E-04	9.57E-08	1.63E-06
Pd109	5.33E-21	1.60E-21	2.21E-20	5.05E-06	1.52E-06	2.10E-05
Pm147	4.62E-18	9.81E-22	1.27E-20	4.38E-03	9.30E-07	1.21E-05
Po210	1.72E-13	3.58E-17	3.70E-16	1.63E+02	3.39E-02	3.51E-01
Pu238	4.49E-15	2.09E-16	3.56E-18	4.26E+00	1.98E-01	3.37E-03
Pu239	4.89E-15	2.27E-16	3.86E-18	4.63E+00	2.16E-01	3.66E-03
Pu240	4.89E-15	2.27E-16	3.86E-18	4.63E+00	2.16E-01	3.66E-03
Pu241	9.35E-17	4.39E-18	7.69E-20	8.87E-02	4.17E-03	7.30E-05
Pu242	4.69E-15	2.18E-16	3.70E-18	4.45E+00	2.07E-01	3.51E-03
Ra224	4.95E-17	2.35E-19	9.81E-19	4.70E-02	2.23E-04	9.30E-04
Ra225	5.71E-16	3.41E-19	1.52E-18	5.42E-01	3.23E-04	1.44E-03
Ra226	6.31E-15	2.89E-18	4.45E-18	5.98E+00	2.74E-03	4.22E-03
Rb86	2.47E-17	1.43E-19	2.10E-21	2.34E-02	1.36E-04	1.99E-06
Rh105	1.78E-20	3.75E-21	3.88E-21	1.69E-05	3.56E-06	3.68E-06
Rh107	1.14E-21	1.14E-22	9.63E-25	1.08E-06	1.08E-07	9.13E-10
Rn222	1.11E-24	2.70E-30	1.94E-48	1.05E-09	2.56E-15	1.84E-33
Ru103	9.44E-18	3.44E-19	9.44E-20	8.96E-03	3.26E-04	8.96E-05
Ru106	1.30E-16	1.19E-18	3.24E-19	1.23E-01	1.13E-03	3.07E-04
S35	7.34E-17	1.60E-20	1.19E-22	6.97E-02	1.51E-05	1.13E-07
Sb124	4.07E-17	3.14E-19	2.59E-19	3.86E-02	2.98E-04	2.45E-04
Sb125	5.67E-17	2.93E-19	1.28E-19	5.37E-02	2.78E-04	1.22E-04
Se75	5.94E-17	1.89E-19	4.02E-18	5.63E-02	1.79E-04	3.82E-03
Sn113	1.41E-17	1.24E-18	5.93E-17	1.34E-02	1.17E-03	5.63E-02
Sr85	1.03E-17	2.45E-20	2.92E-22	9.74E-03	2.32E-05	2.77E-07
Sr87m	4.02E-21	1.13E-22	7.00E-24	3.81E-06	1.07E-07	6.64E-09

Sr89	3.23E-17	1.09E-20	1.21E-21	3.06E-02	1.03E-05	1.15E-06
Sr90	5.57E-16	1.11E-19	1.30E-20	5.29E-01	1.05E-04	1.23E-05
Tc99	4.30E-17	1.49E-21	7.90E-21	4.08E-02	1.41E-06	7.50E-06
Tc99m	2.99E-21	4.79E-23	1.98E-22	2.83E-06	4.55E-08	1.88E-07
Te125m	1.14E-17	3.80E-21	1.35E-19	1.08E-02	3.61E-06	1.28E-04
Te127m	3.51E-17	9.99E-21	3.55E-19	3.33E-02	9.48E-06	3.37E-04
Te129m	3.03E-17	1.30E-20	4.62E-19	2.88E-02	1.23E-05	4.38E-04
Te131m	1.85E-19	8.06E-21	2.75E-19	1.76E-04	7.64E-06	2.61E-04
Te132	1.46E-18	1.63E-20	5.74E-19	1.38E-03	1.55E-05	5.44E-04
Th228	1.60E-15	2.99E-17	1.15E-15	1.52E+00	2.83E-02	1.09E+00
Th230	5.25E-15	4.35E-17	3.29E-15	4.98E+00	4.13E-02	3.12E+00
Th232	6.05E-15	5.94E-17	3.61E-15	5.74E+00	5.63E-02	3.43E+00
Tl201	5.10E-20	2.47E-21	1.44E-19	4.83E-05	2.34E-06	1.37E-04
Tl202	2.55E-18	4.47E-20	7.05E-19	2.42E-03	4.24E-05	6.69E-04
U232	7.26E-15	7.97E-19	9.99E-20	6.89E+00	7.56E-04	9.48E-05
U234	2.14E-15	1.30E-19	6.75E-20	2.03E+00	1.23E-04	6.40E-05
U235	1.05E-15	1.13E-19	1.28E-20	1.00E+00	1.07E-04	1.22E-05
U238	2.11E-15	1.21E-19	7.00E-20	2.00E+00	1.15E-04	6.64E-05
Xe133	8.00E-23	4.33E-28	3.14E-46	7.59E-08	4.10E-13	2.97E-31
Y87	3.34E-19	2.81E-21	1.63E-19	3.17E-04	2.66E-06	1.55E-04
Y90	2.67E-19	6.59E-21	5.83E-20	2.53E-04	6.25E-06	5.53E-05
Y91	3.56E-17	9.53E-21	3.10E-19	3.37E-02	9.04E-06	2.94E-04
Zn65	1.16E-16	1.30E-19	1.97E-18	1.10E-01	1.23E-04	1.87E-03
Zr95	2.03E-17	5.76E-20	8.34E-18	1.92E-02	5.46E-05	7.91E-03

Table A.3 – CGM characterisation factors for emissions into air, fresh water and sea water obtained for a distance of 2.5 km

Nuclide	Emission to		Nuclide	Emission to	
	Air	Coastal water		Air	Coastal water
	Risk/Bq			*Risk/Bq*	
Ac228	9.44E-20	9.01E-20	**Pa231**	5.75E-15	1.22E-17
Ag110m	4.41E-17	6.58E-18	**Pa233**	3.23E-18	5.15E-18
Am241	1.61E-15	6.72E-18	**Pb210**	5.05E-15	8.96E-18
As76	2.41E-20	7.80E-20	**Pd103**	4.61E-19	4.74E-21
At211	4.10E-19	7.42E-21	**Pd107**	3.12E-19	6.40E-22
Au198	7.51E-20	1.73E-20	**Pd109**	2.15E-21	6.75E-21
Bi206	1.70E-18	4.68E-20	**Pm147**	1.89E-18	1.05E-20
Bi210	7.60E-19	1.83E-21	**Po210**	7.04E-14	1.38E-16
Bi212	8.94E-20	3.63E-24	**Pu238**	1.84E-15	1.35E-18
Br82	1.07E-18	3.52E-22	**Pu239**	2.00E-15	1.45E-18
C14	2.32E-19	0.00E+00	**Pu240**	2.00E-15	1.47E-18
Cd109	1.49E-17	7.32E-19	**Pu241**	3.81E-17	5.27E-20
Ce141	2.89E-18	2.16E-18	**Pu242**	1.92E-15	1.40E-18
Ce144	3.71E-17	2.66E-17	**Ra224**	2.02E-17	3.67E-19
Cm242	9.51E-17	1.74E-19	**Ra225**	2.33E-16	5.73E-19
Cm244	9.66E-16	8.76E-19	**Ra226**	2.58E-15	2.68E-18
Co58	6.34E-18	1.77E-17	**Rb86**	1.01E-17	1.49E-20
Co60	1.77E-16	1.52E-16	**Rh105**	7.22E-21	2.77E-20
Cr51	1.77E-19	4.81E-20	**Rh107**	3.11E-22	2.40E-27
Cs134	1.99E-16	1.67E-18	**Rn222**	4.51E-25	1.68E-47
Cs135	1.74E-17	1.16E-20	**Ru103**	3.85E-18	8.37E-19
Cs136	7.41E-18	1.45E-19	**Ru106**	5.32E-17	2.87E-18
Cs137	1.72E-16	7.41E-19	**S35**	3.00E-17	4.43E-23
Cu64	4.32E-21	4.11E-21	**Sb124**	1.66E-17	3.32E-19
Eu154	1.34E-16	2.29E-16	**Sb125**	2.31E-17	2.70E-19
Eu155	6.00E-18	1.11E-17	**Se75**	2.43E-17	1.64E-18
Fe55	2.48E-18	5.71E-19	**Sn113**	5.77E-18	4.77E-17
Fe59	1.04E-17	6.20E-17	**Sr85**	4.18E-18	3.97E-22
Ga67	3.20E-20	9.81E-20	**Sr87m**	1.56E-21	9.91E-25
H3	3.24E-21	2.27E-22	**Sr89**	1.32E-17	4.81E-22
Hg197	1.67E-20	3.70E-19	**Sr90**	2.28E-16	5.10E-21
Hg197m	6.05E-21	6.64E-19	**Tc99**	1.76E-17	2.95E-21
Hg203	9.67E-18	3.32E-18	**Tc99m**	1.19E-21	4.72E-23
I123	5.26E-21	8.09E-23	**Te125m**	4.65E-18	5.25E-20
I125	8.82E-17	7.97E-21	**Te127m**	1.43E-17	1.35E-19
I129	9.33E-16	5.76E-20	**Te129m**	1.24E-17	1.74E-19
I131	2.46E-17	1.14E-20	**Te131m**	7.53E-20	9.52E-20
I132	9.46E-21	2.07E-23	**Te132**	5.96E-19	2.15E-19
I133	1.13E-19	1.81E-21	**Th228**	6.57E-16	6.93E-16

I134	4.98E-21	2.65E-25	**Th230**	2.15E-15	1.61E-15
I135	1.78E-20	2.50E-22	**Th232**	2.47E-15	1.88E-15
In111	5.09E-20	6.37E-20	**Tl201**	2.08E-20	5.71E-20
In113m	8.35E-22	9.89E-23	**Tl202**	1.04E-18	3.86E-19
Kr85	6.00E-24	3.05E-44	**U232**	2.96E-15	4.58E-19
Mn54	1.36E-17	1.12E-16	**U234**	8.71E-16	5.37E-19
Mo99	6.54E-20	4.16E-20	**U235**	4.30E-16	5.24E-20
Na22	9.92E-17	2.40E-22	**U238**	8.60E-16	5.65E-19
Na24	9.73E-20	1.82E-23	**Xe133**	3.26E-23	2.74E-45
Nb95	3.29E-18	1.38E-17	**Y87**	1.36E-19	1.33E-18
Ni59	5.00E-19	3.63E-21	**Y90**	1.09E-19	1.67E-19
Ni63	1.18E-18	8.65E-21	**Y91**	1.45E-17	2.45E-18
Np237	9.45E-16	7.39E-20	**Zn65**	4.76E-17	7.33E-18
Np239	3.13E-20	4.61E-22	**Zr95**	8.28E-18	7.31E-17
P32	8.63E-18	4.09E-19			

Table A.4 – CGM characterisation factors for emissions into air, fresh water and sea water obtained for a distance of 100 km

Nuclide	Emission to, at 100 km distance					
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Risk/Bq			*Bq U235 air-eq./Bq*		
Ac228	2.79E-23	5.18E-22	5.60E-35	4.75E-05	8.83E-04	9.55E-17
Ag110m	6.01E-20	2.18E-17	7.83E-20	1.03E-01	3.72E+01	1.33E-01
Am241	2.20E-18	3.25E-18	8.25E-20	3.75E+00	5.55E+00	1.41E-01
As76	2.30E-23	4.71E-21	7.75E-25	3.92E-05	8.03E-03	1.32E-06
At211	1.52E-22	2.15E-21	4.50E-34	2.59E-04	3.66E-03	7.67E-16
Au198	8.86E-23	4.57E-20	1.16E-23	1.51E-04	7.78E-02	1.98E-05
Bi206	2.18E-21	5.51E-20	1.63E-22	3.72E-03	9.39E-02	2.78E-04
Bi210	9.63E-22	3.09E-21	4.72E-24	1.64E-03	5.27E-03	8.05E-06
Bi212	1.10E-26	1.47E-29	5.92E-107	1.88E-08	2.50E-11	1.01E-88
Br82	1.12E-21	9.90E-21	2.10E-26	1.91E-03	1.69E-02	3.58E-08
C14	3.17E-22	-	-	5.41E-04	-	-
Cd109	2.03E-20	2.02E-19	8.82E-21	3.45E-02	3.44E-01	1.50E-02
Ce141	3.89E-21	2.58E-19	2.09E-20	6.63E-03	4.39E-01	3.56E-02
Ce144	5.05E-20	3.21E-18	3.17E-19	8.61E-02	5.47E+00	5.41E-01
Cm242	1.29E-19	4.41E-20	2.04E-21	2.20E-01	7.52E-02	3.47E-03
Cm244	1.32E-18	4.40E-19	1.07E-20	2.25E+00	7.50E-01	1.83E-02
Co58	8.60E-21	1.61E-18	1.95E-19	1.47E-02	2.75E+00	3.33E-01
Co60	2.42E-19	1.39E-17	1.86E-18	4.13E-01	2.38E+01	3.17E+00
Cr51	2.37E-22	9.81E-21	4.45E-22	4.05E-04	1.67E-02	7.58E-04
Cs134	2.71E-19	5.84E-18	2.03E-20	4.63E-01	9.95E+00	3.45E-02
Cs135	2.37E-20	3.45E-20	1.42E-22	4.05E-02	5.88E-02	2.42E-04
Cs136	9.81E-21	4.83E-19	9.81E-22	1.67E-02	8.23E-01	1.67E-03
Cs137	2.34E-19	2.59E-18	9.08E-21	3.98E-01	4.41E+00	1.55E-02
Cu64	2.81E-24	7.88E-22	1.84E-29	4.78E-06	1.34E-03	3.14E-11
Eu154	1.82E-19	9.17E-20	2.81E-18	3.11E-01	1.56E-01	4.78E+00
Eu155	8.18E-21	5.12E-21	1.36E-19	1.39E-02	8.73E-03	2.31E-01
Fe55	3.38E-21	1.01E-21	6.93E-21	5.77E-03	1.72E-03	1.18E-02
Fe59	1.40E-20	1.51E-19	6.40E-19	2.39E-02	2.58E-01	1.09E+00
Ga67	3.87E-23	9.26E-21	1.09E-22	6.59E-05	1.58E-02	1.86E-04
H3	4.41E-24	1.13E-22	2.77E-24	7.52E-06	1.92E-04	4.72E-06
Hg197	1.96E-23	1.72E-20	2.40E-22	3.34E-05	2.94E-02	4.09E-04
Hg197m	5.56E-24	9.35E-21	3.03E-24	9.48E-06	1.59E-02	5.16E-06
Hg203	1.31E-20	1.09E-18	3.44E-20	2.23E-02	1.86E+00	5.86E-02
I123	3.52E-24	3.56E-22	6.51E-31	6.00E-06	6.06E-04	1.11E-12
I125	1.19E-19	9.26E-20	8.56E-23	2.03E-01	1.58E-01	1.46E-04
I129	1.27E-18	6.57E-19	7.05E-22	2.17E+00	1.12E+00	1.20E-03
I131	3.19E-20	1.22E-19	5.26E-23	5.44E-02	2.08E-01	8.97E-05
I132	2.17E-25	8.68E-25	8.32E-61	3.70E-07	1.48E-06	1.42E-42
I133	9.81E-23	1.06E-20	2.65E-27	1.67E-04	1.81E-02	4.52E-09

I134	1.49E-28	1.08E-30	2.26E-120	2.55E-10	1.84E-12	3.86E-102
I135	5.89E-24	4.05E-22	1.46E-36	1.00E-05	6.91E-04	2.48E-18
In111	6.04E-23	1.84E-19	4.90E-23	1.03E-04	3.14E-01	8.36E-05
In113m	3.99E-27	3.11E-25	9.26E-74	6.80E-09	5.30E-07	1.58E-55
Kr85	8.18E-27	6.37E-26	3.73E-46	1.39E-08	1.09E-07	6.36E-28
Mn54	1.85E-20	6.13E-18	1.34E-18	3.16E-02	1.05E+01	2.28E+00
Mo99	7.74E-23	9.90E-21	2.97E-23	1.32E-04	1.69E-02	5.06E-05
Na22	1.35E-19	7.81E-18	2.92E-24	2.30E-01	1.33E+01	4.97E-06
Na24	7.11E-23	1.25E-20	8.47E-31	1.21E-04	2.13E-02	1.44E-12
Nb95	4.44E-21	3.61E-21	1.36E-19	7.56E-03	6.16E-03	2.31E-01
Ni59	6.82E-22	1.71E-22	4.45E-23	1.16E-03	2.91E-04	7.58E-05
Ni63	1.61E-21	4.05E-22	1.05E-22	2.75E-03	6.91E-04	1.80E-04
Np237	1.29E-18	2.62E-19	9.08E-22	2.20E+00	4.47E-01	1.55E-03
Np239	3.61E-23	1.38E-21	2.05E-25	6.16E-05	2.36E-03	3.50E-07
P32	1.15E-20	1.38E-17	2.90E-21	1.95E-02	2.36E+01	4.94E-03
Pa231	7.84E-18	1.61E-18	1.50E-19	1.34E+01	2.75E+00	2.56E-01
Pa233	4.35E-21	2.08E-21	4.74E-20	7.41E-03	3.55E-03	8.08E-02
Pb210	6.89E-18	2.82E-18	1.10E-19	1.18E+01	4.81E+00	1.88E-01
Pd103	6.14E-22	1.70E-21	3.67E-23	1.05E-03	2.89E-03	6.25E-05
Pd107	4.25E-22	1.01E-22	7.84E-24	7.25E-04	1.72E-04	1.34E-05
Pd109	1.46E-24	4.25E-22	6.60E-29	2.48E-06	7.25E-04	1.13E-10
Pm147	2.58E-21	9.81E-22	1.28E-22	4.39E-03	1.67E-03	2.19E-04
Po210	9.53E-17	3.56E-17	1.60E-18	1.63E+02	6.06E+01	2.72E+00
Pu238	2.50E-18	2.09E-16	1.65E-20	4.27E+00	3.56E+02	2.81E-02
Pu239	2.72E-18	2.27E-16	1.78E-20	4.64E+00	3.88E+02	3.03E-02
Pu240	2.72E-18	2.27E-16	1.80E-20	4.64E+00	3.88E+02	3.06E-02
Pu241	5.20E-20	4.39E-18	6.45E-22	8.86E-02	7.48E+00	1.10E-03
Pu242	2.61E-18	2.18E-16	1.71E-20	4.45E+00	3.72E+02	2.92E-02
Ra224	2.48E-20	1.92E-19	5.31E-22	4.22E-02	3.27E-01	9.05E-04
Ra225	3.10E-19	3.25E-19	4.13E-21	5.28E-01	5.53E-01	7.05E-03
Ra226	3.51E-18	2.89E-18	3.29E-20	5.98E+00	4.92E+00	5.61E-02
Rb86	1.35E-20	1.38E-19	1.21E-22	2.30E-02	2.34E-01	2.06E-04
Rh105	7.55E-24	2.27E-21	1.66E-24	1.29E-05	3.88E-03	2.83E-06
Rh107	2.31E-36	6.02E-44	1.60E-254	3.94E-18	1.03E-25	2.72E-236
Rn222	5.55E-28	2.23E-30	2.67E-50	9.45E-10	3.80E-12	4.55E-32
Ru103	5.20E-21	3.37E-19	8.43E-21	8.86E-03	5.75E-01	1.44E-02
Ru106	7.25E-20	1.19E-18	3.46E-20	1.24E-01	2.03E+00	5.89E-02
S35	4.07E-20	1.58E-20	4.96E-25	6.94E-02	2.69E-02	8.45E-07
Sb124	2.26E-20	3.11E-19	3.58E-21	3.84E-02	5.30E-01	6.11E-03
Sb125	3.15E-20	2.92E-19	3.29E-21	5.38E-02	4.98E-01	5.61E-03
Se75	3.30E-20	1.88E-19	1.88E-20	5.63E-02	3.20E-01	3.20E-02
Sn113	7.85E-21	1.23E-18	5.46E-19	1.34E-02	2.09E+00	9.31E-01
Sr85	5.67E-21	2.42E-20	4.32E-24	9.66E-03	4.13E-02	7.36E-06
Sr87m	7.42E-26	2.00E-25	8.95E-56	1.26E-07	3.41E-07	1.53E-37

Sr89	1.78E-20	1.07E-20	5.04E-24	3.03E-02	1.83E-02	8.59E-06
Sr90	3.11E-19	1.11E-19	6.24E-23	5.30E-01	1.89E-01	1.06E-04
Tc99	2.40E-20	1.49E-21	3.61E-23	4.09E-02	2.53E-03	6.16E-05
Tc99m	3.42E-25	2.52E-24	1.63E-38	5.83E-07	4.30E-06	2.78E-20
Te125m	6.30E-21	3.75E-21	5.62E-22	1.07E-02	6.39E-03	9.58E-04
Te127m	1.94E-20	9.90E-21	1.54E-21	3.31E-02	1.69E-02	2.63E-03
Te129m	1.67E-20	1.27E-20	1.69E-21	2.84E-02	2.16E-02	2.88E-03
Te131m	7.51E-23	4.46E-21	2.23E-24	1.28E-04	7.61E-03	3.80E-06
Te132	7.21E-22	1.30E-20	2.39E-22	1.23E-03	2.22E-02	4.08E-04
Th228	8.96E-19	2.99E-17	8.40E-18	1.53E+00	5.09E+01	1.43E+01
Th230	2.92E-18	4.35E-17	1.97E-17	4.98E+00	7.42E+01	3.36E+01
Th232	3.37E-18	5.94E-17	2.30E-17	5.75E+00	1.01E+02	3.92E+01
Tl201	2.49E-23	1.93E-21	5.33E-23	4.25E-05	3.30E-03	9.08E-05
Tl202	1.38E-21	4.22E-20	2.49E-21	2.34E-03	7.19E-02	4.25E-03
U232	4.04E-18	7.97E-19	5.62E-21	6.89E+00	1.36E+00	9.58E-03
U234	1.19E-18	1.30E-19	6.60E-21	2.03E+00	2.22E-01	1.13E-02
U235	5.87E-19	1.13E-19	6.44E-22	1.00E+00	1.92E-01	1.10E-03
U238	1.17E-18	1.21E-19	6.95E-21	2.00E+00	2.06E-01	1.18E-02
Xe133	4.13E-26	3.76E-28	7.56E-48	7.05E-08	6.41E-10	1.29E-29
Y87	1.65E-22	2.25E-21	1.58E-21	2.81E-04	3.83E-03	2.69E-03
Y90	1.28E-22	5.00E-21	1.09E-22	2.19E-04	8.52E-03	1.86E-04
Y91	1.97E-20	9.44E-21	2.63E-20	3.36E-02	1.61E-02	4.48E-02
Zn65	6.49E-20	1.30E-19	8.72E-20	1.11E-01	2.22E-01	1.49E-01
Zr95	1.12E-20	5.69E-20	7.95E-19	1.91E-02	9.70E-02	1.35E+00

Table A.5 – CGM characterisation factors for emissions into air, fresh water and sea water obtained for a distance of 1000 km

Nuclide	Emission to, at 1 000 km distance					
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Risk/Bq			*Bq U235 air-eq./Bq*		
Ac228	3.10E-31	1.99E-33	7.02E-159	3.47E-11	2.23E-13	7.87E-139
Ag110m	9.01E-22	2.12E-17	3.97E-21	1.01E-01	2.37E+03	4.45E-01
Am241	3.35E-20	3.25E-18	5.57E-21	3.75E+00	3.65E+02	6.25E-01
As76	1.31E-26	1.05E-23	1.53E-54	1.47E-06	1.17E-03	1.72E-34
At211	1.40E-29	4.20E-31	1.33E-139	1.57E-09	4.71E-11	1.49E-119
Au198	3.52E-25	3.77E-21	1.77E-36	3.95E-05	4.22E-01	1.98E-16
Bi206	1.86E-23	1.87E-20	9.99E-29	2.09E-03	2.10E+00	1.12E-08
Bi210	7.10E-24	8.10E-22	1.78E-31	7.97E-04	9.09E-02	1.99E-11
Bi212	7.88E-66	5.24E-99	0.00E+00	8.84E-46	5.88E-79	0.00E+00
Br82	1.47E-24	1.03E-22	6.48E-49	1.64E-04	1.15E-02	7.27E-29
C14	4.82E-24	0.00E+00	0.00E+00	5.41E-04	0.00E+00	0.00E+00
Cd109	3.05E-22	1.99E-19	5.11E-22	3.42E-02	2.23E+01	5.72E-02
Ce141	5.29E-23	2.09E-19	1.53E-22	5.93E-03	2.34E+01	1.72E-02
Ce144	7.58E-22	3.14E-18	1.66E-20	8.50E-02	3.51E+02	1.86E+00
Cm242	1.93E-21	4.24E-20	8.82E-23	2.16E-01	4.75E+00	9.89E-03
Cm244	2.00E-20	4.39E-19	7.17E-22	2.24E+00	4.92E+01	8.04E-02
Co58	1.25E-22	1.47E-18	4.77E-21	1.40E-02	1.64E+02	5.34E-01
Co60	3.67E-21	1.39E-17	1.21E-19	4.11E-01	1.56E+03	1.36E+01
Cr51	3.17E-24	7.66E-21	2.21E-24	3.56E-04	8.59E-01	2.48E-04
Cs134	4.11E-21	5.78E-18	1.25E-21	4.60E-01	6.49E+02	1.40E-01
Cs135	3.60E-22	3.45E-20	9.53E-24	4.04E-02	3.86E+00	1.07E-03
Cs136	1.14E-22	2.89E-19	2.66E-25	1.27E-02	3.24E+01	2.98E-05
Cs137	3.56E-21	2.59E-18	6.11E-22	3.99E-01	2.90E+02	6.84E-02
Cu64	4.57E-29	2.35E-27	4.83E-90	5.12E-09	2.63E-07	5.42E-70
Eu154	2.78E-21	9.17E-20	1.85E-19	3.11E-01	1.03E+01	2.08E+01
Eu155	1.24E-22	5.11E-21	8.83E-21	1.39E-02	5.72E-01	9.90E-01
Fe55	5.12E-23	9.99E-22	4.35E-22	5.75E-03	1.12E-01	4.88E-02
Fe59	1.97E-22	1.30E-19	8.55E-21	2.21E-02	1.46E+01	9.59E-01
Ga67	1.94E-25	1.18E-21	1.79E-33	2.18E-05	1.33E-01	2.00E-13
H3	6.71E-26	1.12E-22	1.84E-25	7.52E-06	1.25E-02	2.07E-05
Hg197	7.72E-26	1.39E-21	2.80E-35	8.65E-06	1.56E-01	3.13E-15
Hg197m	2.21E-27	1.05E-23	4.47E-57	2.48E-07	1.18E-03	5.02E-37
Hg203	1.84E-22	9.44E-19	4.94E-22	2.07E-02	1.06E+02	5.54E-02
I123	7.52E-29	1.75E-27	3.78E-89	8.43E-09	1.96E-07	4.23E-69
I125	1.71E-21	8.28E-20	1.73E-24	1.92E-01	9.28E+00	1.94E-04
I129	1.93E-20	6.57E-19	4.77E-23	2.17E+00	7.37E+01	5.34E-03
I131	3.10E-22	5.29E-20	4.47E-28	3.47E-02	5.93E+00	5.02E-08
I132	1.38E-43	2.95E-55	0.00E+00	1.55E-23	3.31E-35	0.00E+00
I133	2.32E-26	4.57E-24	1.15E-64	2.60E-06	5.12E-04	1.28E-44

I134	2.30E-73	1.10E-110	0.00E+00	2.58E-53	1.23E-90	0.00E+00
I135	1.84E-31	1.06E-32	1.79E-151	2.07E-11	1.19E-12	2.00E-131
In111	2.56E-25	1.71E-20	2.63E-35	2.87E-05	1.91E+00	2.95E-15
In113m	1.29E-51	2.07E-67	0.00E+00	1.45E-31	2.32E-47	0.00E+00
Kr85	1.25E-28	6.36E-26	2.48E-47	1.40E-08	7.13E-06	2.77E-27
Mn54	2.79E-22	6.00E-18	7.17E-20	3.12E-02	6.73E+02	8.04E+00
Mo99	3.16E-25	8.59E-22	7.75E-36	3.55E-05	9.63E-02	8.68E-16
Na22	2.05E-21	7.76E-18	1.82E-25	2.30E-01	8.69E+02	2.05E-05
Na24	3.41E-27	2.76E-25	5.34E-82	3.82E-07	3.09E-05	5.98E-62
Nb95	6.09E-23	2.98E-21	1.16E-21	6.82E-03	3.34E-01	1.31E-01
Ni59	1.04E-23	1.71E-22	3.01E-24	1.16E-03	1.91E-02	3.37E-04
Ni63	2.46E-23	4.05E-22	7.14E-24	2.75E-03	4.54E-02	8.01E-04
Np237	1.96E-20	2.62E-19	6.13E-23	2.20E+00	2.94E+01	6.88E-03
Np239	1.19E-25	8.04E-23	7.14E-40	1.34E-05	9.01E-03	8.01E-20
P32	1.36E-22	8.64E-18	1.26E-24	1.52E-02	9.69E+02	1.41E-04
Pa231	1.19E-19	1.61E-18	1.02E-20	1.34E+01	1.81E+02	1.14E+00
Pa233	5.78E-23	1.62E-21	2.21E-22	6.49E-03	1.82E-01	2.48E-02
Pb210	1.05E-19	2.82E-18	7.35E-21	1.17E+01	3.17E+02	8.24E-01
Pd103	7.56E-24	1.14E-21	3.54E-26	8.48E-04	1.27E-01	3.97E-06
Pd107	6.47E-24	1.01E-22	5.30E-25	7.26E-04	1.13E-02	5.94E-05
Pd109	3.39E-29	2.48E-27	2.32E-86	3.80E-09	2.77E-07	2.60E-66
Pm147	3.90E-23	9.72E-22	8.04E-24	4.37E-03	1.09E-01	9.01E-04
Po210	1.42E-18	3.38E-17	6.40E-20	1.59E+02	3.79E+03	7.17E+00
Pu238	3.80E-20	2.09E-16	1.12E-21	4.27E+00	2.34E+04	1.25E-01
Pu239	4.14E-20	2.27E-16	1.20E-21	4.65E+00	2.55E+04	1.35E-01
Pu240	4.14E-20	2.27E-16	1.21E-21	4.65E+00	2.55E+04	1.36E-01
Pu241	7.90E-22	4.38E-18	4.30E-23	8.86E-02	4.91E+02	4.82E-03
Pu242	3.98E-20	2.18E-16	1.16E-21	4.46E+00	2.45E+04	1.31E-01
Ra224	1.40E-22	3.06E-20	9.90E-32	1.57E-02	3.43E+00	1.11E-11
Ra225	3.69E-21	2.06E-19	2.13E-24	4.14E-01	2.31E+01	2.38E-04
Ra226	5.34E-20	2.89E-18	2.22E-21	5.99E+00	3.24E+02	2.49E-01
Rb86	1.69E-22	9.63E-20	1.72E-25	1.89E-02	1.08E+01	1.93E-05
Rh105	9.81E-27	2.35E-23	5.11E-47	1.10E-06	2.63E-03	5.72E-27
Rh107	3.77E-142	2.24E-237	0.00E+00	4.22E-122	2.51E-217	0.00E+00
Rn222	3.28E-30	3.83E-31	1.12E-59	3.68E-10	4.30E-11	1.25E-39
Ru103	7.21E-23	2.84E-19	9.09E-23	8.09E-03	3.19E+01	1.02E-02
Ru106	1.09E-21	1.17E-18	1.92E-21	1.22E-01	1.32E+02	2.15E-01
S35	5.94E-22	1.46E-20	1.47E-26	6.66E-02	1.63E+00	1.64E-06
Sb124	3.23E-22	2.79E-19	7.32E-23	3.62E-02	3.12E+01	8.20E-03
Sb125	4.79E-22	2.91E-19	2.07E-22	5.36E-02	3.26E+01	2.32E-02
Se75	4.88E-22	1.78E-19	6.95E-22	5.47E-02	1.99E+01	7.79E-02
Sn113	1.16E-22	1.16E-18	1.97E-20	1.29E-02	1.29E+02	2.21E+00
Sr85	8.14E-23	2.18E-20	9.53E-26	9.13E-03	2.45E+00	1.07E-05
Sr87m	4.04E-41	1.93E-50	0.00E+00	4.53E-21	2.16E-30	0.00E+00

Sr89	2.52E-22	9.44E-21	8.14E-26	2.83E-02	1.06E+00	9.13E-06
Sr90	4.72E-21	1.10E-19	4.19E-24	5.29E-01	1.23E+01	4.70E-04
Tc99	3.65E-22	1.49E-21	2.45E-24	4.09E-02	1.66E-01	2.74E-04
Tc99m	2.90E-33	5.84E-36	9.26E-165	3.25E-13	6.55E-16	1.04E-144
Te125m	9.00E-23	3.35E-21	1.10E-23	1.01E-02	3.75E-01	1.23E-03
Te127m	2.86E-22	9.35E-21	5.35E-23	3.21E-02	1.05E+00	6.00E-03
Te129m	2.28E-22	1.04E-20	1.33E-23	2.56E-02	1.16E+00	1.49E-03
Te131m	6.35E-26	2.06E-23	1.22E-50	7.12E-06	2.31E-03	1.37E-30
Te132	3.62E-24	1.66E-21	3.92E-33	4.06E-04	1.86E-01	4.40E-13
Th228	1.36E-20	2.95E-17	5.12E-19	1.52E+00	3.31E+03	5.73E+01
Th230	4.46E-20	4.35E-17	1.34E-18	4.99E+00	4.88E+03	1.50E+02
Th232	5.12E-20	5.94E-17	1.55E-18	5.75E+00	6.66E+03	1.74E+02
Tl201	1.16E-25	2.12E-22	1.73E-34	1.29E-05	2.37E-02	1.94E-14
Tl202	1.56E-23	2.43E-20	4.52E-25	1.75E-03	2.72E+00	5.07E-05
U232	6.15E-20	7.97E-19	3.79E-22	6.90E+00	8.93E+01	4.24E-02
U234	1.81E-20	1.30E-19	4.46E-22	2.02E+00	1.46E+01	5.01E-02
U235	8.92E-21	1.13E-19	4.35E-23	1.00E+00	1.26E+01	4.88E-03
U238	1.79E-20	1.21E-19	4.69E-22	2.00E+00	1.36E+01	5.26E-02
Xe133	3.15E-28	1.05E-28	5.35E-55	3.54E-08	1.17E-08	6.00E-35
Y87	8.52E-25	3.02E-22	4.44E-32	9.55E-05	3.38E-02	4.97E-12
Y90	5.02E-25	4.02E-22	1.27E-35	5.63E-05	4.50E-02	1.42E-15
Y91	2.81E-22	8.38E-21	5.18E-22	3.16E-02	9.39E-01	5.81E-02
Zn65	9.72E-22	1.27E-19	4.38E-21	1.09E-01	1.42E+01	4.91E-01
Zr95	1.61E-22	5.12E-20	1.74E-20	1.81E-02	5.75E+00	1.95E+00

Table A.6 – CGM characterisation factors for emissions into air, fresh water and sea water obtained for a distance of 10 000 km

Nuclide	Emission to, at 10 000 km distance					
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Risk/Bq			*Bq U235 air-eq./Bq*		
Ac228	2.03E-94	1.35E-147	0.00E+	1.49E-72	9.93E-126	0
Ag110m	1.18E-23	1.62E-17	1.49E-23	8.72E-02	1.20E+05	1.10E-01
Am241	5.09E-22	3.25E-18	3.75E-22	3.75E+00	2.40E+04	2.76E+00
As76	1.08E-42	3.00E-50	0.00E+00	7.97E-21	2.21E-28	0.00E+00
At211	1.40E-83	3.50E-128	0.00E+00	1.03E-61	2.58E-106	0.00E+00
Au198	8.04E-33	5.50E-32	3.98E-154	5.93E-11	4.05E-10	2.93E-132
Bi206	8.51E-28	3.82E-25	2.57E-80	6.27E-06	2.82E-03	1.89E-58
Bi210	8.07E-29	1.24E-27	3.48E-95	5.95E-07	9.12E-06	2.57E-73
Bi212	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Br82	4.75E-37	1.44E-42	1.70E-263	3.50E-15	1.06E-20	1.25E-241
C14	7.33E-26	-	-	5.41E-04	-	-
Cd109	4.30E-24	1.72E-19	7.27E-24	3.17E-02	1.27E+03	5.36E-02
Ce141	2.65E-25	2.65E-20	2.29E-33	1.95E-03	1.95E+02	1.69E-11
Ce144	1.02E-23	2.48E-18	8.81E-23	7.50E-02	1.82E+04	6.49E-01
Cm242	2.35E-23	2.81E-20	7.05E-26	1.73E-01	2.07E+02	5.20E-04
Cm244	3.03E-22	4.35E-19	4.35E-23	2.23E+00	3.21E+03	3.20E-01
Co58	1.14E-24	5.69E-19	1.24E-26	8.38E-03	4.20E+03	9.12E-05
Co60	5.47E-23	1.35E-17	5.64E-21	4.03E-01	9.93E+04	4.16E+01
Cr51	1.31E-26	6.77E-22	6.91E-37	9.66E-05	4.99E+00	5.09E-15
Cs134	5.95E-23	5.29E-18	3.21E-23	4.39E-01	3.90E+04	2.36E-01
Cs135	5.48E-24	3.45E-20	6.47E-25	4.04E-02	2.54E+02	4.77E-03
Cs136	1.09E-25	1.71E-21	1.97E-50	8.04E-04	1.26E+01	1.45E-28
Cs137	5.39E-23	2.57E-18	3.86E-23	3.97E-01	1.89E+04	2.84E-01
Cu64	1.37E-60	1.27E-82	0.00E+00	1.01E-38	9.39E-61	0.00E+00
Eu154	4.17E-23	8.98E-20	9.99E-21	3.07E-01	6.62E+02	7.36E+01
Eu155	1.85E-24	4.91E-21	4.01E-22	1.36E-02	3.62E+01	2.95E+00
Fe55	7.52E-25	9.35E-22	1.41E-23	5.54E-03	6.89E+00	1.04E-01
Fe59	1.33E-24	2.88E-20	5.33E-29	9.80E-03	2.12E+02	3.93E-07
Ga67	4.60E-32	1.34E-30	8.52E-131	3.39E-10	9.86E-09	6.28E-109
H3	1.01E-27	9.63E-23	1.06E-26	7.43E-06	7.09E-01	7.84E-05
Hg197	1.54E-33	1.58E-32	4.23E-154	1.14E-11	1.16E-10	3.11E-132
Hg197m	4.97E-45	3.74E-53	0.00E+00	3.66E-23	2.76E-31	0.00E+00
Hg203	1.29E-24	2.25E-19	6.33E-30	9.53E-03	1.66E+03	4.66E-08
I123	3.35E-59	1.44E-80	0.00E+00	2.47E-37	1.06E-58	0.00E+00
I125	1.42E-23	2.70E-20	6.78E-31	1.05E-01	1.99E+02	5.00E-09
I129	2.94E-22	6.57E-19	3.23E-24	2.17E+00	4.84E+03	2.38E-02
I131	5.28E-26	1.25E-23	2.97E-68	3.89E-04	9.19E-02	2.19E-46
I132	3.56E-209	0.00E+00	0.00E+00	2.62E-187	0.00E+00	0.00E+00
I133	2.81E-46	9.81E-58	0.00E+00	2.07E-24	7.23E-36	0.00E+00

I134	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
I135	3.77E-90	1.67E-138	0.00E+00	2.78E-68	1.23E-116	0.00E+00
In111	1.10E-32	8.07E-31	1.75E-147	8.11E-11	5.95E-09	1.29E-125
In113m	3.91E-280	0.00E+00	0.00E+00	2.89E-258	0.00E+00	0.00E+00
Kr85	1.87E-30	6.25E-26	1.39E-48	1.38E-08	4.61E-04	1.03E-26
Mn54	3.78E-24	4.84E-18	4.79E-22	2.78E-02	3.57E+04	3.53E+00
Mo99	9.44E-33	2.07E-32	3.86E-151	6.96E-11	1.53E-10	2.84E-129
Na22	3.00E-23	7.23E-18	5.77E-27	2.21E-01	5.33E+04	4.25E-05
Na24	5.01E-54	7.96E-72	0.00E+00	3.69E-32	5.86E-50	0.00E+00
Nb95	3.30E-25	4.38E-22	8.84E-32	2.43E-03	3.23E+00	6.51E-10
Ni59	1.58E-25	1.71E-22	2.04E-25	1.16E-03	1.26E+00	1.50E-03
Ni63	3.73E-25	4.04E-22	4.73E-25	2.75E-03	2.98E+00	3.49E-03
Np237	2.98E-22	2.62E-19	4.14E-24	2.20E+00	1.93E+03	3.05E-02
Np239	4.11E-34	3.49E-35	6.16E-174	3.03E-12	2.57E-13	4.54E-152
P32	1.65E-25	7.88E-20	1.01E-47	1.22E-03	5.81E+02	7.43E-26
Pa231	1.82E-21	1.61E-18	6.86E-22	1.34E+01	1.19E+04	5.05E+00
Pa233	2.31E-25	1.35E-22	3.68E-35	1.70E-03	9.93E-01	2.71E-13
Pb210	1.59E-21	2.81E-18	4.55E-22	1.17E+01	2.07E+04	3.35E+00
Pd103	1.38E-26	2.19E-23	8.52E-46	1.01E-04	1.61E-01	6.28E-24
Pd107	9.81E-26	1.01E-22	3.58E-26	7.23E-04	7.43E-01	2.64E-04
Pd109	3.72E-59	1.08E-79	0.00E+00	2.74E-37	7.97E-58	0.00E+00
Pm147	5.71E-25	9.08E-22	2.56E-25	4.21E-03	6.70E+00	1.89E-03
Po210	1.66E-20	2.08E-17	2.30E-23	1.22E+02	1.53E+05	1.70E-01
Pu238	5.78E-22	2.09E-16	7.37E-23	4.26E+00	1.54E+06	5.43E-01
Pu239	6.30E-22	2.27E-16	8.12E-23	4.64E+00	1.68E+06	5.99E-01
Pu240	6.30E-22	2.27E-16	8.20E-23	4.64E+00	1.68E+06	6.04E-01
Pu241	1.19E-23	4.33E-18	2.53E-24	8.78E-02	3.19E+04	1.86E-02
Pu242	6.04E-22	2.18E-16	7.86E-23	4.45E+00	1.61E+06	5.79E-01
Ra224	1.12E-28	3.35E-28	1.68E-118	8.24E-07	2.47E-06	1.24E-96
Ra225	4.90E-24	2.21E-21	9.35E-47	3.61E-02	1.63E+01	6.89E-25
Ra226	8.13E-22	2.89E-18	1.50E-22	5.99E+00	2.13E+04	1.11E+00
Rb86	3.73E-25	2.65E-21	1.98E-43	2.75E-03	1.95E+01	1.46E-21
Rh105	3.20E-39	3.30E-43	1.34E-261	2.36E-17	2.43E-21	9.86E-240
Rh107	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Rn222	3.92E-36	8.87E-39	6.26E-143	2.89E-14	6.54E-17	4.61E-121
Ru103	4.38E-25	5.15E-20	6.53E-32	3.23E-03	3.80E+02	4.81E-10
Ru106	1.50E-23	9.72E-19	1.82E-23	1.11E-01	7.16E+03	1.34E-01
S35	5.98E-24	6.77E-21	2.54E-31	4.41E-02	4.99E+01	1.87E-09
Sb124	2.70E-24	9.14E-20	3.13E-29	1.99E-02	6.74E+02	2.30E-07
Sb125	7.02E-24	2.72E-19	6.87E-24	5.18E-02	2.01E+03	5.06E-02
Se75	5.48E-24	1.02E-19	1.14E-25	4.04E-02	7.50E+02	8.38E-04
Sn113	1.28E-24	6.44E-19	2.49E-24	9.46E-03	4.75E+03	1.84E-02
Sr85	7.09E-25	7.72E-21	9.17E-32	5.22E-03	5.69E+01	6.76E-10
Sr87m	2.15E-177	1.36E-300	0.00E+00	1.58E-155	1.00E-278	0.00E+00

Sr89	1.88E-24	2.48E-21	3.36E-33	1.39E-02	1.83E+01	2.47E-11
Sr90	7.16E-23	1.10E-19	2.65E-25	5.28E-01	8.11E+02	1.95E-03
Tc99	5.55E-24	1.49E-21	1.65E-25	4.09E-02	1.09E+01	1.22E-03
Tc99m	1.27E-97	2.61E-152	0.00E+00	9.39E-76	1.93E-130	0.00E+00
Te125m	7.35E-25	1.05E-21	3.00E-30	5.42E-03	7.77E+00	2.21E-08
Te127m	3.13E-24	5.04E-21	4.77E-27	2.30E-02	3.72E+01	3.51E-05
Te129m	1.18E-24	1.40E-21	4.09E-34	8.72E-03	1.03E+01	3.01E-12
Te131m	2.74E-40	9.35E-47	9.53E-303	2.02E-18	6.89E-25	7.03E-281
Te132	8.58E-31	1.88E-30	1.86E-130	6.32E-09	1.39E-08	1.37E-108
Th228	1.96E-22	2.69E-17	1.23E-20	1.45E+00	1.98E+05	9.05E+01
Th230	6.77E-22	4.35E-17	9.03E-20	4.99E+00	3.21E+05	6.66E+02
Th232	7.80E-22	5.94E-17	1.05E-19	5.75E+00	4.38E+05	7.77E+02
Tl201	1.22E-32	5.34E-32	7.57E-139	8.99E-11	3.93E-10	5.58E-117
Tl202	1.23E-26	9.81E-23	5.83E-52	9.05E-05	7.23E-01	4.30E-30
U232	9.35E-22	7.95E-19	2.49E-23	6.89E+00	5.86E+03	1.84E-01
U234	2.75E-22	1.30E-19	3.02E-23	2.03E+00	9.59E+02	2.22E-01
U235	1.36E-22	1.13E-19	2.94E-24	1.00E+00	8.31E+02	2.17E-02
U238	2.71E-22	1.21E-19	3.17E-23	2.00E+00	8.92E+02	2.34E-01
Xe133	4.91E-33	2.86E-34	5.71E-116	3.62E-11	2.11E-12	4.21E-94
Y87	2.64E-31	5.66E-31	4.68E-127	1.95E-09	4.17E-09	3.45E-105
Y90	9.99E-33	4.57E-33	1.91E-154	7.36E-11	3.37E-11	1.41E-132
Y91	2.31E-24	2.66E-21	1.55E-28	1.70E-02	1.96E+01	1.14E-06
Zn65	1.27E-23	9.63E-20	1.53E-23	9.39E-02	7.09E+02	1.13E-01
Zr95	1.40E-24	1.80E-20	1.53E-26	1.03E-02	1.32E+02	1.13E-04

Table A.7 – UCrad characterisation factors for radionuclides contained in HLW, I-LLW, SF, HEU, DNLEU and Pu waste disposed in a Geological Disposal Facility. Characterisation factors are expressed in terms of risk per Bq disposed.

Nuclide	Emissions from HLW		Nuclide	Emissions from I-LLW	
	risk/Bq	*Bq U238 HLW-eq./Bq*		*risk/Bq*	*Bq U238 HLW-eq./Bq*
Be10	6.51E-31	4.52E-02	**Cl36**	2.31E-27	1.60E+02
Cl36	1.58E-28	1.10E+01	**I129**	8.67E-26	6.01E+03
Cs135	3.08E-31	2.14E-02	**Np237**	5.34E-31	3.70E-02
I129	1.30E-26	8.99E+02	**Pb210**	5.94E-32	4.12E-03
Np237	3.58E-33	2.48E-04	**Ra226**	5.95E-31	4.12E-02
Se79	2.15E-30	1.49E-01	**TC99**	6.88E-31	4.77E-02
Sn126	9.86E-35	6.83E-06	**Th229**	6.86E-28	4.75E+01
Tc99	1.52E-34	1.05E-05	**Th230**	2.03E-28	1.41E+01
Th229	1.16E-29	8.07E-01	**U233**	5.89E-29	4.08E+00
U233	1.05E-29	7.30E-01	**U234**	3.68E-30	2.55E-01
			U238	1.44E-29	1.00E+00
Nuclide	**Emissions from SF**		**Nuclide**	**Emissions from HEU**	
	risk/Bq	*Bq U238 HLW-eq./Bq*		*risk/Bq*	*Bq U238 HLW-eq./Bq*
Cl36	7.05E-29	1.91E+01	**Ac227**	8.61E-34	2.34E-04
Cs135	7.30E-32	1.98E-02	**Pa231**	2.85E-31	7.75E-02
I129	1.08E-26	2.94E+03	**Pb210**	2.37E-33	6.45E-04
Ni59	1.04E-35	2.82E-06	**Ra226**	2.26E-30	6.14E-01
Np237	2.00E-34	5.44E-05	**Th230**	1.21E-33	3.29E-04
Pa231	4.59E-30	1.25E+00	**U234**	4.07E-34	1.11E-04
Ra226	6.22E-31	1.69E-01	**U235**	1.15E-33	3.12E-04
Se79	4.02E-31	1.09E-01	**U238**	1.04E-32	2.81E-03
Sn126	6.64E-36	1.80E-06			
U233	3.10E-31	8.43E-02			
Nuclide	**Emissions from DNLEU**		**Nuclide**	**Emissions from Pu**	
	risk/Bq	*Bq U238 HLW-eq./Bq*		*risk/Bq*	*Bq U238 HLW-eq./Bq*
Pb210	3.28E-28	8.92E+01	**Np237**	8.65E-34	2.35E-04
Ra226	2.97E-26	8.08E+03	**Pa231**	7.79E-29	2.12E+01
Th230	3.32E-29	9.02E+00	**Ra226**	4.33E-30	1.18E+00
U234	5.11E-30	1.39E+00	**U233**	5.74E-30	1.56E+00
U235	3.69E-31	1.00E-01	**U236**	2.42E-32	6.57E-03
U236	1.48E-31	4.01E-02			
U238	2.56E-30	6.95E-01			

Table A.8 – CGM characterisation factors for radionuclides contained in HLW, I-LLW, SF, HEU, DNLEU and Pu waste disposed in a Geological Disposal Facility

Nuclide	Emissions from HLW		Nuclide	Emissions from I-LLW	
	risk/Bq	*Bq U238 ILLW-eq./Bq*		*risk/Bq*	*Bq U238 ILLW -eq./Bq*
Be10	3.43E-23	8.17E-01	**Cl36**	2.65E-22	6.31E+00
Cl36	1.81E-23	4.31E-01	**I129**	1.70E-20	4.05E+02
Cs135	4.35E-23	1.04E+00	**Np237**	2.69E-23	6.40E-01
I129	2.54E-21	6.05E+01	**Pb210**	2.33E-22	5.55E+00
Np237	1.80E-25	4.29E-03	**Ra226**	6.02E-23	1.43E+00
Se79	1.14E-22	2.71E+00	**TC99**	1.22E-24	2.90E-02
Sn126	4.04E-26	9.62E-04	**Th229**	4.11E-19	9.79E+03
Tc99	2.70E-28	6.43E-06	**Th230**	2.49E-20	5.93E+02
Th229	6.99E-21	1.66E+02	**U233**	3.17E-21	7.55E+01
U233	5.66E-22	1.35E+01	**U234**	1.34E-23	3.19E-01
			U238	4.20E-23	1.00E+00
Nuclide	**Emissions from SF**		**Nuclide**	**Emissions from HEU**	
	risk/Bq	*Bq U238 ILLW -eq./Bq*		*risk/Bq*	*Bq U238 ILLW -eq./Bq*
Cl36	1.04E-23	7.75E-01	**Ac227**	3.24E-24	2.41E-01
Cs135	1.03E-23	7.70E-01	**Pa231**	4.48E-23	3.35E+00
I129	2.12E-21	1.58E+02	**Pb210**	9.30E-24	6.94E-01
Ni59	4.19E-28	3.13E-05	**Ra226**	2.29E-22	1.71E+01
Np237	1.01E-26	7.53E-04	**Th230**	1.48E-25	1.11E-02
Pa231	7.20E-22	5.38E+01	**U234**	1.48E-27	1.10E-04
Ra226	6.29E-23	4.69E+00	**U235**	2.36E-26	1.76E-03
Se79	2.13E-23	1.59E+00	**U238**	3.01E-26	2.25E-03
Sn126	2.72E-27	2.03E-04			
U233	1.67E-23	1.25E+00			
Nuclide	**Emissions from DNLEU**		**Nuclide**	**Emissions from Pu**	
	risk/Bq	*Bq U238 ILLW -eq./Bq*		*risk/Bq*	*Bq U238 ILLW -eq./Bq*
Pb210	1.29E-18	9.60E+04	**Np237**	4.36E-26	3.25E-03
Ra226	3.01E-18	2.25E+05	**Pa231**	1.22E-20	9.14E+02
Th230	4.07E-21	3.04E+02	**Ra226**	4.39E-22	3.27E+01
U234	1.86E-23	1.38E+00	**U233**	3.09E-22	2.30E+01
U235	7.57E-24	5.65E-01	**U236**	1.22E-24	9.14E-02
U236	7.47E-24	5.57E-01			
U238	7.45E-24	5.56E-01			

Table A.9 – CGM characterisation factors of toxic substances for emissions into air, fresh water and sea water obtained for 1, 100, 1 000 and 10 000 km

Substance	Emission to, at 1 km			Emission to, at 100 km		
	Air	Fresh water	Sea water	Air	Fresh water	Sea water
	Cases per year/Bq			*Cases per year/Bq*		
B	4.46E-09	9.63E-07	2.46E-07	2.48E-12	9.63E-07	1.13E-09
Hg	2.91E-12	7.76E-15	2.20E-13	1.55E-15	7.61E-15	8.16E-16
PeCB	1.30E-10	6.27E-11	1.91E-09	7.24E-14	6.25E-11	8.35E-12
	at 1 000 km			**at 10 000 km**		
B	3.78E-14	9.63E-07	7.61E-11	5.74E-16	9.63E-07	5.15E-12
Hg	1.56E-17	6.39E-15	8.33E-18	3.77E-21	1.10E-15	3.49E-27
PeCB	1.08E-15	6.02E-11	3.77E-13	1.35E-17	4.13E-11	4.44E-16

A.2 Inventory for normalisation factors

Table A.10 –CML inventory for radioactive emission from Eu25+3

Nuclide	Compartment	Emission *kBq*	Nuclide	Compartment	Emission *kBq*
Am241	air	5.10E+05	**Pu240**	air	3.29E+05
Am241	fresh water	3.40E+08	**Pu240**	fresh water	6.80E+08
Ar41	air	6.78E+13	**Pu241**	air	3.06E+06
C14	air	2.12E+11	**Pu241**	fresh water	3.63E+10
C14	fresh water	5.90E+10	**Rn222**	air	2.49E+10
Ce144	fresh water	6.23E+09	**Ru106**	air	1.25E+07
Co60	air	3.74E+05	**Ru106**	fresh water	3.06E+10
Co60	fresh water	1.38E+10	**S35**	air	1.20E+10
Cs134	fresh water	2.61E+09	**S35**	fresh water	2.07E+10
Cs137	air	1.12E+07	**Sb125**	air	2.04E+06
Cs137	fresh water	8.77E+10	**Se75**	air	3.06E+06
H3	air	8.44E+12	**Sr90**	air	6.12E+05
H3	fresh water	4.66E+13	**Sr90**	fresh water	2.27E+11
I125	air	5.35E+07	**Tc99**	fresh water	4.99E+11
I125	fresh water	1.27E+08	**Th230**	fresh water	7.82E+08
I129	air	2.83E+08	**Th232**	fresh water	9.63E+06
I129	fresh water	5.33E+09	**U234**	air	7.05E+06
I131	air	6.80E+07	**U234**	fresh water	3.35E+08
Kr85	air	8.38E+14	**U235**	air	3.06E+05
Nb95	fresh water	1.08E+09	**U235**	fresh water	1.47E+07
Np237	fresh water	5.61E+06	**U238**	air	6.60E+06
Pu239	air	3.29E+05	**U238**	fresh water	4.20E+08
Pu239	fresh water	6.80E+08	**Zr95**	fresh water	1.08E+09

Table A.11 – ILCD inventory for radioactive emission from EU27

Nuclide	Compartment	Emission (kBq)	Nuclide	Compartment	Emission (kBq)
Ag110m	air	7.68E+02	**Pu (alpha)**	air	2.01E-05
Ar41	air	3.68E+09	**Pu238**	air	8.78E-06
Ba140	air	1.36E+05	**Pu241**	air	1.03E+06
Ba140	water (unsp)	3.53E+05	**Pu241**	water (unsp)	3.53E+09
C14	air	4.69E+10	**Ra224**	water (unsp)	5.87E+04
C14	water (unsp)	1.16E+10	**Ra228**	water (unsp)	3.39E+08
Ce141	air	3.29E+04	**Ra228**	air	8.13E+06
Ce141	water (unsp)	1.69E+05	**Rn220**	air	1.74E+09
Ce144	water (unsp)	1.18E+05	**Ru103**	air	2.81E+01
Co57	water (unsp)	2.19E+06	**Ru103**	water (unsp)	7.19E+04
Co60	water (unsp)	1.35E+10	**Sb122**	water (unsp)	2.22E+05
Cr51	water (unsp)	3.95E+07	**Sb124**	air	2.74E+02
Cr51	air	2.11E+03	**Sb125**	air	5.74E+03
Cs134	water (unsp)	4.87E+06	**Sr89**	water (unsp)	2.77E+06
Cs136	water (unsp)	6.89E+04	**Sr90**	air	6.20E+05
Cs137	water (unsp)	6.71E+09	**Tc99**	water (unsp)	4.51E+10
Fe59	water (unsp)	1.27E+05	**Te123**	water (unsp)	9.03E+05
I129	air	1.67E+07	**Te132**	water (unsp)	2.07E+04
I129	water (unsp)	1.54E+09	**Th228**	water (unsp)	1.25E+06
I133	water (unsp)	2.87E+05	**Th228**	air	4.40E+06
K40	water (unsp)	7.80E+02	**Th230**	air	2.22E+05
K40	air	2.78E+07	**Th230**	water (unsp)	9.80E+04
Kr87	air	5.67E+08	**Th232**	water (unsp)	1.41E+02
Kr88	air	6.79E+08	**Th232**	air	6.83E+06
Kr89	air	2.53E+08	**Th234**	water (unsp)	7.47E+02
La140	air	1.16E+04	**Th234**	air	2.22E+05
La140	water (unsp)	4.53E+05	**U234**	air	4.35E+05
Mn	air	1.08E+03	**U235**	air	2.29E+01
Mo99	water (unsp)	1.31E+05	**Xe135**	air	6.60E+10
Na24	water (unsp)	2.54E+06	**Xe137**	air	6.93E+08
Nb95	air	1.46E+02	**Xe138**	air	5.36E+09
Nb95	water (unsp)	9.55E+05	**Zn65**	air	5.39E+03
Pa234	water (unsp)	7.47E+02	**Zn65**	water (unsp)	1.33E+07
Pa234	air	2.22E+05	**Zr95**	air	1.43E+04
Pb210	water (unsp)	7.76E+07	**Zr95**	water (unsp)	3.88E+05
Po210	water (unsp)	4.15E+06			

Notes:
* unsp: unspecified. The water body is not known; it can be either freshwater or seawater.

Appendix B

B.1 Inventory data

Table B.1 – THORP output streams for a reference throughput of 1 teU of uranium pre-irradiation

Stream	to	Amount		Source
Uranium Oxide	Valuable product	9.69E-01	tHM	THORP operational flowsheet
Plutonium Oxide	Valuable product	5.20E-03	tHM	
Highly Active Liquor	HALES	8.82E+00	m^3	Personal communication with THORP technical team. Reference is financial year 2015-16
Salt Evaporate Concentrate	EARP	8.36E+00*	m^3	2016 UKRWI [322] & EARP (SEC) operational flowsheet
Salt Evaporate Concentrate floc		*1.38E+02*	kg	2016 UKRWI [322]
Low Active Effluent	SETP	5.86E+02	m^3	SETP operational flowsheet
AGR cladding	WEP	1.95E+02	kg	2016 UKRWI [322]
Multi-Element Bottle crud	WEP	6.58E+00	kg	2016 UKRWI [322]
Centrifuge cake	WEP	3.51E+01**	kg	2016 UKRWI [322]
Maintenance scrap	WEP	8.45E+00	kg	2016 UKRWI [322]
Plutonium Contaminated Material	WTC	2.78E+01	kg	2016 UKRWI [322]
AGR graphite components	*tbd*	4.89E+01	kg	2016 UKRWI [322]
AGR stainless steel components	*tbd*	2.69E+01	kg	2016 UKRWI [322]
Dissolver Off-Gas stream	DOG plant	1.51E+02	m^3	Operational flowsheet for feed clarification, accountancy and buffer storage for AGR fuel.

Notes:

*SEC needed to produce amount of floc to match UKRWI figure (line below).

**Double-checked with operational flowsheet for feed clarification, accountancy and buffer storage for AGR.

Table B.2 – THORP Chemical and reagents consumption

Reference: 2015-16 financial year
Source: THORP reagents and utilities monitor

	Total FY 2015-16		per teU reprocessed*	
Chemicals				
Nitric Acid (12 M)	1.14E+03	m^3	2.66E+00	m^3
Sodium Hydroxide (7.2 M)	8.04E+02	m^3	1.87E+00	m^3
Sodium Carbonate (0.25 M)	1.89E+03	m^3	4.39E+00	m^3
Sodium Nitrite (6.5 M)	4.44E+02	m^3	1.03E+00	m^3
Odourless Kerosene	1.14E+02	m^3	2.65E-01	m^3
Tri-Butyl Phosphate	2.42E+02	200l drum	1.13E-01	m^3
Hydrazine Nitrate (4.5 M)	1.05E+02	1000L -IBC	2.44E-01	m^3
Hydroxylamine Nitrate (4.3 M)	2.11E+02	1000L -IBC	4.91E-01	m^3
Gadolinium Nitrate (545 g/l)	3.20E+01	200L drum	1.49E-02	m^3
Barium Nitrate (powder)	2.59E+02	25kg -pack	1.51E+01	kg
Nitrogen Dioxide cylinder	5.00E+00	1.25te cyl.	1.16E-02	te
UNL (450 g/l)	3.55E+01	te	8.26E-02	te
Gases				
Nitrogen	3.75E+04	m^3	8.73E+01	m^3
Utilities				
Demineralised Water	5.36E+04	m^3	1.25E+02	m^3
Domestic Water	3.44E+02	m^3	8.01E-01	m^3
High Pressure Steam	9.35E+04	te	1.33E+05	te

Notes:
*Notionally related to 430 teU throughput.

Table B.3 – Liquid and gaseous waste treatment plant input/output and consumption

HALES

Reference: Financial year 2015-16
Source: Personal communication with THORP technical team

Feed	Total from THORP	3.79E+03	m^3
	To HA Liquor Tanks	1.46E+03	m^3
	To Evap C	2.33E+03	m^3
Evaporator C	Total volume processed	4.32E+03	m^3
	MAGNOX	1.27E+02	m^3
Output	HALES/HAST to WVP	2.10E+02	m^3
	THORP part	2.04E+02	m^3
	LAE to SETP	3.60E+03	m^3

EARP

Source: EARP (SEC) operational flowsheet

Feed	SEC	1.00E+03	m^3/batch
Chemicals	Fe(NO3)3	7.74E+00	m^3/batch
	NaOH strong (7.2 M)	6.18E+01	m^3/batch
	NaOH weak (0.2 M)	2.02E+01	m^3/batch
	$Fe(CN)_6$	1.25E+01	m^3/batch
Output	Floc to WPEP	1.50E+01	m^3/batch
	Filter permeate to sea	1.26E+03	m^3/batch

DOG treatment

Source: DOG operational flowsheet

Feed	DOG stream	4.92E+02	m^3
Chemicals	NaOH (pure)	1.71E+02	kg
Output	Off gas to stack	1.14E+04	m^3
	Spent caustic to C14	1.65E+03	l

C14 abatement system

Source: C14 operational flowsheet

Chemicals	Barium Nitrate	2.60E+01	kg
	Sodium Carbonate	1.08E+00	kg
	Demin. Water	2.00E+03	l
Output	Barium Carbonate slurry to WEP	8.84E+01	l

SETP

Source: SETP operational flowsheet

Feed	Acidic effluent THORP	2.52E+05	m^3/year
Chemicals	Sodium Hydroxide (pure)	2.21E+06	kg/year

Table B.4 – Data for I-LLW encapsulation

Source: Derived Inventory for I-LLW [271]

Waste stream	Loading	Conditioning material			Container
		Grout*	Composition		
	(kg)	*(kg)*	Cement**	Water	
at Waste Encapsulation Plant					
AGR Cladding	269	900	74%	26%	500l drum
Barium Carbonate Slurry MEB crud	276	336	100%	0%	
Centrifuge Cake	255	315	100%	0%	
Maintenance Scrap	317	1000	74%	26%	
at Waste Packaging and Encapsulation Plant					
Salt Evaporate Concentrate floc	363	408	100%	0%	500l drum
at Waste Treatment Complex					
Plutonium Contaminated Materials	152	620	58%	42%	6X200l drums supercompacted in 500l drum
at To-be-defined					
AGR Graphite Components	204	434	58%	42%	500l drum
AGR Stainless Steel Components	83	437	58%	42%	500l drum

Notes:

*Includes conditioning and capping grout.

**Cement composition is 75% OPC (Ordinary Portland Cement) and 25% BFS (Blast Furnace Slag) or PFA (Pulverised Fuel Ash).

Table B.5 – Data for HLW encapsulation.

Source: Derived Inventory for HLW [272]

Waste stream	Loading	Borosilicate glass	Container
	(m^3)	*(kg)*	
at Waste Vitrification Plant			
Highly Active Liquor	0.41*	298	WVP canister

Notes:

*This refers to the amount of HAL before evaporation. Reference: 2D02 waste stream information sheet in 2016 UKRWI.

Table B.6 – Data for I-LLW and HLW containers

Source: Derived Inventory for I-LLW and HLW [271], [272]

Container	Material	Empty weight	Payload	Displacement volume
		(kg)	*(m^3)*	*(m^3)*
500 litre drum	316L SS	130	0.470	0.571
WVP canister	309 SS	85	0.150*	0.196

Notes:

*Obtained from 2F01/C waste stream information sheet in 2016 UKRWI.

Table B.7 – Data for HLW disposal canister

Source: Derived Inventory for HLW [3]

	Materials			Empty weight	Payload	Displacement volume
	Copper	Cast Iron	Cast steel	(t)	(m^3)	(m^3)
Disposal Canister	36.59%	61.94%	1.48%	12.19	0.39	2.04

Table B.8 – Average specific radioactivity in waste, including only radionuclides for which characterisation factors are available

Source: Waste stream information sheets data from 2016 UYKRWI [322].

Waste	Nuclide (TBq/m³)										
	Be 10	**Cl 36**	**Cs 135**	**I 129**	**Np 237**	**Se 79**	**Sn 126**	**Tc 99**	**Th 229**	**U 233**	
Vitrified High Level Waste – Encapsulated	2.32E-06	4.79E-04	2.72E-02	1.53E-05	9.34E-03	2.91E-03	1.71E-02	6.66E-01	4.41E-09	4.79E-07	
	Cl36	**I129**	**Pb210**	**Pu238**	**Ra226**	**Tc99**	**Th229**	**Th230**	**U233**	**U234**	**U238**
AGR Cladding – Encapsulated	1.91E-05	2.54E-06	6.06E-12	1.09E-01	4.86E-11	1.21E-03	1.17E-11	1.70E-08	1.41E-09	1.26E-04	3.35E-05
BC and MEB crud – Encapsulated	3.78E-10	2.89E-04	1.28E-14	8.13E-14	1.64E-06	1.78E-14	9.09E-06	2.78E-12	1.70E-07	4.58E-08	
Centrifuge cake – Encapsulated	8.43E-04	6.32E-07	2.42E-12	-	1.54E-11	1.73E-01	3.36E-12	1.52E-05	5.25E-10	1.05E-03	1.85E-05
Plutonium Contaminated Material	-	-	-	-	-	-	-	-	-	-	-
AGR Graphite components	1.72E-05	-	-	-	-	-	-	-	-	-	-
AGR Stainless Steel components	6.18E-05	-	-	-	-	-	-	-	-	-	-
Maintenance Scrap – Encapsulated	-	-	-	-	-	-	-	-	-	-	-

Notes:
Specific activity refers to volume of conditioned waste (waste + conditioning material). In the case of encapsulated waste, specific activity refers to volume of waste + canister

Table B.9 – Inventory data for construction, decommissioning and operation of one geological disposal facility hosting the UK baseline inventory

Material	Amount		Source
Construction			
Concrete	5.60E+05	m^3	RWM's generic disposal design [319]
Reinforcement Steel	2.00E+03	t	RWM's generic disposal design [319]
Electricity	7.25E+08	kWh	Ecoinvent report No 6-VII [253]
Industrial machine	5.69E+07	kg	Ecoinvent report No 6-VII [253]
Diesel for building machine	7.04E+07	MJ	Ecoinvent report No 6-VII [253]
Building of facilities	1.76E+06	m^3	Ecoinvent report No 6-VII [253]
Decommissioning			
Bentonite	1.36E+10	kg	RWM's generic disposal design [319]
Crushed Rocks	2.56E+10	m^3	
Operation			
HLW/SF/Pu/HEU			
Waste	2.49E+04	m^3	RWM's generic disposal design [319]
Bentonite	4.09E+08	kg	
Crushed Rocks	4.19E+09	kg	
I-LLW			
Waste	4.73E+05	m^3	RWM's generic disposal design [319] And Nirex Reference Vault Backfill composition [353]
Ordinary Portland Cement	5.63E+08	kg	
Limestone flour	2.13E+08	kg	
Hydrated lime	6.19E+08	kg	
Water	7.69E+08	kg	

Table B.10 – Atmospheric discharges (Bq)

Reference: 2016 calendar year
Source: THORP environmental team

Source	Throughput*		H3	C14	Kr85	Sr90	Ru106	Sb125	I129	I131	Cs137	Pu 239+240	Pu241	Am241
THORP**	430	teU *	8.96E+06	2.54E+05	5.58E+10	1.73E+01	1.58E+02		8.18E+03	1.66E+02	7.81E+00	6.50E-01	1.73E+01	3.00E+00
FHP	430	teU	-	-	-	2.62E+06	-	1.24E+10	-	-	1.48E+07	-	-	-
WVP	38	dm^3 VHAL	-	1.54E+00	-	-	2.81E+00	-	9.02E-01	6.57E-01	-	-	-	-
HALES**	4190	m3 HAL	9.70E+03	-	8.73E+04	2.94E+10	-	3.34E+08	6.75E+07	-	-	-	-	-
WEP	1751	kg	-	1.01E+08	-	-	-	-	1.72E+06	-	-	-	-	-
Cladding	269	kg		1.55E+07	-	-	-	-	2.64E+05	-	-	-	-	-
Barium carbonate slurry	406	kg		1.59E+07	-	-	-	-	2.71E+05				-	-
Centrifuge cake	276	kg	-	1.47E+07	-	-	-	-	2.50E+05	-	-	-	-	-
MEB crud	255	kg	-	1.59E+07	-	-	-	-	2.71E+05	-	-	-	-	-
Maintenance scrap	276	kg	-	1.55E+07	-	-	-	-	2.64E+05	-	-	-	-	-

Notes:

*Reference throughput for THORP obtained from personal communications with THORP technical team; for FHP, WVP, HALES from plant-specific operational flowsheets; for WEP from 2016 UKRWI and scaled to annual production.

**Data refers to emission occurring at THORP stack and Analytical Service & Plutonium Finishing line (AS and PF&S) with a 50% allocation based on the number of plants contributing.

**HALES radioactive emissions are discharged through STP stack. Sample point name: HLWP.

Table B.11 – Sea water discharges (GBQ)

Reference: 2016 calendar year
Source: THORP environmental team

Source	Reference throughput*		H3	C14	Co60	Sr90	Zr+Nb95	Tc99	Ru106	I129	Cs134
THORP R&S	4.30E+02	teU	3.23E+01	-	2.57E+01	-	-	-	1.89E+01	-	4.52E+00
THORP DOG treatment	2.18E+05	m^3 caustic feed	5.33E+02	1.72E+02	-	-	-	-	-	3.13E+02	-
EARP (Concentrates)**	3.60E+03	SEC treated	5.75E+02	1.26E+02	-	9.15E+01	-	2.33E+01	1.23E+02	-	-
SETP			1.51E+06	4.45E+03	1.18E+01	5.79E+02	5.70E+01	-	1.73E+02	5.20E+01	4.77E+01
			(1.84%)	*(1.84%)*	*(1.84%)*	*(1.84%)*	*(1.84%)*	-	*(1.84%)*	*(1.84%)*	*(1.84%)*
THORP***	2.52E+05	m^3 LAE	2.78E+04	8.19E+01	2.17E-01	1.07E+01	1.05E+00	0.00E+00	3.18E+00	9.57E-01	8.78E-01
			(0.82%)	*(0.82%)*	*(0.82%)*	*(0.82%)*	*(0.82%)*	-	*(0.82%)*	*(0.82%)*	*(0.82%)*
HALES***	3.60E+03	m^3 LAE	1.23E+04	3.63E+01	9.64E-02	4.73E+00	4.66E-01	0.00E+00	1.41E+00	4.25E-01	3.90E-01

Notes:

*Reference throughput for THORP R&S obtained from personal communications with THORP technical team; for THORP DOG treatment, EARP, SETP (THORP and HALES) obtained from plant-specific operational flowsheets.

**Physical allocation based on number of streams. Allocation values is 50% as only 2 streams contribute: Magnox and THORP.

***Physical allocation based on radioactive contribution of each radionuclide of each stream to total feed to SETP.

Table B.12 – Sea water discharges (GBQ) – cont' d

Reference: 2016 calendar year
Source: THORP environmental team

Source	Reference throughput*		Cs137	Ce144	Np237	Pu238	Pu239+240	Pu241	Am241
THORP R&S	430	teU	3.05E+02	-	-	1.54E+00	2.54E+00	7.92E+01	-
THORP DOG treatment	2.18E+05	m3 of caustic feed	-	-	-	-	-	-	-
EARP (Concentrates)**	3.60E+03	SEC treated	4.82E+00	-	3.90E-01	1.50E-02	7.50E-02	5.45E-01	3.64E+00
SETP			1.41E+03	8.95E+01	2.93E+01	1.34E+01	2.54E+01	6.95E+02	1.51E+01
			(1.84%)	*1(.)84%*	*(10.78%)*	*1(0.78%)*	*(10.78%)*	*(1.84%)*	*(10.78%)*
THORP***	2.52E+05	m3 LAE	2.59E+01	1.65E+00	3.16E+00	1.44E+00	2.74E+00	1.28E+01	1.63E+00
			(0.82%)	*(0.82%)*	*(6.86%)*	*(6.86%)*	*(6.86%)*	*(0.82%)*	*(6.86%)*
HALES***	3.60E+03	m3 LAE	1.15E+01	7.31E-01	2.01E+00	9.19E-01	1.74E+00	5.68E+00	1.04E+00

Notes:
*Reference throughput for THORP R&S obtained from personal communications with THORP technical team; for THORP DOG treatment, EARP, SETP (THORP and HALES) obtained from plant-specific operational flowsheets.

Table B.13 – Electricity consumption data

Reference: 2015-16 financial year
Source: Sellafield energy champion

Plant	Consumption	Reference throughput		Allocation	Specific consumption
THORP	2.04E+08 MJ	4.30E+02	teU SNF Processed		4.74E+05 MJ/teU
SETP	4.80E+06 MJ	3.60E+03	m3 LAE from HALES to SETP	28.0%*	3.73E+02 MJ/m³
		2.52E+05	m3 LAE from THORP to SETP	0.4%*	7.61E-02 MJ/m³
EARP	1.05E+07 MJ	8.36E+00	m3 SEC from THORP treated	25.0%**	3.15E+05 MJ/m³
SIXEP	6.83E+06 MJ		Not considered		
STP	5.84E+06 MJ		Not considered		
HALES	1.96E+07 MJ	2.10E+02	m3 HAL to HALES (FY 15/16)		9.32E+04 MJ/m³
WVP	2.86E+07 MJ	2.04E+02	m3 HAL from HALES (FY 15/16)		1.40E+05 MJ/m³
WPEP	1.03E+07 MJ	8.90E+02	m3 SEC floc treated (FY 15/16)		1.16E+04 MJ/m³
WEP	1.91E+07 MJ	1.17E+05	kg All waste types treated (FY 15/16)		1.63E+02 MJ/kg
		2.69E+02	kg AGR cladding	15.4%	2.50E+01 MJ/kg
		4.06E+02	kg LWR cladding	23.4%	3.77E+01 MJ/kg
		2.76E+02	kg BC slurry	15.8%	2.57E+01 MJ/kg
		2.55E+02	kg Centrifuge slurry	14.6%	2.37E+01 MJ/kg
		2.76E+02	kg MEB crud	15.8%	2.57E+01 MJ/kg
		2.69E+02	kg Maintenance scrap	15.4%	2.50E+01 MJ/kg
WTC	*Assumed to have the same specific consumption of WEP*				1.63E+02 MJ/kg
tbd					1.63E+02 MJ/kg

Notes:
*Based on treated volumetric flow
**Based on number of flow types treated by EARP. 50% is for SEC/Bulks, and 50% for SEC/MAC

Table B.14 – Non-radioactive sea discharges

Reference: 2015-16 financial year
Source: Sellafield Environmental team

Substance	Release point to sea	Discharge	Alloc.*	Plant	Reference throughput		Alloc.**	Specific discharge	
Mercury	SETP, SIXEP, EARP, Laundry, Inactive Tank Farm Neutralising Pit, Thorp-C14 Removal Plant, Water Treatment	1.20E-01 kg	14.3%	SETP	3.60E+03	*m3 amount of LAE sent from HALES*	28%	1.33E-06	kg/m³
					2.52E+05	*m3 amount of LAE sent from THORP*	0.40%	2.72E-10	kg/m³
			14.3%	EARP	3.60E+03	m3 amount of SEC from THORP	25%	1.19E-06	kg/m³
			14.3%	THORP-C14	2.18E+05	m3 spent caustic processed	-	7.85E-08	kg/m³
Chromium	SIXEP, SETP, EARP	4.70E+01 kg	33.3%	SETP	3.60E+03	*m3 amount of LAE sent from HALES*	28%	1.22E-03	kg/m³
					2.52E+05	*m3 amount of LAE sent from THORP*	0.40%	2.49E-07	kg/m³
			33.3%	EARP	3.60E+03	m3 amount of SEC from THORP	25%	1.09E-03	kg/m³
N as NO2 and NO3	SETP, EARP	9.00E+05 kg	50%	SETP	3.60E+03	*m3 amount of LAE sent from HALES*	28%	3.50E+01	kg/m³
					2.52E+05	*m3 amount of LAE sent from THORP*	0.40%	7.14E-03	kg/m³
			50%		3.60E+03	m3 amount of SEC from THORP	25%	3.13E+01	kg/m³
N as NO2 and NO3	Thorp-C14 Removal Plant	4.50E+03 kg			2.18E+05	m3 spent caustic processed	-	2.06E-02	kg/m³
Glycol	SETP, SIXEP, EARP, Lagoon	5.30E+02 kg	25%	SETP	3.60E+03	*m3 amount of LAE sent from HALES*	28%	1.37E-02	kg/m³
					2.52E+05	*m3 amount of LAE sent from THORP*	0.40%	2.80E-06	kg/m³
			25%	EARP	3.60E+03	m3 amount of SEC from THORP	25%	1.23E-02	kg/m³

Notes:
*Allocation based on number of plants contributing
**Allocation based on volumetric flows

Table B.15 – Atmospheric discharges from Sellafield site obtained from "Monitoring Our Environment" report [51]

Nuclide	SL report
Americium (Am241) + Curium (Cm242)	1.00E-02
Antimony (Sb125)	8.80E+00
Carbon (C14)	3.40E+02
Caesium (Cs137)	1.50E-01
Hydrogen-3, tritium	9.50E+04
Iodine (I129)	1.20E+01
Iodine (I131)	3.50E-01
Krypton (Kr85)	5.60E+07
Plutonium (Pu alpha)	2.00E-02
Plutonium (Pu241)	2.00E-01
Radon (Rn222)	4.30E+01
Ruthenium (Ru 106)	7.80E-01
Strontium (Sr90)	3.00E-02
Total	**5.61E+07**

Table B.16 – Coastal water discharges from Sellafield site obtained from "Monitoring Our Environment" report [51]

Nuclide	SL report
Americium (Am241)	2.00E+01
Antimony (Sb125)	1.00E+02
Carbon (C14)	4.70E+03
Cerium (Ce144)	1.60E+02
Cesium (Cs134)	6.00E+01
Cesium (Cs137)	2.60E+03
Cobalt (Co60)	5.00E+01
Curium (Cm243-244)	2.00E+00
Europium-152	2.00E+01
Europium-154	1.00E+01
Europium-155	2.00E+01
Hydrogen-3, tritium	1.30E+06
Iodine (I129)	3.60E+02
Neptunium (Np237)	4.00E+01
Plutonium (Pu238)	-
Plutonium (Pu239-240)	1.50E+02
Plutonium (Pu241)	2.90E+03
Ruthenium (Ru106)	1.10E+03
Strontium (Sr90)	1.60E+03
Technetium (Tc99)	1.30E+03
Zinc-65	1.00E+01
Zirconium (Zr95) + Niobium (Ni95)	8.00E+01
Total	**1.32E+06**

B.2 LCIA Results

Table B.17 – Life cycle impact scores for the UK "nominal" Twice Through Cycle

	TOTAL	THORP	WTP	GDF
Acidification midpoint [Mole of H+ eq.]	2.07E+03	4.43E+02	1.04E+03	5.82E+02
Climate change [kg CO_2 eq.]	2.38E+05	1.15E+05	4.16E+04	8.17E+04
Ecotoxicity, freshwater [CTUe]	1.08E+07	5.49E+05	7.93E+06	2.29E+06
Eutrophication freshwater midpoint [kg P eq]	2.73E+02	1.86E+01	1.98E+02	5.65E+01
Eutrophication marine midpoint [kg N-Equiv.]	8.72E+02	6.43E+02	1.04E+02	1.25E+02
Eutrophication [Mole of N eq.]	3.61E+03	1.44E+03	8.36E+02	1.33E+03
Human toxicity midpoint, cancer effects [CTUh	3.87E-02	1.70E-03	2.38E-02	1.32E-02
Human toxicity midpoint, non-cancer effects [CTUh]	5.00E-01	3.09E-02	3.77E-01	9.20E-02
USErad (equiv) [Bq U235 air-equiv]	1.88E+09	1.61E+09	2.66E+08	1.77E+06
USErad – GDF (equiv) [Bq U238 ILLW-equiv]	6.70E+10	0.00E+00	0.00E+00	6.70E+10
Ozone depletion midpoint [kg CFC-11 eq]	3.21E-02	1.73E-02	7.43E-03	7.38E-03
Particulate matter/Respiratory inorganics midpoint [kg PM2,5-Equiv.]	1.95E+02	4.24E+01	9.10E+01	6.18E+01
Photochemical ozone formation midpoint, human health [kg NMVOC]	1.03E+03	3.82E+02	2.55E+02	3.92E+02
Resource depletion, mineral, fossils and renewables, midpoint [kg Sb-Equiv.]	4.85E+01	2.63E+01	1.62E+01	6.02E+00
Resource depletion water, midpoint [m^3 eq.]	1.65E+03	4.64E+02	3.73E+02	8.11E+02

Appendix C

C.1 Inventory Data

Table C.1 – Composition of titanium-based ceramic for plutonium immobilisation

Source: Derived Inventory for U and Pu [273]

Oxide	Composition (% by mass)
PuO2	11.9
UO2	23.7
HfO2	10.6
GD2O3	7.9
CaO	10.0
TiO2	35.9

Table C.2 – Properties of disposal canister for plutonium. Mass refers to amount contained in the disposal canister

Source: Derived Inventory for U and Pu [273]

Ceramic		
	Mass (20 x 28 pucks)	0.280 t
(PuO2)	Mass (20 x 28 pucks)	0.0333 t
(Pu)	Mass (20 x 28 pucks)	0.0294 t
Stainless steel cans		
	Material	SS 316
	Payload	20 pucks
	Mass (28 cans)	0.1 t
Borosilicate glass		
	Mass	1.3 t
Steel canister		
	Payload	28 cans
	Mass (1 canister)	2.3 t
Disposal canister		
	Displacement volume	2.04 m^3
	Payload	1 steel canister
	Mass (1 canister)	9.57 t
Copper	Mass (1 canister)	4.46 t
Cast Iron	Mass (1 canister)	4.93 t
Cast steel	Mass (1 canister)	0.18 t

Table C.3 – Properties of disposal canister for depleted uranium (Repu and tails). Mass refers to the amount contained in the disposal canister

Source: Derived inventory for U and Pu [273]

Uranium Oxide		
	Mass	1.14 t
U	Mass	0.967 t
Encapsulating grout		
	Mass*	0.72 t
Cement**		70.5%
Water***		29.5%
Steel drum		
	Material	SS 316
	Displacement volume	0.571 m^3
	Mass	0.13 t

Notes:

*Includes conditioning and capping grout.

**Composition is 1:1 PFA/OPC for conditioning and 3:1 PFA/OPC for capping grout. In this study we assume to use BFS rather than PFA, as due to this being not available in Ecoinvent.

***Water composition of capping grout was not reported and has been assumed to be 25%.

Table C.4 – Properties of disposal canister for AGR UNF. Mass refers to amount contained in the disposal canister

Source: Derived inventory for HLW and SNF [272]

AGR fuel		
	Mass (8 slotted cans)	1.03 tHM
Slotted can		
	Material	SS (not specified)
	Mass (8 slotted cans)	98.4 kg
Disposal canister		
	Displacement volume	1.59 m^3
	Payload	8 slotted cans
	Mass (1 canister)	8.83 t
Copper	*Mass (1 canister)*	*3.62 t*
Cast Iron	*Mass (1 canister)*	*5.03 t*
Cast steel	*Mass (1 canister)*	*0.18 t*

Table C.5 – Average specific radioactivity in waste, including only radionuclides for which characterisation factors are available

Source: Derived inventory for Pu and U [273]*, and HLW and SNF* [272]

Nuclide (TBq/m³)	Pu	RepU	DepU	AGR
Cl36	-	-	-	2.63E-03
Cs135	-	-	-	1.34E-01
I129	-	-	-	4.52E-03
Ni59	-	-	-	1.38E-03
Np237	1.44E-05	-	-	-
Pa231	1.93E-12	-	-	-
Pb210	-	9.84E-14	-	-
Ra226	1.06E-12	9.57E-12	2.18E-09	3.14E-08
Se79	-	-	-	4.03E-03
Sn126	-	-	-	1.21E-01
Th230		4.43E-08	-	-
U233	2.54E-11	-	-	-
U234	-	4.81E-03	3.71E-03	-
U235	-	4.78E-05	3.40E-04	-
U236	1.14E-05	7.66E-04	-	-
U238	-	1.27E-03	2.10E-02	-

Table C.6 – Transport data

Source: Environmental Life Cycle Assessment of the Nuclear Fuel Cycle Thesis [220]

Type	From	to	Mode	Distance
	ISL mine in Kazakhstan*	Novorossiysk	Rail	3580 km
Yellowcake (U_3O_8)	Novorossiysk	Generic shipping port in UK**	Sea	6580 km
	Generic shipping port in UK	Springfields	Road and rail***	345 km
Uranium hexafluoride (UF_6) (Natural)	Springfields	Capenhurst	Road	95 km
Uranium hexafluoride (UF_6) (Enriched)	Capenhurst	Springfields	Road	95 km
Uranium hexafluoride (UF_6) (Depleted)	Capenhurst	Sellafield	Road	225 km
RepU (UO_3)	Sellafield	Springfields	Road	145 km
Packaged Pu (PuO_2), RepU (UO_3), UNF	Sellafield	GDF location****	Rail	350 km

Notes:
Transportation for yellowcake from ISL mine to UK has been estimated from www.searates.com
*Assumed to be located at the centre of Kazakhstan.
** Average of the 4 main shipping ports in the UK: Liverpool, Southampton, Fekuxstiwer and Tlbury
*** Assume 2/3 road and 1/3 rail
****Centroid of 7 zones covering England and Wales, in which the GDF is assumed to be placed [318]

Table C.7 – Additional transport data for yellowcake from other uranium mines

Mine	From	to	Mode	Distance	
Underground mine, CA	McArthur river/Cigar Lake	Winnipeg	Road	1500	km
	Winnipeg	Generic shipping port in UK*	Sea	7020	km
Open pit/ Ranger Uranium mine, AU	Ranger	Darwin port	Road	255	km
	Darwin port	Generic shipping port in UK*	Sea	18300	km
ISL mine, AU	Four mile	Adelaide	Road	690	km
	Adelaide	Generic shipping port in UK*	Sea	19720	km

Notes:
Transportation distances and modes have been estimated from *searates* (www.searates.com)
* Average of the 4 main shipping ports in the UK: Liverpool, Southampton, Fekuxstiwer and Tlbury.

C.2 Calculation of MOX mixing ratio

The mixing ratios have been obtained from the WISE uranium project [354], according to the following equation:

$$M_{Pu} \times C_{Puf-U235eq} + M_U \times C_{U235} = M_{MOx}\left(C_{U235eq} + C_{U236} \times \alpha\right)$$

Where:

M_{Pu}, M_U and M_{Mox}	are the mass of Pu, U and MOX in ton of heavy metals (tHM);
$C_{Puf-U235eq}$	is the concentration of fissile Pu that produced the same energy in the reactor as U235, and is equal to 1.5 gPu/gU [354]. The concentration of fissile plutonium is taken to be 66% per ton of heavy metals of plutonium [354];
C_{U235}	is the concentration of U235;
C_{U235eq}	is the concentration of fissile plutonium and uranium 235 in MOX, expressed as weight-percent equivalent of U-235;
C_{U236}	is the concentration of U235 in MOX;
α	is the excess concentration of U-235 required to offset the neutron-absorbing effect of U236 present in MOX. This is taken to be equal to 0.25 [354].

C.3 GDF footprint

Calculation of GDFs' footprint for each scenario and approach are based on the RWM (Radioactive Waste Management Ltd) "generic Post-Closure Safety Assessment" [267]. This report estimated the footprint of the future UK national GDF based on the amount of nuclear waste to be disposed (reported in [271]–[273]). From the area required by each type of waste and their amount to be disposed, a specific area per volume of disposed waste has been calculated. Finally, it has been assumed that roadways and support area are dependent on the footprint of the GDF, rather than the volume of disposed wastes; according to this, a specific roadways and support are has been calculated. Data is reported in Table C.8.

Table C.8 – GDF specific area per amount of disposed waste and specific roadways and support area per area of GDF

Source: Generic Post-Closure Safety Assessment [267]*, and derived inventories for HLW* [272]*, ILW and LLW* [271]*, and U and Pu* [273]

	Area (m^2)	**Disposed waste** (m^3)	**Area per amount of disposed waste** (m^2/m^3)
HLW/SF/Pu/HEU deposition tunnels	2246000	24954	90.01
UILW/SILW/LLW vaults	435000	472724	0.92
		Roadways and support area per m^2 of area of GDF	
Roadways and support area	2910000	1.09	

C.4 LCIA Results

Table C.9 – Environmental impact of reprocessing scenarios and direct disposal for a marginal technology corresponding to a generic underground mine located in Canada. The colour scale goes from red: highest, to green: lowest.

		Reprocessing				Direct disposal
		S1	S2	S3	S4	
A	[Mole of H+ eq.]	2.68E+03 *(8%)*	-2.66E+03 *(-207%)*	-3.60E+03 *(-245%)*	-3.63E+03 *(-246%)*	2.48E+03
CC	[kg CO2 eq.]	3.01E+05 *(116%)*	-1.64E+05 *(-218%)*	-2.62E+05 *(-288%)*	-2.62E+05 *(-288%)*	1.39E+05
ECf	[CTUe]	1.41E+07 *(-19%)*	5.09E+06 *(-71%)*	3.96E+06 *(-77%)*	3.90E+06 *(-78%)*	1.75E+07
Ef	[kg P eq.]	3.49E+02 *(-20%)*	1.70E+02 *(-61%)*	1.47E+02 *(-66%)*	1.47E+02 *(-66%)*	4.34E+02
Em	[kg N eq.]	4.45E+02 *(40%)*	-1.37E+03 *(-531%)*	-1.69E+03 *(-630%)*	-1.71E+03 *(-635%)*	3.19E+02
Et	[Mole of N eq.]	4.39E+03 *(51%)*	-1.19E+04 *(-511%)*	-1.48E+04 *(-610%)*	-1.49E+04 *(-615%)*	2.90E+03
HT-c	[CTUh]	5.06E-02 *(-17%)*	-8.25E-03 *(-114%)*	-1.70E-02 *(-128%)*	-1.77E-02 *(-129%)*	6.08E-02
HT-nc	[CTUh]	6.35E-01 *(-22%)*	3.16E-01 *(-61%)*	2.80E-01 *(-65%)*	2.79E-01 *(-66%)*	8.12E-01
IR	[Bq U235 air eq.]	2.34E+09 *(69740%)*	-1.26E+10 *(-375762%)*	-1.50E+10 *(-449259%)*	-1.52E+10 *(-454707%)*	3.35E+06
IRw	[Bq U238 ILLW eq.]	7.12E+10 *(-10%)*	3.77E+10 *(-52%)*	3.17E+10 *(-60%)*	3.13E+10 *(-60%)*	7.88E+10
OD	[kg CFC-11 eq.]	3.85E-02 *(218%)*	-2.42E-02 *(-300%)*	-3.66E-02 *(-403%)*	-3.70E-02 *(-405%)*	1.21E-02
PM/RI	[kg PM2.5 eq.]	2.47E+02 *(7%)*	-3.84E+02 *(-266%)*	-4.94E+02 *(-314%)*	-4.98E+02 *(-316%)*	2.31E+02
POF	[kg NMVOC]	1.28E+03 *(46%)*	-3.37E+03 *(-485%)*	-4.19E+03 *(-579%)*	-4.23E+03 *(-584%)*	8.74E+02
RDm	[kg Sb eq.]	5.58E+01 *(54%)*	-7.64E+02 *(-2216%)*	-9.00E+02 *(-2593%)*	-9.09E+02 *(-2619%)*	3.61E+01
RDw	[m^3 eq.]	2.01E+03 *(78%)*	-3.22E+03 *(-385%)*	-4.38E+03 *(-488%)*	-4.36E+03 *(-485%)*	1.13E+03

Table C.10 – Environmental impact of reprocessing scenarios and direct disposal for a marginal technology corresponding to a generic In-Situ Leaching (ISL) mine located in Australia. The colour scale goes from red: highest, to green: lowest.

		Reprocessing				Direct disposal
		S1	S2	S3	S4	
A	[Mole of H+ eq.]	2.68E+03 *(8%)*	-2.47E+03 *(-199%)*	-3.94E+03 *(-259%)*	-3.97E+03 *(-260%)*	2.68E+03
CC	[kg CO2 eq.]	3.01E+05 *(116%)*	-1.88E+05 *(-235%)*	-3.18E+05 *(-328%)*	-3.18E+05 *(-329%)*	1.39E+05
ECf	[CTUe]	1.42E+07 *(-19%)*	5.46E+06 *(-69%)*	6.40E+05 *(-96%)*	5.49E+05 *(-97%)*	1.75E+07
Ef	[kg P eq.]	3.58E+02 *(-18%)*	-3.86E+01 *(-109%)*	-1.90E+02 *(-144%)*	-1.94E+02 *(-145%)*	4.34E+02
Em	[kg N eq.]	9.63E+02 *(202%)*	-1.77E+04 *(-5657%)*	-2.09E+04 *(-6649%)*	-2.11E+04 *(-6719%)*	3.19E+02
Et	[Mole of N eq.]	4.49E+03 *(55%)*	-1.50E+04 *(-618%)*	-1.91E+04 *(-758%)*	-1.93E+04 *(-764%)*	2.90E+03
HT-c	[CTUh]	5.01E-02 *(-18%)*	1.40E-02 *(-77%)*	-2.75E-03 *(-105%)*	-3.18E-03 *(-105%)*	6.08E-02
HT-nc	[CTUh]	6.56E-01 *(-19%)*	-2.12E-01 *(-126%)*	-5.11E-01 *(-163%)*	-5.20E-01 *(-164%)*	8.12E-01
IR	[Bq U235 air eq.]	1.88E+09 *(56008%)*	1.83E+09 *(54624%)*	1.82E+09 *(54214%)*	1.82E+09 *(54247%)*	3.35E+06
IRw	[Bq U238 ILLW eq.]	7.12E+10 *(-10%)*	4.09E+10 *(-48%)*	3.49E+10 *(-56%)*	3.45E+10 *(-56%)*	7.88E+10
OD	[kg CFC-11 eq.]	3.88E-02 *(221%)*	-3.70E-02 *(-406%)*	-5.37E-02 *(-544%)*	-5.41E-02 *(-547%)*	1.21E-02
PM/RI	[kg PM2.5 eq.]	2.50E+02 *(8%)*	-4.37E+02 *(-289%)*	-6.07E+02 *(-363%)*	-6.12E+02 *(-365%)*	2.31E+02
POF	[kg NMVOC]	1.29E+03 *(48%)*	-3.97E+03 *(-554%)*	-5.09E+03 *(-682%)*	-5.13E+03 *(-687%)*	8.74E+02
RDm	[kg Sb eq.]	5.69E+01 *(57%)*	-7.94E+02 *(-2298%)*	-9.44E+02 *(-2714%)*	-9.53E+02 *(-2741%)*	3.61E+01
RDw	[m^3 eq.]	2.11E+03 *(87%)*	-6.26E+03 *(-654%)*	-8.18E+03 *(-824%)*	-8.20E+03 *(-825%)*	1.13E+03

Table C.11 – Environmental impact of reprocessing scenarios and direct disposal for a marginal technology corresponding to a generic open pit mine located in Australia. The colour scale goes from red: highest, to green: lowest.

		Reprocessing				Direct disposal
		S1	S2	S3	S4	
A	[Mole of H+ eq.]	2.67E+03 *(8%)*	-2.44E+03 *(-199%)*	-3.35E+03 *(-235%)*	-3.37E+03 *(-236%)*	2.48E+03
CC	[kg CO2 eq.]	3.01E+05 *(116%)*	-1.86E+05 *(-234%)*	-2.87E+05 *(-307%)*	-2.88E+05 *(-307%)*	1.39E+05
ECf	[CTUe]	1.41E+07 *(-19%)*	4.35E+06 *(-75%)*	3.10E+06 *(-82%)*	3.03E+06 *(-83%)*	1.75E+07
Ef	[kg P eq.]	3.50E+02 *(-19%)*	1.47E+02 *(-66%)*	1.19E+02 *(-72%)*	1.19E+02 *(-73%)*	4.34E+02
Em	[kg N eq.]	4.28E+02 *(34%)*	-8.36E+02 *(-362%)*	-1.06E+03 *(-432%)*	-1.07E+03 *(-435%)*	3.19E+02
Et	[Mole of N eq.]	4.26E+03 *(47%)*	-7.86E+03 *(-371%)*	-1.00E+04 *(-446%)*	-1.01E+04 *(-449%)*	2.90E+03
HT-c	[CTUh]	5.06E-02 *(-17%)*	-7.94E-03 *(-113%)*	-1.67E-02 *(-127%)*	-1.73E-02 *(-128%)*	6.08E-02
HT-nc	[CTUh]	6.35E-01 *(-22%)*	3.08E-01 *(-62%)*	2.71E-01 *(-67%)*	2.70E-01 *(-67%)*	8.12E-01
IR	[Bq U235 air eq.]	2.91E+09 *(86754%)*	-3.07E+10 *(-916623%)*	-3.62E+10 *(-1081966%)*	-3.67E+10 *(-1094298%)*	3.35E+06
IRw	[Bq U238 ILLW eq.]	7.12E+10 *(-10%)*	3.77E+10 *(-52%)*	3.17E+10 *(-60%)*	3.13E+10 *(-60%)*	7.88E+10
OD	[kg CFC-11 eq.]	3.81E-02 *(215%)*	-1.04E-02 *(-186%)*	-2.04E-02 *(-268%)*	-2.05E-02 *(-270%)*	1.21E-02
PM/RI	[kg PM2.5 eq.]	2.44E+02 *(6%)*	-2.88E+02 *(-225%)*	-3.82E+02 *(-265%)*	-3.85E+02 *(-267%)*	2.31E+02
POF	[kg NMVOC]	1.24E+03 *(42%)*	-2.32E+03 *(-365%)*	-2.96E+03 *(-439%)*	-2.99E+03 *(-442%)*	8.74E+02
RDm	[kg Sb eq.]	5.58E+01 *(54%)*	-7.64E+02 *(-2216%)*	-9.00E+02 *(-2593)%*	-9.09E+02 *(-2619%)*	3.61E+01
RDw	[m^3 eq.]	*2.11E+03 (87%)*	*-6.33E+03 (-660%)*	*-8.02E+03 (-809%)*	*-8.04E+03 (-810%)*	1.13E+03

Table C.12 – Environmental impact of reprocessing scenarios and direct disposal for a marginal technology corresponding to the Ranger mine in Australia. The colour scale goes from red: highest, to green: lowest.

		Reprocessing				Direct disposal
		S1	S2	S3	S4	
A	[Mole of H+ eq.]	2.58E+03 *(4%)*	4.33E+02 *(-83%)*	1.78E+01 *(-99%)*	2.74E+01 *(-99%)*	2.48E+03
CC	[kg CO2 eq.]	2.96E+05 *(113%)*	-2.00E+04 *(-114%)*	-9.29E+04 *(-167%)*	-9.15E+04 *(-166%)*	1.39E+05
ECf	[CTUe]	1.40E+07 (-20%)	9.31E+06 (-47%)	8.90E+06 (-49%)	8.89E+06 (-49%)	1.75E+07
Ef	[kg P eq.]	3.47E+02 *(-20%)*	2.38E+02 *(-45%)*	2.26E+02 *(-48%)*	2.27E+02 *(-48%)*	4.34E+02
Em	[kg N eq.]	3.99E+02 *(25%)*	9.32E+01 *(-71%)*	2.65E+01 *(-92%)*	2.86E+01 *(-91%)*	3.19E+02
Et	[Mole of N eq.]	3.99E+03 *(37%)*	7.55E+02 *(-74%)*	3.22E+01 *(-99%)*	5.38E+01 *(-98%)*	2.90E+03
HT-c	[CTUh]	4.98E-02 *(-18%)*	1.79E-02 *(-71%)*	1.36E-02 *(-78%)*	1.33E-02 *(-78%)*	6.08E-02
HT-nc	[CTUh]	6.31E-01 *(-22%)*	4.42E-01 *(-46%)*	4.27E-01 *(-47%)*	4.27E-01 *(-47%)*	8.12E-01
IR	[Bq U235 air eq.]	1.88E+09 *(56008%)*	1.83E+09 *(54534%)*	1.82E+09 *(54140%)*	1.82E+09 *(54172%)*	3.35E+06
IRw	[Bq U238 ILLW eq.]	7.12E+10 *(-10%)*	3.77E+10 *(-52%)*	3.17E+10 *(-60%)*	3.13E+10 *(-60%)*	7.88E+10
OD	[kg CFC-11 eq.]	3.73E-02 *(208%)*	1.29E-02 *(7%)*	6.90E-03 *(-43%)*	7.03E-03 *(-42%)*	1.21E-02
PM/RI	[kg PM2.5 eq.]	2.33E+02 *(1%)*	5.79E+01 *(-75%)*	2.28E+01 *(-90%)*	2.41E+01 *(-90%)*	2.31E+02
POF	[kg NMVOC]	1.16E+03 *(33%)*	2.02E+02 *(-77%)*	-1.30E+01 *(-101%)*	-6.79E+00 *(-101%)*	8.74E+02
RDm	[kg Sb eq.]	5.29E+01 *(46%)*	-6.72E+02 *(-1960%)*	-7.92E+02 *(-2294%)*	-8.00E+02 *-(2317%)*	3.61E+01
RDw	[m^3 eq.]	1.95E+03 *(72%)*	-1.22E+03 *(-208%)*	-2.04E+03 *(-281%)*	-1.99E+03 *(-276%)*	1.13E+03

List of publications

Paulillo, A., Clift, R., Dodds, J., Milliken, A., Palethorpe, S., Lettieri, P., 2018. Radiological Impact Assessment for Life Cycle Assessment studies: A Review and Possible Ways Forward. Environ. Rev. doi:10.1139/er-2018-0004. (In press.)

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