Naturally occurring radioactive material (NORM) in North Sea produced water: environmental consequences

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Preface

This report has been prepared for Norsk Olje og Gass by Dag Øistein Eriksen, Primus.inter,pares, and Ketil Hylland, Hylland Environment. Einar Lystad and Egil Dragsund have represented Norsk Olje og Gass.

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Abstract

Produced water from offshore gas or oil production platforms in the North Sea have been reported to contain varying amounts of radioactive isotopes, NORM[1], but good data only exists for ^{226}Ra. There are large differences in the concentration and total emissions of NORM from different platforms and production areas. The estimated total amount of radioactivity released annually is high in absolute numbers, but resulting increases of total radioactivity, or indeed the isotopes in question, are not measurable in North Sea ecosystems.

The aim of this report is to provide an overview of NORM released with produced water with a focus on (i) which isotopes will be present, (ii) distribution and compartmental partitioning in marine ecosystems in general and the North Sea in particular, (iii) factors that may affect partitioning and behaviour, (iv) bioavailability and bioaccumulation in marine organisms and, finally, (v) possible consequences for the marine environment.

The project concluded that there is no evidence for the presence of increased concentrations of ^{226}Ra or ^{228}Ra, in sediments in the Norwegian Trench or for increased accumulation of ^{226}Ra in marine organisms of the North Sea due to produced water inputs of this isotope (or ^{228}Ra). With current knowledge of produced water composition, there will be insignificant additional ^{210}Po from this source present in seafood for human consumption and it is unlikely that the observed levels of ^{226}Ra in seawater or sediments in the North Sea, from natural sources or produced water, will cause effects in marine organisms.

[1] NORM = Naturally Occurring Radioactive Materials

Background

Produced water from offshore gas or oil production platforms in the North Sea have been reported to contain varying amounts of naturally occurring radioactive materials (NORM), predominantly ^{226}Ra, ^{228}Ra and ^{210}Pb (Gäfvert et al., 2007). Good data only exists for ^{226}Ra and there is some uncertainty as how much of other radioactive isotopes would be expected to be present due to biogeochemical processes in the reservoir and properties of the different daughters in the series (see below; Varskog and Schultz, 2012). There are also large differences in the concentration and total emissions of NORM from different platforms and production areas (Gäfvert et al., 2007). The estimated total amount of radioactivity released annually is in the order of TBq (10^{12} Bq), but resulting increases of total radioactivity, or indeed the isotopes in question, are not measurable in North Sea ecosystems (cf. Betti et al., 2004).

A series of recent reviews have been published concerning NORM in marine ecosystems with regard to concentrations (Paschoa and Steinhäusler, 2010; Vives i Batlle, 2012), concentration ratios between water and organisms (Hosseini et al., 2010, 2008; Thorne, 2003) and possible impacts (Adam-Guillermin et al., 2012; Hosseini et al., 2012; Tracy et al., 2013).

The aim of this report is to provide an overview of NORM released with produced water with a focus on (i) which isotopes will be present, (ii) distribution and compartmental partitioning in marine ecosystems in general and the North Sea in particular, (iii) factors that may affect partitioning and behaviour, (iv) bioavailability and bioaccumulation in marine organisms and, finally, (v) possible consequences for the marine environment.

The 2003 strategy of the OSPAR Commission ".. is to prevent pollution of the maritime area from ionising radiation through progressive and substantial reductions of discharges, emissions and losses of radioactive substances, with the ultimate aim of concentrations in the environment near background values for naturally occurring radioactive substances and close to zero for artificial

radioactive substances" (OSPAR reference 2003-21). As for other natural substances, e.g. metals, there are some obvious challenges inherent in this statement, such as regional differences in background levels and the challenges involved in separating naturally present metal or radioactive isotopes from those added through anthropogenic activity such as oil and gas production.

Radioactivity in produced water

The most significant primordial[2] nuclides are ^{40}K, ^{232}Th, ^{235}U and ^{238}U. The latter three also give rise to the well-known radioactive series presented with their major decay branches in Figure 1. ^{40}K is the nuclide contributing most to the total radioactivity in the oceans (Solomon, 1988).

Non-primordial, naturally occurring radioactive nuclides are mainly ^{3}H and ^{14}C, both continuously produced in the upper atmosphere by nuclear reactions induced in nitrogen nuclei by cosmic radiation. Both nuclides will, after creation, react chemically with oxygen and will subsequently enter the ecological cycles as $^{3}H^{1}HO$ and $^{14}CO_2$, respectively. Thus, the nuclides will be parts of the water and carbonate systems, and enter all biologically active organisms.

[2]Radioactive substances may be categorized in several ways, but in this context the geological division into primordial, naturally-occurring nuclides not primordial, and man-made radioactivity has been chosen.

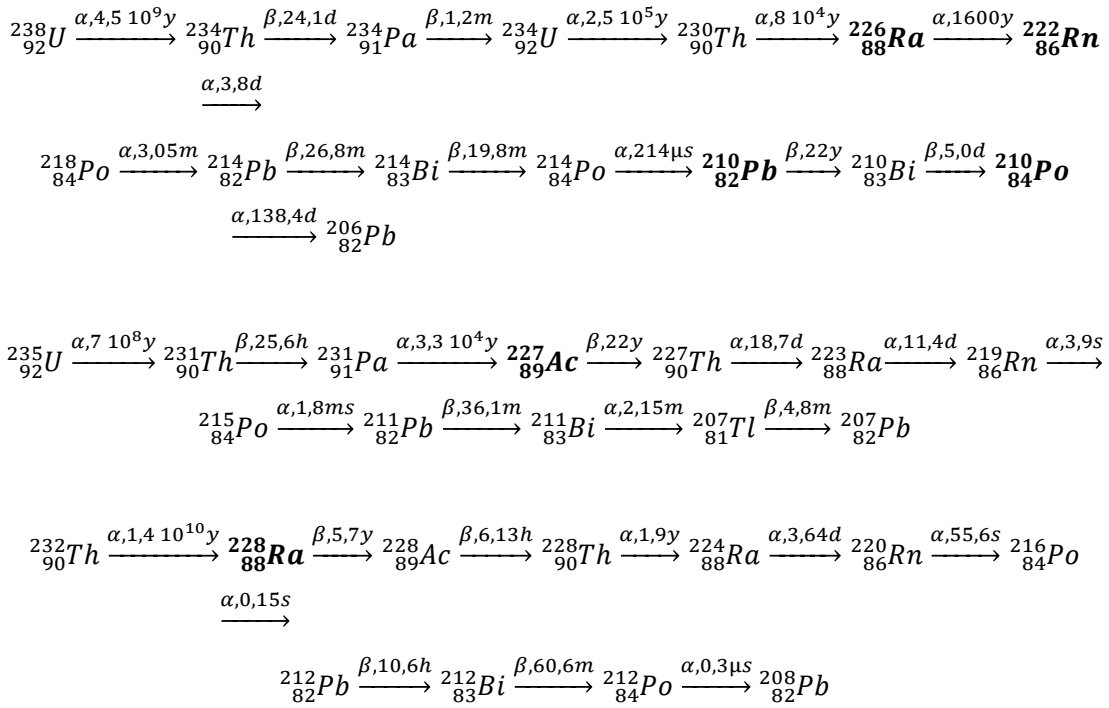

Figure 1. *The three radioactive series stemming from the primordial nuclides ^{232}Th, ^{235}U, and ^{238}U with their main routes of decay. The decay mode (α or β) is indicated and so is the half-life of the nuclide. The most relevant daughter nuclides for this report are in bold.*

Man-made (anthropogenic) radioactivity is mainly due to fallout from nuclear tests in the 1950s and 60s, effluents from nuclear recycling establishments, e.g. Sellafield and La Hague, releases from nuclear accidents such as Chernobyl, effluents from landfills and mine tailings, as well as inputs from fertilizer production and use (see e.g. Baxter, 1996; European Commission. Directorate-General Environment. Radiation Protection, 2003; Navarrete et al., 2012; Paschoa and Steinhäusler, 2010; Santos et al., 2006; Vives i Batlle, 2012). During the last few decades, oil- and gas-producing installations at sea have also been contributing to such inputs, as they emit produced water, containing ^{226}Ra in particular. As mentioned above, a recent report shows that it is only ^{226}Ra and ^{228}Ra that contribute to radioactivity in produced water; other radioactive isotopes such as U, Th, Po, and Pb were close to detection limits if at all detectable (Schultz and Varskog, 2012).

Effluents from nuclear recycling establishments mainly contain long-lived fission products such as ^{90}Sr, ^{99}Tc, and ^{137}Cs, whereas effluents from landfills and mine tailings are similar to the primordial ones. Atmospheric fallout has further contributed to increased levels of the fission product mentioned above, as well as to levels of ^{3}H and ^{14}C in marine ecosystems.

The fertilizer industry dissolves phosphate-containing minerals, e.g. apatite, in sulphuric or nitric acid. The sulphuric route creates huge piles of gypsum as waste. These deposited piles contain traces of uranium and daughter nuclides. Following weathering, uranium is dissolved whereas radium stays in the sulphate-rich environment (Betti et al., 2004; Carvalho, 1995). In Europe, effluents from such piles have diminished as the fertilizer industry has stopped its production and moved to other continents. The fertilizer industry in Norway has used the alternative technology, i.e. the nitric acid route, which has no releases of radioactivity.

Walker and Rose (1990) assessed the dose contributions from all radionuclides present in the oceans. The average concentration of radioactivity in surface waters is 13.6 Bq/kg water. ^{40}K alone contributed 12 Bq/kg (88%).

Ocean waters are saline, but the salinity varies from brackish oceans, e.g. the Baltic Sea, to tropical seas such as e.g. the Persian Gulf. The higher the salinity, the higher the content of radioactive species. For example, the Persian Gulf contains 22 Bq/kg, whereas the Baltic Sea only has 4 Bq/kg (Walker and Rose, 1990). The correlation between radioactivity and salinity, i.e. mainly the concentration of Na^+ and Cl^-, can be explained by K^+ behaving similar to and having a carrier in Na^+, formation of chloride complexes, pH, and the higher carbonate concentration in saltwater compared to freshwater. This is for example important for keeping uranium as uranyl, UO_2^{2+}, in solution. Uranium can exist in several oxidation states, but in marine waters uranyl, U(VI), is by far the most dominant species. Uranyl will form complexes with carbonate at seawater pH (Langmuir, 1978). For pH ≥ 8 $UO_2(CO_3)_3^{4-}$ is the dominant complex. Following Langmuir the aqueous phase must be very anoxic to reduce U(VI) to

U(IV), but at marine pH the uranium will still be in solution as a complex, i.e. $U(OH)_5^-$.

For thorium the situation is quite different. For seawater, Choppin (1989) reported $\log\beta_4 = -17.26$[3] for the hydroxide, indicating that thorium is a solid at this pH. This is also in accordance with measurements of the species in seawater - ^{232}Th is present at ultra-low concentrations (Walker and Rose, 1990), i.e. 0.5 µBq/kg compared to 41 mBq/kg for ^{238}U. Thus, the Th-isotopes present in seawater will nearly entirely be the daughter nuclides from decay of various U-isotopes (cfr. Fig.1), i.e. ^{234}Th 41 mBq/kg, ^{231}Th 1.9 mBq/kg, ^{228}Th 120 µBq/kg, and ^{230}Th 52 µBq/kg (Walker and Rose, 1990). In a more recent study, Hosseini et al. (2010) report the following concentrations in filtered (coastal) seawater: ^{234}Th 12 mBq/L, ^{232}Th 80µBq/L, ^{228}Th 300 µBq/L, and ^{230}Th 150 µBq/L, and ^{238}U 41 mBq/L. Their Th-concentration corresponds to 85 nM.[4] These nuclides born in their parent nuclide's chemical environment have long enough half-lives to establish their own chemical surroundings if possible. Most thorium will therefore form hydroxides and tend to precipitate or be absorbed onto solids present in the water column. Successively, these nuclides will form daughter nuclides, some of which are of radiotoxicological relevance.

From an effect perspective, the most important radioactive nuclides in the marine environment are ^{226}Ra, ^{228}Ra, ^{210}Pb, and ^{210}Po, as they have sufficiently long half-lives to be released into the environment, but on the other hand sufficiently short half-lives to give them high specific radioactivity. Walker and Rose (1990) reported the concentrations of ^{226}Ra as 3.6, ^{210}Po as 3.7, and ^{210}Pb as 5.0 mBq/kg while Hosseini et al. (2010) reported 3.4, 2.0 and 2.0 mBq/L for the same nuclides, respectively. ^{228}Ra was not quoted by Walker and Rose, but Hosseini et al. reported 10 mBq/L for ^{228}Ra. Radium is a "bone seeker" in vertebrates and may thus have a particularly high radiotoxicity in this group of

[3] β_n is the equilibrium constant for the reaction: $M^{n+} + nL^- = ML_n$.
[4] The discrepancy between these authors is not easy to explain. An increase in Th-concentration in the ocean waters of 160 times in 20 years is unlikely. Their U-concentrations are equal indicating the measurement of α-radiation and sample preparations are acceptable.

organisms, including fish (see e.g. Pyle and Clulow, 1998). The speciation of radium in seawater, and hence its bioavailability, is therefore of particular interest.

Seawater has on average 15 mM of sulphate, (Choppin, 1989). The solubility product for $RaSO_4$ is $3.66x10^{-11}$ (Thiers, 2010), but this will require a Ra-concentration of 2 nM to form a precipitate. 3.6 mBq/kg corresponds to approximately 430 aM ($4.3x10^{-16}$M), which is seven orders of magnitude lower. It is thus more probable that the Ra^{2+} will form complexes with sulphate ions than that it will be precipitated. However, the presence of Ba^{2+} can alter the situation as Ba^{2+} and Ra^{2+} are known to behave quite similar in most chemical environments (Kirby and Salutsky, 1964). This is understandable simply from looking at their ionic radii, $r(Ra^{2+})$ = 148 vs $r(Ba^{2+})$ = 142 pm, i.e. an increase of only 4,2%. Ba^{2+} is therefore the best chemical carrier for Ra^{2+}. The solubility product for $BaSO_4$ is $1.08x10^{-11}$ (Thiers, 2010). There are however some differences in the marine geochemistry of barium and radium, as reported by Li et al. (1973).

Li et al. (1973) showed that the ratio between the concentrations of radium and barium is almost constant at $7.1x10^{-9}$ by weight, and that there is a strong correlation between these two elements and the concentration of silicate in oceanic waters (Li et al, 1973). While Li et al. focused on the Antarctic ocean, Dehairs et al. (1980) have studied the barium cycle in the open ocean in general. These authors showed that the budget for barium in the ocean is strongly correlated with biological activity. Radium and barium can form particulate forms and absorb to particulate matter present in the water phase. This is believed to be the reason for the correlation between barium and silicate. Dymond et al. (1992) showed that organic carbon in the water column is important for the sedimentation of barium to the seafloor. They showed that in the settling flux of barium 50 – 70% was as barite and the remaining absorbed to particles or bound to carbonates. Beneš and co-workers have studied these properties in a series of published papers, e.g. Beneš et al. (1981). Their main

focus was, however, on rivers and wastewaters, not seawater and marine chemistry.

^{210}Po is the major source of the collective human dose due to the presence of polonium in seafood (e.g. Aközcan and Uğur, 2013; Aközcan, 2013; Gouvea et al., 1992; Shannon et al., 1970), see further discussion below. Polonium forms lipophilic chemical complexes and will accumulate in the organism's fat. With a short half-life of 138 days the nuclide is produced from decay of ^{226}Ra and ^{210}Pb in the environment (Betti et al., 2004). Thus, the influx of polonium to the ocean water comes mainly from the water itself and pore-water in sediments.

Of the anthropogenic isotopes, ^{90}Sr, ^{99}Tc, and ^{137}Cs are fission products from nuclear wastes. Strontium has, as an alkaline earth, chemical properties in between calcium and barium forming carbonates and sulphates of low solubility. What is applicable for Ba^{2+} is usually the same for Sr^{2+}, but with an ionic radius of 126 pm its chemical properties are closer to Ca^{2+} with an ionic radius of 112 pm. ^{99}Tc is exclusively present in the oceans as TcO_4^-, pertechnetate. This compound is known to be chemically stable and water soluble, and thus mobile. It has been observed that seaweed tend to absorb pertechnetate (Shi et al, 2012). However, at anoxic conditions, e.g. in fjords with low exchange of water, the uptake of technetium increases (Keith-Roach et al., 2003). Caesium behaves much like potassium although Cs^+ has a high and well known affinity for adsorption onto clay minerals, e.g. montmorillonite. Radium has also been shown to adsorb to clay, but it is not selective relative to barium (Zhang et al., 2001).

Sources and their relative contributions

The pH of oceanic seawater is just above 8 and it is quite stable due to the buffer capacities of the carbonate and dissolved organic acids present. Thus, the marine environment does not offer conditions where U(VI) will be precipitated. Seawater is therefore a sink for uranium. The element has been accumulated in the seawater for thousands of years. Goldberg and Koide (1962) estimated the mean residence time of U in ocean water to 500 000 years.

Uranium, thorium, radium, polonium, and radioactive lead in seawater have different chemical behaviour and mobility. The mobility and speciation of various radionuclides were discussed by Ivanovich (1994). A simple measure of the different mobilities of uranium and thorium is to quantify the decay-rates of ^{238}U and ^{230}Th. With no mobility the rates should be equal, but it is commonly found that uranium has moved away from thorium (Ivanovich, 1994). Figure 2 shows a schematic model of the geochemical sources and fluxes for the natural radioactive species. i.e. uranium and thorium, in the near-surface environment. Thus, the elements have to be treated individually, but there are some common possible sources. Such sources include rivers and glaciers transporting elements from the shore, sediments releasing radionuclides into the sea, bedrock erosion underwater or in the littoral, groundwater and surface water discharges, pollution from sunken nuclear submarines or deposits and fall-out, effluents from mines and deposits, petroleum production, and other human activities.

Cochran (1992) described four sources, referred to by Dowdall and Lepland (2012):

1. Riverine supply of particle bound uranium, radium, thorium and daughters to estuarine areas with desorption in the mixing zone and release to the seawater.
2. A presence of mother nuclides with mobilisation to sediment pore water by alpha recoil generate nuclides in marine sediments.
3. Decay of uranium, radium and radon leading to *in situ* generation of daughter nuclides in seawater.
4. Atmospheric deposition due to the emanation of ^{222}Rn from land with transport over the seas and decay to ^{210}Pb, scavenged from the atmosphere by precipitation.

In addition, diffusion inside and eventually out of solids, in particular of radon, must be included.

Barium and radium have different mineralogical origins. Ba comes from barite, $BaSO_4$, or as impurity in carbonaceous rocks like limestone, chalk, or dolomite, while Ra's origins are rocks containing uranium, e.g. granite and sandstone. Nevertheless, in the oceans the concentrations of barium varies between 5 and 25 µg/kg (35 – 180 nM) (Li et al, 1973) compared to ^{226}Ra of 430 aM.

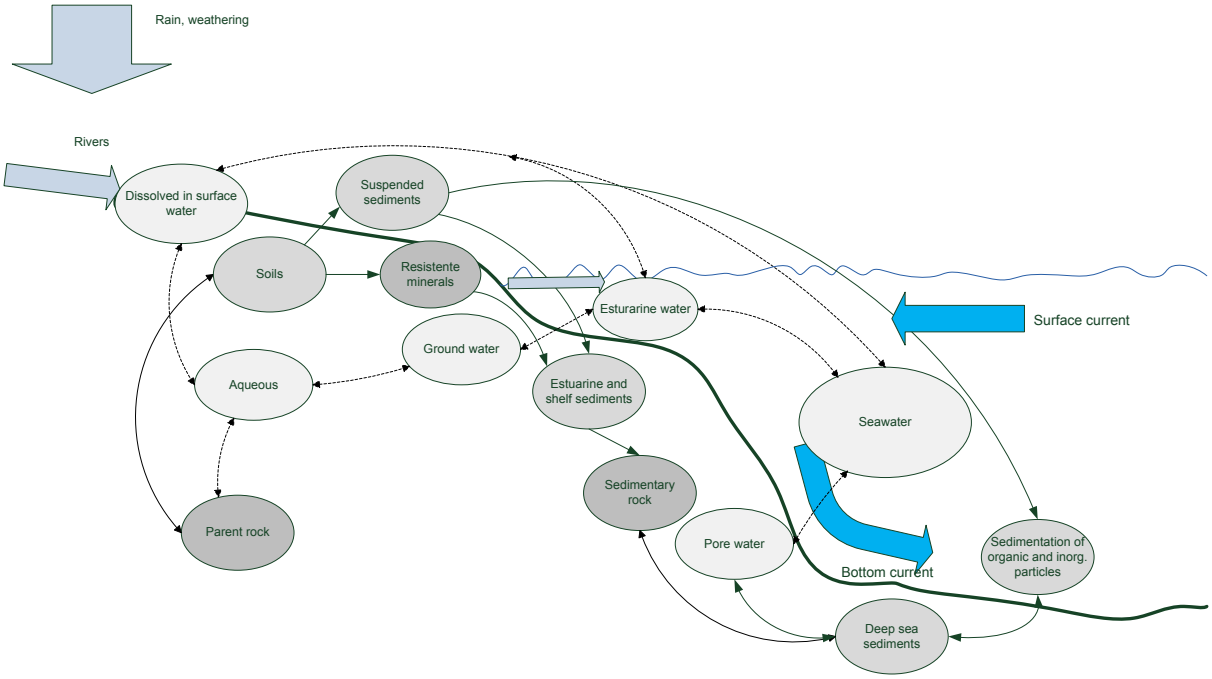

Figure 2. *Schematic of the U and Th geochemical cycle in the near-surface environment. Light coloured ellipses indicate aqueous solutions, darker indicates more immobile phases. Broken arrows indicate mobility of liquids while solid arrows indicate mobility of solids. Figure based on one presented by Ivanovitch, 1994.*

Rivers and estuaries

Rivers bring dissolved as well as particulate matter to the sea. However, the big rivers carrying the largest volumes of solids also generally have extensive estuaries, where the major fraction of the solids will sediment. Large rivers may deliver particles far into the sea, thereby decreasing the salinity and increasing turbidity even far from the shore. The smaller the particles, the more mobile they are. The flux of suspended particles as well as the water flows will have seasonal changes, and the mixing with ocean water will have tidal variations. Thus, to model the flux of radionuclides through an estuary is a complicated task.

Laissaoui and co-workers studied the effects of salinity and suspended load concentrations for barium and radium in estuarine environment where tidal and seasonal changes imply varying salinity and concentration of suspended solids (Laissaoui et al. 1998). Their aim was to obtain transfer coefficients for radionuclides. The coefficients were tested using ^{133}Ba as a tracer in a natural estuary. A similar, but more direct measurement of ^{226}Ra and ^{228}Ra in The Wash, Eastern England, was reported by Plater et al. (1995). They concluded that the ratio between the two Ra-isotopes could be attributed to a combination of conservative mixing and desorption from suspended sediments, while more uniform ^{226}Ra contents may have reflected the bottom sediment diffusion. These processes were mainly controlled by the salinity.

Sediments

Sediments are usually porous rock materials consisting of cemented particles with interstitial water, pore-water. In the sediments the water-soluble radionuclides may then go into solution in the pore-water. The smaller the particles, the more mobile they will be. This has been shown by measurements of radium on sediments from the relatively shallow North Sea and from the much deeper Norwegian Trench (Eriksen et al., 2009). These authors report a content of 1 – 15 Bq/kg of ^{226}Ra in sediments of grain size from 100 µm to 3 mm at the shallow sites, while for the samples from the Trench 10 – 60 Bq/kg of ^{226}Ra at particle sizes of 1 – 100 µm are reported. For small particles to settle, the movements in the sea must be minute, i.e. below the depth where surface waves can influence. From model calculations, Li et al. (1973) concluded that 99% of the radium in the oceans is derived from sediments whereas the influx of barium from sediments is insignificant compared to input from rivers.

Dowdall and Lepland (2012) reported activity from ^{226}Ra, ^{228}Ra, ^{40}K, and ^{137}Cs in sediments from the Norwegian Trench. They also showed profiles of the radioactive components as a function of sediment depth. The signature from ^{137}Cs stems from the Chernobyl accident in 1986 and is used for dating of the sediments, showing a sedimentation rate of ca 10 mm/y. The samples also show that bioturbation will cause the Cs-peaks to broaden (e.g. from 0 – 200 mm at

station 189). The variations in the peak's positions and width indicate that there are different mineral compositions in their samples. It is imperative to indicate the kind of mineralogy and depth (correlated with particle size distribution) to enable a correct evaluation of such data. Dowdall and Lepland's data show that ^{226}Ra and ^{228}Ra have almost identical depth distributions. With the vast difference in half-lives, 1600 vs 5.7 years, and with sediment age of > 25 years, ^{228}Ra must be formed from ^{232}Th present in the sediment. It is therefore not probable that the peaks of both radium-isotopes close to the surface reflected a physical condition, as there are no known emissions of either uranium or thorium. The peaks show no bioturbation and there is no reason for the observed variation in depth, i.e. time, from one site to the neighbouring site. The radium-peaks close to the sediment surface presented by Dowdall and Lepland (2012) must therefore be considered as artefacts.

Produced water

The offshore oil and gas production has created new sources for emission of radioactive nuclides to the sea far from estuaries and close to the ocean surface. Thus, this represents a novel (< 40 years) source without the possible hold-up and absorptions as a river or ground water source represent. In the Norwegian sector the emission of radioactivity from the installations is monitored and reported. In other sectors there are other reporting regimes.

Produced water is water brought up from the reservoir and then separated from oil or gas. It is sometimes referred to as "production water" or "formation water". Thus, the properties of the reservoir are important for the concentration of water-soluble radioactive species present. In general, radium is the most water-soluble element in the radioactive series and radon the most oil- and gas-soluble. In sand stone reservoirs there can therefore be more radium expected in produced water than from chalk reservoir. The recent report by Zpire (Varskog and Schultz, 2012) shows this explicitly: sand stone reservoirs like Troll and Brage have produced waters with 8 – 10 Bq/L of ^{226}Ra and 8 Bq/L of ^{228}Ra, while chalk reservoirs like Valhall and Ekofisk have < 2.5 and < 1 Bq/L for ^{226}Ra and ^{228}Ra, respectively. However, a sand stone reservoir does not have to yield high

radium-concentrations, Statfjord and Gullfaks both have produced waters with < 1 Bq/L of the Ra-isotopes. No other radioactive nuclides are present at detectable levels in produced water from North Sea installations.

The release of produced water from installations in the Norwegian sector in 2011 was approximately 129 Mm^3. This corresponds to a volume 1 km square and 129 m deep. This is a small volume compared to the size of the North Sea. Therefore the contribution of radioactivity to the seawater from produced water would be expected to be below detection limits for radiation effects, as indeed shown by Eriksen et al. (2009). This has been confirmed by annual surveillance activities in accordance with requirements from NRPA[5].

Bioavailability

The bioavailability of any substance will in the following be defined as the fraction of the substance in question that is available for interaction with the cell(s) of aquatic organisms. Such interaction may or may not be the result of accumulation. In contrast to other substances that will require molecular interaction with membranes, and possible subsequent interaction with cellular components, radioactive isotopes may exert an additional influence by radiation from outside the cell (see e.g. Thomas and Liber, 2001). In seawater, nearly all radium would be present as the neutral $RaSO_4$, a molecule that would not be expected to be taken up through ion channels or ion pumps to any extent. Marine fish need to drink seawater and processes in the gastrointestinal system may lead to some uptake of radium. Finally, there has been some discussion as to whether there may be some uptake through endocytosis, either on the gills or in the intestinal tract (see e.g. Streit, 1998).

There is a scarcity of experimental studies actually measuring the bioavailability of e.g. radium in water or sediment. One of the few, Grung et al. (2009) found low bioavailability of radium in sediment to the ragworm *Hediste diversicolor* (see below). The bioavailability was somewhat increased in the presence of a

[5] NRPA = Norwegian Radiation Protection Authority

complexing agent, scale inhibitor, relevant to produced water (Grung et al., 2009), but not to an extent that will have significant consequences in the environment.

Bioaccumulation

Bioaccumulation is the difference between uptake and biotransformation/excretion. For metals and radioactive isotopes there is of course no biotransformation, although they will be associated with different components in or outside cells. Excretion will be proportional to the concentration in tissues, and the persistence of a metal or radioactive isotope in tissues will depend on how it is associated with relevant components and the efficiency of excretion through bile/the gastrointestinal tract and/or urine.

Concentration factors (CF) or concentration ratios (CR) will reflect the bioaccumulation potential of an isotope, but assume equilibrium, which is not always the case (Hosseini et al., 2008). CRs have been compiled for radium for a range of organisms, reported within the ERICA tool (Hosseini et al., 2010, 2008) and for a range of other isotopes in those two studies and others (e.g. Strand et al., 2009; Tagami and Uchida, 2012). Averages reviewed by Hosseini et al. (2008) were 91 L/kg for microalgae, 81 L/kg for zooplankton, 110 L/kg for zooplankton and 200 L/kg for fish, but with huge variation within each group. These values are in contrast to the results found by Grung et al. (2009), in which CRs for the ragworm *Hediste diversicolor* was 0.2-0.5 L/kg ww (calculated from pore water concentrations). There is no clear explanation for this discrepancy, as the study with the ragworm lasted four weeks, which should be sufficient for equilibrium to be established.

Biomagnification

Persistent and lipid soluble molecules may accumulate to higher concentrations in organisms higher in food chains, a process referred to as biomagnification. This process is however only relevant for substances that are taken up efficiently through the gastrointestinal tract, such as legacy contaminants PCBs and DDT.

Inorganic substances such as metals will not biomagnify although they may accumulate and have long biological half-lives in organisms, mainly because intestinal uptake is low.

Specific ions may accumulate to high concentrations in some species or tissues, such as ^{210}Po in some molluscs (Gouvea et al., 1992), but that is a result of species-specific traits, not general ecological processes.

There has been suggestions of biomagnification of some radioactive isotopes (see e.g. Ishikawa et al., 2004), but the study did not address that process, but species and tissue differences in accumulation. The species analysed were either filter-feeders or algae. Hence, the results do not show biomagnification.

Accumulation of NORM in seafood

As referred to briefly above, several studies have indicated ^{210}Po to be the main contributor to human exposure through seafood (Aarkrog et al., 2002; Aközcan and Uğur, 2013; Aközcan, 2013; Cherry et al., 1994; Gouvea et al., 1992). Earlier studies have indicated that it is unlikely that appreciable amounts of ^{210}Po will be released in produced water and this has been corroborated by analyses, albeit not that many (Schultz and Varskog, 2012). Current knowledge therefore indicate that produced water inputs will not contribute to increased concentrations of ^{210}Po in seafood, which would otherwise be an issue with regard to human or food chain exposure. As will be apparent from the discussion of bioaccumulation above, radioactivity contributed by ^{226}Ra in marine organisms in the North Sea will be low, as has indeed been shown (Eriksen et al., 2009). This is both due to low levels and low bioavailability.

Effects

Natural radioactive isotopes in seawater, the main of which is ^{40}K, will result in a continuous background radiation for marine organisms. Potassium is an essential element and will be taken up by all organisms, including unicellular organisms such as algae and bacteria. Intracellular levels will however be

regulated, resulting in a virtually identical concentration in all organisms (Hosseini et al., 2010). The isotope contributing to the highest relative dose when weighed is the alpha-emitter ^{210}Po (Hosseini et al., 2010). As mentioned above, any assessment of radiation effects will need to include both internal and external exposure, which will vary for different isotopes. For the alpha-emitters such as ^{210}Po and ^{226}Ra, uptake will be required. NORM found in produced water are also found naturally in the sea.

There is some understanding as to which radiation level will cause acute toxicity in various organisms (Figure 3), but fewer studies on sublethal responses in aquatic organisms. There is no reason why the relative sensitivity should be identical for acute and sublethal effects.

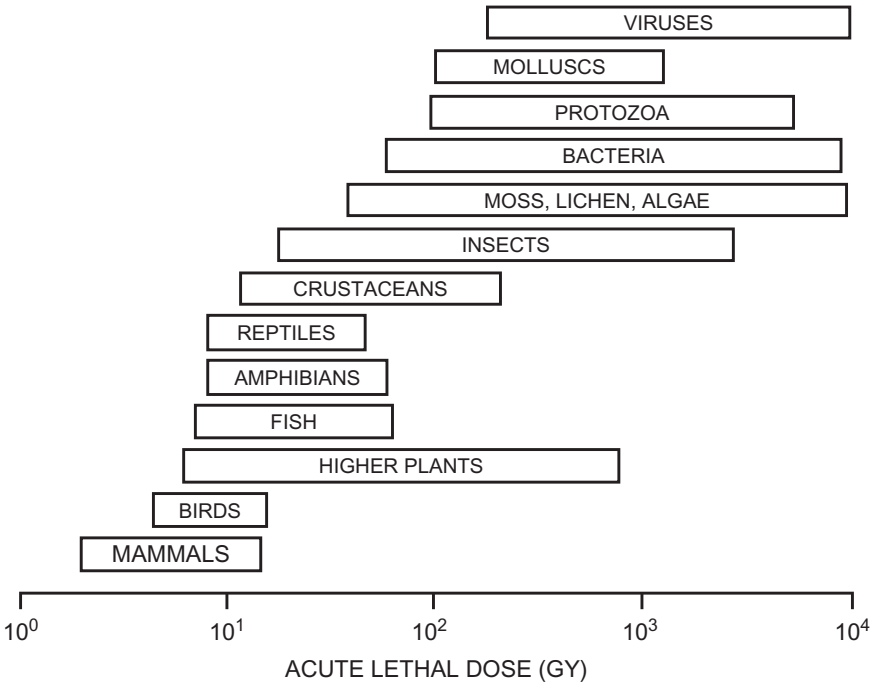

Figure 3. *Generalized overview of the predicted radiation sensitivity for different taxonomic groups (from Airey et al., 2012).*

Effects from gamma radiation on marine organisms have been comprehensively reviewed recently (Adam-Guillermin et al., 2012) and other studies have also addressed radiation effects from a range of isotopes in freshwater (Gilbin et al., 2008; Lagauzère et al., 2009; Thomas and Liber, 2001), marine (Adam-

Guillermin et al., 2012; AlAmri et al., 2012) and terrestrial (Antunes et al., 2013; Lourenço et al., 2012) ecosystems, but there is a scarcity of studies on the biological effects of radium. Many of the available studies were recently reviewed in Hosseini et al. (2012).

Grung et al. (2009) measured responses to oxidative stress in ragworm exposed to ^{226}Ra in spiked sediments for four weeks at activities of 470–6600 Bq kg^{-1} sediment. The polychaete accumulated the isotope in a dose-dependent manner (see above), but there were no effects on the antioxidant defence of the organism. The lowest activity was 5-10 times higher than activity of ^{226}Ra found in North Sea sediments.

In a second study within the same project, Olsvik et al. (2012) exposed blastula-stage cells from cod embryos to 2.11, 23 and 117 Bq/L ^{226}Ra for up to 72 hours. The experiment also included cells isolated from the gastrula stage and cadmium as a positive control. Olsvik et al. observed significant changes in the expression of some genes at some time points and concentrations, e.g. and glutathione peroxidase and heat shock protein 70 (HSP70), at the lowest concentration after 48 hours' exposure, and other genes, e.g. metallothionein and thioredoxin after 48 hours at higher concentrations. Due to the large number of comparisons performed in such a study with quantification of many genes at many time points there is a need to be careful in the interpretation of the results, but the lowest concentration at which responses were seen in two genes is within the range of ^{226}Ra in produced water. The activity of ^{226}Ra in seawater, even in the vicinity of platforms, is orders of magnitude lower (see above).

Conclusions

There is no evidence for the presence of increased concentrations of ^{226}Ra or ^{228}Ra, in sediments in the Norwegian Trench.

There is no evidence for increased accumulation of ^{226}Ra in marine organisms of the North Sea due to produced water inputs of this isotope (or ^{228}Ra).

With current knowledge of produced water composition, there will be insignificant additional ^{210}Po from this source present in seafood for human consumption.

It is unlikely that the observed levels of ^{226}Ra in seawater or sediments in the North Sea, from natural sources or produced water, will cause effects in marine organisms. This is in accordance with the conclusion by NRPA in their report on emissions of radioactive substances to the Norwegian Sea (Liland et al., 2012).

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