Choosing Your Nuclear Fuel Cycle: A Life Cycle Assessment Perspective – 16425

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ABSTRACT
Nuclear waste management has always been a much debated topic and a key driver for decisions within the nuclear industry. A standardised global approach has not yet been developed and at present many countries are re-considering their position. In the UK the reprocessing of Spent Nuclear Fuel is projected to stop, but no clear indication about the future approach has been given. This contribution sets out the early stages of an approach to compare the possible alternatives based on Life Cycle Assessment (LCA), which is widely used in other sectors to assess the overall environmental burdens of a product or a service across the whole life cycle – i.e. from cradle to grave. The LCA may serve to support decision-making processes within the nuclear industry and, provided that it is used in an open and transparent way, to improve public attitudes towards nuclear energy. To date few LCA studies within the nuclear industry have been carried out, the main issue being the lack of a standardised methodology to evaluate the impacts of radionuclides. The present study proposes a new framework for this purpose, and demonstrates its application in an LCA study of the current UK approach to the management of spent nuclear fuel based on specific data from plants at Sellafield. The final results of the study will show the overall environmental footprint of the reprocessing of spent nuclear fuel, in terms of both radiological and non-radiological impacts. A "hot spot" analysis will also be performed to highlight the critical processing steps.

INTRODUCTION
In the modern industrial economy, energy production and use has the greatest impact on the environment of any human activity. Furthermore, as the United Nations (UN) estimates, world population is expected to increase up to eight billion by 2020 [1] and energy consumption to grow by 53% by 2040 [2]. Consequently, there is an urgent need to develop sustainable strategies for energy supply, while minimising environmental impacts. In the 1987 report of the World Commission on Environment and Development, sustainable development was defined as "… development that meets the needs of the present without compromising the ability of future generations to meet their own needs". Within the goal of sustainability there are three main areas that are generally considered relevant in decision-making as practical constraints on

human activities [3]: Environmental, Economic and Societal. Energy generation has links with all these areas, because energy services are essential for economic and social development. Therefore, as energy consumption is predicted to grow, its environmental impact will have to be controlled and alleviated in order to achieve sustainability goals. The role of nuclear energy is central as a source of low-carbon dispatchable or base-load electricity [4].

In 2013, nuclear energy accounted for 17% of UK, 24% of European and 11% of worldwide electricity supply [2] - the lowest value since 1982 [5], mainly due to the 2008 economic crisis and the Fukushima-Daichii accident which led to a total shutdown of Japan’s nuclear reactors. However, nuclear energy seems to be on the rise again: according to the World Nuclear Association [6], there are currently 65 nuclear reactors under construction around the world. This figure includes the first plants announced in the 21st century for the UK and USA.

One of the biggest challenges facing the expansion of nuclear energy is to find a sustainable approach for the management of solid nuclear wastes. For the purpose of this paper, two main approaches may be identified: The direct disposal of spent nuclear fuel assemblies (i.e. open fuel cycle) and The reprocessing of spent nuclear fuel with the aim of manufacturing fuel to be recycled back into the Nuclear Power Plants (NPPs) (i.e. closed fuel cycle). In the open fuel cycle the spent fuel can be “dry stored”, generally on site at the location of the NPP, or “wet stored” in a Spent Fuel Storage Pond, generally located at the reprocessing plant, pending final disposal. In the closed fuel cycle, on the other hand, the spent fuel assemblies are reprocessed: fissile products are separated from fission products and used to manufacture fuel for further use. In both cases, waste is processed into a final manageable form, either containing spent fuel assemblies or High (HLW), Intermediate (ILW) or Low Level Waste (LLW); notably HLW and ILW are together known as Higher Active Wastes (HAWs) An internationally accepted approach for disposal of HAW (by its very nature in a solid form at this stage) has yet to be agreed, but many countries are planning to dispose of them in deep repositories built several hundred meters underground in a geologically stable environment. The eventual approach to be taken in the UK has not been finalised. The current UK approach involves reprocessing most (but not all) of the UK’s spent nuclear fuel at the Sellafield site. Spent fuel from the UK’s Magnox reactors is reprocessed in the Magnox Reprocessing Plant, due to be closed at the end of 2020. The spent reactor fuel from the UK’s Advanced Gas-Cooled Reactors (AGR’s) is sent to the Thermal Oxide Reprocessing Plant (THORP), due to be closed at the end of 2018. It is the intention of the Nuclear Decommissioning Authority that future arisings from nuclear generation in the UK will be stored without intermediate reprocessing pending final processing and disposal in the Geological Disposal Facility (GDF). After the failure of the 2013 consultation exercise [7], the process to decide on siting a repository has been reviewed and restarted. According to the timeline set up by the revised siting 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 [7]. Thus the UK is facing a number of important decisions over the future of its nuclear energy industry.

Life Cycle Assessment (LCA) is a tool that could be used to compare alternative strategic options within the nuclear industry by evaluating their environmental performances. The use of LCA methodology has spread considerably all over the world during the last decade. The results of an LCA study of the nuclear cycle might be used to support the decision making-process within the nuclear industry and, provided that it is used in an open and transparent way, to improve public attitudes towards the nuclear energy. Until now, few LCA studies of the nuclear cycle have been reported, the main obstacle being the 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 nor from nuclear waste.

The goal of the present study is twofold: firstly to introduce a new approach for assessing the impact of radionuclide releases both as direct discharges (i.e. liquid and aerial) and from management of solid wastes in a GDF within the LCIA framework; and secondly to demonstrate the approach by evaluating the life cycle environmental impact of the UK approach to the management of spent nuclear fuel (i.e. closed cycle). Several environmental impact categories will be analysed and a “hot spot” analysis will be performed to identify the more polluting sections of the process. A risk-based methodology, developed by Solberg-Johansen [8] and modified here to use more recent data, is adopted to assess the environmental impact of releases of radionuclides from operation and management of solid nuclear waste facilities. Particular emphasis is given to the difference in terms of impact between operation and waste management. Results of the study will show the overall environmental footprint of the UK reprocessing step of spent nuclear fuel in terms of both radiological and non-radiological impacts.

LIFE CYCLE METHODOLOGY

Life cycle assessment is one of the most developed and widely used environmental assessment tools for comparing alternative technologies when the location of the activity is already defined [9]. LCA identifies and quantifies all the energy and materials used and the emissions and waste generated over the complete supply chain (i.e. life cycles) of goods and services. Moreover, it helps in determining the “hot spots” in the system, i.e. those activities that have the most significant environmental impacts and should be improved in the first instance, thus enabling identification of more environmentally sustainable options [10]. LCA belongs to a whole family of environmental assessment tools, some more focused on physical metrics (Environmental Risk Assessment, Environmental Impact Assessment) and others more focused on economic metrics (Cost-benefit Analysis), but differs from them in the way system boundaries are drawn. LCA encompasses the whole life cycle of a product, which extends from the extraction and processing of raw materials through manufacture, delivery and use to waste management; for this reason this approach is often termed as 'cradle to grave' assessment. The LCA structure and methodology has been applied in standardised [11] form since 1997. It is applied into four successive phases, shown schematically in Figure 1; the process is iterative in that earlier phases may be revisited in the light of the outcome of later phases.

Goal and Scope Definition: It is essential to define unambiguously the purpose or the intended application of the study and what decision is to be informed by the results. LCA may be applied to a single process in order to identify the stages in the life cycle with the greatest impact ("*hot spot" analysis)* or may be carried out to compare alternative ways of delivering a specified product or service. The basis for comparison is generally termed as the *functional unit* whereas the scope of the study is expressed in terms of the *system boundary*.

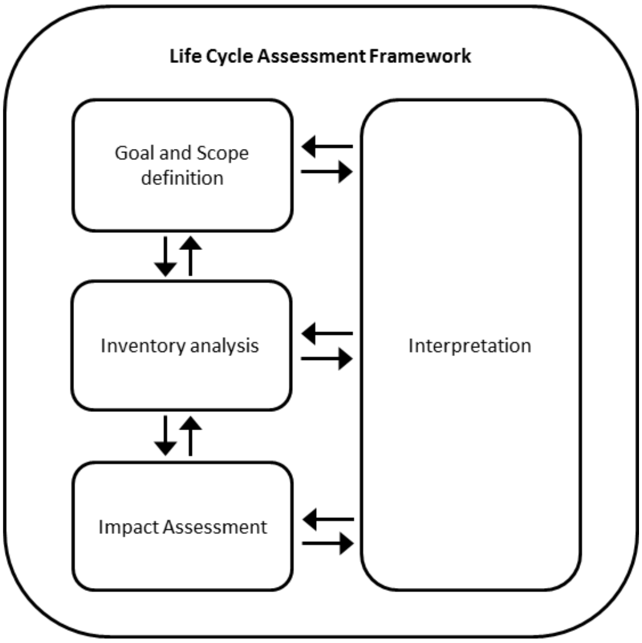

Figure 1: Phases in Life Cycle Assessment (after ISO 144040 [11]**)**

Inventory Analysis: In this stage, all emissions, wastes and use of resources over the life cycle per functional unit are identified and quantified. The complete set of burdens per Functional Unit constitutes the *Inventory Table.* For multiple-output processes, it is necessary to find a rational basis for allocating the environmental burdens between different outputs. The approaches available for allocation are specified in ISO 14041 [11], where system expansion [12] is recommended as the preferred approach.

Impact Assessment: The principal feature of the analysis or comparison is rarely clear from the inventory alone. According to the ISO standard [11], Life Cycle Impact Assessment phase aims at understanding and evaluating the magnitude and significance of the environmental impacts of the product system. Once a set of significant impact categories are selected, the inventory data are assigned to the categories (*classification*) and then the potential contribution of each input and output to different impact categories is calculated (*characterization*).

Interpretation: In this phase, the results of either the Data Inventory or the Impact Assessment or both are combined to address the goal and scope of the study. The results are used to makes recommendation for environmental improvements or as input to some form of decision process.

Following the methodological approach of Clift et al. [9] for Integrated Waste Management (IWM), a pragmatic distinction is made between Foreground and Background, considering the former as 'the set of processes whose selection or mode of operation is affected directly by decisions based on the study' and the latter as 'all other processes which interact with the Foreground, usually by supplying or receiving material or energy'. The burdens evaluated here are considered within two categories [9]: direct burdens, associated with the use phase of the process/service; and indirect burdens, due to upstream and downstream processes (e.g. energy provision for electricity or diesel for transportation). Following conventional practices [13], secondary data for the indirect burdens are taken as the averages for the Background system, while primary data are used for the Foreground operations.

Currently more than thirty software packages exist to perform LCA analysis, with differing scope and capacity: some are specific for certain applications, while others have been directly developed by industrial organisations [14]. In this study GaBi 6 has been used [15]. It contains databases developed by PE International and incorporates industry organisations' databases (e.g. Plastics Europe, Aluminium producers, etc.) and also regional and national databases (e.g. Ecoinvent, Japan database, US database, etc.).

Functional unit and system boundary

The aim of the present study is the application of the LCA methodology to the management of Spent Nuclear Fuel. The functional unit chosen is the management of the amount of LWR and AGR spent fuel assemblies which produced 1 TJ of electricity. If considering a typical fuel irradiation of 40GWd/Te Uranium and a conversion efficiency (thermal to electric) of 1/3 [16], such amount corresponds to 0.87 kgU/TJ. The foreground system data used is wherever possible site-specific; otherwise, average data from the literature, specific data-sets, and general models is considered. The background data used has been taken from the Ecoinvent data-set [17] and consists of averaged values referring to the UK or Europe. The transportation of SNF from the Nuclear Power Plants (NPPs) to the reprocessing plant and from the reprocessing plant to the GDF or the MOX fuel manufacturing plant has not been considered. The valuable output from the process is the MOX fuel returned for further use.

Life Cycle Impact categories

As mentioned above, in the impact assessment phase the emissions and inputs, quantified in the inventory phase, are translated into a smaller number of impacts. This study focuses on 11 impact categories among the conventional ones - showed in Table I - which are considered most significant for the purpose of this paper.

TABLE I – Impact categories and indicators used in this study

Impact category	Impact indicator	Acronym	Units
Acidification	Acidification Potential	AP	kg SO_2 eq
Climate change	Global Warming Potential	GWP	kg CO_2eq
Ecotoxicity	Freshwater Aquatic Ecotoxicity Potential	FAETP	kg DCB[a] eq
	Marine Aquatic Ecotoxicity Potential	MAETP	kg DCB eq
	Terrestrial Aquatic Ecotoxicity Potential	FAETP	kg DCB eq
Eutrophication	Eutrophication Potential	EP	kg phosphate eq
Human toxicity	Human Toxicity Potential	HTP	kg DCB eq
Ozone depletion	Ozone Layer Depletion Potential	OLDP	kg R11[b] eq
Photochemical ozone formation	Photochemical Ozone Creation Potential	POCP	kg ethene eq
Resources depletion (elements)	Abiotic Depletion Potential (elements)	ADP el.	kg antimony eq
Resources depletion (fossil)	Abiotic Depletion Potential (fossil)	ADP f.	MJ

The GWP characterises and calculates the impact of greenhouse gases based on the extent to which these gases enhance radiative forcing. GWP values for specific gases, developed by the Intergovernmental Panel on Climate Change (IPCC), express the cumulative radiative forcing over a given time period following a pulse emission in terms of the quantity of carbon dioxide giving the same effect [18]. Following common convention, for example in the Kyoto Protocol, the 100-year values have been used here. The acidification potential indicator quantifies the impact of acid substances and precursors such as SO_2, NOx, and HCl. Rain, fog and snow trap the atmospheric pollutants and lead to environmental damage such as fish mortality, leaching of toxic metals from soil and rocks, and damage to forests and to buildings and monuments. Abiotic Depletion addresses the diminishing pool of mineral resources such as metal ores and crude oil. The measurement unit of abiotic depletion is MJ as the majority of non-renewable resources represent energy sources. The eutrophication potential includes all pollutants that promote microbiological growth leading to oxygen consumption. Nitrogen and phosphorus are the two main nutrients implicated in eutrophication leading to shifts of species composition and biological productivity. The photochemical ozone creation potential is an indicator of potential to create tropospheric ozone, expressed in equivalents to ethene as the reference species. The Ecotoxicity impact category considers emissions of toxic substances that have an effect on ecosystems.

[a] DCB: dichlorobenzene
[b] R11: trichlorofluoromethane

Freshwater and marine aquatic ecotoxicity potential (FAETP and MAETP) assess the toxic effects of polluting compounds to surface and sea waters respectively; whereas the indicator terrestrial ecotoxicity potential (TETP) is related to land based ecosystems. The Human Toxicity Potential reflects instead the risk of harm to humans from chemical species released into the environment, based on both the inherent toxicity of a compound and the potential human exposure. Finally, Ozone Depletion Potential is a measure of the destructive effects of gases on the stratospheric ozone layer.

LIFE CYCLE INVENTORY

In this analysis, the information about the materials and energy used throughout the process as well as the emissions and waste produced have been collated from different, reliable sources. Three different units have been identified in the foreground system for the process at issue: Thermal Oxide Reprocessing Plant (THORP), MOX fuel manufacturing plant and Geological Disposal Facility (GDF). In Figure 2 the process as a whole and the main links between the different units is depicted.

The Thermal Oxide Reprocessing Plant (THORP) is the latest of the reprocessing plants at Sellafield, where the reprocessing of national AGR and international LWR spent fuel assemblies is carried out. The assemblies are delivered to the site in transport flasks. They are held in THORP's Receipt and Storage Pond for a number of years to allow decay of the highly radioactive fission products with short half-lives, thus addressing the major handling difficulties associated with these short-lived isotopes. The fuel then undergoes a multi-stage reprocessing. From Fuel Receipt and Storage the fuel elements are transferred to the section known as Head End where the fuel assemblies are sheared and dissolved in nitric acid; the undissolved solids are removed by centrifugation. The fumes arising from the dissolvers are treated by the dedicated Dissolver Off-Gas (DOG) Plant. From Head End, the clarified fuel liquor solution is transferred to the Chemical Separation Plant where, firstly, uranium and plutonium are separated from the fission products and then uranium is separated from plutonium (PUREX Process). These liquors are then transferred to the Uranium Finishing and Plutonium Finishing Plants. The process as a whole produces a number of aerial, liquid and solid streams which are treated in ancillary plants. Intermediate level liquid wastes (medium active) are sent to the Low Active Effluent Monitoring Tank (LAEMT) for marine discharge, while intermediate level solid waste from fuel dissolution (medium active) is sent to Cement Encapsulation and storage and low level solid waste to Vault Storage. Off-gases are sent to gas treatment before stack discharge. Information about disposal containers and encapsulation procedures have been taken from the UK Derived Inventory [19].

The MOX fuel manufacturing plant is simulated in the model to manufacture MOX fuel from the Plutonium and Uranium powders produced by THORP. Notably the MOX plant was shut down in 2011 and its flowsheets and datasets are currently not available. Thus it has been modelled using the data available in the Ecoinvent database [17].

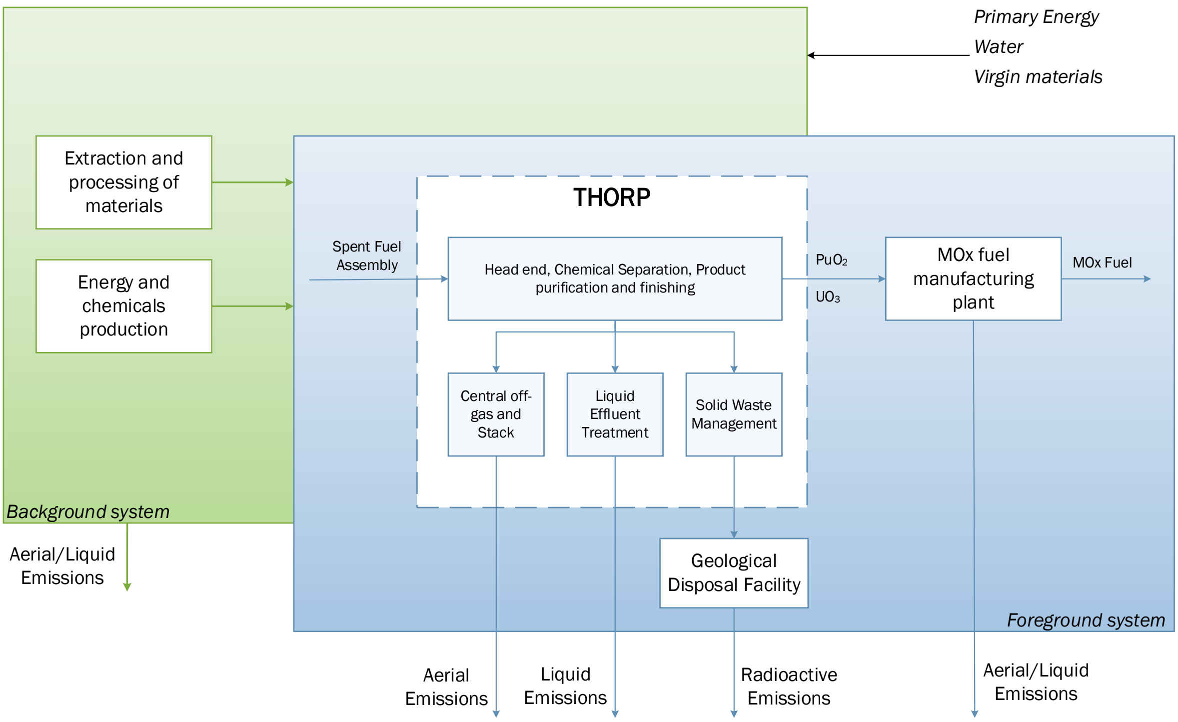

Figure 2 – Schematic diagram of the reprocessing step of Spent Nuclear Fuel

In the analysis, the encapsulated solid wastes are modelled as sent to the national GDF; this is the long-term plan although, as mentioned above, the facility does not yet exist. Information about the construction, operational requirements and capacity of the GDF has been taken from the UK Nuclear Decommissioning Authority's (NDA) Generic Disposal Facility Designs [20].

Notably this analysis is based upon the current UK approach and status of plants; therefore, with regard to the THORP and MOX plants, only operational environmental impacts have been taken into account. This is not the case for the GDF; as it has not been built, the construction has also been considered along with the proposed operation. However, this assumption makes the assessment specific to the UK.

THE RISK-BASED METHODOLOGY

The standard impact categories, conventionally used in LCA studies, do not take into account the impacts of radionuclide releases either as process emissions in the form of liquid and gaseous discharges or arising over time from radioactive solid wastes. However, exclusion of these impacts means that the assessment is not complete. A number of approaches have been proposed to fill this gap but none has so far been operationalized and generally accepted. A new framework for the assessment of both direct emissions of radionuclides and solid radioactive waste in the impact assessment phase of LCA is proposed in this work. It is based on the PhD work of Solberg-Johansen (1998), modified using more current data. A simple diagram of this methodology is shown in Figure 3.

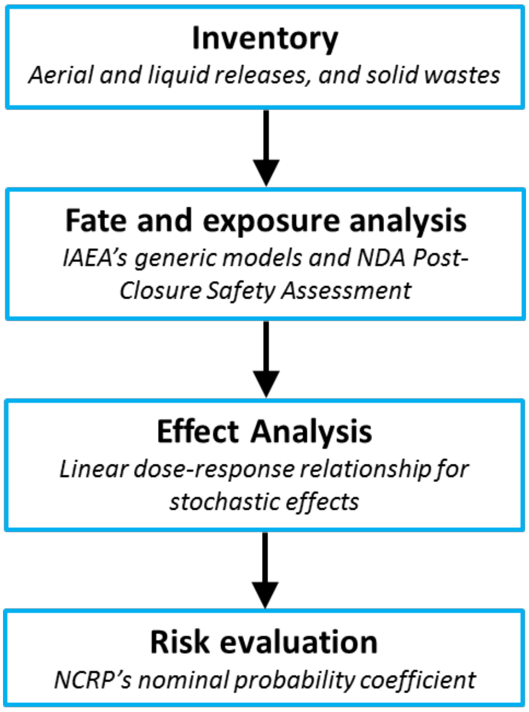

Figure 3 – Diagram of the risk-based approach

The starting point is the inventory, in terms of Becquerels, of liquid and aerial releases, and solid wastes. A fate and exposure analysis is carried out in order to model the transport of radionuclides through the environment (from the source to the receptor or environmental compartment) and through the food chain (from the environment to human beings). The fate and exposure analysis is based on two models: the International Atomic Energy Agency's (IAEA) generic models [21] and the UK NDA Generic Post–Closure Safety Assessment (PCSA) [22].

The former constitutes a simple but robust assessment methodology for the estimation of radioactive doses from routine releases of aerial and liquid radioactive emissions. The latter is a generic exercise performed by the Radioactive Waste Management Ltd. with the aim of supporting the environmental safety case of the future national GDF, in which the impact of the storage of nuclear solid waste in a GDF located in a higher strength rock is assessed. The emission pathway considered most probable and therefore analysed in the assessment is leakage of radionuclides from the corrosion of canisters, followed by dissolution of the contents and transport to the surface in groundwater. The end point of these models is the adsorbed dose, defined as the quantity of ionizing radiation adsorbed per unit mass of the recipient, evaluated for a specific receptor commonly referred as "critical group". The critical group is defined as the member(s) of the public most exposed to radiation due to operations at a given site.

The main difference between the two models lies in the timeframe considered. The IAEA models assess the actual impact of current radionuclide releases, whereas the NDA PCSA estimates the future risks arising from solid nuclear wastes buried in a deep GDF. The latter risks start to arise a thousand years after the closure of the GDF and peak after about a million years. The peak value has been considered as the reference risk value for each radionuclide and type of waste. Figure 4 shows an example of the radiological risk as a function of time from different waste types buried in a GDF as estimated by the NDA.

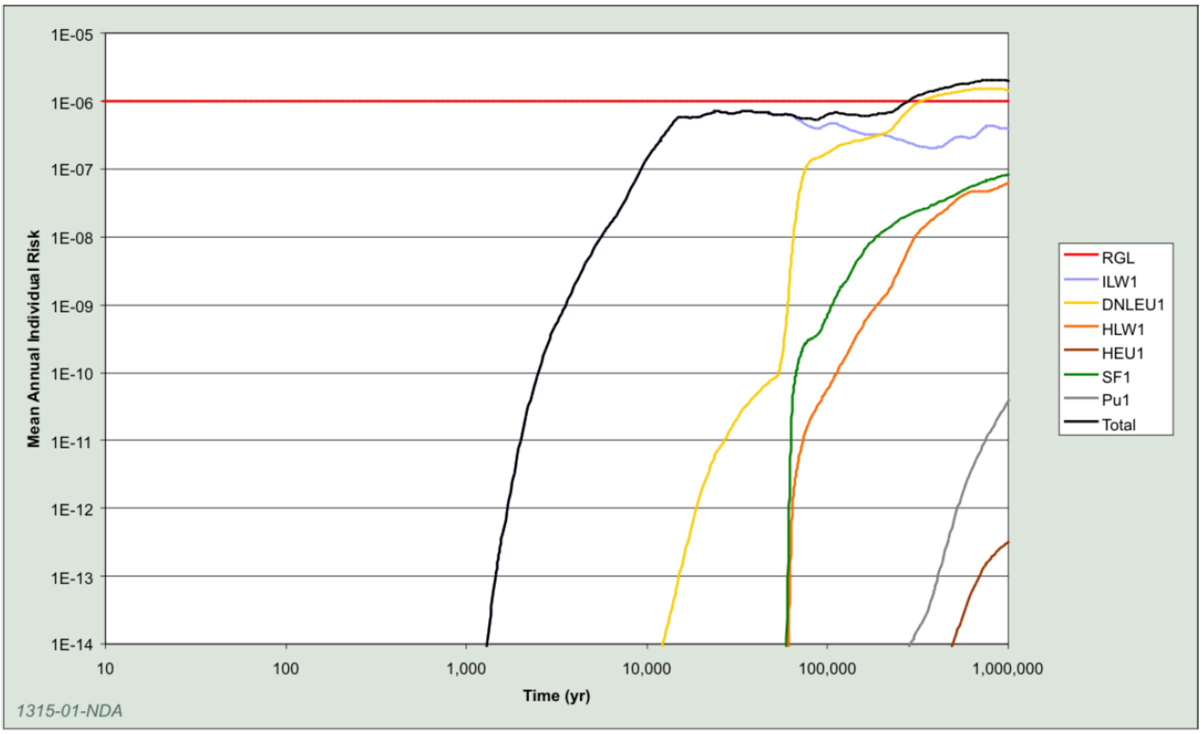

Figure 4 – Total mean radiological risk against time showing contributions from the different waste types buried in a GDF. The risk relates to the amount of wastes included in the RWM reference case [23]**.**

However, different types of radiation can cause different effects in biological tissues and different organs may be more or less susceptible to irradiation. The exposure analysis takes in consideration those aspects by converting the adsorbed dose into an effective dose by means of a linear dose-response relationship for stochastic effects at low doses [24].

Eventually the estimated effective dose is converted into a risk of detrimental effect by using the International Commission on Radiological Protection's (ICRP) nominal probability coefficient for stochastic effects [24].

Notably, the different timeframes of the impacts of direct discharges (aerial and liquid) and solid wastes constitute a big challenge. As the risk associated with direct discharges refers to a current impact and the one arising from solid waste represents a potential impact in the distant future, they are not directly comparable; therefore, their relative significance requires fundamental consideration and discussion. A number of approaches have been used in LCA to deal with different timeframes (e.g. [25]), but none encompasses such great differences in scale. Therefore, at this stage the impact of direct discharges and solid wastes is kept separate within different impact categories. However, in this analysis a preliminary comparison between those two impacts will be drawn, but it must be clear that this is done recognising that a more consistent approach must be developed.

CONCLUSIONS

A Life Cycle Assessment study is being carried out in order to evaluate the environmental impact of reprocessing Spent Nuclear Fuel in the UK. A risk-based methodology has been developed and operationalised with the aim of assessing and comparing the impact of radionuclides releases and radioactive solid wastes. The system has been modelled on the UK approach and consists of three main units: Thermal Oxide Reprocessing Plant (THORP), MOX fuel manufacturing plant and Geological Disposal Facility. Real data from these plants have been used for the foreground system. Preliminary results will be presented at the conference as they are still under review by the National Nuclear Laboratory and Sellafield ltd. Future work will involve the development of a more consistent approach to compare the impact of solid wastes and direct discharges. Moreover, further existing and future nuclear fuel cycles will be modelled and a comparison between them will be drawn.

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