IMPACT ASSESSMENT OF ENHANCED EXPOSURE FROM NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORM) WITHIN LCA.

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Abstract

The potential impact of ionising radiation from enhanced exposure to Naturally Occurring Radioactive Materials (NORM) to humans and the environment is not currently accounted for sufficiently in Life Cycle Assessment (LCA). Here we present midpoint and endpoint characterisation factors resulting from the implementation of impact assessment models for human health and ecosystems for NORM exposure. These models build upon existing fate, exposure and effect models from the LCA and radiological literature. The newly developed models are applied to a theoretical study of the utilisation of bauxite residue, a by-product of alumina processing enriched in natural radionuclides, in building materials. The ecosystem models have significant sensitivity to uncertainties surrounding the differential environmental fate of parent and daughter radionuclides that are produced as a part of decay chains, and to assumptions regarding long term releases from landfill sites. However, conservative results for environmental exposure suggest that in addition to landfill of materials, power consumption (burning coal and mining uranium) is a potentially significant source of radiological impact to the environment. From a human perspective, exposure to NORM in the use phase of building materials is the dominant source of impact, with environmental releases of nuclides playing a comparatively minor role. At an endpoint level, the impact of NORM exposure is highly significant in comparison to other impact categories in the area of protection of human health. This highlights the importance within LCA of having sufficient impact assessment models to capture all potential impacts, such that issues of burden shifting between impact measures can be captured, interpreted and resolved in the optimisation of product systems.

Highlights

- A new life cycle impact assessment model for exposure to NORM is presented
- Midpoint and endpoint characterisation factors for humans and ecosystems are given
- The models are validated with respect to bauxite residue valorisation systems
- The importance of burden shifting within life cycle impact assessment is raised

Keywords

LCA impact category; NORM; Construction materials; Bauxite residue; Burden shifting

1. Introduction

Ionising Radiation has long been recognised as an impact worthy of focus in LCA (Heijungs et al., 1992; Solberg-Johansen et al., 1997). Impact models for ionising radiation resulting from the nuclear fuel cycle have subsequently been developed within LCA for human impact (Frischknecht and Braunschweig, 2000), and for freshwater organisms (Garnier-Laplace et al., 2009), however exposure to natural sources of radiation, the most prevalent source of ionising radiation exposure (UNSCEAR, 2000) is not currently accounted for.

As stated in the ILCD handbook (European Commission, 2011) extension of the number of radionuclides covered by ionising radiation life cycle impact assessment models for both human health and biota is a high priority task. Joyce et al. (2016) have set out a framework for the inclusion of Naturally Occurring Radioactive Materials (NORM) exposure in the LCA framework. In this paper, we outline implementation of this framework and the validation of the models produced. To fulfil this aim, scenarios for Bauxite Residue (BR) utilisation in construction materials are assessed. Bauxite residue is a by-product of alumina industries and is produced in vast quantities worldwide (an estimated 150 million tonnes per year (Evans, 2016)). There is an increasing interest towards the valorisation of this residue (European Commission, 2014, 2008; MSCA-ETN REDMUD, 2015) with its incorporation into construction materials representing a potentially viable solution (Klauber et al., 2011). BR however has concentrations of ^{238}U and ^{232}Th greater than in the bauxite ore from which it is derived, and as such its utilisation raises the issue of NORM exposure.

In this paper, we outline the implementation of the Joyce et al. framework in the development of new impact assessment models for exposure to NORM for both humans and ecosystems. We then demonstrate the application of the models in the assessment of BR valorisation. We use this assessment to validate and evaluate the models and their suitability for application in LCIA.

2. Methods

LCIA characterisation models for enhanced exposure to NORM were developed according to the framework set out in Joyce et al. (2016). This implementation is described in section 3 below, and is available as an Excel file in the online supplementary materials. Characterisation factors derived from the models were imported into SimaPro 8 (Pre Sustainability, 2014). A cradle-to-grave assessment of options for the utilization of BR in building materials, based on theoretical data, was carried out using the newly derived characterisation factors. This assessment is described in section 4.

To validate and verify the new model developed from the Joyce et al. (2016) framework, we need to display that the model (a) is useable, (b) represents the real world system with a sufficient level of accuracy and (c) matches current state of the art (Carson, 2002). To achieve this, we selected an approach, similar to the one described by Harder et al. (2014) and Heimersson et al. (2014) and performed impact method evaluation, sensitivity checks, and midpoint and endpoint indicator cross comparison.

Impact method evaluation was performed in accordance to scientific criteria set in the ILCD handbook (European Commission, 2010): completeness of scope; environmental relevance; scientific robustness and certainty; documentation, transparency and reproducibility; applicability. A sensitivity analysis of the modelling assumptions was performed, showing how choices and changes in the knowledge regarding model parameters may affect data and calculated outcomes.

Comparison of midpoint indicators for the human impact category was performed with impact indicators previously derived for a subset of the natural radionuclides (Frischknecht and Braunschweig, 2000) for environmental releases and for indoor exposure with Meijer et al. (2005a, 2005b). In the case of environmental impact assessment there is no previous data which we can use for comparison, therefore only a sensitivity check was performed.

The endpoint indicator for human health (DALY) was cross validated against existing human impact assessment models in the ReCiPe set of methods (Goedkoop et al., 2009), using the Hierarchist set of endpoint measures. These are human toxicity, particulate matter formation, photochemical ozone formation, climate change, ionising radiation and ozone depletion. The climate change and ozone depletion endpoint models are designated as interim by the ILCD (Hauschild et al., 2013).

3. Model framework implementation

The connection between numerical models selected for the NORM impact method structure are presented in Fig. 1 and Fig. 2 for humans health and ecosystems, following the framework proposed by Joyce et al. The framework and submodels included are further described below. The modelling process is divided into three main steps - assessment of radionuclide fate, exposure to concerned organisms and damage to the population or ecosystem.

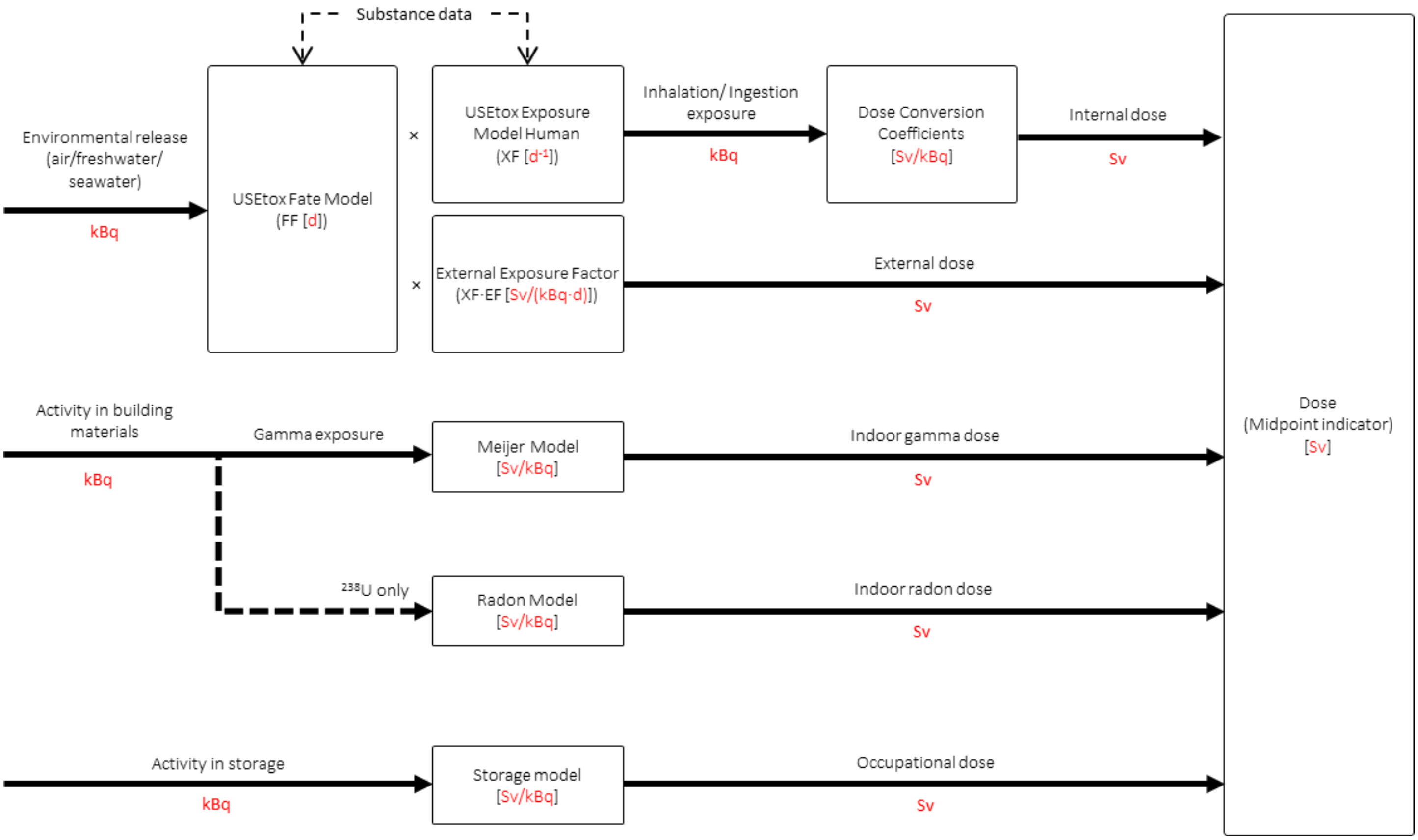

Fig. 1. Diagram of human impact assessment model. Units are presented in red.

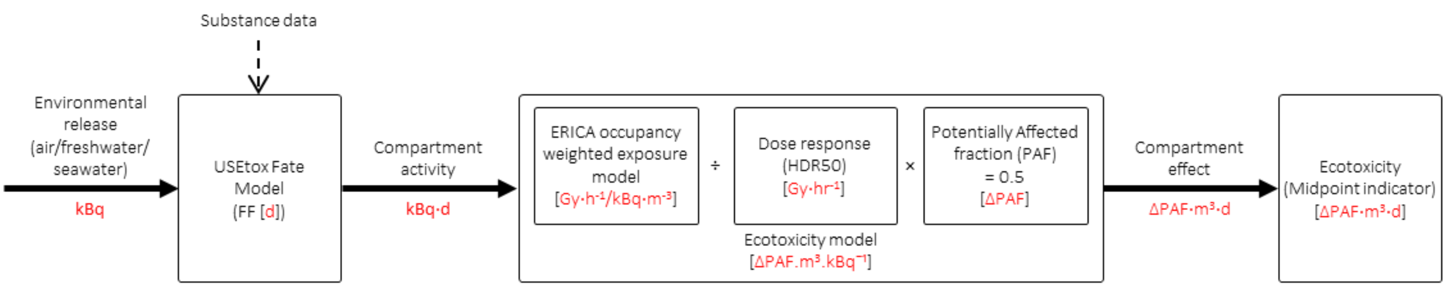

Fig. 2 Diagram of environmental impact assessment model for three ecosystems (terrestrial , freshwater and marine). The terrestrial ecosystems represents both aerial and land living organisms. Units are presented in red.

3.1 Fate and exposure analysis – midpoint model

3.1.1 Environmental releases, human health and ecosystem impact

The human impact assessment model for environmental releases is described in the upper part of Fig. 1. USEtox is a Life Cycle Impact Assessment model developed for characterization of human and ecotoxicological impacts (Huijbregts et al., 2015a; Rosenbaum et al., 2008). Implementation of the USEtox (current version 2.01; Huijbregts et al., 2015b) fate (FF) and exposure (XF) models for environmental releases of radionuclides required the collection of substance data for the NORM nuclides considered. These data and the data sources are shown in Appendix A (Sheet: Ecotox Effect Parameters).

For human receptors, the USEtox model calculates exposure factors for exposure via inhalation and ingestion routes. These factors are multiplied by the Dose Conversion Coefficients (DCCs) taken from UNSCEAR (2000) (units of Sv/kBq) to provide the effective dose via these routes. DCCs are used to convert radionuclide concentration into dose to recipients based on the decay mode and decay energy of the radionuclide. It is common to distinguish between external (exposure from radionuclide presence near recipient) and internal (exposure from inhalation or ingestion of radionuclide) DCCs.

In addition to the routes included in USEtox, external exposure to radionuclides present in soil is also a possible impact pathway. This was calculated using DCCs from Eckerman and Ryman (1993) for soil contaminated to infinite depth (original units $(Sv \cdot m^3)/(Bq \cdot s)$ converted to $(Sv \cdot m^3)/(kBq \cdot d)$). This is consistent with the assumption of instantaneous homogenous mixing made in the derivation of fate factors by USEtox. For a known soil volume (V_s), these DCCs can be converted to act as a combined exposure and effect factor for an isotope i (XF, units: $1/d$; EF, units: Sv/kBq; XF·EF, units: $Sv/(kBq \cdot d)$), as presented in equation (1).

$$(XF \cdot EF)_i = \frac{DCC_i}{V_s} \tag{1}$$

The environmental impact assessment model is shown in Fig 2. USEtox is used for the fate model. The effect model in USEtox only includes freshwater ecotoxicity. Here we also include potential impact for organisms in marine and terrestrial environments. For biota, the radiation dose received depends on the habitats the organism occupies within the environment. In order to address this, we implemented an occupancy weighted exposure model allowing to weight exposure coming from multiple environmental compartments to a single species (e.g. freshwater reptiles spending part of their time in water and part inside sediment). The occupancy factors were taken from the ERICA database. The USEtox model does not include an explicit compartment for freshwater and marine sediments, as it only considers freshwater toxicity, with exposure via water only. To estimate exposure from sediment, the model utilises Kd values (represented as ratio of concentration in sediment over concentration in water) obtained from the ERICA database (Brown et al., 2008), to derive equilibrium sediment concentration.

For external exposure DCCs were directly applied to convert radionuclide concentration into doses to species. For internal exposure Concentration Ratios (CR), the ratio of the equilibrium concentration of the isotope within an organism compared to the concentration of that isotope in the environmental compartment where the organism resides, were applied to calculate internal concentrations and then internal exposure DCCs used to calculate doses.

For biota, ^{238}U and ^{232}Th decay product DCCs, CRs and Kd values are available in the ERICA database for groups of reference organisms (ICRP, 2008) selected to represent each ecosystem (terrestrial, marine, freshwater). ^{40}K is not fully represented within the ERICA database; however the ERICA tool allows DCCs for ^{40}K to be derived for all of the reference organisms. For CR, values for a biogeochemical analogue element (caesium), which has similar behaviour under same conditions (IAEA, 2014; Newman, 2010) were obtained from ERICA. For water Kd values, recent measurements by Takata et al. (2010) have determined partitioning values for potassium in the estuaries of four rivers of Japan to be equal 7.9 l/kg. These measurements were performed for river estuaries, intermediary systems between river and sea, thus we assume this value for both seawater and freshwater compartments.

At this stage, we assess environmental fate of ^{238}U and ^{232}Th, assuming that their decay products reside in the same compartment as the mother nuclides. Sensitivity assessment of this assumption is provided in section 5.4.

3.1.2 Exposure from materials (human impact only)

Where NORM nuclides are incorporated into building materials, exposure of building inhabitants via gamma exposure and radon inhalation must be taken into account. The gamma exposure is described by the room model provided by Meijer et al. (2005a). Meijer et al. also provide empirical values for radon exhalation for a range of materials; however there is no mechanism to assess the potential radon exhalation of novel materials. In order to enable the inclusion of such materials, we use the radon surface exhalation model from UNSCEAR (2000) to obtain radon flux density and then concentration of radon in inhabited area based on ^{226}Ra concentration and material properties.

3.1.3 Exposure from material storage and landfill (human impact only)

NORM material has the potential to expose workers in close proximity to ionising radiation during its storage at industrial sites, particularly in landfill. However, the impact of a given kg of material deposited in a landfill will be attenuated by the shielding effect of subsequent layers of material. We use Markkanen's (1995) model in order to assess dose rates to workers near the storage sites based on the timing of deposition and thickness of the site. The modelling assumptions are listed in Appendix A (Sheet: Managed Storage).

3.1.4 Midpoint indicator, human impact

For impact to humans, each of the models described above yields collective radiation dose for a given inventory flow (kBq), measured in Sieverts (Sv). These can be summed to provide the midpoint indicator for human impacts, with the unit of man.Sv (a unit, used to represent collective dose for the entire population).

3.1.5 Midpoint indicator, environmental impact

For impact to ecosystems, we incorporate the methodology developed by Garnier-Laplace et al. (G-L) (2009), which utilizes the concept of Potentially Affected Fraction of species (ΔPAF) in line with established LCA methodology (Pennington et al., 2004) and allows us to obtain a single indicator considering all reference organisms within a given ecosystem. The concept unites different lethal and non-lethal effects to organism groups based on the level of chronic exposure in such a way, that we end up with a single effect factor for every ecosystem (terrestrial, freshwater, marine) per 1kBq radionuclide released. In addition to the freshwater ecosystem studied in G-L model, we consider marine and terrestrial environments based on the data available from the FREDERICA database (Agüero et al., 2006; Copplestone and Hingston, 2006). This database contains summaries of experimental studies on species sensitivity to chronic ionising radiation exposure.

Equation (2) predicts ΔPAF_e for an ecosystem *e* (terrestrial, marine, freshwater), where ΔC_e is an isotope concentration change in the receiving compartment and $HC_{50,e}$ is Hazardous Concentration affecting 50 % of species per ecosystem.

$$\Delta PAF_e = \frac{0.5 \cdot \Delta C_e}{HC_{50,e}} \qquad (2)$$

$$EC_{50,e,o} = \frac{HDR_{50,e}}{F_o} \qquad (3)$$

$HC_{50,e}$ is a geometric mean of half maximal effective concentrations $EC_{50,e,o}$ for all reference organisms *o* per ecosystem *e* defined in the equation (3) as the ratio of dose rate associated with 10 % effect for 50 % of species over full dose conversion coefficient F_o. This coefficient represents combined internal and external dose to an organism from an isotope. $HDR_{50,e}$ is a geometric mean value for an ecosystem of $EDR_{10,e,o}$ which shows chronic dose rate giving 10 % effect increase per organism per effect (mortality, morbidity, reproduction reduction) (Copplestone et al., 2008). It should be noted that this is a major difference from chemical toxicity models (including USEtox (Rosenbaum et al., 2008)) where instead of EDR_{10}, EC_{50} (the effective concentration at which 50% of organisms are affected) are used. Full dose conversion coefficient F_o is derived in equation (4) (Beresford et al., 2007) for terrestrial species (DCC_{ext} is derived for different exposures based on species habitats- in soil, on soil or in air). For marine and freshwater ecosystems, we use equations (4) - (6); for species living in water column (4), on sediment surface (5) and in sediment (6) using external and internal dose conversion coefficients.

$$F_o = DCC_{ext,o} + DCC_{int,o} \cdot CR_o \tag{4}$$

$$F_o = 0.5 \cdot DCC_{ext,o} \cdot (1 + Kd) + DCC_{int,o} \cdot CR_o \tag{5}$$

$$F_o = DCC_{ext,o} \cdot Kd + DCC_{int,o} \cdot CR_o \tag{6}$$

The model above yields characterisation factors in units of ΔPAF·m³·d/kBq for each ecosystem (freshwater/marine/terrestrial) for a given inventory flow. Hence, the midpoint indicator for environmental impacts has units of ΔPAF·m³·d.

3.2 Damage analysis – endpoint model

3.2.1 Endpoint indicator, human impact

The Disability Adjusted Life Years (DALY) concept (Murray, 1994) is used as a damage criterion. A Sv to DALY conversion factor (1.51 DALY/man.Sv) was obtained from Frischknecht and Braunschweig (F & B model) (Frischknecht and Braunschweig, 2000) for egalitarian/hierarchical scenario, yielding a final unit of DALY as an endpoint indicator.

3.2.2 Endpoint indicator, environmental impact

The potentially disappeared fraction (PDF) concept is used as the endpoint indicator for ecosystems. The severity factor from the USEtox model (0.5 PDF/PAF) is used (Huijbregts et al., 2015a) to convert from the midpoint indicator. The final unit is ΔPDF·m³·d as the endpoint indicator.

4. Case study

4.1.1 Goal and Scope

The application of the impact assessment models presented above was demonstrated for the valorisation of bauxite residue (BR) in the European Union. This takes the form of a cradle to grave assessment of BR utilisation in building materials. Three scenarios were considered, (1) the 'business as usual' scenario, in which the BR is landfilled at the production site; (2) utilisation of BR in bricks at a pre-fired composition of 30% BR; (3) utilisation in ordinary Portland cement, produced from a raw meal including 3% BR. The functional unit for all three scenarios is the treatment of 1 kg of bauxite residue, and both cement and bricks are used in the construction of dwellings.

The outputs of each scenario are different, and therefore an 'equal basket of benefits' approach was taken, a technique which has been used for previous comparisons of waste valorisation systems (e.g. Barrera et al., 2016; Vandermeersch et al., 2014). This approach is based on the concept of system expansion, which is a common approach for life cycle assessments of waste management systems (Finnveden, 1999, Eriksson et al, 2002). The reference flow for each of these scenarios is functionally equivalent, and where products are not produced by the valorisation system, they are considered to be produced by the traditional supply chain (TSC) (Fig. 3).

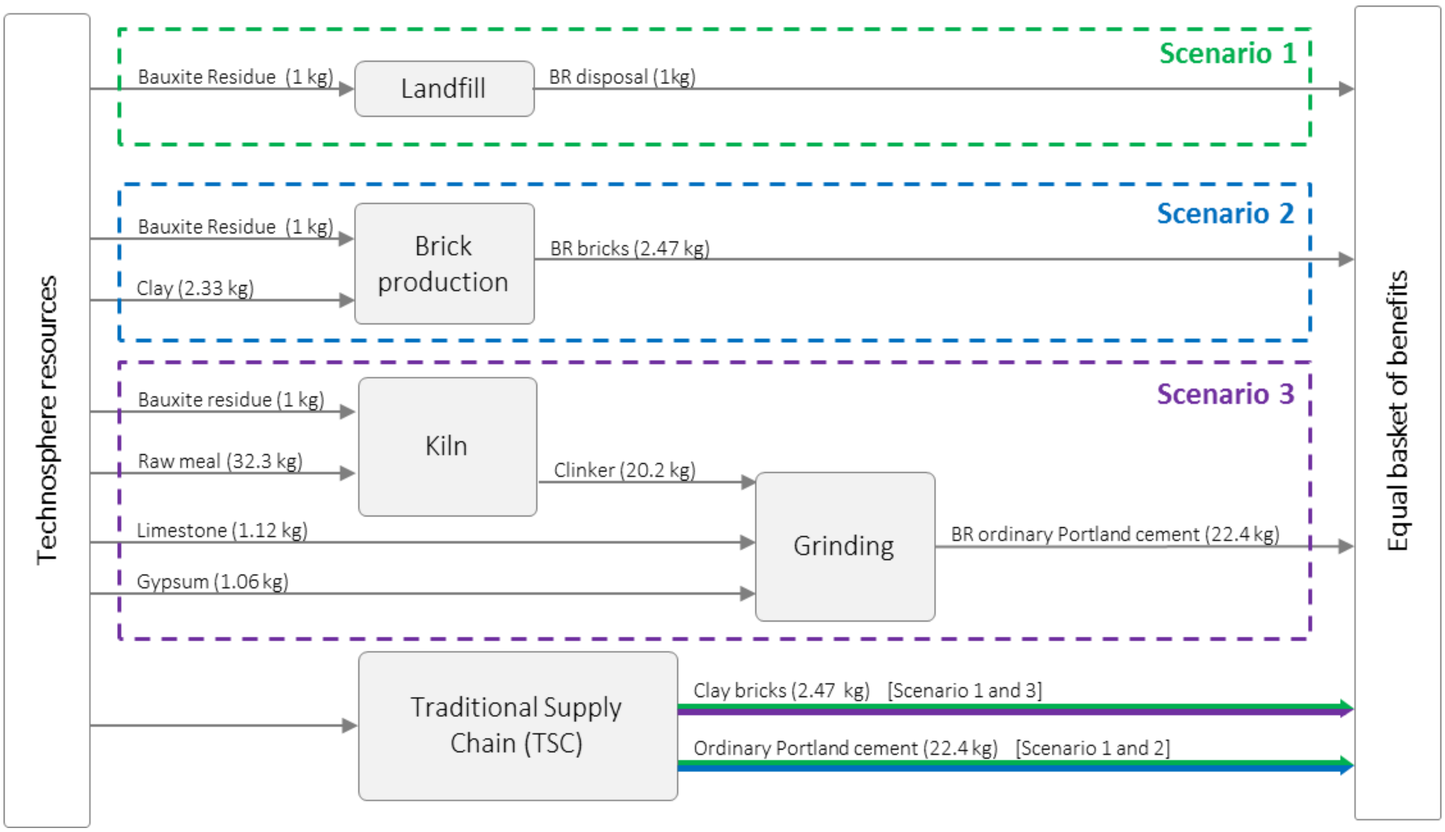

Fig. 3 Scenario flow chart. Mass differences are due to firing of bricks and cement.

An attributional system model is used. BR is considered to be a waste product of the Bayer process. Since all systems treat the same amount of waste, all processes upstream are identical in all studied cases and can be deleted in the comparison (c.f. Clift et al., 2000; Finnveden, 1999). In other words, BR therefore enters the system 'burden free'.

The production of the raw materials required, processing of these into building materials, residential use of the finished materials and their subsequent disposal are also included in the analysis. A lifetime of 75 years is assumed for the building materials. It is assumed that building materials are disposed of in an inert materials landfill, i.e. a landfill which is assumed to have no leaching. This is a simplification which is further discussed below.

Transport of the finished material to the site, the building of the residence and its subsequent demolition are excluded from the analysis. These data are likely to be highly variable, but sufficiently similar between scenarios that their exclusion is warranted.

4.1.2 Inventory analysis

We used the Ecoinvent 3.2 database (Weidema et al., 2013) as the basis of the inventory analysis. For each of the valorisation options, additional assumptions were required regarding the addition of BR. Despite the publication of over 1200 patents and numerous successful trials, the valorisation of BR at an industrial scale is still very much in its infancy: less than 3% of the BR produced annually is productively utilised (Evans, 2016). As a result, there is no mature industrial process for which data for the use of BR as an input for brick or cement production can be gathered. That said, there have been numerous studies in which BR has been used as an addition to the standard brick and cement making technologies without further modifications to these processes, and with resulting products functionally equivalent to the reference material (e.g. Pontikes, 2007; Tsakiridis et al., 2004; Vangelatos et al., 2009).

From a life cycle inventory standpoint this allows us to use the inventory data for the original processes, substituting a certain proportion of the input materials with BR. For brick production, Pontikes (Pontikes, 2007) has shown that for additions to standard brick production of up to 30% BR result in bricks with similar mechanical properties. Likewise, Vangelatos et al. (2009) demonstrated that addition of up to 3% BR to ordinary Portland cement (OPC) production yielded material with comparable properties.

As a result, for scenario 2 we assume that BR replaces 30% of the wet (pre-firing) mass of the clay described in the Ecoinvent 3.2 process as being used to make the brick. For scenario 3, we assume 3% of the mass of the raw meal entering clinker production is BR, with all other constituents of the raw meal in the Ecoinvent 3.2 process for clinker production reduced in the same proportion. The output of this modified clinker production is then used as an input to the Ecoinvent 3.2 cement production process, with no further modification to this process.

Average European production processes are used in all cases, to represent a generic EU-wide system.

4.1.3 Radiological properties of materials

The application of the NORM impact models described above require that the activity of NORM nuclides in the final product is known. The radiological characteristics of the BR used in this assessment are those of Greek bauxite residue from Aluminium of Greece (own measurements, Table 1 ; primary data in Appendix A (Sheet: Greek BR measurements)).

Table 1 Activity concentration of ^{238}U, ^{232}Th and ^{40}K per kg (dry weight) of Greek bauxite residue and TSC materials

Material	Activity concentration (Bq/kg)			Reference
	^{238}U	^{232}Th	^{40}K	
Bauxite Residue	147	426	26	Own measurements
Brick	47	48	598	Trevisi et al. 2012
Cement	45	31	216	Trevisi et al. 2012
Gypsum	15	9	91	Trevisi et al. 2012 (Natural gypsum)
Limestone	21	24	333	Trevisi et al. 2012 (Sedimentary stones)
Clinker	48	33	217	Calculated from activity concentration in cement accounting for contribution from gypsum and limestone

The radiological properties of materials from the traditional supply chain utilised in the three scenarios are taken from Trevisi et al. (2012), the overall average values for the EU are used. Data for clinker are not directly available, so activity concentration was calculated from that for cement, accounting for the contribution of gypsum and limestone to the total (cement composition: 90.25% clinker, 4.75% gypsum, 5% limestone (Ecoinvent, 2015)).

Radon emanation fraction (amount of radon atoms leaving material granules, unitless) is taken to be 0.2 for concrete (Markkanen, 1995) and 0.035 for bricks (Bossew, 2003); diffusion length (units $m^2 \cdot s^{-1}$) for concrete is $3.0 \cdot 10^{-8}$ and for bricks $1.9 \cdot 10^{-7}$ (Nazaroff and Nero, 1988). ^{238}U and ^{232}Th decay series are assumed to be in secular equilibrium in all materials.

4.1.4 BR disposal

Residual material landfill processes in the Ecoinvent database include the short term (present to 100 years) and long term (100 to 60,000 years) release of elements in the waste to groundwater. In order to apply the NORM impact models to the landfilling of BR in residual material landfill, the subsequent release of these nuclides to groundwater had to be approximated.

Data for potassium emissions from residual landfill (specifically *'Redmud from bauxite digestion| treatment of, residual material landfill'*) are available in Ecoinvent (Doka, 2009), with 28.19% of emissions occurring in the short term, and the remainder (71.81%) in the long term. No data are available for uranium and thorium, therefore following the recommendation of Joyce et al. (2016), we begin with the default assumption that in the long term 100% of the nuclides are emitted to the environment from landfill over the

60,000 year period, and that this release is linear, with 1/600 of the releases taking place over the first 100 years. The sensitivity of the model to this assumption is assessed.

For disposal of materials in inert material landfill (such as those for construction and demolition waste) the Ecoinvent database assumes no emissions to groundwater, short or long term. The sensitivity of the model to this assumption is assessed also.

5. Case study results

5.1 NORM Exposure (Human) – emissions vs materials

The potential human health impacts of NORM exposure in the different scenarios are presented in Fig. 4 and Table 2**Error! Reference source not found.**. From a human health perspective, the overall impact at the midpoint level (dose) is similar across the three scenarios, with the landfill of BR scenario having the lowest overall impact, and utilisation of BR in cement the highest (Table 2). In all three scenarios the impact to human health resulting from NORM contained in building materials is many orders of magnitude greater than that caused as a result of emissions of NORM nuclides to the environment. Looking at the processes in the life cycle which contribute to radiological impact it is clear that while the majority of NORM exposure is derived from the presence of NORM nuclides in standard building materials (those from the traditional supply chain), the addition of BR to construction materials increases this exposure (red) in Scenarios 2 and 3. The slightly higher permeability of concrete than bricks to radon leads to a higher radon dose due to the inclusion of BR in cement (Scenario 3). Radiological impacts from the production of building materials (orange, although not visible) and additional exposure to workers at the BR disposal site (purple, although not visible) are negligible.

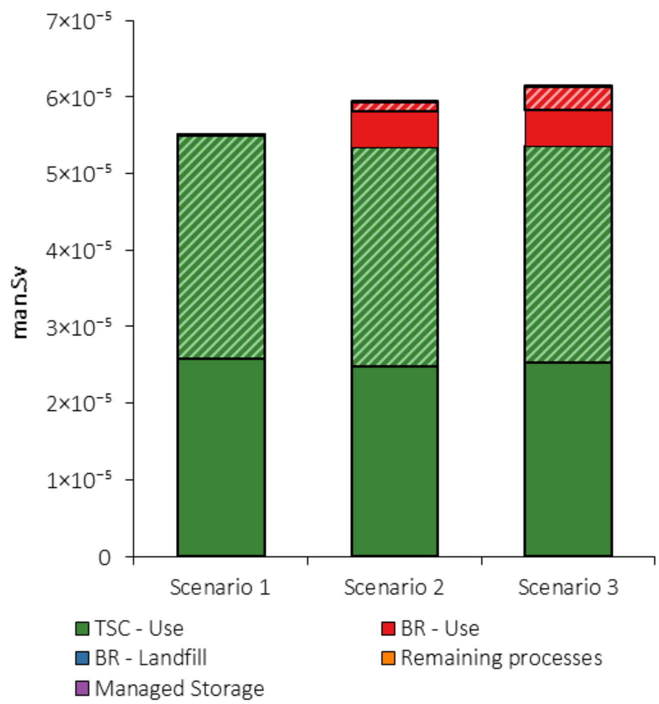

Fig. 4. Processes contributing to NORM exposure in humans.
TSC: Traditional Supply Chain. Solid bar: gamma doses, hatched bar: radon dose

5.2 NORM Exposure and potential impacts (Ecosystems)

The potential ecosystem impacts of NORM exposure in the different scenarios are presented in Table 2 and Fig. 5. Disposal of BR in residual material landfill (Scenario 1) has the highest radiological impact to biota across all three ecosystems (Table 2). This is entirely driven by long term emissions from landfill of radionuclides to groundwater. Fig. 5 presents results for midpoint impact to ecosystems resulting from each NORM nuclide release, along with the compartment into which these flows are released. For freshwater ecosystems the impact as a result of these long term emissions is multiple orders of magnitude greater than any other source of radiological impact across the life cycle. Releases of thorium are the main source of impact here.

For marine ecosystems, long term emissions of uranium from landfill are the major contributor, although releases of polonium to air and freshwater from coal power, resulting from the burning of the coal and the disposal of wastewater from flue gas desulphurisation respectively, represent a substantial contribution to this impact in all three scenarios.

For terrestrial ecosystems, while long term emissions of uranium and thorium from landfill of BR are the differentiating emissions between scenarios, the highest source of impact across all three scenarios is the release of ^{226}Ra to air and freshwater as a result of uranium mining. The use of nuclear energy in the production of clinker in France (as part of average EU production) is the ultimate source of this impact. Airborne releases of ^{210}Pb from coal power production also contribute to this impact.

It should be noted that these results are highly contingent on the assumption that there are no releases to groundwater from inert material landfill in either the short or long term. Consequently there are no releases of the radionuclides that occur in both BR and traditional building materials to groundwater in scenarios 2 and 3. The sensitivity of the results to this assumption is presented in section 5.4.3 below.

Table 2 Midpoint results for NORM impact assessment models

Area of Protection	Impact	Unit	Scenario 1	Scenario 2	Scenario 3
Human Health	**Releases to Environment**	**man.SV**	$1.5 \cdot 10^{-8}$	$4.2 \cdot 10^{-9}$	$4.2 \cdot 10^{-9}$
Human Health	**Built Environment - Gamma dose**	**man.SV**	$2.6 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$3.0 \cdot 10^{-5}$
Human Health	**Built Environment - Radon dose**	**man.SV**	$2.9 \cdot 10^{-5}$	$3.0 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$
Human Health	**Managed Storage**	**man.SV**	$4.7 \cdot 10^{-10}$	-	-
Human Health	***Total***	***man.SV***	*$5.5 \cdot 10^{-5}$*	*$5.9 \cdot 10^{-5}$*	*$6.1 \cdot 10^{-5}$*
Ecosystems	**Freshwater**	**PAF·m³·d**	$7.0 \cdot 10^{-1}$	$8.3 \cdot 10^{-3}$	$8.2 \cdot 10^{-3}$
Ecosystems	**Marine**	**PAF·m³·d**	1200	302	300
Ecosystems	**Terrestrial**	**PAF·m³·d**	$2.4 \cdot 10^{-4}$	$1.7 \cdot 10^{-4}$	$1.7 \cdot 10^{-4}$

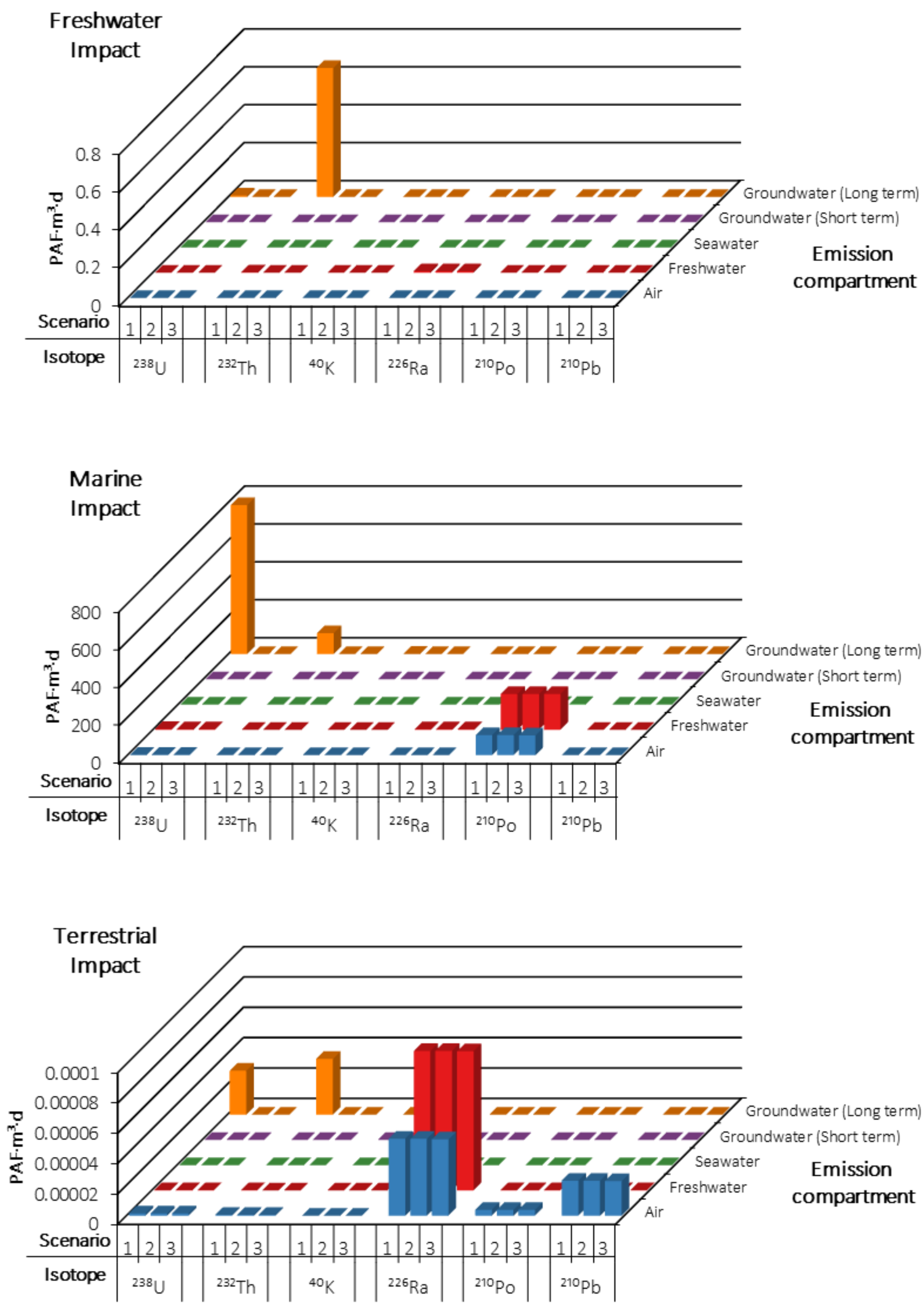

Fig. 5 Inventory flows contributing to midpoint NORM impact to ecosystems by compartment.
Scenario 1: BR to landfill, Scenario 2: BR utilised in brick production, Scenario 3: BR utilised in cement production

5.3 Impact method evaluation

In order to evaluate a newly developed characterisation method for inclusion of NORM in LCIA, we use guidelines developed and used by the ILCD Handbook, as applied by Heimersson et al. (2014) to conduct a qualitative assessment of the proposed methods. The results are displayed in Table 3.

Table 3 Impact evaluation

Assessment criterion	Assessment
Completeness of scope	The developed method covers most relevant impact mechanisms for human health and natural environment. The methodology is overall globally applicable (while for construction materials some additional information regarding material properties is needed).
Environmental relevance	All critical parts of environmental mechanisms are covered in the method by approved and widely used toxicological and radiological concepts.
Scientific robustness and certainty	The method relies on scientifically accepted characterization factors and numerical models. It can be easily improved and extended once updated information becomes available.
Documentation, transparency and reproducibility	This article provides description of the method and main equations used, while the modeling steps and input data either are referenced or are published in the form of appendixes. All the assumptions used and value choices made are explicitly reported.
Applicability	The characterization factors presented can be directly applied to describe natural radionuclides, or the method provided can be used to update existing or derive new factors.

5.4 Sensitivity analyses

During the modelling stages, some assumptions have been made, where there was a lack of knowledge (i.e. radionuclide leaching from landfill sites) or adequate models did not exist (i.e. redistribution of radionuclide decay products in the environment). In the current section model sensitivity to most significant assumptions is tested and their possible influence on the obtained results is discussed.

5.4.1 Fate of daughter nuclides

The proposed model is based on the assumption that daughter nuclides reside in the same compartment as their mother nuclides. This is true for isotopes incorporated into construction materials (with the exception of radon, which we consider using a separate model). However, in the environment mechanisms for radionuclides transfer might differ, meaning that daughter nuclides would have different fate compared to primordial isotopes. To assess the influence of this assumption we derive damage factors with and without considering radionuclide redistribution in the environment and present them in Table 4. Damage factors for humans and terrestrial/marine/freshwater ecosystems derived for three considered release pathways- air, freshwater and seawater are presented for a scenario assuming no redistribution (used in the current study) and for a scenario assuming isotope redistribution. The latter is modelled by applying the following principle: when decay happens and a new isotope is produced, it is released from this compartment (we consider isotopes with half-lives > 10 days, assuming that shorter lived ones would decay before they can leave current compartment). The fate of the new isotope is found by applying USEtox model and exposure is calculated for the newly obtained environmental concentrations. The process is repeated through the whole decay chain and then cumulative damage factors for ^{238}U are obtained and compared with no redistribution scenarios (currently proposed model). Thus we have two boundary scenarios (with zero or all daughter nuclides release, whilst the real case would be somewhere in between) providing us with the ranges of possible uncertainty due to assumption introduced.

Table 4 Environmental damage factors derived for ^{238}U considering proposed scenario in the article (no daughter radionuclide redistribution) and scenario with redistribution of daugher radionuclides. Damage factors are cumulative, considering whole decay chain of ^{238}U.

Cumulative damage factor for ^{238}U	Proposed scenario			Scenario with redistribution		
	Release compartment			Release compartment		
	Air	Freshwater	Seawater	Air	Freshwater	Seawater
Damage to humans (endpoint, DALY/kBq$_{emitted}$)	$2.39 \cdot 10^{-6}$	$1.86 \cdot 10^{-6}$	$8.0 \cdot 10^{-7}$	$5.95 \cdot 10^{-7}$	$8.11 \cdot 10^{-7}$	$7.92 \cdot 10^{-7}$
PDF terrestrial (midpoint, PDF.m3.day/kBq$_{emitted}$)	$3.72 \cdot 10^{-2}$	$5.12 \cdot 10^{-4}$	$5.10 \cdot 10^{-20}$	$1.42 \cdot 10^{-2}$	$4.59 \cdot 10^{-4}$	$5.17 \cdot 10^{-20}$
PDF freshwater (midpoint, PDF.m3.day/kBq$_{emitted}$)	$9.03 \cdot 10^{-1}$	2.55	$1.13 \cdot 10^{-18}$	$4.39 \cdot 10^{-2}$	$7.24 \cdot 10^{-2}$	$1.16 \cdot 10^{-18}$
PDF marine (midpoint, PDF.m3.day/kBq$_{emitted}$)	$9.75 \cdot 10^{5}$	$1.44 \cdot 10^{6}$	$1.45 \cdot 10^{6}$	$9.83 \cdot 10^{5}$	$1.44 \cdot 10^{6}$	$1.45 \cdot 10^{6}$

The main observations are that the current assumption, i.e. "no redistribution":

1. Differs 4 and 2 times for air and freshwater releases from "redistribution" scenario, while being more conservative. In the USETOX model seawater compartment serves a role of the sink, where most of the elements eventually end-up. With every redistribution iteration, more radionuclides leave their environmental compartments and end up in the seawater.
2. Has minor effect for terrestrial and marine ecosystem, as well as for seawater releases. Even though more radionuclides end up in seawater compartment with every iteration, their number is minor compared to initial ^{238}U amount.
3. Strongly affects damage factor for freshwater ecosystem in case of freshwater release.

In summary, for human and seawater ecosystem, the current assumption provides good level of accuracy, while for terrestrial and freshwater ecosystems our approach tends to be conservative and provides an interim solution only for radionuclides with complex decay chain (i.e. ^{40}K and ^{210}Po do not have secondary decay products and therefore are modelled accurately).

5.4.2 Landfill – short vs long term releases of radionuclides to groundwater

Based on the recommendations in Joyce et al. (2016), for residual material landfill in Scenario 1 it was assumed that in the long term 100% of ^{232}Th and ^{238}U are released to the environment, with 1/600th of the emissions occuring in the short term. As shown in Fig. 5, long term emissions of these two nuclides to groundwater contribute significantly to all ecosystem impact categories, accounting for 98.7% of total impact for freshwater ecosystems, 74.7% of total impact for marine ecosystems, and 27.7% of total impact for terrestrial ecosystems. In each ecosystem, ^{238}U and ^{232}Th are the major contributors to this impact.

Long term emissions to groundwater account for 55% of the impact to human health as a result of releases to the environment in Scenario 1. While this only represents 0.015% of the total human health impact of this particular scenario (as a result of the overriding influence of NORM in building matrerials), for systems where NORM materials are not utilised in construction this may represent a substantial contribution to NORM exposure. However, the impact mainly derives from long term releases of ^{40}K (Fig. 6) for which ecoinvent data is available (28.19% released in the short term, with 100% eventual release). The impact of the assumption of 100% eventual release of ^{238}U and ^{232}Th is therefore diminished, as long term releases of these nuclides only account for 14% of the total impact from environmental releases.

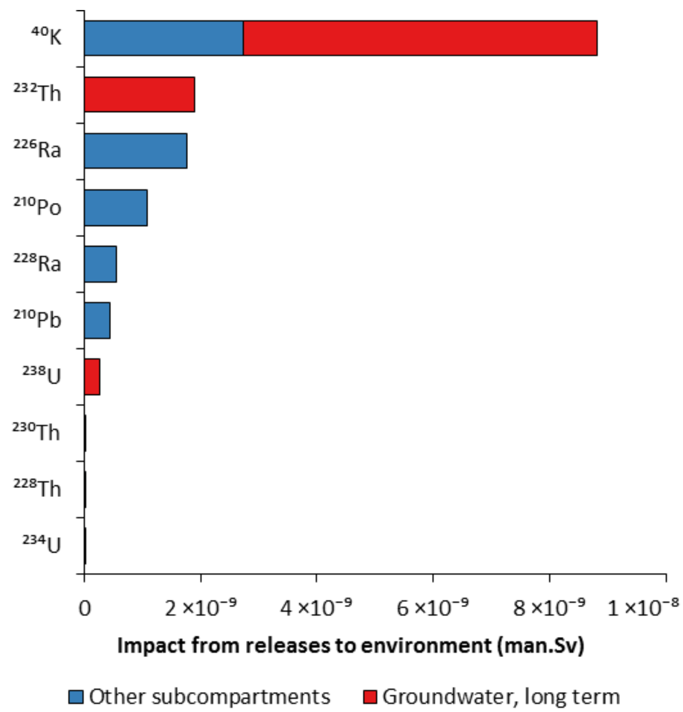

Fig. 6. Human Health model contribution of long-term emissions to releases midpoint

5.4.3 Emissions from inert landfill

Applying the same assumptions regarding the short and long term releases of radionuclides to groundwater from residual landfill to inert landfill has a limited effect on the overall results for the human health models, with the impact increasing by 3.8%, 3.2% and 3.3% for scenarios 1, 2 and 3 respectively. However, as above, for systems where NORM materials are not utilised in building products this is a potentially important assumption, as the impact from radionuclide releases from the 2.47 kg of brick and 22.4 kg of cement in inert landfill is nearly 200 times greater than that from the 1 kg of BR in residual material landfill in Scenario 1.

The effect is far greater for the ecosystem models (Fig. 7), where there is a higher sensitivity to long term emissions to groundwater. The effects of ^{238}U and ^{232}Th on all ecosytems are exaggerated, due to their presence in the TSC materials. TSC materials also contain substantially higher levels of ^{40}K than BR (which is comparatively depleted in this element). For marine ecosystems, the higher levels of ^{40}K in TSC building materials means that the contribution of these emissions is almost equal to that of ^{232}Th (ratio ^{232}Th : ^{40}K = 1 : 0.92). In comparison, when only emissions from BR in residual material landfill are considered the impact of ^{232}Th to marine ecosystems is 87 times higher than that of ^{40}K (ratio ^{232}Th : ^{40}K = 1 : 0.0114).

For terrestrial ecosystems, inclusion of emissions from inert landfill means that long term groundwater emissions of ^{238}U from landfill overtake emissions of ^{226}Ra from the nuclear fuel cycle to become the dominant source of impact.

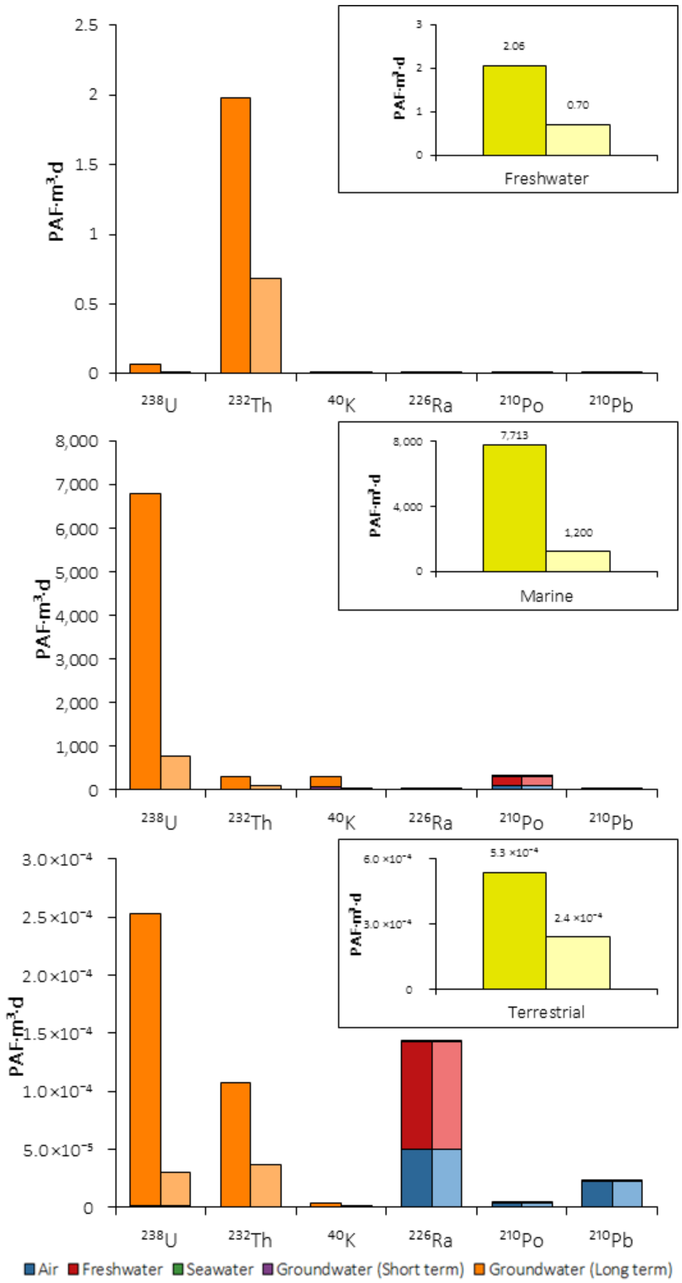

Fig. 7. Change in Ecosystem midpoint results as a result of the inclusion of emissions from inert landfill. Subplots show the effect on the overall total. Dark tones indicate inclusion of inert landfill emissions.

5.5 Characterisation factor cross-comparison

After developing a new impact assessment method and a set of characterisation factors, we would like to compare our results with existing state of knowledge. Human damage factors for environmental releases are compared in Table 3 to the Frischknecht and Braunschweig model for isotopes ^{226}Ra, ^{230}Th, ^{234}U and ^{238}U (isotopes that are presented in both methods). Results for uranium and thorium isotopes are within an order of magnitude, while for radium we predict damage factors 2-4 orders of magnitude higher than that in F & B model. The difference in results for ^{226}Ra is mainly attributed to ingestion part of assessment. F & B use data provided in (Dreicer et al., 1995) which is derived, assuming local radionuclide distribution (gas plume model, or river transfer models were used) while our approach utilises USEtox model, which shows significantly higher radium transfer to agricultural products, due to its chemical similarity to calcium.

Table 5 Damage factor comparison due to radionuclide release into environemnt.

Isotope	Damage Factor [DALY]					
	Air release		Freshwater release		Seawater release	
	F & B	Proposed model	F & B	Proposed model	F & B	Proposed model
^{226}Ra	$9.1 \cdot 10^{-10}$	$2.04 \cdot 10^{-6}$	$1.3 \cdot 10^{-10}$	$8.45 \cdot 10^{-8}$	-[1]	$5.32 \cdot 10^{-9}$
^{230}Th	$4.5 \cdot 10^{-8}$	$1.47 \cdot 10^{-7}$	-	$6.14 \cdot 10^{-9}$	-	$1.82 \cdot 10^{-9}$
^{234}U	$9.7 \cdot 10^{-8}$	$2.25 \cdot 10^{-8}$	$2.4 \cdot 10^{-9}$	$2.94 \cdot 10^{-9}$	$2.3 \cdot 10^{-11}$	$6.07 \cdot 10^{-10}$
^{238}U	$8.2 \cdot 10^{-9}$	$1.97 \cdot 10^{-8}$	$2.3 \cdot 10^{-9}$	$2.7 \cdot 10^{-9}$	$2.3 \cdot 10^{-11}$	$5.57 \cdot 10^{-10}$

For external gamma exposure assessment, we used the Meijer model, which is a recognised and accepted tool to describe indoor impact to humans. It also characterizes impact from radon exhaled from construction materials, while we have chosen UNSCEAR model. Our choice of model on the one hand is more basic and considers a house made completely of a single material, while on the other hand allows us to outline differences between various materials used, as well as to model novel types of materials which are produced using NORM. The damage factors for one Bq of ^{222}Rn for Meijer is $1.88 \cdot 10^{-10}$ DALY and is different from our numbers obtained for completely concrete and brick house respectively – $3.9 \cdot 10^{-9}$ and $1.89 \cdot 10^{-8}$ DALYs. This mainly comes from the differences in the modelled house- Meijer describes a house with a floor area of 39 m^2, while in UNSCEAR floor area is 100 m^2, thus total wall and exhalation area is significantly higher. The difference between exposure from concrete and bricks in our calculations is explained by radon exhalation rate and such is experimentally justified (Stoulos et al., 2003).

For environmental impact assessment, there are no damage factors for natural radionuclides in literature. In addition, we cannot directly compare derived damage factors with existing LCA impact categories (e.g. ecotoxicity) since we use a different method of damage factor derivation (EDR_{10} vs EC_{50}).

5.6 NORM Exposure (Human) – Endpoint cross validation

At the endpoint level (DALY) it is possible to compare the additional human health impact caused by NORM exposure to that caused through other means throughout the lifecycle of the products in question.

[1] No data

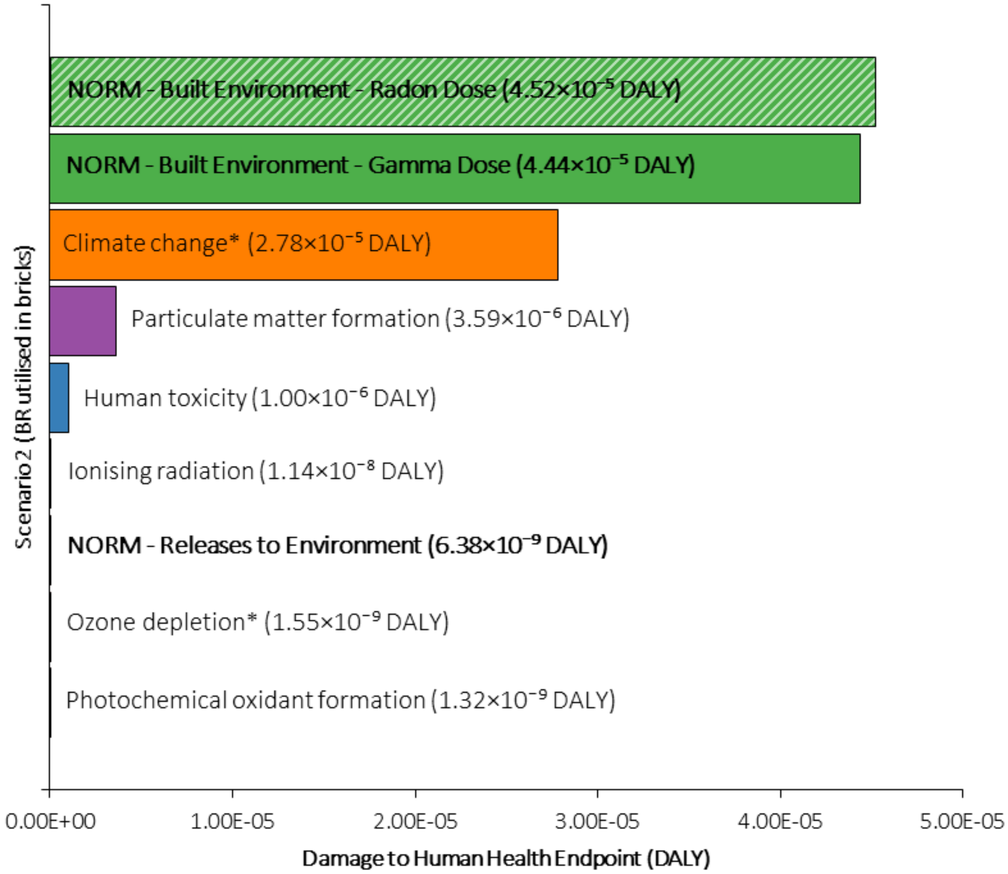

*Fig. 8. Total potential contribution to human health from different impact categories for Scenario 2. Bold type indicates new NORM endpoint measures, * indicates endpoint methods considered interim by ILCD*

Fig. 8 presents the total potential human health impacts resulting from Scenario 2. NORM exposure from the built environment is the overriding impact in all three scenarios. Climate change makes a substantial contribution to overall human health impact, while particulate matter formation and human toxicity are minor contributors. The effect of ionising radiation (from the nuclear fuel cycle), NORM exposure from releases to the environment, ozone depletion and photochemical oxidant formation are negligible in comparison.

5.7 Comparative impact of NORM exposure through materials

As a part of the method of validating and verifying the method, the results can also be compared with other sources of radiation. Using the reference dwelling of Meijer et al. and substituting the clay brick component of the building for 30% BR bricks leads to an additional collective dose over a 75-year lifespan of 0.026 man.Sv. This is equivalent to an extra dose of 0.12 mSv per occupant per year. Additionally substituting sand-lime bricks in the reference dwelling for BR augmented clay bricks results in an annual increase in per capita dose of 0.89 mSv.

Fig. 9 shows this dose in comparison to other common radiation sources. The level of increased annual dose resulting from clay brick substitution is smaller than the dose received as a result of some medical procedures and only slightly higher than the cosmic radiation dose received by taking a single transatlantic flight. Substituting all bricks results in an increase in dose comparable to about 68.5% of that of the average UK annual radon dose.

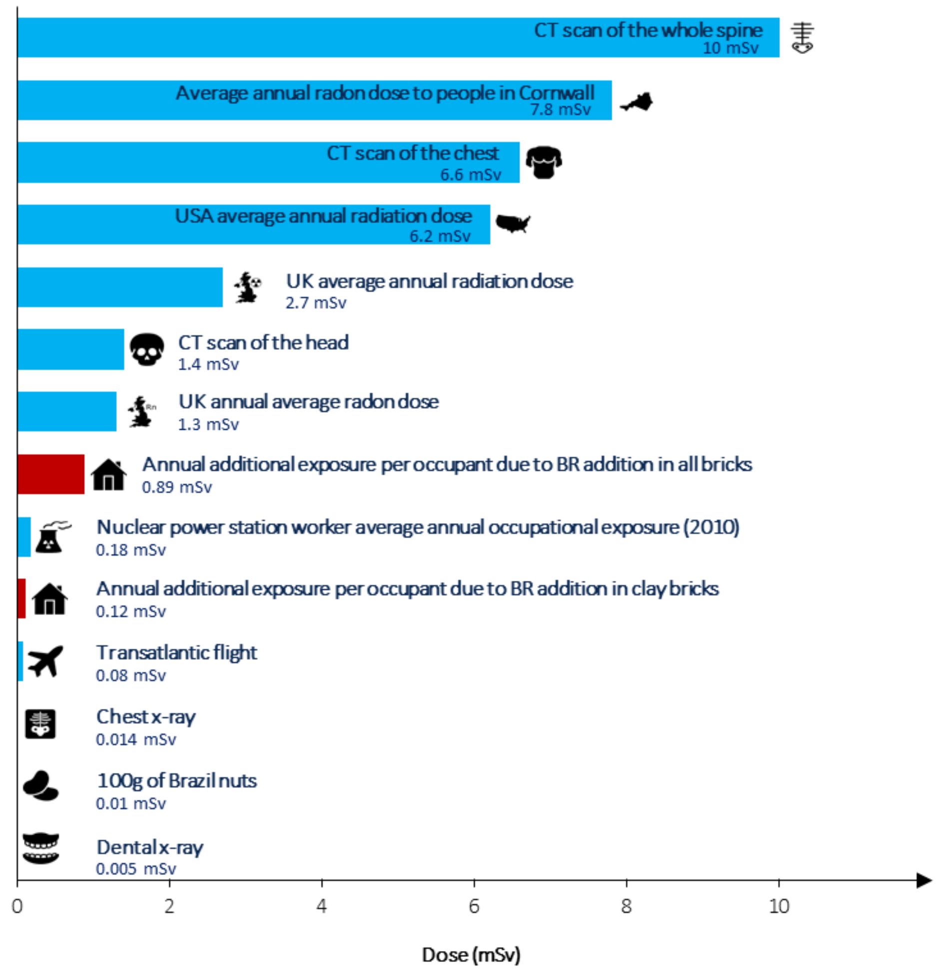

Fig. 9. Comparison of increased radiation doses as a result of various activities (Public Health England, 2011), including living in a house made of 30% BR augmented bricks (in place of just clay bricks and both clay and sand-lime bricks).

6. Discussion

The implementation of the Joyce et al. framework has yielded usable and transparent sets of characterisation factors for enhanced exposure to NORM via all major exposure routes and mechanisms for both humans and ecosystems. These factors are most robust for exposure of humans via the built environment. This was shown to be the most important exposure route for human health in the case of BR valorisation in building materials.

The models are useful but less robust from an ecosystems standpoint. For freshwater ecosystem the model displays sensitivity to uncertainties surrounding the potential redistribution of daughter nuclides. In addition, the effect of assumptions regarding the release of radionuclides from landfill has a pronounced effect on the conclusions that can be drawn with regard to the ecosystem impacts of NORM materials. The default scenarios presented in the case studies represent a worst case assumption with regards to bauxite residue disposal, and a best case assumption with regard to inert material landfills. The reality is likely to be somewhere in between. The default assumption of zero emissions of any kind from inert landfill taken in the Ecoinvent database has implications not only for NORM exposure, but for toxicity resulting from other heavy metal contaminants also, particularly in the case of long term emissions.

Although fate factors are provided for all environmental compartments in the USEtox model framework, effect factors for ecotoxicity are restricted to freshwater ecosystems. The authors of the USEtox methodology note that the long residence time of metals in the marine compartment results in unrealistically high characterisation factors (Rosenbaum et al., 2008). As a result they consider the deep sea to be a sink. In combination with the fact that few experimental data exist for marine organisms, this explains the omission of toxicity effects on marine organisms in USEtox.

The characterisation factors calculated for marine NORM exposure in this study are several orders of magnitude higher than those for freshwater exposure. One key difference however is that all of the emissions considered in the NORM models are persistent, thus a systematic bias related to this persistence should have the same effect across the board. $PAF \cdot m^3 \cdot d$ is a complex composite unit that does not truly represent a physical relationship in the real world. Rather if offers a common yardstick by which to make coherent internal comparisons. So long as the marine ecosystem results are not combined with other ecosystems or compared to other ecosystems when analysed, the fact that the numbers may not truly represent the physical reality of the system should not dissuade us from drawing, albeit uncertain, conclusions from the application of these characterisation factors.

Terrestrial ecotoxicity factors are not included in USEtox on the basis of the paucity of experimental results for terrestrial organisms. Here the restricted number of substances considered helps in this respect, allowing terrestrial ecosystems to be included.

The uncertainties and limitations of the ecosystem models are such that these characterisation factors should be considered indicative, and the results of these models interpreted accordingly.

From the case studies, two main conclusions can be drawn with regard to ecosystem impacts. Firstly, long term groundwater releases (should they exist) are a potentially significant source of ionising radiation impact for freshwater ecosystems. Thus if BR can be stored in such a way that NORM nuclides are effectively immobilised, either through the design of the BR disposal areas or through stabilisation of the BR prior to disposal this will reduce the negative environmental effects of BR. Secondly, power generation – both through the burning of coal and the mining of uranium – has the potential to have a radiological impact on marine and terrestrial organisms. Accordingly, measures such as the increased use of renewable energy can potentially synergistically reduce both climate change and ecoradiological impacts.

From a human health perspective, NORM exposure via use phase exposure to building materials was by far the most significant source of impact. NORM exposure via releases of nuclides to the environment on the other hand is negligible. Current results have been obtained assuming inert landfill of construction materials with no leaching. The sensitivity test of this assumption, considering the potential for the eventual release of 100% of nuclides from these building materials in inert landfill into the environment, still resulted in a use phase effect of NORM over 30 times higher than that from environmental releases.

It is interesting to note the level to which NORM contained in standard building materials contribute to NORM exposure in the scenarios presented here. The increased levels of NORM nuclides in BR are such that it is overrepresented in terms of use phase impact. BR represents 4% of the mass of building materials in Scenarios 2 and 3 and contributes 10% and 13% of the use phase impact respectively. However this also means in these scenarios 90% and 87% of the use phase NORM impact results from radionuclides contained within standard building materials. This suggests that NORM impacts should be given much more attention in environmental assessments of standard building materials. There are studies looking at differences in emissions of greenhouse gases and energy use for buildings using different structural alternatives (e.g. concrete and wood) (Brown, 2013). The results here suggest that it would be interesting to include NORM exposure in these assessments.

At an endpoint level, NORM exposure via the use phase is the overriding impact to human health in all scenarios. In comparison to the ionising radiation impact calculated by the Frischknecht and Braunschweig method, NORM exposure is far more significant, almost 6000 times higher in Scenarios 2 and 3. This suggests that the addition of the NORM exposure impact model is worthwhile in the assessment of human health impact.

For the utilisation of NORM materials in building materials, the predominance of use phase impacts raises the issue of burden shifting. Waste valorisation has two potential sources of environmental benefit – the avoidance of waste treatment processes, and the substitution of materials that would otherwise have had to have been produced. For some impact categories, the combined effect of these two sources of benefit often outweighs the impact associated with the valorisation process, leading to a net benefit. However, for NORM exposure in the utilisation of BR in building materials, the relatively low impacts associated with both the disposal of BR and the production of TSC materials in comparison with the use phase effects result in the opposite conclusion. This leads to potential trade-offs between impact categories and thus requires a more nuanced interpretation of LCA results of such valorisation systems.

The fact that NORM exposure is relatively well understood and increasingly well legislated for provides a useful contextualising factor in this interpretation. The increased radiation dose to humans as a result of BR inclusion in clay bricks, while still capable of causing health impacts, is comparable to lifestyle and/or stochastic factors.

The use phase radiation dose received from a given material is contingent on both its activity and its application, with use in dwellings providing the highest opportunity for exposure. Acknowledging and understanding the potential impacts associated with the use of NORM materials can allow the products of environmentally beneficial valorisation systems to be tailored to reduce the effects of NORM, through both composition and application. Since the impacts during the use phase are significant, it would be of interest to study BR valorisation where the use of the building materials is different. One example could be if the concrete including BR is used for construction of road infrastructure instead of buildings. This would change the use phase and thus the results.

7. Conclusions and outlook

We have developed a new impact method for the LCA methodology and demonstrated its applicability, providing case studies of different BR valorisation options. Our findings suggest that exposure from NORM materials should be considered during human health impact assessment.

The ecosystem model has more limited applicability, i.e. there is lack of knowledge regarding environmental fate of radionuclides that are produced as a part of decay chain, resulting for conservative results for environmental exposure due to freshwater releases. Additionally, the long term fate of the radionuclides at the landfill sites is not well known. However, we were able to demonstrate that the major exposure for humans comes from the use stage of NORM materials, and this stage can be modelled with sufficient level of accuracy.

The application of the impact assessment models to the valorisation of BR demonstrates the importance within LCA of having sufficient impact assessment models to capture all potential impacts, such that issues of burden shifting between impact measures can be captured, interpreted and resolved in the optimisation of product systems.

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