



A new approach for assessing the radioecological risk associated with the legacy discharge of oil derived natural radioactivity in the UK North Sea

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ABSTRACT

Although oil and gas (O&G) derived produced waters and drill cuttings are known to contain enhanced levels of naturally occurring radium-228 (²²⁸Ra) and radium-226 (²²⁶Ra), most relevant ecological impact assessments have excluded radiological hazards and focus on other important contaminants, such as hydrocarbons and metals. Also, due to restricted access to the delimiting safety zone around operational O&G platforms, the few previous radioecological risk assessment studies have been conducted using seawater samples collected far from the main discharge point and applying default dilution and transfer factors to estimate concentrations of contaminants in biota. In this case study, sediment cores were collected close to a former O&G platform, Northwest Hutton (NWH), that used to be in the UK North Sea (61.11N, 1.31E). The sediment materials were analysed by gamma spectrometry and ICP-MS to confirm the presence of particles enriched in natural radioactivity. Benthic macrofaunal assemblages in the surrounding seabed were also characterised and one of the dominant species was selected for additional nano-hard X-Ray Fluorescence (nano-XRF) imaging to confirm the exposure pathways and refine the radioecological risk assessment using the ERICA tool. This novel approach for estimating dose rates was found to be less conservative than more traditional approaches using the ERICA default concentration ratio for ²²⁸Ra and ²²⁶Ra. The dose rate estimations were confirmed to be significantly lower than the ERICA screening level of 10µGy/h, in agreement with findings from previous studies.

1. Introduction

Offshore oil and gas (O&G) platforms installation, drilling, cementing and production operations can cause physical damage to the seabed and its associated benthic habitats (Wolfson et al., 1979; Montagna and Harper, 1996; Gray et al., 1999; Currie and Isaacs, 2005; Terlizzi et al., 2008; Manoukian et al., 2010; Punzo et al., 2015; Chen et al., 2024). The installation of O&G structures in the North Sea may also provide a habitat for a wide range of organisms that would not be found in the soft sediment communities, often producing an ‘artificial reef’ with increased local biodiversity (Fortune et al., 2024). The decommissioning of offshore facilities at the end of their operational life presents its own set of challenges. The removal or abandonment of structures can have ecological consequences, requiring cautious considerations and

planning (Murray et al., 2018; MacIntosh et al., 2022).

The release of chemicals during O&G exploration, production or following accidental oil spills can also have harmful effects on marine ecosystems. Spills and discharges of oil, drilling fluids, cutting, and chemical additives used in the extraction process can contaminate sediments, affecting benthic life and disrupting food chains (Sharma et al., 2024). Extraction of hydrocarbons is accompanied by the production of wastewater, known as produced water (PW). The chemical composition of PW can vary widely depending on the formation geology, operating conditions, practices, and age of the extraction zone (Neff et al., 2011). Produced water typically contains enhanced levels of organic and inorganic chemicals such as phenols, salts, polycyclic aromatic hydrocarbons (PAHs), heavy and alkaline earth metals, in particular lead, barium, strontium, and less notably, their respective naturally occurring

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radioelemental analogues, radium-228 (^{228}Ra), radium-226 (^{226}Ra) and lead-210 (^{210}Pb). Many environmental impact assessment studies in relation to the discharges from offshore O&G activities have been conducted, however, there are still large gaps in our knowledge of the radioecological components surrounding these sites (MacIntosh et al., 2021).

Operational discharges of PW constitute one of the largest inputs of industrially derived contaminants, including naturally occurring radioelements in many marine regions of the world where oil extraction occurs. In the region covered by the Oslo-Paris conventions (OSPAR), monitoring programmes are in place mainly to manage PW discharges from these man-made installations (Beyer et al., 2020). As soon as PW is discharged into the marine environment, it is monitored regularly to identify the main chemicals present and verify it complies with local regulations.

In 2001, OSPAR issued a recommendation for the management of PW from offshore installations (OSPAR, 2001). A separate recommendation was published in 2012 to ensure that the chemical risk posed from ongoing O&G production activity was being adequately assessed and that measures were in place to identify operations that had the potential to cause toxic harm to the marine environment (OSPAR, 2012). While the Best Available Technique (BAT) is to reinject the PW into an existing well, its application is not always possible. Instead, many offshore installations in operation treat PW to reach regulatory levels and subsequently discharge treated water directly to the sea.

Risk-based approaches and modelling tools were subsequently developed for identifying chemical inputs to the marine environment. The Dose-related Risk and Effects Assessment Model (DREAM) is commonly used in Europe (Reed and Rye, 2011). The DREAM model uses inputs from chemical analysis or whole effluent toxicity testing to establish the volume of water from which environmental thresholds could be exceeded. Many stable naturally occurring substances (NOS) are identified and accounted for in the DREAM model, but naturally occurring radioactive elements or materials (NORM) are not (OSPAR, 2014). The DREAM model has been used in previous studies to assess the risk of NORM discharges (Rye et al., 2009). The study published by Eriksen et al. (2009) indicated low dose levels.

Over the last two decades, discharges of industrially derived naturally occurring radioactive materials, also known as NORM, in the European Seas has received substantial attention owing to the OSPAR Radioactive Substances Strategy and more recently under strategic objective three of the 2030 North-East Atlantic Environment Strategy (NEAES), which aims to achieve “concentrations in the environment

near background values for naturally occurring radioactive substances” (OSPAR, 2021). As seen in Fig. 1, 0.59 TBq of ^{228}Ra , 0.70 TBq of ^{226}Ra , and 0.04 TBq of ^{210}Pb present in approximately 300 million cubic meters were discharged into the OSPAR regions in 2021, with the UK and Norwegian O&G industries identified as the two biggest contributors (OSPAR, 2023).

The naturally occurring radioelements identified in O&G waste streams belong to the decay chains of uranium-238 (^{238}U) and thorium-232 (^{232}Th) (Fig. 2). The activity concentrations of some decay products of these two naturally occurring radioelements are known to concentrate in the waste streams during extraction and production of hydrocarbons (Heaton and Lambley, 1995; Jonkers et al., 1997; IAEA, 2003). In more detail, when a hydrocarbon-containing geological formation is open to radioelement migration, changes in chemical and physical conditions can result in mobile radioelements being transferred from the hydrocarbon reservoir to PW, flowback fluid and production equipment. These radioelements are primarily radium, and to a lesser extent, lead, and polonium. Owing to its insolubility, thorium is almost wholly transported in gas suspension and therefore remains in the underground reservoir when oil is brought to the surface (Jonkers et al., 1997). Also, as the redox potential around a hydrocarbon reservoir is usually strongly reducing, tetravalent uranium also remains effectively immobile (Collins, 1975; Bloch and Key, 1981). Initially, when oil is conventionally extracted from the underground reservoir, the amount of PW discharged is low. This water, coupled with a drop in pressure and temperature at the seabed surface, leads to the precipitation of insoluble barite alongside the hydrocarbons, which are brought to the surface and result in the formation of radium enriched scales (Smith, 1987). These scales are classed as Naturally Occurring Radioactive Materials (NORM) and could present a substantial occupational health risk for the operators of these industrial platforms if not managed appropriately (SEPA, 2014; Al Nabhani et al., 2016). If released directly in the marine environment, these scales can cause local ecological harm (Koppel et al., 2022). The main types of NORM waste generated during conventional oil extraction and production are presented (Table 1) (IAEA, 2003).

Although the transport mechanisms of radium during oil extraction and production are well understood (Fig. 2), knowledge of its long-term fate once deposited onto the seabed and associated toxicokinetic mechanisms in benthic organisms remains incomplete. Radium, alongside its key decay products, ^{210}Pb and ^{210}Po , and its main chemical analogues, barium, and strontium, have been found at enhanced concentrations in PW (Al-Masri, 2006). While the typical natural ^{226}Ra background level in seawater from the North Sea is in the mBq/l range,

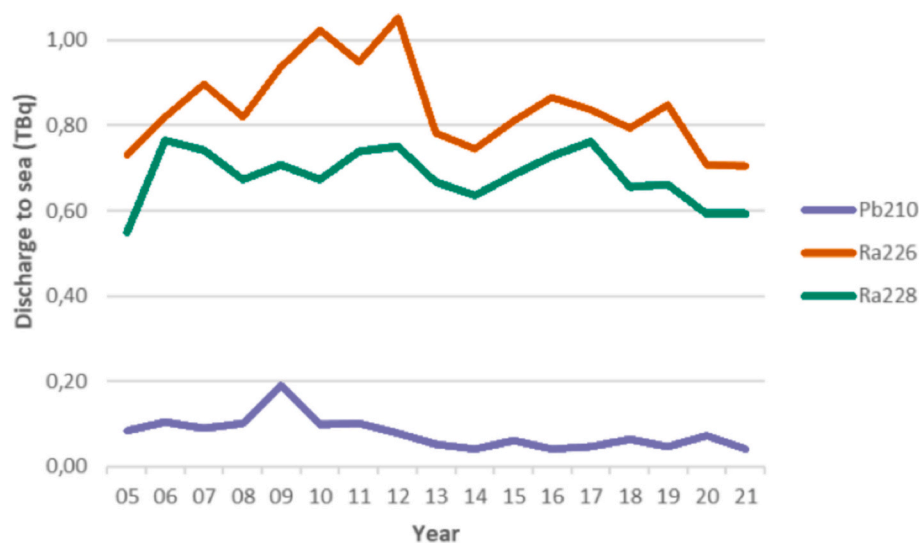


Fig. 1. Total discharges of ^{210}Pb , ^{226}Ra and ^{228}Ra present in PW from the O&G sector in the OSPAR regions between 2005 and 2021 (OSPAR, 2023).



Fig. 2. Transport of (a) ²³⁸U and (b) ²³²Th and their key decay products during O&G production (adapted from Jonkers et al., 1997; IAEA, 2003). Note: Short lived radioelements (i.e. $t_{1/2} < 5$ min) are not represented in this illustration.

Table 1
Main forms of Naturally Occurring Radioactive Material (NORM) generated during conventional oil extraction operations.

Form	Radioelement(s)	Characteristics	Occurrence
Produced water	²²⁶ Ra, ²²⁸ Ra and/or ²¹⁰ Pb	Large volumes in oil production, usually saline	All production facilities
Radium scales	²²⁶ Ra, ²²⁸ Ra and decay products	Hard deposits of calcium, strontium and barium sulphates and carbonates	Wet parts of production installations, well completions
Radium sludge	²²⁶ Ra, ²²⁸ Ra and decay products	Sand, clay, paraffins, heavy metals	Separators, skimmer tanks
Lead deposits	²¹⁰ Pb and decay products	Stable lead deposits, PbS, PbSO ₄ and metallic lead	Wet parts of production installations, well completions, dry gas pipelines

levels of up to several hundred Bq/l in PW have been reported in the literature (Hosseini et al., 2012). A study conducted by Ahmad et al. in 2021 confirmed the presence of industrially derived radiostrontio-barite (Ba(Sr,Ra)SO₄) particles in sediment materials collected in the direct vicinity of a PW discharge site (Ahmad et al., 2021). Enhanced levels of natural radioactivity were also found in drill cuttings (Breuer et al., 2004). Increased levels of barium were also recently measured in marine sediments collected from different locations of the Norwegian and Barents Seas, known to be directly impacted by O&G extraction and drilling activities, respectively (Haanes et al., 2023). Radium-226 sourced from legacy NORM waste disposal activities, including recently from the O&G industry has been shown to accumulate in bivalve soft tissue (Donaher et al., 2023; Pankratz and Warner, 2024).

The ERICA (Environmental Risk from Ionising Contaminants: Assessment and management) tool has been used to assess the

radioecological impact in areas directly affected by major nuclear accidents as well as in relation to the development of new nuclear builds (Brown et al., 2008; Brown et al., 2016; Penfold et al., 2016). For example, model validation work was carried out within the Chernobyl exclusion zone, with agreement found between estimated and predicted doses (Beresford et al., 2008). It was also used to compare the estimated damage to marine biota following the Fukushima Daiichi Nuclear Power Plant (FNPP) disaster in 2011, finding the most exposed taxonomic group to be polychaete worms with an estimated dose of 2 µGy/h, approximately 20 % of the recommended screening value of 10 µGy/h and requiring further assessment (Aliyu et al., 2014). The ERICA tool was also used to further understand the effects of NORM arising from many non-nuclear industrial activities. Nevertheless, only a few studies were specifically related to the direct effect of O&G PW. Beresford et al. (2011) used a dispersion model and the ERICA tool to assess the radioecological risk associated with the releases of O&G PW within three Scottish Natura 2000 sites, by carrying out a conservative screening approach (i.e., Tier 1 ERICA screening) (Beresford et al., 2011). Marine organisms, such as sea anemones, corals and polychaete worms were found to be the most affected, but the level of risk was deemed negligible (Beresford et al., 2011). Other radioecological studies focused on different exposure scenarios and other O&G waste streams. A study conducted by MacIntosh et al. (2022) concluded that polychaetes and corals could receive an estimated dose of 860 and 730 µGy/h in direct contact with barite scale inside pipe structures, respectively. In addition to being well above the ERICA screening value of 10 µGy/h, these dose rates were also found to be above 400 µGy/h, the lower dose band limit for reference crabs defined by the International Commission on Radiological Protection (ICRP), suggesting potential impacts to local benthic vertebrates, invertebrates, seaweeds and phytoplankton (ICRP, 2014; MacIntosh et al., 2022). These same NORM-contaminated scales were also found to leach both ²²⁶Ra and ²¹⁰Po into surrounding sediments, leading to uptake by amphipod species *Victoripisa australiensis*

(MacIntosh et al., 2023). Subsequent radiological dose from this uptake was estimated to be above the ERICA screening value of 10 $\mu\text{Gy/h}$, ranging between 66 and 515 $\mu\text{Gy/h}$.

In 2021, as part of a study aiming to understand better legacy impacts of decommissioning, including naturally occurring organic carbon dynamics, around offshore O&G structures, sediment and benthic biota samples were collected as close as 50 m away from a recently decommissioned platform located in the UK North Sea. This constituted a unique opportunity to refine knowledge of the spatial and radiological impact of legacy discharges of PW and drill cuttings in the local environment, focusing on benthic macrofaunal communities (>500 μm) abundantly present in this marine region (Basford et al., 1990; Kunitzer et al., 1992; Fortune et al., 2024).

The main aims of the presented work were 1: to test a novel approach for estimating radium activity concentrations in biota using a correlation between radium and barium in subsurface sediments and 2: use the resulting estimated concentrations to carry out a dose assessment for the most representative local benthic macrofaunal community using the latest version of the ERICA tool (Brown et al., 2016). We hypothesise that this method is less conservative than traditional modelling approaches using generic and empirically derived concentration ratios (CR) and will help generate more accurate dose rate estimates for biota.

2. Material and methods

2.1. Selected sampling site and sample preparation

Sediment materials near the former location of the North West Hutton (NWH) platform (61.107565N, 1.30915E, in the UK Northern North Sea) were sampled in June 2021, during a research survey on the MRV *Scotia*. The NWH platform was commissioned in 1981 and produced oil between 1983 and 2003 (Blacklaws and Johnston, 2013). The topsides of the platform were removed in 2008, and removal of the jacket structure was completed in 2009. The seawater depth around the platform was over 140 m, with the main currents being in a NE/SW direction (BP, 2005). Sediment cores (20 cm deep maximum) and grabs were taken at different distances along northern and southern transects away from the decommissioned platform (50, 100, 200, 400, 800 and 1600 m). Control sites of similar sediment composition were selected at 3200 m away along each of the gradients. Sediment cores were sampled at each distance from the platform using a multi-corer of 9.8 cm inner diameter, sliced every centimetre and freeze dried prior to elemental and radiometric analysis. Sediment grabs were also collected alongside cores and were used to extract biota species through on-board sieving, that were then preserved in formaldehyde prior to taxonomic identification.

2.2. Taxonomic identification and functional ecological traits

Benthic biota were identified where possible to species level according to the species directory of the marine fauna and flora of the British Isles and surrounding seas (Marine Conservation Society, 1997, WoRMS and NMBAQC Guidelines). Macrofaunal diversity analysis was also undertaken using Plymouth Routines In Multivariate Ecological Research (PRIMER version 6). The most representative taxa were selected for further analytical investigations (Sections 2.3 and 2.4). The functional ecological traits of selected biota (Table 2), particularly their position in the sediment substrates (i.e. epibenthic or infaunal) and feeding mode, were then used to define default occupancy factors (OFs) in the estimation of external dose rates using the ERICA tool (see Section 2.5).

2.3. Trace elemental and radiometric analysis

Following freeze-drying, approximately 200 mg aliquots of each sediment core slice collected at different distances from the NWH

Table 2

Most abundant benthic organisms identified to the species level and present in sediment materials collected around the former location of the NWH platform. Species identified according to World Register of Marine Species (WoRMS, 2024) and NE Atlantic Marine Biological Analytical Quality Control Scheme (NMBAQC, 2024).

Taxa	Class	Sediment Position	Feeding Mode	Occupancy Factors (Sediment: Sediment surface)
<i>Nephtys hombergii</i>	Polychaete	Infaunal	Predator & scavenger	1:0
<i>Chaetozone setosa</i>	Polychaete	Infaunal	Sedentary surface deposit / detritus feeder, sub-surface deposit feeder	1:0
<i>Cirratulus cirratus</i>	Polychaete	Infaunal	Surface and sub-surface deposit feeder	0.5:0.5
<i>Notomastus latericeus</i>	Polychaete	Epibenthic/infaunal	Surface and sub-surface deposit feeder	0.5:0.5
<i>Thyasira sarsii</i>	Bivalve	Infaunal	Sulphur oxidising symbiotic bacteria in gills	1:0
<i>Abra nitida/alba</i>	Bivalve	Epibenthic/infaunal	Passive /active suspension feeder, surface and subsurface deposit feeder	0.25:0.75

platform were ground, sieved down to 63 μm and acid digested using 15 mL of a mixture of ultra-pure $\text{HNO}_3/\text{HCl}/\text{HF}$ (8/4/3) in a microwave system for 60 min. The resulting acid digests were then further diluted prior to ICP-MS analysis. Quantification of trace elements, including barium (Ba) and strontium (Sr), as chemical analogues of radium, was performed by external calibration using standard solutions ranging between 0 and 500 $\mu\text{g/L}$, and using indium as internal standard. With the absence of a reference sediment material presenting a certified value for Ba, the expanded 2-sigma uncertainty was estimated to be 30 % based on actual calculated expanded uncertainties observed for other certified elements.

After air drying for 10 min, selected specimens of molluscs and polychaete worms initially extracted from sediment grab samples collected 50 and 3200 m N of the NWH platform were weighed and fully pre-digested using 10 mL of a 9 M HNO_3 solution for 2 h. Only the shells of mollusc samples were first removed before weighing using plastic tweezers. A blank solution and certified reference materials (approx. 500 mg of Dorm 5: Dogfish protein, Tort 3: Lobster hepatopancreas and Dolt-5: Dogfish liver; National Research Council Canada) were also prepared and run alongside the sample digests. After pre-digestion, the microwave vessels were sealed and digested for 30 min at 200 °C using a different microwave system (Anton Paar Multiwave 6000, Austria). Sample digest solutions were then topped up to 40 mL using deionised water, diluted further 5-fold using a gold diluent solution (5 mg/L Au, 5 % HNO_3 and 0.5 % HCl) and analysed by ICP-MS.

Approximately 27 g of each sediment slice was mechanically compressed in a plastic container that was then sealed and kept for at least for 21 days prior to for high resolution gamma spectrometry analysis (High Purity Germanium detector, low background carbon fibre endcap window, 100 mm Pb shield enclosure plus Cu/Cd linings) to determine the activity concentration levels of ^{210}Pb , ^{226}Ra (via lead-214 (^{214}Pb)), ^{228}Ra (via actinium-228 (^{228}Ac)), at 46.5, 352 and 911 keV, respectively (Joshi, 1987). Each sample was counted for a minimum of 24 h to minimise the limit of detection and counting uncertainty as low as practically possible. The activity concentrations of ^{228}Ra were decay

corrected to account for the period between sampling and analysis time ($t_{1/2}$: 5.75 years). A mixed radioelement standard solution in 4 M HCl (National Physical Laboratory, UK) and the certified reference material (CRM) IAEA-465 (Baltic Sea sediment), presented in the same geometry as other sediment materials, were used to perform the energy and efficiency calibrations of each BeGe detector utilised in this study, respectively. Radium-226 and ^{228}Ra measurement uncertainties were estimated from gamma counting only (2 sigma).

2.4. Nano-hard X-ray fluorescence analysis

Nanoprobe XRF mapping at beamline I14, Diamond Light Source, UK was used to provide evidence of the presence of co-localised Ba and Sr particles in selected benthic nematode individuals collected 50 m away from the NWH platform (Quinn et al., 2021). A full nematode sample was first infiltrated, embedded in an epoxy resin and mounted onto a 5×5 mm silicon nitride window (Fig. 3). Additional $100 \mu\text{m}$ longitudinal histological slices of other nematode samples collected from the same area were also prepared to investigate the presence of these industrially derived Ba/Sr particles inside the selected organisms. Monochromatic X-rays of 18 keV energy were focused to different regions of the nematode, that were continuously scanned through the X-ray focus using step sizes of 2 mm (for coarse overview maps) or 100 nm (for high resolution maps) and an XRF spectra collected at each pixel position. Maps of Ca, Ba and Sr distributions were generated by windowing and summing total counts of the Ca-K α , Ba-L α and Sr-K α fluorescence peaks in the XRF spectra at each position.

2.5. ERICA dose estimation

Dose rate received by the selected organisms was estimated using the ERICA tool (Brown et al., 2016). Due to the customisable nature of the tool, it is possible to use a mixture of inbuilt default, and user input parameters. The initial and highly conservative Tier 1 assessment was carried out by only using environmental activities. Then, a more specific Tier 2 assessment was conducted examining each specimen selected in this study. Situations requiring a Tier 3 assessment are rare and was not required for this study. Two methods were used to allow for comparison of estimated dose rates from activity concentrations calculated using the Ba:Ra ratio method (Method A), versus estimated dose rates using default ERICA CR values (Method B).

For method A, the measured sediment activity concentration values were used here as reference media, alongside the estimated biota activity concentrations. In more detail, correlations between anthropogenic Ba and $^{226}\text{Ra}/^{228}\text{Ra}$ observed in the top 5 cm slices of sediment cores collected at 50, 100 and 200 m North (N) of the NWH platform

enabled the estimation of ^{226}Ra and ^{228}Ra in selected biota, using Ba as a proxy, and assuming a similar $^{138}\text{Ba}:^{226}\text{Ra}$ ratio in biota. The natural levels of Ba, ^{226}Ra and ^{228}Ra in sediment were estimated using averaged concentrations observed in the top 5 cm fraction of the core collected at 3200 m N of the NWH platform (control) and were subtracted from the total measured concentrations (individual data are presented in Appendix 1).

The ERICA tool includes default radiological and environmental parameters, such as dose conversion coefficients (DCCs) as well as a comprehensive list of reference organisms and associated biological parameters, such as CR and occupational factors (OFs) (Fig. 4). Reference organism selection was guided by the ICRP, designed to cover EU protected species over a broad range of habitats (ICRP, 2008). For the marine environment, the ERICA list of reference organisms contains both bivalve molluscs and polychaetes, therefore these were selected for the corresponding selected species, providing both appropriate geometries, which specify the size and shape of an organism, and CR values,

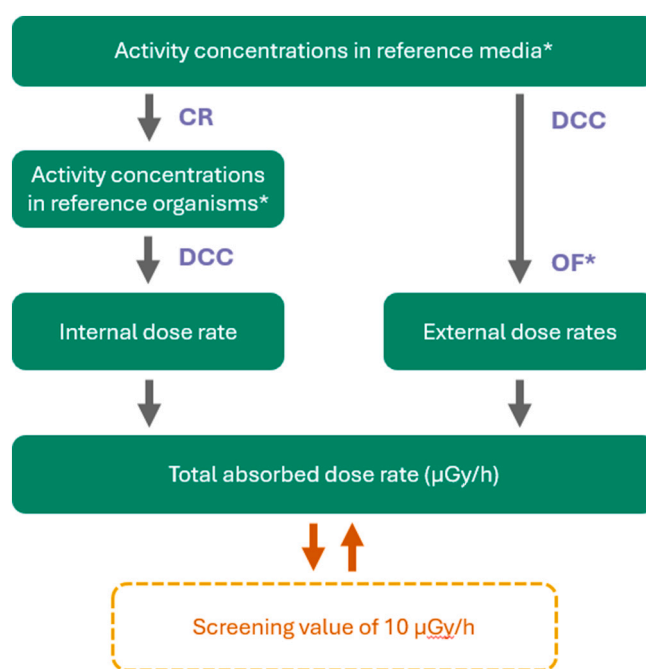


Fig. 4. ERICA compartmental model (adapted from erica-tool.com/about/)
Note: Asterisk indicates where study specific calculated or estimated values were used.

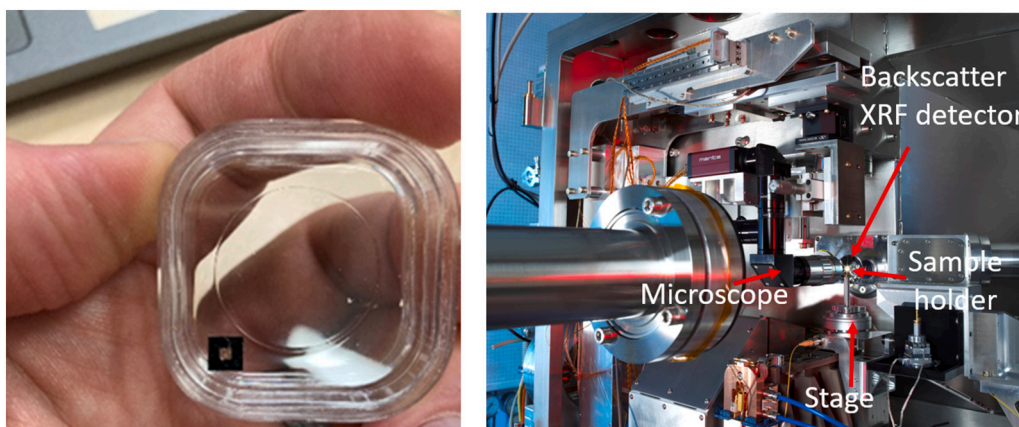


Fig. 3. Left: Photo of a target 5×5 mm silicon nitride window where the nematode sample was embedded in an epoxy resin. Right: Sample introduction of the I14 end station for XRF analysis.

which quantify the uptake of a radioelement from the organism's environmental media. For method B, the default CR for the selected organisms was used to estimate activity concentration using the measured sediment activity. Where the appropriate reference organism was not available for certain radioelements, the ERICA tool suggests the use of a similar group. Occupancy factors (OFs), used to calculate external dose, were also adjusted to account for the more infaunal characteristics of selected organisms in comparison with default reference organisms (Table 2). The model then calculates internal and external DCCs, and subsequent associated total absorbed dose rates.

The ERICA tool is integrated with the FREDERICA effects database, a collation of data on the effects of ionising radiation at specific doses (Copplestone et al., 2008). To assess the risk associated with radioactivity exposure, ERICA uses a screening value of 10 $\mu\text{Gy/h}$, derived from evidence within the FREDERICA database. A received dose below this value indicates, no risk and the assessment can be concluded. Other benchmark screening values are organism dependent. Estimated dose rate was compared against these screening values and effects to gauge risk and determine any potential effects received by the sampled organism.

3. Results and discussion

Six dominant benthic species were identified as polychaetes *Nephtys hombergii*, *Chaetozone setosa*, *Cirratulus cirratus* and *Notomastus latericeus*, and bivalves' molluscs *Thyasira sarsii* and *Abra* spp. (see Table 2). All six species occupy a majority infaunal position in the sediment and expected in the area surrounding the platform. In particular, *Cirratulus cirratus* is known to be found in moderate densities near disturbed areas such as O&G platforms as it is tolerant of the conditions they create (Levell et al., 1989). These species are also representative of the local assemblage, found to be 51.2 % Annelida and 21.1 % Mollusca ($n = 1041$). There was some variation in feeding modes. Of the polychaetes, three are deposit or detritus feeders, ingesting the sediment directly. The exception to this is *Nephtys hombergii*, which is carnivorous and feeds through predation and scavenging. An OF of 1:0 was assigned to species with entire life histories spent within the sediment. When considering *Notomastus latericeus* and *Cirratulus cirratus*, both are surface deposit

feeders, feeding on the sediment surface, therefore these taxa were assigned an OF of 0.5:0.5. *Abra* sp. is both an active suspension feeder, and surface deposit feeder. It remains situated within the sediment, however, uses a siphon to filter particles from the water a few millimetres above the surface when suspension feeding, and to suck up detritus from the surface when deposit feeding (Amouroux et al., 1989). Therefore, an OF of 0.25:0.75 was assigned.

Trends were observed between anthropogenic Ba and $^{226}\text{Ra}/^{228}\text{Ra}$ measured in the top 5 cm sediment materials collected within 200 m N of the former NWH platform, with the highest levels obtained at 50 and 100 m (see Fig. 5). These elemental signatures could not be distinguished from the natural background at 400, 800 and 3200 m. Ratios of 2850 and 2155 were then extracted from correlating linearly anthropogenically derived Ba (expressed in mg/kg) with NORM derived ^{228}Ra and ^{226}Ra (expressed in Bq/kg), respectively, and assuming both Ba and Ra were coming from the same source (i.e., no distinction between drill cuttings and produced water). The concentrations of ^{228}Ra and ^{226}Ra in all taxa selected in this study could then be estimated using these sediment derived ratios, assuming the transfer mechanisms of both elements to be identical between sediment and biota. Both ^{226}Ra and Ba, as alkaline earth metals, are analogues of calcium (Ca), an essential element for living organisms, therefore all follow the same metabolic pathway during uptake. In plankton, there is little preference shown between uptake of Ba and ^{226}Ra (Szabo, 1967). In Bivalve molluscs, Ca, Ba and ^{226}Ra have been shown to share the same distribution within the tissues (Jeffrey and Simpson, 1984). Ba/ ^{226}Ra ratios have also been used for dating marine bivalve mollusc fossils as Ba can be used as a stable isotope for ^{226}Ra (Berkman and Ku, 1998). These estimated $^{226}\text{Ra}/^{228}\text{Ra}$ activity concentrations in biota were applied in ERICA to determine the associated external, internal and total dose rates (see Table 3).

Attempts to correlate between anthropogenic Pb at mass 206 (^{206}Pb) and NORM derived ^{210}Pb in the same sediment samples did not lead to a conclusive outcome, possibly due to the presence of significantly lower levels of NORM derived ^{210}Pb and ^{206}Pb observed in sediment materials (OSPAR, 2023). Additionally, a small fraction of ^{210}Pb present in the seabed would have been sourced from atmospheric fallout and this specific fraction of ^{210}Pb could not be distinguished from the anthropogenic source of unsupported ^{210}Pb (Dal Molin et al., 2023).

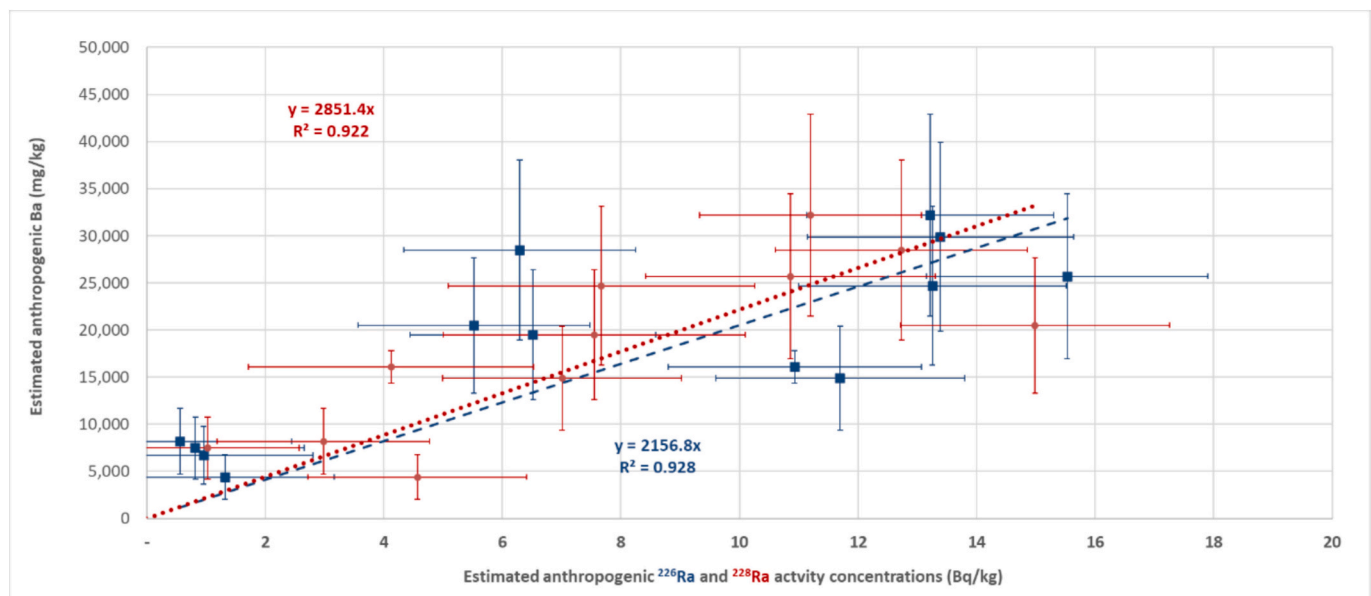


Fig. 5. Trends observed between anthropogenically derived barium and NORM-derived ^{226}Ra (in blue) / ^{228}Ra (in red) in top 5 cm sediment materials collected within 200 m N of the studied NWH platform. Error bars represent 2-sigma uncertainties from measurement. Radium-228 activity concentrations were decay corrected to account for the period between sampling and analysis. Raw data from sediment analysis are provided in Appendix 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3

Barium, ^{228}Ra and ^{226}Ra activity concentrations in selected organisms collected near and away from the site (50 and 3200 m N (control)), estimated total (Tot.) dose rates (DR) using derived average Ra concentrations from Ba/Ra ratios observed in sediment materials (method A) and Tot. DR using the default ERICA CR values (method B). Uncertainty of total dose rate estimations via method A was assumed to be 50 %.

Distance from platform	Species	Mass	Ba	Ra-226	Ra-228	Tot. DR (A)	Tot. DR (B)
		g	mg/kg	mBq/kg	mBq/kg	$\mu\text{Gy/h}$	$\mu\text{Gy/h}$
3200 m	<i>Abra alba</i>	0.092	0.46 ± 0.14	0.16	0.21	0.009–0.027	0.028
	<i>Nephtys hombergii</i>	0.425	<0.1	0.0011	0.0014	0.010–0.030	0.166
	<i>Notomastus latericeus</i>	0.012	2.89 ± 0.87	1	1.3	0.010–0.030	0.166
50 m	<i>Thyasira sarsii</i>	0.186	46.5 ± 14.0	16	22	0.017–0.052	0.068
	<i>Thyasira sarsii</i>	0.093	48.1 ± 14.4	17	22	0.017–0.052	0.068
	<i>Nephtys hombergii</i>	0.137	759 ± 228	270	350	0.044–0.134	0.329
	<i>Notomastus latericeus</i>	0.017	4440 ± 1332	1600	2100	0.163–0.490	0.322
	<i>Chaetozone setosa</i>	0.157	112 ± 34	39	52	0.023–0.069	0.329
	<i>Cirratulus cirratus</i>	0.266	664 ± 199	230	310	0.037–0.111	0.322

The Ca-K α Hard XRF map using a scan at 2 μm pixel resolution showed the outline of the selected nematode (Fig. 6A). When focusing more closely on one region of the nematode (surrounded in red), co-localisation of Sr and Ba was revealed from Sr-K α and Ba-K α mapping using a scan at 100 nm pixel resolution, respectively (Fig. 6B and C). This confirmed the presence of high concentrations of both Sr and Ba that could be related to the presence of one anthropogenically-derived or many particles of submicron size. Ultimately, the mode of preservation and the selected preparative route used prior to analysis implied these elements had been ingested and were present inside the selected nematode specimen. Although the exact nature and sources could not be fully confirmed, the internal exposure pathway was also included as part of the ERICA dose assessment, in addition to the external exposure pathway, assuming Ba and Ra had bioaccumulated in all selected biota specimens.

Barium concentrations ranged between <0.01 and 2.98 mg/kg in the selected organisms collected at the control site (3200 m N from the platform) and between 46.5 and 4440 mg/kg in the organisms collected at the exposed site (50 m N) (see Table 3). This resulted in estimated ^{226}Ra and ^{228}Ra activity concentrations between 1 and 6 orders of magnitude higher at the exposed site in comparison with those estimated in the control site. In both the control and the exposed sites,

Notomastus latericeus presented the highest Ba content and highest total dose rate, approximately 0.02 $\mu\text{Gy/h}$ at the control site and 2.1 $\mu\text{Gy/h}$ at the exposed site. Dose rates at both sampling distances were found to be well below the 10 $\mu\text{Gy/h}$ ERICA screening value and therefore well below the 400 $\mu\text{Gy/h}$ value recommended by ICRP for marine invertebrates. For both ^{228}Ra and ^{226}Ra , external dose rates were found to be significantly higher at the exposed site, with $P = 0.006$ and $P = 0.002$, respectively (unpaired t -Test). However, there was no significant difference observed between the sites in relation to the internal dose rate.

Dose rate figures estimated using derived Ra concentrations from Ba/Ra ratios observed in sediment materials (approach A) were generally found to be lower at both the control and exposed sites in comparison with those estimated using the default CR values suggested by ERICA (approach B) for each species. For the bivalve species *Abra alba* and *Thyasira sarsii*, total dose rates estimated from approach A were on average between 36 % and 49 % lower than from approach B. In the polychaete species, this difference was even more pronounced, with values estimated using approach A were on average between 73 % and 88 % lower than using approach B. The one exception to this was the exposed *Notomastus latericeus*, which exhibited the highest Ba concentration and therefore the highest estimated Ra activity and total dose rate. Estimated total dose range for this species was between 0.163 and

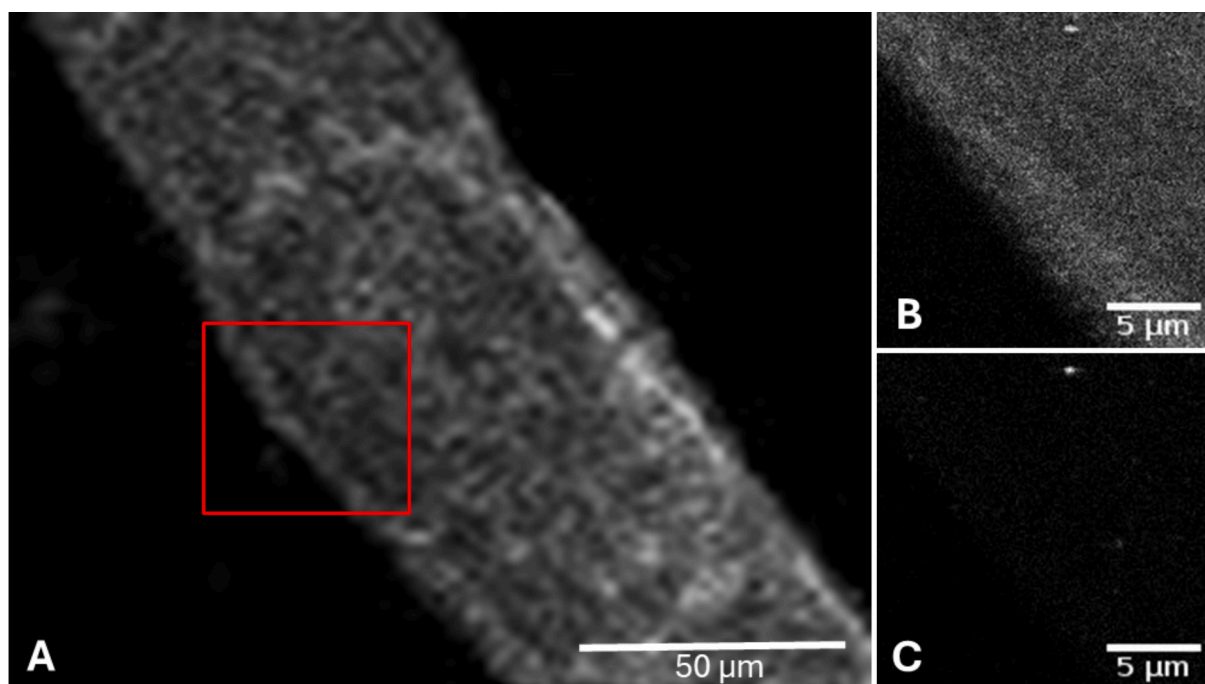


Fig. 6. A: Ca-K α XRF map at 2 μm pixel resolution showing the outline of the selected nematode. B: Sr-K α XRF map at 100 nm pixel and C: Ba-K α XRF map at 100 nm pixel.

0.490 $\mu\text{Gy/h}$, in agreement with the estimated value of 0.322, obtained from approach B.

The ability to estimate activity concentration in organisms without the use of transfer coefficient has numerous advantages. Empirically derived CR values are often quoted for large and diverse groups of organisms. Although shown to be suitable for most applications, there are sometimes exceptions where taxa specific values would be more appropriate (Hunt et al., 2024). In addition, empirically derived CR values are not available for all radionuclides for every reference organism. Although both polychaete worms and bivalve molluscs are ERICA reference organisms for marine ecosystems, there is no CR available for Ra for polychaetes. In this case, ERICA recommends the use of the crustacean CR, suggesting that this is a similar reference organism within the marine ecosystem. ERICA estimated similar dose rates for all polychaetes and all bivalves at each site as this is calculated using the same environmental parameters and CR, with the only difference between species being the OF. The comparison of both approaches suggests that the Ba derived method (A) is less conservative, particularly at lower activity concentrations, supporting this study's hypothesis.

4. Conclusions

Access within the 500 m safety zone of operating offshore O&G platforms can be very challenging and thus, previous radioecological risk assessment studies conducted around these man-made structures have been using many conservative assumptions, leading to risk or dose rate overestimations. In this study, the elemental and radioelemental signatures from the legacy discharges of PW and drill cuttings were identified in sediment materials collected within 200 m of a former offshore O&G platform located in the northern region of the UK North Sea. This was an opportunity to compare different risk assessment approaches using the ERICA tool. The results obtained in this study also implied that these elemental signatures could also be used as forensic tools to characterise and predict long-term effect on local benthic species and sedimentary processes (e.g., from ^{210}Pb based modelling).

Despite the use of several assumptions and relatively large uncertainties associated with measurements, the correlations observed between Ba and Ra analogues in subsurface sediment materials enabled the estimation of ^{228}Ra and ^{226}Ra levels in several benthic biota species present in abundance in this region of the North Sea. In addition, exposure was visually confirmed via 2D Hard-XRF mapping, showing Ba and Sr particles co-localised in a targeted screening area of a selected nematode, suggesting the presence of anthropogenically derived micro-/nano-particles onto or within a selected nematode and providing first visual evidence of exposure in this particular context. Although the exact location of these particles could not be fully confirmed due to the nature of the sample preparative route selected in this study, dose rates could still be estimated by integrating species specific OFs and assuming the presence of both external and internal exposure pathways. Higher Ba concentrations were observed in selected species collected 50 m from the platform, subsequently leading to higher but negligible dose rate estimations in comparison with those obtained from the control site. These observations also suggested the potential use of some of these representative benthic species as chemical and radiological bio monitors in this specific offshore marine region. This has also been observed in a separate study conducted in a US riverine system affected by legacy O&G PW discharge (Pankratz and Warner, 2024). The total dose rate estimation obtained from this approach was also found to be lower than the dose rate derived from the use of more generic CR values provided in the ERICA tool, presenting this newly developed approach slightly less conservative.

While the two latest major nuclear accidents at Chernobyl and

Fukushima have significantly raised concerns around the direct impact of ionising radiation on human and animal health, it also diverted public attention from the potential radiological impact of non-nuclear industrial activities in the offshore marine environment, more publicly known for discharging other hazardous substances in the aquatic environment. The growing use of our offshore marine space for sustainable energy production as well as our need to extract other natural resources will require us to assess the indirect and direct radioecological impact. (Bittner et al., 2023; Volz et al., 2023).

Sampling and analysis of environmental materials as close as possible of the main discharge point is primordial to determine less conservatively the associated radiological risk and subsequently ensure the current regulatory framework for the management of NORM industrial discharges in the offshore marine environment is fit for purpose. Nevertheless, the results observed in this study were found to be lower in most incidences than findings from previous radioecological risk assessment studies and from using default ERICA CR values (Hosseini et al., 2012), suggesting the presented approach was less conservative. It is worth noting there was not previous studies looking at benthic polychaetes and bivalves specifically. Another major knowledge gap is related to our capacity to simulate and predict the transport and deposition of NORM-derived solid and insoluble particulates in the marine environment.

Finally, the rapid growth of nuclear power production and development of large-scale heavy industrial operations worldwide, mainly located in the coastal environment of developing countries, combined with the potential effect of climate change and other persistent pollutants on radioelemental bioavailability and the urgent need to ensure a sustainable exploitation of aquatic food resources oblige us to integrate our traditional radiological risk assessment methodologies into One Health approaches, capable of assessing multiple stressors simultaneously at different ecosystemic and development levels (Stentiford et al., 2022).

CRedit authorship contribution statement

F. Dal Molin: Writing – review & editing, Writing – original draft, Resources, Project administration, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **D. Hunt:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Data curation. **A. Dewar:** Writing – review & editing. **S. Lozach:** Writing – review & editing, Methodology. **C. Phillips:** Writing – original draft. **B. Thomas:** Formal analysis. **L. Warford:** Writing – original draft, Data curation. **J.E. Parker:** Writing – original draft, Formal analysis, Data curation. **J. Walker:** Writing – review & editing, Formal analysis, Data curation. **M. Chocholek:** Formal analysis, Data curation. **D.M. Paterson:** Writing – review & editing, Data curation. **H. Woodward-Rowe:** Data curation. **N. Hicks:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix 1. Radium-226, ²²⁸Ra and Ba concentrations in top 5 cm sediment materials collected within 200m North of the former NWH platform.

Distance from NWH platform North (m)	Core depth	Total Ra-226 (Bq/kg)*	Anthropogenic Ra-226 (Bq/kg)**	Total Ra-228 (Bq/kg)*	Anthropogenic Ra-228 (Bq/kg)†	Total Ba (mg/kg)‡	Anthropogenic Ba (mg/kg)
50	0–1 cm	21.5 ± 1.3	11.7 ± 2.1	14.0 ± 1.9	7.0 ± 2.5	18300 ± 5490	14890 ± 5500
	1–2 cm	23.0 ± 1.3	13.2 ± 2.1	17.4 ± 1.8	11.2 ± 2.3	35600 ± 10680	32190 ± 10690
	3–4 cm	16.1 ± 1.0	6.3 ± 1.9	18.6 ± 2.0	12.7 ± 2.6	31900 ± 9570	28490 ± 9580
	4–5 cm	15.3 ± 1.0	5.5 ± 1.9	20.5 ± 2.2	15.0 ± 2.8	23900 ± 7170	20490 ± 7180
100	0–1 cm	20.7 ± 1.3	10.9 ± 2.1	11.6 ± 2.3	4.1 ± 2.9	19500 ± 1670	16090 ± 1710
	1–2 cm	23.2 ± 1.5	13.4 ± 2.2	<8.2		33300 ± 9990	29890 ± 10000
	2–3 cm	25.3 ± 1.7	15.5 ± 2.3	17.1 ± 2.3	10.9 ± 3.0	29100 ± 8730	25690 ± 8740
	3–4 cm	23.0 ± 1.5	13.2 ± 2.2	14.5 ± 2.5	7.7 ± 3.2	28100 ± 8430	24690 ± 8440
	4–5 cm	16.3 ± 1.2	6.5 ± 2.0	14.4 ± 2.1	7.5 ± 3.1	22900 ± 6870	19490 ± 6880
200	0–1 cm	10.4 ± 0.9	0.6 ± 1.9	10.7 ± 1.7	3.0 ± 2.2	11600 ± 3480	8190 ± 3500
	1–2 cm	10.6 ± 0.8	0.8 ± 1.8	9.1 ± 1.4	1.0 ± 1.9	10900 ± 3270	7490 ± 3290
	2–3 cm	11.1 ± 0.8	1.3 ± 1.8	12.0 ± 1.7	4.6 ± 2.3	7800 ± 2340	4390 ± 2370
	3–4 cm	10.8 ± 0.8	1.0 ± 1.8	<8.2		10100 ± 3030	6690 ± 3060
3200§	0–5 cm	9.8 ± 1.7		8.2 ± 0.6		3410 ± 390	

* 2σ counting uncertainty.

** Propagated 2σ counting uncertainty.

† Decay corrected (number of days between sampling and analysis time), associated with propagated 2σ counting uncertainty.

‡ Default 2σ uncertainty assumed to be 30 %.

§ Background mean ± SD values from control station (n=3).

Data availability

Data will be made available on request.

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