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A dynamic and mechanistic model of PCB bioaccumulation in the European hake (*Merluccius merluccius*)

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ABSTRACT

Bioaccumulation is difficult to document because responses differ among chemical compounds, with environmental conditions, and physiological processes characteristic of each species. We use a mechanistic model, based on the Dynamic Energy Budget (DEB) theory, to take into account this complexity and study factors impacting accumulation of organic pollutants in fish through ontogeny. The bioaccumulation model proposed is a comprehensive approach that relates evolution of hake PCB contamination to physiological information about the fish, such as diet, metabolism, reserve and reproduction status. The species studied is the European hake (*Merluccius merluccius*, L. 1758). The model is applied to study the total concentration and the lipid normalised concentration of 4 PCB congeners in male and female hakes from the Gulf of Lions (NW Mediterranean sea) and the Bay of Biscay (NE Atlantic ocean). Outputs of the model compare consistently to measurements over the life span of fish. Simulation results clearly demonstrate the relative effects of food contamination, growth and reproduction on the PCB bioaccumulation in hake. The same species living in different habitats and exposed to different PCB prey concentrations exhibit marked difference in the body accumulation of PCBs. At the adult stage, female hakes have a lower PCB concentration compared to males for a given length. We successfully simulated these sex-specific PCB concentrations by considering two mechanisms: a higher energy allocation to growth for females and a transfer of PCBs from the female to its eggs when allocating lipids from reserve to eggs. Finally, by its mechanistic description of physiological processes, the model is relevant for other species and sets the stage for a mechanistic understanding of toxicity and ecological effects of organic contaminants in marine organisms.

1. Introduction

Assessment of bioaccumulation and effects of pollutants in marine species is a fundamental step for environmental management. The development of a methodological framework for assessing bioaccumulation is therefore essential for the scientific evaluation of risks that chemicals may pose to the environment and by enlargement to humans. However, in the case of organic contaminants (OCs), bioaccumulation is difficult to document because responses differ among chemical compounds, with environmental conditions, and physiological processes characteristic of each species (Gobas, 1993; Connolly and Glaser, 2002). In early empirical approaches, the quantification of the bioaccumulation phenomena was deduced from concentrations in the organism (e.g. fish) and concentration of their prey and in water, to calculate bioaccumulation and bioconcentration factors (BAF and BCF). However, interpreting bioaccumulation data reveals difficulties related to the contaminant dynamics, possible changes in the environment (temperature, trophic conditions, prey contamination levels) and interactions with the physiology of animals.(Mackay and Fraser, 2000). To improve those tools, it is important to better characterize the dominant underlying processes driving the bioaccumulation phenomenon. Physiological-based models, (Norstrom et al., 1976; Gobas, 1993; Loizeau et al., 2001) take into account some of these processes. Gobas (1993) consider for example that contaminants may be taken up from food and from seawater by transfer through the gills, and lost by fecal egestion, metabolic degradation, and again, by transfer through the gills. Contaminant dilution resulting from growth was also considered as a pseudo-elimination process. These studies assume implicitly steady-state conditions for the physiological processes in the organism. But the kinetics and concentrations of the xenobiotic are affected by the changing physiological condition of fish. Consequently, these models do not always fit well the data, especially in conditions where the physiological state (*ie.* size, energy reserve and reproductive cycle) of

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the organism is changing (van Haren et al., 1994). Moreover, their lack of explicit physiological formulation make impossible a mechanistic understanding of the toxic effects of contaminants on organism physiology. Unifying concepts are needed to better tie together such complexities (Connolly and Glaser, 2002).

The uptake/elimination model proposed by Kooijman and van Haren (1990) for organic contaminants such as PAHs (Polycyclic Aromatic Hydrocarbons) and PCBs (PolyChlorinated Biphenyls) has been designed to account for changes in the physiological conditions of the organism. It is based on the Dynamic Energy Budget (DEB) theory (Kooijman, 2000) which describes mechanistically growth, reserve dynamics and reproduction as a function of body size. Such a model is presented below for fish. The purpose of this paper is to test the applicability to fish of the DEB-based model to assess uptake-excretion kinetics for OCs by taking example of PCBs bioaccumulation in the European hake (*Merluccius merluccius* L. 1758). These contaminants are considered among the most dangerous pollutants because of their toxicity (Ahlborg and Hanberg, 1994; Loonen et al., 1996), stability, long biological half-life, and high liposolubility. They may also be bioaccumulated and biomagnified along food chains involving a wide range of trophic levels, with a potential risk for high trophic level predators (Borga et al., 2001; Hoekstra et al., 2003; Nfon and Cousins, 2006). Having a high trophic level (Ferraton et al., 2007), the European hake is exposed to the bioaccumulation of these organic compounds (Bodiguel et al., 2008). The model was built by coupling a PCB kinetic model through an organism to the standard DEB model. It was calibrated and tested on field data from the Gulf of Lions (Mediterranean sea) and the Bay of Biscay (Atlantic ocean). DEB parameters were estimated with field and experimental data or found in literature. In the model, we took into account contaminant uptake from food, contaminant losses due to spawning and we assessed the influence of physiological variables, such as body size and lipid content on PCB kinetics. Moreover, biotransformation of organic xenobiotics can affect elimination kinetics. But biotransformation increases the complexity of the analysis of the already complicated toxico-kinetic studies. Therefore, to simplify the analysis, we studied toxico-kinetics without biotransformation.Concerning PCBs, this phenomenon is negligible because of the high stability of these compounds. We also assumed that the physiological condition is not affected by the compounds although, one of the major interest of the DEB-based model is to offer a useful basis to describe such effects.

2. Method

2.1. Sampling and chemical analysis

The study areas are the Gulf of Lions, and the northern part of the Bay of Biscay, two regions off the French coasts (Fig. 1). The Gulf of Lions, situated in the NW Mediterranean, is one of most important continental shelf in the Mediterranean sea (Roussiez et al., 2005). It receives high freshwater discharges from the Rhône River watershed, which drains heavily industrialized regions of southern France. The Bay of Biscay, situated in the NE Atlantic, receives freshwater discharges from the Loire River, in western France. Hakes and its various preys were caught during bottom-trawl cruises, on the continental shelves (between 50 and 120 meters depth), in spring 2001, spring 2002 and autumn 2002 in the Bay of Biscay, and in spring 2004 and autumn 2004 in the Gulf of Lions. Largest specimens were caught on the edge of the continental shelf by gillnets, in autumn 2006 in the Gulf of Lions. A total of 202 hakes with total length (L) from 7 to 70 cm and 59 prey pools (sardines, anchovies, pouts, blue whitings, horse mackerels, gobiids, shrimps and zooplankton) were sampled for organic contaminant analysis. Fish were primarily pre-treated in the laboratory for total body weight, total length and macroscopic sex determination for hakes.

Chemical analysis have been described previously (Bodiguel et al., 2008) and are summarized below. The various biological species were solvent extracted using Soxtec apparatus. For samples from the gulf of Lions, extracts were weighed to give the amount of extractible material and thus a rough estimation of the total fat content. Appropriate clean-up of the extracts was then performed before the final instrumental analysis by gas chromatography with electron capture detector, on a HP 5890 series II equipped with a CP-Sil19 capillary column following optimized conditions described by Jaouen-Madoulet (2000). PCBs were quantified individually using a standard solution. Finally, concentrations of the different PCB congeners are available for each prey pool and at the individual level for hakes, for both sexes in the Gulf of Lions but without sex distinction in the Bay of Biscay. 4 PCB congeners described as few metabolisable were chosen because of different chlorination levels, two penta-chlorinated biphenyls: CBs 101 and 118, one hexa-chlorinated biphenyl: CB153, and one hepta-chlorinated biphenyl: CB180.

2.2. Model structure

Our bioenergetic approach is based on a mechanistic model, the DEB theory (Kooijman, 2000). It describes the acquisition and expenditures of energy in an organism, and the consequences for physiological organisation throughout its life cycle, in a potentially variable environment. A kinetic model of PCBs was then built, coupled to the bioenergetic one. It integrates the effects of growth and incorporates the chemical affinity of PCBs to reserve lipids. It is applied by taking the example of bioaccumulation of 4 PCB congeners (CBs) in the European hake (*Merluccius merluccius*, L.) from the Gulf of Lions (Mediterranean sea) and the northern part of the Bay of Biscay (Atlantic ocean). The 4 CBs were chosen among the seven indicators currently measured in pollution monitoring programs: CB101 (2,2′,4,5,5′-pentachlobiphenyl), CB118 (2,3′,4,4′,5-pentachlobiphenyl), CB153 (2,2′,4,4′,5,5′-hexachlobiphenyl) and CB180 (2,2′,3,4,4′,5,5′-heptachlobiphenyl). They describe a large range of chlorination (from 5 to 8 chlorine molecules on the biphenyl core).

2.2.1. DEB growth model

DEB models are based on simple assumptions to describe the rates at which individual organisms assimilate and utilize energy from food,

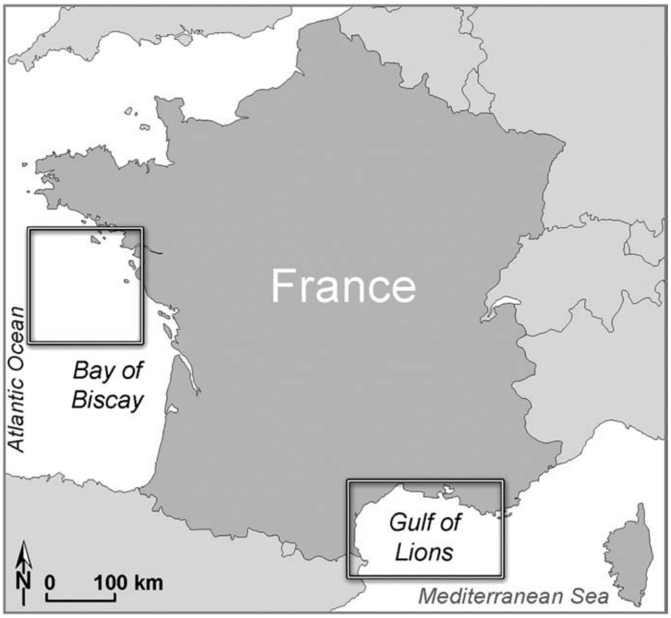

Fig. 1. Map of the study area showing the two sampled sites: the Bay of Biscay and the Gulf of Lions.