A steady-state model of PCB accumulation in dab food web

Model
PCB
Food web
Benthic invertebrates
Dab

Modèle
PCB
Chaîne trophique
Invertébrés benthiques
Limande

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ABSTRACT

A five-compartment steady-state food web model is proposed for the benthic food web leading to the dab. Three exposure pathways are considered in the description of accumulation by benthic animals: ingestion of particulate contaminants associated with either sediment or phytoplankton, and respiratory uptake of free dissolved contaminant in overlaying water. Application of the model to a simple food web in the Bay of Seine (Eastern Channel) indicates that : a) feeding is the principal route of contamination, especially for PCB which have more than four chlorine atoms in the molecule ; b) excretion and growth rates, phytoplankton lipid fraction and organic carbon content of sediment are the parameters which mostly determine the chemical bioaccumulation in the food web.

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RÉSUMÉ

Modèle, à l'état stable, de la bioaccumulation des PCB dans le réseau trophique de la limande

Un modèle à cinq compartiments a été réalisé pour simuler la bioaccumulation des PCB à travers le réseau trophique benthique aboutissant à la limande. Trois voies de contamination des organismes sont considérées : l'ingestion de contaminants à partir du sédiment ou du phytoplancton ainsi que l'absorption de PCB dissous lors de la respiration. L'application de ce modèle en baie de Seine (Manche orientale) indique que : a) l'alimentation est la principale source de contamination, tout particulièrement pour les congénères ayant un nombre d'atomes de chlore supérieur à quatre ; b) les taux d'excrétion et de croissance, le pourcentage lipidique dans le phytoplancton ainsi que la fraction de carbone organique dans le sédiment sont les paramètres auxquels la bioaccumulation des PCB dans la chaîne trophique se montre la plus sensible.

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INTRODUCTION

The bioaccumulation of persistent toxic compounds constitutes a major hazard to marine life, and hence to human beings through seafood and fish consumption. Many authors have investigated the processes leading to the enrichment of xenobiotic compounds within foodwebs, particularly in order to build predictive models of bioaccumulation (Thomann and Connolly, 1984; O'Connor and Pizza, 1987; Connolly, 1991).

Fishes take up contaminants both from water and from food. Flatfishes, which consume mainly benthic species, are thus heavily exposed to hydrophobic contaminants, especially in coastal and estuarine waters (Köhler *et al.*, 1986; Cossa *et al.*, 1992). In our study, the dab (*Limanda limanda*) has been chosen as the top predator of a very

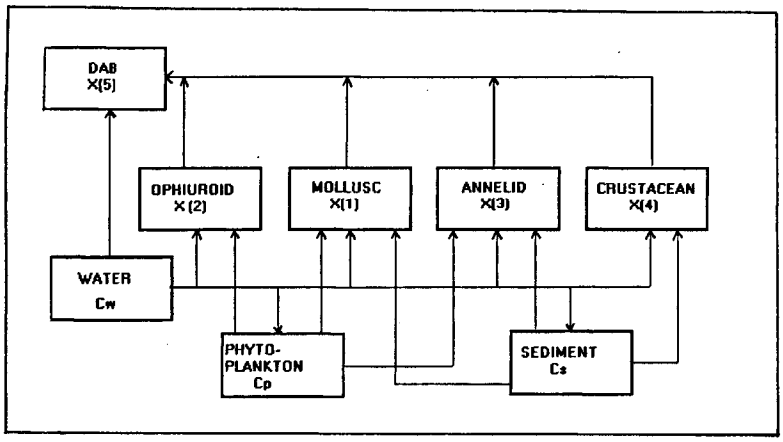

Figure 1

Schematic diagram of five-compartment food-web model.

Diagramme des cinq compartiments de la chaîne trophique.

simple food web. This sedentary species, which is commonly found in European coastal waters, has been studied for PCB accumulation (Knicmeyer and Steinhart, 1989), and its biology is well documented (Deniel, 1981; Quiniou, 1986; Tassel, 1988).

As monitoring programmes have stressed the high levels of PCB contamination in mussels from the Seine estuary and its surrounding area (Abarnou and Simon, 1986; Claisse, 1989), we located our study in the Bay of Seine.

The purpose of this paper is to present a generic modelling framework for the accumulation of PCBs in aquatic systems which takes account of interaction with benthic chemical and biotic compartments.

MODEL STRUCTURE AND EQUATIONS

The compartmental structure of the food web examined in this model is shown in Figure 1. Five biological compartments are considered together with dissolved contaminant concentration in the water column and particulate sediment. In order to provide a generic tractable analysis across the food web, the system is assumed to be at steady state. While it is recognized that natural systems are fluctuating, the suitable averaging of compartment parameters (*e.g.*, growth rates) and calibration data can provide estimates of a steady-state condition. Benthic invertebrates are contaminated *via* uptake from water and *via* direct ingestion of particles (sediment and/or phytoplankton). Benthic fishes accumulate chemicals directly from water and from benthic food. Interaction of the benthic community both with the sediment and the overlying water column implies that the chemical transfer from both these abiotic compartments must be taken into account.

Model equations

The basic model equations are an extension of the equilibrium model of the pelagic food chain given in Thomann (1989).

For all the model state variables, let X(i) be the chemical concentration in the i^{th} compartment on a wet weight basis [$ng.g^{-1}$(w.w.)], Cw be the free dissolved chemical concentration ($ng.l^{-1}$) in the water column, and Cs be concentration in sediment, on a dry weight basis, [$ng.g^{-1}$(d.w.)].

For the biotic compartment i, the rate of chemical uptake from the available dissolved pool is Ri ($l.g^{-1}.d^{-1}$) ; the excretion rate is Ei and the growth rate is Gi, both in (day^{-1}) units. The specific feeding rate of organism i on j (or on sediment, or phytoplankton) is Qij [g[prey(w.w.)].g^{-1} [predator(w.w.)].d^{-1}]. The chemical assimilation efficiency of ingested chemical is Ai,j (%). The feeding preferences of benthic compartment i for sediment and phytoplankton are denominated ais and aip respectively (ais + aip = 1). Finally, the general equation for the i^{th} benthic compartment is:

$$\begin{aligned} dX(i)/dt = 0 = {} & Ri * Cw * Aiw && \text{uptake from water} \\ & + Qip * aip * Aip * Cp && \text{uptake from phytoplankton} \\ & + Qis * ais * Ais * Cs && \text{uptake from sediment} \\ & - (Ei + Gi) * X(i) && \text{excretion and growth} \end{aligned} \quad (1)$$

For the phytoplankton/detritus compartment and for the sediment compartment, a simple partioning formulation is used:

$$Cp = Cw * Kp \quad (2)$$

$$\text{with } Kp = [(Kow^{0.87} * Flip) + 10.35]/1000 \quad (3)$$

Flip is the lipid fraction of phytoplankton, Kp is partition coefficient (Brown *et al.*, 1982) and Kow is the octanol/water partition coefficient.

$$Cs = Cw * Ks \quad (4)$$

$$\text{with } Ks = (Kow^{0.6} * Foc) + 4.81 \quad (5)$$

Foc is the organic carbon fraction of sediment and Ks is the partition coefficient (Van Zoest and Van Eck, 1986: "The behaviour of PCBs in the Scheldt Estuary", personal communication).

The equation for the dab compartment (5) is given by:

$$\begin{aligned} dX(5)/dt = 0 = {} & R5 * Cw * A5w \\ & + Q_{5,1} * X(1) * a5,1 * A5,1 + Q_{5,2} * X(2) \\ & * a5,2 * A5,2 \\ & + Q_{5,3} * X(3) * a5,3 * A5,3 + Q_{5,4} * X(4) \\ & * a5,4 * A5,4 \\ & - (E5 + G5)X(5) \end{aligned} \quad (6)$$

In this equation, $a_{i,j}$ represents the relative feeding preference of the dab for the jth benthic compartment ($\Sigma a_{i,j} = 1$).

Parameter estimation

The model framework contains two broad classes of parameters : those associated with the specific chemical (*i.e.*, uptake rate and chemical assimilation efficiency); and those associated with organism ecology (*e.g.*, growth, respiration and nutrition rates, feeding preferences). The strategy for parameter determination is twofold: estimates of some parameters are obtained from other independent studies in the literature, and some from model calibration.

PCB uptake rate from water

The rate constant Ri parameterizes the uptake of chemical from water to blood across the gill. For oxygen, the uptake rate is given by equation 7:

$$Ri_{O_2}/C_{O_2} = r'i \quad (7)$$

where r'i is the respiration rate [$mlO_2.g^{-1}(w.w.).d^{-1}$] and C_{O_2} the oxygen concentration in water. This uptake rate may also be described in terms of a mass-transfer rate constant through the gill (Rli_{O_2}), the gill surface (A), and the weight of the animal (W) :

$$Ri_{O_2} = Rli_{O_2} * A/W \quad (8)$$

In a similar manner, the mass-transfer rate for a chemical is:

$$Ri = Rli * A/W \quad (9)$$

So, the uptake rate of the chemical can be written as:

$$Ri = Rli * r'i/Ri_{O_2} * C_{O_2} \quad (10)$$

From this equation it follows that the uptake rate of a chemical can be estimated from the respiration rate of the animal, provided that the ratio of the mass-transfer coefficients for the chemical and oxygen is known.

In this model the chemical/oxygen uptake efficiency ratio was supposed to be equal to 1. Measurements of oxygen and organic chemical uptake efficiencies indicate that they are approximately equal for chemicals with logKow values between about 3 and 7 [Mc Kim *et al.*, 1985 (*see* Tab. 1)].

Chemical assimilation efficiency from water

The chemical assimilation efficiency is a simple function of Kow (Thomann *et al.*, 1992):

$$\log Aw = 2.9\text{-}0.5 * \log Kow \quad (6.5 < \log Kow < 10) \quad (11)$$

$$Aw = 0.8 \quad (4.5 < \log Kow < 6.5) \quad (12)$$

Food consumption rate

The rate of consumption of food $Q_{i,j}$ is calculated from the rate of energy usage, and is generally temperature-dependent. These parameters are obtained from other independent studies in literature (*see* Tab. 1).

Table 1

Bioenergetic related parameters used in food chain model calibration (coefficient of variation).

Paramètres bioénergétiques utilisés pour la calibration du modèle (coefficient de variation).

N°	Weight Wet(g)	Growth d^{-1}	Feeding Preferency	Respiration rate	Food consumption rate ($mg.ind^{-1}.d^{-1}$)	Excretion rate ($mg.ind^{-1}.d^{-1}$)
1	0.2836	0.0013	0.75 phyto 0.25sed.	$r = 0.0508Ww*e^{0.0269*T}$ $gO_2.d^{-1}$	Qep = 0.75Ww* [- 11.4(phyto) + 0.0704)] Qes = 0.25ww* [5.5(Foc) + 0.0756)]	0.247 (15 %)
		(10 %)	(10 %)	Salzwedel (1980)	Hughes (1973)	Salzwedel (1980)
2	0.212	0.009	1 phyto.	$r = 0.128Ww*e0.^{0412*T}$ $mlO_2.h^{-1}$	$Qe = 6.379(MES)*e^{0.0062T}$	0147 (15 %)
		(10 %)		Buchanan (1964)	Buchanan (1964)	Olsher and Fedra (1964)
3	0.209	0.0017	0.25 phyto. 0.75 sed.	$r = 1.704Ww*e^{0.0337*T}$ $ulO_2.d^{-1}$	log(Qep) = 0.25log(phyto) + 0.324log(Wd) + 0.702 log(Qes) = 0.75log(MES) + 0.324log(Wd) + 0.702	0.16 15 %
		(10 %)	(10 %)	Nichols (1975)	Arntz and Brunswig (1976)	Arntz and Brunswig (1976)
4	0.0623	0.0018	1 sed	$r = 0.0082Ww*e^{0.0015*T}$ $gO_2.d^{-1}$	$Qe = 0{,}294Wd*e^{0.014*T}$	$Qx = 0.827Ww*e^{0.00067*T}$
		(10 %)		Hawkins (1983)	Hawkins (1983)	Hawkins (1983)
5	150.00	0.0009	0.60 mol. 0.17 oph. 0.13 ann. 0.09 crus.	log(r) = 7.63 - 0.4015log(T) - 4.032log(Wd) $gO_2.d^{-1}$	$Qe = 0.294Wd*e^{0.0014*T}$	Qx = 0.388Qe + 0.055T + 1.765
		(10 %)	(15 %)	Paul *et al.* (1990)	Pandian (1970)	

Table 2

A: Environmental parameters used in food chain model calibration (coefficient of variation).; B: Chemical parameters used in food chain model calibration (coefficient of variation).

A : Paramètres environnementaux utilisés pour la calibration du modèle (coefficient de variation). ; B : Paramètres chimiques utilisés pour la calibration du modèle (coefficient de variation).

A:	**Temperature °C**	**Oxygen ($ml.l^{-1}$)**	**MES density ($g.l^{-1}$)**	**Phytoplankton density ($g.l^{-1}$)**	**Phytoplankton lipid fraction**	**Organic carbon in sediment**
Mean	9.07	6.24	0.103	0.207.10-3	0.0249	0.029
S.D.	(10 %)	(12 %)	(15 %)	(20 %)	(10 %)	(20 %)

B:	**CB 28**	**CB 52**	**CB 101**	**CB 118**	**CB 153**	**CB 138**	**CB 180**
Ass. Eff. (mol/pk)	0.27 (10 %)	0.137 (10 %)	0.61 (10 %)	0.73 (10 %)	0.45 (10 %)	0.62 (10 %)	0.25 (10 %)
Ass. Eff. (mol/sed)	0.012 (10 %)	0.141 (10 %)	0.30 (10 %)	0.88 (10 %)	0.60 (10 %)	0.40 (10 %)	0.70 (10 %)
Ass. Eff. (oph/pk)	0.062 (10 %)	0.135 (10 %)	0.59 (10 %)	0.75 (10 %)	0.48 (10 %)	0.59 (10 %)	0.28 (10 %)
Ass. Eff. (ann/pk)	0.031 (10 %)	0.154 (10 %)	0.54 (10 %)	0.84 (10 %)	0.46 (10 %)	0.601 (10 %)	0.31 (10 %)
Ass. Eff. (ann/sed)	0.50 (10 %)	0.0017 (10 %)	0.203 (10 %)	0.29 (10 %)	0.71 (10 %)	0.65 (10 %)	0.32 (10 %)
Ass. Eff. (cru/sed)	0.80 (10 %)	0.145 (10 %)	0.52 (10 %)	0.92 (10 %)	0.94 (10 %)	0.88 (10 %)	0.90 (10 %)
Ass. Eff. (dab/bth)	0.75 (10 %)	0.75 (10 %)	0.75 (10 %)	0.75 (10 %)	0.75 (10 %)	0.75 (10 %)	0.75 (10 %)-

Assimilation efficiency for ingested PCBs

Several experimental studies have examinated the fraction of ingested PCB that is adsorbed (Tanabe *et al.*, 1982; Goerke and Weber, 1971; Niimi and Oliver, 1983; Opperhuizen and Scharp, 1988). The PCB assimilation efficiency values reported in the above studies range from about 20 to 90 %. Differences between studies may be related to the use of different sizes and species of animals and to differences in the type of food used. Because of the obviously high degree of uncertainty with regard to this parameter, appropriate values for each animal and each PCB congener were not established *a priori*. Rather, the experimental data were used to define an appropriate parameter range as a guide in model calibration (*see* Tab. 2)

Excretion rate

These parameters are obtained from other independent studies in the literature (*see* Tab. 1).

Growth rate

The growth rates were established from observations about length-weight, age-length, and weight-age relationships, given in the literature (*see* Tab. 2).

The daily growth can be written as :

$$Gi = (e^{gi} - 1) \quad (13)$$

$$\text{with } gi = (\ln(W_{t2}/W_{t1}))/(t2-t1) \quad (14)$$

RESULTS AND MODEL CALIBRATION

The model was calibrated according to measurements obtained during the 1990-1992 sampling cruises in the Bay of Seine (PCB analysis procedure was described in Loizeau and Abarnou (1993)). For all the biotic compartments, the computed steady-state concentration was compared to the arithmetic average of the observed concentration.

The food chain model was separately calibrated for 7 PCB congeners: trichlorobiphenyl (28), tetrachlorobiphenyl (52), pentachlorobiphenyl (101), pentachlorobiphenyl (118), hexachlorobiphenyl (153), hexachlorobiphenyl (138), heptachlorobiphenyl (180).

A constant concentration of water-dissolved PCB was assumed (Tab. 2), and hence, species sampled were considered as being exposed to this constant concentration throughout their life.

The comparison for seven congeners between observed and calculated concentrations is presented in Figure 2.

Generally speaking, calibration provides a good agreement between the observed data and the calculated concentration. However, it seems that the model overates the concentration of weakly chlorinated congeners, particularly for dabs and crustaceans. These animals are probably capable of metabolism, especially for congeners Nos 28, 101, 52 (Boon *et al.*, 1989), a process which has

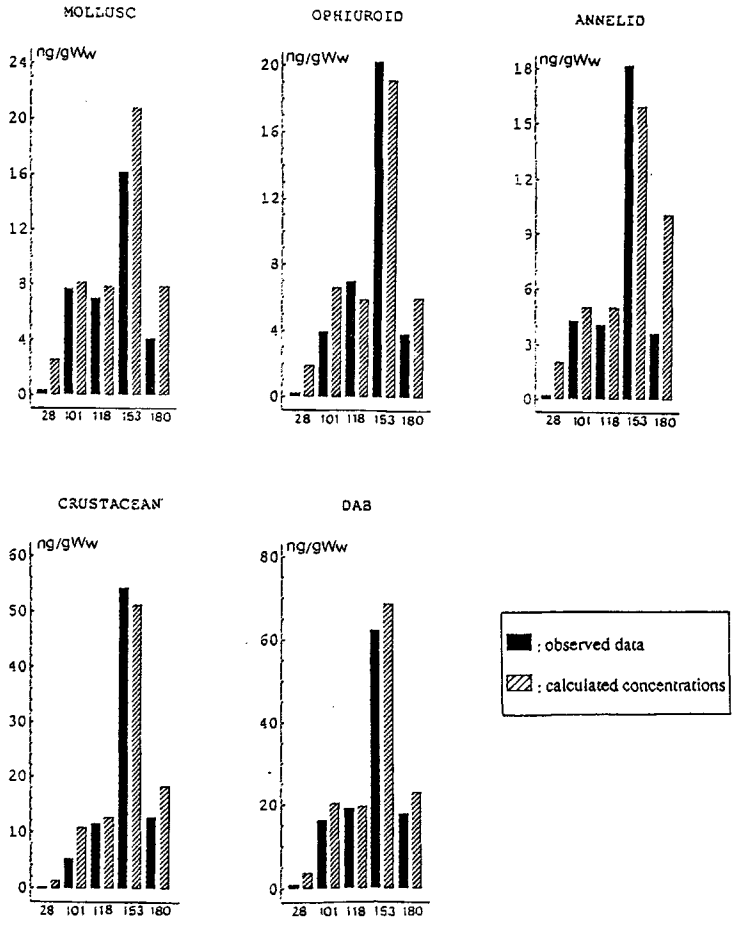

Figure 2

Comparison of observed with calculated concentrations for five compartments.

Comparaison des valeurs observées avec les valeurs calculées pour les cinq compartiments du modèle.

not been included in our model. Moreover, the model fails in simulating heptachlorobiphenyl. Presumably, the chemical assimilation efficiency drops more rapidly at high Kow than assumed in the model.

The importance of fluxes originating in food and water in relation to computed PCB accumulation by molluscs, annelids and dabs is illustrated in Figure 3. Accumulation appears to be mainly due to food consumption rather than to direct uptake from water, particularly when the number of chlorine atoms in the molecule increases. The more hydrophobic and lipophilic a chemical, the more concentrated it will be in the phytoplankton and sediment that constitute the basis of the food chain. However, accumulation from water remains important for CB 28, which is more water-soluble than the others.

Because food constitutes the major vector of contamination, the assimilation efficiency of the ingested chemical is a controlling parameter in the model. Laboratory measurements of assimilation efficiency of PCB congeners (Connolly, 1991) suggest a decline of assimilation efficiency when Kow increases.

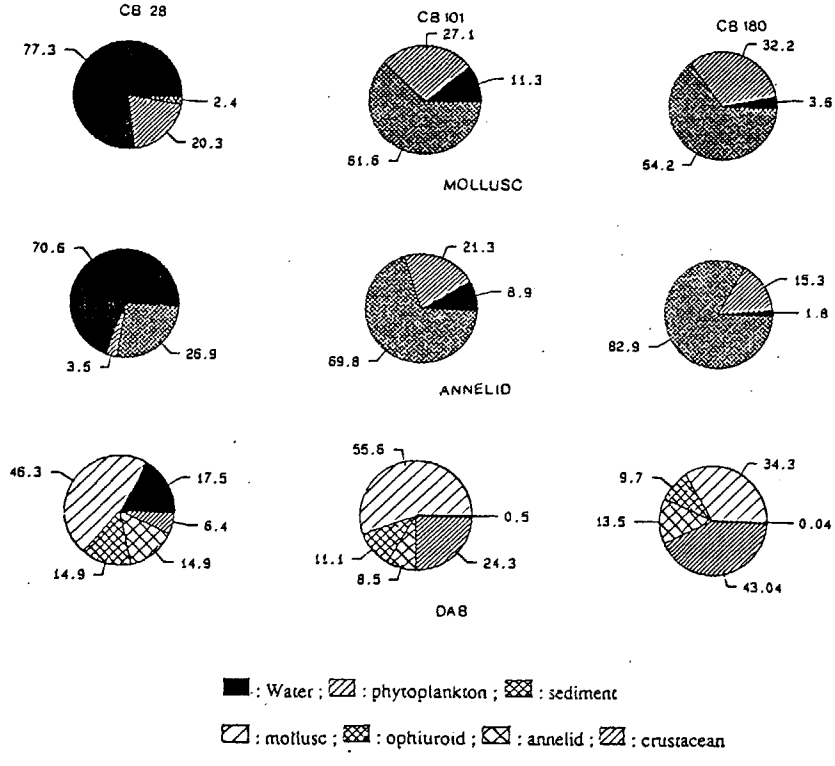

Figure 3

Six first piecharts: calculated contribution of ingestion of sediment and phytoplankton and water exposure to mollusc and annelid contamination for three congeners.

Les six premiers diagrammes circulaires représentent la contribution relative de l'ingestion de sédiment, de phytoplancton et d'eau pour la contamination des mollusques et des annélides par trois congénères.

MODEL SENSITIVITY

The model sensitivity to all parameters has been performed only for 2,2', 4,4', 5,5' hexachlorobiphenyl, the most concentrated congener in all compartments.

The purpose of the sensitivity analysis is to evaluate and describe the variation in the n output state variables induced by perturbations of the p parameters. Parameters can be sampled randomly from normal distributions (Press *et al.*, 1989), (parameters, means and standard deviations showed in Tab. 1, 2, 3). Two hundred sets of parameters have been sampled, each set leading to a specific steady state ; the data generated are gathered in a matrix with 200 rows and (p + n) columns.

Scaling of this matrix is necessary: so, each column has been divided by the value in the nominal run. The scaled matrix may be called a sensitivity matrix (Huson, 1982).

A principal component analysis (PCA) may be performed on the whole sensitivity matrix to estimate the similarities between parameters actions on the system, between the responses of state variables to parameter perturbations and finally, the link between specific parameter perturbations and observed effect on state variables. (PCA has been performed using PC software package Addad).

Two pairs of highly correlated state variables do appear [X(1)-X(5) and X(3)-X(4)], which are roughly independent from each other. X(2) is correlated with the first pair. Figure 4A represents the projection of matrix columns on the first two principal components, which extracts 18 % of total variance. In this figure, only vectors which have a good contribution to components 1 and 2 are plotted. Figure 4B gives the projection on the 2-3 plane, which represents 16 % of total variance.

Parameters SPM, LIP, A1S, and A5N are positively correlated with the pair X(1)-X(5), whereas QX1 is negatively correlated with it. Parameters CO and A4S are positively correlated with the pair X(3)-X(4), whereas Qx3 is negatively correlated whit it.

This results reveal that CB 153 contamination of molluscs [X(1)] and ophiuroid [X(2)] is highly sensitive to the lipid fraction in phytoplankton and SPM abundance in water, whereas CB 153 contamination of crustacean and annelids is most sensitive to organic carbon in sediment. Dab contamination is most sensitive to mollusc contamination.

Finally, phytoplankton lipid content, organic carbon in sediment, excretion and growth rates of benthic species can be considered as parameters to which the model is the most sensitive. Molluscs appear to be the main path controlling the contamination of dab.

DISCUSSION

Although a five-compartment food chain model may be regarded as an oversimplification of the real food web, the successful application of this model to seven PCB congeners, which differ by number and position of chlorine atoms, indicates that such a simple model can describe the food chain relatively well.

Connolly (1991) has developed a similar model of bioaccumulation for lobster and winter flounder food chains in New Bedford Harbor. His model successfully reproduces tri-, tetra-, penta-, and hexachlorobiphenyl concentrations observed at all levels of the food chain. In his study, as in ours, dietary uptake exceeds uptake across the gill and becomes the dominant route of the higher chlorinated homologues.

Our work can also be compared to the model developed by Thomann *et al.* (1992). Again, these authors developed an equilibrium model of organic chemicals accumulation in aquatic food webs with sediment interaction. In their study, they performed a normalization by using organism lipid-based chemical concentrations and sediment organic carbon-based chemical concentrations. Application of their model to an amphipod-sculpin web for Lake Ontario indicates that amphipod water contamination is relatively important for logKow < 5.5. For logKow in the range 6.5 to 7.5, contamination is almost entirely due to food web transfer from the sediment.

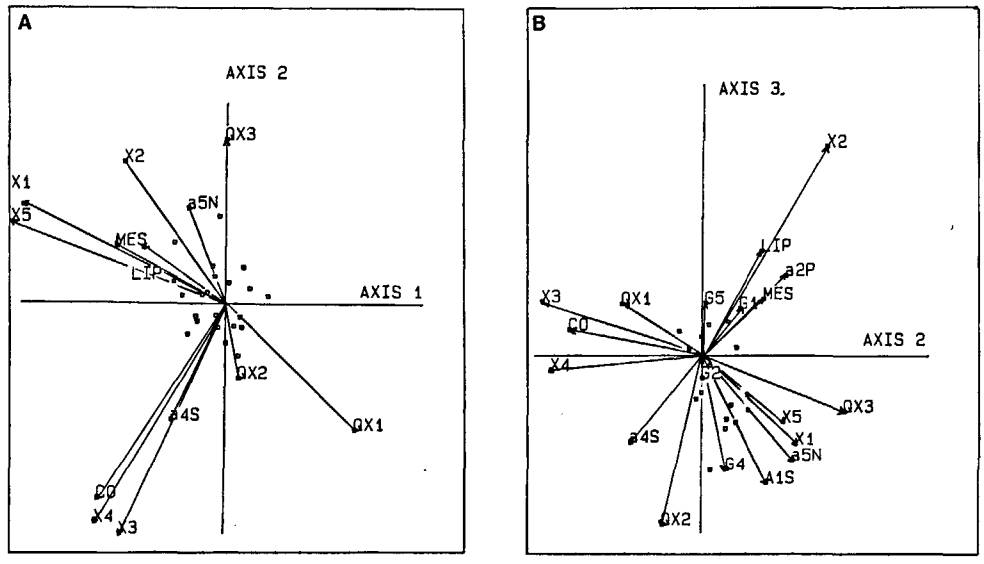

Figure 4

Principal component analysis of the sensitivities. A: first two principal components (1-2 plane); B: 2-3 plane (only vectors which have a good contribution to exposed plane are plotted).

Analyse en composantes principales sur la sensibilité. A : deux premières composantes principales (plan 1-2) ; B : plan 2-3 (seuls les vecteurs ayant une contribution relative importante aux deux plans sont représentés).

CONCLUSION

In summary, the calibration of the PCB bioaccumulation food chain model to data from the Bay of Seine shows the following results :

- The model can reproduce the magnitude of PCB contamination observed in dab and benthic species from the bay.

- Transfer of PCB through the food chain is the major contribution to observed PCB contamination. However, for congeners with few chlorine atoms, water remains an important source.

- For benthic species, sediment is the major source of contamination.

It is concluded from this work that a simple steady-state model of transfer of PCB in the food chain can help to explain the observed concentration factors. This simple steady-state model indicates that parameters of phytoplankton lipid fraction, organic carbon in sediment, excretion and growth rates are important characteristics that determine the degree to which a chemical bioaccumulates in the food chain.

In the future, other processes, such as metabolism and reproduction, will be introduced into the model. For that purpose, dab demography needs to be considered. This model can also be used dynamically to investigate seasonal variations of contamination, with a special interest in the role of gamete release as a possible way of decontamination.

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