Science of the Total Environment 563–564 (2016) 396–404

Contents lists available at ScienceDirect

Science of the Total Environment

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

Predicting the bioaccumulation of polyaromatic hydrocarbons and polychlorinated biphenyls in benthic animals in sediments

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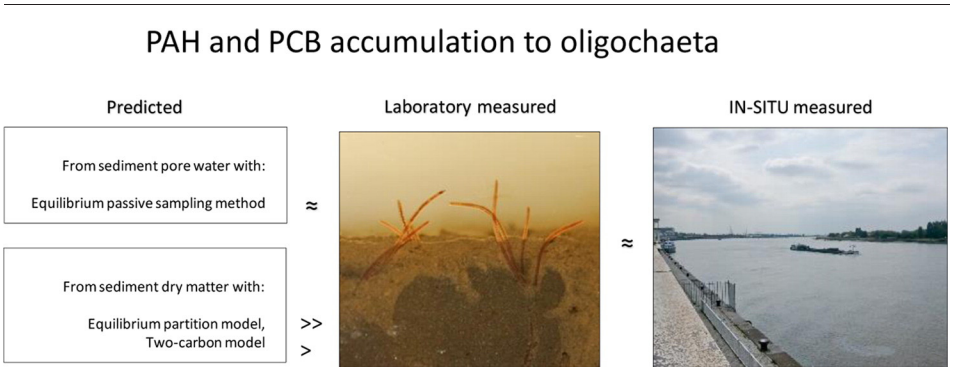
HIGHLIGHTS

- Determination of exposure is needed for estimation the risks of polluted sediments.
- Efficiency of different methods in predicting PAH and PCB accumulation was compared.
- Accumulation was overestimated with traditional organic carbon-water partition model.
- Additional correction for black carbon with two-carbon model improved the prediction.
- Chemical freely dissolved pore water concentrations gave accurate predictions.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 January 2016
Received in revised form 15 April 2016
Accepted 16 April 2016
Available online xxxx

ABSTRACT

There were two main objectives in this study. The first was to compare the accuracy of different prediction methods for the chemical concentrations of polyaromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) in the organism, based on the measured chemical concentrations existing in sediment dry matter or pore water. The predicted tissue concentrations were compared to the measured ones after 28-day laboratory test using oligochaeta worms (*Lumbriculus variegatus*). The second objective was to compare the

Abbreviations: AOC, amorphic organic carbon; BC, black carbon; BCF, bioconcentration factor; BSAF, biota to sediment accumulation factor; EqL, equilibrium partition model; EqPS, equilibrium passive sampling; MB, model bias; msPAF, multi-substance Potentially Affected Fraction of the species; OC, organic carbon; PAH, polyaromatic hydrocarbon; PCB, polychlorinated biphenyl; POP, persistent organic pollutant; TOC, total organic carbon; TPH, total petroleum hydrocarbons; TU, toxic unit; A1, Jorba sediment; A3, Martorell sediment; LB, Lippenbroek sediment; PAR, Pardubice sediment; PLC, Prelouc sediment; SE, Eenhoorn sediment; SRV, Rundvoortbrug sediment.

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http://dx.doi.org/10.1016/j.scitotenv.2016.04.110

Keywords:
PAH
PCB
Black carbon
Equilibrium passive sampling
Bioconcentration factor
Lumbriculus variegatus

bioaccumulation of PAHs and PCBs in the laboratory test with the in situ bioaccumulation of these compounds. Using the traditional organic carbon-water partitioning model, tissue concentrations were greatly overestimated, based on the concentrations in the sediment dry matter. Use of an additional correction factor for black carbon with a two-carbon model, significantly improved the bioaccumulation predictions, thus confirming that black carbon was important in binding the chemicals and reducing their accumulation. The predicted PAH tissue concentrations were, however, high compared to the observed values. The chemical concentrations were most accurately predicted from their freely dissolved pore water concentrations, determined using equilibrium passive sampling. The patterns of PCB and PAH accumulation in sediments for laboratory-exposed *L. variegatus* were similar to those in field-collected Lumbriculidae worms. Field-collected benthic invertebrates and *L. variegatus* accumulated less PAHs than PCBs with similar lipophilicity. The biota to sediment accumulation factors of PAHs tended to decrease with increasing sediment organic carbon normalized concentrations. The presented data yields bioconcentration factors (BCF) describing the chemical water-lipid partition, which were found to be higher than the octanol-water partition coefficients, but on a similar level with BCFs drawn from relevant literature. In conclusion, using the two-carbon model method, or the measured freely dissolved pore water concentrations method is recommended for predicting the bioaccumulation of PAHs and PCBs.

1. Introduction

When the toxicity of persistent organic contaminants for organisms in freshwater sediments needs to be assessed, a realistic approximation of the contaminant bioaccumulation in the organism is needed. Bioaccumulation depends on the characteristics of the contaminant, the organism and the environment (Newman, 2010). The most accurate method in assessing bioaccumulation is to measure contaminant concentrations in the tissue or target organ of each species of concern. Due to economic and practical reasons, often the contaminant concentrations in sediments or sediment pore water, rather than in biota, are measured. To translate these concentrations into risks for biota, predictive tools for estimating tissue concentrations are required.

According to the equilibrium partitioning theory, the chemical activity of a nonionic organic contaminant is equal in sediment organic carbon (OC), in exposed biota and in pore water (freely dissolved concentration), if equilibrium is attained between these phases (Di Toro et al., 1991). By using this theory, it is possible to predict the contaminant concentrations in one phase from those in another, if the phases can be well defined and the partition coefficients of the chemicals are known. The concentrations of different types of hydrophobic organic contaminants in benthic organism were shown to be predictable by measuring the freely dissolved concentrations in sediment pore water and using the bioconcentration factor, which describes the chemical accumulation from water to animal lipid (Kraaij et al., 2003). The freely dissolved concentrations of contaminants can be measured by different equilibrium passive sampling methods (EqPS) using e.g. silicone rubber (Mayer et al., 2000a, 2000b; Reichenberg et al., 2008; Smedes, 2007). Predicting the tissue concentration of contaminants from the concentration in sediment organic carbon (OC) is more complicated. The reason for this is that, usually only a fraction of a nonionic organic chemical in the sediment is bound by soft amorphous organic carbon (AOC) and accessible, while another fraction is strongly bound to a soot-like organic matter fraction, called black carbon (BC), or has diffused into the micro pores of sediment particles and is no longer readily bioavailable (Luthy et al., 1997). Consequently, the actually bioavailable concentration, corresponding with the freely dissolved concentration, will be lower than predicted from concentration in the sediment (total) organic carbon fraction. Therefore, more realistic estimates of bioavailable concentrations should be possible, if the heterogeneity of the sediment is taken into account, by using separate partition coefficients for black carbon and amorphous organic carbon. Koelmans et al. (2006) summarized current understanding of sorption mechanisms of persistent organic pollutants (POPs, such as PCBs and PAHs) for black carbon, and the role of black carbon in reducing the accumulation of pollutants in aquatic organisms. Their findings indicated that the sorption of chemicals in black carbon is a surface adsorption, including the surfaces of pores. The strength of sorption, is mainly governed by the specific surface area of black carbon, organic carbon, the contact area and the sorbate thickness. The two-carbon model, including partitioning for black carbon, has been used in predicting bioaccumulation of PAHs and PCBs (e. g. Hauck et al., 2007; Moermond et al., 2005; Sun and Ghosh, 2008; Werner et al., 2010). The use of the two-carbon model in estimating POP partitioning in sediments has improved the prediction of their bioaccumulation in organisms, despite reported challenges (Hauck et al., 2007; Werner et al., 2010). In recent years, alternatives to one-carbon partition models have been introduced. These assume that chemical sorption in total organic carbon is similar to coal-tar sorption. Such models have been used in predicting the partitioning of POPs in sediments and soil, and their accuracy has been compared with the both one and two-carbon models (Arp et al., 2009, 2014; Hawthorne et al., 2011).

The objectives of this study were to assess the accuracy of different equilibrium partition-based models to predict the bioavailability of PCBs and PAHs in sediment dwelling organisms. The models were compared with observed bioavailability in biotests in the laboratory and in collected field data. This was done by passive sampling, and applying one and two-carbon models with literature data on partitioning, to calculate tissue concentrations in benthic organisms. These were then compared to measured tissue concentrations from 28-day bioaccumulation tests with *Lumbriculus variegatus*. In addition, the PAH and PCB bioaccumulation in *L. variegatus* was presented as biota to sediment accumulation factors (BSAFs). The BSAFs were used for comparison of accumulation of these chemicals in *L. variegatus* and in field collected benthic animals, in the same sampling sites of river sediments. The strength of this study is that the feasibility of different bioaccumulation predicting models was studied with two major groups of persistent organic pollutants, using in-situ contaminated sediments, which represents over wide range of contaminants and geochemical conditions. The challenges in the modelling are discussed and recommendations are provided.

2. Materials and methods

2.1. Description of the sediments and chemical analyses

The sediments used in the present study were sampled in three different river basins, two sites from each: the Elbe in the Czech Republic, the Scheldt in Belgium and the Llobregat in Catalonia, Spain (Table 1). These six sediments were used for testing the bioaccumulation of PAHs and PCBs in *L. variegatus*, and for the prediction of bioaccumulation from chemical concentrations in sediment. Equilibrium passive sampling, to estimate the freely dissolved pore water concentrations, was applied only to the two Llobregat sediments. The river basins and sites were used as case studies, in the EU project MODELKEY (Brack et al., 2005), and a map and description of the sampling sites are