North sea produced water PAH exposure and uptake in early life stages of Atlantic Cod
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A B S T R A C T
Produced water discharges from offshore oil and gas platforms represent a significant source of petroleum components such as polycyclic aromatic hydrocarbons (PAHs) released to the ocean. High molecular weight PAHs are persistent in the environment and have a potential for bioaccumulation, and the investigation of their fate and uptake pathways in marine life are relevant when assessing environmental risk of produced water discharges. To study the exposure and uptake of 2–5 ring PAHs in early life stages of Atlantic Cod in the North Sea, we run a coupled fate and individual-based numerical model that includes discharges from 26 platforms. We consider 26 different PAH components in produced water which biodegrade with primary degradation rates; intermediate degradation products are not included. Model simulations are run covering multiple years (2009–2012) to study annual exposure variability, while a one-day time slice of spawning products from the peak spawning season are followed. By covering multiple release points and large spatio-temporal scales, we show how individuals can be exposed to produced water from multiple regions in the North Sea. We find that a combination of oceanic fate processes and toxicokinetics lead to markedly different compositions in the predicted internal concentrations of PAHs compared to discharge concentrations; for instance, naphthalene makes up 30% of the total discharged PAHs, but contributes to at most 1% of internal concentrations. In all simulations we find the predicted total internal PAH concentration (26 components) to be below 1.2 nmol/g, a factor of 1000 less than concentrations commonly associated with acute narcotic effects.

1. Introduction
Produced water (PW) discharges from offshore oil and gas production represent the largest discharge to the marine environment worldwide, with approximately $1.3 \times 10^8$ m$^3$ released annually on the Norwegian continental shelf (NCS). PW contains a mixture of formation water, oil, gas, injected fresh/brine water and added production chemicals. The formation water contains a mixture of dissolved inorganic and organic components, and the composition varies between different reservoirs (Neff et al., 2011). Total PW discharges from activities on the NCS in 2017 included 129 tons polycyclic aromatic hydrocarbons (PAHs) (NOROG, 2018). Despite being a small fraction (0.306%) of the total composition of produced water, PAHs are considered a key risk component (Beyer et al., 2020). These PAHs can be dissolved in the water, present in dispersed oil droplets and/or adsorbed to particulates (Faksness et al., 2004). Different PAHs display varying lifetimes and biological uptake properties in the ocean. In general biodegradation rates tend to decrease with increasing molecular weight of PAHs (Loftus et al., 2018), and the potential for bioaccumulation (bioconcentration) tend to increase with molecular weight (De Hoop et al., 2013).

The Atlantic cod (Gadus morhua) is found throughout the North-ern Atlantic Ocean where it has been one of the most commercially important species for centuries (Otterå, 2004). Cod populations in the North Sea have declined (Brander, 2005), and it is labelled vulnerable on the IUCN Red List of Threatened species. Apart from overfishing, recruitment failure is generally believed to be a main contributor to the decline, the causes of which are complex (Husserbråten et al., 2018; Akimova et al., 2019). Important spawning grounds for Atlantic cod in the North and Norwegian Seas are found in vicinity of oil production fields with produced water discharges (see Fig. 1 and Akimova et al., 2019; Sundby et al., 2017). The spawning season for cod in North Sea extends from January to May, with a peak in February and March (Sundby et al., 2017), and the eggs develop over a period of approximately 100 degree-days before hatching (Fraser et al., 1988). After fertilization, eggs are positively buoyant in seawater and are therefore found in the upper water masses. After hatching the larvae drift with ocean currents and perform vertical migration (Vikebø et al., 2007).

Exposure of early developmental life stages (ELS) to petrogenic compounds has shown to cause acute toxicity as well as a range of sub-lethal effects. Atlantic cod and Atlantic haddock (Melanogrammus aeglefinus) embryos exposed to oil dispersions showed craniofacial deformations in hatched larvae. This occurred for exposure concentrations above 0.76 (haddock) and 2.8 (cod) μg TPAH/L, and associated with TPAH internal concentrations of 542 ng/g (wet weight) for cod (Sørensen et al., 2017). In another study, cardiotoxicity and severe craniofacial deformations were observed in cod larvae after embryonic exposure to PW extracts corresponding to 50-500-fold dilutions of PW (ranging 2.5–34 μg TPAH/L) (Hansen et al., 2019). These studies have only measured

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a sub-set of components (PAHs and alkylated homologues) present in the oil dispersions and PW, and the contribution of non-quantified components to toxicity has not been determined. Studies with single PAH components have also shown embryotoxicity; a study with phenanthrene exposure (45–85 μg/L) resulted in severe deformations, with body residues in the range of 20–30 μg/g (wet weight) (Sørensen et al., 2019).

In this study, the DREAM model (Reed and Hetland, 2002; Reed and Rye, 2011) was used to evaluate the potential for bioaccumulation of PAHs from PW in cod populations ELS. DREAM is routinely used to predict the physical transport and fate of produced water, and in risk assessment by calculating ratio of predicted environmental concentration (PEC) and predicted no effect concentration (PNEC) of individual PW substances (see e.g. Smit et al., 2011; Beyer et al., 2012). Several studies have also compared predictions from the model with field measurement (Durell et al., 2006; Niu et al., 2016).

Given the temporal and spatial distribution of concentrations in a PW plume, impact on marine life can be assessed through coupling with individual-based models (IBM) (Grimm and Railsback, 2005; Scheffer et al., 1995) and toxicokinetic models (Mccarthy and Mackay, 1993; Hendriks et al., 2001; Jager et al., 2011). Whereas the individual-based models are used to model the behaviour of population individuals or groups of these, toxicokinetic models are used to predict internal concentrations given the external concentration of components in the PW plume. The resulting internal concentrations, also known as body residues or body burdens, are more closely related to the effective dose than the environmental exposure concentrations, and are thus more relevant for predicting effects and environmental risks (Meador et al., 2008; Ashauer and Escher, 2010; Jager et al., 2011). Internal concentrations can also be determined for oceanic organisms sampled in the field (see e.g. Durell et al., 2006; Hansen et al., 2020), which may potentially be useful for comparison with model predictions.

2. Materials and method

2.1. Study area and produced water discharge data

PW discharge concentration profiles containing 26 different PAH components from 26 distinct offshore platforms in the North Sea were used as input to the simulations (Fig. 1). A list of the individual PAH components and their relevant physico-chemical properties, concentrations in the discharge, and details on each release site are given in the Supplementary Information. Site-specific release rates, varying from 3000–66 000 m$^3$/day, and composition profiles for each platform are shown in Fig. 2. These were obtained from the Norwegian Environment Agency based on annual reports from oil and gas operators (see e.g. Equinor, 2018). Release rates represent a yearly average rate, and the depths of the discharges vary from 1 to 40 m. The geographic positions of the offshore platforms are based on data from the Norwegian Petroleum Directorate fact pages. An overview of the study area, showing release sites and spawning grounds, is shown in Fig. 1 where the release sources have been grouped within three different regions marked: Tampen, Utsira High and Ekofisk. The named component groups used in the figure are naphthalenes (naphthalene and C1–C3 alkyl homologs), phenanthrenes (phenanthrene and C1–C3 alkyl homologs) and dibenzothiophenes (dibenzothiophene and C1–C3 alkyl homologs), while the remaining 14 components are included in
the “Rest” category. The three named groups constitute the majority of the PAHs in the discharge, and are the most relevant for analysing internal concentrations. In the discussion of simulation results, the full 26 components are referred to as total PAH, in the sense of total PAH in the simulated discharge. A complete list of names and properties of the PAHs included in the simulations is given in Table 2 in the Supplementary Information.

2.2. Ocean circulation data

Simulating advective transport of contaminants and biological individuals require an external circulation field. To investigate the potential variation in exposure in different years, we used an archive of ocean circulation model data covering the model domain over four years (2009–2012), with a horizontal resolution of 4 km. Monthly mean surface currents from the archive for March 2009 are shown in Fig. 1. This archive was produced with the hydrodynamic component of the SINMOD model system, which is based on the primitive Navier-Stokes equations (Slagstad and McClimans, 2005). SINMOD uses a z-coordinate grid, and in the present archive uses 33 vertical levels, with layer thicknesses ranging from 5 m in the upper layers (down to 40 m depth), increasing to 50 m in the intermediate layers (100 m–1000 m depth), then to 500 m in deeper layers (below 1000 m depth) (Alver et al., 2016).

2.3. Produced water transport and fate

Transport of produced water in DREAM is handled by a Lagrangian particle approach (DREAM, Reed and Hetland, 2002; Reed and Rye, 2011) which tracks each discharge component in the receiving environment. The produced water release does not include dispersed oil droplets, treating the discharge as a release of purely dissolved compounds. Eulerian concentration grids are calculated from the mass and distribution of the particles which provides the input to the exposure model. Two resolution levels of grids are used: near the discharge droplets, treating the discharge as a release of purely dissolved compounds. Eulerian concentration grids are calculated from the mass and distribution of the particles which provides the input to the exposure model. Two resolution levels of grids are used: near the discharge sources 1 km x 1 km cell size grids are used for improved resolution, corresponding approximately to the regional outlines (dotted lines) in Fig. 1), while the whole computational domain (dashed outline in Fig. 1) is covered by a second, coarser grid (4 km x 4 km). To cover the peak spawning season in the North Sea the model was run for the months of March, April and May for each of the years from 2009 to 2012.

In addition to advection and turbulent diffusion, the model calculates bacterial degradation of PAHs based on first-order kinetics and primary biodegradation reference rates (Loftus et al., 2018) given in Table 5 in the Supplementary Information. By using primary degradation rates we track fate of the original PW components in the water column, and do not follow further degradation products. Each PW component in is associated with a specific biodegradation rate. For the component groups that have been used, half lives vary from a few days for the low molecular weight compounds to over a year for the heaviest compounds. Water temperature affects the biodegradation rates, which is accounted for by scaling the rates according to the Q10-approach (Bagni et al., 2013), using the temperature at the location of each Lagrangian particle. In the present simulations, a constant sea water temperature of 10 °C was used. A constant temperature was selected to focus the analysis on the variation of ocean currents between the chosen months and years in the study. The temperature of 10 °C is in the high end but within observed values for March, April and May in the North Sea. Another fate outcome for produced water is evaporation through the sea surface. Evaporation is included by default in DREAM, but contributes to a negligible fraction of produced water’s mass balance.

To represent the produced water discharges in the simulations a maximum total number of 70 000 particles was used, with 10 particles being released from each discharge site in each time step, and the main model time step was 15 set to minutes. Sub-grid transport and mixing not resolved by the ocean model are typically accounted for by adding a horizontal diffusivity in the particle model. The value of this parameter depends on the resolution of the ocean model, with higher values typically used for coarser models. Different values have been reported in recent model studies: 10 m²/s was used in studies of the Deep Water Horizon oil spill (Boufadel et al., 2014; Berenshtein et al., 2020), while Simonsen et al. (2017) experimented with different horizontal diffusivity values (0 and 100 m²/s) in a 4 km ocean model setup in a study of radionuclide tracer transport in the North Sea and found that it had only minor impacts on model skill. In the present study we used a fixed horizontal diffusivity value of 50 m²/s, which is comparable to median values calculated on the ocean model flow field with the Smagorinsky method, which accounts for resolution and shear in the mean flow field (Smagorinsky, 1963). A sensitivity test was also performed on this value, which is given in the Supplementary Information.

The vertical diffusivity was set to a constant value 10⁻⁴ m²/s, which corresponds to low vertical mixing conditions. Higher values of vertical diffusivity, caused by e.g. strong winds, can occur near the surface, and would lead to increased dilution and lower concentrations. Deeper in the water column lower diffusivity values are generally expected, with a large seasonal variation in the surface mixed layer in the North Sea. Uniform values have been used in previous model studies, such as Paris et al. (2012), where the Macondo oil well blowout was modelled using a fixed diffusivity value of 10⁻⁵ m²/s, although focusing on deeper waters than here (around 1000 m). Differences between uniform and depth-varying profiles of vertical diffusivities have been explored by several authors, for some recent in-depth discussions see e.g. Nordam et al. (2019), Boufadel et al. (2020). See Supplementary Information for sensitivity analysis on the effect of choice of vertical diffusivity parametrization.

2.4. Individual-based model and toxicokinetics

Individuals are modelled as “biological particles” which are transported by ocean currents and experience growth and uptake and depuration of produced water components. In this section, we give a description of the parameters and processes that describe and affect the
The produced water environmental concentrations tend to be highest near the point sources with the largest emissions, and most of these are found in the Tampen region (cf. Fig. 2). Consequently this is where the potential is highest for PW exposure to super-individuals, as seen in Fig. 4 from the areas indicating where concentration levels above 20 ng/L total PAH can occur (time- and depth-maximum). While these areas are largest around Tampen, smaller areas are also found around Utøya and Ekofisk. At the end of the simulations (all years), about 86% by mass of the total discharged PAHs has undergone primary biodegradation, while 0.1–0.5% or less has been advected outside the model domain.
Following spawning, the super-individuals representing eggs are transported with currents, as shown in Fig. 4, where the colour coding indicates the spawning ground origin. For each spawning ground 100 individual trajectories are shown, corresponding to those super-individuals which incurred the highest total internal concentration. Based on the prevailing patterns in the circulation (Fig. 1), super-individuals from the same spawning ground have common patterns in their trajectories. From the southernmost spawning ground (Fisher Bank), most super-individuals are transported eastward, away from the platforms in the Ekofisk region, into the Skagerrak “loop” (Huserbråten et al., 2018), and end up in the Norwegian coastal current, being eventually transported northward. The super-individuals from the Ling Bank are transported northward, with some intersecting the Utsira High region. Finally, the Viking Bank super-individuals, starting out in the Tampen region, tend to follow northward trajectories, entraining into the North Atlantic current. This transport pattern is seen in the period 2009–2011. However, transport in 2012 is different, where there is a general advection of super-individuals in a south–southwest direction. In this year, super-individuals from VB end up in the Utsira High region, and super-individuals from LB end up in the Skagerrak loop, while there is little net transport from FB, and the end positions remain in the vicinity of the spawning ground. This can be seen in Fig. 4, where the left pane shows trajectories for 2010 as an example of the 2009 to 2011 years, and the right pane shows trajectories for 2012. Inset figures in Fig. 4 show the depth of bioparticles as a function of time. Vertical turbulent diffusivity is not high enough to overcome positive buoyancy in the egg stage, but causes gradual vertical mixing once the particles reach the larvae stage (see Supplementary Information for a sensitivity analysis of vertical diffusivity on model results).

In terms of uptake of PW components, we find that total internal PAH concentrations vary significantly between individuals, covering several orders of magnitude, both in the egg and larval stages. To quantify the overall population internal total PAH concentrations (26 components), we calculate summary statistics over all super-individuals, shown in Fig. 5. Statistical summary values for this figure may be found in Table 2 in the Supplementary Information. Here, the median and 95-percentile values (top and middle panel in Fig. 5) provide the total molar internal PAH concentration level below which 50% and 95% of the super-individuals fall, respectively. The highest inter-annual variability is found in the population-median value (upper panel), as indicated by the shaded region, covering one standard deviation from the simulations mean (over different years). We observe that the 95-percentiles (middle panel) attain maximum values at the end of the egg stage (around simulation day 25), for all years, and that the values are quite similar between years. Since the equilibrium value of internal concentrations, related to the partition coefficient $P_{\text{lw}}$, is higher for the egg stage than the larval stage due to differences in lipid fraction, it is plausible that just such a maximum can occur during the egg stage. Its position at the end of the stage following a continuous increase, when lipid fraction change in the super-individuals, further indicate that equilibrium was not reached. The figure also shows the maximum total internal PAH concentration reached among all super-individuals (lower panel), which in all scenarios is less than 1.24 nmol/g. These values are the highest observed over the entire population at any given time, and may originate from different super-individuals at different times. As such, they are more prone to random fluctuations (super-individual diffusive transport is a random process), and too much should no be read into the specific time development of the curves, but they do provide an upper bound on the observed internal concentration values.
The relative compositions of total internal PAH concentrations are shown in Fig. 6, summed into three different component groups, which combined make up more that 98% of the total internal concentrations. For reference, the total PAH discharge composition is shown, similarly grouped; also in this case the three groups constitute more than 98% of the total. The vertical black dashed lines delineate egg spawning start (leftmost line) and end of the egg stage (rightmost line) for the super-individuals. Top panel: population-median total internal PAH concentration. Middle panel: population 95-percentile total internal PAH concentration; 95% of super-individuals have a total internal concentration less than this. Lower panel: population-maximum total internal PAH concentration.

Fig. 5. Population total internal PAH concentration percentiles and maximum values over time, for different simulation years. The black lines are mean values taken over simulation years, while the shaded areas are ± one standard deviation from that mean. The vertical black dashed lines delineate egg spawning start (leftmost line) and end of the egg stage (rightmost line) for the super-individuals. Top panel: population-median total internal PAH concentration. Middle panel: population 95-percentile total internal PAH concentration; 95% of super-individuals have a total internal concentration less than this. Lower panel: population-maximum total internal PAH concentration.

4. Discussion

A large part of the total produced water discharge undergoes bacterial degradation (biodegradation) in the simulations, with only 14% of the mass of the original components remaining after 90 days simulation time (not shown). Since biodegradation rates tend to decrease with increasing molecular size, smaller components such as naphthalene were rapidly degraded (see Supplementary Information Table 1 and Lofthus et al., 2018). This makes the biodegradation process an important regulator of individual exposure concentrations. A biodegradable organic component will eventually turn into water and carbon dioxide through a degradation pathway that includes intermediate products. In this investigation the intermediate products are not explicitly tracked: primary (first step) degradation rates are used as input, and biodegraded masses are removed from the simulation, thus concentrations of intermediate degradation products are not considered. Heavier components (e.g. chrysene), which have lower biodegradation rates, will persist longer in the water column, and become more dominating in the exposure concentrations at greater distance from the discharge points. Conversely, the lighter components (e.g. naphthalene) occur mostly near the discharge points, due to their higher biodegradation rates.

In our simulations we released and followed spawning products from a single day. However, we expect that our conclusions will hold also for other dates within the peak spawning period, since the period large-scale transport patterns are broadly similar, and the PW discharge rates are constant. We also find that inter-year variability in the population internal concentration statistics is not very large. While the transport patterns for the 2009–2011 simulations are similar, the 2012 simulation has a markedly different pattern (Fig. 4), but this difference is not reflected in the general population internal concentrations 95-percentile (middle panel) and maximum values (lower panel) shown in Fig. 5. The population-median (upper panel) does show more variability, with a difference of almost an order of magnitude between highest and lowest at the end of the egg stage. It may be that since there are large single emission sources in each of the regions, similar internal concentrations levels can result from different transport pathways as PW plumes and spawning products are transported together. We note here that the super-individuals with the highest internal concentrations at the end of the simulation are not concentrated in a small region, but rather spread out (see Fig. 4). In terms of originating spawning ground, it is the sub-population from VB (Tampen area, blue in Fig. 4) that have the highest internal concentration (data not shown), which perhaps not surprising due to the greater number of high volume PW discharge sources in this area, and partial overlap of these with the spawning ground.

Previous studies of produced water discharges in the North Sea have found that environmental risks associated with these discharges are largely local, restricted to the immediate vicinity of the emission source (see e.g. Hylland et al., 2008; Brooks et al., 2011; Durell et al., 2006; Bakke et al., 2013). In a model study focusing on the alkylphenol components of produced water, Beyer et al. (2012) found that the environmental exposure concentration and internal concentrations (C2 – C3 and Cn+3 components) were too low to have an impact on the reproductive health of three different species of fish. From the bi-annual monitoring program conducted on the NCS, total internal PAH concentration in caged blue mussels placed near a produce water discharge (Tampen region) was in one study found to be up to 205 ng/g (w.w.) (Durell et al., 2006). A more recent iteration of the program included copepod sampling in the Tampen area, and found a mean total internal PAH concentration of 231 ± 102 ng/g (w.w.) (Hansen et al., 2020). The present simulations show a maximum value of 198 ng/g (1.2 nmol/g), and the 95-percentile values are all below 27 ng/g (0.15 nmol/g). Due to the difference in species (mussels, copepods,
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5. Conclusion

We have developed a toxicokinetic-individual-based model coupled to an existing transport and fate model (DREAM), and used it to study uptake of polyaromatic hydrocarbons from produced water sources in the North Sea (Norwegian side) in early life stages of Atlantic cod. The highest levels of total internal PAH concentration we find are 1.2 nmol/g (198 ng/g), with 95-percentiles generally below 0.15 nmol/g (27 ng/g), which are below thresholds that are expected to cause chronic effects. Inter-annual variations are found in the transport patterns of the eggs and larvae, but the population-level internal concentrations remain similar between the years. A large variation in relative composition of internal concentrations compared to discharges are found, demonstrating the importance of toxicokinetic models and their usefulness as a building block in assessment of environmental effects and risk.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.marenvres.2020.105203.

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CRediT authorship contribution statement

Raymond Nepstad: Conceptualization, Methodology, Software, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization, Project administration, Funding acquisition.

Bjørn Henrik Hansen: Conceptualization, Writing - original draft, Writing - review & editing.

Jørgen Skancke: Conceptualization, Data curation, Writing - original draft, 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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