# Radiological risk assessment to marine biota from exposure to NORM from a decommissioned offshore oil and gas pipeline

Amy MacIntosh [a,b,*], Darren J. Koppel [c,1], Mathew P. Johansen [a], Nicholas A. Beresford [d], David Copplestone [e], Beth Penrose [f], Tom Cresswell [a]

[a] Australian Nuclear Science and Technology Organisation, Lucas Heights, NSW, Australia
[b] School of Natural Sciences, Macquarie University, Sydney, NSW, Australia
[c] Curtin University Oil and Gas Innovation Centre, Faculty of Science and Engineering, Curtin University, Perth, WA, Australia
[d] UK Centre for Ecology & Hydrology, Lancaster Environment Centre, Lancaster, UK
[e] Faculty of Natural Sciences, University of Stirling, Stirling, United Kingdom
[f] Tasmanian Institute of Agriculture, University of Tasmania, Tasmania, Australia

## ABSTRACT

Scale residues can accumulate on the interior surfaces of subsea petroleum pipes and may incorporate naturally occurring radioactive materials (NORM). The persistent nature of 'NORM scale' may result in a radiological dose to the organisms living on or near intact pipelines. Following a scenario of *in-situ* decommissioning of a subsea pipeline, marine organisms occupying the exteriors or interiors of petroleum structures may have close contact with the scale or other NORM-associated contaminated substances and suffer subsequent radiological effects. This case study used radiological dose modelling software, including the ERICA Tool (v2.0), MicroShield® Pro and mathematical equations, to estimate the likely radiological doses and risks of effects from NORM-contaminated scale to marine biota from a decommissioned offshore oil and gas pipeline. Using activity concentrations of NORM ($^{226}$Ra, $^{210}$Po, $^{210}$Pb, $^{228}$Ra, $^{228}$Th) from a subsea pipeline from Australia, environmental realistic exposure scenarios including radiological exposures from both an intact pipe (external only; accounting for radiation shielding by a cylindrical carbon steel pipe) and a decommissioned pipeline with corrosive breakthrough (resulting in both internal and external radiological exposure) were simulated to estimate doses to model marine organisms. Predicted dose rates for both the external only exposure (ranging from 26 μGy/h to 33 μGy/h) and a corroded pipeline (ranging from 300 μGy/h to 16,000 μGy/h) exceeded screening levels for radiological doses to environmental receptors. The study highlighted the importance of using scale-specific solubility data (i.e., $K_d$ values) for individual NORM radionuclides for ERICA assessments. This study provides an approach for conducting marine organism dose assessments for NORM-contaminated subsea pipelines and highlights scientific gaps required to undertake risk assessments necessary to inform infrastructure decommissioning planning.

## 1. Introduction

From 2018 to 2040, thousands of offshore petroleum fields are nearing or have reached the end of their productive life, resulting in a substantial amount of infrastructure requiring to be decommissioned (Cresswell et al., 2021; MacIntosh et al., 2021; Koppel et al., 2022). Decommissioning options for all infrastructure include complete or partial removal from the seabed or leaving *in situ* (leave in place), with no intervention or with options for burial (Bull and Love 2019). The

decision-making processes for decommissioning in most countries with offshore petroleum infrastructure is on a case-by-case basis and requires environmental impact assessments (EIA). This is to ensure that operators and licence holders demonstrate that minimal harm to the marine ecosystem (and human consumers) occurs during decommissioning and potentially after if infrastructure is left *in situ*. The prospect of extensive decommissioning has, only recently, given rise to the need for a better understanding of the associated environmental risks, benefits to biodiversity, and improvement of the assessment tools needed to address

them (Cresswell et al., 2021; MacIntosh et al., 2021; Melbourne-Thomas et al., 2021; Koppel et al., 2022). Research into the environmental impacts of abandoned structures, particularly of associated contaminants has only emerged in the last five years (MacIntosh et al., 2021). Of the range of contaminants, very little is known about the long-term fate and impacts of naturally occurring radioactive material (NORM).

Oil and gas extraction and transport can lead to the deposition of solid deposits of inorganic salts known as scale on the interior surfaces of petroleum infrastructure such as pipes, vessels and platforms. NORM radionuclides can become incorporated into these scales, which make them an important consideration for decommissioning. Scales can have the highest radioactivity of any NORM-contaminated product due to high activities of radium (Ra) radioisotopes ($^{226}$Ra and $^{228}$Ra can range from 40 to 1000 Bq/g (Ali et al., 2019; Koppel et al., 2022).

The natural decay of parent radionuclides, the uranium series ($^{238}$U), thorium series ($^{238}$Th) and actinium series ($^{235}$U), can lead to elevated levels of Ra in formation waters, but it is the chemical similarities of Ra and scale-forming minerals such as barium (Ba), strontium (Sr) and calcium (Ca) that leads to its elevated activity of progeny radionuclides within the various forms of scale (e.g., barium sulphate, calcium carbonate). The deposition of scale containing the NORM radionuclides, $^{226}$Ra, $^{228}$Ra, $^{210}$Pb and $^{210}$Po, in addition to co-occurring metals, are known to be present for years beyond cessation of operations in petroleum infrastructure and could be at concentrations that lead to potential ecological harm if released to the marine environment (Grung et al., 2009; Olsvik et al., 2010; Jensen et al., 2016; MacIntosh et al., 2021).

A key challenge for petroleum operators is to predict potential impacts of NORM contaminants in production infrastructure to inform decommissioning plans. Several modelling tools and software programs have been created to assess the risk to non-human biota from radiation and radioactive contamination, including the ERICA Tool (Environmental Risk of Ionising Contaminants: Risk and Management; hereafter referred to as ERICA) and RESRAD- BIOTA (RESidual RADioactive materials) (DOE 2019; IAEA 2021). ERICA is one of the most widely used software packages globally that allows for simplified estimations of dose rates to biota in terrestrial, freshwater, and marine ecosystems (Brown et al., 2016). ERICA has been used for environmental risk assessments for different sources of potential radiological concerns, including aquatic ecosystems close to uranium industry sites (Goulet et al., 2022), nuclear power plant discharges (Nedveckaite et al., 2011; Vandenhove et al., 2013; Li et al., 2015), waste disposal facilities (Robinson et al., 2010; Johansen et al., 2012; Posiva Oy 2014) and releases from medical facilities (Carolan et al., 2011).

Decommissioning scenarios for NORM-contaminated infrastructure that include *in situ* disposal may eventually lead to the NORM constituents being released following infrastructure corrosion. This means that NORM could be directly available to marine organisms in the surrounding benthic environment where bioaccumulation and subsequent ecotoxicological effects from the chemical and radiological properties of the scale material could occur. Prior to the corrosion of infrastructure, marine organisms inhabiting the exteriors of subsea production infrastructure may receive doses from NORM contained inside the pipes due to gamma emissions of some radionuclides penetrating through the pipeline material. This may lead to ecotoxicological effects for organisms (Hosseini et al., 2012; MacIntosh et al., 2021), especially of sessile organisms colonising the pipeline. However, these exposures are not well understood.

Two key challenges for the prediction of NORM impacts from decommissioning decisions include the inability of ERICA to calculate shielded dose rates from radionuclides contained within infrastructure and data limitations for some radionuclides resulting in some default parameters being derived using extrapolation approaches (Brown et al., 2016). The ERICA Tool includes a standard set of exposure geometries that are generally applicable to most aquatic exposure scenarios (e.g. organisms surrounded by water only, organisms at sediment surface). However, the geometries do not include important features required when modelling decommissioning structures. Specifically needed are exposures that include shielding in cases where there is a steel structure between the source and the organism and cylindrical sources (e.g., accumulated scale within a pipe during normal pipeline operations; MacIntosh et al., 2021).

The ERICA tool provides conservative default parameters to ensure that assessments are protective to a wide range of environments and organisms. Parameters, such as those describing the solid to liquid phase partitioning of radionuclides (the $k_d$ value), are based on assumptions of the radionuclide being in equilibrium in the environment. This may not be the case when NORM-contamination associated with infrastructure is exposed to marine waters or sediments. Cresswell et al. (2021) recommended using results from NORM scale leachate experiments (in seawater) to derive pipe scale-specific $k_d$ values for use in ERICA modelling. This approach should ensure more environmentally relevant assessments are conducted and reduce the level of uncertainty and conservatism in dose assessments. However, this has not yet been assessed in a comparative modelling study.

The first aim of this paper was to investigate the likely radiological dose rates from NORM-contaminated scale under environmental realistic exposure scenarios to model marine organisms. This was achieved by using available radiological dose modelling software, including the ERICA Tool (v2.0), MicroShield® Pro and mathematical equations, to estimate the radiological doses and subsequent risks from NORM-contaminated scale to marine biota from a decommissioned offshore oil and gas pipeline. Radiological exposures from both external sources (accounting for radiation shielding by a cylindrical carbon steel pipe) and internal sources over two scenarios of *in situ* pipeline decommissioning are provided using measured concentrations of NORMs from subsea pipelines. The secondary aim of this study was to investigate the impact of different model parameterisations on the overall assessment outcomes of using the ERICA Tool and to determine the best approach to apply to subsea pipeline NORM scale assessments.

## 2. Methodology

This case study presents two radiological exposure scenarios of marine biota exposed to radiological doses from NORM-contaminated subsea scale i) external only to sessile organisms colonising the exteriors of the pipe and ii) organisms seeking refuge inside a recently opened pipe (via corrosive breakthrough) where organisms are exposed via non-shielded external radiation and via internal radiation following respiration and/or ingestion of soluble radionuclides.

### 2.1. Radionuclides considered in the modelling: NORM scale

For all the radiological dose modelling assessments, we used measured concentrations of NORMs associated with barium sulphate ($BaSO_4$; hereby referred to as barite) scale. The scale collected was from a decommissioned subsea well tubular (construction details and dimensions in section 2.2) carrying predominantly oil with minor fractions of gas and formation water from the Griffin Field, located on the North West Shelf of Australia, approximately 68 km offshore of Onslow, Western Australia. The tubular was brought to shore, where solid scale was collected from the internal surfaces, homogenised (pulverised to $P_{80}$ 231 μm) and was analysed by alpha and gamma spectrometry at the Australian Science and Technology Organisation (ANSTO), Lucas Heights (Sydney, Australia) to determine the presence of the radionuclides present in the $^{238}$U, $^{235}$U and $^{232}$Th day chains (Table 1; radioanalytical methodology is provided in Supporting Information 1.1). No activity was detected for radionuclides from the $^{235}$U decay chain or for head of chains $^{238}$U and $^{228}$Th.

**Table 1**

Mean activity concentrations (Bq/g dry mass) of radionuclides with >10 day half-lives determined in pulverised barite scale samples from a Griffin Field well tubular (±SE; n = 3).

| Radionuclide | Half-life | Activity concentration in scale (Bq/g dry weight) |
|---|---|---|
| *Uranium-238 series* | | |
| $^{226}$Ra | 1, 600 years | 180 ± 18 |
| $^{210}$Pb | 22.2 years | 65 ± 7 |
| $^{210}$Po | 138 days | 64 ± 4 |
| *Thorium-232 series* | | |
| $^{228}$Ra | 5.75 years | 44 ± 4 |
| $^{228}$Th | 1.91 years | 63 ± 6 |

## 2.2. Exposure scenario 1: external doses from an intact pipe to marine organisms

To simulate a scenario where scale radionuclides are contained within a pipeline (i.e., during operation or following recent decommissioning), it was assumed a decommissioned pipe was left *in situ*, closed at either end and retained its structural integrity. Therefore, organisms will not be directly in contact with the NORM-contaminated scale, but only exposed though x-rays and gamma radiation that penetrates the pipe. As the ERICA tool cannot account for a cylindrical curved source or shielding effects, the computational program MicroShield ® Pro was used (Version 12.12, Grove Software, further details in Supporting Information 1.2). MicroShield® Pro was used to calculate external-only dose rates from a subsea pipeline containing Ra-contaminated scale to a receptor on the outside of the pipe. The geometry included a radioactive source layer, a shield of steel pipe (representative of a standard API 5LC Seamless 12% Cr steel pipeline, 12.06 cm external diameter) and the pipe hollow middle contained seawater representative of Australian marine environments (Table 2). The modelled pipe had dimensions of 100 cm length and NORM-contaminated barite scale was used as the radioactive source. The source contained radionuclides in the $^{226}$Ra and $^{228}$Ra decay series (Table 1) and allowed the evaluation of the influence of 52x discrete gamma photons with primary energies >0.1 MeV and emission probabilities >1% (Supporting Table 1). All progeny with half-lives <10 days were assumed to be in secular equilibrium with their respective parent. Attenuated gamma radiation, as well as secondary radiation or 'build up' was calculated for the following range of variables: scale activity concentrations (45–1350 Bq/cm$^3$ of $^{226}$Ra and associated progeny) and estimating the likely external dose rates to a biological receptor at different distances from the exterior surface of the pipe (1 cm–20 cm; 1 cm representing organisms attached to the exterior surface of pipe) (Table 2). Radiation build up is a correction factor that considers the influence of the scattered radiation from a material (i.e., a radiation shield) plus any secondary particles in the shielding medium. Micro-Shield calculations that varied receptor distance away from the pipe were assumed to be in air and were calculated for build-up and no build up. MicroShield did not allow for a seawater layer outside the pipe, so additional calculations were needed to account for attenuation by seawater (see Section 2.3). The geometry of the scenario and an illustration of the different input parameters are shown in Fig. 1.

### 2.2.1. Calculation of external-only dose rates with attenuation of radiation though a pipeline to marine organisms

To account for the subsea environment in our assessment, the output from the MicroShield computations required additional manual calculations to be conducted to account for attenuation of x-rays and gamma radiation by seawater outside the pipe (see Supporting Information Section 1.2.1). These calculations had two purposes. Firstly, to investigate the external-only dose rates sessile organisms may experience from Ra-contaminated scale on the surface of the pipe, and secondly, to calculate the distance away from a pipeline required to reduce the external only dose rate to 10 μGy/h (see Section 2.6). The first set of

**Table 2**

Data parameterisations, inputs and justifications for the models simulated in MicroShield® Pro for a 12.06 cm external diameter pipe.

| Parameter | Parameter in MicroShield | Simulated value | Justification |
|---|---|---|---|
| Steel thickness | Outer Cyl Thickness | 0.87 cm | Range of steel pipe thicknesses (in mm) by API 5L specifications |
| Scale thickness | Source | 1.5 cm | A range reflecting limited scale precipitation to 3/4 max pipeline diameter (in mm) |
| Inner void diameter | Inner Cyl Radius | 6.03 cm | Calculated by (outer cylinder thickness - (2 × scale thickness) - (2 × steel thickness)) |

**Additional materials in Micro Shield**

| | Seawater (Representative of Australian marine environment) | Steel pipeline (Outer Cyl) | Barite scale (Source) | |
|---|---|---|---|---|
| **Elemental components and percentage of makeup (%)** | O; 87% | | | |
| | H; 10% | Fe; 78% | Ba; 58.9 | |
| | Cl; 2% | Cr; 12% | O; 27.4% | |
| | Na; 1% | Ni; 8% | S; 13.7% | |
| **Density (g/cm$^3$)** | 1.035 | 7.784 | 4.5 | |

| Activity of source material | Activity concentration (Bq/cm$^3$)[b] | | | | |
|---|---|---|---|---|---|
| Radionuclide[a] | Simulation 1 | Simulation 2[c] | Simulation 3[c] | Simulation 4[c] | Simulation 5[c] |
| Ra-226 | 810 | 45 | 135 | 675 | 1350 |
| Bi-214 | 810 | 45 | 135 | 675 | 1350 |
| Pb-214 | 810 | 45 | 135 | 675 | 1350 |
| | | | | | |
| Ra-228 | 198 | 45 | 135 | 675 | 1350 |
| Ac-228 | 198 | 45 | 135 | 675 | 1350 |
| Bi-212 | 284 | 68 | 203 | 1013 | 2025 |
| Pb-212 | 284 | 68 | 203 | 1013 | 2025 |
| Ra-224 | 284 | 68 | 203 | 1013 | 2025 |
| Tl-208 | 284 | 68 | 203 | 1013 | 2025 |

[a] Only radionuclides with gamma energies >0.1 MeV and >1% emission probabilities were included.

[b] Activity concentrations (Bq/cm3) of each radionuclide in the barite scale multiplied by the density of scale (4.5 g/cm3).

[c] Simulations 2-5 activity concentrations of each radionuclide in the barite scale increase by a factor of 3 to examine impacts of an activity concentration series.
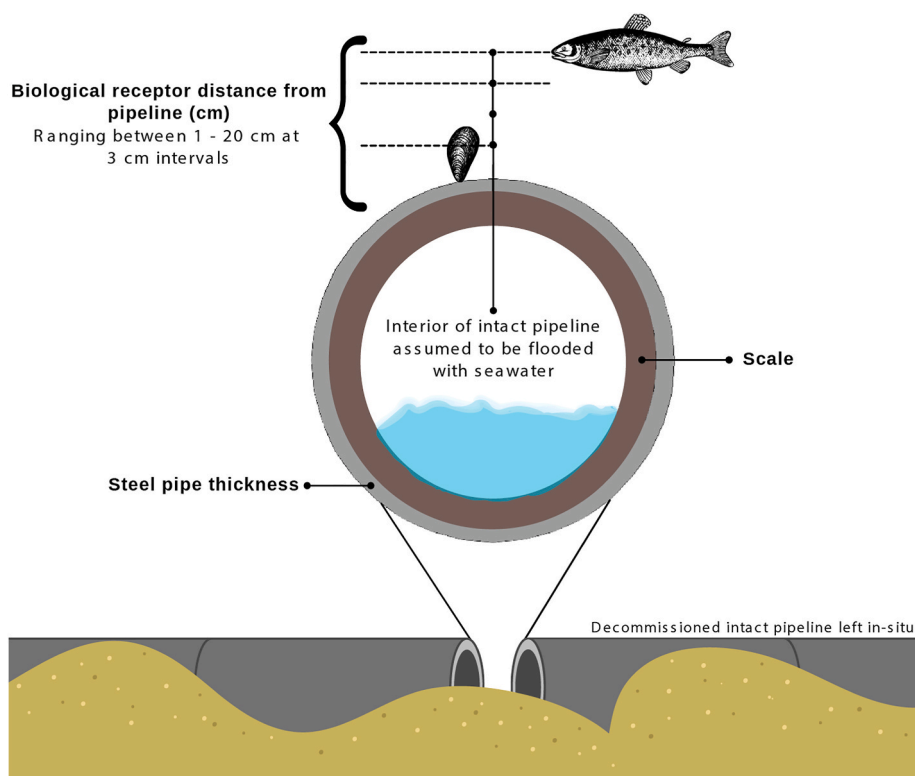
**Fig. 1.** The input parameters used for each computation using MicroShield® Pro to simulate a contained scenario in the environment (external exposure only) of Ra-contaminated scale contained within a decommissioned intact pipeline (100 cm length and 12.06 cm external diameter) left in-situ on the seafloor and flooded with seawater.

calculations used the NORM-scale described in Section 2.2 which varied the dose point distance from the pipe surface (Simulation 1, Table 2). The second set of calculations used an arbitrary activity series of $^{226}$Ra and $^{228}$Ra at a 1:1 ratio in equilibrium with their progeny (Simulation 2–5, Table 2). We investigated an activity concentration series of 45–1350 Bq/g$^{226}$Ra and $^{228}$Ra and progeny to determine the activity of Ra-contaminated scale radionuclides ($^{226}$Ra and $^{228}$Ra in equal proportions and in secular or transient equilibrium with their progeny) that results in a 10 μGy/h dose rate at different distances from the pipe were then calculated by interpolation of linear regressions fitted to model outputs.

### 2.3. Exposure field scenario for ERICA: organisms within internals of pipe after corrosive breakthrough

This scenario models corrosive breakthrough of the pipe and subsequent flooding of pipe internals with seawater, where it is assumed, there are holes and openings that allow some biota to seek refuge inside. The scenario conservatively assumes the openings do not allow for significant water exchange within the structure such that the activity concentrations of radionuclides in the water can be estimated from the scale using partitioning coefficients. In this assessment, the potential radiological doses from the radionuclides of concern were determined for marine organisms such as benthic fish, invertebrates, and zooplankton, that can enter and occupy the interiors of the pipe (assumed to be 12.06 cm external diameter). Hence this scenario considers radiological exposure directly from exposure though contact with the scale and accumulation of radionuclides within the organisms, as well as externally to organisms inhabiting the interior surfaces of the pipe (i.e., molluscs, crustaceans, plankton). There is no standard geometry for curved pipe surfaces within the ERICA Tool. Therefore, it was assumed that the organisms within the pipe are exposed to a planar source (infinite distance) of contamination, such as would be

experienced if an organism was on the surface of contaminated sediment. Two assessments were performed using the variations of data parameters discussed below.

### 2.4. Assessment data for ERICA

For the purposes of this study, we used the ERICA Tool version 2.0 and ran all assessments at the Tier 2 stage using a marine ecosystem. Default occupancy factors, concentration ratios (CR) percentage dry mass of sediments, uncertainty factors, and radiation weighting factors were used and are considered conservative. The ERICA tool contains a well-defined set of representative organisms for which default values are provided for all parameters needed for dose rate calculations (ICRP 2017, Table 3). Only representative organisms that are characteristic of biota identified at Australian offshore petroleum facilities were used

**Table 3**
Representative organisms selected for the ERICA assessments that are likely present in, on or near offshore petroleum subsea pipelines.

| Representative Organism | Mass (kg) | Geometry defined by | Reference |
|---|---|---|---|
| Benthic fish | 1.31E+00 | ICRP Flat fish | ICRP (2017) |
| Crustacean | 7.54E-01 | ICRP Crab | ICRP (2017) |
| Macroalgae | 6.52E-01 | ICRP Brown seaweed | ICRP (2017) |
| Mollusc - bivalve | 1.64E-02 | FASSET Benthic mollusc | Larsson (2004) |
| Pelagic fish | 5.65E-01 | FASSET Pelagic fish | Larsson (2004) |
| Phytoplankton | 1.00E-06 | FASSET Phytoplankton | Larsson (2004) |
| Polychaete worm | 1.73E-02 | FASSET Worm (benthic) | Larsson (2004) |
| Sea anemones & True coral (Polyp) | 1.77E-03 | ICRP Polyp | ICRP (2017) |
| Zooplankton | 6.14E-05 | FASSET Zooplankton | Larsson (2004) |

(Bond et al. 2018a, 2018b; McLean et al. 2019, 2020). Details about the organisms used for the ERICA assessments are provided in Supporting Table 3.

### 2.5. Assessing the influence of different parameterisations

To investigate how different parameterisations may affect the assessment of dose rates, several sub-assessments were performed with different input data, including: (1) comparing default radionuclide $K_d$ values vs. measured $K_d$ values from seawater leach experiments (hereafter referred to as "scale-specific $K_d$ value"; see below) with scale samples, and (2) manual input of short-lived daughter products vs only inputting head of chain parents.

#### 2.5.1. Scale-specific versus default partitioning coefficients of radionuclides

For each ERICA assessment, total concentrations of the radionuclides in the barite scale (Table 4) were used to estimate absorbed weighted dose rates to marine organisms. These were entered as 'sediment' concentrations. Corresponding water concentrations were calculated using partitioning coefficients ($K_d$ values) for each radionuclide. Default and laboratory-derived $K_d$ values were used to investigate the differences to resulting dose rates.

Default partition coefficient ($K_d$) values from the ERICA Tool were based on ocean margin values taken from IAEA (2004) (Table 4). Dose rates using these values were then compared to those estimated using a range of $K_d$ values determined by a seawater leach experiment where various scales samples were continuously rolled in seawater for 1 month in the laboratory (as per Cresswell et al. (2021)). Details on the seawater leachate experiments and radiochemical methods are provided in Supporting Information Section 1.3. The resulting average seawater activity of each radionuclide ($^{210}$Pb, $^{210}$Po and $^{228}$Th) was then divided by the activity concentrations in the total scale ("Scale-specific" values, Table 4). As seawater $^{226}$Ra activities were below detection limits in some replicates of the seawater leach experiments, the minimum detected value of 0.12 Bq/L was used. This approach allows a better estimate of radionuclide solubility from the specific barite matrix of the pipe scale used in our assessment.

#### 2.5.2. Comparison of head of chain only versus the inclusion of short-lived daughter radionuclides

For the barite scales in this study all radionuclides in the $^{226}$Ra and $^{228}$Ra decay chains are represented by the activity concentrations of the parent radionuclides (>10 days half-lives) $^{226}$Ra, $^{210}$Pb, $^{210}$Po, $^{228}$Ra and $^{228}$Th (as there were no detectable activities of $^{238}$U, $^{235}$U and $^{232}$Th). As the short-lived radionuclides (<10 days) have unique $K_d$ and concentration ratio values and are not accounted for, this leads to deviations

from secular equilibrium of the short-lived radionuclides in the water and organisms. Therefore, to investigate a scenario for when the parents and short-lived progeny are not assumed to be in equilibrium, a further assessment was performed to include the daughter radionuclides of the $^{226}$Ra and $^{228}$Ra decay chains with a physical half-life shorter than 10 days; $^{218}$Po, $^{214}$Pb, $^{214}$Bi, $^{214}$Po, $^{210}$Bi, $^{206}$Tl, $^{228}$Ac, $^{224}$Ra, $^{216}$Po, $^{212}$Pb, $^{212}$Bi, $^{212}$Po, $^{208}$Tl (i.e., these radionuclides were explicitly selected for inclusion in the assessment as well as the parent; see Supporting Table 3 for additional details). By including the short-lived daughter products, it is possible to input water or sediment activity concentrations for each and enable $K_d$ values for the progeny to be used. The assessment for the progeny, when added to the primary assessment, had to be conducted in two steps: (i) inputting our measured activity concentrations in scale to estimate the internal dose rate (via application of CR values): (ii) input an activity concentration of each progeny appropriate to estimating the external dose rate (this was calculated as the scale activity concentration multiplied by the radionuclide-specific media activity correction factor from the ERICA Tool database). The media activity correction factor estimates the average unsupported progeny activity concentration in scale over the 1-year integration period assumed in the ERICA Tool (for more details, see Supporting Information Section 1.4.1).

### 2.6. Calculated dose rates, comparative benchmarks and data analysis

The output from MicroShield® Pro and manual calculations were converted from mGy/hr to µGy/hr for comparisons with radiological benchmarks for non-human biota. We used benchmarks of 1) ERICA generic screening level of 10 µGy/h, which is the ecological screening value representing the dose rate at which 95% of species in the ecosystem are expected to be protected (Commonwealth of Australia, 2015), and 2) ICRP Derived Consideration Reference Levels (DCRLs) that are bands of dose rates within which there is likely to be some chance of deleterious effects of ionising radiation occurring to individuals for representative aquatic organisms: 40–400 µGy/h for a Reference Flatfish and Reference brown seaweed and 400–4000 µGy/h for Reference Crab (ICRP, 2014). For the relevant measured organisms with no specific DCRLs, the 40–400 µGy/h band was used for marine benthic invertebrates and the 400–4000 µGy/h band for polychaete worms. If dose rates exceeded the ERICA screening value and the lower value of the ICRP DCRLs, comparisons were made with relevant dose-effect relationships between the relevant radiation exposure and the likely associated radiobiological effects on organisms collated within the FREDERICA database (Copplestone et al., 2008).

Linear regression models were applied to the external-only exposure scenario using manual dose rate calculations and the output from MicroShield® using the parameters from simulations 1–5 (Table 2). We

### Table 4

Data parameterisation inputs for the ERICA assessment assessing the dose rate from only the long-lived radionuclides and comparing the scale-specific versus default ERICA $K_d$ values. Total activity concentrations of the long-lived radionuclides barite scale (Bq/kg) were input as sediment, ERICA default $K_d$ values are retrieved from IAEA (2004) guidelines.

| Radionuclide[a] | Activity concentrations in the scale (Bq/kg dry mass) | $K_d$ | | Estimated seawater concentrations calculated by ERICA (Bq/L) | |
|---|---|---|---|---|---|
| | Neat[b] | Default ERICA value | Scale-specific value[c] | Default ERICA value | Scale-specific value[d] |
| **Assessment inputting long-lived radionuclides only** | | | | | |
| Ra-226 | 180000 | 5.33E+03 | 1.50E+06 | 3.38E+01 | 1.20E-01 |
| Pb-210 | 65000 | 2.66E+05 | 9.03E+05 | 2.44E-01 | 7.20E-02 |
| Po-210 | 64000 | 5.33E+07 | 1.5E+06 | 1.20E-03 | 4.00E-02 |
| Ra-228 | 44000 | 5.33E+03 | 1.5E+06 | 8.26E+00 | 2.90E-02 |
| Th-228 | 63000 | 7.99E+06 | 1.3E+06 | 7.88E-03 | 5.00E-02 |

[a] Naturally occurring radionuclides included in the ERICA assessments.

[b] Neat is representative of 100% of the radionuclide activity concentrations analysed in the pulverised barite scale sample.

[c] $K_d$ values for each of the long-lived radionuclides were calculated from: average total activity concentrations in pulverised scale ÷ average activity concentrations detected from filtered (<0.45 µm) seawater leach tests.

[d] The water activity concentrations were calculated from using the input sediment activity concentrations and the $K_d$ values for each respective radionuclide.

tested the correlations between the external-only exposure models calculated from MicroShield (air attenuation) and the manual calculations for water attenuation by performing linear regression models. All linear regression models, data plotting and visualisations were performed in R Studio software version 9.2 (R Development Core Team, 2016).

## 3. Results and discussion

### 3.1. Contained field scenario of external radiation doses (accounting for shielding from pipeline)

The shielded dose rates for an external only exposure based on a scale activity of 180 Bq/g of $^{226}$Ra (assuming $^{226}$Ra and $^{228}$Ra and their progeny are in secular or transient equilibrium with their head of chain; see Simulation 1 in Table 2) varied between 26 µGy/h (build up and attenuated by water) to 33 µGy/h (no build up and attenuated in air), at the point of external colonisation by marine biota (i.e. 1 cm from pipe surface; Fig. 2). At direct contact with the exteriors of the pipe surface (i. e. d = 0 cm), the dose rate ranged between 28 µGy/h (build up and attenuated by water) and 45 µGy/h (build up from steel and attenuated in air). The calculated external dose rates alone are above the ERICA screening value of 10 µGy/h, and therefore there may be a non-negligible dose to organisms colonising the external surfaces of NORM-contaminated subsea pipelines at the modelled radionuclide concentrations. This is relevant for small organisms attached to the surface that are in the early life stages of development into larger organisms (e.g. eggs, larvae) and would be the most exposed organisms to receive a direct radiological dose.

Our models suggest that organisms need to be between 14.0 cm (assuming only build up from steel and in air) and 17.9 cm (build up from the steel pipe and water attenuation) to receive a dose rate of 10 µGy/h. The water attenuation modelling approach assumed a linear

collimated beam moving away from the pipe (represented as a point source of radiation), whilst MicroShield® uses the actual geometry of a cylindrical pipe to represent a curved, planar source. These approaches illustrated that the receiving dose rate will decrease with the increase of the distance between the source (pipe) and the biological receptor (Fig. 3.). Simplified linear attenuation models to calculate the dose-rate of gamma emissions from the $^{226}$Ra and $^{228}$Ra decay chains assume an inverse square fit from a point source, which is unlikely to occur in an actual exposure scenario that involves both a shield (pipeline) and source (Ra-contaminated scale material) from a cylindrical geometry. However, if the Beer-Lambert law was applied (attenuation in air) to calculate the external dose rates from NORM-contaminated scale inside pipelines, the activity or dose of a collimated monoenergetic photon beam passing through a shield would decrease exponentially. The latter is more applicable for an exposure scenario with a decommissioned pipeline containing NORM-contaminated scale. However, as these external exposure radiological dose models and calculations are theoretical and have a number of underlining assumptions, further work is needed to investigate if these models can be extrapolated to a real-world scenario where marine organisms inhabit the exteriors of an intact decommissioned pipe containing NORM-contaminated scale e.g. conducting external exposure experiments by locating marine organisms at varying distances to shielded NORM-contaminated scale and measuring the receiving external dose rates.

The dose assessment approach adopted in the ICRP Publication 136 (2017) is based on the uniform isotropic model where radioactive sources are homogenously distributed in the environment and organisms which share the same density and elemental composition. However, this is unlikely to occur in the marine environment around a subsea NORM-contaminated structure, along with the interactions with surrounding marine organisms. The interaction between photons and a shielding medium such as seawater is energy and shielding specific for both the attenuation of the primary photon source, as well as the
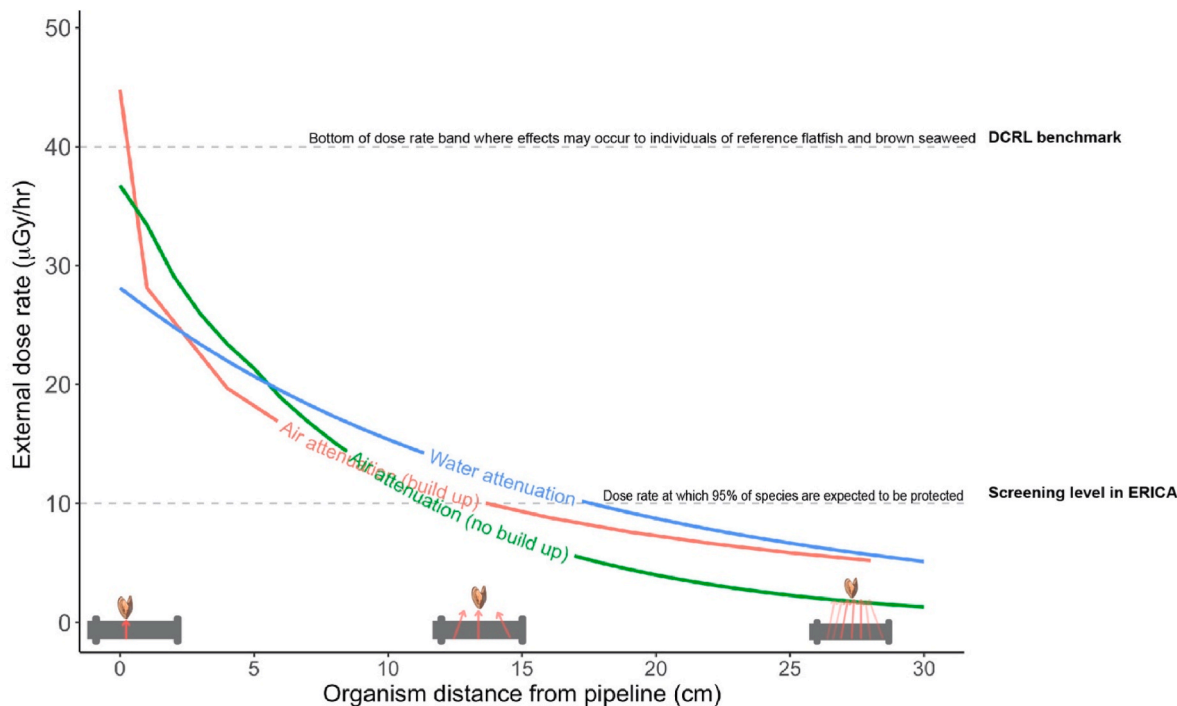


**Fig. 2.** Modelled external dose rates based on the organism distance from a subsea pipeline (cm) using MicroShield® for air attenuation (build up in red; no build up in green) and additional manual calculations to account for attenuation from seawater (blue line; see section 2.3). Comparisons were made to the following radiological benchmarks: ERICA default screening value of 10 µGy/h (lower grey dashed line) and the Derived Conservative Reference Level (DCRL) lower level for reference flatfish and brown seaweed of 40 µGy/h (upper grey dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
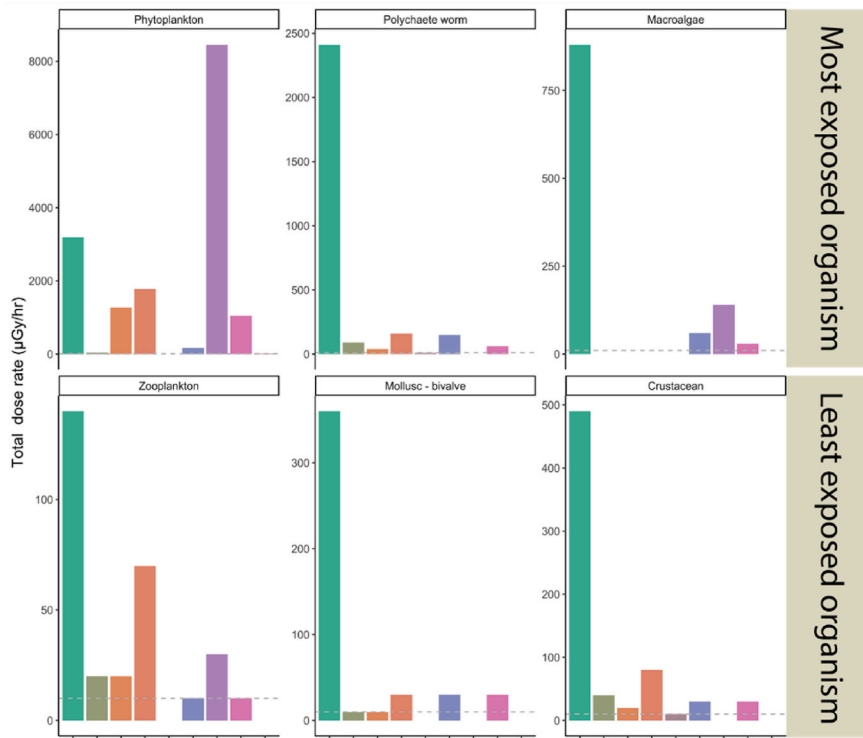
**Fig. 3.** Individual radionuclide contributions to the total absorbed dose rates (μGy/hr; only dose rates above 1 μGy/h are illustrated) to the three most (i.e. highest total dose rate) and least (i.e. (lowest total dose rate) exposed marine organisms using **A)** Default ERICA $K_d$ values and **B)** scale-specific experimental $K_d$ values. Dose rates were calculated from the scale-specific activity concentrations from Ra-contaminated barium sulphate scale (Table 4) from a decommissioned offshore well tubular. All details and assumptions on the modelled scenario are provided in Sections 2.4 and 2.6. The ERICA default 10 μGy/h screening value is depicted by a grey dashed line. Radionuclides are in the order of their decay series.
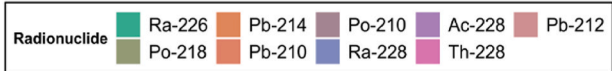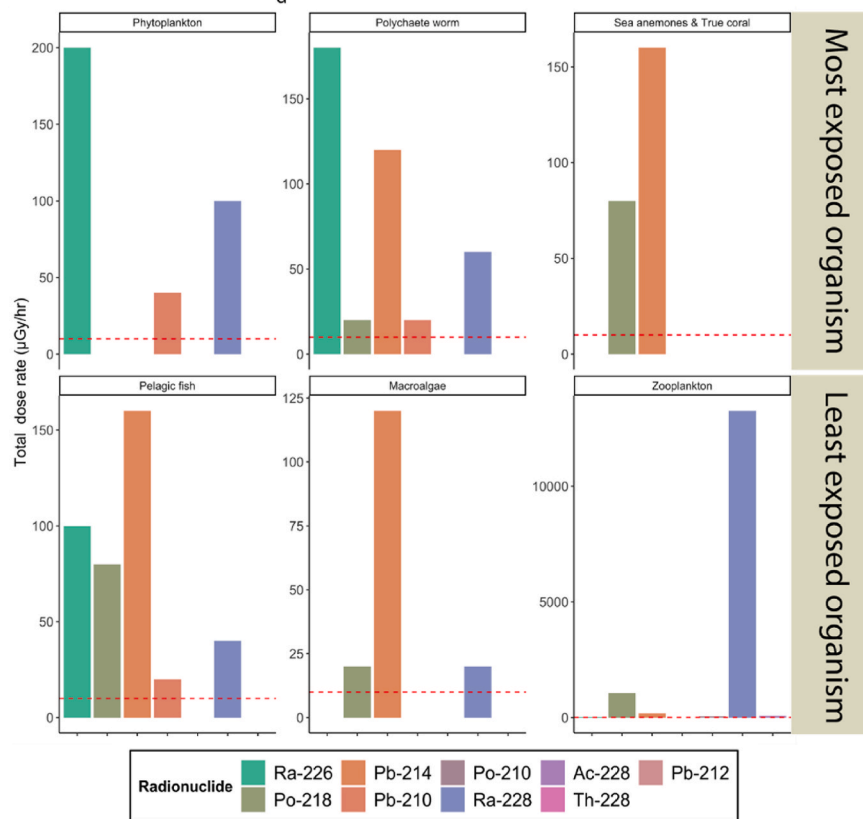
potential generation of secondary radiation from other effects e.g., photoelectric effect, Compton scattering and pair production. The utilisation of MicroShield ® can account for these processes in relation to the complex materials and cylindrical geometries of subsea oil and gas pipelines.

### 3.2. Dose rates to marine organisms within a decommissioned pipe after corrosive breakthrough

#### 3.2.1. Default and scale-specific dose rates to marine organisms

The estimated total doses for all the reference organisms using default ERICA $K_d$ are considerably higher than the ERICA generic screening value (10 μGy/h; Table 5) and were above the ICRP 40–400 μGy/h dose bands for flatfish and brown seaweed and within the ICRP 400–4000 μGy/h dose band that for reference crabs, suggesting potential impacts to vertebrates, invertebrates and marine flora. The dose rates varied substantially between organisms, ranging from 300 μGy/h to 16,000 μGy/h and the most exposed organisms with the highest dose rates being for phytoplankton (16,000 μGy/h), polychaetes (2900 μGy/h), and macroalgae (1100 μGy/h; Table 5 and Fig. 3).

When using scale-specific $K_d$ values (Table 4), the total dose rates were considerably lower ranging from a 17%–86% decrease in the predicted dose rates when using scale-specific $K_d$ values (Table 5). However, the dose rates were still above the ERICA screening dose rate, with four organisms exceeding the ICRP 400 μGy/h DCRL lower band for crabs (Table 5). The most exposed organisms with the highest dose rates were phytoplankton (7300 μGy/h), polychaete (860 μGy/h) and sea anemones and true coral (730 μGy/h; Table 5), the latter were comparable to the default ERICA calculated assessment previously described. The total dose rates of all the modelled marine organisms for both assessments are shown in Supporting Figs. 1 and 2. However, when determining the potential risk of radiobiological effects from this exposure, there is plausible data on radiobiological effects. Hence, more investigation is warranted e.g. conduct laboratory-dose response experiments with endpoints such as growth and reproduction to better determine radiobiological effects.

For both assessments, the largest dose contributors were $^{226}$Ra, $^{210}$Pb and $^{228}$Ra with the dominant radionuclide being $^{226}$Ra for all organisms, except for the phytoplankton for which it was $^{228}$Ac (Supporting Figs. 2

**Table 5**
Estimated total dose rates (μGy hr$^{-1}$; 2 significant figures) to ERICA reference marine organisms that are likely to seek refuge and inhabit a corroded pipeline estimating using the neat activity concentrations from NORM-contaminated scale (180 Bq/g of $^{226}$Ra), default ERICA and scale-specific $K_d$ values, and ERICA calculated seawater concentrations. Values highlighted in bold exceed the ERICA screening value of 10 μGy/h and the respective ICRP Derived Consideration Reference Levels (DCRLs) for representative aquatic organisms; 40–400 μGy/h for a Reference Flatfish (fish) and Reference brown seaweed (macroalgae) and 400–4000 μGy/h for Reference Crab (crustaceans).

| Organism | Default ERICA $K_d$ values | Scale-specific $K_d$ values | Percentage decrease in total dose rate (default vs scale-specific $K_d$ values) |
|---|---|---|---|
| | Total Dose Rate per organism (μGy hr$^{-1}$)[a] | | |
| Benthic fish | **990** | **250** | 74% |
| Crustacean | **700** | **380** | 45% |
| Macroalgae | **1120** | **170** | 84% |
| Mollusc - bivalve | **470** | **210** | 55% |
| Pelagic fish | **860** | **120** | 86% |
| Phytoplankton | **16000** | **7300** | 54% |
| Polychaete | **3000** | **860** | 70% |
| Sea anemones & True coral | **880** | **730** | 17% |
| Zooplankton | **290** | **190** | 35% |

[a] Includes all long-lived radionuclides and short-lived daughter products calculated by default in ERICA.

and 3).

A comparison of the predicted dose rates with the dose-effect relationships from the FREDERICA database (Copplestone et al., 2008) indicates that some of the modelled organisms may experience radiobiological effects when exposed to the activities from the NORM-contaminated scale at activity concentrations used in this assessment. The organisms most vulnerable to exposure of mixed types of radiation were macroalgae, phytoplankton, polychaete, sea anemones and zooplankton, with the FREDERICA database showing dose rates >1000 μGy/h may be sufficient to have some impact on molluscs and polychaetes and <40 μGy/h for pelagic fish, in terms of increased mortality, increased morbidity and reproductive capabilities. However, comparing our assessments with the DCRLs and ERICA default screening values was difficult due to the lack of available data for the respective dose rates for the organisms. For example, the chronic marine toxicity data set in FREDERICA only covers three taxonomic groups; aquatic invertebrates, molluscs, and fish (Garnier-Laplace et al., 2008). Furthermore, a review by MacIntosh et al. (2021) suggested that some organisms may be able to withstand high radiological doses and therefore have no observable radiobiological effects. Thus, relating the exposure dose rate to biological effects needs to be carefully interpreted because of the high variability in inter-species tolerance to radiation.

There is a scarcity of data regarding acute and chronic effects of NORMs to individual and populations of marine organisms (MacIntosh et al., 2021). If dose rates are estimated to be below the 10 μGy/h screening level, one could assume the radiological risks are low. If the screening level and DCRLs are exceeded, as was the case in this assessment, further work is required to understand the potential individual and population level effects of exposure to ionising radiation from NORM scale under the site-specific exposure scenario.

Using default ERICA $K_d$ parameters (Table 4), the estimated internal dose rates ranged between 290 and 16,000 μGy/h, whereas external dose rates were considerably smaller ranging between 0.04 and 270 μGy/h (Table 6). When the scale-specific $K_d$ values were used, instead of default values, the internal dose rates were considerably lower ranging between 39 and 7340 μGy/h, reflecting the lower solubility of key radionuclides such as radium and lead when incorporated within a barite matrix. However, the external dose rates were the same (except for pelagic fish, phytoplankton and zooplankton) as the default ERICA assessment because the input sediment activity concentrations were the same (Table 6). Internal and external dose rates of all the modelled marine organisms for both assessments are illustrated in Supporting Figs. 3–6.

By assessing the potential doses from internal and external exposure using the default ERICA $K_d$ parameters, between 72 and 100% of the total dose rate to representative organisms are the result of internal exposure to the radionuclides with only <1–28% of the total dose rate from external exposure (Table 6). Over 80% of the total internal dose rate to reference organisms, except for phytoplankton, is the result of internal exposure to $^{226}$Ra and $^{210}$Pb as the main internal dose contributors (Fig. 4). In addition, $^{226}$Ra was the dominant radionuclide for external exposure representing on average 73% of the external dose rate to all reference organisms (Fig. 4). The assessment predicted a minor contribution of up to 0.68% to the total dose rates to the organisms from $^{210}$Po (Fig. 4). Using the scale-specific $K_d$ values, the range of internal dose contribution decreased (20–100%), resulting in a higher contribution from the external dose (<1–80%) to the representative organisms. For both assessments, the representative organisms with the highest contributions from external dose were crustaceans and molluscs (Table 6). For the pelagic fish and phytoplankton, virtually all the potential total dose rate was contributed from internal exposure (100%; Table 6).

The radionuclide contributions to the external dose rates were similar between the two different approaches; however, there were considerable differences in internal dose rates (Fig. 4). Using scale-specific solubility data as inputs, over 80% of the total internal dose

**Table 6**

Summary of the calculated total internal and external dose per organism and the percentage contribution of the respective dose rate (% of the total dose) to the total dose rate (μGy/hr$^{-1}$) to representative organisms exposed via direct contact to the radionuclides in the barite scale, estimated by the activity concentrations in the Ra-contaminated barite scale provided in Table 4 and default ERICA $K_d$ values or scale-specific $K_d$ values and input parameters provided in SI Table 4.

| Dose rate (μGy/hr$^{-1}$) | | | | Percentage of total dose rate (%) | |
|---|---|---|---|---|---|
| Organism | Total dose rate per organism | Internal dose rate per organism | External dose rate per organism | Percentage from internal dose rate (%) | Percentage from external dose rate (%) |
| **Default ERICA $K_d$ values** | | | | | |
| Benthic fish | 990 | 860 | 120 | 87 | 13 |
| Crustacean | 700 | 580 | 120 | 83 | 17 |
| Macroalgae | 1130 | 980 | 140 | 87 | 13 |
| Mollusc - bivalve | 470 | 340 | 130 | 72 | 28 |
| Pelagic fish | 830 | 860 | 0.04 | 100 | <1 |
| Phytoplankton | 16000 | 16000 | 0.05 | 100 | <1 |
| Polychaete worm | 2900 | 2600 | 270 | 91 | 9 |
| Sea anemones & True coral | 880 | 740 | 140 | 84 | 16 |
| Zooplankton | 290 | 290 | 0.05 | 100 | <1 |
| **Barite scale-specific $K_d$ values** | | | | | |
| Benthic fish | 250 | 120 | 120 | 50 | 50 |
| Crustacean | 80 | 270 | 120 | 70 | 30 |
| Macroalgae | 170 | 39 | 140 | 20 | 80 |
| Mollusc - bivalve | 210 | 80 | 130 | 40 | 60 |
| Pelagic fish | 120 | 125 | 1.67E-04 | 100 | <1 |
| Phytoplankton | 7340 | 7340 | 2.65E-04 | 100 | <1 |
| Polychaete worm | 860 | 590 | 270 | 70 | 30 |
| Sea anemones & True coral | 730 | 580 | 140 | 80 | 20 |
| Zooplankton | 190 | 190 | 2.27E-04 | 100 | <1 |

rate to modelled organisms, except for phytoplankton, is the result of internal exposure to $^{210}$Po (average 67%) (Fig. 4). Similar to the default ERICA assessment, $^{226}$Ra was the dominant radionuclide in external exposure representing on average 68% of the external dose rate to all reference organisms (Fig. 4). However, the scale-specific assessment predicted minor contributions from the internal dose rate of up to 21% of $^{228}$Th to the total dose rates to the organisms (Fig. 4). This highlights the importance of using scale-specific experimental data (i.e., $K_d$) values, specifically derived for the pipeline scale being assessed for ERICA modelling. Using the default $K_d$ values in ERICA may under- or over-estimate solubility of individual radionuclides, which in turn have a consequence on the estimated internal radionuclide concentration with the representative organisms, and hence influence the estimated internal radiation dose to organisms by ERICA.

Standard IAEA $K_d$ values are derived from marine sediment without contamination and reflect radionuclide activities in the marine environment in equilibrium between the sediment and water that do not consider the behaviour of radium (generally insoluble in anoxic conditions) in addition to not accounting for disequilibrium conditions. As soon as there is the addition of contamination, there is no longer an equilibrium and so understanding radionuclide's behaviour may not be best described by a $K_d$ value. For example, the default radium $K_d$ value is $5.3 \times 10^3$ whereas a $K_d$ based on the RaSO$_4$ solubility in seawater would be $3.2 \times 10^8$ (Matyskin, 2016). A recent study found using literature-retrieved $K_d$ values for uranium milling associated NORMS varied over six orders of magnitude for $^{210}$Po and $^{230}$Th and thee orders of magnitude for $^{226}$Ra, clearly demonstrating there is high variability of partition coefficient values and the assessment outcome is dependent on the selected value (Goulet et al., 2022; Koppel et al., 2022). This illustrates the behaviour of sulphate and carbonate minerals of Ra (individually or co-precipitated with barium, strontium, or calcium) in sediments is still not well understood. However, these geochemical reactions will likely play an important role in moderating the risk of NORM-contaminated scales in the marine ecosystem.

In this case study, benthic and microorganisms (polychaete, macroalgae) are the most exposed group of organisms, given that they reside within or on top of the pipeline. Whilst marine polychaetes had the highest estimated radionuclide activity concentrations, this case study did not focus on radionuclides in the sediment and therefore polychaetes and other sediment-dwelling organisms are realistically not going to be exposed to pipe scale within subsea pipelines. Nevertheless, sediments can be contaminated by the presence of scale and subsequently sediment-dwelling organisms can still be exposed via ingestion of sediment as they burrow or though absorption directly though their soft tissue by endocytosis and is likely reflected in the estimated dose rates. Re-precipitated sulphate phases of radium can be rapidly absorbed or transported by particulate matter and can likely have small enough particle sizes to be susceptible for filter-feeding and microorganisms, such as phytoplankton, molluscs, sea anemones and coral (Lepland et al., 2000; Ahmad et al., 2021). This is especially important because these habitat forming organisms are key ecological components of artificial reef ecosystems formed on decommissioned offshore petroleum infrastructure (Bull and Love 2019; McLean et al., 2022).

*3.2.2. Inclusion of unsupported short-lived daughter progeny on dose rates*

When the unsupported fraction of the short-lived daughter radionuclides (half-life shorter than 10 days) of the $^{226}$Ra and $^{228}$Ra decay chains are included in the ERICA V2.0 assessment, equilibrium between the parents and the progeny is no longer assumed and the activity concentration in the environmental media correspond to the average unsupported fraction for a one-year integration period. In brief, this scenario investigated the likelihood that disequilibrium will occur between the organism tissues and the surrounding environmental concentrations, reflecting differences in individual radionuclide aqueous partitioning and subsequent bioaccumulation into organisms' tissues. The main outcomes of the two ERICA assessments using the calculated unsupported fraction of the short-lived daughter progeny (default $K_d$ values vs scale-specific $K_d$ values) are as follows, with comprehensive results and discussion provided in Supporting Information Section 2.1:

I) The total dose rate to the organisms increases for only the mollusc using default $K_d$ values, whilst it decreases for macroalgae, pelagic fish and phytoplankton for both assessments (Table 7).
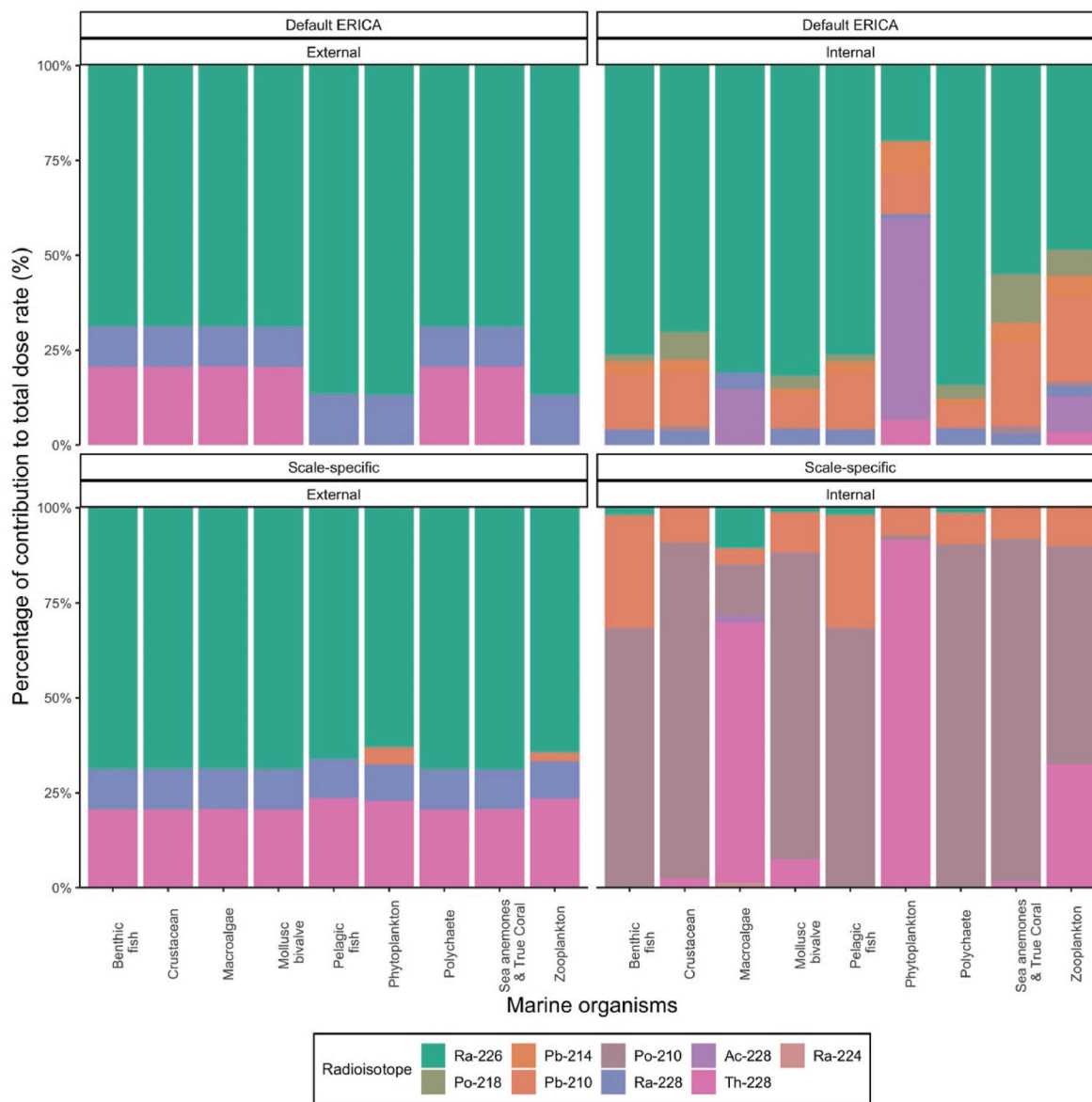
**Fig. 4.** Percentage contribution of each radionuclide to the total dose rate (%) for all the modelled marine organisms. Only radionuclides contributing more than 1% to the total dose rate are shown. Dose rates were calculated from the scale-specific activity concentrations from radium-contaminated barium sulphate scale (Table 4) from a decommissioned offshore subsea oil and gas pipe for ERICA assessments using default ERICA $K_d$ values and scale-specific experimental $K_d$ values. All details and assumptions on the modelled scenario are provided in Sections 2.4 and 2.6.

There were no significant differences between the total dose rates for all the other organisms (Table 7).

II) For both assessments, the internal dose decreases substantially although the dose rates still exceed screening and radioecological protection benchmark values (Supporting Fig. 7B and Supporting Fig. 8B.).

III) The external dose rate for the organisms did not differ between the two assessments (Supporting Fig. 7A and Supporting Fig. 8A).

IV) When the unsupported fraction of the short-lived progeny is accounted for, though the dose rates decrease, there are greater dose contributions from $^{226}$Ra, $^{210}$Pb and $^{228}$Th for all organisms (Supporting Fig. 7 and Supporting Fig. 8).

Overall, there were no substantial differences when conducting a separate assessment using radionuclide specific $K_{ds}$ and concentration ratios for the short-lived radionuclides. The ICRP published an approach whereby progeny need to be considered in a separate assessment, and can account for complications of the numerous progeny products in both

the $^{238}$U and $^{228}$Th day series and the individual partitioning of these radionuclides between seawater and marine sediments (ICRP, 2017). Furthermore, parent and daughter radionuclides that have similar geochemical and biochemical behaviours within organisms are assumed to have additive effects that further increases the overall risk. Therefore, environmental risk assessments for NORM-contaminated products should therefore consider the likelihood of the daughter product that is associated with the parent product, and which may contribute significantly to the overall dose.

The need to include the unsupported short-lived radionuclides has been raised in previous model and field-based studies using the ERICA Tool (Strand et al., 2014). Over time, the shorter-lived radionuclides will continue to decay, and thus may contribute a radiation dose to an exposed marine organism in that brief period of exposure. In the short-term, this has been shown post-accident at Chernobyl and Fukushima where the short-lived radionuclides contribute a dose to freshwater and marine biota (Strand et al., 2014; Li et al., 2019; Beresford et al., 2020; Lu et al., 2021). Accounting for the short-lived

**Table 7**

Comparison of the total dose rates (µGy hr; 2 s.f.) between two ERICA assessments using i) inclusion of only the head of chain long-lived parents and supported short-lived radionuclides and ii) inclusion of the unsupported fraction of short-lived radionuclides, to reference marine organisms surrounding a corroded subsea pipeline using the neat activity concentrations from NORM-contaminated scale (180 Bq/g of $^{226}$Ra), default ERICA $K_d$ values and scale-specific $K_d$ values, and ERICA calculated seawater concentrations (Table 4). All details and assumptions on the modelled scenario are provided in Section 2.4 and 2.6.2. All values exceed the ERICA screening value of 10 µGy/h and the respective ICRP Derived Consideration Reference Levels (DCRLs) that have a band of dose rates within which there is likely to be some chance of deleterious effects of ionising radiation occurring to individuals for representative aquatic organisms; 40–400 µGy/h for a Reference Flatfish (fish) and Reference brown seaweed and 400–4000 µGy/h for Reference Crab (crustaceans and worms). Differences between the two assessments are represented in bold and '*'.

| Assessment | | Only including the head of chain parents and supported short-lived radionuclides [a] | Inclusion of unsupported fraction of short-lived daughter radionuclides [b] |
|---|---|---|---|
| **Default $K_d$ values** | **Organism** | **Total Dose Rate per organism (µGy hr$^{-1}$)** | |
| | Benthic fish | 990 | 990 |
| | Crustacean | 700 | 690 |
| | Macroalgae | **1120*** | **1040*** |
| | Mollusc - bivalve | **470*** | **510*** |
| | Pelagic fish | **860*** | **810*** |
| | Phytoplankton | **16000*** | **6190*** |
| | Polychaete | 3000 | 2910 |
| | Sea anemones & True coral | 880 | 810 |
| | Zooplankton | 290 | 230 |
| **Scale-specific $K_d$ values** | Benthic fish | 250 | 250 |
| | Crustacean | 380 | 380 |
| | Macroalgae | **170*** | **150*** |
| | Mollusc - bivalve | **210*** | **200*** |
| | Pelagic fish | 120 | 120 |
| | Phytoplankton | **7340*** | **1670*** |
| | Polychaete | 860 | 860 |
| | Sea anemones & True coral | 730 | 720 |
| | Zooplankton | 190 | 140 |

[a] Includes all long-lived radionuclides (>10 days) and short-lived daughter products (<10 days).

[b] The unsupported fraction of each of the short-lived radionuclides were calculated via the methodology outlined in Section 2.6.2. Unsupported concentration (corrected external dose rate = media correction factor x concentration of radionuclide in sediment).

radionuclides is complicated due to the varying radiation dose, dose rate, temporal and spatial variations and the radio sensitivities of the different marine biota to the short-lived progeny. Furthermore, there is insufficient evidence of accumulation in marine organisms of short-lived radionuclides in NORM-contaminated by-products (MacIntosh et al., 2021; Koppel et al., 2022). This case study is an example of assessing what could be the overall result of accounting for the head of chain and all the decay products when assessing marine biota exposure to radiation, in the absence of scale-specific data.

### 3.3. Assessment uncertainties

The use of the ERICA Tool for modelling worst-case environmental exposure scenarios of NORM-contaminated petroleum products to marine biota has several underlining uncertainties that could impact the certainty and applicability of the results from this case study. Whilst the ICRP DCRL 400 µGy/h lower benchmark was exceeded for all the invertebrate representative organism types it applies to (i.e., crustacean, mollusc-bivalve, phytoplankton, polychaete), it is difficult to

extrapolate the dose rate exceedances as there are no data regarding the full extent of effects from exceeding the 10 µGy/h screening level for benthic marine organisms in a subsea marine environment. It is worth noting the 10 µGy/h screening dose rate is used as a baseline for all potentially exposed reference plants and animals and was derived with very little data for marine organisms (Garnier-Laplace et al., 2008). However, as there are large uncertainties in using default parameter values, they may not be protective of all marine organisms.

This exposure scenario is limited to assuming environmental conditions within a pipe where only a small percentage of pelagic and benthic organism populations could reside and the realistic extent and area of corrosive breakthrough. In addition, the exceedances are only relevant for the short-term (i.e., acute) period when breakthrough happens and when no substantial water exchanged has occurred. However, these assessments illustrate that NORM-contaminated products are still likely to pose a degree of risk to surrounding marine life, if left *in situ* and assume 100% organism occupancy within the flooded pipe.

By illustrating a comparison between using scale-specific and ERICA's default $K_d$ values, it demonstrates a difference between global averages and case-specific data. Using site-specific data is recommended in the ERICA approach wherever possible. This case study supports this approach and has highlighted the large differences in dose rates when applicable for scale-contaminated products. The $K_d$ approach assumes equilibrium between the sediment and water, which is highly unlikely to occur in the marine environment following large-scale corrosive breakdown of contaminated pipelines due to natural sea currents interacting with the uncontaminated water across the small area of internal pipeline scale. An initial pulse of radionuclides, such as from an emergency or planned release scenario may lead to an increase in sediment activity that will persist much longer than the water activities. This means that using equilibrium-based $K_d$ values will overestimate sediment activities at the initial release and underestimate sediment activities once seawater activities have returned to a background state (Periañez et al., 2018). The consequence to an ecological risk assessment using default parameters would depend on the media being sampled and the time it is sampled relative to a release event. It is important for subsea oil and gas infrastructure operators to consider that sediment activity concentrations may increase over time, and the $K_d$ values will change as radionuclide concentrations within sediments increase (Kusakabe and Takata 2020). Therefore, this suggests the exposure scenario needs to be considered highly conservative, given the time that sufficient corrosive breakthrough of the pipe has allowed macro-roganisms to enter and there would likely be seawater exchange to reduce the internal activity concentrations of NORM-contaminated barite scale.

## 4. Future research directions

To better understand the effects of short and long-term radiological risks of NORM exposure to marine biota from offshore oil and gas infrastructure, we recommend the following approaches to improve the use of radiological dose modelling tools for incorporation into environmental risk assessments:

### 4.1. Derive contaminant, speciation specific $K_d$ values to better model NORM behaviour and partitioning

This case study has demonstrated that default ERICA $K_d$ values may not be appropriate when considering the behaviour of NORM-contaminated products in the marine environment. Default $K_d$ values can lead to under or overestimation of radionuclide exposure which is why the ERICA Tool recommends the use of site-specific $K_d$ values. However, in the absence of contaminated environments, laboratory testing may provide a more applicable value. The speciation of the radionuclides in NORM-contaminated material is unlikely to reflect a state of equilibrium state when released into the environment. Therefore,

seawater leachate tests to determine the solubility of radionuclides in scale is recommended (Cresswell et al., 2021). A leach test gives a conservative estimate of radionuclides that would be solubilised over time, because it is conducted in a closed system, and the material undergoing leaching has greater surface area than when attached as scale to the internal surfaces of a pipeline. However, a leach test will not predict speciation changes from NORM release to different environment conditions, i.e. anoxic sediments, where solubilities may be much greater because of chemical processes such as reductive dissolution. If this is relevant to the release of NORM-contaminated material, then assessing the diffusive flux of radionuclides from the material in sediments to overlying waters could be used as a better measurement of radionuclide partitioning. This has been investigated in freshwater systems but not marine systems.

### 4.2. Derive concentration ratios for scale-specific radionuclides and relevant marine species to make the model relevant to oil and gas infrastructure and organisms that are likely to colonise them

The concentration ratio may be a poor predictor of radionuclide bioaccumulation in marine organisms. The Wildlife Transfer Database contains data on concentration ratio values that are used to parameterise ERICA assessments. However these values may not reflect the scale-specific conditions or the unique biology of local species (Copplestone et al., 2013). In addition, not all scale-based radionuclides have concentration ratio values for all organisms, which means other approaches or analogues are used (Hosseini et al., 2008). Gaps in the database still exist with respect to marine data related to NORM exposure. As a large amount of subsea oil and gas infrastructure to be decommissioned may contain NORM-scale, there is a requirement for more data on the parameters of NORM transfer to marine organisms (Hirth et al., 2017). Focus needs to be given to the creation of an inventory of concentration ratio values for scale-specific radionuclides in local organisms and of particular importance to local communities and fisheries (Hirth et al., 2017; Koppel et al., 2022).

### 4.3. Develop marine-specific radiotoxicity data and guidelines to improve model accuracy in marine environments

Using ERICA for this case study highlighted difficulties in comparing the assessments with dose-effect relationships from databases, due to the lack of, or no available data for the dose rates for the representative organisms. The predicted 95% species protection level (Garnier-Laplace et al., 2008) of 10 µGy/h that is used as default in ERICA, incorporates data for all species in all ecosystems (i.e., terrestrial, freshwater and marine) and so may be overly conservative for marine ecosystems. Radiotoxicological data from marine organisms exposed to NORM-contaminated products is limited (MacIntosh et al., 2021). Research is still needed to understand the bioavailability and bioaccumulation potential of radionuclides from NORM-contaminated products to marine organisms. This will increase certainty on dose-response relationships and refine estimates of radiation dose and subsequent acute and chronic radiation-induced effects.

## 5. Recommendations for offshore petroleum decommissioning risk assessments

Both MicroShield® and the ERICA Tool can be used as radiological dose assessment tools for risk assessments of NORM-contaminated products. The use of different approaches (default, scale-specific partition coefficients, considering short-lived progeny of the radionuclide decay chains) showed variable results. Fully integrating all potential NORM-associated contaminants is the best approach to providing a robust demonstration of the level of associated radiological risk to marine flora and fauna and whether there is going to be a risk in the short and long-term following decommissioning of NORM contaminated

pipelines. Hence, we recommend a checklist of how to effectively use biota dose modelling tools for their application in offshore petroleum decommissioning environmental risk assessments, which considers the nature of scale-contaminated products and all exposure pathways to marine biota.

### 5.1. Demonstrate conservatism for planned exposure scenarios

- There is a need to demonstrate that assessments are conservative where future radionuclide releases are planned. This is to meet the requirements of the precautionary principle under ecologically sustainable development and recognises that there are limited data describing radionuclide behaviour and impacts in the marine environment.
- External-only exposure assessments should be performed using MicroShield or similar radiological dose software that can account for exposure geometries to marine organisms located on the external surfaces of enclosed pipes and associated shielding effects.
- In the absence of measured radionuclide activities for progeny, assume secular equilibrium of progeny with measured parent radionuclide.

### 5.2. Site specific parameterisations

- Site-specific environmental risk assessments should include either or both radiological biota dose tools in the efforts to predict the ecological and radiological impacts from scale-specific NORM relevant to scale-contaminated infrastructure.
- Scale-specific data should ideally be used as it provides a better approach to understanding the radiological risks to the marine environment for a given decommissioned structure.
- Seawater leachate tests is recommended to determine the solubility of scale-specific potential to retrieve scale-specific data.
- Using the ERICA Tool to assess NORM-contaminated products should include representative marine organisms that colonise pipelines.

### 5.3. Calculation methods

- Where scale may reside in contained infrastructure for long periods of time, external-only exposure assessments should be performed using MicroShield or similar radiological dose software.
- The manual calculation and inclusion of the shorter-lived radionuclides should be considered in a separate assessment, although not a necessity it is recommended by the ICRP (2017).

### 5.4. Consider all impacts and risks

- Organism effects testing in controlled laboratory conditions, or the field should be undertaken along with the creation of food-web models to provide an in-depth analysis of the tropic transfer of radionuclides among the marine organisms.

**CRediT authorship contribution statement**

Amy MacIntosh: Conceptualisation, Methodology, Formal analysis, Investigation, Data Curation, Writing-Original draft preparation, Writing-reviewing and editing, Visualization, Tom Cresswell: Conceptualisation, Methodology, Supervision, Funding acquisition, Validation, Writing-Original draft preparation, Writing-reviewing and editing, Project administration., Darren Koppel: Conceptualisation, Methodology, Validation, Writing-Original draft preparation, Writing-reviewing and editing., Nick Beresford: Methodology, Software, Validation, Writing-Original draft preparation, Writing-reviewing and editing., David Copplestone: Methodology, Software, Validation, Writing-reviewing and editing, Mathew Johansen: Methodology, Validation, Writing-reviewing and editing., Beth Penrose: Supervision, Writing-

reviewing and editing.

## Funding

This study was partially supported by BHP Petroleum Pty Ltd, a wholly owned subsidiary of Woodside Energy Group Ltd as part of a long-term collaborative project between the Australian Nuclear Science and Technology Organization (ANSTO) and Macquarie University (Project ID 113793469: Developing an ecological framework for closure of offshore petroleum structures). This work was supported by an Australian Government Research Training Program scholarship and a New South Wales (NSW) FutureNow Scholarship to A.M.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Tom Cresswell reports financial support was provided by BHP Petroleum Inc.

## Data availability

Data will be made available on request.

## Acknowledgements

The authors acknowledge all the Aboriginal and Torres Strait Islander custodians on whose Traditional Land and Countries this research was conducted. We strongly support equity, diversity and inclusion in science. We, the authors, come from different countries (Scotland, England, South Africa, the United States, Australia and New Zealand) and represent different career stages.

## Appendix A.  Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2022.106979.

## References

Ahmad, Faraaz, Morris, Katherine, Law, Gareth T.W., Taylor, Kevin G., Shaw, Samuel, 2021. Fate of radium on the discharge of oil and gas produced water to the marine environment. Chemosphere 273, 129550.

Ali, Mohsen M.M., Zhao, Hongtao, Li, Zhongyu, Maglas, Najeeb N.M., 2019. Concentrations of TENORMs in the petroleum industry and their environmental and health effects. RSC Adv. 9 (67), 39201–39229.

Beresford, N.A., Scott, E.M., Copplestone, D., 2020. Field effects studies in the Chernobyl exclusion zone: lessons to be learnt. J. Environ. Radioact. 211, 105893.

Bond, T., Partridge, J.C., Taylor, M.D., Cooper, T.F., McLean, D.L., 2018a. The influence of depth and a subsea pipeline on fish assemblages and commercially fished species. PLoS One 13 (11).

Bond, T., Partridge, J.C., Taylor, M.D., Langlois, T.J., Malseed, B.E., Smith, L.D., McLean, D.L., 2018b. Fish associated with a subsea pipeline and adjacent seafloor of the North West Shelf of Western Australia. Mar. Environ. Res. 141, 53–65.

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.

Bull, Ann Scarborough, Love, Milton S., 2019. Worldwide oil and gas platform decommissioning: a review of practices and reefing options. Ocean Coast Manag. 168, 274–306.

Carolan, J.V., Hughes, C.E., Hoffmann, E.L., 2011. Dose assessment for marine biota and humans from discharge of (131)l to the marine environment and uptake by algae in Sydney, Australia. J. Environ. Radioact. 102 (10), 953–963.

Copplestone, David, Hingston, J.L., Real, Almudena, 2008. The development and purpose of the FREDERICA radiation effects database. J. Environ. Radioact. 99, 1456–1463.

Commonwealth of Australia, 2015. Guide for Radiation Protection of the Environment. Radiation Protection Series Publication G-1, Canberra.

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.

Cresswell, Tom, Brown, Sue, Wong, Henri, Apte, Simon, 2021. Assessing the impacts of scale residues from offshore oil and gas decommissioning on marine organisms. The APPEA Journal 61 (2), 379–383.

DOE, 2019. A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota. U. D. o. Energy, Washington D.C.

Garnier-Laplace, Jacqueline, Copplestone, David, Gilbin, Rodolphe, Alonzo, Frederic, Ciffroy, Philippe, Gilek, Michael, Agüero, A., Björk, Mikael, Oughton, D.H., Jaworska, Alicja, 2008. Issues and practices in the use of effects data from FREDERICA in the ERICA Integrated Approach. J. Environ. Radioact. 99 (9), 1474–1483.

Goulet, Richard R., Newsome, Laura, Vandenhove, Hildegarde, Keum, Dong-Kwon, Horyna, Jan, Kamboj, Sunita, Brown, Justin, Johansen, Mathew P., Twining, John, Wood, Michael D., Černe, Marko, Beaugelin-Seiller, Karine, Beresford, Nicholas A., 2022. Best practices for predictions of radionuclide activity concentrations and total absorbed dose rates to freshwater organisms exposed to uranium mining/milling. J. Environ. Radioact. 244–245, 106826.

Grung, Merete, Ruus, Anders, Holth, Tor Fredrik, Sidhu, Rajdeep Singh, Eriksen, Dag Øistein, Hylland, Ketil, 2009. Bioaccumulation and lack of oxidative stress response in the ragworm H. diversicolor following exposure to 226Ra in sediment. J. Environ. Radioact. 100 (5), 429–434.

Hirth, Gillian A., Johansen, Mathew P., Carpenter, Julia G., Bollhöfer, Andreas, Beresford, Nicholas, A., 2017. Whole-organism concentration ratios in wildlife inhabiting Australian uranium mining environments. J. Environ. Radioact. 178–179, 385–393.

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 (9), 1408–1429.

Hosseini, Ali, Brown, Justin E., Gwynn, Justin P., Dowdall, Mark, 2012. Review of research on impacts to biota of discharges of naturally occurring radionuclides in produced water to the marine environment. Sci. Total Environ. 438, 325–333.

IAEA, 2004. Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna.

IAEA, 2021. Radiation Protection of Wildlife: Modelling the Exposure and Effects. INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna.

ICRP, 2014. Protection of the environment under different exposure situations. ICRP publication 124. Ann. ICRP 43 (1), 1–62.

ICRP, 2017. Dose coefficients for nonhuman biota environmentally exposed to radiation. Ann. ICRP 46, 1-136.

Jensen, L.ouise Kiel, Halvorsen, Elizabeth, Song, Yu, Hallanger, Ingeborg, Hansen, Elizabeth Lindbo, Brooks, Steven J., Hansen, Bjørn Henrik, Tollefsen, Knut Erik, 2016. Individual and molecular level effects of produced water contaminants on nauplii and adult females of *Calanus finmarchicus*. J. Toxicol. Environ. Health, Part A 79 (13–15), 585–601. https://doi.org/10.1080/15287394.2016.1171988.

Johansen, M.P., Barnett, C.L., Beresford, N.A., Brown, J.E., Černe, M., Howard, B.J., Kamboj, S., Keum, D.K., Smodiš, B., Twining, J.R., Vandenhove, H., Vives i Batlle, J., Wood, M.D., Yu, C., 2012. Assessing doses to terrestrial wildlife at a radioactive waste disposal site: inter-comparison of modelling approaches. Sci. Total Environ. 427–428, 238–246.

Koppel, Darren J., Kho, Fenny, Hastings, Astley, Crouch, Dean, MacIntosh, Amy, Cresswell, Tom, Higgins, Stuart, 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.

Kusakabe, Masashi, Takata, Hyoe, 2020. Temporal trends of 137Cs concentration in seawaters and bottom sediments in coastal waters around Japan: implications for the Kd concept in the dynamic marine environment. J. Radioanal. Nucl. Chem. 323 (1), 567–580.

Larsson, C.M., 2004. The FASSET framework for assessment of environmental impact of ionising radiation in European ecosystems–an overview. J. Radiol. Protect.: Off. J. Soc. Radiol. Protect. 24 (4A), Ai–A12. https://doi.org/10.1088/0952-4746/24/4a/001.

Lepland, Aivo, Sæther, Ola, Thorsnes, Terje, 2000. Accumulation of barium in recent Skagerrak sediments: sources and distribution controls. Mar. Geol. 163 (1), 13–26.

Li, J., Liu, S., Zhang, Y., Chen, L., Yan, Y., Cheng, W., Lou, H., Zhang, Y., 2015. Pre-assessment of dose rates of (134)Cs, (137)Cs, and (60)Co for marine biota from discharge of Haiyang Nuclear Power Plant, China. J. Environ. Radioact. 147, 8–13.

Li, Xinpeng, Sun, Sida, Hu, Xiaofeng, Huang, Hong, Li, Hong, Morino, Yu, Wang, Shuntan, Yang, Xingtuan, Shi, Jiasong, Fang, Sheng, 2019. Source inversion of both long- and short-lived radionuclide releases from the Fukushima Daiichi nuclear accident using on-site gamma dose rates. J. Hazard Mater. 379, 120770.

Lu, Yonglong, Yuan, Jingjing, Du, Di, Sun, Bin, Yi, Xiaojie, 2021. Monitoring long-term ecological impacts from release of Fukushima radiation water into ocean. Geography and Sustainability 2 (2), 95–98.

MacIntosh, Amy, Dafforn, Katherine, Penrose, Beth, Chariton, Anthony, Cresswell, Tom, 2021. Ecotoxicological effects of decommissioning offshore petroleum infrastructure: a systematic review. Crit. Rev. Environ. Sci. Technol. 1–39.

Matyskin, A.V. (2016). On the solubility of radium sulfate and carbonate. [Doctoral thesis, Chalmers University of Technology]. Chalmers Publication Library. https://publications.lib.chalmers.se/records/fulltext/233368/233368.pdf.

McLean, D.L., Taylor, M.D., Ospina, A. Giraldo, Partridge, J.C., 2019. An assessment of fish and marine growth associated with an oil and gas platform jacket using an augmented remotely operated vehicle. Continent. Shelf Res. 179, 66–84.

McLean, Dianne L., Parsons, Miles J.G., Gates, Andrew R., Benfield, Mark C., Bond, Todd, Booth, David J., Bunce, Michael, Fowler, Ashley M., Harvey, Euan S., Macreadie, Peter I., Pattiaratchi, Charitha B., Rouse, Sally, Partridge, Julian C., Thomson, Paul G., Todd, Victoria L.G., Jones, Daniel O.B. 2020. Enhancing the scientific value of industry remotely operated vehicles (ROVs) in our oceans. Front. Mar. Sci. 7.

McLean, Dianne L., Ferreira, Luciana C., Benthuysen, Jessica A., Miller, Karen J., Marie-Lise, Schläppy, Ajemian, Matthew J., Berry, Oliver, Birchenough, Silvana N.R.,

Bond, Todd, Boschetti, Fabio, Bull, Ann S., Claisse, Jeremy T., Condie, Scott A., Consoli, Pierpaolo, Coolen, Joop W.P., Elliott, Michael, Fortune, Irene S., Fowler, Ashley M., Gillanders, Bronwyn M., Harrison, Hugo B., Hart, Kristen M., Henry, Lea-Anne, Hewitt, Chad L., Hicks, Natalie, Hock, Karlo, Hyder, Kieran, Love, Milton, Macreadie, Peter I., Miller, Robert J., Montevecchi, William A., Nishimoto, Mary M., Page, Henry M., Paterson, David M., Pattiaratchi, Charitha B., Pecl, Gretta T., Porter, Joanne S., Reeves, David B., Riginos, Cynthia, Rouse, Sally, Russell, Debbie J.F., Sherman, Craig D.H., Teilmann, Jonas, Todd, Victoria L.G., Treml, Eric A., Williamson, David H., Thums, Michele, 2022. Influence of offshore oil and gas structures on seascape ecological connectivity. Glob. Change Biol. 28, 3515–3536.

Melbourne-Thomas, Jess, Hayes, Keith R., Hobday, Alistair J., Little, Richard, Strzelecki, Joanna, Thomson, Damian, van Putten, Ingrid, Elizabeth, Hook, Sharon E., 2021. Decommissioning research needs for offshore oil and gas infrastructure in Australia. Front. Mar. Sci. 8, 1007.

Nedveckaite, T., Filistovic, V., Marciulioniene, D., Prokoptchuk, N., Plukiene, R., Gudelis, A., Remeikis, V., Yankovich, T., Beresford, N.A., 2011. Background and anthropogenic radionuclide derived dose rates to freshwater ecosystem: nuclear power plant cooling pond: reference organisms. J. Environ. Radioact. 102 (8), 788–795.

Olsvik, P.A., Nordtug, T., Altin, D., Lie, K.K., Overrein, I., Hansen, B.H., 2010. Transcriptional effects on glutathione S-transferases in first feeding Atlantic cod (Gadus morhua) larvae exposed to crude oil. Chemosphere 79 (9), 905–913.

Oy, Posiva, Helsinki, 2014. Safety Case for the Disposal of Spent Nuclear Fuel at Olkiluoto Data Basis for the Biosphere Assessment BSA-2012 Part 1-2, Appendices, p. 1448. Finland.

Periañez, 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.

Robinson, C.A., Smith, K.L., Norris, S., 2010. Impacts on non-human biota from a generic geological disposal facility for radioactive waste: some key assessment issues. J. Radiol. Prot. 30 (2), 161–173.

Strand, P., Aono, T., Brown, J.E., Garnier-Laplace, J., Hosseini, A., Sazykina, T., Steenhuisen, F., Vives i Batlle, J., 2014. Assessment of fukushima-derived radiation doses and effects on wildlife in Japan. Environ. Sci. Technol. Lett. 1 (3), 198–203.

Vandenhove, H., Sweeck, L., Vives i Batlle, J., Wannijn, J., Van Hees, M., Camps, J., Olyslaegers, G., Miliche, C., Lance, B., 2013. Predicting the environmental risks of radioactive discharges from Belgian nuclear power plants. J. Environ. Radioact. 126, 61–76.