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Report

Long-term fate of Ra-226 originating from offshore produced water discharges

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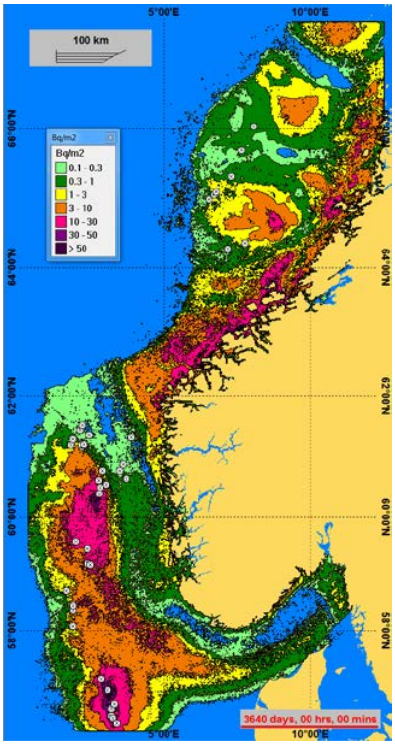

2016-06-23

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CLIENTS
NRPA

CLIENT'S REF.
Eivind Andreas Stubø

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ABSTRACT

Formation water in oil and gas fields contain elevated concentrations of several radionuclides compared with seawater. It is therefore of interest to track the fate of these radionuclides when they are released into the ocean along with the produced water stream. In this work we have simulated the fate of radionuclides in produced water discharge from all relevant installations on the Norwegian continental shelf for periods up to 20 years. We investigated separately the fate of the radionuclide Ra-226 in solid and in dissolved phase. Our findings show that when Ra-226 spreads as a solid it has the highest potential for reaching the sediments in the areas around the Oseberg and Ekofisk fields. The estimated contribution above background at these sites over a 30 year period was estimated to be from 2 Bq/kg to 10 Bq/kg, depending on the degree of burial, bioturbation and sediment mixing. Our simulations of dissolved Ra-226 showed transport along the Norwegian coast up past Svalbard. We found that the maximal contribution of dissolved Ra-226 to radioactivity in sediments over a 30 year period lies in the range 0.6 - 2 Bq/kg at several locations along the Norwegian coast.

for

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Table of contents

1 Introduction

Produced water from offshore oil and gas installations represent an anthropogenic source of radioactive discharges to the ocean, most notably of the radionuclide Ra-226. Once released, radionuclides may precipitate as particulate matter or may dissolve into sea water and spread with the ocean circulation. For radionuclides in both phases there is a potential for adsorption to seabed sediments. It is therefore of interest to investigate the fate of discharged radionuclides from offshore oil and gas installations. If they may accumulate in sediments, it is of interest to compare the added concentration to the natural background. For the Norwegian continental shelf (NCS) there is now historical data for produced water discharges going back two decades. In this report, we make use of these historical discharge data to simulate the long-term fate of radionuclides discharged in produced water to assess the resulting concentrations in the water column and the sediments.

In previous work, we have followed the fate of discharged radioactive nuclides in produced water on the NCS for a period of up to 5 years (Skancke *et al*., 2014). In this report, we present model simulations that build on the previous work but extend it in several ways. In this work we make use of all available reported data on radioactivity and volume of produced water discharges for all installations on the NCS from 1997 to 2014. This is an improvement on the previous work where discharge data from 2012 was used for all 5 years (Skancke *et al*., 2014). Second, we run the model at a higher resolutions over longer time periods: one simulation using 1.3 km resolution forcing data over a period of 10 years, and another using 12 km resolution forcing data over a period of 20 years. Together, these improvements lead to a more accurate assessment of the fate of discharged radionuclides.

2 Input data

2.1 Radionuclides in produced water

The radionuclides reported in produced water discharges on the NCS are 228-Ra, 226-Ra, 210-Pb, and 228-Th (source: NRPA). Data for Ra-226 is available starting from 2002; data for Ra-228 and Pb-210 is available starting from 2005; while data for 228-Th is available starting from year 2013. Table 1 gives an overview of properties of the radioactive discharges: activity, mass, half-life, and whether the radionuclide is an alpha or beta emitter. Ra-226, an alpha emitter, represents the largest release in terms of mass and activity. We therefore focus on this radionuclide in this report.

	Annual mass [g]	Emission type	Half life [years]	Mean annual activity [GBq]
Ra-226	12.16785	alpha	1600	445.5498
Ra-228	0.037438	beta	5.75	378.1115
Pb-210	0.014278	beta	22	40.9189
Th-228	0.00062	alpha	1.9	18.95681

Table 1: Properties and activity of radionuclides discharged in produced water on the NCS.

2.2 Produced water and Ra-226 discharge overview

In this work, we have used data of Ra-226 concentrations in produced water from 2002 to 2014, and volumes of produced water discharge from the years 1997 to 2014 (data provided by NRPA). We have summarized this input data in several figures. Figures 1 and 2 show the yearly total amount of released produced water [m3] and Ra-226 activity [MBq], respectively. Figure 3 shows the yearly variation in the distribution of Ra-

226 activity concentration [Bq/L] across all discharging installations. Figure 4 shows the distribution of Ra-226 activity concentration [Bq/L] for each installation.

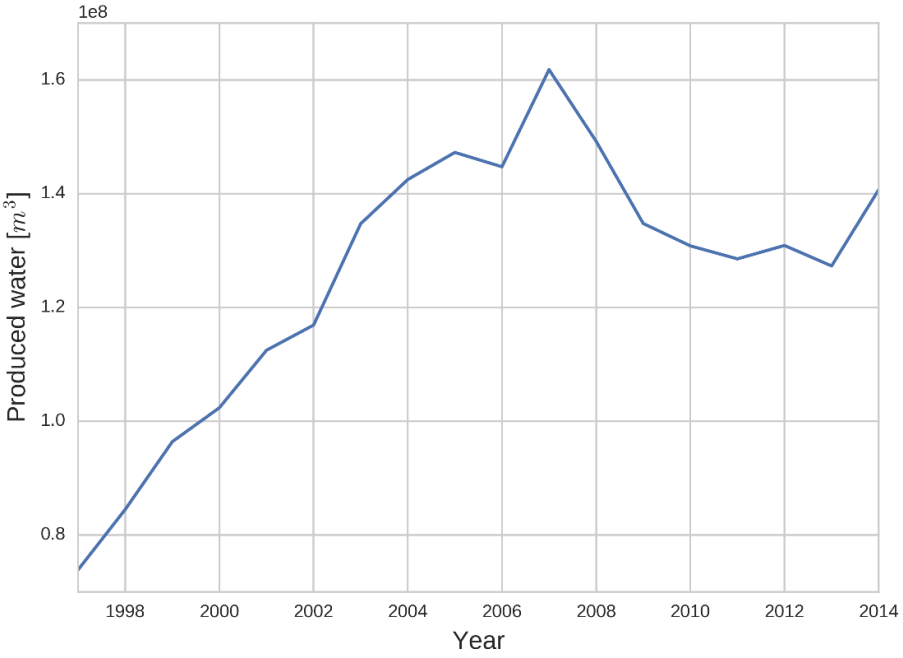

Figure 1: Yearly volumes of produced water discharge from offshore installations on the NCS

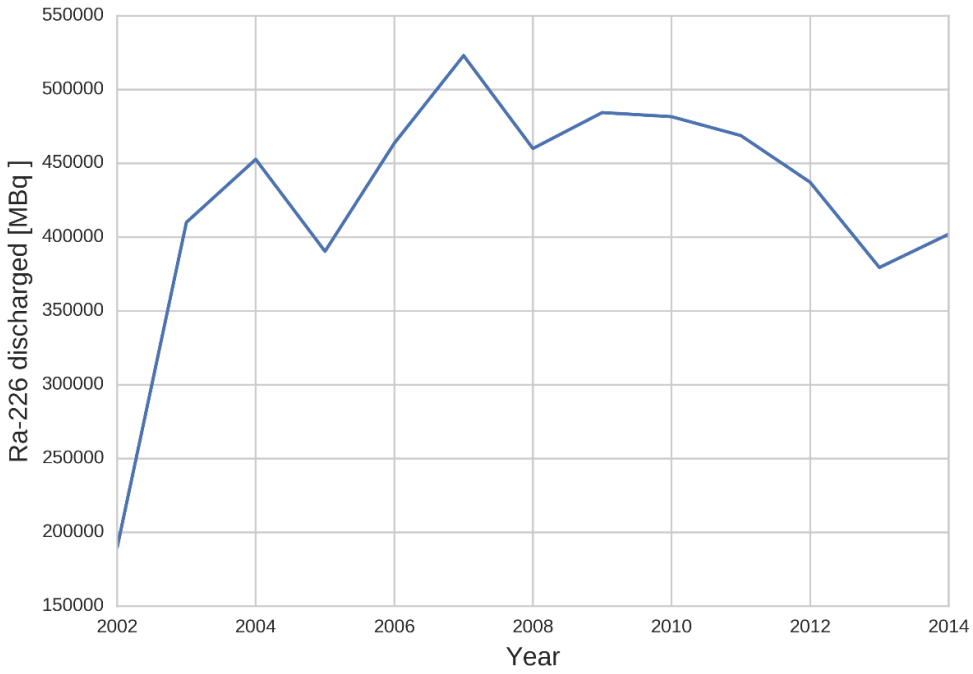

Figure 2: Yearly activity of Ra-226 originating from produced water discharges on the NCS

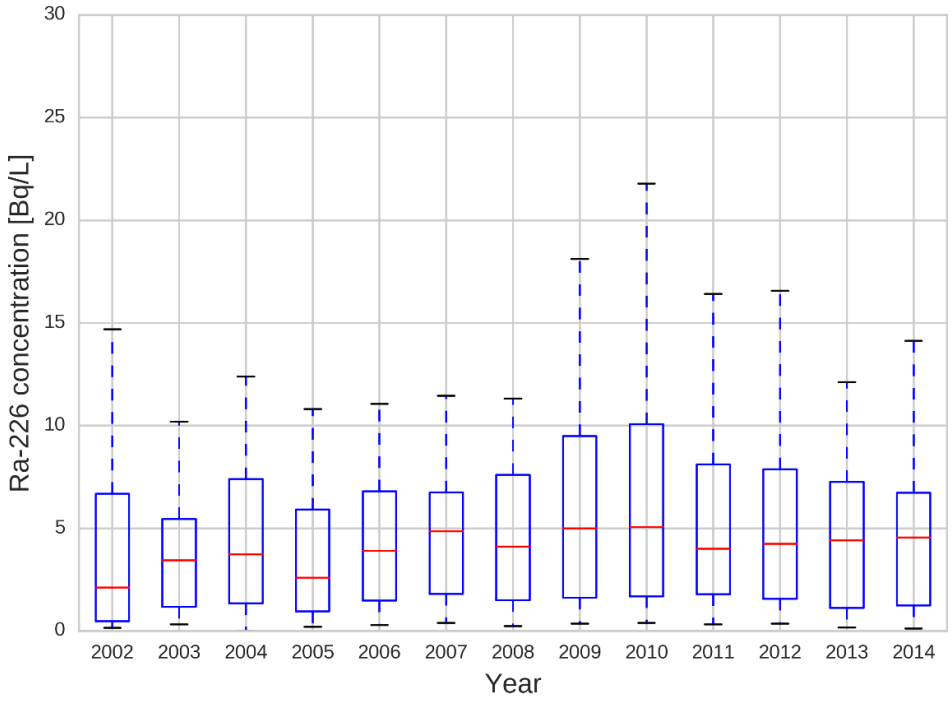

Figure 3: Distribution of Ra-226 activity concentrations in produced water discharges on the NCS by year.

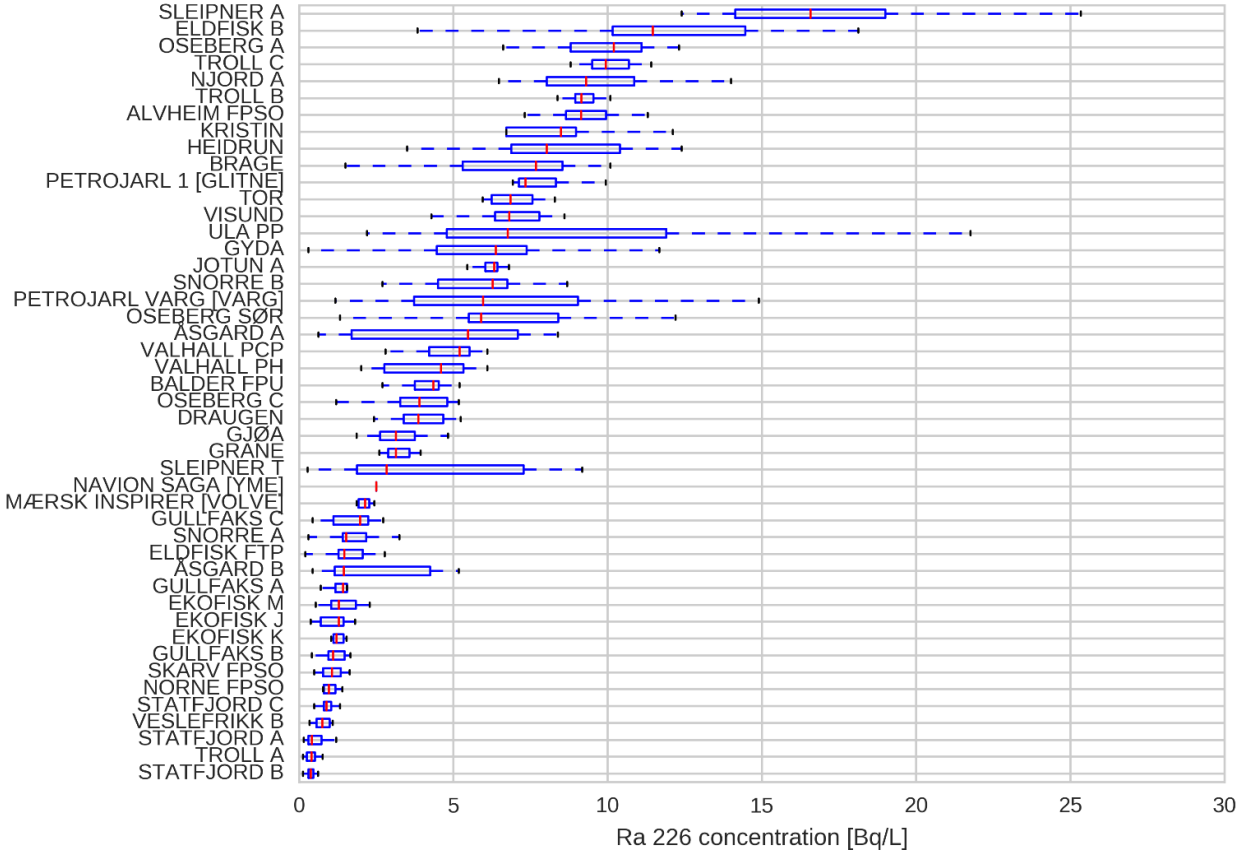

Figure 4: Distribution of Ra-226 concentration for all installations and fields, years 2002 – 2014.

2.3 Environmental data

Two environmental datasets have been used for the model simulations. The first dataset consists of wind and ocean currents data that span 10 years, from 2005 to 2014, with a horizontal resolution of 1.3 km, and a temporal resolution of 2 hrs. The model area was chosen as to cover all discharge sites on the NCS. The second dataset consists of wind and ocean current data that span 20 years, from 1995 to 2014, with a horizontal resolution of 12 km, and a temporal resolution of 1 hr. The model area was chosen to be able to follow dissolved radionuclides for long times and over long distances. The geographical extents of the model areas are shown in Figure 5.

Figure 5: Boundaries of current and wind data. Green outline: 1.3 km model. Blue outline: 12 km model.

2.4 The DREAM model

DREAM (Dose Related Risks and Effects Assessment Model) has been used to simulate the discharges of produced water and the resulting concentrations of Ra-226 in the recipient. The model calculates spreading based on turbulent diffusion and calculates transport from three-dimensional ocean current fields. More details about the DREAM model can be found in the published literature (Reed and Rye 2011; Reed and Hetland 2002). Resuspension in the model is calculated according to shear stress through the Shields criterion (Rye *et al.* 2006). The model uses a 1 hr timestep and a 1.5 km sediment grid cell size.

The model output does not include natural background levels. Therefore, all results shown with the DREAM model are the additional contributions of radioactivity in the sediments due to discharged Ra-226.

2.5 The parallel transport model

Simulations of transport of Ra-226 in the dissolved phase have been carried out using an experimental transport model developed at SINTEF called the parallel transport model. Like DREAM, it represents matter as numerical "particles", where each particle represents a certain mass. The particles are transported with the ocean currents using a variable time step approach, and diffusion is implemented as a random-walk process. The horizontal diffusion coefficient is calculated from the current data according to Smagorinsky (Smagorinsky, 1963), and vertical diffusion is set to 10% of the horizontal. Step lengths are scaled with the square root of the time step duration. This model is characterized by being able to process a very large number of particles, which makes it well suited for long-term simulation of dissolved matter in the ocean where each particle may have a long lifetime.

The simulations presented here were run for 20 years (1995 – 2014), using the current dataset with 12 km spatial resolution and 1 hour time step. At each release site, 40 particles per day was released. Particles are treated as dissolved, i.e., as passive tracers with no positive or negative buoyancy. When a particle comes into contact with the sea floor or the coast through diffusion, it is removed from the simulation. Land detection is done at the end of every timestep. The number of active particles in the simulation varies as particles are continuously produced at the discharge origin and lost as they come into contact with the sea floor. The maximum number of particles in circulation during the simulations was about 1.2 million, and the total number of particles released was about 43 million. It is stressed that this is an experimental model, with somewhat simplified diffusion and land detection to allow the model to run with a large number of particles.

For the part of the simulation covering 1995 to 1996 we have used produced water discharge data from 1997. As for DREAM, the model output does not include natural background levels. All results shown indicate the additional contribution of radioactivity due to discharged Ra-226.

2.6 Simulation scenario overview

After discharge into the ocean Ra-226 is likely present in both dissolved and solid form. As discussed previously (Skancke, 2014), it is not clear how Ra-226 is fractionated into these two phases. Neither is it clear how this fraction changes with dilution in seawater. We therefore investigate the fate of Ra-226 in two different scenarios, where Ra-226 is present either as 100% dissolved or 100% solid. In this regard, the simulation results can be considered extreme cases of the real situation.

When Ra-226 spreads in dissolved form it is neutrally buoyant and therefore less likely to interact with the sea bed, which means it will spread further from the discharge site. To simulate the long-term fate of dissolved Ra-226 we modelled this scenario using a parallelized particle model, as described in the previous section. This model used a large simulation area (covering the northern North Atlantic Ocean and most of the Arctic Ocean) with 12 km resolution. When Ra-226 spreads in solid form it is affected by gravity and is more likely to sediment closer to the discharge point. After sedimentation, Ra-226 may spread further as it resuspends from the sea floor. For the processes of sedimentation and resuspension, it is necessary to have a higher resolution representation of the ocean bathymetry. Therefore, we simulate the spread of solid Ra-226 using a high resolution 1.3 km dataset. Since this dataset has a high resolution, it also covers a smaller area than the 12 km model (Figure 1). Simulations of solid Ra-226 are performed with the DREAM model. To compare the extent of resuspension, we also run a simulation without resuspension. An overview of the three different simulations that have been run is given in Table 2.

Table 2: Overview of simulation scenarios

	Model	Time span	Resuspension	Phase	Model area
1	DREAM	2005-2014	No	Solid	Norway
2	DREAM	2005-2014	Yes	Solid	Norway
3	Parallel	1995-2014	No	Dissolved	North Atlantic and Arctic

2.7 Processing of input data for simulations

A total of 44 offshore installations were reported to discharge Ra-226 between 2002 and 2014. The activity concentration in the discharge from a given installation varies year by year (Figure 4), and not all installations have discharges every year. This results in a highly complex model release scenario. To reduce the total complexity of the release scenario we associated each installation with a single activity concentration, calculated from the mean of the activity concentrations for all available years. Since the distributions of activity concentrations for each site are fairly confined (Figure 4) we do not expect that this introduces large inaccuracies in the model result.

2.8 Particle size distribution for precipitated Ra-226

When Ra-226 is discharged into the ocean it can interact with SO_4^{2-} to form $RaSO_4$ (radium sulphate), which may further co-precipitate with the more abundant $BaSO_4$ (barium sulphate). For the simulations that consider the fate of Ra-226 in solid form we make use of a particle size distribution obtained from experimental measurements of $BaSO_4$ particles (Eriksen *et al*. 2006) as previously described (Skancke *et al*., 2014).

3 Results

3.1 Fate of solid Ra-226

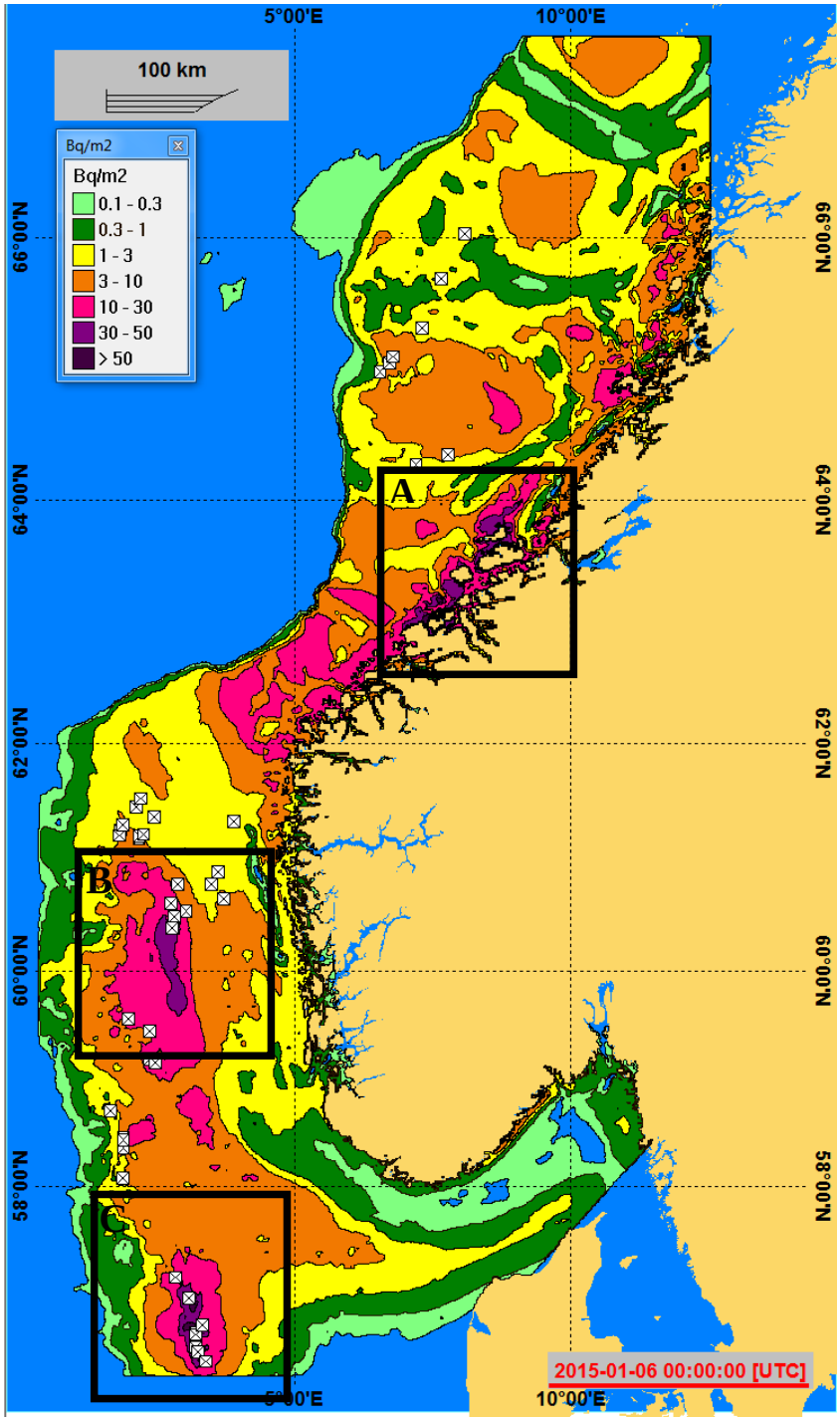

Figure 6: Simulation of dispersion of solid Ra-226 without resuspension after 10 years of discharge.

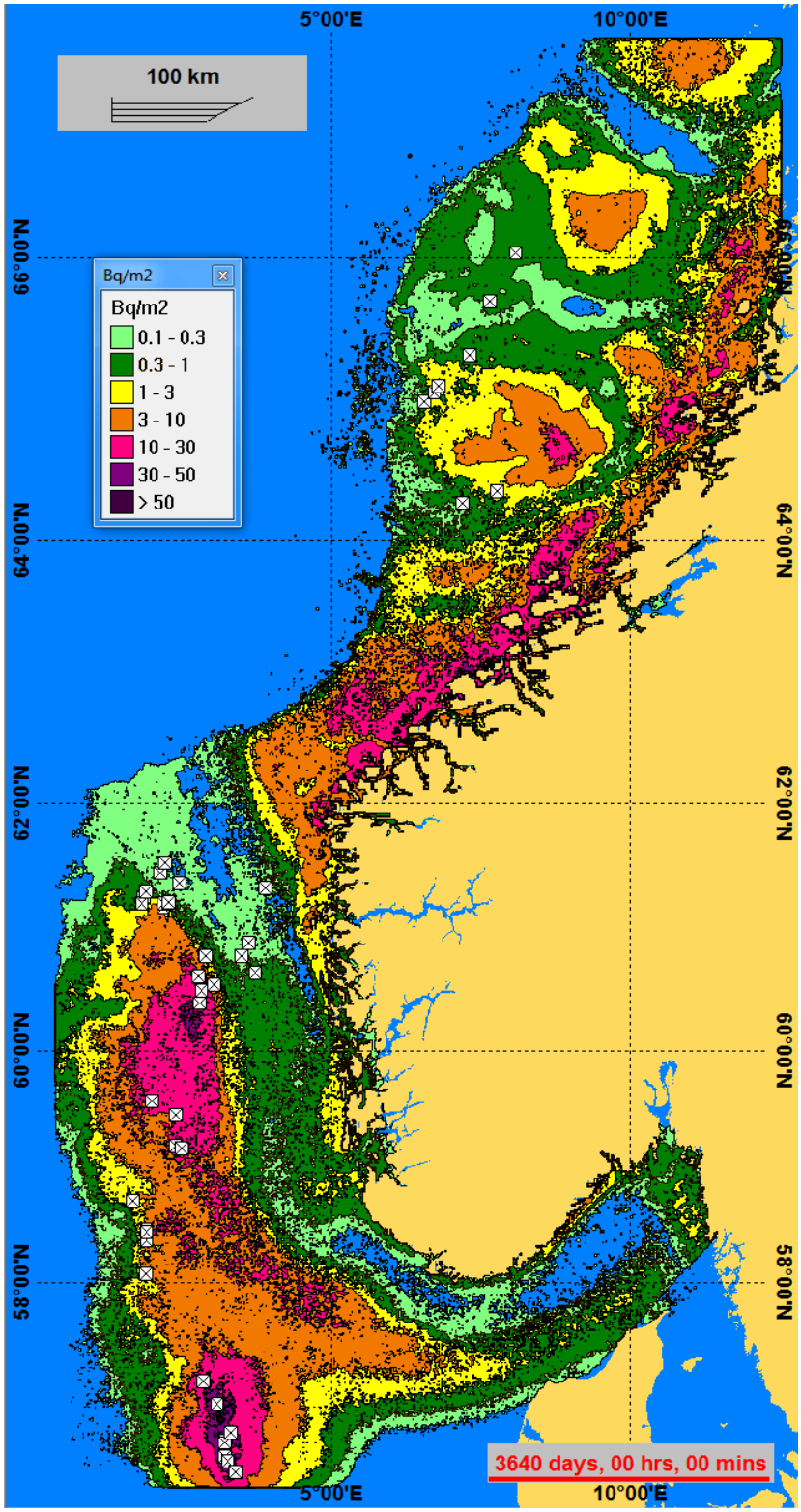

Figure 7: Simulation of dispersion of solid Ra-226 with resuspension after 10 years of discharge

For the simulations of Ra-226 bound within the lattice of $BaSO_4$ we first consider the simulation without resuspension. In this simulation there are three areas that show high accumulation of Ra-226, with values higher than 50 Bq/m^2 being reached after 10 years of discharge (manual inspection revealed the highest numbers to be close to 70 Bq/m^2). The first area is near the coast in mid-Norway (box A in Figure 6), the second is located at the Viking bank, south of the Oseberg field (box B in Figure 6), and the third in the area around the Ekofisk and Eldfisk fields in the south of the Norwegian North Sea sector (box C in Figure 6). These three areas represent two different dispersion scenarios. For the case of accumulation around the installations, the source of Ra-226 in the sediment is the surrounding installations; weak currents in the area, especially around Ekofisk and Eldfisk, cause a large fraction of the released particles to deposit within kilometers of the source. On the other hand, the accumulation along the coast (box A) represents Ra-226 that has been transported from a distance and enters the sediment when encountering the more shallow coastal sea bed.

For the simulations where resuspension is enabled a similar dispersion pattern is observed, but more scattered due to remobilization and secondary transport (Figure 7). In terms of the three main areas of accumulation observed in the scenario without resuspension (Figure 6), the off-shore concentrations around the installations remain similar, but the coastal concentration is reduced (Figure 7). This can be understood as there are larger bed shear forces in coastal areas due to tidal currents and waves. On the other hand, offshore seabed currents are normally weaker, and large waves associated with powerful storms are required to remobilize the seabed in the North Sea.

The reported values for Ra-226 in sediments have been given in units of Bq/m^2, but to compare with observation data it is necessary to convert this number into Bq/kg. The nature of this conversion will vary from location to location, depending on the amount burial of other organic and inorganic matter and sea bed bioturbation. The extent of burial and bioturbation are not known in general for the areas investigated here, so it is necessary to make use of simplifying assumptions. Using the same assumption as previously (Skancke *et al*. 2014) (10 cm bioturbation depth, a sediment porosity of 0.6 and density of 2.5 kg/L), one obtains a conversion from 1 Bq/m^2 to 0.01 Bq/kg since 10 cm of sediments would weight 100 kg. For the highest impacted areas with up to 70 Bq/m^2, we therefore reach a value of up to 0.7 Bq/kg activity concentration contributed by Ra-226 from produced water sources. If we scale this up to releases going on for 30 years, but assume the same bioturbation depth, we obtain a value of 2.1 Bq/kg dry matter. In an extreme case where bioturbation is only 2 cm, this number will be 10.5 Bq/kg. This number is lower than 15 Bq/kg which was estimated previously (Skancke *et al*. 2014), however we believe that the number reported here is a better estimate due to improved model resolution and longer simulation duration.

3.2 Comparison of dispersion of Ra-226 assuming releases from the sea surface

In the main model simulations (Table 2), all discharge sites have been placed at 30 meters depth, since the depth of discharge was unknown for the individual platforms. To investigate the effect of this choice on the model results, we repeated the simulation of the dispersion of solid Ra-226 but positioned all discharge sites at the sea surface. After 5 years, we stopped the simulation. Figure 8 compares the difference between 30 m and 0 m discharge depths. With 30 m discharge depth, there is slightly more sedimentation in the areas underneath the discharging installations, while the difference is less marked further away. There is therefore some sensitivity to the discharge depth of the produced water.

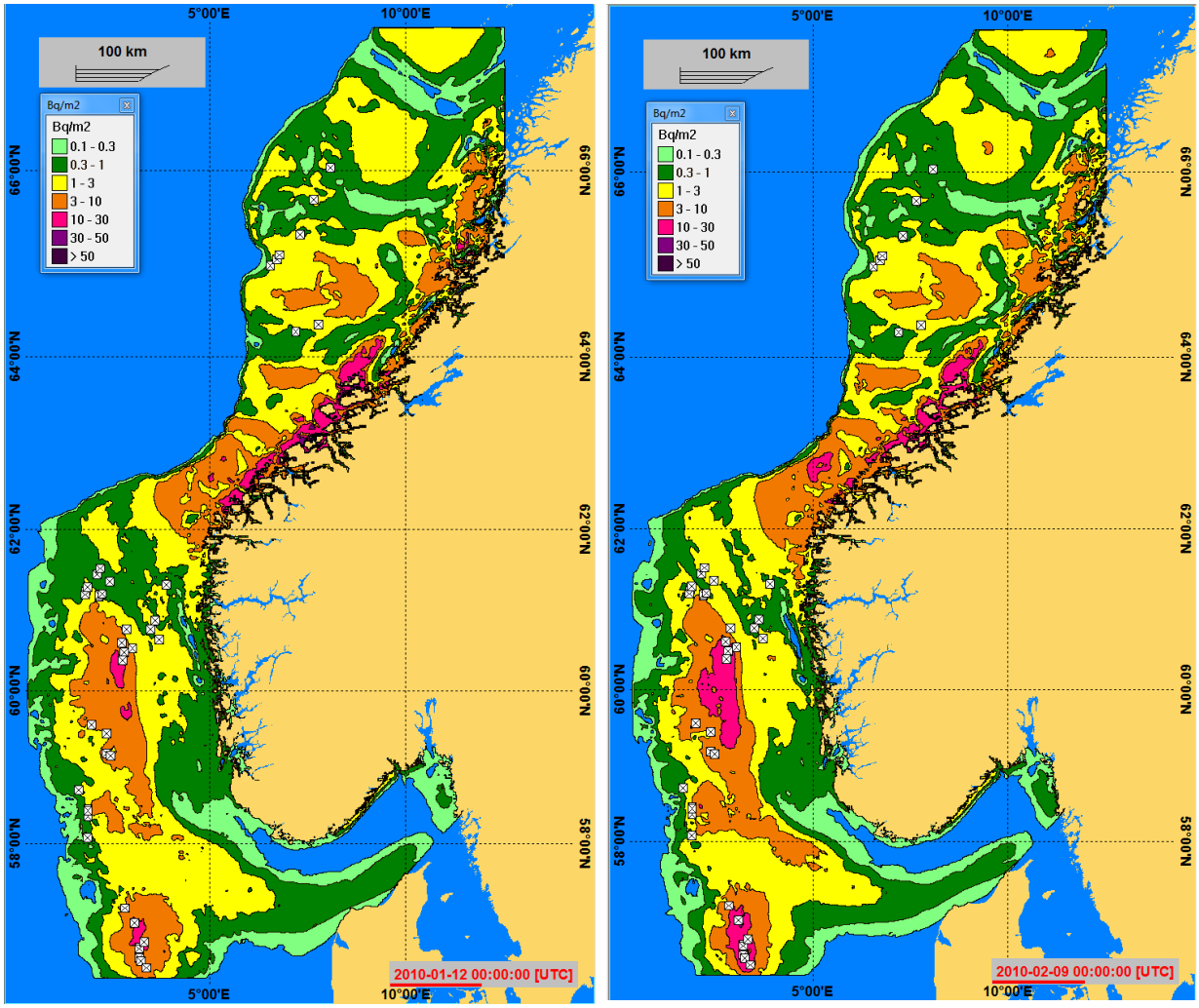

Figure 8: Comparison of discharges of Ra-226 as a solid from the sea surface (left) and from 30 meters depth (right). For the 30 meter discharges, more sedimentation occurs directly underneath the discharge sites.

3.3 Fate of dissolved Ra-226

Figure 9 shows a snapshot of concentrations in October 2014, after 20 years of continuous discharge. The primary transport of the particles is north along the Norwegian coast up past the west coast of Svalbard, however there is also some transport to the east of Svalbard and to the west of Svalbard through the Denmark Strait along the east coast of Greenland. In these locations, far from the source, the concentrations in the water column from discharged Ra-226 are very low, around 10^{-13} Bq/L. However, it illustrates that the model transports particles along the well-known Norwegian, Spitsbergen, and East Greenland currents. The highest concentrations in the water column were obtained along the Norwegian coast, close to the discharging installations, with maximum values reaching 10^{-9} Bq/L (Figure 9). This number must be considered in light of the grid cell size of 12 km x 12 km x 20 m. Previous modelling work using a higher resolution grid has shown that water column concentrations may be elevated with more than 1 Bq/L near discharging installations (Skancke *et al.*, 2014).

Dissolved Ra-226 may end up in the sea bed through adsorption to sediments or to sinking particulate matter. We investigated the potential for partitioning of dissolved Ra-226 to sediments using a conservative

method that transfers Ra-226 to sediments when a particle encounters the sea bed, equivalent to instant and irreversible adsorption. Sediment concentrations were calculated at the end of the 20 year simulation. This showed that dissolved Ra-226 was transferred to sediments in shallow areas where particles were transported to coast lines (Figure 10). As expected the highest concentrations were obtained along the Norwegian coast, shown in higher detail in Figure 11. At several locations concentrations up to 10 Bq/m^2 were obtained, with the highest concentrations reaching 20 Bq/m^2. Since most sediment transfer occurred in coastal waters, discharged Ra-226 does not reach the deep ocean on the time scale of these simulations. In addition to transfer to sediments in coastal areas, some Ra-226 ended up in shallow regions in the southern North Sea (Figure 11), where dispersion may transport these particles to the bed within the 20 year duration of the simulation.

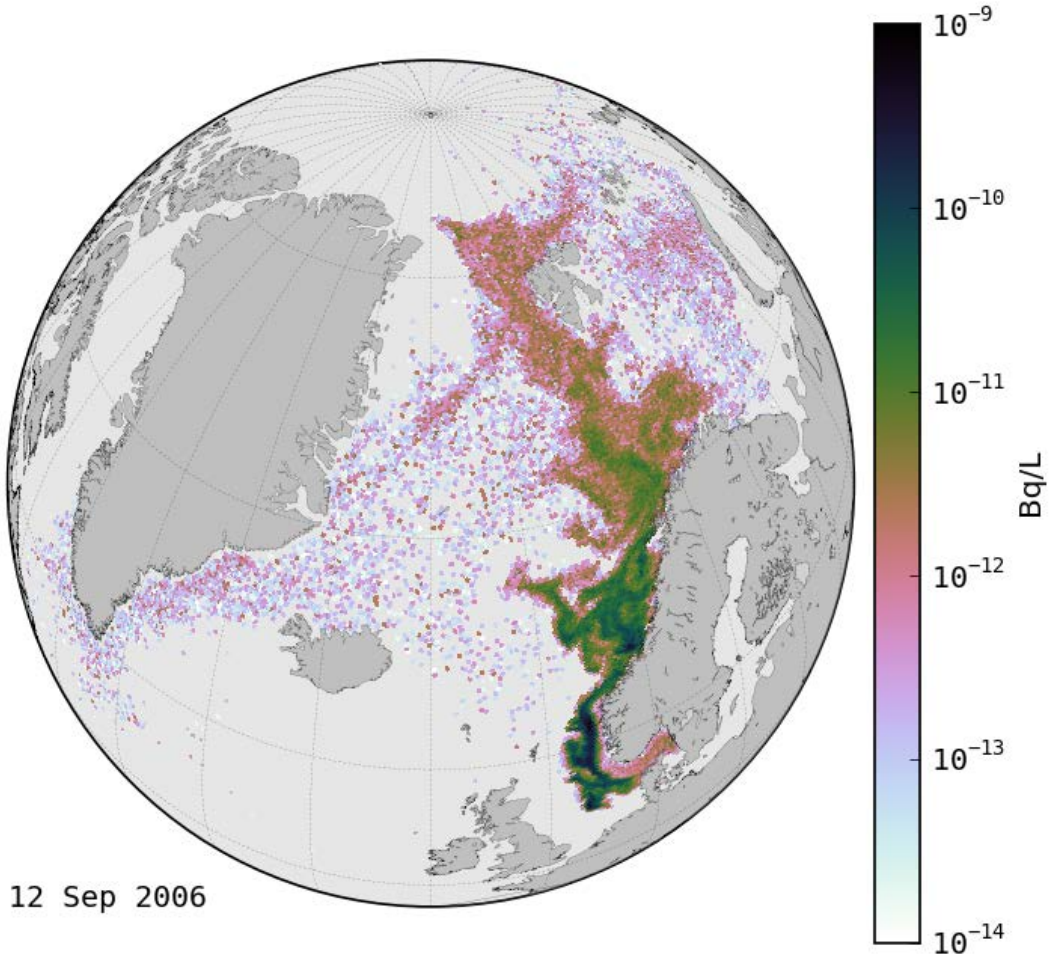

Figure 9: Snapshot showing concentrations of dissolved Ra-226 at 12 September 2006. Concentrations were calculated in cells of 12000 x 12000 x 20 meters, with the map showing the maximum concentration in the vertical for each location. 644915 particles were in circulation at this time.

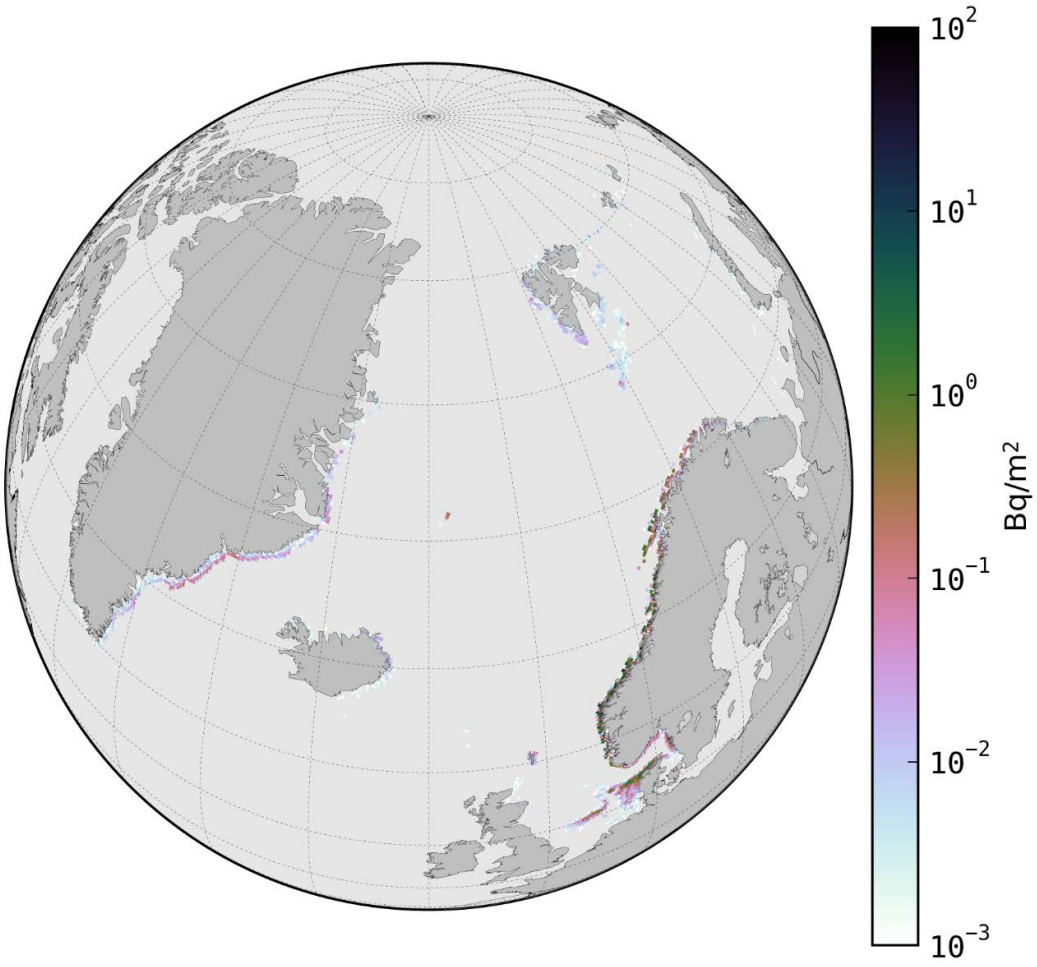

Figure 10. Sediment concentrations calculated after 20 years of simulation. Concentrations are calculated from all particles that were adsorbed by the sea floor on a grid using cell size 12000 x 12000 m.

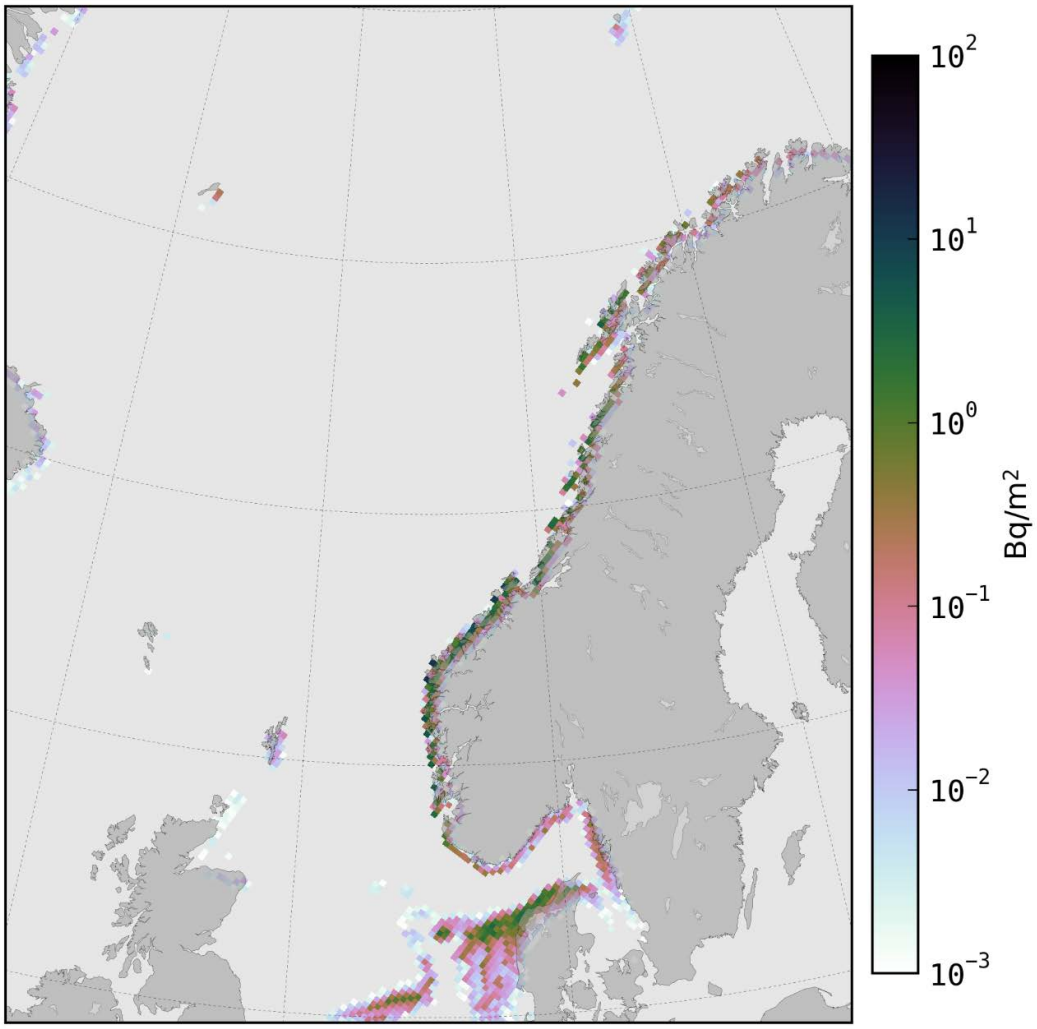

Figure 11. Sediment concentrations calculated after 20 years of simulation.

4 Discussion

In this work we have simulated the long-term fate of Ra-226 from produced water sources in both solid and dissolved form. When Ra-226 is released in solid form, we found that the highest accumulation occurs in areas near the discharging installations at the Oseberg and Ekofisk fields (Figure 7). High accumulation along the coast in mid-Norway was first seen in the simulation without resuspension (Figure 6), but this impact was reduced when resuspension was taken into account (Figure 7). The conclusion from these simulations is that the areas most likely to be experience accumulation of Ra-226 are the areas surrounding the aforementioned installations. For Ra-226 that spreads beyond these installations, dispersion in the water column and resuspension leads to much lower concentrations at the sea bed.

The concentrations of Ra-226 in the most impacted areas was found to be up to 70 Bq/m2 after 10 years of simulation. Extrapolated to 30 years of releases, and converted from Bq/m2 to Bq/kg, this may lead to activity concentrations in the area of 2 Bq/kg, or up to 10 Bq/kg in extreme cases, depending on the rate of sedimentation deposition and extent of mixing in the upper sediment layers (see Results). In comparison, the

average Ra-226 activity from sediment cores in the North Sea was found to lie in the range of 20-35 Bq/kg (Dowdall and Lepland, 2012). If these sediment cores represent natural background (see Skancke *et al*. 2014) this indicates that, given the model assumptions, the activity contribution of Ra-226 at the identified locations close to the Oseberg and Ekofisk/Eldfisk fields may reach up to within 1 – 10 % of the background concentration. However, it must be taken into account that the model-obtained numbers are averaged over the grid cell size of 1.5 km x 1.5 km. Depending on the local bathymetry and hydrodynamic conditions on the sub-gridsize scale, concentrations might be higher or lower at any one location within the area spanned by the grid cell.

Simulations of transport of Ra-226 in the dissolved phase showed long-term transport of Ra-226 from Norwegian offshore installations into the Arctic Ocean (Figure 9). The concentrations obtained in the water column this far from the discharging installations are very low (Figure 9). It is then of more interest to consider the potential for dissolved Ra-226 to remain adsorbed to seabed sediments. Using a conservative method, we calculated the potential for dissolved Ra-226 enter the sediments. Transfer to sediments was obtained primarily in coastal areas as particles encountered shallow coastal waters (Figure 10). The concentrations we obtained ranged up to 20 Bq/m^2, which is about 1/5th of the maximal value obtained from considering discharged Ra-226 in solid form after adjusting for the duration of the simulation (Figure 7). This corresponds to a contribution of 0.6 Bq/kg to 2 Bq/kg following the same assumption as for sedimentation of solid Ra-226. We note that the method applied is conservative in that it assumes instant and irreversible adsorption to sediments upon particle contact. None the less, the simulation demonstrates that transport with ocean currents and turbulence has the potential bring dissolved Ra-226 into contact with the seabed, primarily in coastal areas.

In considering the fate of discharged Ra-226 separately in dissolved and solid phase, the concentrations obtained in the two investigations represent each the largest potential impact from Ra-226 discharge, so the values are not additive. In order to assess the fate of Ra-226 as dissolved and solid matter simultaneously more work is required to identify the partitioning between these two phases. Specifically, it is necessary to assess the initial partitioning of Ra-226 between dissolved phase and the solid phases of free RaSO4 and RaSO4 bound in the lattice of BaSO4, and how this partitioning changes with dilution in seawater.

The simulations presented here demonstrate the usage of dynamic models to investigate the long-term fate of radionuclides discharged into the marine environment. Traditionally, the long-term fate of radionuclides, up to several thousand years, have been investigated using a box-model approach, where the marine environment is compartmentalized into boxes with prescribed flows between boxes, either assuming immediate or gradual dispersion of the released material in each box (see for example Iosjpe and Liland, 2012). When dynamic models have been used, the temporal and spatial resolution has typically been low (for example weekly averaged currents, as in Orre *et al*., 2007). Here we have shown that it is feasible to track radionuclides originating from all produced water sources on the NCS in the North Atlantic and Arctic oceans over 20 years using a 1 h model timestep with 12 km forcing data with more than one million tracking particles. Provided a long enough time-series of forcing data, it is feasible to study the transport of radionuclides using this model for up to 100 years, potentially covering the time starting from the first oil and gas installations in the North Sea and reaching up into the next several decades.

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