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An integrated approach for bioaccumulation assessment in mussels: Towards the development of Environmental Quality Standards for biota

J.M. Zaldívar [a,*], D. Marinov [b], S. Dueri [c], J. Castro-Jiménez [d], C. Micheletti [a], A.P. Worth [a]

[a] European Commission, Joint Research Centre, Institute for Health and Consumer Protection, via E. Fermi 2749, 21027 Ispra (VA), Italy
[b] European Commission, Joint Research Centre, Institute for Environment and Sustainability, via E. Fermi 2749, 21027 Ispra (VA), Italy
[c] Institut de Recherche pour le Développement, Avenue Jean Monnet BP 171, 34203 Sète CEDEX, France
[d] Environnement Industriel et Risques Industriels et Naturels (LGEI), Ecole des Mines d'Alés, 6 Avenue de Clavières, 30319 Ales CEDEX, France

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ABSTRACT

The possible use of chemical concentrations measured in mussels (*Mytillus galloprovincialis*) for compliance checking against Environmental Quality Standards (EQS) established for biota is analyzed with the help of an integrated model. The model consists of a 3D planktonic module that provides biomasses in the different compartments, i.e., phytoplankton, zooplankton and bacteria; a 3D fate module that provides the concentrations of contaminants in the water column and in the sediments; and a 3D bioaccumulation module that calculates internal concentrations in relevant biotic compartments. These modules feed a 0D growth and bioaccumulation module for mussels, based on the Dynamic Energy Budget (DEB) approach. The integrated model has been applied to study the bioaccumulation of persistent organic pollutants (POPs) in the Thau lagoon (France). The model correctly predicts the concentrations of polychlorinated biphenyls (PCBs) and polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) in mussels as a function of the concentrations in the water column and in phytoplankton. It also sheds light on the origin of the complexity associated with the use of EQS for biota and their conversion to water column concentrations. The integrated model is potentially useful for regulatory purposes, for example in the context of the European Water Framework (WFD) and Marine Strategy Framework Directives (MSFD).

1. Introduction

Mussels are frequently employed in contaminant monitoring programmes in transitional, coastal and marine waters. For example, the US Mussel Watch program (Kimbrough et al., 2008) has been monitoring trace metals and organic contaminants in mussels at several estuaries and coastal sites in the US since 1986, and now it covers approximately 140 chemicals. The OSPAR (OSPAR, 1999), HELCOM and Barcelona Conventions have included mussels among the species analyzed in the assessment of chemical contamination in coastal ecosystems (Roose and Brinkman, 2005). In addition, there are several European monitoring programmes in the Member States, e.g. RINBIO (Réseaux Intégrateurs Biologiques, France).

Monitoring contaminant concentrations in mussels has some advantages compared with the measurement of the total contaminant concentration in the water column, especially in the case of hydrophobic compounds that bioaccumulate in the food web and which are found at very low levels. The accumulation of organic contaminants in the tissues of mussels (or other biota) is a time-integrated indicator of pollutant occurrence, bioavailability and its distribution in aquatic ecosystems (Goldberg, 1986; Pereira et al., 1996). In particular, molluscs have been used as bioindicators of pollution in coastal ecosystems because of their feeding behavior and their limited mobility, which make them particularly exposed to contamination both via the water column and sediment, either directly or after resuspension. Mussels are farmed for human consumption and therefore, if contaminated, pose a potential risk for human health.

Although seafood represents a significant means of contamination in the human diet, to date few legal thresholds have been established to protect human health from toxic compounds and their mixtures. The European Commission (EC) introduced several laws to regulate the water quality of bivalves farming zones (EC, 1991) and to restrict farming, transport and purchase (EC, 1979, 1989). However, these rules refer only to microbiological contamination. More recently, several pieces of EC legislation have been put in place to regulate several families of organic contaminants such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), dioxin-like polychlorinated biphenyls (DL-PCBs), polycyclic aromatic hydrocarbons (PAHs) (EC, 2006), and metals such as lead, cadmium, mercury (EC, 2005) for several aquatic species such as fish, molluscs, crustaceans and cephalopods. To supplement the Water Framework Directive (EC, 2000), a new directive has been approved (EC, 2008a) to establish Environmental Quality Standard

* Corresponding author. Fax: +39 0332789328.
E-mail address: jose.zaldivar-comenges@jrc.ec.europa.eu (J.M. Zaldívar).

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(EQS) limits for 33 priority substances and 8 priority hazardous substances in surface waters, but also, for some of these compounds, in sediment and biota. For example EQS of 10 and 55 μg/kg (wet weight in prey tissue), have been established for hexachlorobenzene and hexachlorobutadiene, respectively.

In the risk management of hazardous chemicals, the prediction of bioconcentration and bioaccumulation factors in aquatic organisms has become a very important element in the assessment of environmental and human health effects. A quantitative assessment of uptake, metabolism, excretion, and depuration processes is needed to predict the fate and bioaccumulation of contaminants in the food web (Moriarty and Walker, 1987). These processes are related to specific physiological characteristics, feeding behavior and metabolism of the aquatic organism, to the physico-chemical characteristics of the compound, and to the environmental conditions of the aquatic system in which the organisms resides. Due to the complex influence of these factors, it is difficult to make comparisons between different field studies and results may appear contradictory.

Consequently, it is desirable to have a general modeling tool able to simulate field and laboratory toxicological experiments and integrate all the results into a predictive assessment of the ecotoxicological behavior of a certain substance. In addition, models may complement field studies and monitoring activities, and help to understand the transport and fate of contaminants (Carafa et al., 2006; Jurado et al., 2007; Dueri et al., 2009b, 2010) and their impacts on communities and ecosystems (Carafa et al., 2009; Bacelar et al., 2009; Dueri et al., 2009a; Marinov et al., 2009).

The overall objective of this work was to investigate the problems in developing EQS for biota (*concentrations that should not be exceeded to protect human health and the environment*) and in using these values for compliance checking in the context of the Water Framework Directive (WFD) and the Marine Strategy Framework Directive, MSFD (EC, 2008b). The development of EQS values for biota is particularly important for certain hydrophobic compounds for which bioaccumulation and biomagnification in the food chain may occur. To investigate these issues, we have developed an integrated modeling approach for calculating the contamination level in mussels (*Mytilus galloprovincialis*) from the pollutant concentration values in the water column. This modeling framework has been applied to the Thau lagoon (France) to predict the tissue concentration of selected PCB (28, 52, 101, 118, 138, 153 and 180) and PCDD/F (PeCDD, OCDD, TCDF, PeCDF and HxCDF) congeners. The modeling framework, consisting of 3D hydrodynamics, fate, planktonic and bioaccumulation modules, provides: (a) temperatures and chemical concentrations in the water column, (b) biomasses in the different compartments, i.e., phytoplankton, zooplankton and bacteria (Marinov et al., 2009; Dueri et al., 2010) and (c) internal concentrations of contaminant in the planktonic compartments. These values are used to feed a 0D growth and bioaccumulation model for mussels based on the Dynamic Energy Budget approach (DEB) (Kooijman, 2000; Kooijman and van Haren, 1990; Van Haren et al., 1994).

The Thau lagoon was selected because, besides its ecological interest as a recruitment zone for various species of sea fish, it is of notable economic importance due to shellfish cultivation (about 15 000 tons per year, amongst the highest in the Mediterranean Sea) and it has been extensively studied during the last 20 years (Amanieu et al., 1989; Picot et al., 1990; Plus et al., 2006 and references therein). Even though various numerical models have been developed, focusing on the lagoon hydrodynamics (Lazure, 1992), the nitrogen and oxygen cycles (Chapelle, 1995; Chapelle et al., 2001), the plankton ecosystem (Chapelle et al., 2000), the impact of shellfish farming (Bacher et al., 1997; Gangery et al., 2004a, b) and the macrophytes (Plus et al., 2003a, b); no model has been developed to support the study of the fate and effects of organic contaminants.

Furthermore, for over 20 years IFREMER has been coordinating a monitoring program (RNO, Réseau National d'Observation de la qualité du milieu marin) aimed at evaluating the levels and trends of chemical contaminants in the marine environment (http://www.ifremer.fr/envlit/surveillance/rno.htm). As a consequence, there is a database containing historical and present contaminant levels, as well as the geographical contaminant distribution in the French coastal waters for PAHs, PCBs PBDEs, OCPs and PCDD/Fs in mussels and sediments during the last decades (Johansson et al., 2006; Munschy et al., 2008).

2. Methods and approach

2.1. Study area

The Thau lagoon is 25 km long, 5 km wide and on average 4 m deep. The lagoon is located on the French Mediterranean coast (Fig. 1) and is sheltered by two narrow sea mouths. The catchment area is small (280 km^2) and drained by numerous small streams with intermittent flows. The climate imposes a wide range of water temperatures and salinities with minima of 5 °C in February and salinity near 27‰, and maxima of 29 °C in August and a salinity of 40‰. Precipitation also shows large interannual variation (from 200 to 1000 mm per year). The wind is often strong, particularly when it blows from the Northwest (the so called "Tramontane"). The Thau lagoon hydrodynamics are heavily influenced by wind and precipitation (Lazure, 1992). During the summer, it frequently undergoes anoxia that can lead to important economic losses.

In terms of experimental measurements, there are temporal time series for polycyclic aromatic hydrocarbons (PAHs), PCBs, polybrominated diphenyl ethers (PBDEs), organochlorine pesticides (OCPs) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in mussels and sediments during the past few decades (Munschy et al., 2008; Castro-Jiménez et al., 2008). All these contaminants show a decreasing trend with the exception of PBDEs. This decreasing trend has significantly slower rates in sediments than in mussels, for example the half-lives ($t_{1/2}$) for ΣPCBs are 8 and 32 years in mussels and sediments, respectively (Munschy et al., 2008). As an example, Fig. 2 shows some of the observed trends at two stations (in Zones A and C, see Fig. 1) for PCB 153.

2.2. Bioaccumulation

Bioconcentration refers to the accumulation of a substance dissolved in water by an aquatic organism. The bioconcentration factor (*BCF*) of a compound is defined as the ratio of concentrations of the chemical in the organism (C_b) and in water (C_w) at equilibrium; normally C_w is the dissolved water concentration:

$$BCF = \frac{C_b}{C_w} \quad (1)$$

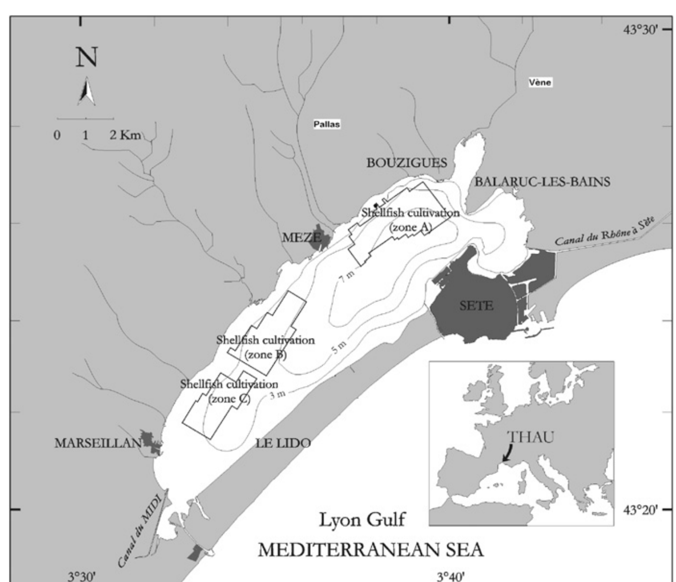

Fig. 1. The Thau lagoon and its watershed. Connections with the Mediterranean Sea are located at the extremities: in the Sète city and near Marseillan village.