Marine Pollution Bulletin 76 (2013) 178–186

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

# Modelling bioaccumulation of oil constituents in aquatic species

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ARTICLE INFO

*Keywords:*
Petroleum
Hydrocarbons
Bioaccumulation model
Absorption
Elimination
Bioconcentration factor

ABSTRACT

Crude oil poses a risk to marine ecosystems due to its toxicity and tendency to accumulate in biota. The present study evaluated the applicability of the OMEGA model for estimating oil accumulation in aquatic species by comparing model predictions of kinetic rates (absorption and elimination) and bioconcentration factors (BCF) with measured values. The model was a better predictor than the means of the measurements for absorption and elimination rate constants, but did not outperform the mean measured BCF. Model estimates and measurements differed less than one order of magnitude for 91%, 80% and 61% of the absorption and elimination rates and BCFs of all oil constituents, respectively. Of the "potentially modifying" factors: exposure duration, biotransformation, molecular mass, and water temperature, the last two tended to influence the performance of the model. Inclusion of more explanatory variables in the bioaccumulation model, like the molecular mass, is expected to improve model performance.


## 1. Introduction

Petroleum industry activities may contribute to contamination of marine waters, for example via the discharge of water produced during oil extraction, and accidental spills from shipping and drilling. In the near future, oil exploitation and transportation is expected to increase due to the large energy demand and a changing environment (Gautier et al., 2009). For instance, the current decline in the extent and thickness of Arctic ice offers opportunities for oil exploitation in hitherto unexplored regions. Simultaneously, oil exploitation might become more risky as the increasing amount of moving, newly formed ice could damage rigs and vessels (Harsem et al., 2011). More petroleum industry activities will thus increase the risk of oil contamination of marine ecosystems (De Hoop et al., 2011).

Since crude oil poses a risk to marine ecosystems due to its toxicity and tendency to accumulate in biota, quantitative information on oil bioaccumulation is important for risk assessment and to establish environmental quality guidelines (Arnot and Gobas, 2004; De Hoop et al., 2011; De Laender et al., 2011). Risk estimates can be obtained by comparing internal concentrations with a critical internal concentration, the so called critical body burden (CBB), at which detrimental lethal or sublethal effects occur in organisms. Internal concentrations can be derived from measurements and by using bioaccumulation models that can estimate internal concentrations based on kinetic parameters, e.g. uptake and elimination rate constants (Baussant et al., 2001). The use of models can limit additional animal testing and inform regulatory decision making.

Although several bioaccumulation models have been developed (Arnot and Gobas, 2004), few have been used to quantify the accumulation of oil constituents in aquatic species. The few studies available have focussed mainly on the accumulation of polycyclic aromatic hydrocarbons (PAHs) in fish species (Baussant et al., 2001; Gobas and Opperhuizen, 1986; Mathew et al., 2008), whereas species other than fish, such as algae and invertebrates, will be exposed to oil constituents as well. Furthermore, oil is a complex mixture of constituents, including not only PAHs but also various alkylphenols and straight-chain, ring and branched structures, such as paraffins (Mendelssohn et al., 2012). In the present study, oil accumulation was therefore estimated for aquatic species using the OMEGA bioaccumulation model (Hendriks et al., 2001). In this model absorption and elimination rate constants are

*Abbreviations:* BCF, bioconcentration factor; CBB, critical body burden; DAH, dicyclic aromatic hydrocarbon; *E*, coefficient of efficiency; HOC, hydrophobic organic chemical; $K_{ow}$, Octanol–water partition coefficient; MAH, monocyclic aromatic hydrocarbon; OMEGA, optimal modelling for ecotoxicological applications; PAH, polycyclic aromatic hydrocarbon; RMSE, root-mean-square-error.

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0025-326X/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.marpolbul.2013.09.006

quantified as a function of the octanol–water partition coefficient ($K_{ow}$) of the constituent and the weight, lipid content, and trophic level of the species (Hendriks et al., 2001). These data are relatively easy to obtain. Additionally, several parameter values in the model have been determined with allometric relations. The OMEGA model therefore facilitates bioaccumulation estimations of many chemicals and species, in contrast to most other bioaccumulation models which depend on experimental chemical- and species-specific data. The OMEGA model has been successfully applied to estimate the internal concentrations of metals and several organic pollutants (e.g. biocides, ethers) for various invertebrate and vertebrate species (De Laender et al., 2010; Hauck et al., 2007; Hendriks et al., 2001; Veltman et al., 2008).

The overall aim of the current study was to evaluate the applicability of the OMEGA model and to explore if the model needed improvements for estimating the accumulation of oil constituents in aquatic organisms. To this end, absorption and elimination rate constants and bioconcentration factors (BCFs) estimated with the OMEGA model were compared with measured values reported in literature for aquatic species from different taxonomic groups (e.g. Crustacea, Mollusca and Osteichthyes) exposed to constituents from different oil groups (i.e. mono-, di-, and polycyclic aromatic hydrocarbons, phenols and n-paraffins). Additionally, differences between the model estimates and measurements were evaluated in relation to water temperature, exposure duration, molecular mass of the oil constituents, and biotransformation rate constants. Finally, model estimates for hydrocarbons were compared with model estimates for other organic compounds (e.g. biocides, ethers) to compare variability among oil constituents with variability among organic compounds in general.

## 2. Methods

### 2.1. Experimental data collection

Laboratory-derived rate constants for oil constituents were collected from publications obtained with the ISI Web of Knowledge and Google Scholar search engines. We used the search terms: (1) oil, petroleum, aromatic, aliphatic, resin, phenol, alkane, alkene, alkyn, paraffin, thiophene, olefin, naphthenic mono- and di-aromatic and (2) elimination, excretion or efflux rate, and uptake, absorption or influx rate. Using the reference lists of papers thus obtained, we searched for additional publications. Our search resulted in 10 papers with 66 absorption and 61 elimination rate constants for 10 aquatic species (crustaceans, fish and molluscs) (Berrojalbiz et al., 2009; Bruner et al., 1994; Djomo et al., 1996; Huckins et al., 2004; Jensen et al., 2012; Jimenez et al., 1987; Jonsson et al., 2004; Jovanovich and Marion, 1987; Ruotsalainen et al., 2010; Tollefsen et al., 1998). Additionally, 80 absorption rate constants for 19 aquatic species and 164 elimination rate constants for 29 aquatic species exposed to aromatics and phenols were derived from four studies that used these data for calibration of the OMEGA model (Hendriks, 1995a; Hendriks et al., 2001; Van der Linde et al., 2001; Veltman et al., 2005). Thus, the data set consisted of kinetic rates found in the literature and of rates used for OMEGA calibration.

To ensure independency, BCF values for oil constituents were searched for in scientific literature sources other than the sources containing absorption and elimination rate constants. In total, 528 BCF values were found for 42 aquatic species (including algae, annelids, crustaceans, diatoms, fish, insects and molluscs) exposed to 26 mono-, di- and polycyclic aromatic hydrocarbons (MAHs, DAHs and PAHs) and n-paraffins in the U.S. EPA Ecotox database (ECOTOX, 2012). The Handbook of Physical–Chemical Properties and Environmental Fate for Organic Chemicals (Mackay et al., 1992) and The National Library of Medicine's Hazardous Substances Data Bank (HSDB, 2012) provided nine studies with oil BCF data for algae, crustaceans, insects, fish and molluscs (Davies and Dobbs, 1984; Freitag et al., 1985; Herman et al., 1991; Lu et al., 1978; McCarthy et al., 1985; Melancon and Lech, 1978; Pedersen and Hill, 2002; Roubal et al., 1978; Tolls and van Dijk, 2002). The ISI Web of Knowledge database provided seven additional studies based on the following search terms: (1) oil, petroleum, aromatic, aliphatic, resin, phenol, alkane, alkene, alkyn, paraffin, thiophene, olefin, naphthenic mono- and di-aromatic and (2) bioconcentration factor and BCF (Baussant et al., 2001; Boese et al., 1999; Fan and Reinfelder, 2003; Mäenpää et al., 2009; Qin et al., 2010; Richter and Nagel, 2007; Yakan et al., 2011).

A contaminant was considered to be an oil constituent when included in the CONCAWE library of the PETROTOX model (PETROTOX, 2012) or when mentioned as such in the literature. Each oil constituent was assigned to one of five oil groups that were considered homogeneous with respect to their chemical structure, i.e. the number of aromatic rings (one, two and more than two, i.e. mono-, di-, and polycyclic aromatics), unbranched hydrocarbons (alkanes i.e. n-paraffins) and hydroxyl groups (phenols) (Reed et al., 2001). The number of data did not allow for a more specific classification. The molecular mass of the constituents typically ranged from 78–162, 128–204, 166–280, 94–220 and 170–310 Da for the MAH, DAH, PAH, phenol and n-paraffin groups, respectively, reflecting the number of (carbon) atoms the molecules were composed of. All oil groups included non-alkylated (C0) and alkylated (C1–C3) constituents, except for the n-paraffins.

For comparison, 253 absorption and 551 elimination rate constants and 143 BCFs for, respectively, 22, 57 and 17 aquatic species exposed to persistent organic compounds other than oil, such as biocides and ethers, were derived from four studies that used these data for calibration of the OMEGA model (Hendriks, 1995a; Hendriks et al., 2001; Van der Linde et al., 2001; Veltman et al., 2005).

### 2.2. Data treatment

The BCFs were based on parent and radiolabelled compounds measured in the water and in the whole organism or its organs, such as the liver and bile. BCFs based on species wet weight were divided by the fat fraction of the whole species or the species organs to normalize differences in lipid fractions between species. To include BCFs reported on dry weight as well, the values were converted with a species- specific dry-to-wet weight ratio or a default ratio for the species' taxonomic group (Table S1, Supporting Information). The geometric mean was used when multiple rate constants or BCF values were available for a single species and single constituent. All absorption and elimination rate constants and BCFs that were collected are available in the Supporting Information.

### 2.3. Model estimates

The OMEGA bioaccumulation model estimates the internal chemical concentration in an organism based on the uptake and elimination rate constants of the chemical. These rate constants are a function of the chemical property $K_{ow}$ and the species' wet weight, lipid content and trophic level (Hendriks et al., 2001). The current study estimates the absorption of a chemical via the water phase ($k_{0,in}$; μg L/μg kg wet weight·day$^{-1}$). Elimination from the species can be estimated via water ($k_{0,out}$), faeces ($k_{1,out}$) and dilution by biomass as a consequence of growth or reproduction ($k_{2,out}$). The total elimination rate constant is the sum of these three elimination rate constants ($\Sigma k_{j,out}$; kg/kg day$^{-1}$). Although measured total elimination rate constants can include elimination via biotransformation of chemicals in organisms, this route was not

included in the total elimination estimates due to a lack of data for the different taxonomic and oil groups (Van der Linde et al., 2001). The steady state BCF (μg L/μg kg lipid wt) is determined as the ratio between the estimated absorption and total elimination rate constants. A conceptual diagram of the OMEGA model, the model equations, parameter values and variables used are available in Text Section 1 and Table S2 in the Supporting Information.

To estimate rate constants and BCFs with the OMEGA model, all $K_{ow}$ values of the constituents included in the two empirical data sets were calculated with the KOWWIN model in the EPI Suite programme (EPI Suite, 2012). The wet weight, lipid fraction, taxonomic classification and trophic level of the species in the data sets were collected from the literature. If weight was not reported in the experimental study, a default adult weight was used, obtained from other studies or estimated from length-to-weight ratios (Hendriks et al., 2001). We assumed all oil constituents to accumulate in the lipid of the organisms. The lipid fractions of species and their tissues were available for 58% of the kinetic rate constants and 17% of the BCFs. These fractions ranged between 0.01 and 0.12 for fish, 0.003 and 0.07 for arthropods, 0.01 and 0.15 for molluscs and 0.01 and 0.08 for annelids. Default values based on the trophic level of species were used if no fat percentage was reported in the experimental study (i.e. 0.01 for unicellular organisms, 0.03 for annelids, arthropods and molluscs and 0.05 for fish, Table S2).

### 2.4. Evaluating model performance

The absorption and elimination rate constants and BCFs estimated with the OMEGA model were compared with the laboratory-derived values collected from the literature. These measured and estimated data were log-transformed.

First, the coefficient of efficiency $E$ (i.e. the predictive squared correlation coefficient $q^2$) was calculated, according to:

$$E = 1 - \frac{\sum_{i=1}^{n}(O_i - P_i)^2}{\sum_{i=1}^{n}(O_i - \bar{O})^2} \quad (1)$$

where $O_i$ is the observed value for case $i$, $P_i$ is the estimated value for case $i$, $n$ is the number of cases and $O$ denotes the mean of the observed values (Legates and McGabe, 1999). $E$ ranges from minus infinity to 1, with a value of 1 indicating perfect model estimation. A positive $E$ indicates that the model estimates rate constants and BCFs more accurately than the average of the observed values.

Second, an absolute error measure was obtained by calculating the average difference between the model estimates and measured values as the root-mean-square-error (RMSE):

$$\mathrm{RMSE} = \sqrt{\frac{1}{n} \times \sum_{i=1}^{n}(O_i - P_i)^2} \quad (2)$$

The RMSE summarizes both random error and systematic bias (Veltman et al., 2009).

Next, differences between estimated and measured absorption and elimination rate constants and BCFs were related to the water temperature, exposure duration, molecular mass of the constituent and biotransformation rates (fish taxa only), as these variables were not accounted for in the OMEGA model. A correction factor for temperature dependence of kinetic rate constants was already included in the model, but this multiplication factor was set at 1 due to a lack of experimental data (Hendriks et al., 2001). The exposure duration may be of relevance for BCF estimates, because steady state is not reached instantly. The molecular mass, a chemical property covarying with hydrophobicity (i.e. $K_{ow}$), the cross section and chain length of a molecule, has been suggested to influence bioaccumulation of organic constituents (Franke et al., 1994; Müller and Nendza, 2007). Finally, labile constituents may be biotransformed into metabolites that are more water soluble, and thus more susceptible to elimination (Newman and Unger, 2003).

Linear regression was applied to assess relationships between model performance and each of the four explanatory variables. Here, we expressed model performance as the ratio between log-transformed estimated and measured rate constants and BCFs, with a positive ratio indicating model overestimates. The geometric mean of rate constants and BCFs was determined prior to applying the linear regression to molecular mass and biotransformation rates. No geometric means were determined for water temperature and exposure duration, since different temperatures and durations were available for a single species and a single constituent. The significance of the linear trends was determined with the Student's $t$-test.

In literature, measured biotransformation rate data are lacking for most species and chemicals (Van der Linde et al., 2001). Estimated biotransformation rate constants for fish were therefore used to evaluate the performance of the model in relation to the biotransformation of chemicals. These rate constants were obtained for oil and non-oil organics from the biotransformation rate constant model in the EPI Suite programme (EPI Suite, 2012). This model estimates whole body primary rate constants for organic chemicals in a 1 kg fish based on the $K_{ow}$, the biological half-life and the molecular weight of a chemical (i.e. quantitative structure–activity relationship) (Arnot et al., 2009). The biotransformation rate constants for 1 kg fish were converted to values corresponding with the actual weight of the fish species by multiplying with weight$^{-\kappa}$, as rate constants scale to organism size with the exponent $-\kappa$ (Hendriks, 1999).

## 3. Results

Overall, model estimates for absorption ($k_{0,in}$) and elimination rate constants ($\Sigma k_{j,out}$) were more accurate than those for the BCFs of oil constituents. Coefficients of efficiency for absorption and elimination were positive, with $E = 0.51$ and 0.16 (Table 1), and model estimates differed by less than one order of magnitude from the measured data for 91% and 80% of the rate constants, respectively (Fig. 1). The average differences between the model estimates and measurements were a factor of 3.4 and 7.6 for absorption and elimination rate constants, respectively (factor = $10^{\wedge \mathrm{RMSE}}$, with RMSE values of 0.53 and 0.88; Table 1). In contrast, the coefficient of efficiency was negative for BCFs ($E = -0.20$) and model estimates differed by less than one order of magnitude from the measured data for 61% of the values (Fig. 1).

Model accuracy of absorption rate constants was high for PAH and phenol oil groups and for annelids, crustaceans and molluscs, based on a positive $E$ (0.42–0.62) (Figs. 1a and S1, Table 1). The uncertainty in the model, i.e. the RMSE, ranged from 0.29 to 0.59. Modelled elimination rate constants were accurate for hydrocarbons with two rings or more (DAHs and PAHs), and for crustaceans (Figs. 1b and S1, Table 1; $E = 0.08$–$0.65$ and RMSE = 0.67–1.08). Model estimates of BCFs were accurate for the Annelida and Chlorophyta groups ($E = 0.07$–$0.64$, RMSE = 0.31–0.91), but the $E$ was negative for all oil groups (Fig. 1c and Table 1). For example, the OMEGA model overestimated all six BCFs for $n$-paraffins by two to four orders of magnitude (Fig. 1c). There were no corresponding uptake or elimination data for n-paraffins so it was difficult to determine the sources of error in these model-data comparisons.

On the whole, absorption rate constants were more accurately predicted for oil constituents than for persistent non-oil organic constituents (Fig. 1a), as indicated by the higher goodness-of-fit measure $E$ and the lower absolute error measure RMSE for oil constituents ($E$ and RMSE were 0.51 and 0.53 for oil and 0.23 and 0.71

**Table 1**
The number of data ($n$), coefficients of efficiency ($E$) and the root-mean-square-errors (RMSE) of log-transformed absorption and elimination rate constants and bioconcentration factors (BCF) divided into various groups of oil and persistent non-oil organic constituents and taxonomic groups of aquatic species.

| Groups | Absorption ($k_{0,in}$) | | | Elimination ($\Sigma k_{j,out}$) | | | Bioconcentration factor[a] | | |
|---|---|---|---|---|---|---|---|---|---|
| | $n$ | $E$ | RMSE | $n$ | $E$ | RMSE | $n$ | $E$ | RMSE |
| Oil constituents | 120 | 0.51 | 0.53 | 165 | 0.16 | 0.88 | 168 | −0.20 | 1.26 |
| *Oil groups* | | | | | | | | | |
| MAHs | 2 | – | – | 3 | −0.87 | 0.65 | 16 | −0.92 | 0.95 |
| DAHs | 12 | −1.45 | 0.74 | 20 | 0.65 | 0.67 | 37 | −0.06 | 0.92 |
| PAHs | 102 | 0.62 | 0.47 | 135 | 0.08 | 0.90 | 103 | −0.49 | 1.22 |
| Phenols | 4 | 0.53 | 0.45 | 7 | −3.64 | 1.16 | 6 | −3.77 | 1.19 |
| n-Paraffins | – | – | – | – | – | – | 6 | −4.27 | 3.11 |
| *Taxonomic groups* | | | | | | | | | |
| Annelida | 9 | 0.42 | 0.59 | 14 | −0.34 | 0.43 | 6 | 0.64 | 0.31 |
| Chlorophyta | – | – | – | – | – | – | 8 | 0.07 | 0.91 |
| Crustacea | 39 | 0.51 | 0.55 | 40 | 0.31 | 1.08 | 41 | −0.24 | 0.92 |
| Insecta | 4 | −3.80 | 0.56 | 4 | −4.55 | 0.94 | 7 | −1.11 | 1.18 |
| Mollusca | 35 | 0.50 | 0.29 | 49 | −1.63 | 0.59 | 33 | −0.18 | 1.25 |
| Osteichthyes [b] | 33 | −0.53 | 0.66 | 58 | −2.45 | 1.00 | 73 | −2.18 | 1.49 |
| Non-oil organic constituents | 156 | 0.23 | 0.71 | 372 | 0.57 | 0.64 | 148 | −0.14 | 1.22 |

[a] Bioconcentration factors are lipid normalized.
[b] Taxonomic group that includes fish.

for non-oil, respectively; Table 1). On average, elimination rate constants were more accurately estimated for non-oil organics ($E = 0.57$, RMSE = 0.64) than for oil constituents ($E = 0.16$, RMSE = 0.88; Fig. 1b). For BCFs, the goodness-of-fit measure showed little difference between oil and non-oil organic constituents (Table 1).

The model performance of BCFs did not show a significant (i.e. $p < 0.05$) trend in relation to water temperature and exposure duration (with and without achieving steady state) (Fig. 2a and b; Table 2). The same held for absorption and elimination rate constants in relation to exposure duration (Fig. 3 and Table 3). There was a significant relationship between the model performance of kinetic rate constants and the water temperature (2–30 ℃), as the model tended to underestimate both absorption and elimination rate constants at higher temperatures (Fig. 3, Table 3). The performance of the model was also related to the molecular mass of the oil constituents (Fig. 2c and Table 2). On average, the model tended to overestimate absorption rate constants and BCFs for constituents with a molecular mass above 200 Da, whereas elimination rates tended to be underestimated for these constituents (Fig. 3 and Table 3). A similar trend was found for BCF model performance and $K_{ow}$, as 91% of the 43 BCF values were overestimated for oil constituents with a log $K_{ow} > 5.5$, i.e. PAHs, phenols and paraffins (Fig. S2). No significant relationship was found between the performance of the model for absorption ($p = 0.99$) and elimination rate constants ($p = 0.86$) and the molecular mass of non-oil organic constituents (Fig. 3). Finally, the ratio between estimated (excluding biotransformation) and measured (possibly including biotransformation) absorption and elimination rate constants was significantly related to the biotransformation rate constant (Fig. 3 and Table 3). The model increasingly overestimated absorption and total elimination rate constants at increasing biotransformation rates. The opposite was found for the BCFs (Fig. 2d and Table 2).

## 4. Discussion

### 4.1. Overall model performance

The OMEGA model estimated absorption and elimination rate constants of all the oil constituents more accurately than the average of the measurements. While for BCFs the average of the measured values was a better predictor than the model. The lower accuracy of the model for the BCFs than for the rate constants may be related to uncertainties in the data set, since the model performance was evaluated with all BCF values that resulted from our literature and database search. Thus, a range of different aquatic species, oil constituents and experimental conditions was included. Yet, no apparent differences were found in the extent of overestimates or underestimates between BCFs based on whole organisms versus organs or between BCFs based on steady state versus non-steady state (Fig. S3). Additionally, the BCF data (both those determined at steady state and non-steady state) were not related to the exposure duration of the experiments. For this analysis only 250 of the total 611 were used since for these data information on the level of steady state was reported. All the 611 BCFs were used to evaluate the performance of the model for oil constituents in order to cover a wider range of constituents and species. Additionally, measurements on both the radiolabelled and non-radiolabelled oil constituents were included in the analysis. In experiments, radiolabelling might lead to the overestimation of the actual amount of parent compounds present. In the current study, the model underestimated and overestimated experimental BCFs of both the radiolabelled and non-radiolabelled oil constituents (Fig. S3). Moreover, the BCF overestimates of radiolabelled constituents were probably biased by fish (Fig. S1).

### 4.2. Model performance in relation to water temperature

The performance of the model for BCFs did not show a trend in relation to the water temperature. In contrast, the bioaccumulation model tended to underestimate absorption and elimination rate constants for the higher temperatures. In other words, measured kinetic rates tended to increase slightly with increasing temperature. Yet, these trends explained only 5–6% of the variation in the estimated/measured ratios. Positive trends in relation to water temperature have also been observed for the BCF of hydrophobic organic chemical (HOC) chlorobenzene and the uptake of Bisphenol A in fish (Muijs and Jonker, 2009). However, the bioaccumulation factors of non-metabolizable HOCs related inversely to temperature, indicating a negative trend between temperature and uptake (Muijs and Jonker, 2009). Additionally, no trend in relation to water temperature was observed for the elimination rates and BCFs for the oil constituents pyrene and benzo(a)pyrene in the zebra mussel *Dreissena polymorpha* (Gossiaux et al., 1996). Based on the current study and literature, relationships between the kinetic rate constants and temperature are ambiguous, requiring in-depth attention in a separate study.

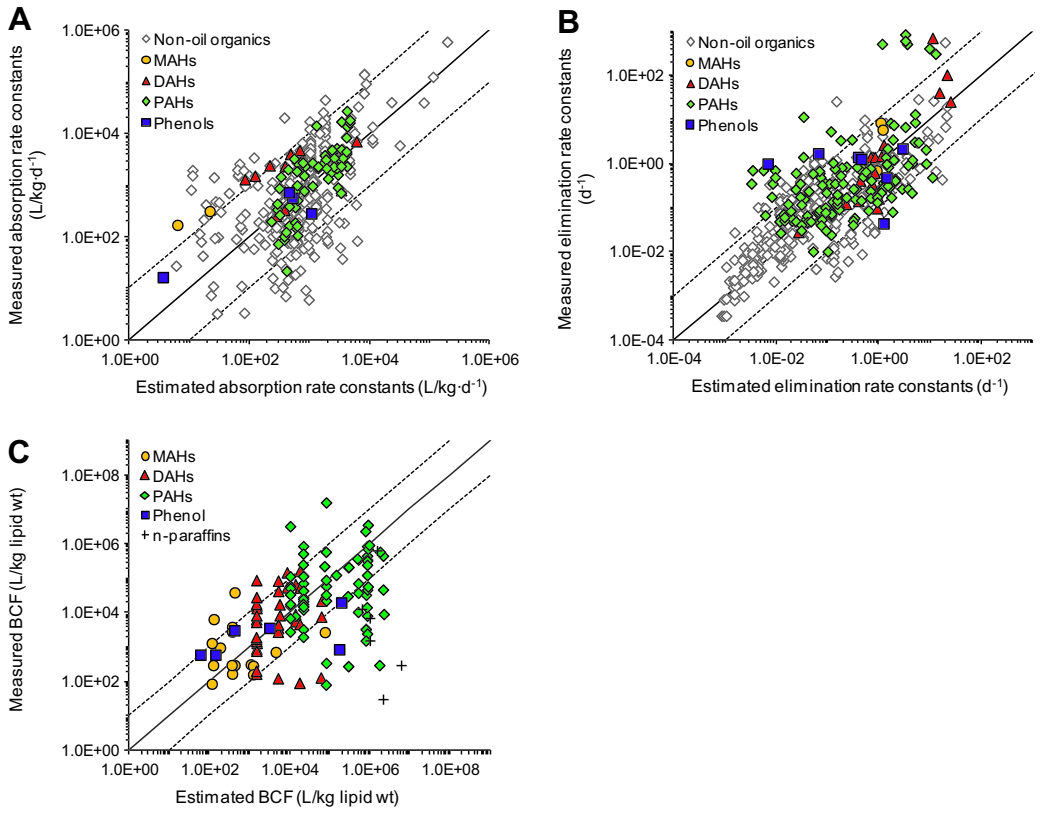


**Fig. 1.** Geometric mean for (A) absorption rate constants, (B) elimination rate constants, and (C) lipid-normalized bioconcentration factors (BCF) of organic constituents measured in experiments versus the geometric mean estimated with the OMEGA bioaccumulation model for aquatic species. The coloured diamonds represent different oil groups. The open diamonds represent persistent organic compounds other than oil that have been addressed in previous studies, e.g. biocides, ethers and halobiphenyls (Hendriks, 1995b; Hendriks and Heikens, 2001; Hendriks et al., 2001). The 1:1 line indicates a perfect model fit. The dashed lines represent a factor of 10 under- and overestimation by OMEGA.

### *4.3. Model performance in relation to molecular mass and biotransformation*

Absorption rate constants were less accurately estimated for DAHs than for PAHs. As the molecular mass for DAHs was 128–204 Da compared to 166–280 Da for PAHs, the tendency of the model to underestimate absorption rate constants for oil constituents with a relatively low molecular mass (approximately below 160 Da) might explain the less accurate DAH estimates. Although only two absorption rate constants for MAHs (78 and 92 Da) were available, these rates were also underestimated by the bioaccumulation model (Fig. 3). The performance of the model for BCFs corresponded with these findings, as BCFs were also underestimated at low molecular mass (Fig. 2).

Additionally, the 91% overestimated BCFs of oil constituents with a log $K_{ow} > 5.5$ implied that the performance of the model for relative hydrophobic compounds was influenced by variables not yet included in the model. One of these variables could be the molecular mass, as most BCFs were overestimated above 200 Da. Furthermore, biotransformation might play a role in the performance of the model, for example if the oil constituent is labile and the species has a biotransformation enzyme system. An increased biotransformation will increase the measured total elimination, causing the model to underestimate elimination rate constants and overestimate BCFs. Yet, in the current study a counterintuitive trend was demonstrated by the tendency of the model to overestimate elimination rate constants at high biotransformation rates (Fig. 3). The causes were unclear for this trend as well as for the remarkable relationship between the biotransformation rates and the performance of the model for absorption rates. The relationship between the model performance of BCFs and the biotransformation rate constants (Fig. 2) was highly dependent on two highly overestimated BCF values, namely for the n-paraffins docosane and hexadecane. After removal of these two values, the performance of the model for BCFs was no longer related to the biotransformation rate constants. More measured kinetic rate constants and BCFs are needed for an extensive evaluation of the performance of the model for oil constituents in relation to (measured) biotransformation rate constants.

In addition to linear regression, we used another method to test the accuracy of the model for oil in relation to biotransformation rates. Biotransformation by fish was added as a fourth elimination route to the OMEGA model by adding the obtained rate constants from the EPI Suite programme to the estimated total elimination rate constant $\Sigma k_{j,out}$. The modelled elimination rate constants (including biotransformation) were compared with measured elimination rate constants (possibly including biotransformation). Although the coefficient of efficiency E remained negative, the E increased from −2.45 to −1.25 (Table S3). Model performance particularly improved for elimination rate constants of PAHs ($E$: −6.57 to −2.21; Fig. S4), which is consistent with previous studies that showed fish to biotransform PAHs, such as benzo(a)pyrene, fluoranthene and benzo(a)anthracene (Bechmann et al., 2010; Moermond et al., 2007). The performance of the model for BCFs improved correspondingly after incorporating biotransformation rate constants in the model. The goodness-of-fit $E$ increased from −2.10 to −0.34 for all oil constituents (Table S4). It may be concluded that in general a minor improvement will be achieved from incorporating biotransformation rates to the model, but the model accuracy can increase for fish exposed to PAHs.

Estimates of elimination rate constants were more accurate for molluscs than for fish, but the model performed best for

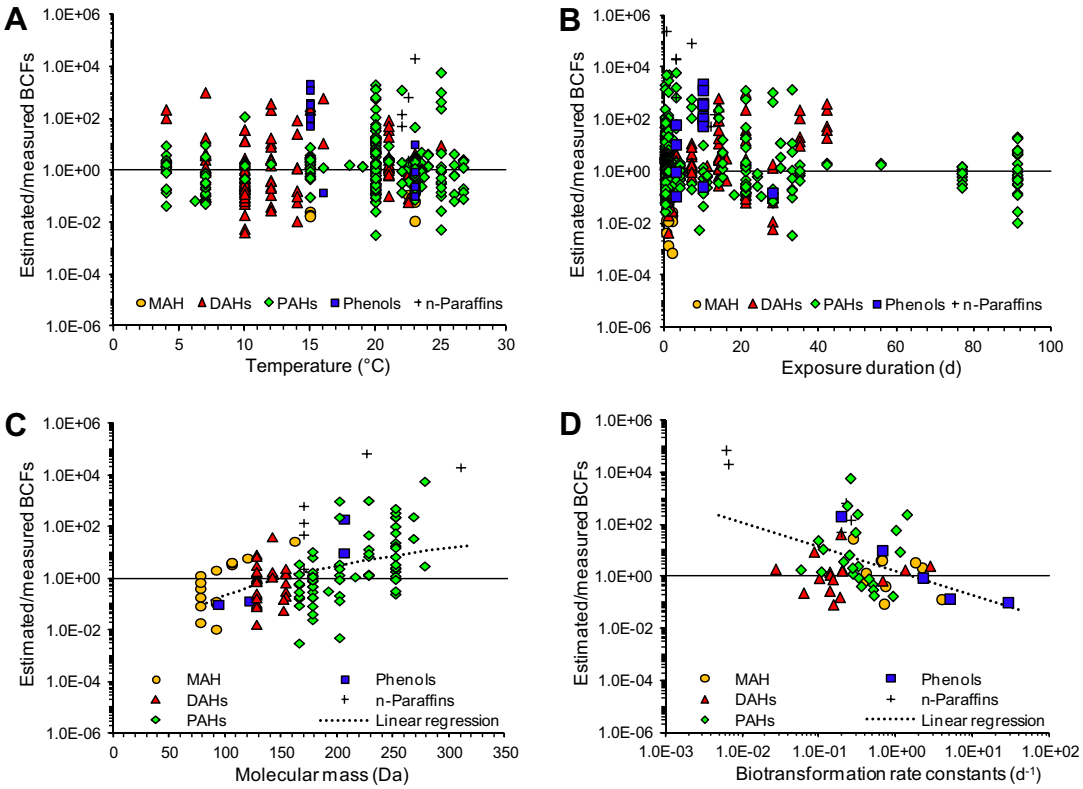


**Fig. 2.** Ratio between estimated and measured bioconcentration factors (BCFs) of aquatic species exposed to oil constituents in relation to (A) water temperature, (B) exposure duration, C) the molecular mass of oil constituents and (D) biotransformation rate constants for fish as obtained from the EPI Suite program (Arnot et al., 2009; EPI Suite, 2012). The horizontal line indicates a perfect model fit. The dotted lines represent linear regression models ($p < 0.05$) fitted through all data points of the oil constituents.

**Table 2**
The coefficient ($\alpha$), intercept ($\beta$), coefficient of determination ($R^2$), $p$-value and the number of data ($n$) for the linear regression log(estimated/measured) bioconcentration factor = $\alpha \log x + \beta$ for four explanatory variables (denoted by $x$).

| Variable ($x$) | $\alpha$[a] | $\beta$[a] | $R^2$ | $p$-Value [b] | $n$ |
|---|---|---|---|---|---|
| Temperature (K) [c] | 7.12 [−3.92; 18.16] | −17.35 [−44.54; 9.84] | <0.01 | 0.21 | 398 |
| *Exposure duration (d)*[d] | | | | | |
| Steady state | −0.06 [−0.23; 0.11] | 0.18 [0.07; 0.29] | <0.01 | 0.49 | 140 |
| Non-steady state | 0.15 [−0.19; 0.50] | −0.39 [−0.57; −0.21] | <0.01 | 0.38 | 100 |
| Information on steady state lacking | −0.29 [−0.44; −0.13] | 0.66 [0.49; 0.83] | 0.04 | <0.01 | 351 |
| Molecular mass (Da) [e] | 3.84 [2.54; 5.14] | −8.34 [−11.26; −5.43] | 0.19 | <0.01 | 149 |
| Biotransformation rate constants ($d^{-1}$) [e,f] | −0.94 [−1.42; −0.46] | 0.19 [−1.18; 0.56] | 0.21 | <0.01 | 61 |

[a] The coefficient ($\alpha$) or intercept ($\beta$) and the [lower; upper] 95% confidence interval.
[b] Student's $t$-test.
[c] The water temperature was converted from Celsius to Kelvin prior to log transformation of the data.
[d] The data set was divided according to the information available on chemical steady state.
[e] In case of multiple BCF values for a single species and a single constituent, the geometric mean was determined.
[f] Only fish were included.

crustaceans. Differences among species in the ability to biotransform hydrocarbons may contribute to these differences in the model performance between taxonomic groups. For example, the biotransformation rates of several PAHs were reported to be higher in vertebrates (e.g. fish) than invertebrates (e.g. molluscs and crustaceans) (Bechmann et al., 2010; Moermond et al., 2007), which may explain the more severely underestimated elimination rates for fish.

### 4.4. Implications and recommendations

The OMEGA bioaccumulation model predicted absorption and elimination rate constants for aquatic species exposed to oil constituents with an accuracy that is consistent with other bioaccumulation models that focus on chemicals other than oil constituents (Arnot and Gobas, 2004; Barber et al., 1991; Thomann et al., 1992). Inclusion of more explanatory variables in the bioaccumulation model can improve the performance of the model. Firstly, correcting absorption rate constants for molecular mass may improve model performances for oil constituents with a relatively low mass (e.g. MAHs and DAHs). The molecular mass could be added as a variable influencing the lipid layer permeation resistance. For example, Gobas and Opperhuizen (1986) related the lipid layer permeation rate to the solute's membrane-water partition coefficient and to factors affecting the diffusion coefficient in the membrane layer, such as the molecular mass (Gobas and Opperhuizen, 1986). In addition to molecular mass, the molecular cross section and chain length have also been suggested to

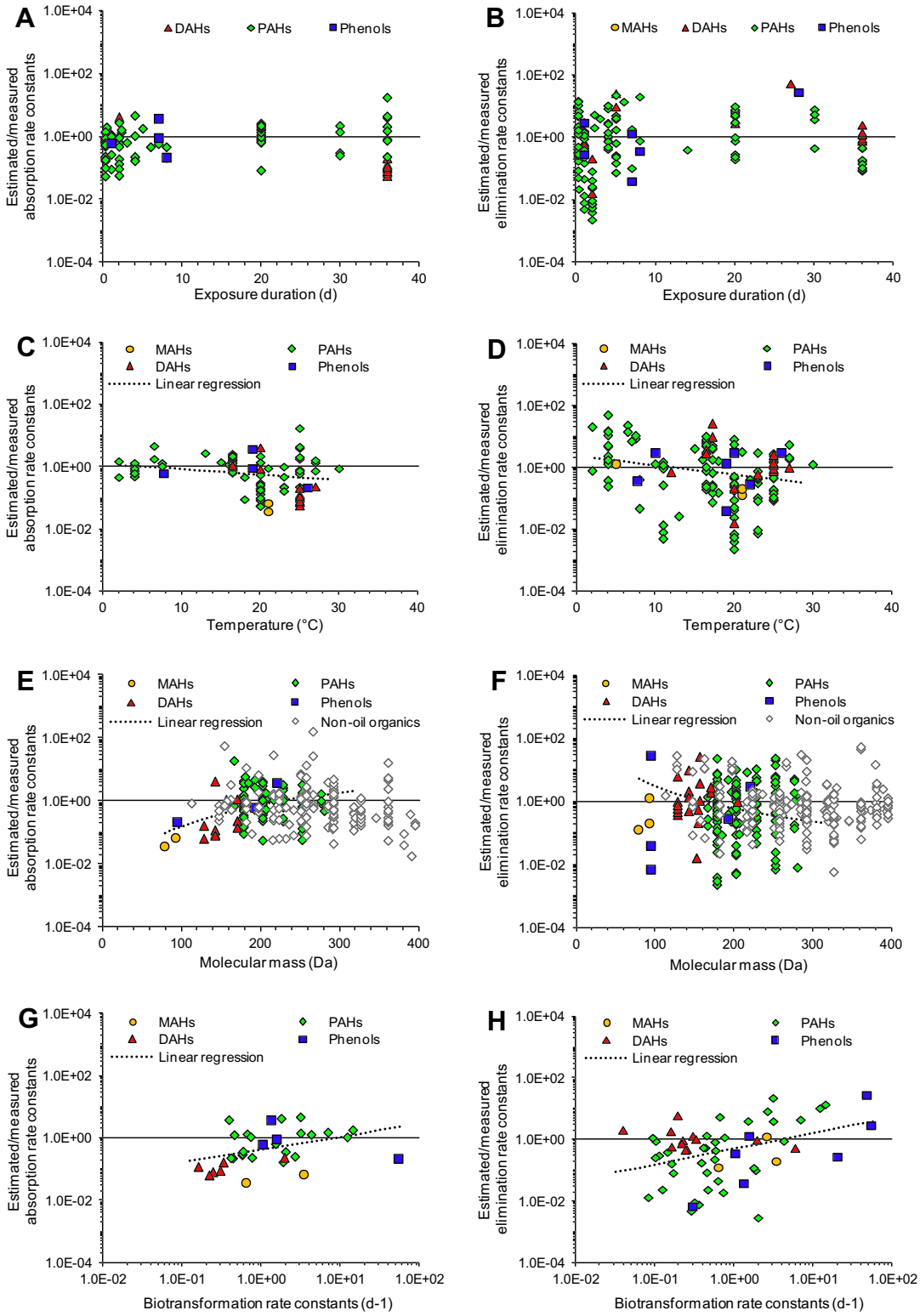


**Fig. 3.** Ratio between estimated and measured absorption (A, C, E, G) and elimination (B, D, F, H) rate constants of aquatic species exposed to oil constituents versus exposure duration (A and B), water temperatures (C and D), molecular mass of oil constituents (E and F), and biotransformation rate constants for fish as obtained from the EPI Suite program (G and H) (Arnot et al., 2009; EPI Suite, 2012). The horizontal line indicates a perfect model fit. The dotted lines represent linear regression models ($p < 0.05$) fitted through all data points of the oil constituents.

influence the bioaccumulation of organic constituents (Franke et al., 1994; Müller and Nendza, 2007). In general, a diameter above 0.95 nm and a chain longer than approximately 4.3 nm may cause a decreased membrane permeation of organic chemicals (Müller and Nendza, 2007). The majority of crude oils contain straight-chained hydrocarbons (i.e. n-paraffins) that can be up to 35 carbon atoms long (Mendelssohn et al., 2012). Secondly, relating the absorption and elimination rate constants to the water

**Table 3**
The coefficient ($\alpha$), intercept ($\beta$), coefficient of determination ($R^2$), $p$-value and the number of data ($n$) for the linear regression log(estimated/measured) absorption and elimination rate constant = $\alpha \log x + \beta$ for four explanatory variables (denoted by $x$).

| Rate constant | Variable ($x$) | $\alpha$[a] | $\beta$[a] | $R^2$ | $p$-Value [b] | $n$ |
|---|---|---|---|---|---|---|
| Absorption | Temperature (K) [c] | −11.33 [−20.66; −2.00] | 27.69 [4.69; 50.68] | 0.05 | 0.02 | 104 |
| | Exposure duration (d) | 0.10 [−0.02; 0.22] | −0.26 [−0.38; −0.14] | 0.02 | 0.11 | 103 |
| | Molecular mass (Da) [d] | 2.27 [1.38; 3.16] | −5.35 [−7.40; −3.30] | 0.18 | <0.01 | 119 |
| | Biotransformation rate constant ($d^{-1}$) [d,e] | 0.41 [0.07; 0.74] | −0.37 [−0.56; 0.18] | 0.16 | 0.02 | 33 |
| Elimination | Temperature (K) [c] | −20.01 [−32.79; −7.24] | 49.14 [17.67; 80.60] | 0.06 | <0.01 | 158 |
| | Exposure duration (d) | 0.18 [−0.02; 0.38] | −0.26 [−0.44; −0.08] | 0.02 | 0.07 | 149 |
| | Molecular mass (Da) [d] | −2.36 [−3.60; −1.12] | 5.21 [2.36; 8.05] | 0.08 | <0.01 | 164 |
| | Biotransformation rate constant ($d^{-1}$) [d,e] | 0.51 [0.17; 0.84] | −0.30 [−0.54; −0.07] | 0.14 | <0.01 | 58 |

[a] The coefficient ($\alpha$) or intercept ($\beta$) and the [lower; upper] 95% confidence interval.
[b] Student's $t$-test.
[c] The temperature was converted from Celsius to Kelvin prior to log transforming the data.
[d] In case of multiple values for a single species and single constituent, the geometric mean was determined.
[e] Only fish were included.

temperature may slightly improve the kinetic rate estimates. Thirdly, biotransformation could be added as an additional elimination route when estimating the bioaccumulation of oil constituents. This will probably improve model estimations for taxonomic groups that are able to biotransform labile constituents, as shown in the current study for fish exposed to PAHs.

In the future, model performances could be evaluated in relation to these physico-chemical properties if more empirical data become available on for instance n-paraffins and other aliphatics. In the current study, the general applicability of the model to oil constituents and the influence of variables on the model were evaluated with all kinetic rate constants and BCFs resulting from our literature and database search. Alternatively, the performance can be evaluated by simulating the kinetics of one chemical in one species and comparing it to experimental data. Afterwards, a sensitivity analysis may be carried out to identify the parameters that require most attention.

## Acknowledgements

F.D.L. is a postdoctoral research fellow from the Fund for Scientific Research (FWO, Flanders). We thank Chiel Jonker, Eric Verbruggen, Aaron Redman and Duncan King for their suggestions and sharing their knowledge on oil accumulation. We thank the Norwegian Research Council (NRC) for support through the PETROMAKS program (BIP project #ES468602). The SYMBIOSES project is a cooperation of 15 research partners, financed by the NRC, BP Exploration Operating Company Limited, ConocoPhillips Skandinavia, ExxonMobil Upstream Research Company, Eni Norway, Shell Technology Norway, Statoil Petroleum and Total E&P Norway.

## Appendix A. Supplementary material

The supporting information includes the empirical data sets for absorption and elimination rate constants and BCF values (Excel file), a description of the OMEGA model (Text Section 1, Table S1), dry to wet weight ratios (Table S2), measured versus estimated kinetic rate constants and BCFs divided into taxonomic groups (Fig. S1), a relationship of BCFs to the $K_{ow}$ (Fig. S2), measured versus estimated BCFs divided into steady state and radiolabelled groups (Fig. S3), measured versus estimated elimination rate constants for fish (Fig. S4) and the corresponding linear regression equations and goodness of fit measures (Tables S3–S4). Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2013.09.006.

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