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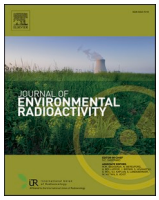

Threshold values for the protection of marine ecosystems from NORM in subsea oil and gas infrastructure

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ABSTRACT

This modelling study uses the ERICA Tool and Bateman's equation to derive sediment threshold values for radiation protection of the marine environment relevant to NORM-contaminated products (radium-contaminated scales, ^{210}Pb films and ^{210}Po films) found in subsea oil and gas infrastructure. Threshold values are calculated as the activity concentration of the NORM-contaminated products' head of chain radionuclide (i.e., ^{226}Ra + ^{228}Ra, ^{210}Pb, or ^{210}Po) that will increase radiation dose rates in sediments by 10 μGy/h to the most exposed organism at a given release time. The minimum threshold value (corresponding to peak radiation dose rates from the ingrowth of progeny) were for radium-contaminated scales, 0.009 Bq/g of ^{226}Ra, 0.029 Bq/g of ^{228}Ra (in the absence of ^{226}Ra) or 0.14 Bq/g of ^{228}Ra (in the presence of ^{226}Ra), followed by 0.015 Bq/g for ^{210}Pb films, and 1.6 Bq/g for ^{210}Po films. These may be used as default threshold values. Added activity concentrations of the NORM-contaminated products to marine sediments below these threshold values implies a low radiological risk to organisms while exceedances imply that further investigation is necessary. Using contaminated product specific parameterisations, such as K_d values derived for Ra from a $BaSO_4$ matrix in seawater, could greatly affect threshold values. Strong consideration should be given to deriving such data as part of specific radiological risk assessments for these products.

1. Introduction

Naturally occurring radionuclides are found at low activity concentrations in oil and gas reservoirs around the world (Smith, 2011). As fluids are extracted from reservoirs, radionuclides may become concentrated in contamination products including inorganic salt scales, films, sludges, and sands within infrastructure such as production pipelines (Nelson et al., 2016; Schmidt, 2000). These naturally occurring radioactive materials (NORM) may be recalcitrant and remain in oil and gas infrastructure at the cessation of operations.

A significant inventory of offshore oil and gas infrastructure is approaching the end of its productive life (Wood, 2017). Pipelines represent a large component of this infrastructure, with hundreds of thousands of kilometres of subsea pipelines laid around the world (Kaiser, 2018). Decommissioning options for this infrastructure include complete or partial removal or leaving it *in situ*. The reported benefits of leaving some infrastructure *in situ* include cost savings, improved health and safety outcomes for workers, and ecological benefits from the provision of productive artificial reef habitat (Bull and Love, 2019; McLean et al., 2022). However, questions remain about the long-term environmental consequence of contaminants, including NORM, in such infrastructure (MacIntosh et al., 2021; Melbourne-Thomas et al., 2021; Schläppy et al., 2021).

The disposal of residual NORM-contaminated products in oil and gas infrastructure to the marine environment via *in situ* abandonment is typically subject to regulatory oversight. For example, nations that are signatories to the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter (London Convention) (IMO, 1972)

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have a requirement to ensure that any infrastructure approved to be abandoned in the marine environment has "*de minimis*" radioactivity levels (IAEA, 2016). Understanding the potential dose rates from NORM-contaminated products to marine organisms is therefore important to help clarify regulatory requirements.

Three important NORM-contaminated products have been identified in oil and gas systems, including radium-contaminated mineral scales, ^{210}Pb films, and ^{210}Po films (Koppel et al., 2022). Radium-contaminated scales occur when radium is precipitated as an inorganic salt, typically co-precipitated with barium sulfate ($Ba(Ra)SO_4$) (Grandia et al., 2008). These are precipitated from water so may occur wherever water from production fluids is transported. Two radium isotopes, ^{226}Ra and ^{228}Ra, contribute to these scales. Both isotopes have a common chemical behaviour; however, they have different decay chains and progeny, emission types and energies, and half-lives, so their radioecological impacts may be different. ^{210}Pb films originate from either deposition from the decay of ^{222}Rn throughout treatment and transportation systems or electrodeposition of ^{210}Pb from fluids to surfaces such as wet parts of gas production systems (Schmidt, 2000; Smith, 2010). Unsupported ^{210}Po films are rarely reported but likely to originate where polonium partitions to lighter fractions such as ethane which are then separated and cooled in pipelines (Schmidt, 2000; Smith, 2010). More comprehensive reviews on the chemistry of NORM-contaminated product formation are provided by Nelson et al. (2015) and Schmidt (2000).

The risk of radionuclides to non-human biota is dependent on the amount of radiation that is absorbed by the organisms, termed the absorbed dose, and the radiosensitivity of the organism (ICRP, 2007). Radiation assessment tools have been developed to simplify calculations of absorbed dose rates to non-human biota and include the ERICA (Environmental Risk from Ionising Contaminants: Assessment and Management) Tool (Brown et al., 2008, 2016) and RESRAD (Residual Radioactive) Biota code (US DoE, 2004). Calculated absorbed dose rates can be compared to various environmental dose rate reference levels that describe levels above which potential radiation effects in organisms may occur. Common dose rate reference levels include 10 μGy/h for initial screening and ecosystem-wide protection (Garnier-Laplace et al., 2008), 400 μGy/h benchmark dose rate for marine organisms (UNSCEAR, 2008), or the derived consideration reference levels (DCRL) of the International Commission on Radiological Protection (ICRP) of 40400 μGy/h for reference flatfish and seaweed or 400–4000 μGy/h for reference crab (ICRP, 2008). For more information on these values and their application see the review by Real and Garnier-Laplace (2020).

The 10 μGy/h screening value was derived using the same approach used to derive environmental quality standards for aquatic ecosystems in jurisdictions including Australia and New Zealand (ANZG, 2018), the United States of America (United States Environment Protection Agency, 1985), and the European Union (Directorate-General for Health and Food Safety (European Commission), 2017). In short, dose rates that elicit a 10% effect to an organism's health, for a range of species, are aggregated into a species sensitivity distribution. The dose rate protecting 95% of all species is then taken and an additional safety factor of 2 applied to account for limitations in input data (Garnier-Laplace et al., 2010).

The ERICA Tool uses the 10 μGy/h reference level as its default screening value. At Tier 1, the ERICA Tool defines Environmental Media Concentration Limits (EMCL), which are the activity concentration of a radionuclide in an environmental media (i.e. soil, sediment, freshwater, or seawater) that will lead to a 95th percentile dose rate of 10 μGy/h to the most exposed organism (Brown et al., 2008). These EMCL values allow for a rapid screening of radionuclide risk with minimal data needs. Scenarios where the summed quotient of radionuclide activity concentrations in the relevant environmental medium by their respective EMCL values is less than one are considered of negligible environmental concern with a high degree of confidence, due to the conservatism of their input parameters and comparison to dose rates where effects are known to occur (Larsson, 2008).

NORM-contaminated products in oil and gas infrastructure are unique because they form in disequilibria with their progeny, become unsupported when the extraction of oil and gas products cease, and are isolated from marine receptors until the containing infrastructure corrodes (Nelson et al., 2015). This means that the ingrowth of the progeny of the products needs to be considered on timescales commensurate to the corrosion of the infrastructure in the marine environment, which may be hundreds to thousands of years (Melchers, 2021).

This study derives NORM-contaminated product-specific threshold values for radium (^{226}Ra + ^{228}Ra) contaminated scale, ^{210}Pb films, and ^{210}Po films. Threshold values can be used to inform initial screening assessments of radiological impacts to marine organisms in ecological risk assessments of subsea oil and gas infrastructure. These thresholds are calculated as the maximum initial activity concentration of the NORM-contaminated product's head of chain radionuclides that when released to sediments will not exceed the screening dose rate criterion of 10 μGy/h. The influence of the NORM release time to the marine environment (i.e., radionuclide ingrowth and decay), and in the case of radium-contaminated scales the contribution of both ^{226}Ra and ^{228}Ra is investigated to account for temporal aspects of contaminant risk. These values may be used as screening values to demonstrate that levels of radioactivity in infrastructure are *de minimis* and thus suitable for *in situ* decommissioning, or whether further assessments are required.

2. Methods

2.1. Calculation of threshold values for NORM-contaminated products in marine sediment

A modelling approach was applied to calculate the activity concentrations of NORM-contaminated products that when released into marine sediments would result in a dose rate of 10 μGy/h to the most exposed marine organism, accounting for radionuclide ingrowth and decay over time. These activity concentrations are termed threshold values and were determined by combining ERICA Tier 1 calculations (and their related simplifications in dosimetry) with the Bateman Equation.

All decay constants were taken from ICRP Publication 107 nuclear decay data (Eckerman and Endo, 2008). Default EMCL values and the associated underlying parameter values including partition coefficients (K_d), concentration ratios (CR), organism occupancy factors, dose conversion coefficients (DCCs), and radiation weighting factors implemented in ERICA Tool 2.0 were used (Brown et al., 2016; Copplestone et al., 2013; ERICA Consortium, 2021). The most exposed organisms for the radionuclides investigated in this study were Phytoplankton (^{226}Ra, ^{210}Pb, ^{228}Ra, and ^{228}Th), and Sea anemones & True corals (^{210}Po). The EMCL values for these radionuclides are given in Table 1.

Calculations were conducted in the open source statistical software R version 4.0.5 (R Core Team, 2016), using the Tidyverse extension packages (Wickham and RStudio, 2021). R scripts implementing Equations (3), (6) and (8) and the production of risk quotient figures are provided in Supplementary Information S1. A table of the half-lives and decay constants are given in Supplementary Information S2.

ERICA Tier 1 EMCLs are calculated by Equation (1) where SDR is the

Table 1
The most exposed organism and EMCL values for the head of chain radionuclides in NORM-contaminated products.

Radionuclide	EMCL (Bq/g dw)	Most exposed organism
^{226}Ra	0.020	Phytoplankton
^{210}Pb	0.015	Phytoplankton
^{210}Po	1.600	Sea anemones & True coral
^{228}Ra	0.094	Phytoplankton
^{228}Th	0.021	Phytoplankton

selected screening dose rate (here, 10 μGy/h) and F is the dose rate that an organism will receive for a unit activity concentration of a given radionuclide in an environmental medium (μGy/h per Bq/kg dry weight (dw) for sediment). F is calculated by an equation that accounts for the habitat of the organism, shown in Equation (2) for 'Phytoplankton' and 'Sea anemones & True coral' which are the most exposed organisms for the radionuclides investigated in this study (Table 1). Probability density functions based on lognormal distributions around K_d and CR values are used in Monte Carlo simulations to give the 95th percentile of the F value, which is then used in Equation (1).

$$EMCL = \frac{SDR}{F} \tag{1}$$

$$F_{Phytoplankton} = DCC_{int}\frac{CR}{K_d} + DCC_{ext}\frac{1}{K_d}$$

$$F_{Sea\ anemones\ \&\ True\ coral} = DCC_{int}\frac{CR}{K_d} + 0.5 \bullet DCC_{ext}\left(1 + \frac{1}{K_d}\right) \tag{2}$$

A risk quotient was calculated following the Tier 1 approach in the ERICA Tool using Equation (3), where RQ_t is the risk quotient for the NORM-contaminated product at time t, $A_{n,t}$ is the activity concentration of radionuclide n in the NORM-contaminated product at time t, and $EMCL_n$ is the EMCL for radionuclide n at a dose rate reference level of 10 μGy/h.

$$RQ_t = \sum_{1}^{n}\frac{A_{n,t}}{EMCL_n} \tag{3}$$

In this study the marine sediment EMCLs were used to reflect the solid-phase nature of NORM-contaminated products. When inputting radionuclide activity concentrations into the ERICA Tool, decay chains are truncated where progeny radionuclides' half-lives exceed 10 days based on the assumption that they are in equilibrium with their parent. This means that ERICA considers the decay for the head of chain radionuclides in NORM-contaminated products as shown in Fig. 1. While the input of short-lived progeny are simplified in this manner, the output of ERICA assessments includes their individual contribution to dose rates to organisms following the updated approach from ICRP Publication 136 (ICRP, 2017).

To calculate NORM-contaminated product threshold values for marine sediments, the radioactive decay and ingrowth equations were solved as follows.

The radioactive decay equation, Equation (4), gives the activity concentration of radionuclide n at time t:

$$A_{n,t} = A_{n,0}e^{-\lambda_n t} \tag{4}$$

where $A_{n,t}$ is the activity concentration of radionuclide n at time t, $A_{n,0}$ is the initial activity concentration of radionuclide n, and λ_n is the decay constant for radionuclide n. Substituting Equation (4) into Equation (3) for a risk quotient of 1 and rearranging for the initial activity concentration gives Equation (5). This is the solution for a one-component decay chain, i.e., ^{210}Po films.

$$A_{n,0} = 1 \Big/ \left(\frac{e^{-\lambda_n t}}{EMCL_n}\right) \tag{5}$$

The Bateman Equation can be used to calculate the activity concentration of any radionuclide in a decay chain at time t given the starting activity concentration of its head of chain radionuclide and their decay constants (Bateman, 1910). The general derivation shown in Equation (6) indicates that radionuclide *i* decays into *i*+1 at the rate of λ_i. J indicates the daughter radionuclide of i when n > 1. Equation (6) assumes that at time 0 only the head of chain radionuclides are present and that all decay chains are linear (i.e., the equation does not allow for branched decay).

$$A_{n,t} = A_{1,0} * \left(\prod_{i=1}^{n-1}\lambda_i\right) * \sum_{i=1}^{n}\frac{e^{-\lambda_j t}}{\prod_{j=1, j\neq i}^{n}(\lambda_j - \lambda_i)} \tag{6}$$

The solution of Equation (6) for the second radionuclide in a decay chain at time t is given in Equation (7).

$$A_{2,t} = A_{1,0}\left(\frac{\lambda_2}{\lambda_2 - \lambda_1}\right)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) \tag{7}$$

Substituting Equation (4) and Equation (7) into Equation (1) for a risk quotient of 1 and rearranging for the initial activity concentration of the head of chain radionuclide gives Equation (8), the function to calculate the threshold value a two-component decay chain, i.e. ^{210}Pb films.

$$A_{1,0} = 1 \Big/ \left(\frac{e^{-\lambda_1 t}}{EMCL_1} + \frac{\left(\frac{\lambda_2}{\lambda_2 - \lambda_1}\right)\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)}{EMCL_2}\right) \tag{8}$$

For radium-contaminated scale, the contribution of both ^{226}Ra and ^{228}Ra must be investigated. The ^{226}Ra chain is a three-component decay

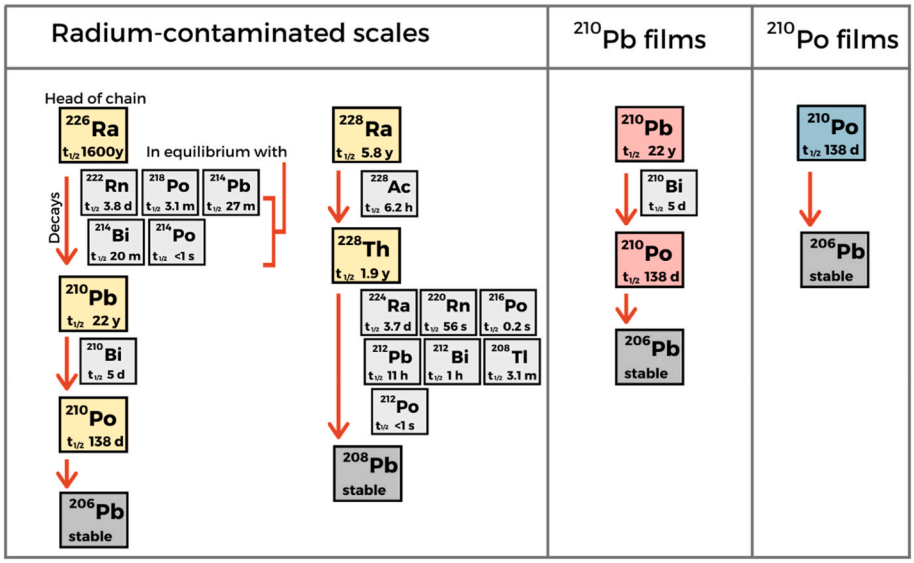

Fig. 1. Decay chains for NORM-contaminated products based on the ERICA Tool. The dose contribution of progeny radionuclides with a half-life of less than 10 days is incorporated into the DCC of its parent radionuclide based on an assumption of secular equilibrium. This is shown for each component of the decay chains for ^{226}Ra, ^{228}Ra, ^{210}Pb, and ^{210}Po. Half-lives are given in years (y), days (d), hours (h), minutes (m) and seconds (s). Note that ^{220}Rn and ^{222}Rn are not included in the ERICA assessment as their primary exposure route is by inhalation.

chain (^{226}Ra to ^{210}Pb to ^{210}Po, where short-lived progeny, i.e., with half-lives <10 d, are assumed to be in equilibrium with these chain segment parents, Fig. 1). The third component can again be calculated using the Bateman Equation and combined with expressions for the first and second component, in Equation (3). This is shown in Equation (9).

$$A_{1,0} = 1 \Big/ \left(\frac{e^{-\lambda_1 t}}{EMCL_1} + \frac{\left(\frac{\lambda_2}{\lambda_2-\lambda_1}\right)(e^{-\lambda_1 t} - e^{-\lambda_2 t})}{EMCL_2} + \frac{\lambda_2\lambda_3 \left(\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right)}{EMCL_3} \right) \quad (9)$$

The ^{228}Ra chain is a two-component decay chain and is solved using Equation (8). The risk quotient for the radium-contaminated scale can thus be expressed as shown in Equation (10).

$$RQ_t = A_{1,0} \left(\frac{e^{-\lambda_1 t}}{EMCL_1} + \frac{\left(\frac{\lambda_2}{\lambda_2-\lambda_1}\right)(e^{-\lambda_1 t} - e^{-\lambda_2 t})}{EMCL_2} + \frac{\lambda_2\lambda_3 \left(\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right)}{EMCL_3} + A_{2,0} \left(\frac{e^{-\lambda_4 t}}{EMCL_4} + \frac{\left(\frac{\lambda_5}{\lambda_5-\lambda_4}\right)(e^{-\lambda_4 t} - e^{-\lambda_5 t})}{EMCL_5} \right) \right) \quad (10)$$

Equation (10) cannot be solved to calculate the initial activity concentration of ^{226}Ra and ^{228}Ra that would not exceed a risk quotient of 1 (as was done in Equation (5) and Equation (8)) because they can occur in different proportions in scale and have different decay rates. Therefore, threshold values were first calculated independently. ^{226}Ra and ^{228}Ra will ingrow progeny and reach a peak in radioactivity at approximately 122 years and 4 years, respectively. Therefore, the maximum starting activity concentration of ^{228}Ra was calculated to give a risk quotient equal to 1 minus the risk quotient of ^{226}Ra at year 4 starting at its minimum head of chain activity. As ^{228}Ra decays faster, its contribution to the risk quotient of the radium scale will be insignificant at 122 years (corresponding to peak ingrowth of ^{226}Ra), meaning its contribution could be ignored. For a given time, the bracketed component of Equation (10) can be solved to give a single value for ^{226}Ra and ^{228}Ra components of the equation. This is shown for t = 4 years in Equation (11).

The solution of Equation 10 for t = 4, $A_{1,0}$ = ^{226}Ra, and $A_{2,0}$ = ^{228}Ra

$$RQ_{t=4} = {}^{226}Ra_{t=0} * 57.3 + {}^{228}Ra_{t=0} * 34.4$$

For a $RQ_{t=4}$ of 1, rearranged for $^{228}Ra_{t=0}$ this becomes

$$^{228}Ra_{t=0} = 0.029 - 1.7 * {}^{226}Ra_{t=0} \quad (11)$$

The accuracy of these equations was confirmed using online calculators and the ERICA Tool (v2.0). The solutions of Equations (3), (6) and (7) for a given time were inputted to the World Information Service on Energy Uranium Project universal decay calculator (https://www.wise-uranium.org/rcc.html) to determine the activities of radionuclides in their decay chains at time t. These values were inputted to the ERICA Tool for a Tier 1 assessment and shown to give a risk quotient of 1.

2.2. Sensitivity analysis

The contribution of each radionuclide to the total dose rate to the two most exposed organisms for the NORM-contaminated products' head of chain radionuclides (Table 1), Phytoplankton and Sea Anemone & True corals, was investigated using a Tier 2 assessment in the ERICA Tool. Note that the output of ERICA assessments includes the contribution of all radionuclides, including those with half-lives <10 days. A sensitivity analysis was conducted using the Tier 3 functions of the ERICA Tool to investigate which parameter estimates contribute the greatest uncertainty to the total dose rate to each organism. To investigate how the provision of site-specific data affects threshold values, K_d values from seawater leach experiments with radium-contaminated scale collected from a subsea oil and gas pipeline measured by Cresswell et al. (2021) and reported in Macintosh et al. (2022) were used. The K_d values for radium, 1.5×10^6 L/kg, and polonium, 1.5×10^6 L/kg, were used to derive new EMCL values for radium-contaminated scales following the probabilistic Tier 1 approach specified in ERICA (Equation (1) and Equation (2)). The scale-specific EMCL values were inputted into Equation (10) and Equation (11) to calculate product-specific threshold values.

The K_d value for polonium is based on measured solid and aqueous activity concentrations whereas the limit of detection for the analytical approach was used for the radium aqueous concentrations. This means that the radium K_d value is a conservative estimate (i.e., will likely overestimate aqueous radium concentrations). Despite this, it is still 1000 times greater than the default value in ERICA of 2×10^3 L/kg. K_d values were also reported for lead and thorium; however, all values were below the limit of detection which did not have the sensitivity to detect the expected aqueous concentrations based on their default K_d values. For that reason, they were not used.

2.3. NORM-contaminated products and assessment assumptions

The three NORM-contaminated products assessed here are based on the types of NORM contamination reported in by-products in oil and gas systems (Ali et al., 2019; Koppel et al., 2022; Schmidt, 2000). In the assessment, the following assumptions are made about the products:

1. Contamination products are comprised of only their head of chain when oil and gas extraction has stopped. This assumption is used as a simplification as contamination products form to varying extents throughout the operational life of the oil and gas system (Yang et al., 2020).
2. There is no loss of radionuclide progeny. This is a conservative assumption that is likely to be true for closed pipe decommissioning scenarios following the cessation of operations, but unlikely to be true for contaminated material in the marine environment where soluble components may emanate or leach.

3. The speciation of NORM-contaminated products does not change between operations ceasing and its release to the marine environment, such as following corrosive breakthrough of pipelines. Decommissioned pipelines that are filled with seawater and capped may lead to reducing conditions that could promote speciation changes of NORM-contaminated products, such as reductive dissolution of sulfate minerals. However, this is not well understood, and the speciation of the contaminants will change to reflect its local receiving environment.
4. The values derived are for sediment concentrations and do not incorporate any mixing of the NORM-contaminated product into the sediment. Application of these values to real world scenarios should account for a conservative dilution of contaminated material in the environment. Dispersion modelling or other approaches should be considered to justify a selected mixing scenario.
5. The ERICA Tool includes default model parameters for radionuclide partitioning between sediments and seawater (K_d values) and from waters to biota (CR values). There is no CR value for ^{226}Ra, ^{210}Pb, or ^{210}Po to Sea anemones and True coral, so values from a taxonomically similar organism (polychaete worm) were used in line with the recommendations of Hosseini et al. (2008). Additionally, the default K_d value for polonium is based on a 'periodically adjacent element' (IAEA, 2004a). More information about the limitations of default ERICA parameter values for this exposure scenario can be found in recent publications from Koppel et al. (2022) and Macintosh et al. (2022).

3. Results and discussion

3.1. Threshold values for NORM-contaminated products in marine sediments

Threshold values are defined as the initial head of chain radionuclide activity concentration in the NORM-contaminated product that will result in a 10 μGy/h dose rate to the most exposed organism in the marine environment at a given time of exposure (i.e., at a given age of the NORM-contaminated product when released into the marine environment). As radionuclides ingrow and decay over time in NORM-contaminated products, their corresponding dose rate to organisms will increase and decrease, as visualised in Fig. 2 for ^{226}Ra and ^{228}Ra. Therefore, it is important to consider radiation risks over time, particularly where the release of radionuclides will occur well after any decision about their disposal.

The results presented here provide for the ability to calculate appropriate threshold values for a given release time. Alternatively, a set of minimum threshold values are provided that are calculated from the highest radiation dose rate that NORM contamination will deliver to a marine organism at any time, reflecting the balance between the increase in dose rate from radionuclide in-growth and the decrease in dose rate from the decay of the parent radionuclide (i.e., the peak of the sum curve in Fig. 2).

Table 2
Threshold values (TV) for the head of chain radionuclides of different NORM-contaminated products in sediments that would result in the most exposed marine organism receiving a dose rate of 10 μGy/h. All values reflect initial activity concentrations, and do not consider mixing and dispersion processes.

NORM-contaminated product	Minimum threshold value (Bq/g)	NORM-contaminated product age at minimum threshold value (years)	Threshold value for 50 years (Bq/g) release time	Threshold value for 100 years (Bq/g) release time
^{226}Ra in scale	0.00898	121.6	0.0098	0.0090
^{228}Ra in scale (no ^{226}Ra)	0.029	3.99	4.98	>1000
^{228}Ra in scale (with ^{226}Ra)	0.014	3.99	0.014	0.0141[a]
^{210}Pb film	0.015	0	0.069	0.330
^{210}Po film	1.6	0	>1000	>1000

[a] Note that the contribution of ^{228}Ra is minimal at this stage due to its relatively short half-life of 5.8 years.

3.1.1. Radium-contaminated scale

The minimum threshold value for ^{226}Ra was 0.009 Bq/g and for ^{228}Ra was 0.029 Bq/g, when considered independently (Table 2). A key difference in the temporal extent of the risk of ^{226}Ra and ^{228}Ra relates to their half-lives. The ingrowth and decay of the ^{228}Ra series is controlled by the 5.8-year half-life of ^{228}Ra, which is much shorter than the 1600-year half-life of ^{226}Ra. This means that the ^{228}Ra series peaks and decays before the ^{210}Pb component of the ^{226}Ra series comes to equilibrium. As a result, ^{226}Ra reaches the peak of its ingrowth at 122 years compared to 4 years for ^{228}Ra (Fig. 2).

It is likely that ^{226}Ra and ^{228}Ra will co-occur in a radium-contaminated scale, given their identical chemical behaviour (Nelson et al., 2015). As ^{226}Ra has a longer half-life and radioactive peak from the ingrowth of ^{210}Pb (Fig. 2) the joint risk quotient was determined to ensure that ^{228}Ra when in the presence of ^{226}Ra will not exceed a risk quotient of 1. ^{226}Ra with an activity concentration of 0.009 Bq/g will contribute a risk quotient of 0.52 after 4 years – corresponding to peak radioactivity from ^{228}Ra ingrowth. Therefore, the maximum initial

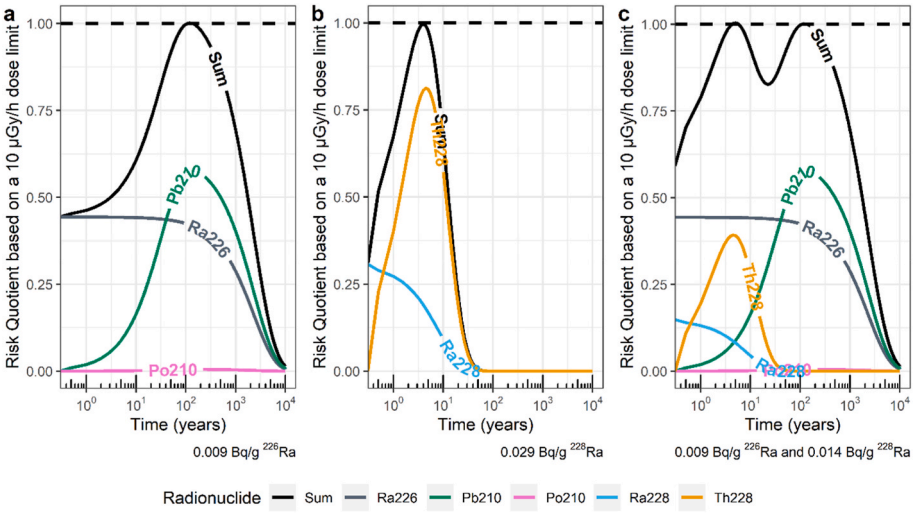

Fig. 2. The changing risk profile of (a) ^{226}Ra, (b) ^{228}Ra, and (c) ^{226}Ra + ^{228}Ra following their ingrowth and decay over time. The contribution of long-lived progeny ($t_{1/2}$ >10 d) are shown while short-lived progeny ($t_{1/2}$ <10 d) are accounted for by their first parent with a $t_{1/2}$ >10 d as per Brown et al. (2008). The starting activity concentration is 0.009 Bq/g for ^{226}Ra and either 0.029 Bq/g for ^{228}Ra in the absence of ^{226}Ra or 0.014 Bq/g for ^{228}Ra in the presence of ^{226}Ra. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

^{228}Ra activity concentration that would give a risk quotient of 0.48 (calculated as 1 minus 0.52, the contribution of ^{226}Ra) was calculated to be 0.014 Bq/g. The impact of ^{228}Ra on the radioactivity peak from ^{226}Ra ingrowth is not necessary because of the relatively short half-life of ^{228}Ra. I.e., a starting ^{228}Ra activity concentration of 0.014 Bq/g would contribute a risk quotient <0.001 after 122 years. For release scenarios where ^{226}Ra activity concentrations will be less than 0.009 Bq/g, a greater amount of ^{228}Ra may be permissible than 0.014 Bq/g. More details on these results can be found in the code provided in Supplementary Information S1.

^{228}Ra and its progeny are decayed >95% after 25 years meaning that on the time scale of pipeline corrosion rates (10s to >1000 of years (Melchers, 2021)) it is unlikely to be present in significant quantities to meaningfully contribute much radiation dose rate if it is isolated from environmental receptors in infrastructure (Fig. 2). In comparison, it will take approximately 8000 years for ^{226}Ra to decay >95% of its initial activity meaning that the risk to the environment will be relevant on timescales relevant to pipeline corrosion. The consequence of these differences depends on the different ^{226}Ra to ^{228}Ra ratio and age of the NORM-contaminated product when released to the marine environment. At proportions of ^{226}Ra <0.4, or release times <20 years the ^{228}Ra ingrowth peak will result in a higher sum of risk quotient peak than the ^{226}Ra ingrowth peak (see Fig. 2). This will reduce the 'acceptable' ^{226}Ra activity concentration to <0.009 Bq/g, and result in the ^{226}Ra ingrowth peak being well below a risk quotient of 1. These temporal risk considerations will have to be considered if NORM-contaminated products are to be released at or shortly after operations cease, such as if they are stored open to the marine environment (e.g., uncapped or cut pipelines).

3.1.2. ^{210}Pb and ^{210}Po films

The minimum threshold value for ^{210}Pb films was 0.015 Bq/g at 0 years due to the low EMCL of ^{210}Pb (0.015 Bq/g) and high EMCL of its progeny ^{210}Po (1.6 Bq/g) (Table 2). That is, the ingrowth of ^{210}Po was less impactful to the risk quotient compared to the decay of ^{210}Pb. This means that the risk from ^{210}Pb films decreases at a rate proportional to the half-life of ^{210}Pb (22.2 years).

^{210}Po films have the highest EMCL of the radionuclides investigated in this study, a short half-life ($t_{1/2}$ = 138 d) relative to ^{210}Pb, ^{228}Ra, or ^{226}Ra, and no radioactive progeny. The minimum threshold value was equal to its EMCL at 1.6 Bq/g which increases at a rate proportional to the half-life of ^{210}Po. This means that unsupported ^{210}Po is unlikely to be a radionuclide of concern in decommissioning contexts where operations have ceased for more than a few years, as shown in Fig. 3. This does not diminish the need to consider the risk of ^{210}Po to human health and safety where public or occupational exposures are possible.

3.1.3. Sensitivity of radioecology parameters

The dose rate to the organisms Phytoplankton and Sea anemones & True corals were explored by a Tier 2 assessment. All NORM-contaminated products were investigated, and input activity concentrations were equal to those present in the NORM-contaminated products at the age that results in the minimum threshold value as defined in Table 2. Dose rates by NORM-contaminated product, organism, and radionuclide are given in Supplementary Information S3. In short, external dose rates are a negligible contribution to the total. For phytoplankton, ^{228}Ac contributes the greatest dose in scale containing ^{226}Ra and ^{228}Ra (~75% of the total dose rate) followed by ^{228}Th and ^{226}Ra (~5% each). In the absence of ^{228}Ra, ^{210}Pb and ^{226}Ra contribute

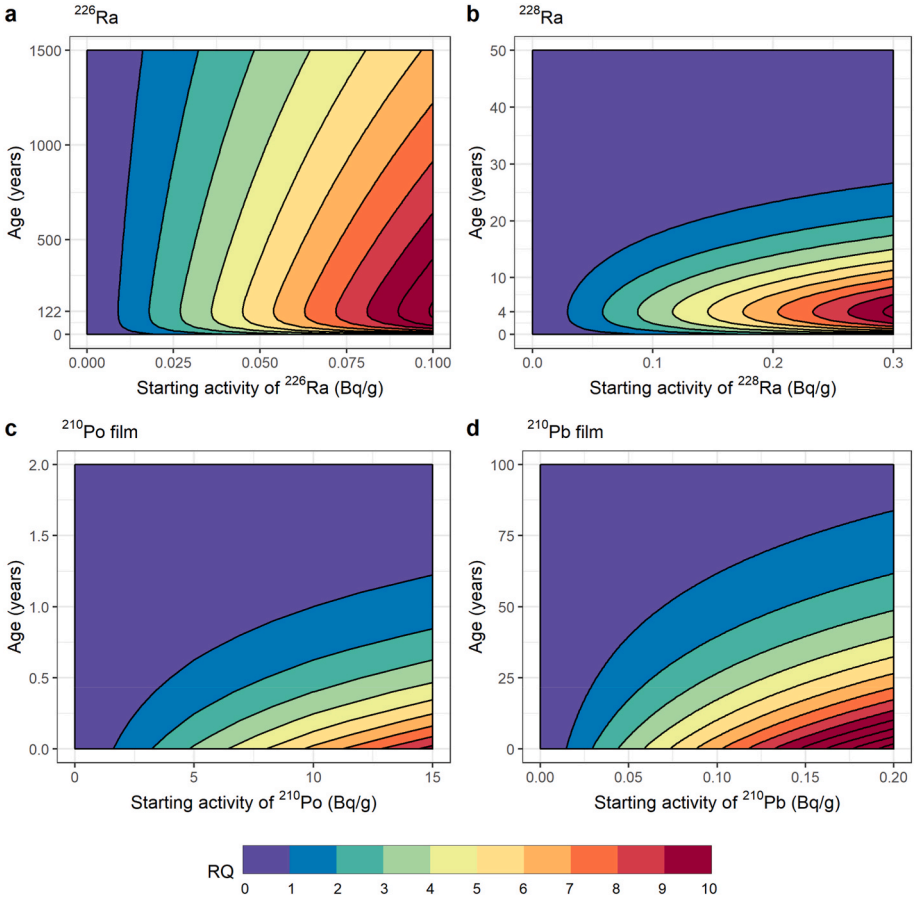

Fig. 3. Temporal patterns of risk for (a) ^{226}Ra, (b) ^{228}Ra, (c) ^{210}Pb, and (d) ^{210}Po at different initial activity concentrations and times. Risk quotients were determined as the sum of the quotient of each radionuclide in the products' decay chains at a given time by their EMCL for a 10 μGy/h dose rate. Default parameters were used to calculate EMCLs in the ERICA Tool (v2.0). Note the different ranges for the age of the NORM contaminant (y axis).

approximately equivalent dose rates to phytoplankton.

A sensitivity analysis was undertaken using the Tier 3 function of the ERICA Tool. Input radionuclides were the ^{226}Ra decay series (^{226}Ra, ^{210}Pb, ^{210}Po) or both the ^{226}Ra and the ^{228}Ra series (^{228}Ra and ^{228}Th) and both Phytoplankton and Sea anemones & True Coral were investigated. The parameter and radionuclide pairs with the greatest Pearson correlation coefficients (positive or negative) are tabulated in Supplementary Information S4. Generally, CR values contribute the greatest variability with correlations between the parameter value and total dose rate to organism greatest for phytoplankton. For example, for phytoplankton exposed to scale with ^{226}Ra and ^{228}Ra, the Th CR was the most impactful parameter with a positive coefficient of 0.33 followed by the K_d for Th with a negative coefficient of −0.11. For exposures of ^{226}Ra in scale to phytoplankton, the CR of Pb was the most impactful parameter with a coefficient of 0.45 followed by K_d for Pb at −0.07. The relatively low sensitivity of K_d values relative to CR values likely reflects the approach adopted to create a distribution around the K_d estimate – where the 5th and 95th percentile values of the distribution are set as 10x lower and higher than the point estimate, respectively (Brown et al., 2016; IAEA, 2004a). This is a much narrower range of potential values than exists for CR values. It is unlikely that this range fully accounts for the possible partitioning of radionuclides from NORM-contaminated products. For example, mineral scales and their associated radionuclides may become more soluble in anoxic conditions (Phillips et al., 2001).

To understand how using NORM-contaminated product specific K_d values will affect EMCLs and TVs, radium-contaminated scale specific Ra and Po K_d values were used. The scale-specific K_d values were derived from a 30-day seawater leaching experiment of scale retrieved from a subsea oil and gas pipeline. These K_d values were used to derive new EMCL values using the same probabilistic Tier 1 approach in ERICA (all distribution parameters used to recalculate EMCL values are given in Supplementary Information S5). The use of scale-specific K_d values greatly increased the EMCL values for ^{226}Ra and ^{228}Ra, increasing from 0.02 Bq/g to 16.3 Bq/g for ^{226}Ra and increasing from 0.094 Bq/g to 70.2 Bq/g for ^{228}Ra. The EMCL for ^{210}Po decreased from 1.6 Bq/g to 0.105 Bq/g (Table 3). The distribution of F values is given in Fig. 4.

The minimum TVs recalculated using the scale-specific K_d values for Ra and Po did not have the same substantial changes as the recalculated EMCL values (Table 4). For example, the minimum TV for ^{226}Ra increased from 0.009 Bq/g to 0.015 Bq/g whereas the EMCL increased from 0.02 to 16.3 Bq/g (Table 4). This is likely because ^{210}Pb and ^{228}Ac contribute the largest doses from ^{226}Ra and ^{228}Ra decay chains, respectively, meaning that the ingrowth of progeny may be more important to the dose rate than the radium isotopes themselves (Supplementary Information S3). K_d values from the MacIntosh et al. (2022) study could not be calculated for all radionuclides detected in radium-contaminated scale due to the detection limits of the analytical approach (Cresswell et al., 2021). More detail is provided in Supplementary Information S5. However, the change in EMCL values for radium isotopes and ^{210}Po demonstrate the benefit of using parameterisations relevant to the exposure scenario.

Unexplored in this study are other contaminant and site-specific considerations that may increase or decrease NORM bioavailability and mobility. Default K_d and CR parameter values assume radionuclide activities in the environment at an equilibrium between sediments, waters, and organisms. This assumption may not be true for point sources of contamination with different chemistries to environmental matrices such as radionuclides originating from NORM-contaminated products (Periáñez et al., 2018). This is clearly the case for radium, which is known to be highly insoluble in oxic seawater due to the low solubility product of $RaSO_4$ ($\log_{10}K_{sp}$ of −10.24) (Brown et al., 2022). This gives greater context to the high K_d value calculated from seawater leach experiments of radium in radium-contaminated barite scale, 1.5×10^6 L/kg, which is ~800 times greater than the default Ra K_d value of 2×10^3 (IAEA, 2004a).

Radioecology and radioecotoxicology data are limited for marine organisms compared to terrestrial and freshwater organisms. Seawater leaching tests to calculate K_d values are inexpensive and so should be actively considered by those managing infrastructure containing NORM-contaminated products.

3.2. Comparison to published radiation criteria and background activities

A comparison of the threshold values derived here to activity concentrations in NORM-contaminated products, background sediments (Koppel et al., 2022), and other criteria for radiation protection (Real and Garnier-Laplace, 2020), suggests that these derived threshold values are conservative. However, these values: are applied in addition to background radionuclide activity concentrations; account for the ingrowth of progeny over the radiological life of the NORM-contaminated product; do not incorporate dispersion or dilution of the NORM-contaminated products in the marine environment; and are proposed to be used as part of a graded approach to radiological protection.

The derived threshold values for radium-contaminated scales and ^{210}Pb films are much lower than the commonly applied exemption criterion for NORM of 1 Bq/g (IAEA, 2014a). Various national jurisdictions apply the 1 Bq/g criterion to NORM-contaminated products from oil and gas extraction to identify material subject to regulatory control (Loy, 2015).This study suggests that the 1 Bq/g criterion may not be suitably protective of the marine ecosystem for this exposure scenario. For example, the peak risk quotient for NORM-contaminated product with a starting activity concentration of 1 Bq/g in sediments will be 111 for ^{226}Ra at 122 years or 68 for ^{210}Pb at 0 years. For ^{226}Ra, risk quotients will persist above a value of 1 for 100–1000s of years (Fig. 3).

Threshold values that reflect release scenarios commensurate with corrosion times for pipelines would be much higher, particularly for ^{210}Po and ^{228}Ra decay series, which will have undergone significant decay. For example, for a release time 100 years after contaminated-product formation and oil and gas operations ceasing there may be no need to consider ^{210}Po films or ^{228}Ra in scale as many half-lives of decay will substantially reduce their activity concentration (Table 2). Where the corrosion of pipelines is expected to take >200 years, there may be no need to consider the risk of ^{210}Pb films. However, this should be considered carefully against expected corrosion timeframes.

3.3. Environmental management of NORM-contaminated products in subsea oil and gas infrastructure

The approach adopted in this study solves a challenge faced by the oil and gas industry and their regulators around the need to understand the radiological risk of NORM-contaminated products to the marine environment over their radiological life. Coupling risk and ingrowth models for exposure scenarios where the radioactive source is not being regenerated, and is at disequilibria from its progeny, has been adopted in

Table 3
Default EMCLs, recalculated EMCLs using default parameters, and recalculated EMCLs for ^{226}Ra, ^{210}Po, and ^{228}Ra using Ra and Po K_d values from radium-contaminated scale leach experiments. Lognormal distribution parameters are given in Supplementary Information S5. Scale-specific EMCLs were not calculated for ^{210}Pb and ^{228}Th because appropriate updated K_d values were not available.

Radionuclide	ERICA default EMCLs (Bq/g)	Recalculated default EMCLs (Bq/g)	EMCLs calculated using scale-specific K_d values (Bq/g)
^{226}Ra	0.020	0.020	16.3
^{210}Pb	0.015	0.014	–
^{210}Po	1.60	1.42	0.105
^{228}Ra	0.094	0.091	75.2
^{228}Th	0.021	0.023	–

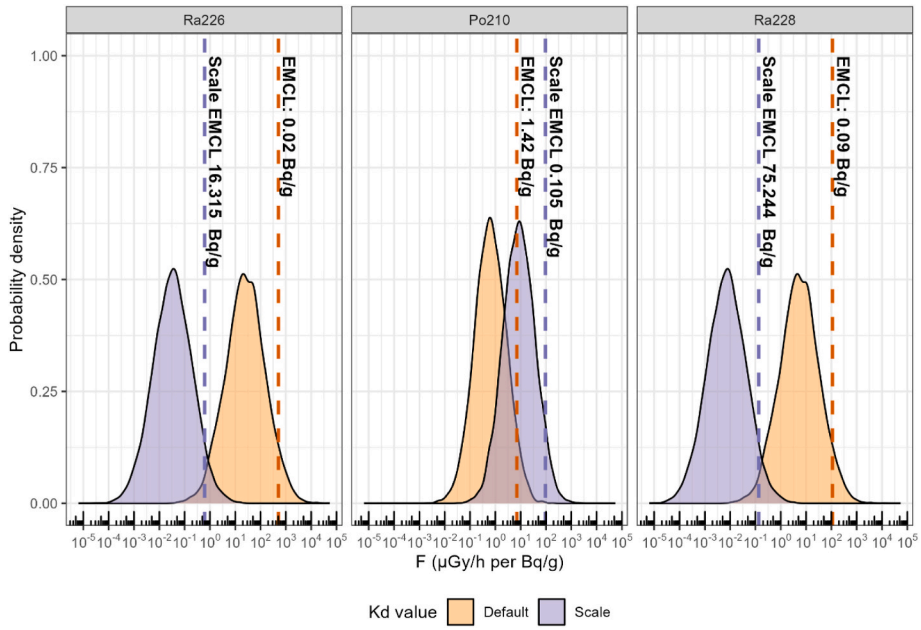

Fig. 4. Probability density distributions of F values calculated using default parameters and radium-contaminated scale specific K_d values for Ra and Po taken from MacIntosh et al. (2022). Dashed lines indicate the 95th percentile of the lognormally distributed F values calculated based on Monte Carlo simulations of the K_d and CR values lognormal distributions. The resulting EMCL from this distribution is given in text adjacent to the lines (calculated by Equation (1)).

Table 4
Minimum threshold values re-calculated using scale-specific K_d values for Ra and Po. Age at minimum threshold value (corresponding to the peak radiological dose to the most exposed organism balancing radionuclide ingrowth and decay) is given in brackets.

NORM-contaminated product	Default minimum threshold value (Bq/g)	Updated minimum threshold value (Bq/g)
^{226}Ra in scale	0.009 (122 years)	0.014 (139 years)
^{228}Ra in scale (no ^{226}Ra)	0.029 (4 years)	0.036 (4.5 years)
^{228}Ra in scale (with ^{226}Ra)	0.14 (4 and 122 years)	0.031 (4.5 and 139 years)
^{210}Pb Films	0.015 (0 years)	0.014 (1.1 years)
^{210}Po films	1.6 (0 years)	0.105 (0 years)

other jurisdictions. For example, with the US EPA's peak preliminary remediation goal model for Superfund sites (Galloway et al., 2020).

3.3.1. *Applying the threshold values*

The threshold values derived here represent the head of chain activity concentration for NORM radionuclides in marine sediments that will prevent exceedance of the screening dose rate of 10 μGy/h and thus pose a negligible radiological risk to the marine ecosystem. These values represent activity concentrations that may be added to the natural background activity concentration of marine sediments, rather than limits on the activity concentrations of the NORM-contaminated products themselves. As such, threshold values could be applied to predicted sediment activity concentrations following the products' dispersion and mixing in the environment.

Exceedances of threshold values do not indicate that there will be radiological effects to organisms. Rather, exceedances should warrant a more detailed investigation. This may include incorporating site-specific data that can be used to better quantify radionuclide partitioning and bioavailability for a specific environment. This could then be assessed against other environmental dose rate reference levels such as the ICRP DCRL bands of 40–400 μGy/h for flatfish and seaweed (ICRP, 2008) or the UNSCEAR marine benchmark dose rate of 400 μGy/h (UNSCEAR, 2008).

For the context of offshore decommissioning, the use of the lower level of a relevant DCRL band (e.g. 40 μGy/h for flatfish and seaweed) may be particularly appropriate given the definition of *de minimis* radiation levels from IAEA (2016) for the London Convention. *De minimis* subsumes the IAEA criteria of exemption and exclusion (IAEA, 2004b). As it relates to NORM contamination in subsea oil and gas infrastructure, a release of NORM-contaminated products that would substantially increase radioactivity at the site requires a specific assessment for marine flora and fauna protection. In the specific assessment, DCRL are used as the radiological criteria for marine biota. Threshold values derived here can be recalculated for a DCRL of 40 μGy/h by multiplying by 4.

3.3.2. *The need for a holistic approach to radiological assessments*

Only exposure scenarios where NORM is released to the marine environment were considered here. NORM-contaminated products may also pose a risk to sessile organisms by external-only radiation exposures. For example, the radionuclides ^{226}Ra, ^{214}Bi, ^{214}Pb of the ^{226}Ra decay chain and ^{228}Ac, ^{212}Bi, ^{212}Pb, ^{224}Ra, and ^{208}Tl from the ^{228}Ra decay chain have gamma emissions with emission probabilities >1% and energies >100 keV (Supplementary Information S6), which may result in a radiological dose rate to organisms colonising the external surfaces of contaminated pipelines (MacIntosh et al., 2022).

The derived threshold values represent a single line of evidence, radiological contamination of sediments, based on an impact of dose rates to marine organisms. A holistic understanding of all impacts and risks from a decommissioning scenario should be considered in an ecological risk assessment (Chapman et al., 2002). This should include the risk of other contaminants, such as mercury (Kho et al., 2022), plastics (Testoff et al., 2022), steel corrosion by-products, and mixtures therein (Koppel et al., 2018). Nonetheless, the derived threshold values provide a quick assessment method to determine whether NORM-contaminated products from oil and gas infrastructure pose a negligible radiological risk for a given release scenario. Importantly, this assessment aligns with IAEA and ICRP recommendations for radiological protection of the environment in a planned exposure scenario (IAEA, 2014b; ICRP, 2014). This approach may also be useful in determining the suitability of disposing NORM-contaminated infrastructure at sea under the '*de minimis*' standard of the London Convention and Protocol.

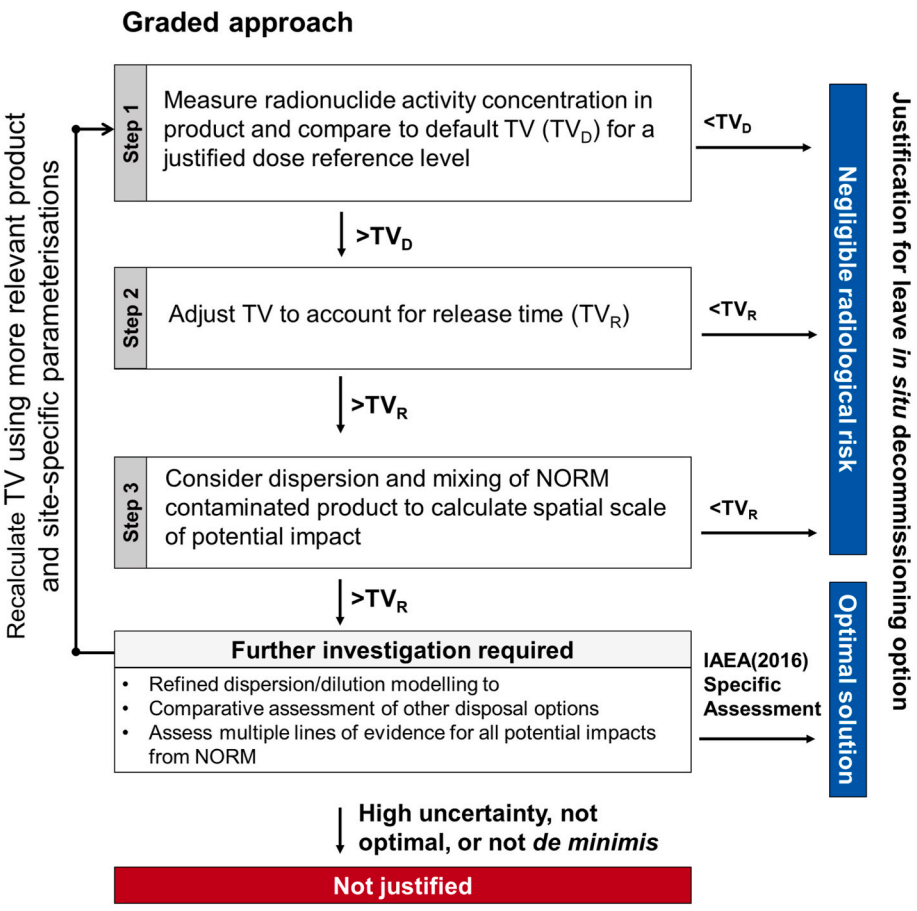

Fig. 5. A proposed implementation of a graded approach to assess NORM risk in the marine environment using threshold values derived in this study. Risk refers to the likelihood of dose rate exceedances of a dose screening level to marine organisms. NORM may have other impacts to marine ecosystems that should be considered holistically in an ecological risk assessment. IAEA (2016) Specific Assessment refers to the assessment provided by IAEA-TECDOC-1759 Determining the Suitability of Materials for Disposal at Sea under the London Convention 1972 and London Protocol 1996: A Radiological Assessment Procedure.

3.4. Case study

To illustrate how these threshold values can be applied, two fictional case studies are considered here: (1) an export gas pipeline containing films of ^{210}Pb at 10 Bq/g; and (2) a flexible flowline containing radium-contaminated scale in a barite matrix at an activity concentration of 5 Bq/g^{226}Ra and 5 Bq/g^{228}Ra. In these hypothetical examples, an oil and gas operator is investigating the possibility of decommissioning their pipeline containing residual NORM contamination by leaving them *in situ*. The proposed assessment approach is visualised in Fig. 5.

3.4.1. Lead film in gas export pipeline

Gas export pipelines may carry traces of ^{222}Rn which decay into the longer lived ^{210}Pb ($t_{1/2} = 22$ years) leading to the formation of thin lead films on the internal surface of pipes. For this hypothetical case study, a 10 Bq/g^{210}Pb film has been measured in a pipeline that will experience corrosive breakthrough (the point where pipeline corrosion leads to the ingress of seawater) after 200 years.

Step 1: The TV$_d$ for ^{210}Pb films is 0.015 Bq/g (Table 2). As the activity concentration of ^{210}Pb in the film is greater than the TV$_d$ the assessment moves to step 2.
Step 2: Gas-export pipelines are often made of carbon steel. Typical corrosion rates mean that corrosive breakthrough may occur after approximately 200 years. This is the point where corrosion penetrates the pipe wall allowing seawater/sediment to contact the scale contamination. For a real scenario, an understanding of the pipeline material and its corrosion rates in its environment would be necessary.

The TV$_R$ for ^{210}Pb with a release time of 200 years is 7.5 Bq/g (i.e., the solution for Equation (8) at t = 200 years). The ^{210}Pb film activity concentration is above the TV$_R$ the assessment moves to step 3.

Step 3: A 10 Bq/g film requires a dilution factor of 1.3 to reduce to 7.5 Bq/g. A highly conservative mixing factor could be 1:1 pipeline to sediment, with the lead film being a small component of overall pipeline mass. This suggests that the resulting release to the marine environment is unlikely to lead to dose rates >10 μGy/h. A persuasive argument could thus be made that the radiological component of the ecological risk assessment for the release of NORM from this pipeline is low.

3.4.2. Radium-contaminated scale in production flowlines

Flexible production flowlines can connect subsea wells to floating production storage and offloading vessels. They carry raw well fluids and so may accumulate NORM residues over their production life.

Step 1: The TV$_d$ for ^{226}Ra is 0.009 Bq/g and ^{228}Ra is 0.014 Bq/g (Table 2). As the activity concentration of the radium scale is greater than the TV$_d$ the assessment moves to step 2.
Step 2: Flexible flowlines often contain coatings and corrosion resistant steels. So corrosive breakthrough is expected after approximately 1000 years.

The release-time adjusted screening value, TV$_R$, is 0.013 Bq/g for ^{226}Ra and >1000 Bq/g for ^{228}Ra due to its 5.8-year half-life (i.e. the solutions to Equation (9) and Equation (8), respectively). This means that the time-adjusted screening value TV$_R$ is now 0.013 Bq/g for ^{226}Ra and ^{228}Ra no longer needs to be considered.

As the activity concentration of ^{226}Ra in the radium scale is greater than the TV$_R$ the assessment moves to step 3.

Step 3: In Step 3, the mixing and dispersion of the scale is considered. This requires some understanding of the local environment. For an activity concentration of 5 Bq/g of ^{226}Ra in NORM-contaminated scale, a dilution factor of 152 is required to dilute the activity concentration to 0.033 Bq/g. Assuming a 10 mm thick scale with a density of 4.5 g/cm^3 in a 6-inch pipeline with a 10 mm thick wall, this would mean diluting 283 g scale in 43 kg for every meter of pipeline. A number of dispersion models exist that could be used to investigate the spatial extent of possible impact (see Periáñez et al. (2015) for a comparison of model performance for radionuclide release from Fukushima Daichi nuclear power plant).

Additional investigation is required to demonstrate the acceptability of this release scenario. The use of by-product-specific K_d values from leach studies (Fig. 4), marine organism specific CR values from bioaccumulation studies, or more refined dispersion or mixing models should be considered.

4. Conclusions

The preservation of marine sediment quality is an important management objective for oil and gas operators decommissioning offshore infrastructure. This study contextualises the 10 μGy/h dose rate screening criterion into threshold values for the types of NORM-contaminated products commonly reported in offshore oil and gas systems. Minimum threshold values provide an activity concentration for a NORM-contaminated product that when released to the sediment at any age will not exceed 10 μGy/h to the most exposed organisms. Minimum threshold values derived were 0.009 Bq/g for ^{226}Ra in scales; 0.029 Bq/g for ^{228}Ra in the absence of ^{226}Ra or 0.014 Bq/g for ^{228}Ra in the presence of ^{226}Ra; 0.015 Bq/g for ^{210}Pb films, and; 1.6 Bq/g for ^{210}Po films. Threshold values may also be calculated for any given NORM-contaminated product age. This may assist risk assessments if the time between the formation of the NORM-contaminated product and its release environment following the corrosion of containing infrastructure can be estimated.

The greatest contribution of dose rates to the most exposed organisms from NORM-contaminated product exposures are modelled to be from the internal dose from the radionuclides ^{228}Ac,^{228}Th, ^{226}Ra, and ^{210}Pb. CR values were the most sensitive parameters according to the ERICA Tier 3 assessment. However, the true variability of K_d values for NORM-contaminated products may not be accounted for in the default parameter ranges. Using K_d values for radium and polonium derived from seawater leach experiments with radium-contaminated barite increased the EMCLs of ^{226}Ra from 0.02 to 16.3 Bq/g, ^{228}Ra from 0.094 to 75.2 Bq/g and decreased the EMCL for ^{210}Po from 1.6 to 0.105 Bq/g. Recalculated threshold values for NORM contaminated products based on the updated EMCLs did not change as much as the EMCL values themselves, particularly for radium-contaminated scales. This is because the progeny of ^{226}Ra and ^{228}Ra contribute the greatest proportion of dose. This demonstrates the need to derive specific parameters for all radionuclides of NORM-contaminated products to improve the environmental relevance of radiological assessments.

The derived values suggest that contamination with ^{226}Ra and ^{210}Pb may pose a long-term risk on timescales commensurate with the corrosion of infrastructure. In contrast, ^{228}Ra and ^{210}Po (if unsupported by ^{210}Pb) will decay on shorter time scales of <50 years and may not pose long-term risks. Depending on the dilution and dispersion of NORM-contaminated products, the IAEA exemption criteria of 1 Bq/g may not be protective of marine environments for these types of NORM-contaminated products and may not satisfy *de minimis* criteria under the London Convention and Protocol.

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Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

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References

Ali, M.M.M., Zhao, H., Li, Z., Maglas, N.N.M., 2019. Concentrations of TENORMs in the petroleum industry and their environmental and health effects. RSC Adv. 9, 39201–39229. https://doi.org/10.1039/C9RA06086C.

ANZG, 2018. Australian and New Zealand guidelines for fresh and marine water quality, Australian and New Zealand governments and Australian state and territory governments. Canberra, ACT, Australia. Available at. www.waterquality.gov.au/anz-guidelines.

Bateman, H., 1910. Solution of a system of differential equations occurring in the theory of radioactive transformations. Proc. Camb. Phil. Soc. 15, 423–427.

Brown, J.E., Alfonso, B., Avila, R., Beresford, N.A., Copplestone, D., Hosseini, A., 2016. A new version of the ERICA tool to facilitate impact assessments of radioactivity on wild plants and animals. J. Environ. Radioact. 153, 141–148. https://doi.org/10.1016/j.jenvrad.2015.12.011.

Brown, J.E., Alfonso, B., Avila, R., Beresford, N.A., Copplestone, D., Pröhl, G., Ulanovsky, A., 2008. The ERICA tool. J. Environ. Radioact., The ERICA Project 99, 1371–1383. https://doi.org/10.1016/j.jenvrad.2008.01.008.

Brown, P.L., Matyskin, A.V., Ekberg, C., 2022. The aqueous chemistry of radium. Radiochim. Acta 110, 505–513. https://doi.org/10.1515/ract-2021-1141.

Bull, A.S., Love, M.S., 2019. Worldwide oil and gas platform decommissioning: a review of practices and reefing options. Ocean Coast Manag. 168, 274–306. https://doi.org/10.1016/j.ocecoaman.2018.10.024.

Chapman, P.M., McDonald, B.G., Lawrence, G.S., 2002. Weight-of-Evidence issues and frameworks for sediment quality (and other) assessments. Hum. Ecol. Risk Assess. 8, 1489–1515. https://doi.org/10.1080/20028091057457.

Copplestone, D., Beresford, N.A., Brown, J.E., Yankovich, T., 2013. An international database of radionuclide concentration ratios for wildlife: development and uses. J. Environ. Radioact. 126, 288–298. https://doi.org/10.1016/j.jenvrad.2013.05.007.

Cresswell, T., Brown, S., Wong, H., Apte, S., Cresswell, T., Brown, S., Wong, H., Apte, S., 2021. Assessing the impacts of scale residues from offshore oil and gas

decommissioning on marine organisms. APPEA J 61, 379–383. https://doi.org/10.1071/AJ20024.
Directorate-General for Health and Food Safety (European Commission), 2017. Technical Guidance for Deriving Environmental Quality Standards. Publications Office of the European Union, LU.
Eckerman, K., Endo, A., 2008. ICRP Publication 107. Nuclear decay data for dosimetric calculations. Ann. ICRP 38, 7–96. https://doi.org/10.1016/j.icrp.2008.10.004.
ERICA Consortium, 2021. ERICA Assessment Tool: Version 2.0 Overview of Changes. No. Version 1).
Galloway, L.D., Bellamy, M.B., Dolislager, F.G., Ringer, H.J., Asano, E.A., Stewart, D.J., Noto, K.A., Manning, K.L., Gross, L.J., Armstrong, A.Q., 2020. BATEMAN EQUATION ADAPTATION FOR SOLVING AND INTEGRATING PEAK ACTIVITY INTO EPA ELCR AND DOSE MODELS (No. ORNL/TM-2020/1780). Oak Ridge National Laboratories, Oak Ridge, TN.
Garnier-Laplace, J., Copplestone, D., Gilbin, R., Alonzo, F., Ciffroy, P., Gilek, M., Agüero, A., Björk, M., Oughton, D.H., Jaworska, A., Larsson, C.M., Hingston, J.L., 2008. Issues and practices in the use of effects data from FREDERICA in the ERICA Integrated Approach. J. Environ. Radioact. 99, 1474–1483. https://doi.org/10.1016/j.jenvrad.2008.04.012.
Garnier-Laplace, J., Della-Vedova, C., Andersson, P., Copplestone, D., Cailes, C., Beresford, N.A., Howard, B.J., Howe, P., Whitehouse, P., 2010. A multi-criteria weight of evidence approach for deriving ecological benchmarks for radioactive substances. J. Radiol. Prot. 30, 215–233. https://doi.org/10.1088/0952-4746/30/2/S02.
Grandia, F., Merino, J., Bruno, J., 2008. Assessment of the Radium-Barium Co-precipitation and its Potential Influence on the Solubility of Ra in the Near-Field. Swedish Nuclear Fuel and Waste Management Co.
Hosseini, A., Thørring, H., Brown, J.E., Saxén, R., Ilus, E., 2008. Transfer of radionuclides in aquatic ecosystems – default concentration ratios for aquatic biota in the Erica Tool. J. Environ. Radioact. 99, 1408–1429. https://doi.org/10.1016/j.jenvrad.2008.01.012.
IAEA, 2016. Determining the Suitability of Materials for Disposal at Sea under the London Convention 1972 and London Protocol 1996: A Radiological Assessment Procedure (TECDOC No. 1759). International Atomic Energy Agency, Vienna, Austria.
IAEA, 2014a. Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards. International Atomic Energy Agency, Vienna.
IAEA, 2014b. Modelling of Biota Dose Effects: Report of Working Group 6 Biota Dose Effects Modelling of EMRAS II Topical Heading Reference Approaches for Biota Dose Assessment, Environmental Modelling for Radiation Safety (EMRAS II) Programme. International Atomic Energy Agency, Vienna.
IAEA, 2004a. In: Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. Technical reports series/International Atomic Energy Agency. Internat. Atomic Energy Agency, Vienna.
IAEA, 2004b. Application of the Concepts of Exclusion, Exemption and Clearance Safety Guide, Safety Standards Series. International Atomic Energy Agency, Vienna.
ICRP, 2017. Dose Coefficients for Non-human Biota Environmentally Exposed to Radiation, 136. ICRP Publ, p. 46, 9781526439086.
ICRP, 2014. Annals of the ICRP 43. Protection of the Environment under Different Exposure Situations, 124. ICRP Publ, 9781446296141.
ICRP, 2008. Annals of the ICRP 38. Environmental Protection - the Concept and Use of Reference Animals and Plants, 108. ICRP Publ, 978-0-444-52934-3.
ICRP, 2007. The 2007 Recommendations of the International Commission on Radiological Protection. ICRP publication. International Commission on Radiological Protection, Oxford.
IMO, 1972. Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter. International Maritime Organisation.
Kaiser, M.J., 2018. The global offshore pipeline construction service market 2017 – Part I. Ships Offshore Struct. 13, 65–95. https://doi.org/10.1080/17445302.2017.1342923.
Kho, F., Koppel, D.J., von Hellfeld, R., Hastings, A., Gissi, F., Cresswell, T., Higgins, S., 2022. Current understanding of the ecological risk of mercury from subsea oil and gas infrastructure to marine ecosystems. J. Hazard Mater. 438, 129348 https://doi.org/10.1016/j.jhazmat.2022.129348.
Koppel, D.J., Adams, M.S., King, C.K., Jolley, D.F., 2018. Chronic toxicity of an environmentally relevant and equitoxic ratio of five metals to two Antarctic marine microalgae shows complex mixture interactivity. Environ. Pollut. 242, 1319–1330. https://doi.org/10.1016/j.envpol.2018.07.110.
Koppel, D.J., Kho, F., Hastings, A., Crouch, D., MacIntosh, A., Cresswell, T., Higgins, S., 2022. Current understanding and research needs for ecological risk assessments of naturally occurring radioactive materials (NORM) in subsea oil and gas pipelines. J. Environ. Radioact. 241, 106774 https://doi.org/10.1016/j.jenvrad.2021.106774.
Larsson, C.-M., 2008. An overview of the ERICA Integrated Approach to the assessment and management of environmental risks from ionising contaminants. J. Environ. Radioact., The ERICA Project 99, 1364–1370. https://doi.org/10.1016/j.jenvrad.2007.11.019.
Loy, J., 2015. What should a radiation regulator do about naturally occurring radioactive material? Ann. ICRP 44, 197–201. https://doi.org/10.1177/0146645315572298.
MacIntosh, A., Dafforn, K., Penrose, B., Chariton, A., Cresswell, T., 2021. Ecotoxicological effects of decommissioning offshore petroleum infrastructure: a systematic review. Crit. Rev. Environ. Sci. Technol. 1–39. https://doi.org/10.1080/10643389.2021.1917949.
MacIntosh, A., Koppel, D.J., Johansen, M.P., Beresford, N.A., Copplestone, D., Penrose, B., Cresswell, T., 2022. Radiological risk assessment to marine biota from exposure to NORM from a decommissioned offshore oil and gas pipeline. J. Environ. Radioact. 251–252, 106979 https://doi.org/10.1016/j.jenvrad.2022.106979.
McLean, D.L., Ferreira, L.C., Benthuysen, J.A., Miller, K.J., Schläppy, M.-L., Ajemian, M. J., Berry, O., Birchenough, S.N.R., Bond, T., Boschetti, F., Bull, A.S., Claisse, J.T., Condie, S.A., Consoli, P., Coolen, J.W.P., Elliott, M., Fortune, I.S., Fowler, A.M., Gillanders, B.M., Harrison, H.B., Hart, K.M., Henry, L.-A., Hewitt, C.L., Hicks, N., Hock, K., Hyder, K., Love, M., Macreadie, P.I., Miller, R.J., Montevecchi, W.A., Nishimoto, M.M., Page, H.M., Paterson, D.M., Pattiaratchi, C.B., Pecl, G.T., Porter, J. S., Reeves, D.B., Riginos, C., Rouse, S., Russell, D.J.F., Sherman, C.D.H., Teilmann, J., Todd, V.L.G., Treml, E.A., Williamson, D.H., Thums, M., 2022. Influence of offshore oil and gas structures on seascape ecological connectivity. Glob. Change Biol. n/a. https://doi.org/10.1111/gcb.16134.
Melbourne-Thomas, J., Hayes, K.R., Hobday, A.J., Little, L.R., Strzelecki, J., Thomson, D. P., van Putten, I., Hook, S.E., 2021. Decommissioning research needs for offshore oil and gas infrastructure in Australia. Front. Mar. Sci. 8.
Melchers, R.E., 2021. Corrosion of steels and irons immersed in natural seawater for up to 600 Y. Corrosion 78, 87–95. https://doi.org/10.5006/3967.
Nelson, A.W., Eitrheim, E.S., Knight, A.W., May, D., Mehrhoff, M.A., Shannon, R., Litman, R., Burnett, W.C., Forbes, T.Z., Schultz, M.K., 2015. Understanding the radioactive ingrowth and decay of naturally occurring radioactive materials in the environment: an analysis of produced fluids from the Marcellus Shale. Environ. Health Perspect. 123, 689–696. https://doi.org/10.1289/ehp.1408855.
Nelson, A.W., Johns, A.J., Eitrheim, E.S., Knight, A.W., Basile, M., Bettis III, E.A., Schultz, MichaelK., Forbes, T.Z., 2016. Partitioning of naturally-occurring radionuclides (NORM) in Marcellus Shale produced fluids influenced by chemical matrix. Environ. Sci. Process. Impacts 18, 456–463. https://doi.org/10.1039/C5EM00540J.
Periáñez, R., Brovchenko, I., Duffa, C., Jung, K.-T., Kobayashi, T., Lamego, F., Maderich, V., Min, B.-I., Nies, H., Osvath, I., Psaltaki, M., Suh, K.-S., 2015. A new comparison of marine dispersion model performances for Fukushima Dai-ichi releases in the frame of IAEA MODARIA program. J. Environ. Radioact. 150, 247–269. https://doi.org/10.1016/j.jenvrad.2015.09.003.
Periáñez, R., Brovchenko, I., Jung, K.T., Kim, K.O., Maderich, V., 2018. The marine kd and water/sediment interaction problem. J. Environ. Radioact. 192, 635–647. https://doi.org/10.1016/j.jenvrad.2018.02.014.
Phillips, E.J.P., Landa, E.R., Kraemer, T., Zielinski, R., 2001. Sulfate-reducing bacteria release barium and radium from naturally occurring radioactive material in oil-field barite. Geomicrobiol. J. 18, 167–182. https://doi.org/10.1080/01490450120549.
R Core Team, 2016. R: A Language and Environment for Statistical Computing.
Real, A., Garnier-Laplace, J., 2020. The importance of deriving adequate wildlife benchmark values to optimize radiological protection in various environmental exposure situations. J. Environ. Radioact. 211, 105902 https://doi.org/10.1016/j.jenvrad.2019.01.014.
Schläppy, M.-L., Robinson, L.M., Camilieri-Asch, V., Miller, K., 2021. Trash or treasure? Considerations for future ecological research to inform oil and gas decommissioning. Front. Mar. Sci. 8.
Schmidt, A.P., 2000. Naturally occurring radioactive materials in the gas and oil industry: origin, transport and deposition of stable lead and 210Pb from Dutch gas reservoirs = Natuurlijk voorkomende radioactieve materialen in de gas- en olie-industrie. Geologica Ultraiectina. Univ, Utrecht.
Smith, A.L., 2011. First correlation of NORM with a specific geological hypothesis. In: Presented at the SPE European Health, Safety and Environmental Conference in Oil and Gas Exploration and Production, SPE-138136-MS. https://doi.org/10.2118/138136-ms.
Smith, A.L., 2010. NORM: the lessons to be learned, new challenges and innovative thinking with decommissioning and radioactive waste. In: Presented at the SPE International Conference on Health, Safety and Environment in Oil and Gas Exploration and Production. https://doi.org/10.2118/125661-MS. Rio de Janeiro, Brazil.
Testoff, A.N., Nelson, N.A., Nicolette, J.P., 2022. A quantitative method for evaluating ecological risks associated with long-term degradation of deep-sea plastic-containing infrastructure. APPEA J 62, 141–158. https://doi.org/10.1071/AJ21113.
United States Environment Protection Agency, 1985. Guidelines for Deriving Numerical National Water Quality Criteria for the Protection of Aquatic Organisms and Their Uses. No. PB85-227049).
UNSCEAR, 2008. Sources and Effects of Ionizing Radiation: UNSCEAR 2008 Report to the General Assembly. United Nations, New York.
US DoE, 2004. RESRAD-BIOTA: a Tool for Implementing a Graded Approach to Biota Dose Evaluation. Washington, D. C., USA.
Wickham, H., RStudio, 2021. Tidyverse: Easily Install and Load the "Tidyverse.
Wood, Mackenzie, 2017. US$32 Billion of Decommissioning Worldwide over the Next Five Years: Is the Industry Ready? (Insight).
Yang, Y., Luo, X., Hong, C., Yadav, A., Rogowska, M., Ambat, R., 2020. Characterization, formation and development of scales on L80 steel tube resulting from seawater injection treatment. J. Pet. Sci. Eng. 193, 107433 https://doi.org/10.1016/j.petrol.2020.107433.