Enhanced levels of ^{226}Ra radiation in sea water and sediment caused by discharges of produced water on the Norwegian Continental Shelf

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Abstract. A numerical model has been applied to simulate the fate of ^{226}Ra radionuclides in the ocean, generated from discharges of produced water in the Norwegian Sector of the North Sea. The model simulates the transport, dilution and the (possible) deposition on the sea floor of ^{226}Ra. Figures are presented for the concentration levels and deposition rates of ^{226}Ra in the ambient for various assumptions for the fate of the ^{226}Ra in the ocean (as dissolved, as adsorbed to ambient organic particles that sink down on the sea floor, and as forming Ba(Ra)SO$_4$ particles). Concentration levels and deposition rates are compared with natural levels and fluxes of ^{226}Ra in the ocean and to the sediment, respectively. The added concentration levels/fluxes are found to be generally relatively small, compared to natural levels/fluxes. The reason for this is attributed to the presence of fairly high natural levels/fluxes of ^{226}Ra.

1. INTRODUCTION

The production of oil and gas offshore from beneath the ocean floor generates large quantities of produced water. Produced water is the aqueous fraction extracted along with oil and gas from geological formations. After separation from the oil and gas and subsequent treatment, produced water is either discharged to the sea of re-injected back into the reservoir. The discharges of produced water contain radionuclides that originate from dissolved metals in the oil/water/gas reservoirs. In order to study the potential environmental risk and doses to man, a research project was initiated by the Norwegian Research Council of Norway (NFR) termed "Radioactivity in Produced Water from Norwegian oil and gas installations – concentrations, bioavailability, and doses to marine biota". Measurements of concentrations of various radionuclides in produced water are presently carried out on the Norwegian offshore installations that discharge produced water. As a part of the NFR project, the fate of the nuclides in the ambient sea has been simulated with a numerical model. The purpose of the simulations was to reveal the magnitude of the added radiation levels, compared to the natural background. Produced water discharges contain radionuclides of various types. ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po are some of the most important ones with respect to potential environmental impact and doses to man. In the NFR project, the ^{226}Ra nuclide was considered to be the most important one. The reason for this is that it appears to be more abundant than the others in produced water [1], and also because it is an α emitter. The compound

[210]Po in the produced water may also pose a risk in the ambient sea as well, because this compound is a daughter of [226]Ra and is also an α emitter. In the present project, focus was directed on the compound [226]Ra only.

2. MATERIALS AND METHODS

The [226]Ra nuclides are generally present in the produced water stream in dissolved state (ionic form). Radium is chemically similar to Barium (Ba), which is also present (as dissolved) in the produced water stream in relatively large amounts. However, when discharged into the sea, the produced water will mix with sea water (which is rich in SO_4^{2-} ions) to form $BaSO_4$ (barium sulphate or barite) and $RaSO_4$. These reactions will transform Ba and Ra into solid state. However, dilution of the produced water will tend to slow down or reverse the particle formation process. Because the sea water (except for very deep waters) is generally unsaturated with barite, $BaSO_4$ will tend to dissolve again for sufficiently large dilutions [2]. Because radium is shown to behave similarly [3], the discharge of [226]Ra into the sea is therefore assumed to be present as partly particle and partly dissolved state. Also, the particle radium is expected to be present along with the $BaSO_4$ particles, and is therefore expected to be bound within the lattice of the $BaSO_4$ to a large extent.

Another process that can impact on the fate of [226]Ra in the recipient is the adsorbtion to organic (particle) matter naturally present in the recipient. Thus, three different paths can be envisaged for the fate of [226]Ra in the recipient after the radium has left the discharge pipe: 1) [226]Ra remains in dissolved state in the recipient and dilutes there along with other water soluble constituents in the discharges 2) [226]Ra remains in dissolved state and adsorbs to natural organic particle matter present in the recipient. The natural particles descend through the water column and down on the sea floor. 3) [226]Ra forms particle matter (together with Ba) consisting of $Ba(Ra)SO_4$ (barite) and then descends down on the sea floor as particles (sinking rates to be dependent on the particle size distribution).

The DREAM (*Dose Related Risks and Effects Assessment Model*) has been used to simulate the discharges of produced water and the resulting concentrations of [226]Ra in the recipient. This model calculates the ocean dilution and transport caused by oceanic turbulence (derived from winds and lateral spreading) and transport (based on output from the ocean current fields generated from three-dimensional and time variable hydrodynamic models). Concentrations of [226]Ra (in terms of Bq/L or mBq/L) are calculated as a function of time and space. The DREAM model also includes multiple sources. Depositions on the sea floor (fluxes) caused by the discharges of [226]Ra radionuclides are calculated by the model as well. The model is fully three-dimensional and includes time variability. Features of the DREAM model are described in [4].

An ocean current database for the North Sea area was used. This database contains outputs from ocean current simulations for the North Sea area for the years 1990–1994 simulated by *Det Norske Meteorologiske Institutt* (met.no) in Oslo, Norway. The time resolution used (for the output storage of the currents simulated) was 2 hours, and the horizontal resolution was 20 km. The depth resolution was variable, with the finest grid resolution closest to the sea surface. The ocean current model operated by (met.no) is denoted ECOM-3D and includes residual (time averaged) currents, tidal currents as well as currents generated from meteorological forcing (pressure and wind).

The North Sea area was selected for the simulations of the produced water discharges. This is the area on the Norwegian Continental Shelf (NCS) with the largest density and sizes on the produced water discharges. Only discharges on the Norwegian part of the shelf were included. The concentrations of the radionuclide [226]Ra for the year 2003 are used as input to the simulations, along with the amounts of produced water discharged for the same year. Total amounts discharged for the year 2003 have been estimated to be about 440 GBq (range 310–590 GBq) for [226]Ra, and 380 GBq (range 270–490 GBq) for [228]Ra. Total amounts of ^{210}Pb for 2003 was lower than 92 GBq. Average levels for [226]Ra and [228]Ra in produced water for 2003 was 3.3 and 2.8 Bq/L, respectively. Average levels of [210]Po in produced water

were all detected to be lower than 10 mBq/L [1]. Because ^{226}Ra was the most abundant component, this one was selected as input to the DREAM model simulations.

Laboratory experiments carried out at Institute for Energy Technology at Kjeller, Norway [5] showed that it was difficult to determine the specific fate of dissolved radium discharged to the ambient sea. Relevant factors were found to be the presence of chemicals in the discharge (slowing down the Ba(Ra)SO$_4$ formation process), the speed of the dilution process and the (speed and size of) particles formed. Therefore, it was agreed that the DREAM simulations to be carried out should include all three possibilities for the fate of ^{226}Ra in the recipient. Consequently, three calculations were made with the DREAM model. Each calculation is based on the assumption that the fate of ^{226}Ra is fully described by only one of the three paths outlined above (conservative assumptions). That is, all ^{226}Ra discharges are assumed to consist of either 1) All dissolved, or 2) All adsorbed to natural organic matter in the recipient, or 3) All forms RaSO$_4$ particles (along with BaSO$_4$) that then descends down on the sea floor. The reason for this approach is because it turned out to be difficult to give reliable numbers for the relevance of each of the three processes outlined above, relative to each other.

3. RESULTS AND DISCUSSION

The model was run for 14 months, starting at 1 January. The yearly variations in the ocean current and turbulence conditions (wind dependent) are therefore included in the simulations. Figure 1 (figure to the left) shows snapshot of the discharges during the initial (startup) phase of the simulation. The discharge water is still located close to the discharge points. Figure 1 (figure to the right) shows the concentration fields for the case where all ^{226}Ra in the discharge is assumed to remain dissolved after discharge. The figure shows a snapshot during summer (snapshot after about 7 months). The concentrations are generally lower than background level, except for some areas close to the discharge sites. Background levels are of order 1–2 mBq/L.

The results shown in Figure 2 represent deposition of ^{226}Ra on the sea floor after one year of discharge. Figure 2 (figure to the left) represents the accumulation of radionuclides on the sea floor caused by deposition of ambient organic matter (with dissolved radionuclides adsorbed to it) on the sea floor. Figure 2 (figure to the right) shows the deposition of nadionuclides in the form of Ba(Ra)SO$_4$ particles formed when the produced water discharges are released into the sea. The formation of the particles is due to dissolved barium in the discharge that mixes with sulphate (SO$_4$) to form BaSO$_4$, along with RaSO$_4$. The largest depositions of radiation occur in the Ekofisk area for both cases. The Ekofisk area is shown in the lower part in Figure 2. The reason for this is that the depths are relatively shallow in the Ekofisk area (about 70 m), combined with a rotating current pattern (tidal currents) in this area. These two effects cause the ^{226}Ra nuclides to remain in the area to a large extent, while they are sinking down to a relatively shallow sea floor. The sizes of the particles that are the carriers of the ^{226}Ra radiation activity are generally small with low sinking velocities. Laboratory experiments on the Ba(Ra)SO$_4$ particle formation gave an average particle size of about 8 μm when no chemical (a scale inhibitor) was added to the produced water stream [5]. When chemicals were added, the average particle size reduced to about 3 μm. A particle size distribution between 1 and 25 μm (average at 8 μm) was used in the simulations. The small particle sizes explain why the ^{226}Ra is carried away relatively long distances before they deposit on the sea floor.

Local maxima for the ^{226}Ra flux down on the sea floor is calculated to be about 10–12 Bq/m^2 and year for the Ba(Ra)SO$_4$ particle case (Figure 2, figure to the right) and about 1–3 Bq/m^2 and year for the adsorbed ^{226}Ra to the organic particles case (Figure 2, figure to the left). Although the ^{226}Ra in particle form deposits in larger rates than the adsorbed ^{226}Ra to organic particles, the absorbed radium will be more bioavailable. The ^{226}Ra present in the barite particles (BaSO$_4$) will be bound within the lattice of the barite particles. Estimates of the natural deposition rates of ^{226}Ra caused by the natural sedimentation of organic as well as inorganic particles were carried out as well. Also, the natural expected levels of dissolved ^{226}Ra in the sediment pore water were considered. Details can be found in [6]. The results
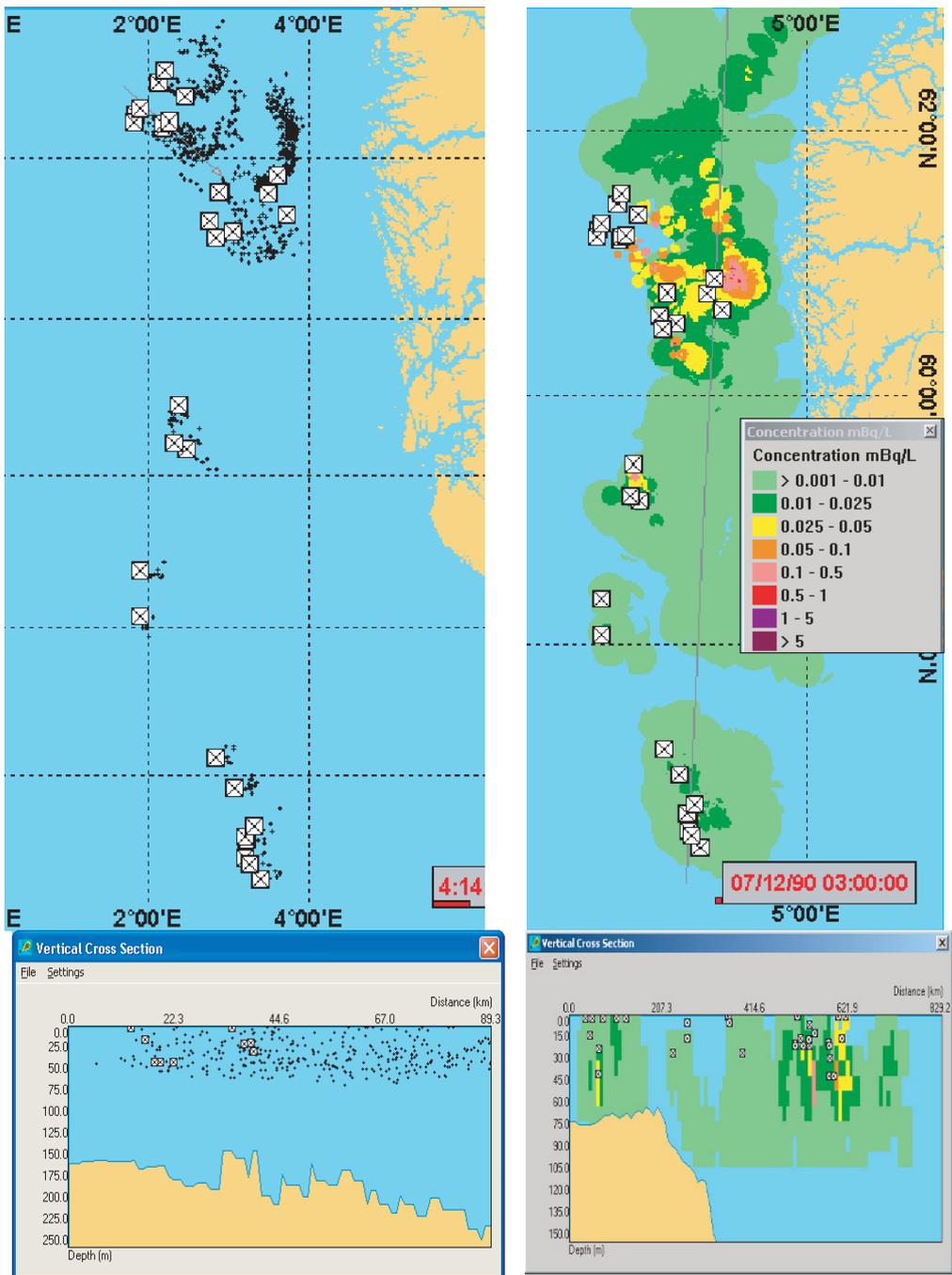
Figure 1. To the left: Discharge of produced water after some days of discharge. Model particles are representing the discharges. Vertical cross section below. To the right. Concentration field calculated for dissolved ^{226}Ra in the water column, assuming all ^{226}Ra to be completely dissolved. Vertical cross section below.
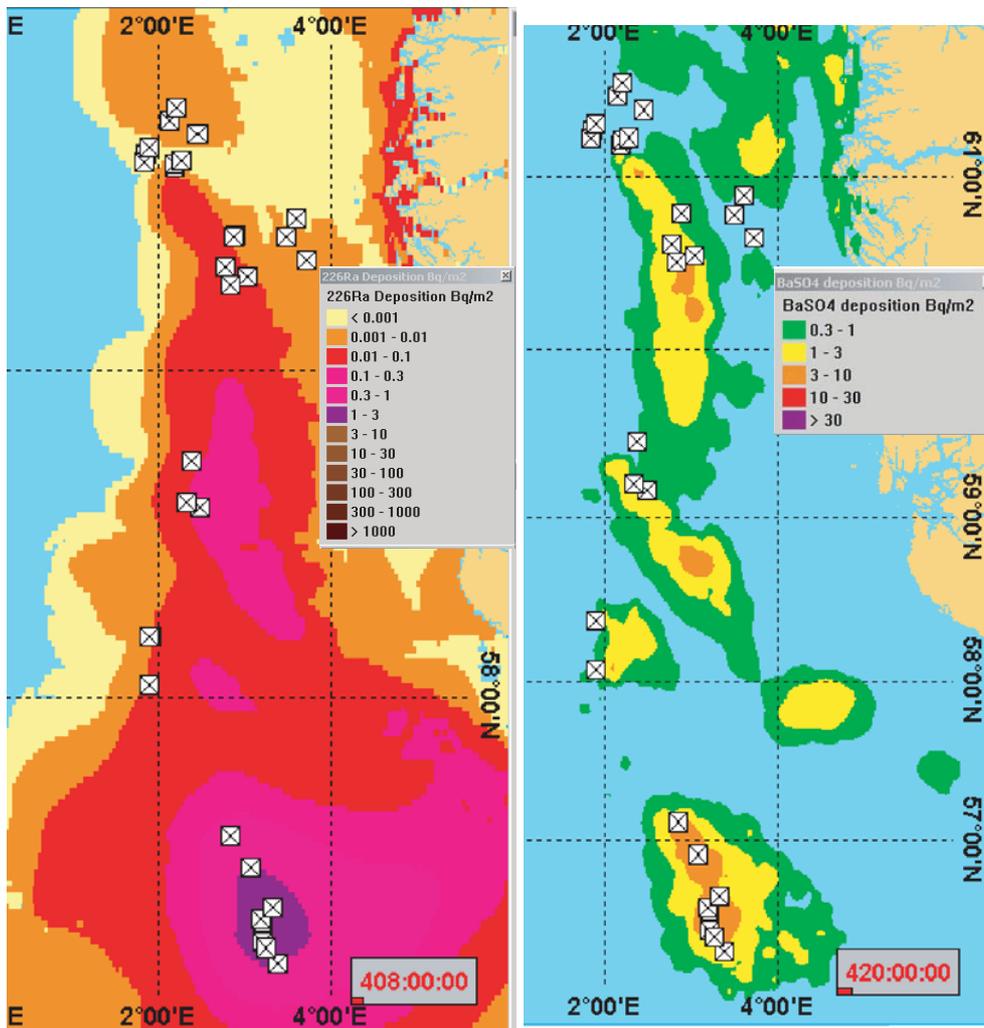
Figure 2. Calculation of deposition on the sea floor of [226]Ra nuclides with the associated radiation, given in Bq/m[2] over one year of discharge. Figure to the left: Deposition of [226]Ra (and associated radiation levels) in sediment caused by adsorption of dissolved [226]Ra to ambient organic matter in the recipient. Figure to the right: The deposition of [226]Ra on the sea floor caused by formation of $RaSO_4$ particles, jointly with the formation of $BaSO_4$ particles. Partition coefficient for Ra to organic matter in the water column is assumed to be 2000 L/kg (figure to the left).

from the study indicate that the added radiation levels on the sea floor caused by the added deposition of [226]Ra will be of limited significance, compared to other natural (and more bioavailable) sources and fluxes of [226]Ra radiation. Further details of the estimates carried out (including estimates of the natural fluxes of [226]Ra to and from sediment) can be found in [6].

Acknowledgments

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