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# Transfer of PCBs from bottom sediment to freshwater river fish: A food-web modelling approach in the Rhône River (France) in support of sediment management

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ABSTRACT

Since 2005, restrictions have been because of fish consumption along the Rhone River because of high polychlorobiphenyl (PCB) concentrations, which have resulted inadverse economic consequences for professional fisheries in affected areas. French environmental authorities have expended considerable efforts to research sediment remediation strategies and development of sediment quality guidelines designed to protect the health of humans consuming Rhône River fish. Here we: (1) develop a bioaccumulation food-web model that describes PCB concentrations in three common freshwater fish species of the Rhône River, using Bayesian inference to estimate the input parameters; (2) test the predictive power of the model in terms of risk assessment for fish consumption; and (3) discuss the use of this approach to develop sediment quality guidelines that protect the health of humans consuming Rhône River fish. The bioaccumulation model predictions are protective for human consumer of fish and are efficient for use in risk assessment. For example, 85% of the predicted values were within a factor of 5 of measured CB153 concentrations in fish. Using sensitivity analyses, the major role played by sediment and diet behaviors on bioaccumulation process is illustrated: the parameters involved in the respiratory process (contamination from water) have little impact on model outputs, whereas the parameters related to diet and digestion processes are the most sensitive. The bioaccumulation model was applied to derive sediment concentrations compatible with safe fish consumption. The resulting PCB sediment thresholds (expressed as the sum of seven PCB indicator congeners) that are protective for the consumption of the fish species ranged from 0.7 to 3 ng/g (dw).



## 1. Introduction

Persistent organic pollutants, such as polychlorinated biphenyls (PCBs), are widespread in the environment and can adversely affect human health and wildlife. Food-web bioaccumulation in aquatic ecosystems has been studied because of its relevance for environmental toxicology and risk assessment in an attempt to implement regulatory measures. In this context, food-web bioaccumulation models can be considered tools for researchers and environmental managers (Arnot and Gobas, 2006; Kelly et al., 2007).

Levels of hydrophobic contaminants in fish have been shown to be highly variable at inter-individual (Kiriluk et al., 1995; Dufour et al., 2001), seasonal (De Laender et al., 2010b), and species (Kiriluk et al., 1995) levels, and between ecosystems (Kidd et al., 1998). This variability can be caused by environmental factors, such as the basal contamination level and pathways for the ecosystem considered (either atmospheric or point sources) (Stapleton et al., 2001; Stapleton et al., 2002), or from physiological factors, such as fish lipid concentrations (Borgå et al., 2004), age (Larsson et al., 1991), and metabolic rates (Burtnyk et al., 2009). Stable isotope methods applied to ecotoxicological studies have revealed that the variability in fish contamination levels can be related to the trophic position of the fish (Kiriluk et al., 1995; Kidd et al., 1998; Kidd et al., 2001) or to feeding habits (Kidd et al., 2001; Lopes et al., 2011). As a consequence, a food-web bioaccumulation model usually must include a large number of parameters, determined from the literature or from laboratory experiments, site-specific measurements, or empirical relationships.

Linkov et al. (2001) showed that the use of point estimates in a bioaccumulation food-web model may significantly overestimate risk. Many studies have used least squares methods to estimate parameters and obtain the "best" match between estimates and measurements (Lundstedt-Enkel et al., 2005). More complicated approaches, such as Bayesian inference or linear inverse modelling,

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have been used to parameterize bioaccumulation models (Watanabe et al., 2005; De Laender et al., 2009). These methods have the advantage of quantifying uncertainty and variability in parameter estimates. Using Monte-Carlo simulations, the uncertainties can be propagated to determine uncertainty in bioaccumulation model predictions.

Since 2005, a series of restrictions on fish consumption and trade have been implemented along the Rhône River and for several tributaries (the Saône, Doubs, Isère, Durance, etc.), as well as for other rivers in France. A national action plan aiming to reduce adverse impacts of PCBs was adopted in February 2008; although remediation actions for sediments are not mentioned explicitly in this plan, substantial research has nevertheless been dedicated to remediation strategies and modeling the transfer of PCB contamination from sediment to fish. Food-web models are viewed as possible tools for supporting the selection of areas for remediation, for helping to determine "safe" concentrations in sediments, and for predicting trends. For this study, three large and long-lived freshwater cyprinids were sampled (the bream *Abramis brama*, the barbel *Barbus barbus* and the chub *Squalius cephalus*) because they all accumulate PCBs over several years, but have different diets, and exploit different habitats. According to a recent epidemiological study relating freshwater fish consumption and PCB levels in human blood and serum (INVS and ANSES, 2011), breams and barbels also are occasionally consumed (two and less than one meals per year, respectively), while chub is not valued for consumption but may represent other appreciated species such as roach (*Rutilus rutilus*, around two meals per year on average). According to the same study, the European eel (*Anguilla anguilla*), highly valued for consumption, is consumed less than three times per year on average (CI 2.1–3.0).

The purposes of the present study were: (1) to develop a bioaccumulation food-web model that describes PCB concentrations in these three common freshwater fish species of the Rhône River, using Bayesian inference to estimate the input parameters; (2) to test the predictive power of the model in terms of risk assessment for fish consumption; and (3) to discuss the use of this approach to develop sediment quality guidelines that protect the health of humans consuming Rhône River fish.

## 2. Material and methods

### 2.1. Data

#### 2.1.1. Study sites

The study was conducted at three sites in France along the Rhône River (Fig. 1) (1) Lône de la Morte (MTE), the relative reference site upstream from Lyon (upstream from the first contaminated stretch); (2) Grand Large (GDL), a fluvial lake within the contaminated area and close to the city of Lyon; and (3) Ile du Beurre (BRE), a site downstream from Lyon.

These sites were chosen on the basis of the expected degree of sediment contamination. Dams that border each site limit fish migration.

#### 2.1.2. Sampling and analyses

Adult specimens were captured with nets by electrico fishing. The fish were killed in accordance with the ethical guidelines on experimentation on animals, as detailed in agreement A69389 10 01 provided by the "Service Protection and Santé animals" (Direction départementale de la protection des populations) to our laboratory in March 2010. For each fish, Length (in cm) and weight (in g) were measured, sex was determined, age was estimated by scalimetry (in years), and gut content was analysed. The number of individuals caught at each site and their morphological characteristics are summarised in Table 1.

Key invertebrates known to be eaten by these fish species (Vieille-Blanchard, 2007) were caught with sieves: Chironomidae (CHI), Gammaridae (GAM), Ephemeroptera (EPH) and bivalves: *Pisidium* (PIS) and *Corbicula* (COR).

Invertebrates were starved in the lab for 24 h to empty their digestive tract of residues that could distort the PCB and isotope analyses. Fish fillets and invertebrate individuals were weighed, frozen at −20 °C, freeze-dried, reweighed, and finely ground.

Lipid content (in %) (g lipid, dry weight/g tissue, dw) and PCB concentrations (in ng/g wet weight) were measured by the CARSO-LSEHL group (Lyon, France), following USEPA standard 1668. Fish were analyzed individually and invertebrates were analyzed by pooling all individuals. The seven PCB indicator congeners ($_i$PCB) were quantified (CB28, CB52, CB101, CB118, CB138, CB153 and CB180). Uncertainty of measured concentrations was about 20%.

One sediment core was collected at each site at the same locations as the invertebrates and fish and radionuclide measurements were used to age the successive sediment layers in each core. PCB analysis was done by the EUROFINS laboratory (Orléans, France).

For further details on the analyses, see (Lopes et al., 2011) for biota and (Desmet et al., submitted for publication) for sediment.

### 2.2. Model development

#### 2.2.1. Choice of the PCB congeners modeled

We modeled fish contamination for the $_i$PCB. These congeners represent a range of chlorination, and thus a range of capacities to be metabolised and bioaccumulated. The most chlorinated congeners (such as CB153) are not (or are only very slightly) metabolized, and thus are the most likely to bioaccumulate (Gagnon et al., 1990; Nfon and Cousins, 2007; Paterson et al., 2007; Morgan and Lohmann, 2010).

#### 2.2.2. Invertebrate model

We used a steady-state approach to estimate the $_i$PCB concentration in invertebrates from the PCB concentration in water and sediment:

$$C_{c,j}(t) = ku_j(t)Cw_c(t) + ks_j(t)Cs_c(t) \quad (1)$$

where $ku_j(t)$ is the uptake of water for an individual of the species $j$ at time $t$ (L/g); $Cw_c(t)$ is the concentration of congener $c$ in water at time $t$ (ng/L); $ks_j(t)$ is the uptake of sediment for an individual of the species $j$ at time $t$ (g/g); and $Cs_c(t)$ is the concentrastion of congener $c$ in sediment at time $t$ (ng/kg dw).

This steady state approach assumes that $_i$PCB concentrations have had sufficient time to equilibrate between water, sediments and invertebrates, and that temporal changes in the $_i$PCB concentrations in the invertebrates therefore reflect those in the sediments.

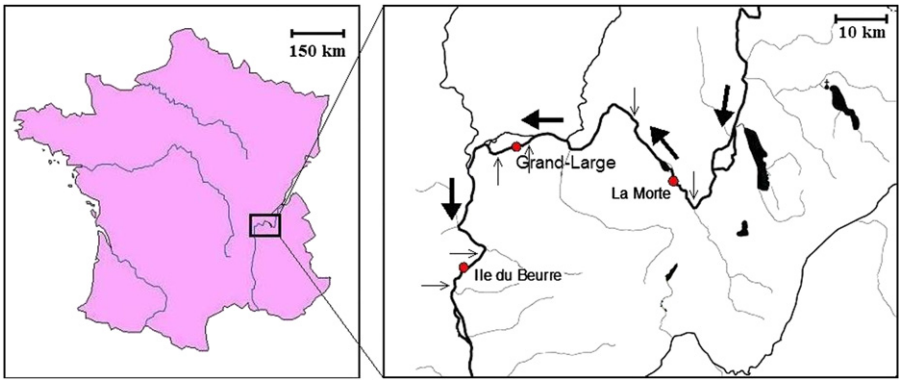


**Fig. 1.** Study sites along the Rhône River. The narrow arrows correspond to dams and the bold arrows correspond to the direction of flow.

**Table 1**
Number of fish and invertebrates collected at each site between August 2008 and January 2009 (N). For fish, the number of females (♀) and males (♂), mean size (± standard deviation), mean weight (± standard deviation), mean age (± standard deviation), and mean lipid content (± standard deviation) are given.

| | N | Size (cm) | Weight (g) | Age (year) | Lipid (%) |
|---|---|---|---|---|---|
| Bream *Abramis brama* | | | | | |
| La Morte | 7 (3♀+4♂) | 53.1 ± 8.18 | 1636 ± 775 | 6.43 ± 1.51 | 9.24 ± 4.76 |
| Grand-Large | 15 (9♀+6♂) | 54.1 ± 5.78 | 2025 ± 695 | 8.67 ± 3.96 | 22.8 ± 13.93 |
| Ile du Beurre | 17 (10♀+7♂) | 52.5 ± 4.69 | 2009 ± 607 | 5.94 ± 2.34 | 28.3 ± 11.22 |
| Chub *Squalius cephalus* | | | | | |
| La Morte | 20 (13♀+7♂) | 40.0 ± 8.76 | 861 ± 500 | 6.3 ± 1.78 | 3.2 ± 1.15 |
| Grand-Large | 15 (6♀+9♂) | 44.0 ± 5.80 | 1075 ± 457 | 5.27 ± 1.98 | 7.77 ± 3.89 |
| Ile du Beurre | 17 (12♀+5♂) | 42.6 ± 7.67 | 1061 ± 533 | 5.82 ± 2.40 | 7.62 ± 3.91 |
| Barbel *Barbus barbus* | | | | | |
| La Morte | 11 (11♀+0♂) | 52.1 ± 2.92 | 1215 ± 117 | 10 ± 1 | 5.85 ± 2.94 |
| Grand-Large | 15 (8♀+7♂) | 54.7 ± 6.36 | 1710 ± 767 | 9.4 ± 2.32 | 13.6 ± 6.14 |
| Ile du Beurre | 5 (3♀+2♂) | 56.6 ± 5.45 | 1853 ± 575 | 9.8 ± 3.56 | 17.6 ± 4.14 |
| *Invertebrates* | *Pisidium* | *Corbicula* | *Chironomids* | *Gammarids* | *Ephemeroptera* |
| La Morte | 140 | 15 | 385 | 260 | 94 |
| Grand-Large | 160 | 13 | 200 | 173 | 21 |
| Ile du Beurre | 130 | 60 | 400 | 130 | – |

#### 2.2.3. Fish model

The fish model is based on the model developed by Arnot and Gobas (2004), which considered different pathways of chemical uptake and elimination and was based on the assumption that the exchange of PCB congener $c$ between the organism and its ambient environment can be described by a single equation:

$$\frac{dC_c(t)}{dt} = U(t)\alpha_c Cw_c(t) - H_c(t)C_c(t) + \beta_c F(t)\sum_j Q_j C_{c,j}(t) - (E_c(t) + G(t) + M_c(t))C_c(t) \quad (2)$$

where $C_c(t)$ is the concentration of the congener $c$ in the organism (ng/g ww) on day $t$; $U(t)$ is the gill uptake rate at time $t$ (L/g/d); $\alpha_c$ is the assimilation efficiency of the dissolved congener $c$ (dimensionless); $Cw_c(t)$ is the concentration of congener $c$ in water at time $t$ (ng/L); $H_c(t)$ is the gill elimination rate of congener $c$ at time $t$ (1/d); $\beta_c$ is the assimilation efficiency of congener $c$ in ingested particles (dimensionless); $F(t)$ is the ingestion rate at time $t$ (1/d); $Q_j$ is the diet preference of the individual for the prey $j$ (dimensionless); $C_{c,j}(t)$ is the concentration of the congener $c$ at time $t$ in the prey $j$ (ng/g ww); $E_c(t)$ is the fecal egestion rate of congener $c$ at time $t$ (1/d); $G(t)$ is the growth dilution rate at time $t$ (g/d); and $M_c(t)$ is the rate for metabolic transformation of congener $c$ (1/d). As mentioned above, the most chlorinated congeners (like CB153) are not (or only very slightly) metabolized. To simplify the model, we assumed that no congeners are metabolized and set $M_c(t)$ at 0. This assumption might be excessive for the two least chlorinated congeners (CB28 and CB52).

Given that $_i$PCB sediment concentrations varied over time (and, accordingly, the concentration in prey as well) and by study site, the fish model described above should be adapted to the age of fish at the sampling date and sampling site.

In this data set, fish age ranged from 2 to 15 years. Thirteen 1-year age classes were created, leading to integration of 117 equations (13 age classes for each of the three species at each of the three sites).

### 2.3. Input functions

The functions and parameters in the fish bioaccumulation model are detailed in Table 2. Environmental conditions are taken into account through the function $T_k(t)$, which describes the water temperature at site $k$ at time $t$. Physico-chemical properties of the PCB congener considered are accounted for by *Kow*, the octanol-water partition coefficient. The physiological conditions of each fish are accounted for by the functions $W_{i,n}(t)$, the weight of an individual of the species $i$ and age $n$ at time $t$, and $Lf_{i,n}(t)$, the lipid fraction of an individual of the species $i$ and age $n$ at time $t$. These dependent variables are described below. The specific functions implied in the elimination rate $Ec(t)$ and the diet preference $Q$ (estimated from analysis of gut content) are described in Supporting Information (Appendix A, Table S1).

### 2.4. Dependent variables

#### 2.4.1. Water temperature

Water temperature for the Rhône River for the past 20 years at available for the Bugey site (between La Morte and Grand-Large Fig. S1, SI). Given that we did not have the specific water temperature at each site, and to gain an idea of the daily variability that can occurs from 1 year to another, we compiled the daily temperature data for 1 year from 1994 to 2008 and with Bayesian inference adjusted a Gaussian model describing the variation in daily temperature over 1 year (Fig. S2, SI). We thus obtained a credibility interval for daily water temperatures over the year. The daily variability in water temperature observed at the Bugey site thus was taken into account, with the assumption that this variability represents the range of water temperatures at the three sites studied. Greater detail on the procedure is given in Supporting Information (Appendix B).

#### 2.4.2. Growth kinetics

For each species, we modeled growth kinetics in three steps. First, we fitted a log-linear model relating the size and weight of individuals:

$$\log(W(t)) = a_1 * \log(L(t)) - b_1 \quad (3)$$

where $W$ is the weight (g), $L$ the size (cm) and $t$ the time (days).

The difference between sites was tested for each species.

Second, we fit a von Bertalanffy growth model:

$$L(t) = L_\infty - (L_\infty - L_0)e^{(-kt)} \quad (4)$$

where $L_\infty$ is the asymptotic size (cm), $L_0$ is the size at birth (cm) and $k$ is the growth rate ($day^{-1}$).

To take into account the effect of temperature on growth of these species in this region, we assumed that growth did not occur for the 6 months between November and April (Philippart, 1977; Baras and Philippart, 1999). The estimated growth rate $k$ therefore was limited to the effective growth period and assumed to be null for the remaining months.

Third, we integrated the von Bertalanffy growth model obtained in the second step to the log-linear relation obtained in the first step, so as to obtain weight kinetics.

The parameters of each model were estimated by Bayesian inference using the Monte-Carlo Markov-Chain (MCMC) and computed using the WinBugs software (Lunn et al., 2000). For each data set, the inference was made on $6*10^3$ iterations following $10^4$ adaptive iterations on three independent MCMCs (with three different initial parameter values), resulting in a total of $18*10^3$ parameter sets.

#### 2.4.3. Lipid fraction

For each species, we modeled the variation in lipid fraction over time by fitting a linear model to weight versus lipid data using Bayesian inference, as explained above.

### 2.5. Simulation method, uncertainty, and sensitivity analyses

The bioaccumulation model was implemented in R software (R Development Core Team, 2007). We assumed that all individuals were born uncontaminated at the end of May (i.e., no transfer of PCBs from the mother): $C(0)=0$. Time 0 in the model depends on the age of fish at the date of capture. For each age class, time 0 thus is different: from 1994 for the oldest fish to 2006 for the youngest.

Uncertainty of model predictions was tested by Monte Carlo simulations: we drew 1000 samples from within the 95% credibility i interval of dependent variables, i.e., water temperature, growth kinetics and lipid fraction. The mean contamination kinetics and its credibility interval can thus be estimated.

A sensitivity analysis was carried out to test the influence of each function and each parameter on model outputs. It was run by successively modifying each function or parameter by ±10% and by measuring the effect on two model outputs: (i) the variation in the number of false-negatives, i.e., the number of individuals for which the predictions allowed consumption, but measured

**Table 2**
Input functions and parameters (more details in SI). The indices correspond to: *c* for the congener, *i* for the species, *n* for the age class, and *k* for the site.

| Function | Definition | Units | Equation | Reference |
|---|---|---|---|---|
| Fish | | | | |
| $U$ | Uptake rate from water | L/g/d | $U_{i,n,k}(t)=(1400*(W_{i,n}(t)/1000)^{0.65}/Cox_k(t))$ | (Arnot and Gobas, 2004) |
| | $O_2$ content in water | mg $O_2$/L | $Cox_k(t)=(14.45-0.413*T_k(t)+0.00556*T_k(t)^2)$ | (Norstrom et al., 1976) |
| $\alpha_c$ | Efficiency of assimilation of dissolved particles | – | $\alpha_c=(1.85+155/Kow_c)^{-1}$ | (Drouillard et al., 2009) |
| $H$ | Gill elimination rate | /d | $H_{c,i,n}(t)=\frac{\alpha_c U_{i,n}(t)}{lf_{i,n}(t)Kow_c}$ | (Gobas, 1993) |
| $\beta_c$ | Efficiency of assimilation of ingested particles | – | $\beta_c=(3*10^{-7}*Kow_c+2)^{-1}$ | (Arnot and Gobas, 2004) |
| $F$ | Uptake rate from food | /d | $F_{i,n,k}(t)=(0.022*(W_{i,n}(t))^{0.85}*e^{(0.06*T_k(t))})$ | (Gobas et al., 1999) |
| $Q$ | Diet preferences | – | Values in Supporting Information | (Lopes et al., 2011) |
| $E$ | | /d | $\log(E_c)=-0.11*\log(Kow_c-2.86)$ | (Paterson et al., 2010) |
| $G$ | | g/d | $G_{i,n}(t)=e^{(g_{i,n}(t))}-1$ with $g_{i,n}(t)=\ln\left(\frac{W_{i,n}(t+1)}{W_{i,n}(t)}\right)$ | This study |
| Invertebrates | | | | |
| $ku$ | Uptake of water | L/g | $ku_k(t)=3.54+0.166*T_k(t)$ [a] | (Landrum et al., 1998) |
| $ks$ | Uptake of sediment | g/g | $ks_k(t)=0.142+0.037*T_k(t)$ [b] | (Landrum et al., 1998; Lopes et al., 2011) |

[a] Equation used for all the prey species.
[b] Equation used for gammarids only and adapted for other species, more details in the text.

concentrations exceeded the regulatory limit for fish consumption (Index $I_1$, Eq. (5)); and (ii) the percentage of variation in the sum of squared differences between predictions and observations (Index $I_2$, Eq. (6)):

$$I_1 = FN_{\text{mean}} - FN_{\text{modif}} \tag{5}$$

where $FN_{\text{mean}}$ is the "reference" number of individuals for which the model predictions are in error in terms of fish consumption (obtained with the mean value of each parameter) and $FN_{\text{modif}}$ is the number of individuals obtained with the modified parameter.

$$I_2 = 100*\left(\frac{\sum_{i=1}^{n}(\text{Cmodif}_i-\text{Cobs}_i)^2}{\sum_{i=1}^{n}(\text{Cmean}_i-\text{Cobs}_i)^2}-1\right) \tag{6}$$

where $\text{Cobs}_i$ is the observed concentration of the individual $i$, $\text{Cmean}_i$ is the mean predicted concentration for the individual $i$, $\text{Cmodif}_i$ is the predicted concentration for the individual $i$ with the modified parameter, and $n$ is the total number of sampled individuals. A negative value of $I_2$ means a reduction of $I_2$% of the sum of squared differences and thus a better prediction.

### 2.6. Application of the model to derive sediment guidelines

The bioaccumulation model can be used in a predictive way to extrapolate the maximum concentration in the sediment to which each individual can be exposed during its lifetime without ever exceeding the regulatory limit for fish consumption, namely 8 pg TEQ $g^{-1}$ at the time of the study (E.C., 2006). Note that this limit is not a safety limit, but was adopted as a risk reduction measure following an ALARA (as low as reasonably achievable) approach. A new regulation was promulgated at the end of 2011, setting the regulatory limit at 6.5 pg TEQ $g^{-1}$. As the TEFs of DL-PCBs were re-evaluated in the meantime, referring now to 2005 WHO TEFs (Van den Berg et al., 2006) the consequences on model predictions will not be significant. The maximum "safe" PCB concentration in sediment was estimated step by step using the general bioaccumulation model applied to the CB153 congener (Eq. (2), without age class differentiation) for each species at each site and considering that the concentration of CB153 in sediment remains constant over time. We used CB153 because it is the main congener found in both fish contamination profiles (Fig. S5, SI) and sediment profiles (Fig. S4, SI), and because it is one of the congeners best correlated to the $\Sigma_i$PCB in fish and sediment (Fig. S6 and Table S3, SI), implying that the regulatory threshold for fish consumption based on CB153 is relevant. The sediment concentration values of interest are: (i) the value for which the individual never exceeds the regulatory limit for fish consumption during its lifetime ($C_{\text{sedmax}}$), and (ii) the values for which the individual exceeds the regulatory threshold 5%, 10% and 20% of its lifetime ($C_{\text{sed5}}$, $C_{\text{sed10}}$, $C_{\text{sed20}}$, respectively).

## 3. Results

### 3.1. Dependent variables

The kinetics equations obtained for the dependent variables are summarised in Table 3. For the function $L(t)$, the equations presented here correspond to those used for the growth period (from May to October each year). During the winter, $dL(t)/dt$ was set at 0 (assuming that individuals did not grow during the cold period). The fit of the different models to the data are presented in SI (Fig. S3).

### 3.2. Forcing variables

The concentrations in water and sediment and the octanol-water partition coefficient (*Kow*) are forcing variables. Their values are reported for each congener in Table 4. The concentration in water is assumed to be constant, but the concentration in sediment is not (Fig. S4, SI). Concentrations of $_i$PCB in the sediment for dates falling between those of age-dated sediment intervals in the cores were estimated by linear interpolation (Fig. S4, SI).

### 3.3. Contamination results

In the fish contamination profiles for each species at each site, the highly chlorinated congeners (such as CB153) are at higher concentrations than the less chlorinated congeners, and are thus more likely to bioaccumulate (Fig. S5, SI). Additionally, the $_i$PCB concentrations for each congener are correlated to the sum of the seven $_i$PCBs for all species at all sites (Table S3). Using these relations and that between $\Sigma_i$PCB and the TEQ (Babut et al., 2009), we can determine a value for the concentration of each $_i$PCB congener in sediment that corresponds to the regulatory limit in fish for consumption (given that the threshold is set at 8 pg TEQ/g ww, corresponding to $153 \pm 3$ ng of $\Sigma_i$PCB/g ww) (Table S3).

### 3.4. Simulation results

The age-class bioaccumulation model was used to obtain the concentration kinetics for each species at each site, some of which are presented in the SI (Figs. S7 and S8). For invertebrates, we had only observations at the sampling date. The predictions agree with the observations. High seasonal variations were observed for concentrations in fish, as reported previously (Loizeau et al., 2001; Volta et al., 2009; De Laender et al., 2010a), possibly as a result of the large range of temperatures to which fish are exposed.

To test the relevance of the model predictions, the concentration measured in each fish sampled was compared to the predicted concentration (Fig. 2 for CB153, Figs. S9 to S14 for the other congeners), broken out into the contributions of the two routes of contamination (the trophic route by the contribution of each prey type and the respiratory route). The uncertainty

**Table 3**
Equations of dependent variables obtained from our data by Bayesian inference. The mean estimate of each parameter is followed by its standard deviation in parentheses.

| Symbol | Definition | Species | Equation |
|---|---|---|---|
| $T(t)$ | Water temperature (°C) | | $T(t) = 4.75(\pm 0.08) + 15.8(\pm 0.09) * e^{(-0.5 * ((t - 210.8(\pm 0.27))^2 / 72(\pm 8.7)^2))}$ |
| $W(t)$ | Weigh/size log-linear relationship | Bream | $\log(W(t)) = 2.59(\pm 0.30) * \log(L(t)) - 1.192(\pm 0.52)$ |
| | | Chub | $\log(W(t)) = 3.17(\pm 0.15) * \log(L(t)) - 2.20(\pm 0.26)$ |
| | | Barbel | $\log(W(t)) = 4.286(\pm 0.33) * \log(L(t)) - 4.27(\pm 0.59)$ |
| $L(t)$ | Von Bertalanffy growth model | Bream | $L(t) = 58.0(\pm 1.4) - (58.0(\pm 1.4) - 0.1) e^{(-2.8 \times 10^{-3}(\pm 3.1 \times 10^{-4})t)}$ |
| | | Chub | $L(t) = 51.3(\pm 2.4) - (51.3(\pm 2.4) - 0.13) e^{(-2.1 \times 10^{-3}(\pm 2.8 \times 10^{-4})t)}$ |
| | | Barbel | $L(t) = 59.8(\pm 2.5) - (59.8(\pm 2.5) - 0.15) e^{(-1.7 \times 10^{-3}(\pm 2.8 \times 10^{-4})t)}$ |
| $Lf(t)$ | Lipid fraction | Bream | $Lf(t) = 5.0 * 10^{-5} W(t) - 0.035$ |
| | | Chub | $Lf(t) = 8.3 * 10^{-6} W(t) + 0.006$ |
| | | Barbel | $Lf(t) = 1.3 * 10^{-5} W(t) + 0.006$ |

**Table 4**
Concentration of $_i$PCB congeners in water and sediment.

| Symbol<br>Definition | | $Cw_c$<br>Concentration of congener $c$ in water | $Cs_c$<br>Concentration of congener $c$ in sediment | log($Kow$)<br>Octanol-water partition coefficient |
|---|---|---|---|---|
| | Units | (ng/L) | (ng/kg dw) | – |
| Value for each $_i$PCB congener | CB28 | 0.06 | Fig. S4 (SI) | 5.67 |
| | CB52 | 0.05 | Fig. S4 (SI) | 5.84 |
| | CB101 | 0.04 | Fig. S4 (SI) | 6.38 |
| | CB118 | 0.025 | Fig. S4 (SI) | 6.74 |
| | CB138 | 0.015 | Fig. S4 (SI) | 6.83 |
| | CB153 | 0.01 | Fig. S4 (SI) | 6.92 |
| | CB180 | 0.005 | Fig. S4 (SI) | 7.36 |
| References | | (Fruget et al., 2010) | (Desmet et al., submitted for publication) | (Hawker and Connell, 1988) |

associated with the dependent variables is indicated by error bars around individual model predictions.

Overall, the model tends to overestimate concentrations for the least chlorinated congeners, but sometimes underestimates those for the most chlorinated congeners. For example, 85% of the predicted values were within a factor of 5 of measured CB153 concentrations in fish, while this percentage falls to 36% for CB52. Nevertheless, predictions are generally relatively good except for those for chub, for which concentration levels are overestimated, perhaps because this species is opportunistic, and a fixed diet over the lifetime of the fish was assumed here. Furthermore, whatever the PCB congener, the PCB contamination process in fish is essentially via the trophic route, and respiration has a limited effect (Thomann and Connolly, 1984; Thome and Leroy, 2007). No relation was observed between the model predictions and the sex of individuals, their age, or the date on which they were sampled.

From a risk assessment perspective, only one individual of 113 (a bream at the upstream site) had a measured CB153 concentration greater than the regulatory limit for fish consumption and a predicted CB153 concentration less than the regulatory limit.

### 3.5. Sensitivity analysis

Results of the sensitivity analysis are presented here for two chlorinated congeners: CB52 and CB153. No parameter had an effect on the first index ($I_1$, Eq. (5)), which represents the number of individuals for which the predictions allowed consumption while measured concentrations exceeded the regulatory limit.

The effects of parameter modifications on the second index ($I_2$, Eq. (6)) are presented in Fig. 3 for the two congeners for the entire data set (all species combined). For the same variation range of each parameter, the model is equally sensitive for CB52 and CB153. Regardless of the congener, however, some parameters had little or no effect on predictions: water filtration rate ($U(t)$), assimilation efficiency of the dissolved particles ($\alpha$), water concentration ($C_w$), diet preference for *Corbicula* ($Q_4$), concentration in *Corbicula* (*Ccorbi*), growth rate ($G(t)$), and gill elimination rate ($H(t)$). Other parameters had a large influence on predictions. Better predictions (negative value of $I_2$) were obtained with an increase in the excretion rate ($E(t)$) and the rate of metabolic transformation ($M$(t)) and a decrease in the feeding rate ($F$(t)), the efficiency of assimilation of ingested particles ($\beta$), diet preference towards chironomids ($Q_1$), concentration in chironomids (*Cchiro*), and the sediment concentration (*Csed*). Finally, some parameters had different effects depending on the congener tested: the increase of the octanol-water partition coefficient (log($Kow$)) improved the model prediction quality for CB153 more than for CB52, suggesting that the accuracy of the value of this parameter is more important for high-chlorinated congeners that are more bioaccumulated. In contrast, parameters concerning gammarids and Ephemeroptera (diet preference for them ($Q_2$, $Q_3$) and concentration in them (*Cgam*, *Ceph*) have higher effects for CB52 than CB153, while parameters concerning chironomids (diet preference for them ($Q_1$) and concentration in them (*Cchiro*) have higher effects for CB153 than CB52. The most important point to underline from this sensitivity analysis is the impact of metabolic transformation on model predictions. We can observe an higher effect for CB52 than CB153, confirming the fact that such physiological process is more important to consider for low-chlorinated congeners than high-chlorinated one. Nevertheless, this point is often neglected in bioaccumulation model while it has a no negligible impact on predictions.

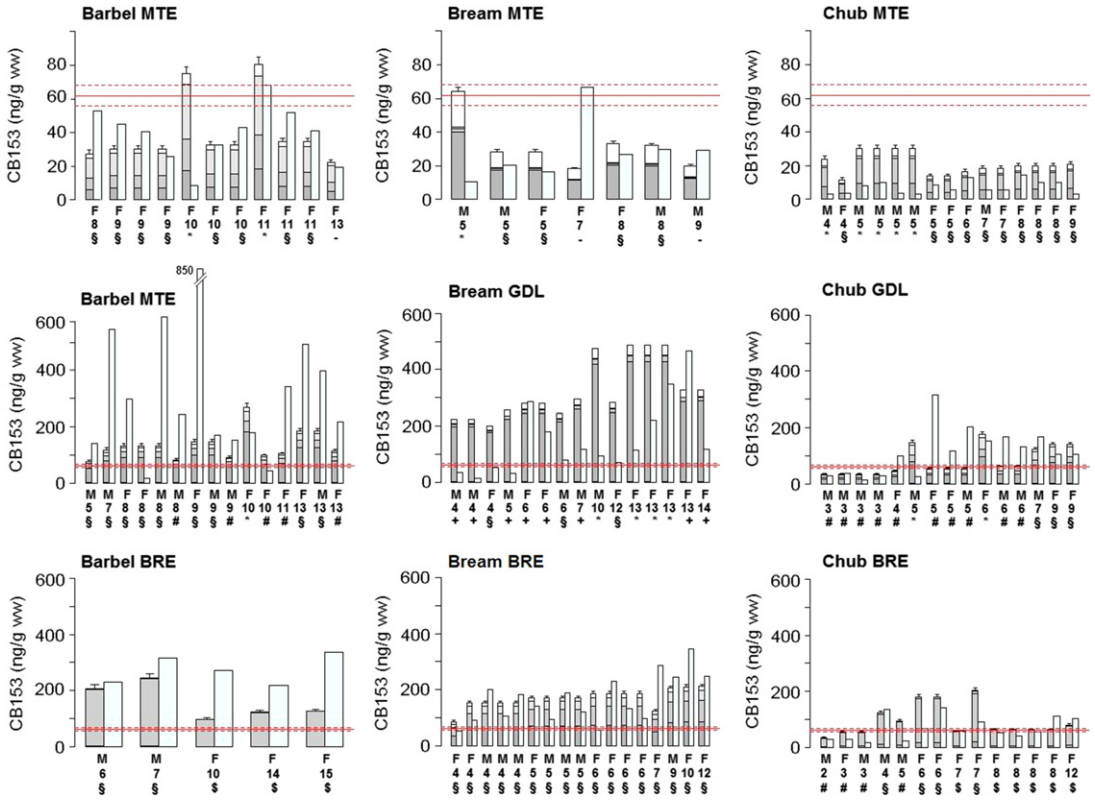


**Fig. 2.** Comparison of model concentration predictions for CB153 (by contamination source: ■ water; ■ chironomids; ■ gammarids; ■ Ephemeroptera; and □ *Corbicula*) and individual observations for each species at each site. The error bars around model predictions correspond to the 95% confidence interval obtained from Monte-Carlo simulations. The horizontal line represents the mean CB153 concentration corresponding to the regulatory limit for fish consumption, and the dotted lines the 95% confidence interval. Below each individual are indicated the sex (*M*, male; *F*, female), the age (in years), and the sampling date (* for August 2008; + for September 2008; § for October 2008; # for November 2008; – for December 2008 and $ for January 2009).

### 3.6. Sediment guidelines

The results are presented in Table 5. There are differences between species at the same site and differences among sites for the same species. The barbel tends to generate more conservative sediment maximum concentrations, and the chub less ones. These differences can be explained by the differences in trophic behaviours among species and between sites, as supported by the results of the sensitivity analysis, suggesting that parameters related to diet behaviour control PCB contamination. Because the sediment guidelines associated with barbel are the most conservative, the threshold value recommended for sediment associated with this species in the study area should be either $_{i}$PCB = 0.7 ng/g (dw), which is protective for all three species at all sites, or a threshold for each site varying from 0.7 to 1 ng/g (dw).

## 4. Discussion

### 4.1. Model structure and parameterisation

Model parameterisation involves the selection of the state variable to ensure that the model is representative of conditions in the Rhône River. Ecological data and the associated uncertainty in bioaccumulation modeling have been increasingly incorporated in recent years (Linkov et al., 2001; Watanabe et al., 2005; De Laender et al., 2009; De Laender et al., 2010b). Here, we used measured data as often as possible to estimate the uncertainty associated with ecological information and to transfer it to bioaccumulation model predictions. Water temperature is the main environmental factor driving many physiological processes in poikilotherm organisms (De Laender et al., 2010a; Hallanger et al., 2011). The daily measurements in the Rhône River between the two upstream sites and the procedure used to select the temperature each day provide a good overview of the environmental conditions to which fish were subjected. Furthermore, individual data were used to parameterise individual conditions such as growth, lipid content, and diet, and thus provide a realistic description of fish conditions in the Rhône River.

Some physiological processes were not included in the model. The reproductive process was not included as a way of PCB elimination. Some studies have shown that egg deposition transfers a substantial fraction of the PCB body burden from the adult female fish to the eggs (Bodiguel et al., 2009; Madenjian et al., 2010). However, this effect on female internal PCB concentrations seems limited in the sense that some authors, such as Mayon et al. (2006), did not show sex-dependent PCB accumulation in chubs, perhaps because, following spawning, females rapidly reached the contamination levels of males. The underlying processes are not well known: some authors suggested that, to restore energy reserves, event, females considerably increase their food consumption after a reproductive event and thus their PCB contamination level (Loizeau et al., 2001). Other authors showed that differences in PCB concentrations between sexes were not necessary because of an export of PCBs during egg laying (Madenjian et al., 2010), and that the lipid equivalent concentration within the organism remained the same (Russell et al., 1999). Another physiological process ignored by the model is the rate for metabolic transformation of PCBs. This process depends on the PCB congener and the species. Some studies have shown that the metabolic transformation in fish decreases for the

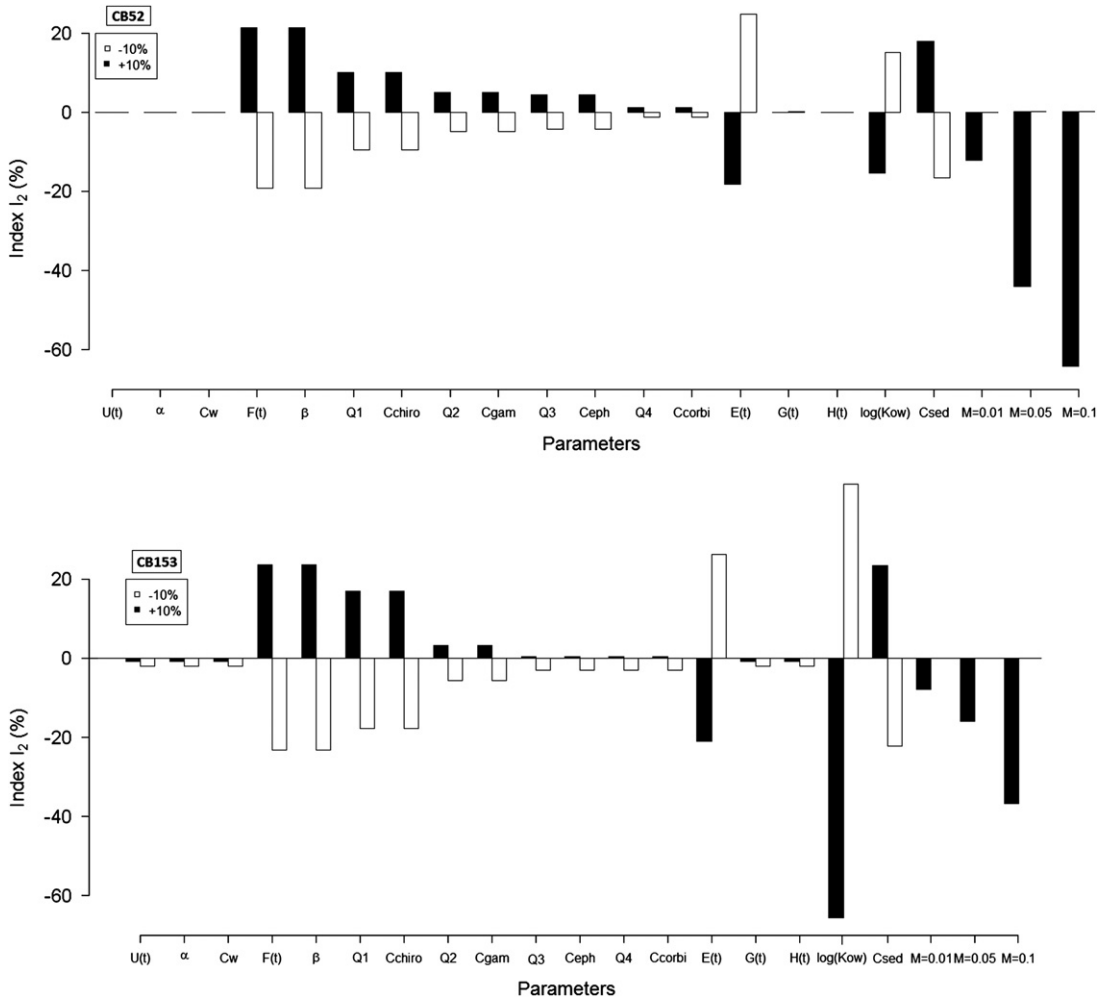


**Fig. 3.** Changes in the index $I_2$ (Eq. (6)) for CB153 (top) and CB52 (bottom) caused by a 10% reduction (white) or a 10% increase (black) of each parameter: water filtration rate ($U(t)$), assimilation efficiency of the dissolved particles ($\alpha$), water concentration ($C_w$), ingestion rate ($F(t)$), assimilation efficiency of ingested particles ($\beta$), diet preferences towards chironomids ($Q_1$), gammarids ($Q_2$), Ephemeroptera ($Q_3$) and *Corbicula* ($Q_4$), concentration in chironomids (*Cchiro*), gammarids (*Cgam*), Ephemeroptera (*Ceph*) and *Corbicula* (*Ccorbi*), fecal egestion rate ($E(t)$), growth rate ($G(t)$), gill elimination rate ($H(t)$), the octanol-water partition coefficient (log(*Kow*)), the sediment concentration (*Csed*) and the metabolisation rate ($M$), fixed to 0.01, 0.05 and 0.1).

most chlorinated congeners and that it can be ignored for CB153 (Gagnon et al., 1990; Nfon and Cousins, 2007; Paterson et al., 2007). The results of the sensitivity analysis confirm the importance of such physiological processes in PCB bioaccumulation and thus underline the necessity of estimating the rates, at least for low-chlorinated congeners for which such parameters have a higher impact.

We assumed that the diet of the three species, estimated from gut content analysis, remained constant during the entire lifetime of the fish. It is known, however, that diet differs according to the age of the individuals. As reported in previous studies for the bream *Abramis brama* (Kangur et al., 1999; Persson and Brönmark, 2002), young breams feed on zooplankton and visible macro-invertebrates, whereas, with increasing size, bream gradually become benthos feeders. For the chub and the barbel, some studies also have shown a change in diet with age (Markovic et al., 2007; Piria et al., 2005).

All these processes could influence PCB concentrations and thus have a long-term effect on model predictions. Future studies may wish to integrate them into the modelling framework for more realistic results and to study their potential impacts on bioaccumulation predictions. Nevertheless, without measured data, it would difficult to add such processes to the model without increasing uncertainty in model predictions. This is a typical illustration of the parsimony principle underlying modelling, which encourages a compromise between the complexity of the processes considered and the uncertainty around them and the realism of the predictions.

### 4.2. Model application for risk assessment

The bioaccumulation of hydrophobic organic chemicals in the aquatic food web contributes to adverse effects on both aquatic organisms and their predators. Monitoring the chemical residues in various organisms in the food web can provide necessary information for ecological risk assessment (ERA) (Wang et al., 2011). The risks that persistent organic compounds may pose to humans and the environment have recently increased the interest of researchers and environmental managers in the use of food-web bioaccumulation models as reliable tools in risk assessment (Arnot and Gobas, 2006; Gobas and Arnot, 2010).

The bioaccumulation kinetics of PCBs in fish obtained with the present model show high seasonal variations, with higher concentrations in summer than in winter, as observed in previous studies (Loizeau et al., 2001; Volta et al., 2009; De Laender et al., 2010a). This is explained by the higher water temperature in summer, which significantly increases physiological (respiration,

**Table 5**
Maximum concentration in the sediment (ng/g dw) to which each individual can be exposed without concentration in the fish exceeding the health-based benchmark for fish consumption ($C_{sedmax}$), and for which the concentration would exceed the benchmark 5%, 10% and 20% of its lifetime ($C_{sed5}$, $C_{sed10}$, $C_{sed20}$, respectively).

| | Barbel | | Bream | | Chub | |
|---|---|---|---|---|---|---|
| | CB153 | $\Sigma_i$PCB | CB153 | $\Sigma_i$PCB | CB153 | $\Sigma_i$PCB |
| $C_{sedmax}$ | | | | | | |
| La Morte | 0.4 | 1 | 0.4 | 1 | 0.8 | 2 |
| Grand Large | 0.3 | 0.7 | 0.2 | 0.5 | 0.4 | 1 |
| Ile du Beurre | 0.3 | 0.7 | 0.6 | 1.5 | 0.5 | 1.2 |
| $C_{sed5}$ | | | | | | |
| La Morte | 0.5 | 1.2 | 0.5 | 1.2 | 0.9 | 2.3 |
| Grand Large | 0.4 | 1 | 0.3 | 0.7 | 0.4 | 1 |
| Ile du Beurre | 0.4 | 1 | 0.7 | 1.7 | 0.6 | 1.5 |
| $C_{sed10}$ | | | | | | |
| La Morte | 0.6 | 1.5 | 0.6 | 1.5 | 1 | 2.5 |
| Grand Large | 0.5 | 1.2 | 0.3 | 0.7 | 0.6 | 1.5 |
| Ile du Beurre | 0.5 | 1.2 | 0.8 | 2 | 0.8 | 2 |
| $C_{sed20}$ | | | | | | |
| La Morte | 0.9 | 2.3 | 0.7 | 1.7 | 1.3 | 3.3 |
| Grand Large | 0.7 | 1.7 | 0.4 | 1 | 0.8 | 2 |
| Ile du Beurre | 0.8 | 2 | 1.2 | 3 | 1.2 | 3 |

excretion) and feeding processes. Stapleton et al. (2002) also highlighted the effect of the changes measured in fish lipid content associated with gamete production and spawning. Their results suggested that accumulation of PCBs by biota on seasonal scales is appreciably controlled by growth and lipid dynamics as well as foraging behavior. Nevertheless, supplementary data are needed to improve such effective seasonal variations in the field.

The credibility interval observed around the model predictions (Fig. 2) reflects the uncertainty propagated from the estimation of dependent variables (water temperature, growth, and lipidic fraction kinetic) on the model predictions, whereas the sensitivity analysis assesses the impact of variability and/or error in the model input functions and forcing variables on the model predictions. The sensitivity analysis is useful in the analysis of the internal mechanisms of the model and can be used to characterise potential errors in the model and to develop a better understanding of the relationship between the processes that control the behavior of PCBs in the food web of freshwater fish species. Here, we use the sensitivity analysis to illustrate the major role of sediment and diet behaviour on bioaccumulation process (Fig. 3). As reported by Gobas and Arnot (2010), the parameters involved in the respiratory process (contamination from water) have little impact on model outputs, while parameters related to diet or digestion processes are the most important.

The bioaccumulation model proposed here was not developed for descriptive purposes, but in a risk assessment perspective. As a consequence, we did not try to refine the predictions in order to have a perfect fit between observations and predictions, but attempted rather to study how observations and predictions match from a risk assessment point of view. We thus wanted to limit the cases where the predicted concentration was below the regulatory limit for fish consumption when the observed concentration was above (false-negative cases). On the contrary, a false-positive corresponds to a case where the prediction is above the regulatory limit while the observation is below. The latter scenario, which occurs mainly for the chub, is less disadvantageous than the first one in the sense that it does not underestimate the risk for the consumer. In this way, the bioaccumulation model proposed here is sufficiently protective for the consumer and efficient for risk assessment use because, for CB153, only one individual out of 113 was a false-negative case, i.e., a bream at MTE that exceeded the regulatory limit. Nevertheless, more data are needed to validate the model's predictive capability in term of risk assessment.

### 4.3. Potential use of this model in sediment risk management

Because bottom sediments are the main source of fish contamination by PCBs (Lopes et al., 2011), the fish advisories for the Rhône River and its tributaries have an impact on sediment management as a whole. Concerns are raised when it comes to dredging a lock or a fluvial harbour, or to partially removing dykes or groynes so as to restore, to some extent, the ability of the river bed to slow floods and its original ecological functioning. Moreover, clean-up decisions may be made at some specific sites. Catchment authorities therefore require sediment quality guidelines (SQGs) or assessment tools for these purposes. A first approach using biota-sediment accumulation factors (BSAF) was attempted and made it possible to derive a SQG intended to screen sediment quality throughout the catchment (Babut et al., 2012). Nevertheless, this SQG had poor predictive ability, because only 62% of the fish specimens recorded in the catchment database on fish contamination for which sediment quality data were available were correctly classified. Clearly such an SQG would not be appropriate in site-specific remediation decisions. Either a site-specific study using our model or an application of one of the values calculated above (range, 0.7–3.0 ng/g (dw) for ¡PCBs, depending on the site, the species and the level of protection granted) would be more efficient and protective. These values are comparable (although little lower) to the results obtained by Gobas and Arnot (2010) for white croaker consumption in San Francisco Bay, i.e., PCB concentrations in sediment equal to or less than 3.8 ng/g (dw). The different conditions retained for SQG calculations, from "no exceedance at all during fish lifetime" to "exceedance allowed up to 25% of the time" logically yields increasing SQGs. The selection between these is out of the scope of this study; we nevertheless point out that exceedance of the fish regulatory consumption limit, if allowed, would occur in spring and summer, which is also the period where amateur fishermen will likely consume their catch (see Fig. S8 in SI). Considering that the barbel and the bream are currently the species that accumulate the most PCBs in the Rhône River (except eels, but the regulatory threshold applicable to this species is higher), it is unlikely that more restrictive thresholds would be determined if the model was adapted to other species. Moreover, the range of values obtained from the model is limited. Consequently, proposing different SQGs is not justifiable, and a single value in the above-mentioned range would be sufficient. One advantage of using the model instead of simply the SQG is that it could help to test different remediation scenarios and predict the proportion of fish exceeding the current regulatory threshold in each scenario.

## 5. Conclusion

The bioaccumulation model was developed to describe the accumulation of PCBs in the food web of three freshwater fish species in the Rhône River and to determine sediment quality guidelines that would protect the health of humans consuming Rhône River fish. We illustrated here that this food-web model is a feasible method for predicting the extent of bioaccumulation of hydrophobic compounds in organisms as a step to evaluate their risk on aquatic ecosystems. Nevertheless, in such an approach, it is important to consider the uncertainties associated with ecological information on the ecosystem considered in order to screen the risk management of such high-risk pollutants.

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## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.ecoenv.2012.04.007.

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