Reprocessing Vs Direct Disposal of Used Nuclear Fuels: The environmental impacts of future scenarios for the UK nuclear fuel cycle

Abstract

The UK recently switched from a "nominal" twice-through cycle - whereby used nuclear fuels were reprocessed, but uranium and plutonium were not routinely reintroduced in the fuel cycle – to a once-through cycle, where used nuclear fuels are stored pending disposal. However, it is also the current strategy to keep other options open, including a twice-through cycle based on a different chemical separation process from the conventional PUREX. This article presents a comprehensive Life Cycle Assessment 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. Several marginal technologies are explored because of the uncertainty regarding the actual response of the market. Results of the study show that recycling of uranium, but especially of plutonium is of paramount importance because of the avoided burdens associated with production of nuclear fuel from mined uranium. The reprocessing scenarios envisaging reprocessing of used nuclear fuels and recycling of both plutonium and uranium represent the most favourable options. The direct disposal approach may be advantageous only in terms of radiological impacts depending on the marginal technology chosen.

Keywords: Life Cycle Assessment; used nuclear fuels; reprocessing; direct disposal

Highlights:
- A comprehensive LCA study on future UK scenarios for managing used nuclear fuels.
- The consequential approach is adopted with inclusion of short-term effects.
- Plutonium recycling is key for improving the environmental performance.
- Reprocessing with recycling of plutonium and uranium represent the best option.
- Direct disposal may be advantageous only in terms of radiological impacts.

1   Introduction

In 2019 the UK government enshrined in law an amendment to the Climate Change Act that established a legally binding target to reduce UK's greenhouse gas emissions to net-zero by 2050 [1]. This is in line with the EU's proposed green deal [2] and falls within a global effort for restraining increase in global average temperature to well below 2 °C above pre-industrial levels, agreed in Paris in 2015 [3]. The power generation sector represents the largest contributor to greenhouse gas emissions and has the potential to decarbonise almost fully and more quickly than other industrial sectors. At present, the UK is home-producing around 95% of the total electricity supply. Natural gas represents the largest source at ~40%, followed by renewables at ~33% and nuclear at ~20%. Coal, the largest source of electricity since the industrial revolution, has fallen steeply in recent years, and currently accounts for ~5% [4]. The UK government outlined in its Carbon Plan [5] a number of scenarios for achieving reduction targets. 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 [5,6].

Historically, the UK operated a "nominal" twice-through cycle [7]: used nuclear fuels (UNFs) were reprocessed at the Sellafield site, with fission products being vitrified into a final, manageable form suitable for disposal. The twice-through cycle was just "nominal" because plutonium (Pu) and uranium (RepU[1]) – especially from oxide fuels - were not routinely reintroduced in the fuel cycle, rather they were stored in an oxide form at Sellafield site pending a future decision by the Government on their fate. Except for uranium mining and milling, the UK had full fuel cycle facilities and was self-sufficient in both the front (conversion, enrichment and fuel fabrication) and the back end (reprocessing and waste treatment) [8].

At present, the UK operates a once-through cycle. Reprocessing of oxide fuels ceased in 2018 with the closure of the Thermal Oxide Reprocessing Plant (THORP); the Magnox reprocessing plant, which deals with metal fuels, is planned to shut down in 2020 after all remaining Magnox fuels are reprocessed. Remaining and future UNFs arisings, which will come primarily from existing Advanced-Gas Cooled Reactors (AGRs), are planned to be wet-stored at Sellafield site or dry-stored in newly developed cask storage facilities (such as that at Sizewell), pending disposal in a deep repository built several hundred metres underground in a geologically stable environment; this is known as Geological Disposal Facility (GDF) [9]. After the failure of the 2013 consultation exercise, 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 and its operation is projected to last for approximately 100 years [10].

Although the UK has moved to a once-through cycle, it is the intention of the Government to keep other options open, including a twice-through cycle based on a different chemical separation process from the conventional PUREX (e.g. Advanced PUREX , i-SANEX and GANEX [11]) and with a full integrated management of nuclear waste and pre-determined fate of RepU and Pu. The work presented in this article quantifies and compares the environmental performances of alternative strategic options for the back-end of the UK nuclear fuel cycle by means of Life Cycle Assessment (LCA), with the ultimate objective of supporting the decision-making process. The focus is on the once-through cycle approach and several scenarios for a reprocessing approach that assume operations of an equivalent of the THORP plant (which was based on the PUREX process), ignoring any future

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[1] The acronym 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) or depleted uranium (DepU).

newbuild. The article is organised as follows: Section 2 reports the study's goal and scope, the systems boundaries, life cycle inventory and impact categories analysed; Section 3 presents results of the LCA study for reprocessing and direct disposal approaches, which are discussed in Section 4. Finally, the key findings of the analysis are summarised in Section 5, and a glossary is reported in Section 6.

2 Methods

2.1 Goal and scope

According to the framework developed by Sanden et al., LCA studies are distinguished according to responsibility and time perspective [12,13]. Responsibility differentiates between attributional and consequential approaches: the former aims at mapping the share of environmental impacts a system is responsible for, whilst the latter focuses on describing the environmental effects of choices. The time perspective distinguishes between studies looking at historic (retrospective LCA) or future (prospective LCA) environmental impacts.

The goal of this study is to quantify and compare the environmental impacts of two alternative approaches for managing UNFs in the UK: one envisaging their direct disposal (i.e. once-through cycle), and the other their reprocessing in a twice-through cycle. The focus is on future environmental impacts that are consequence of strategic choices made today. Therefore, 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 be difficult to predict, only short-term effects are considered. These include first order direct physical effects, but not second and third order effects (known as negative and positive feedback effects) [14]. A consequential study with such features is in effect identical to a retrospective 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 the management of UNFs from AGR containing 1 tonne of uranium pre-irradiation, which refer to the quantity of uranium before being partially transformed in nuclear reactors, either by fission or transmutation, into other elements.

2.2 System boundary

We divide the system boundaries into a foreground and background system [15]. The former identifies all those processes that are the main focus of the study and that may be directly affected by decisions based on the study's results; the latter encompasses all other processes that exchange materials and energy with the foreground, usually through an homogenous market. Figure 1 reports the system boundaries for the reprocessing and direct disposal approaches.
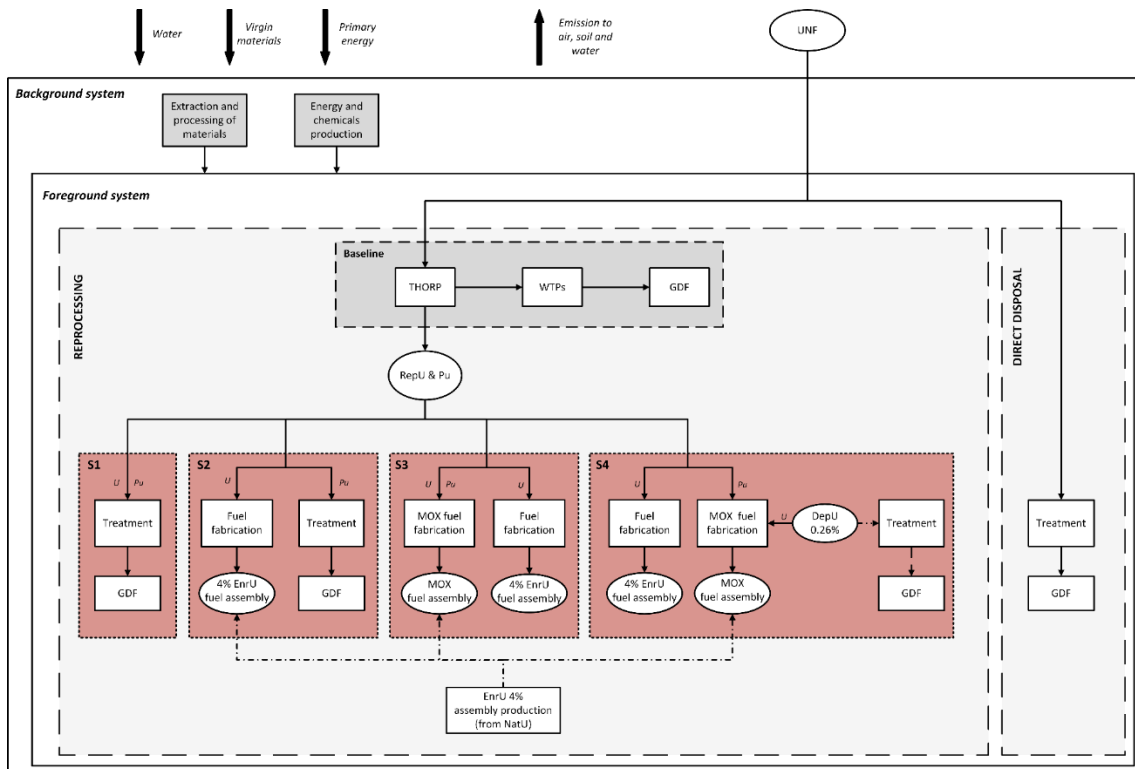
Figure 1 - System boundary for direct disposal and 4 different reprocessing scenarios. For clarity, tranportation has not been included. WTPs: Waste Treatment Plants. GDF: Geological Disposal Facility.

2.2.1 Reprocessing

The foreground system for the reprocessing approach includes the historical approach to UNFs management in the UK (i.e., the "nominal" twice-through cycle mentioned in Section 1, hereafter termed "baseline") and four scenarios for disposal and/or reuse of RepU and Pu. The baseline includes not only processes that were carried out, but also other processes that were part of policy but not yet operationalised; one notable example is the Geological Disposal Facility (GDF). The environmental peformance of the baseline were assessed in [7] , and all the assumptions made there also apply here. For instance, 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. We assumed that Pu is encapsulated according to the can-in-canister approach [16] developed in the US for disposal of Pu alongside high level waste (HLW) glass - although the current strategy favours other approaches, including the development of alternative ceramic forms made by HiPing [17]. The approach envisages plutonium oxide to be immobilised in a titanium-based matrix (whose composition is reported in Table S1 in the Supporting Information), to form a puck 6.9 cm wide and 2.5 cm thick. 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 about 20 pucks, and 28 cans are loaded into each canister. Finally, we assume that each steel canister is packaged in a single high-integrity copper disposal canister, whose design is inspired to the Swedish KBS-3V concept [18]; the design and its underpinning assumptions are reported in [19]. However, this is only one amongst other concepts currently being considered for implementation in the UK. RepU is assumed to be encapsulated in grout and packaged in 500 litres stainless steel drums, in line with the approach used for other intermediate level wastes (ILWs) [19].

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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. Finally, both Pu and RepU packaged wastes are disposed in a GDF based on the NDA's generic design for higher strength rock [7,20,21], to be built in a location to be defined in the UK. Pu is assumed to be disposed in disposition tunnels alongside HLW and/or UNFs, whilst RepU is disposed in vaults with other ILWs.

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. RepU is re-enriched to a concentration of 4.1% (equivalent to 4% of EnrU from NatU)[2] and used for fabrication of new fuel assemblies to be inserted into nuclear reactors for electricity generation purposes. Figure 2 reports the production process of fuel assemblies from RepU. As the enrichment process requires uranium to be in a gaseous state at relatively low temperature, RepU is first transported to a fuel manufacturing and conversion plant, where it is converted into uranium hexafluoride (UF_6), and then to a centrifuge enrichment plant, where it is enriched to 4.1%. Finally, the Enriched Uranium (EnrU) is transported back to the fuel manufacturing and conversionl plant, where it is reconverted into an oxide form, put in the form of pellets and inserted into fuel assemblies. It must be noted that typically fuel manufacturing and conversion, and enrichment plants have been designed to treat NatU rather than RepU. Handling of RepU requires alterations of the existing design, such as e.g. thicker barriers to protect against additional radiation due to U234, a strong alpha emitter with a moderate half-life [22]. Such modifications have not been taken into account because of lack of data in the literature; but they are 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 following the same approach as RepU (Scenario 1), i.e. the working assumption (as this is not currently carried out) is that they are encapsulated in grout, packaged in 500 litres drums and disposed in a GDF.
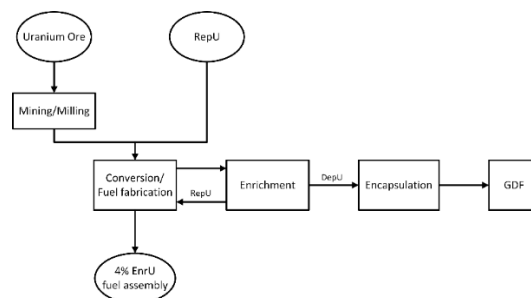

Figure 2 – Schematic outline for the production of 4.1% enriched uranium (EnrU) from reprocessed uranium (RepU); and from uranium ore (i.e. natural uranium, NatU).

Scenario 3

Both uranium and plutonium in their oxide states are combined to produce a mixed oxide (MOx) fuel. The mixing proportion of uranium and plutonium (calculated according to the equation reported in Section S1.1 of the Supporting Information) 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 of a specific level of enriched uranium [23]. This study assumes that the MOx produced is equivalent to 4% EnrU from NatU. Other parameters that affect the mixing proportion are the concentration of fissile plutonium (239 and 249) and uranium (235) in both oxides,

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[2] This is due to poison radionuclides like uranium 234 (U234) and uranium 236 (U236) [22].

and the concentration of the isotope 236 of uranium (U236) 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. Concentration of U235 and U236 in RepU and the mixing ratio used for MOx are reported in Table 1 and Table 2. The uranium in excess of what is 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 2.

Table 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 [22]	0.26%	Ecoinvent database
U236	0.39%	Figure 5 in IAEA tecdoc 1529 [22]	0.00%[**]	

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

Table 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 2

~~Table 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 from NatU); this is due to the 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. Finally, like in Scenarios 2 and 3 RepU is re-enriched and used to produce new uranium fuel assemblies.

Avoided Burdens

The consequential perspective with inclusion of only short-term effects represented by first order direct effects (see Section 2.1) is implemented by accounting for the additional or avoided burdens associated with changes in demand or production. Unlike Scenario 1, Scenario 2, 3 and 4 represent multifunctional product systems: they deliver the twofold function of 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 2.3.1.

2.2.2 Direct disposal

The direct disposal approach (Figure 1) envisages 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 down and short-lived fission products to decay. In the UK, wet ponds at Sellafield site will continue to be used for this purpose, but new dry storage facilities have also been constructed to take some of the UNF and this will increasingly be used as the capacity at Sellafiled is filled.Both approaches have minor routine discharges and operational consumptions; therefore, they were not included in the system boundaries. (Note that the same assumption was made for the baseline.) Following interim

storage, UNFs are assumed to be encapsulated, packaged into disposal canisters and disposed in a GDF. The approach to encapsulation and packaging of UNFs is based on [24]. 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. 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 8 slotted cans, corresponding to approximately 24 AGR fuel elements [20,24]; its design is inspired to the Swedish KBS-3V concept for LWR fuel [18] - 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 (Figure 3). As noted above, this is one of many concepts currently being considered in the UK.
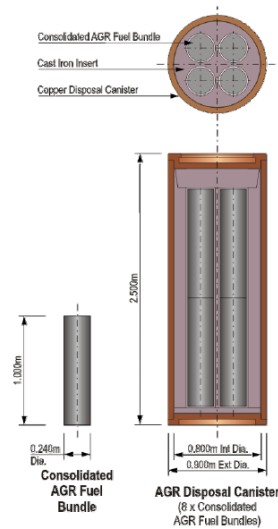

Figure 3 – Modified version of the KBS-3V concept for disposal of AGR UNF (adapted from [25]*).*

2.3 Life cycle inventory

Primary data, including process-specific operational or design data, have been used for the foreground system, whilst secondary data from the Ecoinvent database v3.3. have been used for the background system. A comprehensive description of the inventory used for the baseline processes (see Figure 1) is reported in [7]. Table 3 reports mass balances for key intermediate and final products and wastes related to the functional unit.

Table 3 - Mass balances for management of 1 tonne of uranium in AGR fuel, for reprocessing (S1, S2, S3 and S4 scenarios) and direct disposal approach

Approach		RepU	Pu	Packaged Pu	Packaged U	Packaged DepU	Packaged UNF	MOx	EnrU
		(kgHM)	(kgHM)	(m³)	(m³)	(m³)	(m³)	(kgHM)	(kgHM)
Reprocessing	S1			3.5	0.57	-	-	-	-
	S2	970	30	3.5	-	0.34	-	-	384
	S3			-	-	0.31	-	96	351
	S4			-	-	0.34	-	64	388
Direct disposal		-	-	-	-	-	1.54	-	-

Data for encapsulation and packaging of RepU/Pu and UNFs have been obtained from [19] and [24] respectively; these are reported in Tables S2, S3 and S4 in the Supporting Information. Operational and design data for the processes of enrichment of RepU and NatU, production of MOx and fabrication of fuel assemblies were not available; 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 [26]; but

some processes have been amended to reflect specific features of the 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.

Data for construction, operation and decommissioning of the GDF have been obtained from the generic design developed by the Radioactive Waste Mangament Ltd. for the UK [21] and is fully reported in [7]. Radioactivity levels of disposed wastes (Pu, RepU and UNFs) have been obtained from [19,24], and are reported in Table S5 in the Supporting Information. Notably, the radiaoctivity reported does not refer to the time at which wastes are generated, rather to that at which disposal occurs. Immediate disposal is assumed for all waste types except HLW and Pu 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 [27] and are reported in Table S6 in the Supporting Information.

2.3.1   Marginal technologies

As noted in Section 2.2.1, production of MOx and RepU fuels for use in nuclear power plants is assumed to induce a decline in demand for enriched fuel from NatU. Because production of uranium does not represent a constrained market, the procedure developed by Bjorn et al. [28] 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 et al. [29] (1999). 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 competitive technology cannot be determined objectively. Thus, in this study the technology that is deemed to contribute most to the supply of uranium in the UK is taken to represent the marginal technology.

As noted in Section 1, the UK had (prior to ceasing reprocessing) full fuel cycle facilities in both the front- and back-end of the nuclear fuel cycle apart for uranium mining and milling. Therefore, the marginal technologies for enrichment and fuel manufacturing are represented by those processes currently carried out in the UK. On the other hand, uranium in the form of yellowcake is purchased on the open market. According to figures published by the World Nuclear Association (WNA), uranium production increased by ~5% in the period from 2009 to 2018, from ~50 to ~53 thousand tonnes of U [30]. Uranium is typically purchased through contracts that may last from 2 up to 10 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, which through the years has sourced uranium from about all uranium-producing states [31]. Although during the first decades Australia was the main exporter, present indications are that an increasing amount of uranium is sourced from Kazakhstan, which from 2009 became the largest producer (at the expense of Canada, 2nd, and Australia, 3rd) and has since steadily increased its production [32]. All major mines in Kazakhstan use the In-Situ Leaching technology [33], which involves leaving the ore in the ground, dissolving minerals and pumping the pregnant solution to the surface [34]. The marginal technology is thus represented by a generic In-Situ Leaching mine in Kazakhstan (see Figure 2) and the yellowcake is assumed to be transported to the UK via rail and sea freight according to transportation data reported in Table S6 in the Supporting Information. The In-Situ Leaching mine in Kazakhstan is modelled based on generic data included in the Ecoinvent database v3.3.

Because the actual response of the market to marginal changes in uranium demand is uncertain, it is crucial not to limit the analysis to one, but to consider a range of marginal technologies [35]. 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 [36]. 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 In-Situ Leaching and underground mines [37]. 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 [29] with respect to uranium production and thus it is not considered in this study. McArthur River, Cigar Lake, Ranger and Four Mile mines have been modelled based on generic datasets for underground, open-pit and In-Situ Leaching mines included in the Ecoinvent database v3.3 and enhanced 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 [27]. Transportation data data for each marginal technologies are reported in Table S7 in the Supporting Information.

2.4 Impact assessment

In the Impact Assessment phase, the emissions and inputs quantified in the inventory phase are translated into a smaller number of impacts. Two general approaches are available, using so-called mid-points or end-points [38]. This study uses the mid-point approach based on the ILCD (International Life Cycle Data System) recommendations [39,40]. All impact categories with the exception of land use and ionising radiations, have been included. The former was excluded due to lack of data for the foreground system, whilst the latter was replaced by two impact categories developed by Paulillo et al. [41–44] for direct discharges (named ionising radiations) and for emissions from solid waste disposed in a GDF (named ionising radiations, waste). Table 4Table 4 reports the impact categories considered in this study, along with their metrics and acronyms used in results charts.

Table 4 - Impact categories analysed

Impact category	Metric	Acronym
Acidification	[Mole of H+ eq.]	A
Climate change	[kg CO2-Equiv.]	CC
Ecotoxicity freshwater	[CTUe]	ECf
Eutrophication freshwater	[kg P eq]	Ef
Eutrophication marine	[kg N-Equiv.]	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-equiv]	IR
Ionizing radiations, waste	[Bq U238 ILW-equiv]	IRw
Ozone depletion	[kg CFC-11 eq]	OD
Particulate matter/respiratory inorganics, human health	[kg PM2,5-Equiv.]	PM/RI
Photochemical ozone formation, human health	[kg NMVOC]	POF
Resource depletion, mineral, fossils and renewables	[kg Sb-Equiv.]	RDm
Resource depletion water	[m³ eq.]	RDw

3 Results

The environmental impacts of the product system described in Section 2 were calculated with Gabi sustainability software version 8 [45]. The results section is divided into four sub-sections. First, Sections 3.1 and 3.2 report hot-spot analyses for the reprocessing scenarios and the direct disposal approach; then these options are compared in Section 3.3; and finally, Section 3.4 presents a comparison of several marginal technologies for uranium mining (see Section 2.3.1).

3.1   Reprocessing scenarios

Figure 4 shows results of the impact assessment phase for reprocessing Scenarios 1, 2, 3 and 4. Impacts are expressed as percentual additions to the impact of the baseline (see Section 2.2) of each process within a scenario (reported in full colour) and of each scenario, as net sum 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; 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.
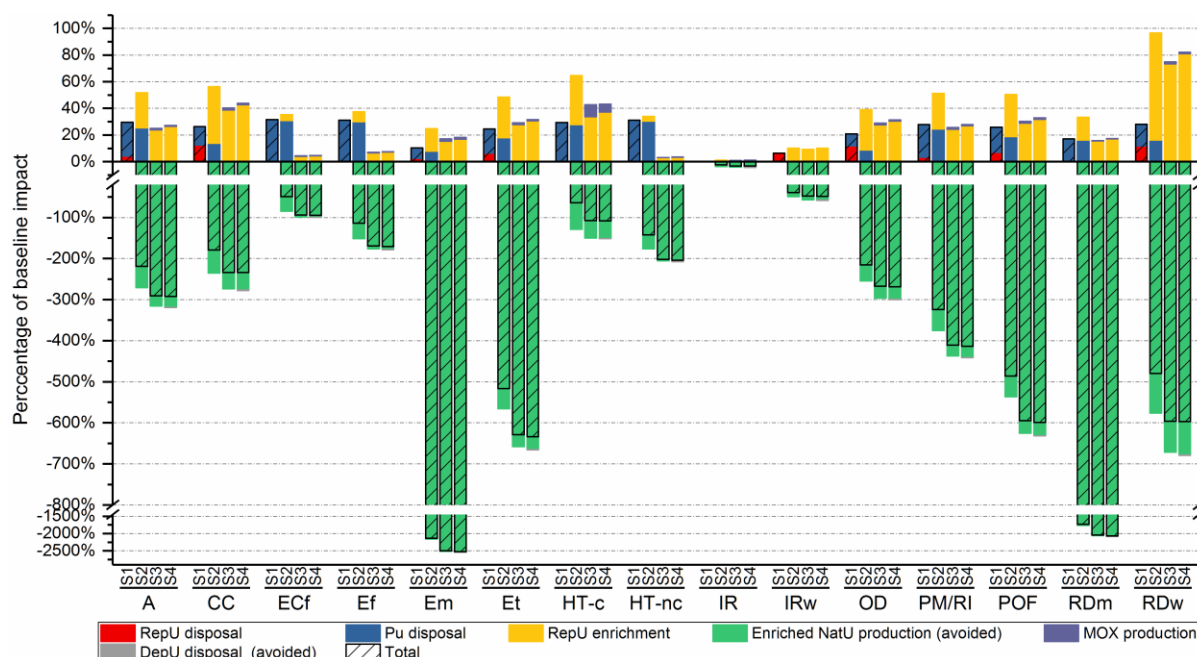

Figure 4 - Environmental impacts of reprocessing scenarios (S1, S2, S3 and S4) reported as percentage of the impact of the baseline.

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 disposal of both Pu and RepU; and ionising radiations from nuclear waste (IRw), which by contrast is entirely attributable to disposal of RepU. More specifically, IRw impact is chiefly caused by two radionuclides, namely uranium-235 (U234) and uranium-238 (U238), contributing to about 99.5% (see Figure S1 in the Supporting Information). The other ionising radiations category, which is concerned with direct discharges (IR), results in having negligible impact, lower than 1%, compared to the baseline. Scenario 1 does not include any avoided burdens, thus the net impacts are positive in sign, and range from 5% for IRw, to ~20% for the majority of impact categories, and up to 30% in the freshwater ecotoxicity (ECf) and eutrophication (Ef) category. For instance, a value of 20% entails 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.

In Scenario 2 RepU is recycled (rather than disposed), 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, 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 contributions in the remaining categories. 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. Net impacts (i.e. sum of positive and negative) of all categories are 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 (i.e. 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 and fabrication of MOx fuel has negligible impacts, also the positive impacts are lower than Scenario 2. The highest reductions occur for 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 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 in 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.

3.2   Direct disposal

Figure 5 reports the hot-spot analysis for the direct disposal approach. The chart shows that all impact categories with the exception of IRw are dominated by construction and decomissioning of the GDF and by manufacturing of disposal canisters. The former represents the major contributor to the categories CC, Em, Et, IR, OD, POF and RDw, and its environmental impacts (Figure S2) are caused by operational consumption of electricity and by bentonite, which is primarily used for backfilling the GDF during decommissioning. The environmental impacts of disposal canisters manufacturing, which dominates the remaining categories, are primarily due to copper and cast iron (Figure S3). By definition, the impacts in the IRw category are associated with radioactive emissions arising from nuclear waste, which we attribute to the operational phase of the GDF. Notably, over 95% of the IRw impacts is attributable to two specific radionuclides, namely caesium-135 and tin-126 (Figure S4). Finally, the enviornmental impacts related to transportation of UNFs, operation of GDF and slotted cans production (see Section 2.2) are in practice negligible.
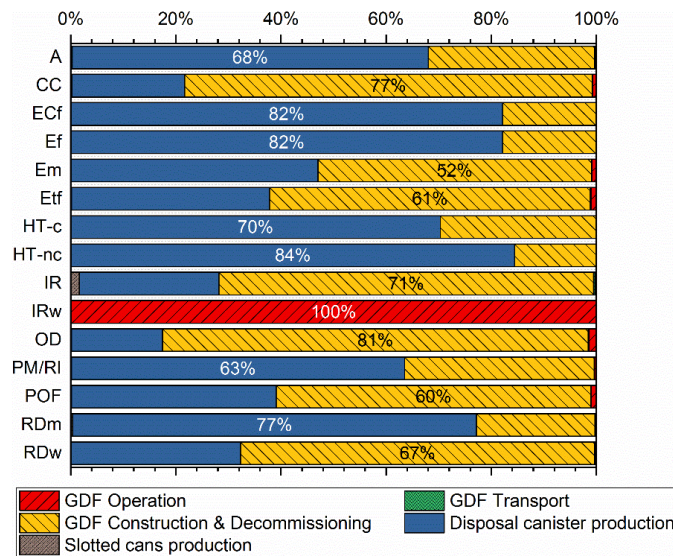
Figure 5 – Hot-spot analysis of the direct disposal approach

3.3 Comparison of reprocessing and direct disposal

Table 5 presents a comparison of the environmental performances of the approaches considered in this article for managing UNFs. The table reports both absolute impacts and the difference between reprocessing scenarios and direct disposal impacts expressed as percentage of the direct disposal impacts. The figures refer to all activities required to manage UNFs, meaning that the reprocessing scenarios also include the impacts of the baseline. For each category, impacts are ranked from lowest (highlighted in green) to highest (highlighted in red).

		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	[m³ eq.]	2.11E+03 *(87%)*	-6.26E+03 *(-654%)*	-8.18E+03 *(-824%)*	-8.18E+03 *(-825%)*	1.13E+03

Table 5 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-nc, HT-c and IRw after direct disposal (from 10% to 19% lower). The direct disposal approach yields the highest or the second highest environmental impacts in all categories with the exception of IR, for which it features the lowest figure (by three orders of magnitude) amongst the options considered. This is explained by the absence of direct radioactive discharges when directly disposing of UNFs, which in contrast occur during UNFs reprocessing. On the other end of the spectrum, Scenarios 3 and 4 represent 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. Scenarios 2, 3 and 4 yield negative impacts in all categories concerning non-radiological impacts; no approach yields negative impacts in the radiological categories.

3.4 Comparison of marginal technologies

Table 6 presents a comparative analysis of the environmental impacts associated with four marginal technologies for uranium mining, namely an underground mine in Canada, an In-Situ Leaching 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 - 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 CO2 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	[m³ 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. For some categories (e.g. A and CC) the variation is as little as ~10%. The IR category features variations as high as five orders of magnitude between the ISL mines and the generic open pit mine in Australia. Overall, the generic In-Situ Leaching mines in Kazakhstan and Australia feature the highest environmental impacts in 10 out of 14 categories, and the lowest IR impacts amongst the technologies considered; whilst the Australian Ranger mine yields 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. 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 Tables S9 to S12 in the Supporting Information; 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 option for all impact categories considered. The comparative analysis of marginal technologies also demonstrates that using a specific inventory dataset from the Ranger mine would lead to changes in favour of the direct disposal approach in all impact categories. However, the LCA results reported in the Supporting Information show that Scenarios 3 and 4 are still the most preferred option for all impact categories but IR.

4   Discussion

4.1   Reprocessing of used nuclear fuels

The comparative analysis between reprocessing and direct disposal (Section 3.1 and Table 5) shows that, with the exception of radiological impacts due to direct discharges, the reprocessing approach is more environmentally beneficial than direct disposal when either uranium only or both uranium and

plutonium are recycled (Scenarios 2, 3 and 4). Similar results were obtained for the French nuclear fuel cycle [46,47]. The production of nuclear fuel from recycled materials avoids mining and enrichment of uranium ore – the two activities of the nuclear fuel cycle with the greatest impact [27,46,48]. The avoided production of NatU fuel, displaced by either only RepU fuel or RepU fuel and MOx, offsets the additional environmental impacts and generates significant environmental gains, which are even higher than impacts linked with UNFs reprocessing and disposal of fission products, namely the baseline (see Figure 5 and Table 5)

The analysis also demonstrates that recycling of Pu (Scenario 3 and 4) is of paramount importance, for three main reasons. First, disposal of plutonium is a high-impact activity (see Figure 4). Second, plutonium generates 50% more energy than uranium on a mass basis; this implies 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 - another high-impact, energy-intensive process. Third, plutonium, like HLW and UNF, is disposed of in the GDF's disposition tunnels, which occupy an area per volume of waste considerably higher than vaults used for ILW. This implies that avoiding disposal of plutonium through recycling also entails a lower land footprint of the GDF (Table S8 and S9).

It is also evident why Scenario 1 represents the worst available option: the application of an intensive chemical process to achieve separation of fission products, uranium and plutonium only for them to be sent for disposal 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 do not recognize the value of unused resources contained in UNFs; however, whilst the direct disposal approach envisages UNFs to be disposed altogether, the reprocessing Scenario 1 separates individual waste streams with the final aim of disposing them in the same way as the direct disposal approach. In addition to this, Scenario 1 increases proliferation risks, the amount of wastes generated and the size of the GDF required for their disposal. The approach of Scenario 1 has only one main benefit: the vitrification of fission products - the most troublesome waste stream - in glass generates a homogenous, compact and secure waste form that decays much faster than spent fuels.

Although recycling of valuable materials is an environmentally advantageous practice in all industries, in the nuclear sector it brings additional benefits. Uranium is a relatively concentrated resource (although more widespread than oil); reprocessing and recycling of uranium and plutonium can guarantee the security of supply to those countries with no available resources and protect against market volatility. The results of this study suggest that, in some scenarios, the case for reprocessing (instead of direct disposal) may be based less on the efficient use of fissile resources, but rather on the environmental improvements that are primarily associated with avoiding construction and operation of uranium mines. It is in fact 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 [49]. The deployment of fast reactor technologies could also extend the lifetime of conventional and expected resources up to infinite.

However, the approach of reprocessing used nuclear fuels has two main arguments against it. First, reprocessing is often linked with an increase in the 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, US studies indicated [50,51] that the cost of reprocessing is far higher than that of direct disposal: with uranium price equal to $40/lb (today's price

is ~\$25/lb [52]), a reprocessing-based fuel cycle would increase the cost of nuclear electricity by \$1-2/MWh; otherwise, to break even the price of uranium would need to go up to \$360/lb – a price never reached and not likely to be seen in the coming years, even more so if price rebound effects due to UNFs reprocessing are considered. Additionally, the growing demand to move towards a low-carbon economy will mean that the economic feasibility of nuclear energy will increasingly be based on comparison to the price of renewable energies; only if the price of nuclear-based electricity is competitive will conventional[3] nuclear energy survive.

4.2   Direct disposal of used nuclear fuels

Direct disposal currently represents a straightforward, lower-cost and proliferation-resistant approach for managing UNFs that, unlike reprocessing, 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) when the marginal technology corresponds to a generic In-Situ Leaching mine located in Kazakhstan or Australia, or an open pit mine in Australia described by the Ranger mine site-specific dataset (Section 3.4). If the marginal technology is assumed to be represented by an underground mine in Canada or an open pit mine in Australia, the reprocessing scenarios envisaging recycling of RepU (Scenario 2,3 and 4) will result in having lower ionising radiations impacts 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 In-Situ Leaching technology and the Ranger mine, thus increasing the significance of avoided burdens. Besides IR, the ranking of options for the other impact categories is not significantly affected by the choice of the marginal technology.

The contribution analysis on the direct disposal approach revealed that the impacts are primarily linked to the use of copper as corrosion resistant material in disposal canisters (notably, copper production is also one of the major contributors to the impact of the baseline) and with construction and decommissioning of the GDF.

4.3   Validity of the LCA results

The consequential perspective with inclusion of short-term effects relies on the assumption that MOx/RepU fuel displaces NatU fuel with a one-to-one ratio. This assumption is valid for the assessment of waste management systems when market mechanisms are neglected, but does not apply to assessment of nuclear fuel cycles because MOx and RepU fuels cannot be recycled indefinitely, as the continuous building-up of radionuclides' poisons reduces their efficiency. Studies on the nuclear fuel cycle need to consider appropriate substitution rates based on an number of recycles of used nuclear fuels. To this end, it would be interesting to compare the twice-through approach (where UNFs are recycled only once) with multi-recycling approaches.

5   Conclusion

This study assessed the environmental performances of five scenarios for the management of used nuclear fuels (UNFs) in the UK, including a direct disposal approach and four reprocessing scenarios envisaging different strategies for disposal and/or reuse of uranium and plutonium. The results provides valuable information for UK policy- and decision-makers. The comparative analysis demonstrated that recycling of plutonium and uranium is of paramount importance: the environmental gains linked with avoided production of nuclear fuels from mined uranium offset the

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

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 from reprocessed or depleted uranium and plutonium (namely Scenario 3 and 4) represent the most advantageous options from an environmental perspective, showing negative impacts (i.e. they reduce the impacts of the baseline and/or the entire fuel cycle) 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 the worst options amongst those considered. The direct disposal approach may result favourable with respect to radiological impacts from direct discharges depending on the marginal technology chosen. Notably, the marginal technology does not affect the ranking of the options for the other impact categories.

It is the authors' intention to extend the analysis presented in this study to new advanced reprocessing scenarios and operational regimes for future nuclear generation capacity in the UK. Understanding the whole life environmental impact of those scenarios will be a valuable tool in helping to determine the future direction for nuclear power in the UK.

6   Glossary

AGR	Advanced Gas-cooled Reactor
DepU	Depleted uranium (enrichment tails)
EnrU	Enriched Uranium
GDF	Geological Disposal Facility
HLW	High Level Waste
ILW	Intermediate Level Waste
LWR	Light Water Reactor
MOx	Mixed oxide fuel
NatU	Natural uranium
RepU	Reprocessed uranium
UNF	Used nuclear fuel
WTP	Waste Treatment Plant

7   Acknowledgments

8   References

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