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Modelling bioaccumulation of oil constituents in aquatic species

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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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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