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Mind the gap – Relevant design for laboratory oil exposure of fish as informed by a numerical impact assessment model

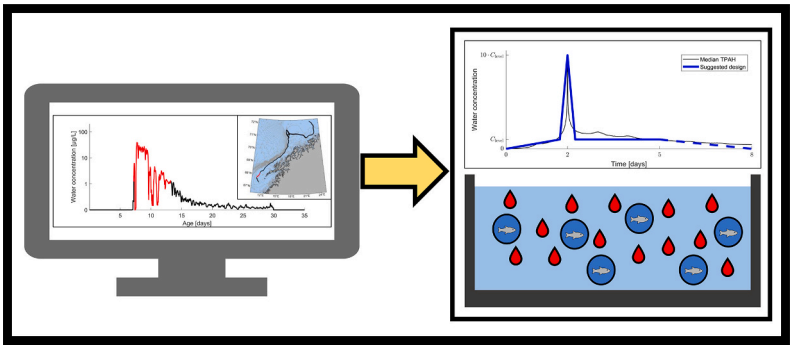
Håvard G. Frøysa [a,*], Raymond Nepstad [b], Sonnich Meier [a], Carey Donald [a], Elin Sørhus [a], Mathias Bockwoldt [c], JoLynn Carroll [c,d], Frode B. Vikebø [a,e]

[a] Institute of Marine Research, PO Box 1870, Nordnes, 5817 Bergen, Norway
[b] SINTEF Ocean, PO Box 4762, Torgarden, 7465 Trondheim, Norway
[c] Department of Geosciences, UiT The Arctic University of Norway, 9037 Tromsø, Norway
[d] Akvaplan-Niva, FRAM – High North Research Centre for Climate and the Environment, 9296 Tromsø, Norway
[e] Geophysical Institute, University of Bergen, PO Box 7830, 5020 Bergen, Norway

HIGHLIGHTS

- Modelling of fish early life stages exposed to a large oil spill
- Individual concentrations have high temporal variability but distinct properties.
- Suggestion of a new laboratory exposure design

GRAPHICAL ABSTRACT



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ABSTRACT

Laboratory experiments provide knowledge of species-specific effects thresholds that are used to parameterize impact assessment models of oil contamination on marine ecosystems. Such experiments typically place individuals of species and life stages in tanks with different contaminant concentrations. Exposure concentrations are usually fixed, and the individuals experience a shock treatment being moved from clean water directly into contaminated water and then back to clean water. In this study, we use a coupled numerical model that simulates ocean currents and state, oil dispersal and fate, and early life stages of fish to quantify oil exposure histories, specifically addressing oil spill scenarios of high rates and long durations. By including uptake modelling we also investigate the potential of buffering transient high peaks in exposure. Our simulation results are the basis for a recommendation on the design of laboratory experiments to improve impact assessment model development and parameterization. We recommend an exposure profile with three main phases: i) a gradual increase in concentration, ii) a transient peak that is well above the subsequent level, and iii) a plateau of fixed concentration lasting ~3 days. In addition, a fourth phase with a slow decrease may be added.

* Corresponding author.
E-mail address: haavard.guldbrandsen.froeysa@hi.no (H.G. Frøysa).

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1. Introduction

Petroleum exploitation comes with the risk of accidental oil spills in the marine environment (Keramea et al., 2021). Furthermore, routine discharges of produced water containing oil components, metals, and a variety of proprietary synthetic compounds represent a continuous contamination source (Nepstad et al., 2021). Risk assessments are carried out both prior to licensing for exploration drilling and during the development of wells (Hodson et al., 2019). A risk assessment involves identification of ecologically and economically valuable marine species, typically including early life stages (ELS) of fish, corals, invertebrates, seabirds and mammals, and a quantification of individual to population losses under various oil spill scenarios (Hjermann et al., 2007; Beyer et al., 2016; Langangen et al., 2017).

Advanced numerical models can be applied to quantify the effects of oil exposure on species-specific stages (Carroll et al., 2018; Carroll et al., 2022; Vikebø et al., 2015) relying on laboratory established LC50 thresholds (Hansen et al., 2021; Sørhus et al., 2021). Uncertainties in these models arise from simplifications because of our limited understanding of processes involved, computational limitations in resolving relevant processes, and limited observational data needed to initialize and run such models.

Combining measured individual responses studied in laboratories with numerical models for population effects is not straightforward. Hodson et al. (2019) conclude that laboratory test methods must be improved, particularly regarding control of exposure concentrations and the particle to dissolved ratio. It is, for example, inaccurate to report a static or semi-static exposure regime as a fixed concentration as these regimes result in a time-varying concentration. And even if these challenges were resolved, e.g. with methods and protocol standardization (Bejarano et al., 2023), the number of replicates and variations in exposure trials are limited. Ideally, it would be desirable to have an experimental design with variation in environmental variables (e.g. temperature and UV radiation) and timing of exposure relative to species stage development in addition to various exposure dynamics and concentrations, but this would require a significant increase of laboratory capacity. Hence, the quantitative results from discrete exposure regimes available for inclusion into impact assessment models limit our ability to develop accurate models for varying exposure scenarios.

A key element of an oil spill impact assessment model is consideration of exposure effects. This involves uptake kinetics, effects thresholds and impacts including both lethal and sub-lethal effects. Lethal effects are quantified as survival probability. Sub-lethal effects may be a diverse composition of reduced functionalities such as the ability to attack prey or escape predators. Because of limited experimental data, parameterizations of impact assessment models extrapolate observational data for other species, stages and exposure scenarios. Carroll et al. (2018, 2022) is one example of this presenting a parameterization of a Dynamic Energy Budget model (Kooijman and Kooijman, 2010). In addition, the basis for most parameter sets is exposure of individuals to fixed oil concentrations and time durations, despite that the exposure an individual experiences during an oil spill may be highly dynamic. Here, we therefore aim at quantifying typical individual exposure histories, specifically for oil spill scenarios with high rates and long durations. Such spills are not frequent but have occurred (Deepwater Horizon (Incardona and Scholz, 2018; National Academies of Sciences, E. a. M, 2020)) and should be considered in a risk assessment. It is known that oil in the ocean is quickly diluted and biodegraded (Lee et al., 2013; Bejarano et al., 2014; Nedwed et al., 2021), but large continuous spills may lead to extended periods of high concentrations. The results will contribute to an informed design of laboratory exposure trials that are more relevant to field exposure experienced by individuals during a large oil spill. This could also improve knowledge of uptake kinetics for dynamic exposures, where observations are scarce.

Specifically, we address the following questions and discuss the results in the context of how to design relevant laboratory exposures for use in risk assessments of large oil spills:

i. What is the typical temporal exposure of fish ELS to oil under this oil spill scenario?
ii. How is the estimated impact of oil exposure on fish ELS affected by uptake kinetics?

To address these questions, we run a large oil spill scenario with a coupled model system described in Carroll et al. (2018, 2022) including model components for the ocean, oil spill and fate, and fish ELS. Note that caution is required when drawing conclusions based on numerical replicates of reality, where especially small-scale processes are typically not resolved by the spatial and temporal model resolution.

2. Materials and methods

We simulated an exposure scenario where ELS of Northeast Arctic cod (NEA cod, *Gadus morhua*) experience a large oil release in the Lofoten area of northern Norway during their spawning season. The focus is on the dynamic exposure characteristics during the early life stages to support a discussion on experimental exposure design. More specifically, we focus on the water concentrations of dissolved total polycyclic aromatic hydrocarbon (TPAH) as this is a commonly reported quantity from oil exposure experiments (Hodson, 2017). Toxicity varies between various PAHs (Hodson, 2017; Carls and Meador, 2009), but water concentration of TPAH correlates well with toxicity of TPAH and is a good proxy for oil induced toxicity (Hodson, 2017; Aranguren-Abadía et al., 2022; Sørhus et al., 2023).

Observations of body burden in Aranguren-Abadía et al. (2022) were compared to modelled uptake kinetics to evaluate if the model enables us to assess the potential of kinetics to buffer transient peaks in exposure. This was done by reproducing the lab exposure with models using two different parameterizations and is relevant since harmful internal concentrations are caused by interaction between exposure and uptake.

2.1. Scenario

Oil is released in the Lofoten region of northern Norway concurrent with the spawning season of NEA cod. The release site is at 10.841 E, 67.700 N (Fig. 1), the same as used in Carroll et al. (2018, 2022). The scenario is a surface release of 4500 m^3/day with the Balder oil blend for 90 days starting on March 1st, which is also the start date for the cod spawning in the model. The simulation runs for 45 days after the release, giving a simulation period of 135 days from Mar 1st until July 14th in 2001. The year was chosen as this is the scenario in Carroll et al. (2018) with the highest oil induced cod ELS mortality. Note that we do not weight our results by empiric spawning intensity. This is because our focus is on the dynamic features of ELS exposure and uptake, and not on assessing the scenario-specific population impact.

2.2. Model system

The data is simulated using the SYMBIOSES coupled model (Carroll et al., 2018; Carroll et al., 2022) consisting of five modules; an ocean model, an oil spill and fate model, a multi-species fish ELS model, a *C. finmarchicus* full life cycle model and a multi-species fish population model (latter two not considered in this study). All modules are mature and well tested as described below. In the classification system of French-McCay et al. (2023) for aquatic toxicity models, SYMBIOSES is a Model Tier 5 with the capability to calculate population effects based on individual exposure histories. Fig. 1 (left panel) shows the model domains and the spawning grounds of NEA cod used in the model. A model state (Fig. 1, right panel) consists of hydrographic conditions, an oil concentration field and ELS individuals, each with time-dependent positions, weights and other properties.

The ocean model SINMOD is a primitive Navier-Stokes equation

model with a spatial grid resolution of 4 by 4 km and a 6 min time step covering the Northeast Atlantic (Alver et al., 2016; Slagstad and McClimans, 2005; Wassmann et al., 2006; Støle-Hansen and Slagstad, 1991). OSCAR is a multi-phase particle based Lagrangian oil model (Johansen et al., 2015; Nordam et al., 2019; Barreto et al., 2021; Reed et al., 2004; Daae et al., 2018; Socolofsky et al., 2015) using a pseudo-component approach (Reed et al., 1999) forced with currents, temperatures, salinities and turbulence from the SINMOD ocean model. It is run with a 12 min time step, 302,000 oil particles and 25 oil pseudo-components (SM Table S1). The particles are interpolated to concentration values on a grid with 1.5 km spatial resolution, which are the values used by the fish ELS model.

LARMOD is an Individual Based Model (IBM) for NEA cod ELS that is run with a 12 min time step (Vikebø et al., 2021; Vikebø et al., 2007). A year class is represented by 189,000 individuals spawned at nine different spawning grounds from March 1st until April 30th. Individuals develop according to their experienced environment and are advected using the velocity fields of the SINMOD ocean model with a Runge Kutta 4th order scheme. Sampling the gridded oil concentrations at the individual positions give the complete exposure histories of all individuals. Note that the time steps for the oil model and fish ELS model are equal, so that individuals always use an updated oil state. Data for each individual is saved every time step, i.e. every 12 min simulated time.

2.3. Individual exposure histories

Exposure is given by the water concentrations of dissolved oil for the 25 pseudo-components (SM Table S1) used in the oil model. This results in the exposure history of an individual being 25 time series of water concentrations. From these, individual time series of total dissolved oil and TPAH are calculated as the sum of all the pseudo-components or the pseudo-components Naphthalenes 1, Naphthalenes 2, PAH-1 and PAH-2 (SM Table S2), respectively.

2.4. Characteristic exposure profile

Having the exposure histories of all individuals, a characteristic exposure profile for the release scenario can be created using the following algorithm:

1. Specify an exposure metric, a selection criterion, a normalization and a statistic.
2. Calculate the time series of the exposure metric for each individual.
3. Apply selection of individuals.
4. Shift the time series in time such that the maximum occurs at relative time zero.
5. Apply normalization of time series.
6. Perform statistics to get a characteristic time series.

Here, the exposure metric is TPAH, the selection criterion is all individuals with a non-zero exposure, the normalization is to divide by the individual max TPAH values, and the statistic is the median. These choices produce a normalized TPAH exposure profile assumed to be typical for the scenario. There are of course several other choices possible in step 1, e.g. to use total dissolved oil or toxic units as exposure metric, but our choice makes the profile easy to calculate and does not depend on specific concentration thresholds for effects.

2.5. Internal concentrations of oil

For every individual, a scaled internal concentration C_D can be calculated for each of the oil pseudo-components. These are equivalent to water concentrations C_V, but with a delay due to uptake and elimination given by the elimination rate k_e using the ordinary differential equation

$$\frac{d}{dt}C_D = k_e \cdot (C_V - C_D)$$

where C_D has an analytical solution for constant C_V, but is otherwise found numerically. This calculation is the first step in the GUTS framework (Jager et al., 2011) and can be the input to a variety of effect models as it considers the speed at which the oil is taken up by an individual. Note that k_e is a lumped parameter (Jager et al., 2011) and could cover different processes in different data sources. An estimate of the body burden that would be measured in real life is given by $C = P_{iw} \cdot C_D$ where P_{iw} is the partitioning internal-water coefficient, also called a bioconcentration factor (BCF).

2.6. Kinetic parameterizations

Values of k_e and P_{iw} depend on properties of both the oil and the fish, making them pseudo-component and species specific. We use two different functions for k_e based on the octanol-water partition coefficient K_{ow} and a water temperature of 7 °C. The first is from Klok et al. (2014),

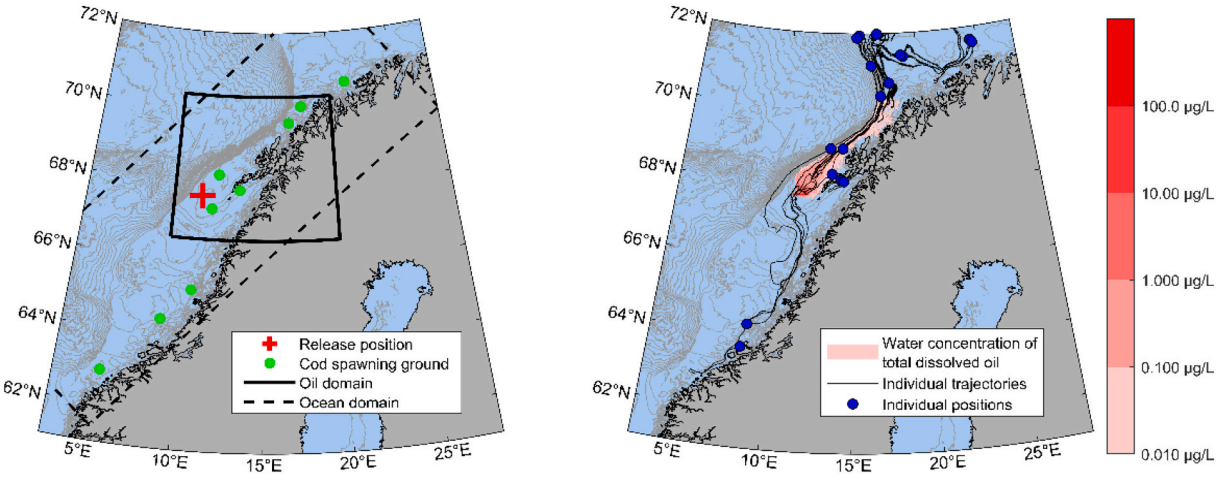

Fig. 1. Overview of the model domains (left) and an example of a model state (right). The oil release position has coordinates 10.841 E, 67.700 N. Dissolved oil concentrations are calculated within the oil domain and assumed to be zero outside. The underlying ocean model has a larger domain enabling the larvae to drift through the oil domain from south to north with the Norwegian Coastal Current. 1000 individuals are released from each of the nine spawning grounds every third day from March 1st until April 30th, giving a total of 189,000 modelled individuals. The right panel shows the complete trajectories of 18 individuals (lines) released March 1st together with the positions of these 18 individuals (blue dots) and the concentration field of total dissolved oil at April 15th.

originally from Baas et al. (2009) for fathead minnow. The second is from the OMEGA model (Hendriks et al., 2001) with parameter values from Nepstad et al. (2021) and Sørensen et al. (2016). Both elimination rate functions are combined with the same function for the BCFs. The function for P_{iw} is from the OMEGA model (Hendriks et al., 2001) with a lipid fraction of 0.6 % based on Sørensen et al. (2016).

For a complete description and numerical values of the kinetic parameters, see the Supplementary Material. This includes Fig. S1 showing the elimination rates as functions of K_{ow} and Fig. S2 for uptake time. OMEGA predicts a much faster uptake than Klok, and the two parametrizations can thus be interpreted as examples of fast and slow uptake, respectively. This is supported by the comparison in Fig. S1 with a more generic formula for the elimination rates from Redman et al. (2022). For both parameterizations, elimination rates decrease with increasing K_{ow} resulting in slower kinetics. However, BCFs increase with K_{ow} resulting in a greater body burden for a given scaled internal concentration. See Fig. S3 for an illustration of this.

2.7. Example of observed body burdens in NEA cod

Species specific data sets reporting both water concentrations and body burdens are scarce and limited to specific exposure scenarios, making it hard to evaluate the kinetics. One example, however, is Aranguren-Abadía et al. (2022) that report measured water concentrations and corresponding body burdens in Atlantic cod embryos for specific oil components after 72 h exposure to crude oil (Supplementary Tables S1 and S2 in Aranguren-Abadía et al. (2022), values for no UV). We have chosen four of these oil components (naphthalene, fluoranthene, pyrene, chrysene) that roughly correspond to the four PAH pseudo-components based on their K_{ow} values (retrieved from ChemSpider.com). To compare the observations with estimated body burdens from the model, we assume constant water concentrations for three days.

3. Results

3.1. Temporal variability in TPAH exposure

An example of the 3-hourly mean exposure experienced by an individual during its dispersal from a spawning ground along the Norwegian Coast is shown in Fig. 2. It experiences no oil for about the first 7 days of its life before a rapid increase when it reaches the impact region of the oil spill. As the egg hatches and continues to drift northwards, the concentration decreases slowly since the oil drifts along in the same direction. The total time the individual experiences a non-zero concentration is about 25 days, where the duration between the first and the last time with TPAH above the plotted threshold of 1.0 µg/L is about six days. For exposure statistics of the full year class, see the Supplementary Material.

Fig. 3 shows the median normalized TPAH water concentration (black) calculated using the algorithm of Section 2.4. The highest value occurs as a spike with a duration on the scale of hours. After the rapid decline of the spike, the value decreases slowly from about 20 % of the spike value right after the peak to about 10 % of the spike value three days after the peak.

Using the slow Klok uptake, internal concentration (blue dashed) is buffered by the uptake speed and stays well below the maximum water concentration. For the fast OMEGA uptake (orange dashed) the internal concentration mostly follows the water concentration except for the transient peak. However, the peak results in a clear response for OMEGA in contrast to for Klok. This can be explained by the inset of Fig. 3 showing the maximum internal concentration as a function of elimination rate assuming the exposure profile of the main figure. For Klok, the PAH pseudo-components range from 2 % (PAH-2) to 12 % (Nap-1) of the maximum water concentration. For OMEGA, they range from 16 % (PAH-2) to 59 % (Nap-1). As TPAH is dominated by PAH-2 with the slowest uptake of the PAH pseudo-components, the internal concentrations (dashed lines in the main part of Fig. 3) have maximum values close to the corresponding left-most (PAH-2) dots in the inset.

3.2. Body burden modelling

Body burden data after three days of constant exposure to crude oil from Aranguren-Abadía et al. (2022) are shown in Fig. 4 together with estimated values from the uptake model. The data follow a linear pattern where the slope of the fitted line increases with increasing K_{ow} except for chrysene with the highest K_{ow}. Modelled dose-response slopes, on the other hand, are strictly increasing with K_{ow}. This is also illustrated in the right panels of Fig. S3 in the SM. Modelled values for naphthalene with the lowest K_{ow} are close to or at steady-state (maximum internal concentration for a constant exposure), but both parameterizations are still below the observations. Using the slower Klok parameterization, modelled values for pyrene and fluoranthene are at 22.8 % and 16.1 % of the steady-state values, respectively, and compare well with the measurements. Contrary, these components are at steady-state using the

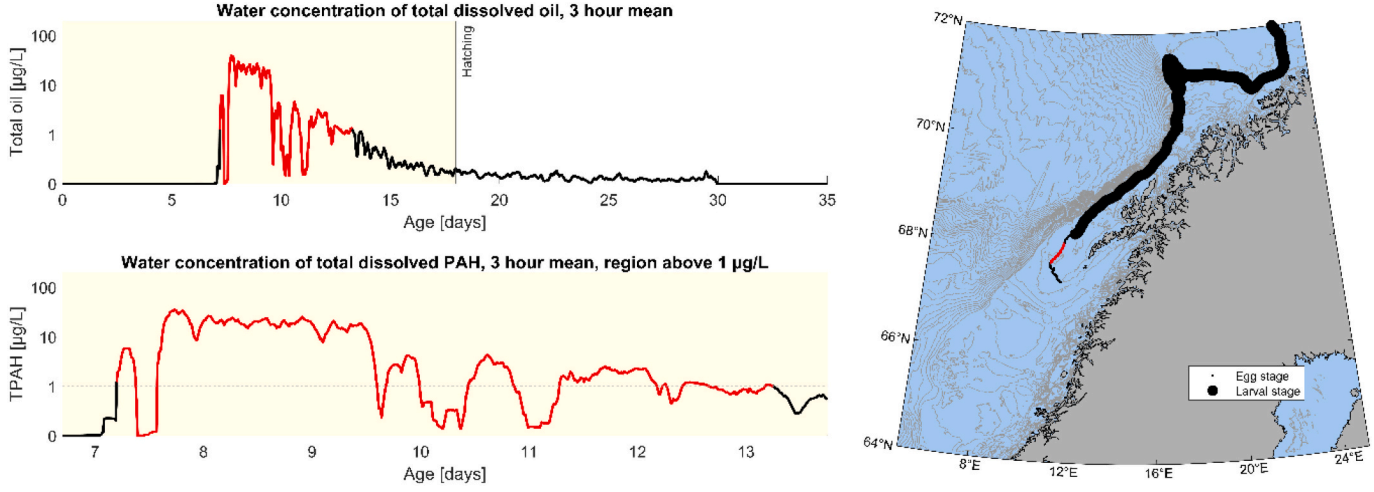

Fig. 2. Example time series of total dissolved oil (upper panel) and total dissolved PAH (TPAH, lower panel) experienced by an individual, and its corresponding spatial trajectory. The red region is from the first time the TPAH value is above the plotted threshold of 1.0 µg/L until the last time the TPAH value is above 1.0 µg/L. Total dissolved oil is calculated as the sum of all the pseudo-components (SM, Table S1), while the TPAH is the sum of four pseudo-components (SM, Table S2). The yellow background shows the duration of the egg stage. Note that the y-axes have a linear scale below 1.0 µg/L and a logarithmic scale above 1.0 µg/L.

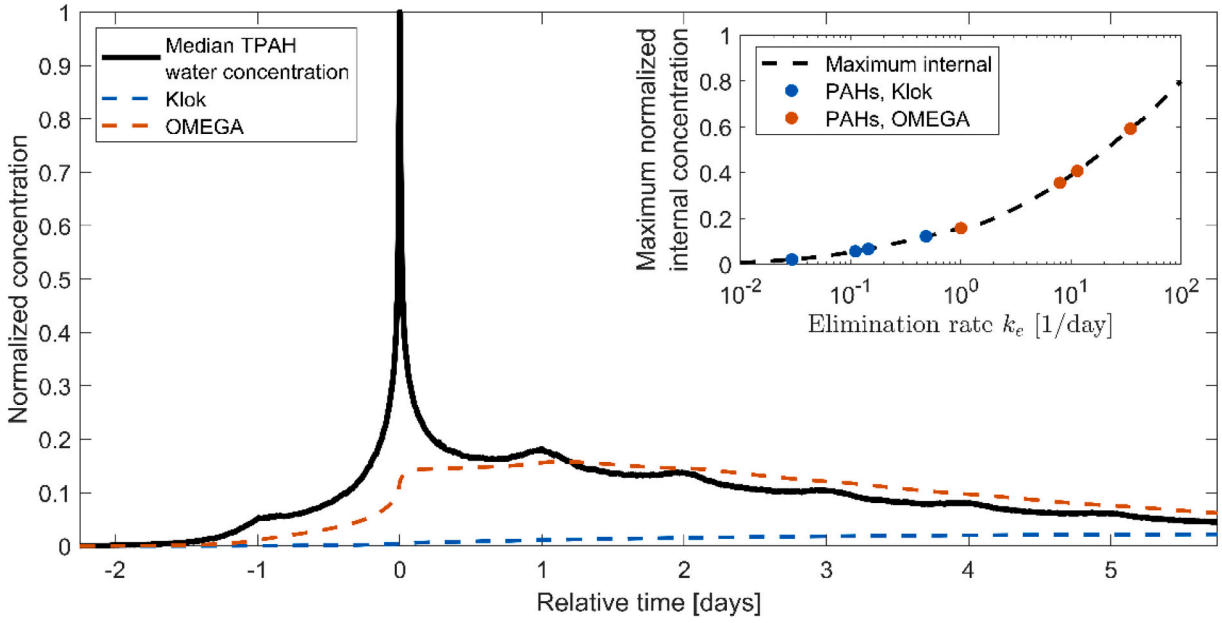

Fig. 3. Median normalized water concentration of dissolved TPAH (black) using the algorithm in Section 2.4. Scaled internal TPAH concentrations (dashed) are calculated using the elimination rates k_e for 7 °C and the dynamic distribution of the four PAH pseudo-components. Note that the y-axis has a linear scale in contrast to the y-axes in Fig. 2. The inset shows the maximum normalized internal concentration as a function of elimination rate for the exposure profile of the main fig. E.g., the peak of the normalized internal concentration for a component with $k_e = 1$ would be about 0.2 for this exposure profile.

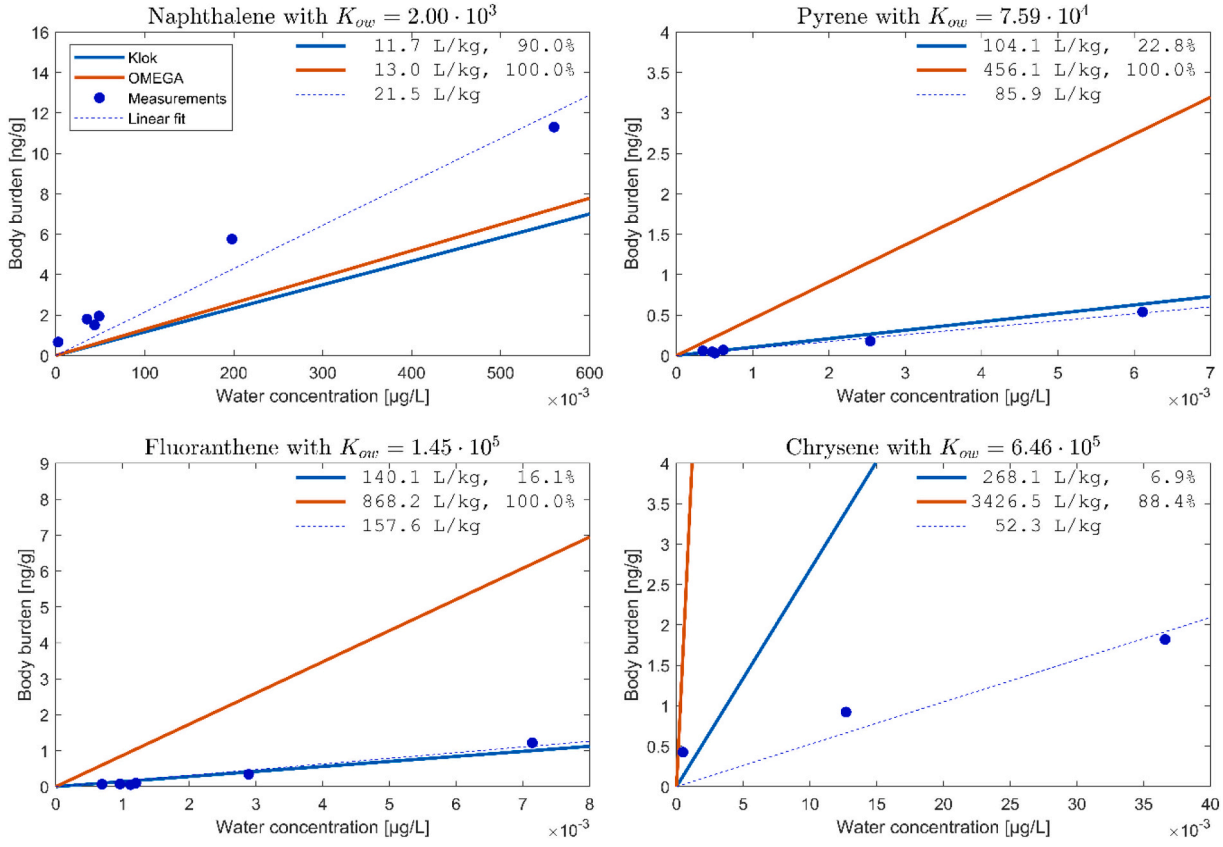

Fig. 4. Body burden as a function of water concentration for various oil components after three days exposure at 7 °C. Dots are measurements from laboratory experiments where cod embryos were exposed to crude oil (Aranguren-Abadía et al., 2022). Dashed lines are fitted to the data while solid lines are model estimates using the parameterizations of Section 2.6. Values in the upper right corners are the slopes of the lines. The solid lines also inform how close the model estimate has reached within the three days relative to the modelled steady-state as percentage. Note that the panels have axes with different scales, and that components in this figure are not the exact PAH pseudo-components used in the oil model (Table S2), but instead components with K_{ow} values close to these.

faster OMEGA parameterization resulting in modelled body burdens well above the measurements. Chrysene, with the highest K_{ow} value of the four components, is close to steady-state for OMEGA with 88.4 %, but far from steady-state for Klok with only 6.9 %, making the two lines for modelled body burden very different. Both modelled lines, however, are well above the measured values.

4. Discussion

4.1. Exposure dynamics and relevance to experimental oil design

Here, we show results from a modelled relevant large oil spill scenario in the Northeast Atlantic, an area where petroleum resources have been mapped and adjacent petroleum production sites are located. The scenario displays exposure dynamics of key importance to assess potential effects on fish ELS according to the numerical model. The modelled exposure history to oil for fish ELS can be summarized in four phases;

i) a slow increase in water concentrations of oil enduring for several days as the individual nears the oil plume
ii) a rapid increase to a transient peak exposure as it meets the bulk of the oil plume, well above the levels before and after, that last for a few hours before rapidly decreasing
iii) a subsequent exposure at a near constant level that endures for ~3 days as it travels with the plume, and ends with
iv) a decrease in concentrations as the oil plume diffuses that is slower than the time from the inception of the exposure to the peak.

A typical design in laboratory exposure studies is a fixed exposure for a limited number of days. Contrary to the numerical results shown here, there is no gradual build-up of the oil. An individual in a laboratory experiment would therefore spend more time reaching an equilibrium between water and internal concentrations of oil than what may be expected in the field. Furthermore, the results shown here indicate that fish ELS in the field likely experience a transient high peak in water concentrations of oil before the above-described third phase with a near-fixed exposure lasting for a few days. This peak is due to individuals being transported through smaller areas of elevated concentrations, and it is not apparent what significance such a transient peak in exposure may have on the different developmental stages of fish ELS with stage-specific sensitivity. Depending on the oil components and the characteristics of the individual under consideration, a peak exposure may or may not lead to a considerable uptake of harmful oil. There are indications that a shorter exposure at higher levels has a greater effect than a longer exposure at lower levels (Sørhus et al., 2016). However, a slow uptake may protect the individual from such transient peaks. Finally, the long tail of exposure suggested by the numerical model may be below known threshold levels for effects but might still be on a level of significance for individuals recovering. Literature suggests that sub-lethal effects may include behavioral changes that hamper prey and predator interactions at oil exposure levels below what is considered to result in visible malformations (Cresci et al., 2020; Heintz et al., 2000; Carls et al., 1999; Wang et al., 2018; Brown et al., 2016).

Though numerical models may inform of ways to increase relevance of oil spill scenario design, there are still limitations to what exposure designs that may be implemented in the lab. A high number of eggs may be divided among tanks in equal numbers before introducing oil to the environment. Note that already here there are limitations, because ideally, we would like to know the effects of individual oil components for a wide range of covarying environmental parameters such as temperature and UV radiation (Aranguren-Abadía et al., 2022). However, accepting that we settle with total oil or a few selected components in a standardized environment, the limited number of tanks available and the need for replicates severely limits the number of different concentrations that may be investigated. Furthermore, though we may extract individuals at different times after exposure initiation, we can only assess a few initiation times and durations. Thus, a standard profile should be established to keep the number of replicates manageable for dynamic exposure experiments.

Based on our results, we suggest the experimental design presented in Fig. 5 to mimic the exposure of an individual during a large oil spill. This consists of the following modifications:

- Begin with a gradual increase in exposure
- Include an early peak in exposure at an order of magnitude higher than the fixed level
- Allow the fixed exposure to last for up to three days
- If possible; end with a gradual decrease in exposure

This experimental design is of course more complicated than the traditional fixed exposure, but a pulsed exposure regime is used in previous lab studies (Sørhus et al., 2015) and can be obtained with the correct lab equipment (Nordtug et al., 2011). However, it could be beneficial to have some replicates with a standard fixed exposure as controls if feasible.

Furthermore, though scenario design involves consideration of complex processes and dynamics, stakeholder interaction is needed to increase relevance and for conveying intrinsic uncertainties that should be taken into consideration when assessing risk based on numerical models versus other types of relevant knowledge. The receivers of assessments need to be carefully explained what prerequisites the linkage between experimental exposure design and development of relevant models are based on.

4.2. Uptake kinetics and importance to experimental oil design

Kinetic parameters used here are not based on cod data, and the need for extrapolation in both temperature and species introduces uncertainties. For the typical exposure of three days, the oil components in the model span from being close to steady-state to far away depending on the K_{ow} value (SM, Fig. S3). When far from steady-state ($k_e \cdot t \ll 1$), the modelled body burden is $C \approx k_e \cdot P_{iw} \cdot C_V \cdot t$ such that only the product $k_e \cdot P_{iw}$ is important, not the individual values of k_e and P_{iw}. Close to steady-state ($k_e \cdot t \gg 1$), the exact value of k_e becomes insignificant as $C \approx P_{iw} \cdot C_V$. The components of the example data (Fig. 4) show that naphthalene with the lowest K_{ow} is close to steady-state for both elimination rates, while the estimated body burden is too low compared to the measurements. To increase the estimated body burden, one would need to increase the P_{iw}. Looking at pyrene and fluoranthene with the middle two K_{ow} values, however, the estimated body burdens for the slower Klok parameterization fit well with the measurements, while the faster OMEGA parameterization predicts too high values. This could indicate that the uptake speed of Klok is more realistic than that of OMEGA, especially if the bioconcentration factors are in the lower end as indicated by the naphthalene data. For chrysene with the greatest K_{ow} value, both parameterizations estimate too high body burden. It is not obvious what the correct adjustment to the parameters is and the parameterizations may be approaching an upper limit of their valid K_{ow} range. This illustrates that to estimate both k_e and P_{iw} accurately, one needs repeated measurements in time using an experimental setup similar to the one of Øverjordet et al. (2018) for *Calanus hyperboreus* that covers both uptake and elimination. The data used for estimation in such an experiment should be measurements of body burden directly, and not only through survival calculated by an effects model (Jager et al., 2017). A complete model for uptake and elimination should also include growth and change in lipid content, which is considered unnecessary here to discuss the consequences of different uptake speeds. For a further discussion on elimination rates and BCFs, see e.g. Baas et al. (2015), Arnot and Gobas (2006) and Hendriks et al. (2001). Note, however, that the elimination of PAHs in the model may in reality be metabolization

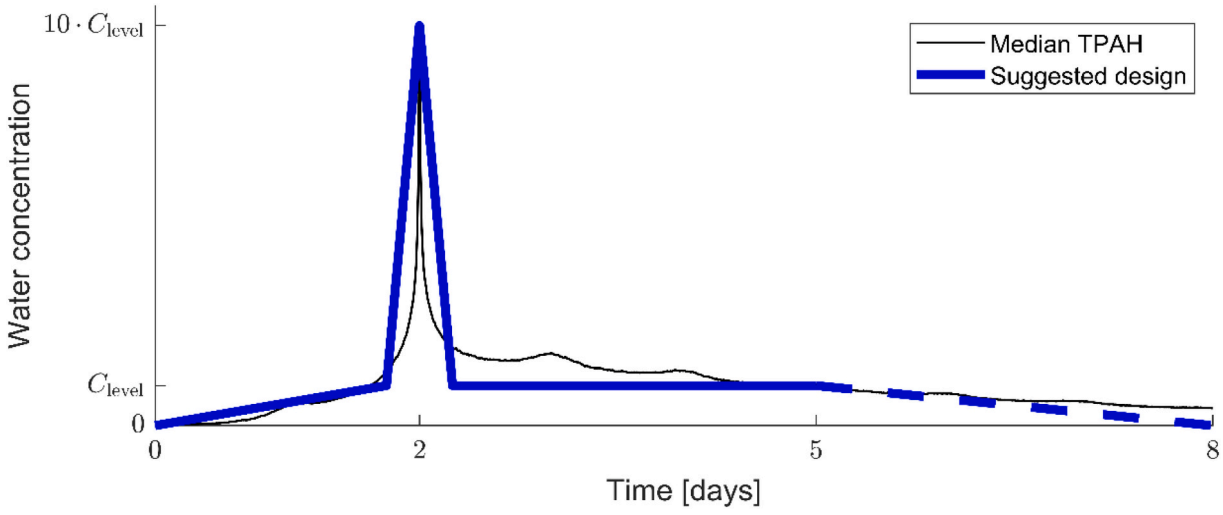

Fig. 5. Suggested design (blue) for an oil exposure experiment based on the median curve of Fig. 3 (black). First a slow increase in concentration lasting for ~2 days, then a transient peak lasting for a few hours with roughly 10 times as high concentration. Finally, a fixed exposure for ~3 days, similar to the traditional design used in experiments with fixed exposure. A final decline stage can be added if possible.

into other toxic compounds (Donald et al., 2023; Tanabe et al., 2022) with an effect dependent on the development stage (Sørhus et al., 2021).

While numerical impact assessment models often address water concentrations of oil components, effects are caused by body burdens that are a combination of exposure and uptake. As shown in Fig. 3, accurate kinetics is increasingly important when exposure is highly dynamic. A slow uptake may buffer transient exposure events dependent on the large range of uptake speeds for various oil components. Hence, there is an urgent need to both; i) revisit exposure design for increased relevance according to what this looks like in the field, and ii) develop more data for parameterizing species, stage and oil component dependent kinetics given the dynamic nature of exposure during a large oil spill.

4.3. Uncertainty in methods and data

Spatial and temporal resolution in the ocean model limits the dynamic scales included that may affect individual exposure history of fish ELS. Both the IBM for fish ELS and the oil model are Lagrangian models where fish and oil are represented by a limited number of particles. Though the particle numbers are high with 189,000 and 302,000, this may affect the accuracy in the modelled description of dynamic exposure, probably in the direction of overestimating the maximum exposure. With an increasing number of particles, distributions typically become smoother, which likely reduces the degree of dynamics in the exposure. It should also be noted that an individual could experience several spikes in TPAH that are smoothed out in the median calculation for Fig. 3, as seen for the example individual in Fig. 2.

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CRediT authorship contribution statement

Håvard G. Frøysa: Conceptualization, Formal analysis, Methodology, Software, Visualization, Writing - original manuscript & editing. **Raymond Nepstad:** Methodology, Software. **Sonnich Meier:** Conceptualization, Writing - review & editing. **Carey Donald:** Conceptualization, Writing - review & editing. **Elin Sørhus:** Conceptualization, Writing - review & editing. **Mathias Bockwoldt:** Data curation, Software. **JoLynn Carroll:** Conceptualization, Funding acquisition, Writing - review & editing. **Frode B. Vikebø:** Conceptualization, Writing - original manuscript & 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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

List of all oil pseudo-components, detailed description of PAH pseudo-components, details about functions for kinetic parameters, and exposure statistics for all individuals (PDF). Supplementary data to this article can be found online at doi:https://doi.org/10.1016/j.scitotenv.2023.166951.

References

Alver, M.O., Broch, O.J., Melle, W., Bagøien, E., Slagstad, D., 2016. Validation of an Eulerian population model for the marine copepod *Calanus finmarchicus* in the Norwegian Sea. J. Mar. Syst. 160, 81–93.

Aranguren-Abadía, L., Yadetie, F., Donald, C.E., Sørhus, E., Myklatun, L.E., Zhang, X., Lie, K.K., Perrichon, P., Nakken, C.L., Durif, C., Karlsen, O.A., 2022. Photo-enhanced toxicity of crude oil on early developmental stages of Atlantic cod (*Gadus morhua*). Sci. Total Environ. 807, 150697.

Arnot, J.A., Gobas, F.A., 2006. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. Environ. Rev. 14 (4), 257–297.

Baas, J., Jager, T., Kooijman, S., 2009. A model to analyze effects of complex mixtures on survival. Ecotoxicol. Environ. Saf. 72 (3), 669–676.
Baas, J., Spurgeon, D., Broerse, M., 2015. A simple mechanistic model to interpret the effects of narcotics. SAR QSAR Environ. Res. 26 (3), 165–180.
Barreto, F.T., Dammann, D.O., Tessarolo, L.F., Skancke, J., Keghouche, I., Innocentini, V., Winther-Kaland, N., Marton, L., 2021. Comparison of the coupled model for oil spill prediction (CMOP) and the oil spill contingency and response model (OSCAR) during the DeepSpill field experiment. Ocean Coast. Manag. 204, 105552.
Bejarano, A.C., Clark, J.R., Coelho, G.M., 2014. Issues and challenges with oil toxicity data and implications for their use in decision making: a quantitative review. Environ. Toxicol. Chem. 33 (4), 732–742.
Bejarano, A.C., Adams, J.E., McDowell, J., Parkerton, T.F., Hanson, M.L., 2023. Recommendations for improving the reporting and communication of aquatic toxicity studies for oil spill planning, response, and environmental assessment. Aquat. Toxicol. 106391.
Beyer, J., Trannum, H.C., Bakke, T., Hodson, P.V., Collier, T.K., 2016. Environmental effects of the Deepwater Horizon oil spill: a review. Mar. Pollut. Bull. 110 (1), 28–51.
Brown, D., Bailey, J., Oliveri, A., Levin, E., Di Giulio, R., 2016. Developmental exposure to a complex PAH mixture causes persistent behavioral effects in naive *Fundulus heteroclitus* (killifish) but not in a population of PAH-adapted killifish. Neurotoxicol. Teratol. 53, 55–63.
Carls, M.G., Meador, J.P., 2009. A perspective on the toxicity of petrogenic PAHs to developing fish embryos related to environmental chemistry. Hum. Ecol. Risk. Assess. 15 (6), 1084–1098.
Carls, M.G., Rice, S.D., Hose, J.E., 1999. Sensitivity of fish embryos to weathered crude oil: part I. low-level exposure during incubation causes malformations, genetic damage, and mortality in larval pacific herring (Clupea pallasi). Environ. Toxicol. Chem. Int. J. 18 (3), 481–493.
Carroll, J., Vikebø, F., Howell, D., Broch, O.J., Nepstad, R., Augustine, S., Skeie, G.M., Bast, R., Juselius, J., 2018. Assessing impacts of simulated oil spills on the Northeast Arctic cod fishery. Mar. Pollut. Bull. 126, 63–73.
Carroll, J., Frøysa, H.G., Vikebø, F., Broch, O.J., Howell, D., Nepstad, R., Augustine, S., Skeie, G.M., Bockwoldt, M., 2022. An annual profile of the impacts of simulated oil spills on the Northeast Arctic cod and haddock fisheries. Mar. Pollut. Bull. 184, 114207.
Cresci, A., Paris, C.B., Browman, H.I., Skiftesvik, A.B., Shema, S., Bjelland, R., Durif, C. M., Foretich, M., Di Persia, C., Lucchese, V., Vikebø, F., Sørhus, E., 2020. Effects of exposure to low concentrations of oil on the expression of cytochrome P4501a and routine swimming speed of Atlantic haddock (*Melanogrammus aeglefinus*) larvae in situ. Environ. Sci. Technol. 54 (21), 13879–13887.
Daae, R.L., Skancke, J., Brandvik, P.J., Faksness, L.-G., 2018. The sensitivity of the surface oil signature to subsurface dispersant injection and weather conditions. Mar. Pollut. Bull. 127, 175–181.
Donald, C.E., Sørhus, E., Perrichon, P., Nakken, C.L., Goksøyr, A., Jørgensen, K.B., Mayer, P., da Silva, D.A., Meier, S., 2023. Co-exposure of phenanthrene and the cyp-inducer 3-methylchrysene leads to altered biotransformation and increased toxicity in fish egg and larvae. Environ. Sci. Technol. 57 (30), 11022–11031.
French-McCay, D.P., Parkerton, T.F., de Jourdan, B., 2023. Bridging the lab to field divide: advancing oil spill biological effects models requires revisiting aquatic toxicity testing. Aquat. Toxicol. 256, 106389.
Hansen, B.H., Nordtug, T., Farkas, J., Khan, E.A., Oteri, E., Kvæstad, B., Faksness, L.-G., Daling, P.S., Arukwe, A., 2021. Toxicity and developmental effects of Arctic fuel oil types on early life stages of Atlantic cod (*Gadus morhua*). Aquat. Toxicol. 237, 105881.
Heintz, R.A., Rice, S.D., Wertheimer, A.C., Bradshaw, R.F., Thrower, F.P., Joyce, J.E., Short, J.W., 2000. Delayed effects on growth and marine survival of pink salmon *Oncorhynchus gorbuscha* after exposure to crude oil during embryonic development. Mar. Ecol. Prog. Ser. 208, 205–216.
Hendriks, A.J., van der Linde, A., Cornelissen, G., Sijm, D.T., 2001. The power of size. 1. Rate constants and equilibrium ratios for accumulation of organic substances related to octanol-water partition ratio and species weight. Environ. Toxicol. Chem. 20 (7), 1399–1420.
Hjermann, D.Ø., Melsom, A., Dingsør, G.E., Durant, J.M., Eikeset, A.M., Røed, L.P., Ottersen, G., Storvik, G., Stenseth, N.C., 2007. Fish and oil in the Lofoten–Barents Sea system: synoptic review of the effect of oil spills on fish populations. Mar. Ecol. Prog. Ser. 339, 283–299.
Hodson, P.V., 2017. The toxicity to fish embryos of PAH in crude and refined oils. Arch. Environ. Contam. Toxicol. 73 (1), 12–18.
Hodson, P.V., Adams, J., Brown, R.S., 2019. Oil toxicity test methods must be improved. Environ. Toxicol. Chem. 38 (2), 302–311.
Incardona, J.P., Scholz, N.L., 2018. Case study: the 2010 Deepwater Horizon oil spill and its environmental developmental impacts. Environ. Dev. 235–283.
Jager, T., Albert, C., Preuss, T.G., Ashauer, R., 2011. General unified threshold model of survival - a toxicokinetic-toxicodynamic framework for ecotoxicology. Environ. Sci. Technol. 45 (7), 2529–2540.
Jager, T., Øverjordet, I.B., Nepstad, R., Hansen, B.H., 2017. Dynamic links between lipid storage, toxicokinetics and mortality in a marine copepod exposed to dimethylnaphthalene. Environ. Sci. Technol. 51 (13), 7707–7713.
Johansen, Ø., Reed, M., Bodsberg, N.R., 2015. Natural dispersion revisited. Mar. Pollut. Bull. 93 (1–2), 20–26.
Keramea, P., Spanoudaki, K., Zodiatis, G., Gikas, G., Sylaios, G., 2021. Oil spill modeling: a critical review on current trends, perspectives, and challenges. J. Mar. Sci. Eng. 9 (2), 181.
Klok, C., Nordtug, T., Tamis, J.E., 2014. Estimating the impact of petroleum substances on survival in early life stages of cod (*Gadus morhua*) using the dynamic energy budget theory. Mar. Environ. Res. 101, 60–68.
Kooijman, B., Kooijman, S., 2010. Dynamic Energy Budget Theory for Metabolic Organisation. Cambridge University Press.
Langangen, Ø., Olsen, E., Stige, L.C., Ohlberger, J., Yaragina, N.A., Vikebø, F.B., Bogstad, B., Stenseth, N.C., Hjermann, D.Ø., 2017. The effects of oil spills on marine fish: implications of spatial variation in natural mortality. Mar. Pollut. Bull. 119 (1), 102–109.
Lee, K., Nedwed, T., Prince, R.C., Palandro, D., 2013. Lab tests on the biodegradation of chemically dispersed oil should consider the rapid dilution that occurs at sea. Mar. Pollut. Bull. 73 (1), 314–318.
National Academies of Sciences, E. a. M, 2020. The Use of Dispersants in Marine Oil Spill Response. The National Academies Press, Washington, DC.
Nedwed, T., Mitchell, D., Konkel, W., Coolbaugh, T., 2021. SMART protocol using polarized infrared cameras. In: International Oil Spill Conference, p. 690093.
Nepstad, R., Hansen, B.H., Skancke, J., 2021. North Sea produced water PAH exposure and uptake in early life stages of Atlantic cod. Mar. Environ. Res. 163, 105203.
Nordam, T., Beegle-Krause, C., Skancke, J., Nepstad, R., Reed, M., 2019. Improving oil spill trajectory modelling in the Arctic. Mar. Pollut. Bull. 140, 65–74.
Nordtug, T., Olsen, A.J., Altin, D., Meier, S., Overrein, I., Hansen, B.H., Johansen, Ø., 2011. Method for generating parameterized ecotoxicity data of dispersed oil for use in environmental modelling. Mar. Pollut. Bull. 62 (10), 2106–2113.
Øverjordet, I.B., Nepstad, R., Hansen, B.H., Jager, T., Farkas, J., Altin, D., Brönner, U., Nordtug, T., 2018. Toxicokinetics of crude oil components in Arctic copepods. Environ. Sci. Technol. 52 (17), 9899–9907.
Redman, A.D., Parkerton, T.F., Letinski, D.J., Sutherland, C.A., Butler, J.D., Di Toro, D. M., 2022. Modeling time-dependent aquatic toxicity of hydrocarbons: role of organism weight, temperature, and substance hydrophobicity. Environ. Toxicol. Chem. 41 (12), 3070–3083.
Reed, M., Johansen, Ø., Brandvik, P.J., Daling, P., Lewis, A., Fiocco, R., Mackay, D., Prentki, R., 1999. Oil spill modeling towards the close of the 20th century: overview of the state of the art. Spill Sci. Technol. Bull. 5 (1), 3–16.
Reed, M., Daling, P., Lewis, A., Ditlevsen, M.K., Brørs, B., Clark, J., Aurand, D., 2004. Modelling of dispersant application to oil spills in shallow coastal waters. Environ. Model. Software 19 (7–8), 681–690.
Slagstad, D., McClimans, T.A., 2005. Modeling the ecosystem dynamics of the Barents Sea including the marginal ice zone: I. Physical and chemical oceanography. J. Mar. Syst. 58 (1–2), 1–18.
Socolofsky, S.A., Adams, E.E., Boufadel, M.C., Aman, Z.M., Johansen, Ø., Konkel, W.J., Lindo, D., Madsen, M.N., North, E.W., Paris, C.B., Nedwed, T.J., 2015. Intercomparison of oil spill prediction models for accidental blowout scenarios with and without subsea chemical dispersant injection. Mar. Pollut. Bull. 96 (1–2), 110–126.
Sørensen, L., Silva, M.S., Booth, A.M., Meier, S., 2016. Optimization and comparison of miniaturized extraction techniques for PAHs from crude oil exposed Atlantic cod and haddock eggs. Anal. Bioanal. Chem. 408 (4), 1023–1032.
Sørhus, E., Edvardsen, R.B., Karlsen, Ø., Nordtug, T., van der Meeren, T., Thorsen, A., Harman, C., Jentoft, S., Meier, S., 2015. Unexpected interaction with dispersed crude oil droplets drives severe toxicity in Atlantic haddock embryos. PloS One 10 (4), e0124376.
Sørhus, E., Incardona, J.P., Karlsen, Ø., Linbo, T., Sørensen, L., Nordtug, T., van der Meeren, T., Thorsen, A., Thorbjørnsen, M., Jentoft, S., Edvardsen, R.B., Meier, S., 2016. Crude oil exposures reveal roles for intracellular calcium cycling in haddock craniofacial and cardiac development. Sci. Rep. 6 (1), 31058.
Sørhus, E., Donald, C.E., Da Silva, D., Thorsen, A., Karlsen, Ø., Meier, S., 2021. Untangling mechanisms of crude oil toxicity: linking gene expression, morphology and PAHs at two developmental stages in a cold-water fish. Sci. Total Environ. 757, 143896.
Sørhus, E., Donald, C.E., Nakken, C.L., Perrichon, P., Durif, C.M.F., Shema, S., Browman, H.I., Skiftesvik, A.B., Lie, K.K., Rasinger, J.D., Müller, M.H.B., Meier, S., 2023. Co-exposure to UV radiation and crude oil increases acute embryotoxicity and sublethal malformations in the early life stages of Atlantic haddock (*Melanogrammus aeglefinus*). Sci. Total Environ. 859, 160080.
Støle-Hansen, K., Slagstad, D., 1991. Simulation of currents, ice melting, and vertical mixing in the Barents Sea using a 3-D baroclinic model. Polar Res. 10 (1), 33–44.
Tanabe, P., Pampanin, D.M., Tiruye, H.M., Jørgensen, K.B., Hammond, R.I., Gadepalli, R. S., Rimoldi, J.M., Schlenk, D., 2022. Relationships between isomeric metabolism and regioselective toxicity of hydroxychrysenes in embryos of Japanese medaka (*Oryzias latipes*). Environ. Sci. Technol. 57 (1), 539–548.
Vikebø, F., Jørgensen, C., Kristiansen, T., Fiksen, Ø., 2007. Drift, growth, and survival of larval Northeast Arctic cod with simple rules of behaviour. Mar. Ecol. Prog. Ser. 347, 207–219.
Vikebø, F.B., Rønningen, P., Meier, S., Grøsvik, B.E., Lien, V.S., 2015. Dispersants have limited effects on exposure rates of oil spills on fish eggs and larvae in shelf seas. Environ. Sci. Technol. 49 (10), 6061–6069.
Vikebø, F.B., Broch, O.J., Kajiya Endo, C.A., Frøysa, H.G., Carroll, J., Juselius, J., Langangen, Ø.O.G., 2021. Northeast Arctic cod and prey match-mismatch in a high-latitude spring-bloom system. Front. Mar. Sci. 8, 767191.
Wang, Y., Shen, C., Wang, C., Zhou, Y., Gao, D., Zuo, Z., 2018. Maternal and embryonic exposure to the water soluble fraction of crude oil or lead induces behavioral abnormalities in zebrafish (*Danio rerio*), and the mechanisms involved. Chemosphere 191, 7–16.
Wassmann, P., Slagstad, D., Riser, C.W., Reigstad, M., 2006. Modelling the ecosystem dynamics of the Barents Sea including the marginal ice zone: II. Carbon flux and interannual variability. J. Mar. Syst. 59 (1–2), 1–24.