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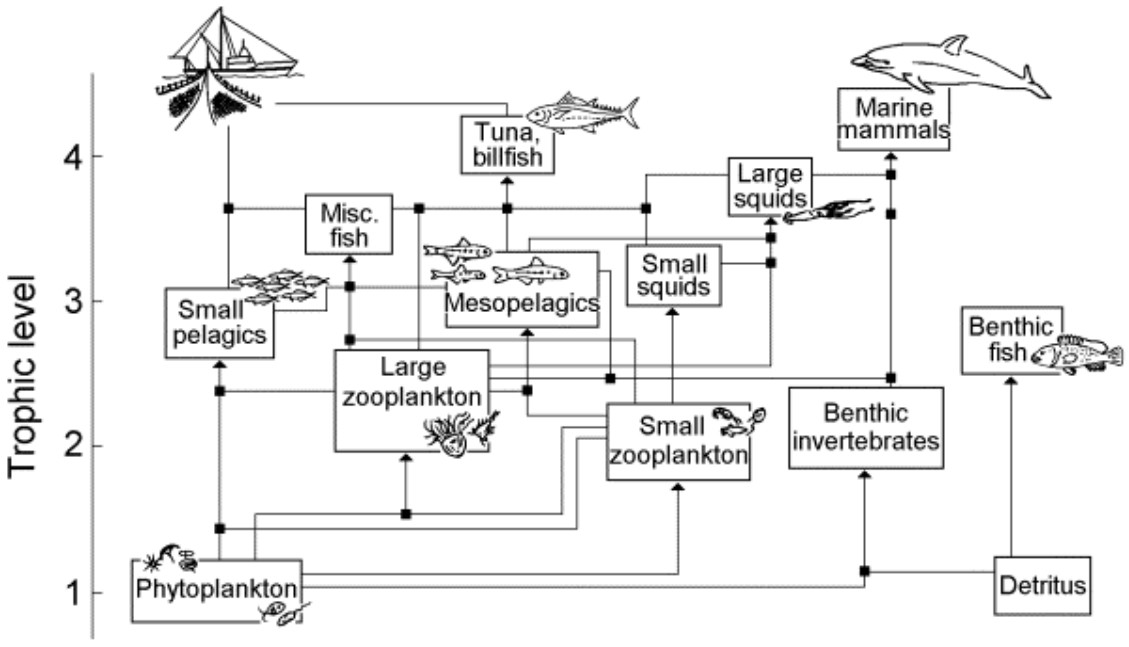
# Towards the definition of Environmental Quality Standards for biota: The influence of food web structure and dynamics on bioaccumulation and biomagnification

**José-Manuel Zaldívar**

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# EXECUTIVE SUMMARY

The analysis of four reconstructed European food webs and the dynamic modelling exercise using a simplified food web with different diet preferences suggest that a greater understanding on the food web structure and the relative contribution of the linkages between species is necessary for tailoring the developed Environmental Quality Standards (EQS) to a particular ecosystem. This is due because there is the possibility, that the same concentration of a hydrophobic and bioaccumulative chemical in the water column, could give between one or two orders of magnitude differences on the concentrations of the same top or pelagic predator when compared to a similar or closely related ecosystem.

# 1. INTRODUCTION

With the development of the Water Framework Directive (WFD, EC 2000) and the daughter directive on Environmental Quality Standards (EC, 2008) the development of EQS values for biota has become an important issue. An EQS has been defined (EC, 2000) as "*the concentration of a particular pollutant or group of pollutants in water, sediment or biota that should not be exceeded in order to protect human health and the environment*".

The development of EQS values for biota is relevant primarily for certain hydrophobic compounds for which bioaccumulation and biomagnification in the food chain that corresponds to a certain food web network may occur. The food chain and food web represent trophic structure and feeding relationships in an entire ecosystem, respectively. In this case, it is important to analyze if depending on the food web of a particular EU ecosystem, bioaccumulation values may be different for the same species and if this occurs, quantify the magnitude of the possible differences. This is important if we aim at defining EU wide EQS for Priority Substances (PS) and Priority Hazardous Substances (PHS) as defined in the Directive on environmental quality standards in the field of water policy (EC, 2008).

A food web, with all links between predators and preys, provides the first block for the assessment of the trophic position. Normally, primary producers are assigned to trophic level (TL) 1, zooplankton and zoobenthos to trophic level 2 and so on. The assignment becomes more difficult when moving to higher trophic levels since; in this case, it depends on the food web structure. However, it is also necessary to quantify the strength of the trophic interactions based on energetic importance or on the biomass-weighted composition of the diet of a certain species. In the past this was calculated using dietary data (stomach analysis contents); actually the application of stable isotope ratios or signatures of biological important elements such as carbon ($\delta^{13}C/ \delta^{12}C$) and nitrogen ($\delta^{15}N/ \delta^{14}N$) has gained considerable importance (Vander Zanden and Rasmussen, 1999). The combination of these two factors, i.e. position in the food web and diet composition, determines the bioaccumulation potential of a certain species in its ecosystem. In addition, trophic level may also change during the life of an organism due to growth and/or habitat change. For example, Vander Zanden and Rasmussen (1996) found analyzing data from several lakes that lake trout trophic position – to differentiate from trophic level which they consider discrete values- ranged from 3.0 to 4.6. Specifically, they found that even though in close lakes like Michigan, Huron, Ontario and Superior, there was a variation in the average TL[1] values, i.e. 4.36, 4.37, 4.38 and 4.44, respectively. They concluded that discrete trophic levels were not able to represent quantitatively the trophic structure. In a subsequent study, where the bioaccumulation factors (BAF) for PCBs were calculated for lake trout, Streets et al. (2006) found that

[1] In this work we do not distinguish between trophic level and trophic position.

the average *log* BAF values were 6.9, 6.7, 7.1 and 7.0 for Michigan, Huron, Ontario and Superior Lakes, respectively. In this case, small differences in TL do not reflect in the values of *log BAF*. However lake trout TLs in these lakes are quite similar and therefore one should expect similar *log BAF* values. The problem is to assess what one should expect in less similar ecosystems and how this could affect the *log BAF* values obtained.

To analyze this effect, we have studied four terrestrial and aquatic trophic network models generated previously for three similar lakes and a costal lagoon (Carafa et al., 2007, 2009a). The objective was to analyse if the similarities and differences between calculated trophic levels could have consequences on the bioaccumulation and biomagnification potential of hydrophobic contaminants. Due to data limitation, diet differences are not considered in trophic network models. To analyze this effect a simplified ecosystem plus bioaccumulation model has been developed and food preferences studied. These results are important for the assessment and development of Environmental Quality Standards for biota that will be applied at European level.

# 2. METHODS AND APPROACH

## 2.1. STUDY AREAS

To assess the main ideas of this study, we have used already developed networks from previous studies (Carafa et al., 2007; 2009a). One network corresponds to Ria Formosa, a sheltered large mesotidal temperate coastal lagoon located on the southern Portuguese coast, Fig. 1 (Carafa et al. 2007); whereas the other networks correspond to three lakes located in the Lombardy region (Northern Italy) at similar altitude, Fig. 2 (Carafa et al., 2009a).

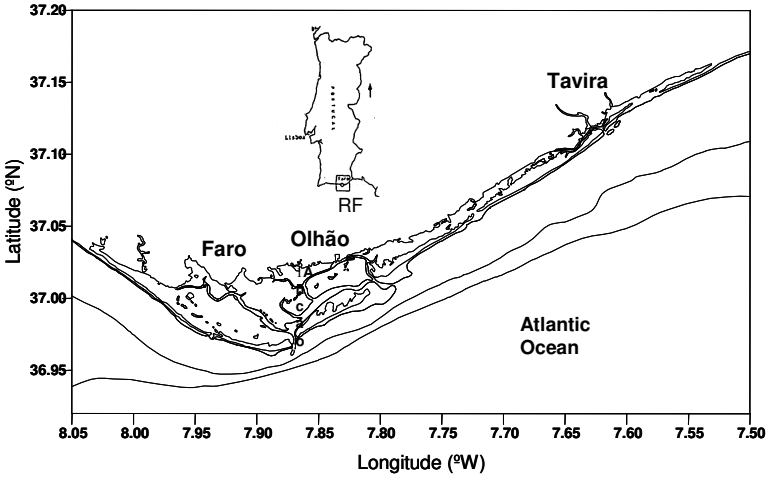



Figure 1. Geographic location of Ria Formosa.

The surface area of Ria Formosa is approximately 160 $km^2$, of which 48 $km^2$ are covered by salt marshes, 32 $km^2$ by a network of tidal channels and about 20 $km^2$ are dedicated to aquaculture ponds

(Aníbal et al., 2006). Only 14% of the surface is permanently submersed (Teixeira and Alvim, 1978) and the lagoon is covered by large beds of macroalgae and macrophytes (Loureiro et al., 2006). Ria Formosa extends for 55 km along the coast and has a mean depth of 3.5 m and is directly connected with the ocean through several inlets (Loureiro et al., 2006). The lagoon does not receive important freshwater input and salinity oscillates between 35.5 and 36.9 ‰ (Falcão et al., 1985).

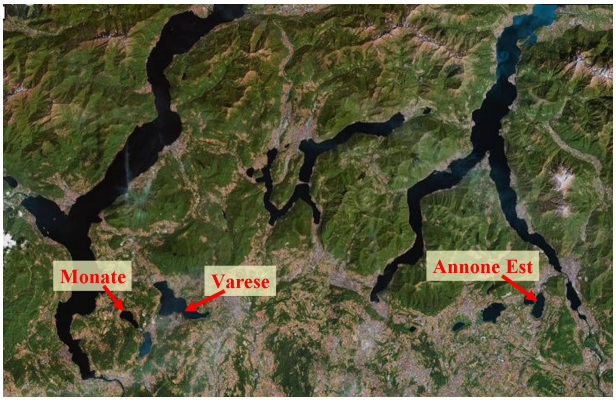


Figure 2. Map of Monate, Varese and Annone lakes.

In the second case, the three lakes are exposed to similar climatic conditions and show similar watersheds geological composition. Monate is a deep lake (max depth about 35 m) of medium dimension, with winter circulation. The low concentration of nutrients (N, P) and the good oxygenation confirm the oligotrophic status of the lake. The lake is not subject to acidification. The use of the lake water for agricultural and commercial activities is forbidden, as well as for fishery and navigation. The lake is suited for swimming and recreation purposes. The lake Varese and its watershed are larger than Monate. The lake presents also winter circulation. This Lake received, before the 90's, the waste water from Varese town. Now, after the introduction of waste water treatment plants and the restrictions on the use of phosphates in domestic detergents, the trophic status of the lake is improving, but it still falls in the eutrophic category. The lake water is used for irrigation, industry, and energy production. Fishery is allowed, but navigation is not permitted. In this lake the water quality is poor and the lake suffers from periodic anoxia and hypoxia of the hypolimnic water layer. Annone Est is a shallow water lake, with spring and autumn turnover, and it is normally covered by ice during winter. The lake receives waste water not completely treated from the watershed and, as the Varese Lake, shows hypoxia and anoxia in the hypolimnion water, most significant during summer. Its trophic state is meso-eutrophic, and the water quality is poor.

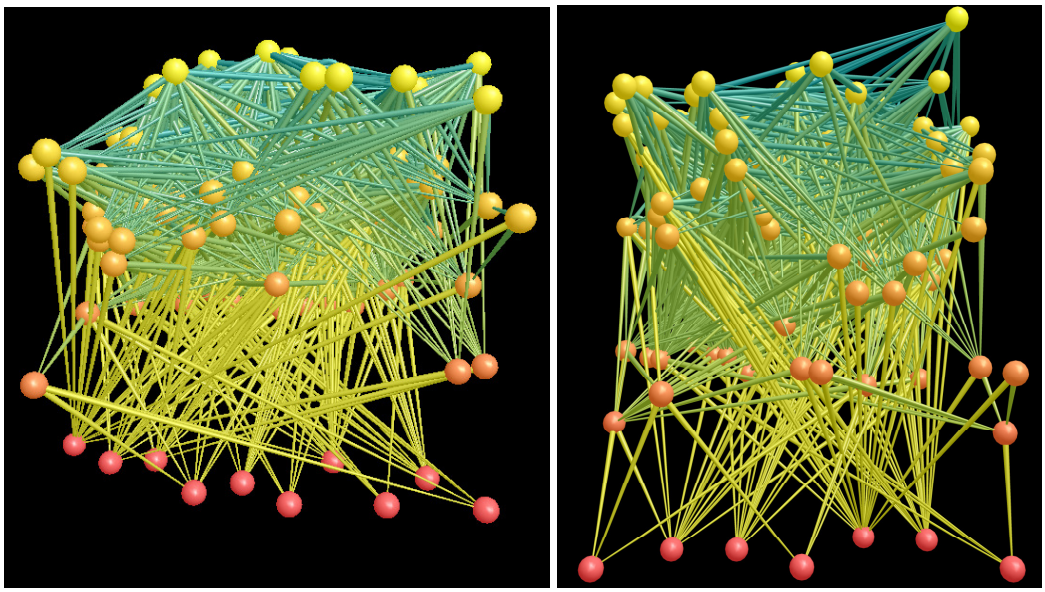
## 2.2. DATA COLLECTION AND NETWORKS DEVELOPMENT

The main problem in building a food-web model is the resolution of all trophic connections and, frequently, the particular sensitivity, the specific knowledge or the special interest of the expert,

strongly affects the results (Pinnegar et al., 2005). In addition, to develop a coherent food-web structure, data on species composition and on the diet of each species from a consolidated database are needed.

A relevant part of information on species composition for the coastal lagoon and the three sub-alpine lakes network has been drawn from CORINE biotopes database and NATURA 2000 database, whereas specific information concerning diet was gathered from several databases and scientific literature. The interested reader is referred to Carafa et al. (2007) and (2009a), and references therein.

Due to the presence of thousands of species in real ecosystems, species with similar ecological behaviour and diet composition are normally pooled in the same ecological node of the network. Therefore the number of nodes is much lower than the number of species. We use automatic classification method to divide species of a subgroup in a cluster. Binary data are transformed in a distance matrix using Jaccardian similarity index to quantify similarity between species diet (Jacard, 1901). Then species aggregation in "tropho species" with same incoming and outgoing lines is made using a cluster linkage method. This choice has been made to avoid topological redundancy, which may affect the calculation of structural network properties. Therefore, in the networks a node may be a single species or a group of species sharing the same set of preys and predators, a taxonomic group (e.g. bacteria) or a basal group (e.g. aquatic detritus or aquatic plants). Moreover lower trophic compartments are more aggregated due to lack of the high resolution information on diets of macroinvertebrates species. As an example, Figure 3 represents two of the four food-web networks generated, with species ordered according to their TL from bottom to top.



a) b)

Figure 3. Example food-web network structures: a) Ria Formosa coastal lagoon; b) Monate Lake. Representation performed with FoodWeb3D (Yoon et al., 2004). Species ordered according to their TL from bottom to top.

Estimation of a species' trophic level, based only on binary link information, has been conducted using the short-weighted trophic level method (Pimm, 1980; 1982), which closely estimates traditional mean flow-based TL (Williams and Martinez, 2004). According to Cohen and Briand (1984), a 'top species' (T) is a species without predator, an 'intermediate species' (I) is a species that is both a predator and a prey and a 'basal species' (B) has predator but no prey.

## 2.3. BIOCONCENTRATION AND BIOACCUMULATION

Transfer mechanisms of persistent hydrophobic contaminants in aquatic organisms are essentially two: the first one is the direct uptake of dissolved phase from water trough skin or gills, named bioconcentration, the second one is the indirect uptake of bound contaminants to suspended particular matter and through consumption of contaminated food (biomagnification).

The bioaccumulation of pollutants may be an important source of hazard for the ecosystem, due to adverse effect not quickly evident (e.g. acute or chronic toxicity) but that became manifested after years in the higher levels of the trophic food web or in a later stage of life of organisms or after several generations (Van der Oost *et al*., 2003). Accumulation is a general term for the net result of absorption (uptake), distribution, metabolism and excretion (ADME) of a substance in an organism. Information on accumulation in aquatic organisms is vital for understanding the fate and effects of a substance in aquatic ecosystems. In addition, it is an important factor when considering whether long-term ecotoxicity testing might be necessary. This is because chemical accumulation may result in internal concentrations of a substance in an organism that cause toxic effects over long-term exposures even when external concentrations are very small. Highly bioaccumulative chemicals may also transfer through the food web, which in some cases may lead to biomagnification.

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 concentration of the chemical in the organism and in water at equilibrium, normally $C_w$ is the dissolved water concentration.

$$BCF = \frac{C_b}{C_w} \tag{1}$$

The existence of equilibrium between the concentration of the chemical in the organism and the concentration in the water is not easy to asses. For example, for rainbow trout Vigano *et al*. (1994) measured a time range between 15 and 256 days to reach equilibrium after exposure to different concentrations of PCBs.

Biomagnification refers to accumulation of substances via the food chain. It may be defined as an increase in the (fat-adjusted) internal concentration of a substance in organisms at succeeding trophic

levels in a food chain. The biomagnification factor (*BMF*) can be expressed as the ratio of the concentration in the predator and the concentration in the prey:

$$BMF = \frac{C_b}{C_p} \tag{2}$$

where $C_b$ is the steady-state chemical concentration in the organism (mg $kg^{-1}$) and $C_p$ is the steady-state chemical concentration in the diet (mg $kg^{-1}$).

The term bioaccumulation refers to uptake from all environmental sources including water, food and sediment. The bioaccumulation factor (*BAF*) can be expressed for simplicity as the steady-state (equilibrium) ratio of the substance concentration in an organism to the concentration in the surrounding medium (e.g. water). Normally, it is evaluated using a multiplicative approach. Therefore, the Bioaccumulation Factor (*BAF*) may be calculated as:

$$BAF = BCF \cdot \prod_{i=1}^{n} BMF_i \tag{3}$$

where the number of biomagnifications factors depends on the trophic level or position of the organism in the food web.

The mass balance of a contaminant (A) in the tissue of an aquatic organism, $C_b$ (mg $kg^{-1}$), can be defined as (adapted from Thomann, 1989 and Thomann *et al.*, 1992):

$$\frac{dC_b}{dt} = k_u C_w + k_f C_p - k_d C_b - k_m C_b - k_g C_b \tag{4}$$

where the first two terms indicate the uptake (*u*) of contaminant from water (*w*) and predation (*p*), respectively, and the third, fourth and fifth terms indicate losses of contaminants through depuration (*d*) (release from gill membranes or excretion through faeces), metabolism (*m*) and dilution effect of growth (*g*), respectively. Removal of chemicals in an aquatic organism is realized essentially through two main pathways: the contaminant is either eliminated by depuration/excretion in the original chemical form (parent molecule) or bio-transformed by the organism. The latter process leads in general to the formation of more hydrophilic compounds. In this case the metabolites are rapidly excreted after a detoxification reaction. These compounds are normally less harmful than the parent compound. However, in some cases the parent compound can be "bioactivated" through metabolic reactions and lead to formation of a metabolite more toxic than the former molecule (Van der Oost, *et al.,* 2003). The velocity and efficiency of metabolic clearance have been demonstrated to be a function of several species-specific characteristics: presence of enzymes, feeding status, stage of life, spawning period (Van der Oost *et al.*, 2003).

Using this model and assuming steady-state conditions, i.e. $dC_b/dt = 0$, then it is possible to calculate the bioconcentration factor ($BCF = C_b/C_w$, see Eq. 1) as:

$$BCF = \frac{k_u}{(k_d + k_m + k_g)} \quad (5)$$

In addition, the biomagnification factor ($BMF=C_b/C_p$, see Eq. 2) defined as the ratio between the uptake of a contaminant from food and its removal by depuration/excretion (*d*), metabolism (*m*) and growth (Sijm *et al*., 1992) is given by:

$$BMF = \frac{k_f}{k_d + k_m + k_g} \quad (6)$$

This simple model considers the organisms as a single compartment, more complete models that take into account changes in lipid contents and size of the animal has been developed, see for example Kooijman and van Haren (1990) and van Haren et al. (1994).

According to the Guidance Document for Environmental Quality Standards, which is based on REACH Guidance, a simple food web is assumed that consists of water – *BCF*→ fish/mussel – $BMF_1$ → fish-eating predator. In the case of marine ecosystems, for marine top predators, an additional biomagnification factor in prey of top predators ($BMF_2$) should be applied. Default values for *BMFs* are provided by the TGD (EC, 2003) and summarized in Table 1. However, *BCF* and *BAFs* should be preferably measured rather than applying default values.

Table 1. Default BMF values for organic substances (EC, TGD, 2003)

| **log $K_{ow}$ of substance** | ***BCF* (fish)** | ***$BMF_1$*** | ***$BMF_2$*** |
|---|---|---|---|
| < 4.5 | < 2000 | 1 | 1 |
| 4.5 - < 5 | 2000-5000 | 2 | 2 |
| 5 – 8 | > 5000 | 10 | 10 |
| > 8 -9 | 2000-5000 | 3 | 3 |
| > 9 | < 2000 | 1 | 1 |

A careful evaluation of these values is important, since the conversion from a biota standard to an equivalent water concentration can introduce uncertainty. This is especially relevant for highly lipophilic substance (i.e. *BCF*>2000). Generally, substances with a *BCF* of 500 or less can be converted to an equivalent water concentration with reasonable confidence. Here we summarize two cases, from the EQS Guidance document, to highly the problems that can occur depending on the values used for the calculation.

- Hexachlorobenzene (HCB) example

| | |
|---|---|
| $PNEC_{water}$ | 13 ng $l^{-1}$ (EQS Substance data sheet, 2005) |
| $PNEC_{oral}$ | 16.7 µg $kg^{-1}$(EQS Substance data sheet, 2005) |
| *BAF* | 52,300 L $kg^{-1}$ (mean value; 26 experimental fish *BAF* values, mean value; min= 8,130 max 550,000, median 51,900), Arnot and Gobas (2006) |

$$EQS_{water} = \frac{EQS_{biota}}{BAF}$$

Extrapolated $PNEC_{water}$

| | |
|---|---|
| Calculated with median *BAF* | 0.3 ng $l^{-1}$ |
| Calculated with minimum *BAF* | 2 ng $l^{-1}$ |
| Calculated with maximum *BAF* | 0.03 ng $l^{-1}$ |

- Lindane example

| | |
|---|---|
| $PNEC_{water}$ | 20 ng $l^{-1}$ (EQS Substance data sheet, 2005) |
| $PNEC_{oral}$ | 33 µg $kg^{-1}$ |
| *BCF* | 1300 (selected in the EQS datasheets, min= 220, max= 2200), (EQS Substance datasheet, 2005) |
| *BMF* | a BMF of 1 was assumed according to the TGD |

$$EQS_{water} = \frac{EQS_{biota}}{BCF.BMF}$$

Extrapolated $PNEC_{water}$

| | |
|---|---|
| Calculated with selected *BCF* | 25 ng $l^{-1}$ |
| Calculated with minimum *BCF* | 150 ng $l^{-1}$ |
| Calculated with maximum *BCF* | 15 ng $l^{-1}$ |

The worked examples for Hexachlorobenzene and Lindane below show that for Hexachlorobenzene the biota EQS is likely to be the critical EQS regardless of the uncertainties of the extrapolation, whereas in the case of Lindane there is uncertainty as to whether the biota EQS or the water EQS are the critical EQS.

Ideally, the *BMFs* should be based on measured data. However, such data are not available for all compounds and all ecosystems. Arnot and Gobas (2006) in a recent review found that only 4% of the chemicals that require review on the Canadian Domestic Substances List have *BCF* or *BAF* values and of these 76% have less than three good quality values. The absence of experimental data has forced the development of estimation models using empirical correlations, quantitative structure-activity relationships (QSARs) (Pavan et al., 2008) or fate models (Thomann, 1989; Thomann et al. 1992).

As it can be seen from the examples above, the use of correct values of *BCF* and *BMF* would allow a better estimation of the critical path when defining EQS values. In addition, since these values depend on the food web, it seems important to develop better approaches to characterize more in detail EU food webs.

In general, the most reliable data on biomagnification originate from trophic magnification studies. In these studies the levels of contaminants in several species in an ecosystem are measured and expressed as a function of the trophic level. The trophic level is mostly derived from stable isotope techniques (Keough et al., 1996; Vander Zanden and Rasmussen, 1999). The advantage of this method is that it takes into account the magnification along the whole food chain and it is not subject to the arbitrary

choice of two species for which a *BMF* is calculated. In field or laboratory studies, where normally lipid normalized concentrations are measured, care should be taken in interpreting these values, because they only represent one link in the food chain and may not represent the overall biomagnification potential of a substance. In these studies the biomagnification factor is restricted to the ratio between the concentrations in the predator and in its prey or food in the case of a laboratory study.

For example, Vander Zanden and Rasmussen (1996) found analyzing the concentration of PCB in lake trout in 21 lakes that log PCB (ng/ gww) correlated with the trophic position, $r^2 = 0.83$, with the following expression:

$$\log PCB = -6.07 + 2.11 \cdot TL \qquad (7)$$

Since lake trout trophic position changed from 3.0 to 4.6 the total concentrations of PCBs spanned over nearly two orders of magnitude.

### 2.3.1. Modelling bioaccumulation dynamics

To analyze the effects of diet preferences on bioaccumulation potential, we have implemented two simple ecological and bioaccumulation models.

*2.3.1.1. Ecological model*

A schematic flow diagram of the aquatic ecosystem model analysed in this work, is summarised in Fig. 4. Besides the presence of available nutrients in the water column, the model accounts for phytoplankton, zooplankton, and fish, as well as for nutrients bound in the sediments. In addition the model contains the so called microbial loop, which accounts for the mineralization of dead organic mater, called detritus, performed by the bacteria.

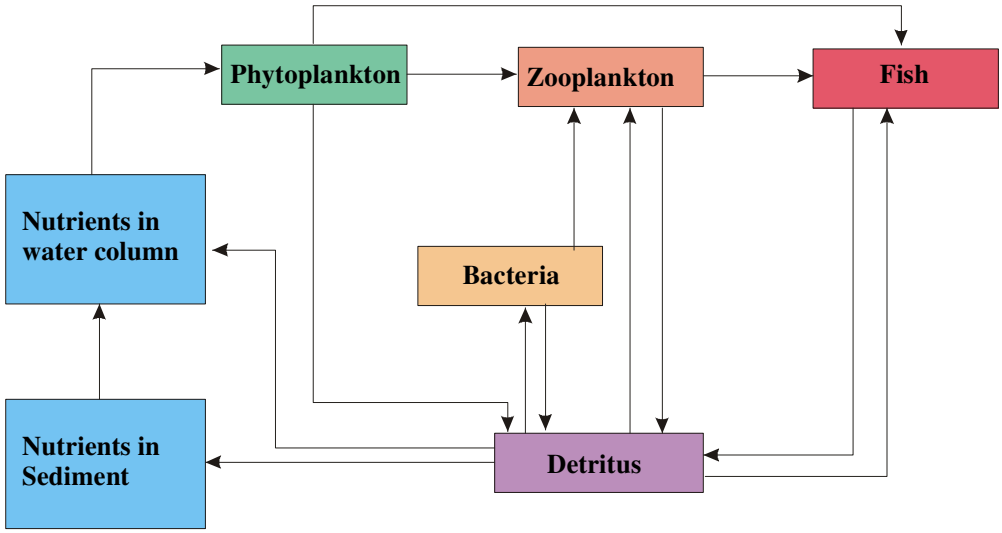



Figure 4. Simplified flow diagram in a typical aquatic ecosystem (Mosekilde,1996).

In this case study, we do not consider pelagic predators since we have stopped the food web at a TL= 3.

The model based on Fig. 4, has seven state equation describing the dynamics of phytoplankton ($P$), zooplankton ($Z$), fish ($F$), bacteria ($B$), nutrients in the sediment ($N_S$), nutrients in the water column ($N_W$) and detritus ($D$). These equations can be written, following Mosekilde (1996), as:

$$\frac{dP}{dt} = c(t)P\mu_{m,P}\frac{N_w}{N_w + K_P} - Z\mu_{m,Z}\frac{P}{P+K_Z}\frac{Y_{P,0}+Y_P}{2} - F\mu_{m,F}\frac{P}{P+K_F}\frac{X_{P,0}+X_P}{2} - \tau_P P - \rho P \quad (8)$$

$$\frac{dZ}{dt} = Z\mu_{m,Z}\left(\frac{P}{P+K_Z}\frac{Y_{P,0}+Y_P}{2} + \frac{B}{B+K_Z}\frac{Y_{B,0}+Y_B}{2} + \frac{D}{D+K_Z}\frac{Y_{D,0}+Y_D}{2}\right) - F\mu_{m,F}\frac{Z}{Z+K_F}\frac{X_{Z,0}+X_Z}{2} - \tau_Z Z - \rho Z \quad (9)$$

$$\frac{dF}{dt} = F\mu_{m,F}\left(\frac{P}{P+K_F}\frac{X_{P,0}+X_P}{2} + \frac{Z}{Z+K_F}\frac{X_{Z,0}+X_Z}{2} + \frac{D}{D+K_F}\frac{X_{D,0}+X_D}{2}\right) - \tau_F F - \rho F \quad (10)$$

$$\frac{dB}{dt} = B\mu_{m,B}\frac{D}{D+K_B}(1-\alpha) - Z\mu_{m,Z}\frac{B}{B+K_Z}\frac{Y_{B,0}+Y_B}{2} - \tau_B B - \rho B \quad (11)$$

$$\frac{dN_S}{dt} = \beta D - \gamma N_S \quad (12)$$

$$\frac{dN_W}{dt} = \gamma N_S + \alpha B\mu_{m,B}\frac{D}{D+K_B} - c(t)P\mu_{m,P}\frac{N_W}{N_W+K_P} + \rho(C_i - N_W) \quad (13)$$

$$\frac{dD}{dt} = \tau_P P + \tau_Z Z + \tau_F F + \tau_B B - Z\mu_{m,Z}\frac{D}{D+K_Z}\frac{Y_{D,0}+Y_D}{2} - F\mu_{m,F}\frac{D}{D+K_F}\frac{X_{D,0}+X_D}{2} - B\mu_{m,B}\frac{D}{D+K_B} - \beta D - \rho D \quad (14)$$

In this model an annual sine function is used to describe the variation in energy input (light and temperature) to the system:

$$c(t) = \frac{1}{2}\left(\sin\left(2\pi\frac{t-80}{365}\right)+1\right) \quad (15)$$

The specific growth rates follow the Monod equation modified for taking into account the case that there is more than one prey. In this case, the growth rate becomes the sum of n terms of Monod type, one term for each of the n prey species that the considered predator eats. Each of these n terms is weighted so the *i-th* contribution reads:

$$k_g^j = \mu_{m,i}\frac{S_i}{S_i+K}\frac{X_{i,0}+X_i}{2} \quad (16)$$

with $X_i = S_i \Big/ \sum_{i=1}^{n} S_i$ and $X_{i,0}$ = constant. Here $S_i$ is the biomass of the i-th prey. $\mu_{m,i}$ and $K$ are the maximal specific growth rate and the half-saturation constant respectively. The parameter $X_{i,0}$ defines

the optimal fraction of the predator's diet deriving from prey of species $i$, and $X_i$ measures the actual value of this fraction. Moreover, $\alpha$ is the mineralization part of the bacterial uptake, $\beta$ the sedimentation rate, $\gamma$ the mineralization rate, $\rho$ the rate of dilution, $C_i$ the inlet concentration of nutrients, $\tau$ is the mortality rate. ($X_P$, $X_Z$, $X_D$) defines the actual composition of the fish diet, and ($Y_P$, $Y_B$, $Y_D$) the composition of the zooplankton diet. Table 2 summarizes the values suggested by Mosekilde (1996).

Table 2. Base case parameters for the aquatic ecosystem.

| Parameter | Unit | Value | Parameter | Unit | Value |
|---|---|---|---|---|---|
| $\mu_{m,P}$ | day$^{-1}$ | 2.5 | $\alpha$ | - | 0.33 |
| $\mu_{m,Z}$ | day$^{-1}$ | 0.5 | $\beta$ | day$^{-1}$ | 0.009 |
| $\mu_{m,F}$ | day$^{-1}$ | 0.047 | $\gamma$ | day$^{-1}$ | 0.0005 |
| $\mu_{m,B}$ | day$^{-1}$ | 2.9 | $\rho$ | day$^{-1}$ | 0.001 |
| $K_P$ | mg/l | 0.6 | $C_i$ | mg/l | 2.0 |
| $K_Z$ | mg/l | 0.6 | $X_{Z,0}$ | - | 0.8 |
| $K_F$ | mg/l | 0.28 | $X_{P,0}$ | - | 0.1 |
| $K_B$ | mg/l | 0.2 | $X_{D,0}$ | - | 0.1 |
| $\tau_P$ | day$^{-1}$ | 0.1 | $Y_{P,0}$ | - | 0.85 |
| $\tau_Z$ | day$^{-1}$ | 0.1 | $Y_{B,0}$ | - | 0.05 |
| $\tau_F$ | day$^{-1}$ | 0.02 | $Y_{D,0}$ | - | 0.1 |
| $\tau_B$ | day$^{-1}$ | 0.9 | | | |

Figures 5 and 6 show an example of a simulation run covering one year after the model has reached steady state (limit cycle). As can be seen this highly idealised model is able to reproduce the phytoplankton spring bloom immediately followed by an increase in zooplankton population, which in turn depletes the phytoplankton population. The fish population, on the other hand, exhibits fewer and smoother fluctuations. Concerning nutrients, Fig. 6, the model follows the typical behaviour of depletion during spring and recovery during winter. It is important to notice that this model does not consider nutrients separately, but assumes a distribution that follows Redfield molar ratio (C:N:P = 106:16:1). This implies that single limiting nutrient can not occur as normally happens in aquatic ecosystems, but for our objectives this does not constitute a problem.

A detailed analysis on attractors, bifurcation points and influence of different parameters on the model dynamic behaviour can be found in Mosekilde (1996). In this work we are interested in analyzing the effects of diet preferences on bioaccumulation potential. For this reason we are going to change only the zooplankton ($Y_P$, $Y_B$, $Y_D$) and fish ($X_P$, $X_Z$, $X_D$) diet parameters.

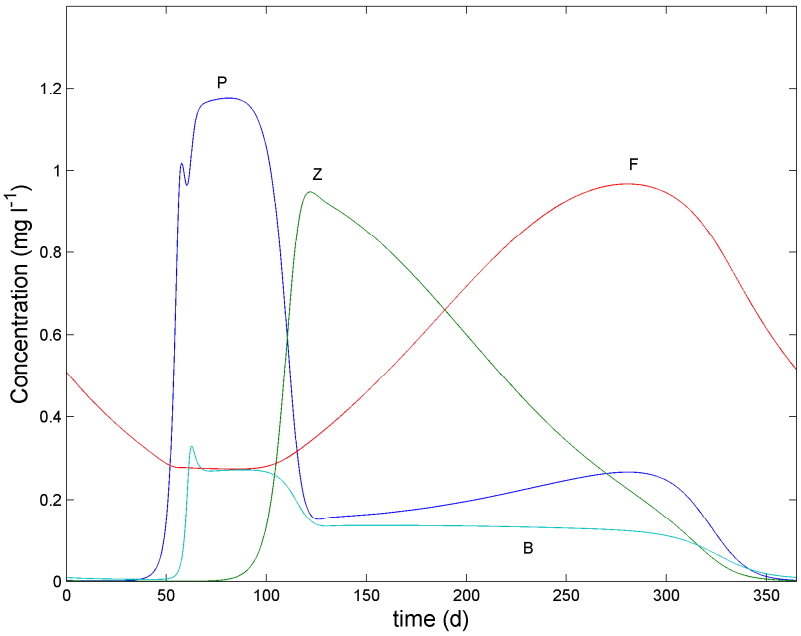



Figure 5. Simulated annual variations of the four species in the model.

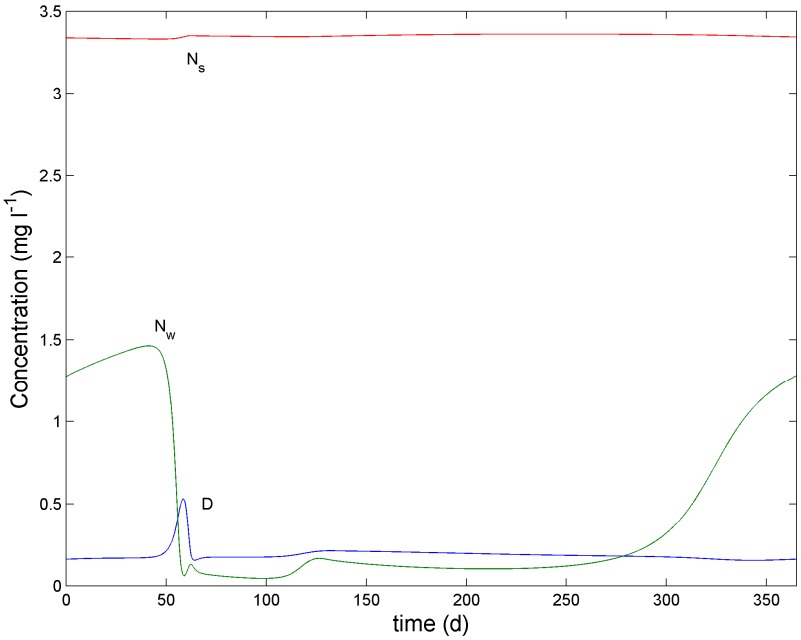



Figure 6. Simulated annual variations of the detritus and nutrients (water column and sediments) in the model.

*2.3.1.2. Bioaccumulation model*

The bioaccumulation model may be defined using Eq. (4), but a distinction between phytoplankton and bacteria that do not have predation from zooplankton and fish should be made.

*- Bioconcentration in phytoplankton and bacteria*

Bioconcentration of contaminants by phytoplankton can be calculated assuming constant uptake and depuration rates and by modelling the water-phytoplankton exchange as shown by Del Vento and Dachs (2002).

The concentration of a chemical (mg kg ww$^{-1}$) in phytoplankton ($C_P$) and bacteria ($C_B$) over time can be expressed using Eq. (4), assuming there is no biomagnification ($k_f$ = 0), a self-sustained phytoplankton community ($k_g$ = 0), and a metabolism rate much lower than the depuration rate. Under these assumptions Eq. (4) becomes:

$$\frac{dC_P}{dt} = k_u^P \cdot C_w^{dis} - k_d^P \cdot C_P \tag{17}$$

$$\frac{dC_B}{dt} = k_u^B \cdot C_w^{dis} - k_d^B \cdot C_B \tag{18}$$

where $k_u$ (m$^3$ kg$^{-1}$ d$^{-1}$) and $k_d$ (d$^{-1}$) are the uptake and depuration rates constants. Bacteria feed on detritus. However, it is assumed that there is no egestion and therefore we do not consider the concentration in the particulate phase ($D$). Uptake and depuration constants can be parameterized as function of bioconcentration factors of the chemical, permeability ($P_r$, m/h) of the cell membrane and specific surface area ($S_p$, m$^2$/kg) (Del Vento and Dachs, 2002):

$$k_{dep} = \frac{S_p \cdot P_r}{BCF}$$
$$k_{upt} = S_p \cdot P_r \tag{19}$$

The specific surface area of phytoplankton and bacteria has been estimated by assuming cylindrical and spherical shapes respectively. Phytoplanton cells are taken as of 11.5 µm of diameter and 31.5 µm of height respectively. The density of phytoplankton ($\rho_{phyto}$) is taken as of 1025 kg m$^{-3}$ (Del Vento and Dachs, 2002). This gives a volume of $3.27 \cdot 10^{-15}$ m$^3$, area of $1.35 \cdot 10^{-9}$ m$^2$, and specific surface area ($Sp$) of 401.29 m$^2$ kg$^{-1}$.The specific surface area ($S_p$) of bacteria has been calculated assuming a diameter of 1 µm, spherical shape and density ($\rho_{bac}$) equal to 1080 kg m$^{-3}$, which gives 2777.78 m$^2$ kg$^{-1}$.

In order to predict uptake and depuration rates it is necessary to know values for $BCF$ and $P_r$. Since estimations of $BCF$ and $P_r$ exist only for a few number of compounds (e.g. Swackhamer and Skoglund, 1993; Arnot and Gobas, 2006), these parameters have been calculated using empirical approximation based on the physical-chemical properties of the contaminant.

It has been demonstrated (Swackhamer and Skoglund, 1993; Stange and Swackhamer, 1994) that, for many organic compounds, the logarithm of the bioconcentration factor plotted against the logarithm of the octanol/water partition coefficient gives two linear correlations (with a plateau in correspondence to log $K_{ow} \approx 6.5$, that can be fitted by least squares and may be represented by the following log linear equations (Del Vento and Dachs, 2002):

$$\log BCF = 1.085 \log K_{ow} - 3.770 \qquad \text{for } \log K_{ow} < 6.4 \tag{20}$$

$$\log BCF = 0.343 \log K_{ow} + 0.913 \qquad \text{for } \log K_{ow} \geq 6.4 \qquad (21)$$

The same considerations can be made for the estimation of permeability of cell membrane and similar regressions have been proposed (Del Vento and Dachs, 2002):

$$\log P_r = 1.340 \log K_{ow} - 8.433 \qquad \text{for } \log K_{ow} < 6.4 \qquad (22)$$

$$\log P_r = 0.078 \qquad \text{for } \log K_{ow} \geq 6.4 \qquad (23)$$

As an example, Table 3 summarizes the uptake and depuration constants used in Eqs. (17)-(18) to calculate the concentrations of several PAHs, PCBs and PCCD/Fs in phytoplankton and bacteria.

Table 3 Uptake ($m^3 \cdot kg^{-1} \cdot d^{-1}$) and depuration ($d^{-1}$) constants for several families of chemicals used in the model.

| **Compound** | ***log $K_{ow}$*** | **Phytoplankton (*P*)** | | **Bacteria (*B*)** | |
|---|---|---|---|---|---|
| **PAHs** | | ***$k_u$*** | ***$k_d$*** | ***$k_u$*** | ***$k_d$*** |
| Naphthalene | 3.37 | 0.0486 | 0.0631 | 0.336 | 0.436 |
| Fluorene | 4.12 | 0.491 | 0.0979 | 3.400 | 0.678 |
| Antracene | 4.54 | 1.795 | 0.125 | 12.425 | 0.868 |
| Phenanthrene | 4.57 | 1.969 | 0.128 | 13.630 | 0.883 |
| Pyrene | 5.17 | 12.539 | 0.181 | 86.796 | 1.256 |
| Fluoranthene | 5.22 | 14.630 | 0.187 | 101.274 | 1.294 |
| Benzo[a]anthrecene | 5.84 | 99.097 | 0.269 | 685.96 | 1.862 |
| Chrysene | 5.84 | 99.097 | 0.269 | 685.96 | 1.862 |
| Benzo [a]pyrene | 6.04 | 183.679 | 0.302 | 1271.446 | 2.094 |
| Benzo[b]fluoranthene | 6.44 | 480.240 | 0.363 | 3324.282 | 2.511 |
| Benzo[k]fluoranthene | 6.44 | 480.240 | 0.363 | 3324.282 | 2.511 |
| Indeno[1,2,3-cd]pyrene | 6.58 | 480.240 | 0.325 | 3324.282 | 2.248 |
| **PCBs** | | | | | |
| PCB28 | 5.67 | 58.649 | 0.243 | 405.974 | 1.685 |
| PCB52 | 5.80 | 87.591 | 0.263 | 606.314 | 1.818 |
| PCB101 | 6.40 | 480.240 | 0.374 | 3324.282 | 2.591 |
| PCB118 | 6.70 | 480.240 | 0.295 | 3324.282 | 2.045 |
| PCB138 | 6.83 | 480.240 | 0.267 | 3324.282 | 1.845 |
| PCB153 | 6.92 | 480.240 | 0.248 | 3324.282 | 1.718 |
| PCB180 | 7.40 | 480.240 | 0.170 | 3324.282 | 1.176 |
| **PCDD/Fs** | | | | | |
| TCDD | 6.9 | 480.240 | 0.252 | 3324.282 | 1.746 |
| PeCDD | 7.4 | 480.240 | 0.170 | 3324.282 | 1.176 |
| HxCDD | 7.8 | 480.240 | 0.124 | 3324.282 | 0.858 |
| HpCDD | 8.0 | 480.240 | 0.106 | 3324.282 | 0.732 |
| OCDD | 8.2 | 480.240 | 0.090 | 3324.282 | 0.625 |
| TCDF | 7.7 | 480.240 | 0.134 | 3324.282 | 0.928 |
| PeCDF | 7.6 | 480.240 | 0.145 | 3324.282 | 1.004 |
| HxCDF | 7.7 | 480.240 | 0.134 | 3324.282 | 0.928 |
| HpCDF | 7.5 | 480.240 | 0.157 | 3324.282 | 1.087 |
| OCDF | 7.6 | 480.240 | 0.145 | 3324.282 | 1.004 |

*- Bioaccumulation in Zooplankton and Fish*

In the case of zooplankton, we have also to consider the intake due to food consumption as well as the egestion and metabolization. In this case the concentration of a chemical in the zooplankton ($C_Z$) over time can be expressed as:

$$\frac{dC_Z}{dt} = k_u^Z \cdot C_w^{dis} + k_g^D \cdot C_D + k_g^P \cdot C_P + k_g^B \cdot C_B - k_d^Z \cdot C_Z - k_e^Z \cdot C_Z - k_m^Z \cdot C_Z \qquad (24)$$

where $k_g^j$ ($m^3 \cdot kg^{-1} \cdot d^{-1}$) are the grazing constants for the different food items (detritus, phytoplankton and bacteria), $k_e$ ($d^{-1}$) and $k_m$ ($d^{-1}$) are the egestion and metabolization rate constants for zooplankton. A similar expression may be written for fish:

$$\frac{dC_F}{dt} = k_u^F \cdot C_w^{dis} + k_g^D \cdot C_D + k_g^P \cdot C_P + k_g^Z \cdot C_Z - k_d^F \cdot C_F - k_e^F \cdot C_F - k_m^F \cdot C_F \quad (25)$$

In this work, values for the constants have been taken from existing correlations in literature. Following Farley et al. (1999) the uptake constant (L $gww^{-1}$ $d^{-1}$) for aquatic species can be expressed as:

$$k_u = \varepsilon \frac{R_{O_2}}{[O_2]} \quad (26)$$

where $\varepsilon$ is a transfer efficiency constant that may be expressed as a function of log $K_{ow}$ as: $\varepsilon = 0.9632 - 0.0892 \cdot \log K_{ow}$, $R_{O2}$ is the respiration rate, and [$O_2$] is the dissolved oxygen concentration, which in our case is assumed constant and equal to 8.0 $10^{-3}$ g $l^{-1}$. The respiration rate in g $O_2$ $gww^{-1}$ $d^{-1}$ can be calculated according with Thomann (1989) as:

$$R_{O_2} = a_{oxygen-carbon} \cdot a_{carbon-drywt} \cdot f_{drywt} \cdot R \quad (27)$$

where for respiration rates in oxygen equivalents $a_{oxygen\text{-}carbon}$ and $a_{carbon\text{-}dry\ wt}$ are taken as 2.67, 0.4 respectively, $f_{dry\ wt}$ is assumed to be equal to 0.2 and 0.25, for zooplankton and fish, respectively and $R$ is calculated as (Farley et al., 1999):

$$R = val_1 \cdot e^{0.06293 \cdot T} \quad (28)$$

with $val_1$ equal to 0.01249 for zooplankton and 0.0047 for fish.

The depuration constant indicates the chemical losses from gill and skin, and they can be expressed as (Farley et al., 1999):

$$k_d = \frac{k_u}{f_{lipid} \cdot K_{ow}} \quad (29)$$

where $f_{lipid}$ is the fraction lipid weight (kg (lp)/kg(ww)), which in zooplankton and fish is 0.06 (Farley et al., 1999).This equation assumes that the same transport mechanisms responsible of chemical uptake from water are active as well in the transport out of lipidic cell membranes.

The excretion constant ($k_e$) was taken from Van der Linde et al. (2001) and set constant to 0.05 $d^{-1}$, which is the average value for chlorinated dioxins, furans and PCBs

The contaminant metabolic rate ($k_m$) is strictly related to specific chemical-physical properties of the compound and to the particular metabolic processes and enzymes of the organism. For the case of PAHs a mean value was estimated as 0.74 $d^{-1}$ by Berrojabiz et al. (2006), whereas for the case of PCBs and PCDD/Fs is normally assumed negligible (Farley et al., 1999).

Concerning grazing, the model uses the values provided by the ecological model taking into account

the diets of zooplankton and fish, see Eq (16). These values are then multiplied by the concentrations of contaminant in each ecosystem compartment with the exception of detritus.

To couple the ecological and fate models the total concentration of the chemical –maintained constant during the simulation- is divided between dissolved and particulate which is associated to the concentration of detritus ($D$). This is done following Carafa et al. (2006, 2009b) and Dueri et al. (2009) as:

$$C_w^{dis} = \frac{C_{tot}}{1 + K_d \cdot D} \tag{30}$$

$$C_{part} = \frac{K_d \cdot D \cdot C_{tot}}{1 + K_d \cdot D} \tag{31}$$

where $K_d$ is the equilibrium partition coefficient between particulate and freely-dissolved phases attached to calculate from the total concentration of the chemical, the dissolved concentration and of the chemical and it may be obtained from $K_{ow}$ and the fraction of organic carbon, $f_{oc}$, in detritus as (Farley et al., 1999):

$$K_d = f_{oc} \cdot K_{ow} \tag{32}$$

The particulate phase concentration is then used as the concentration of the chemical in detritus.

As an example, a five year simulation of the concentrations in phytoplankton, bacteria, zooplankton and fish are shown in Fig. 7 using the ecological model dynamics from Figs. 5-6. We have assumed a total constant concentration in the water column of $10^{-3}$ mg $l^{-1}$ of PCB101. As it can be observed, the system dynamics requires one year for reaching the attractor (limit cycle) and it changes during the year. This is mainly due to the temperature variations, but also to the dynamics of detritus that controls the partitioning between dissolved and particulate phases.

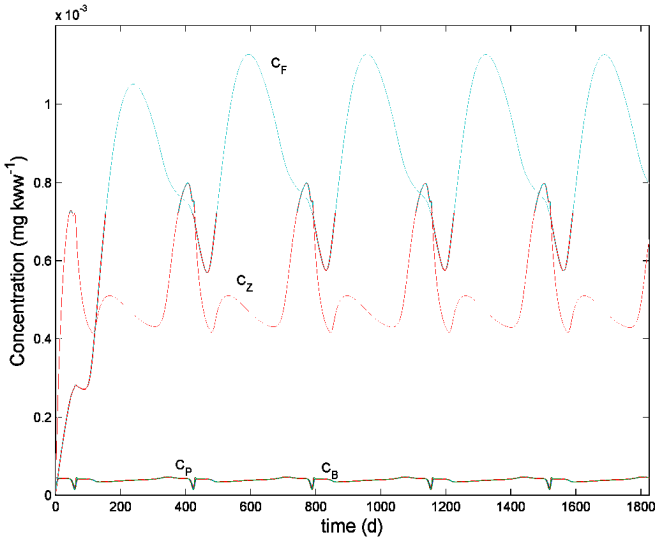



Figure 7. Concentrations of the contaminant in the different ecological compartments: F (fish),Z (zooplankton), P (Phytoplankton), B (Bacteria).

# 3. RESULTS AND DISCUSSION

## 3.1. INFLUENCE OF TROPHIC LEVEL ON BIOACCUMULATION POTENTIAL

Table 4 summarizes the values calculated for the Trophic level as reported in Section 2, whereas Fig. 8 shows the values for the common species in all four ecosystems. As it can be observed there are differences even in close ecosystems like the three sub-alpine lakes. These differences do not take into account diet preferences since we have calculated the species TL based on its position on the food web and we have assigned no diet preferences since there were no enough data for this calculation.

Table 4. Trophic level calculated according to network structure for the selected ecosystems.

| Species | Varese Lake | Monate Lake | Annone Lake | Ria Formosa lagoon |
|---|---|---|---|---|
| Terrestrial detritus | 1 | 1 | 1 | 1 |
| Bacteria (land) | 2 | 2 | 2 | 2 |
| Plankton detritus | 1 | 1 | 1 | 1 |
| Benthic detritus | 1 | 1 | 1 | 1 |
| Bacteria (water) | 2 | 2 | 2 | 2 |
| Benthic Algae | 1 | 1 | 1 | 1 |
| Phytoplankton | 1 | 1 | 1 | 1 |
| Terrestrial plants | 1 | 1 | 1 | 1 |
| Aquatic plants | 1 | 1 | 1 | |
| Macroalgae | | | | 1 |
| Fanerogams | | | | 1 |
| Epiphites | | | | 1 |
| Microzoo plankton | 2.33 | 2.33333 | 2.33333 | 2.33333 |
| Macro-mesozoo plankton | 2.58333 | 2.58333 | 2.58333 | 2.58333 |
| Anellidae | 2.5 | 2.5 | 2.5 | 2.5 |
| Arachinda | 3.45833 | 3.45833 | 3.45833 | 3.45833 |
| Insects large | 2.58333 | 2.58333 | 2.58333 | 2.58333 |
| Insects small | 2.33333 | 2.33333 | 2.33333 | 2.33333 |
| Molluscs gastropoda | 2.5 | 2.5 | 2.5 | 2.5 |
| *Calidris minuta* | | | | 3.3499 |
| *Calidris alpina* | | | | 3.23889 |
| *Charadrius alexandrinus"* | | | | 3.56225 |
| *Aythya fuligula* | | | | 3.67704 |
| *Cyanopica cyanus* | | | | 3.90293 |
| *Anas clypeata* | | | | 3.19434 |
| *Larus ridibundus* | | | | 3.74145 |
| Birds omniv. | | | | 3.56106 |
| Birds pred. 1 | | | | 3.64962 |
| Birds pred. 2 | | | | 3.92972 |
| Clamator glandarius | | | | 3.957 |
| Birds pred. 3 | | | | 4.36904 |
| Birds piscivores | | | | 4.12865 |
| *Anas penelope* | | | | 2.5 |
| *Accipiter nisus* | 4.86313 | 4.93182 | 4.802 | |
| *Acrocephalus arundinaceus* | 4.24186 | 4.23179 | 4.22826 | 3.93544 |
| *Alauda arvensis* | 3.39583 | 3.39583 | 3.39583 | |
| *Alcedo atthis* | 4.55877 | 4.54957 | 4.38341 | 4.12217 |
| Ardea purpurea | 4.59479 | 4.57906 | 4.37725 | 4.19775 |
| Ardeidae 1 | 4.0709 | 4.15992 | 3.92779 | 3.79733 |
| Ardeidae 2 | 4.65225 | 4.64957 | 4.43532 | 4.28371 |
| *Asio flammeus* | 4.82646 | 4.79754 | 4.73191 | |
| Birds carniv./insectiv. | 4.63107 | 4.64642 | 4.59115 | |

| | | | | |
|---|---|---|---|---|
| Birds herb/graniv. | 2.33333 | 2.33333 | 2.33333 | |
| Birds insectiv./graniv. 1 | 3.39583 | 3.39583 | 3.39583 | 3.39583 |
| Birds insectiv./graniv. 2 | 3.10417 | 3.10417 | 3.10417 | |
| Birds insectiv./graniv. 3 | 3.35522 | 3.64453 | 3.07963 | 3.31363 |
| Birds insectiv. | 3.675 | 3.675 | 3.675 | 3.675 |
| Birds invertiv. 1 | 3.71507 | 3.85595 | 3.58393 | 3.54648 |
| Birds invertiv. 2 | 3.76462 | 3.96657 | 3.58393 | 3.64036 |
| *Botaurus stellaris* | 4.37423 | 4.34973 | 4.3301 | |
| *Bubo bubo* | 4.9859 | 5.02741 | 4.91513 | |
| *Chlidonias niger* | 4.04368 | 4.13954 | 3.87104 | 3.72314 |
| *Ciconiidae* | 4.60603 | 4.59204 | 4.35117 | 4.17605 |
| *Cinclus cinclus* | 3.79509 | 4.05813 | 3.64394 | |
| *Circaetus gallicus* | 4.82071 | 4.83661 | 4.77764 | |
| *Circus aeruginosus* | 4.89928 | 4.93574 | 4.82389 | 4.65783 |
| *Circus cyaneus* | 4.83708 | 4.8676 | 4.77433 | 4.51896 |
| *Circus pygargus* | 4.69902 | 4.73121 | 4.64969 | 4.16262 |
| *Columba oenas* | 2.5 | 2.5 | 2.5 | |
| *Dryocopus martius* | 3.10417 | 3.10417 | 3.10417 | |
| *Egretta alba* | 4.73992 | 4.72657 | 4.4725 | |
| *Falco peregrinus* | 4.67671 | 4.70528 | 4.61373 | 4.50349 |
| *Falco subbuteo* | 4.63597 | 4.68338 | 4.60035 | |
| *Falco tinnunculus* | 4.40181 | 4.39378 | 4.36198 | |
| *Gallinula chloropus* | 3.57652 | 3.78849 | 3.39497 | |
| *Glaucidium passerinum* | 4.5854 | 4.64118 | 4.63758 | |
| *Grus grus* | 4.32582 | 4.32591 | 4.2641 | |
| *Haliaeetus albicilla* | 4.8918 | 4.82633 | 4.53206 | |
| *Ixobrychus minutus* | 3.82157 | 4.07665 | 3.65833 | 3.72368 |
| *Lanius collurio* | 4.46336 | 4.49618 | 4.4387 | |
| *Limosa limosa* | 3.36111 | 3.29167 | 3.4213 | 3.26667 |
| *Mergus merganser* | 4.50591 | 4.48541 | 4.06694 | |
| *Milvus migrans* | 4.11951 | 4.45922 | 4.03333 | 4.59293 |
| *Milvus milvus* | 4.40689 | 4.41004 | 4.32709 | |
| *Pandion haliaetus* | 4.89815 | 4.90429 | 4.69708 | 4.60097 |
| *Pernis apivorus* | 4.5552 | 4.57717 | 4.49516 | |
| *Phalacrocorax carbo* | 4.58279 | 4.55665 | 4.26335 | 4.12217 |
| *Phalaropus lobatus* | 3.39167 | 3.5 | 3.59259 | |
| *Podiceps cristatus* | 3.92126 | 4.02691 | 3.84059 | |
| *Rallus aquaticus* | 4.10553 | 4.26735 | 3.93155 | |
| *Strix aluco* | 4.34546 | 4.3385 | 4.31094 | |
| *Lacertidae insectiv.* | | | | 3.79167 |
| *Hemidactylus turcicus* | | | | 3.675 |
| *Lacertidae omniv.* | | | | 3.85521 |
| *Mauremys leprosa* | | | | 3.6975 |
| *Chamaeleo chamaeleon* | | | | 4.69397 |
| *Elaphe scalaris* | | | | 4.5893 |
| *Coronella austriaca* | 5.23926 | 5.26614 | 5.1569 | |
| *Emys orbicularis* | 3.90738 | 3.97371 | 3.80138 | 3.67704 |
| *Lacerta agilis* | 3.79167 | 3.79167 | 3.79167 | |
| *Lacertidae* | 3.675 | 3.675 | 3.675 | |
| *Natrix natrix* | 4.68839 | 4.69274 | 4.64151 | |
| *Natrix tessellata* | 4.64154 | 4.62471 | 4.56471 | |
| *Vipera berus* | 4.40689 | 4.41004 | 4.37304 | |
| *Bufo calamita* | | | | 3.54648 |
| Other amphibians | | | | 3.60833 |
| *Bombina variegata* | 3.60216 | 3.83648 | 3.62438 | |
| *Bufo bufo* | 3.24479 | 3.24479 | 3.24479 | 2.9537 |
| *Hyla arborea* | 3.42262 | 3.42262 | 3.42262 | |
| *Hyla intermedia* | 3.69167 | 3.69167 | 3.69167 | |
| *Rana arvalis* | 3.26569 | 3.4022 | 3.24246 | |

| | | | | |
|---|---|---|---|---|
| *Rana dalmatina* | 3.69167 | 3.69167 | 3.69167 | |
| *Rana temporaria* | 3.70409 | 3.83648 | 3.675 | |
| *Ranidae* | 3.71285 | 2.90774 | 3.6441 | |
| *Triturus cristatus* | 3.67557 | 3.83648 | 3.58389 | |
| *Genetta genetta* | | | | 4.30929 |
| *Herpestes ichneumon* | | | | 4.2807 |
| *Microtus cabrerae* | | | | 2.97222 |
| *Suncus etruscus* | | | | 3.33333 |
| *Felis silvestris* | | | | 4.36385 |
| *Castor fiber* | 2.33333 | 2.33333 | 2.33333 | |
| *Erinaceus europaeus* | 4.42331 | 4.4419 | 4.38754 | |
| *Gliridae* | 4.15824 | 4.19106 | 4.13358 | |
| *Lutra lutra* | 4.57927 | 4.59272 | 4.07881 | 4.28118 |
| *Martes foina* | 4.4365 | 4.45751 | 4.39606 | |
| *Mustela nivalis* | 4.35163 | 4.37632 | 4.29926 | |
| *Mustela putorius* | 4.37658 | 4.38975 | 4.34585 | 4.12791 |
| *Neomys anomalus* | 4.16809 | 4.29797 | 4.03763 | |
| *Neomys fodiens* | 4.30325 | 4.37089 | 4.01478 | |
| *Sciurus vulgaris* | 2.5 | 2.5 | 2.5 | |
| *Vespertilionidae* 1 | 3.79167 | 3.79167 | 3.79167 | |
| *Vespertilionidae* 2 | 4.02083 | 4.02083 | 4.02083 | |
| Macroinvertebrates scavengers | | | | 2.5 |
| Crustaceans scraper/shredder | | | | 2.25 |
| *Polychaetes* and *Hydrobia sp.* | | | | 2.2 |
| *Ruditapes decussatus* | | | | 2.33333 |
| *Tipulidae* (larvae) | | | | 2.33333 |
| *Microdeutopus* sp. | | | | 2.5 |
| *Rissoa membranacea* | | | | 2.33333 |
| *Crabs* sp. | | | | 3.46035 |
| *Hippolyte inermis* | | | | 2.25 |
| *Palaemon serratus* | | | | 2.92143 |
| Crustaceans ominv/pred | | | | 3.22161 |
| *Picnogonidae* | | | | 2.73333 |
| Crustaceans pred | | | | 3.56215 |
| *Bivalvia* | 2.33333 | 2.33333 | 2.33333 | |
| *Ceratopogonidae* | | | 2.94444 | |
| *Chaoborus flavicans* | 2.92778 | | | |
| *Chironomidae* | 2.2 | 2.41667 | 2.25 | 2.33333 |
| *Cyathura carinata* | | 2.5 | | |
| *Coleoptera* | | | 2.33333 | |
| *Erpobdella octoculata"* | | 2.80556 | | |
| *Gastropoda* | | | 2.46667 | 2.5 |
| *Hydracarina* | | 3.33333 | 3.47222 | |
| *Nematoda* | 2.5 | | | |
| *Odonata* | 3.79629 | | | |
| *Oligochaeta* | 2.25 | 2.25 | 2.25 | |
| *Orconectes limosus* | 3.30976 | 3.67786 | | |
| *Phryganea* | | 2.7381 | | |
| *Planorbis carinatus* | | 2.5 | | |
| *Procambarus clarkii* | | 3.69335 | | |
| *Sialidae* | | 3.75 | | |
| *Trichoptera* | | | 2.5 | |
| *Turbellaria* | 2.81667 | | 2.25 | |
| *Zigoptera* | | 4.26429 | | |
| *Cobitis taenia* | 2.33333 | 2.33333 | 2.33333 | |
| *Alburnus alburnus* | 3.03098 | 3.29779 | 3.10729 | |
| *Ameiurus melas* | 3.96007 | 4.07445 | 3.68836 | |
| *Anguilla anguilla* | 4.39735 | 4.38149 | 4.08082 | 4.05509 |
| *Carassius sp.* | 3.87685 | | 3.59259 | |

| | | | | |
|---|---|---|---|---|
| *Coregonus lavaretus* | 3.45267 | 3.60024 | | |
| *Cyprinus carpio* | 3.87518 | 3.31265 | 3.64684 | |
| *Esox lucius* | 4.59434 | 4.53922 | 4.33103 | |
| *Gambusia holbrooki* | | 4.05952 | | |
| *Lampreys and carps* | 3.74846 | | | |
| *Lepomis gibbosus* | 3.48328 | 3.73564 | 3.44022 | |
| *Leuciscus cephalus* | 4.15508 | 4.16362 | | |
| *Lota lota* | 4.56303 | 4.56132 | | |
| *Micropterus salmoides* | 4.32404 | 4.31852 | 4.11425 | |
| *Padogobius martensii* | 3.74846 | 3.96905 | 3.53333 | |
| *Perca fluviatilis* | 4.00036 | 4.18793 | 3.73539 | |
| *Rutilus erythrophtalmus* | 3.27675 | 3.47496 | 3.23333 | |
| *Salmo trutta* | 4.39735 | 4.4172 | | |
| *Sander lucioperca* | 4.57721 | | | |
| *Scardinius erythrophthalmus* | 3.41406 | 3.22794 | 3.30337 | |
| *Silurus glanis* | 4.75149 | | | |
| *Tinca tinca* | 3.95992 | 3.6987 | 3.66645 | |
| *Sarpa salpa* | | | | 2 |
| Fish omniv-detrit 1 | | | | 3.28645 |
| Fish omniv-detrit 2 | | | | 3.28645 |
| *Dicentrarchus labrax* | | | | 3.78379 |
| *Rutilus alburnoides* | | | | 3.28147 |
| *Diplodus sargus* | | | | 3.5379 |
| Fish omniv. 1 | | | | 3.42679 |
| Fish omniv. 2 | | | | 3.45189 |
| Fish omniv-pred 1 | | | | 3.28411 |
| Fish omniv-pred 2 | | | | 3.69631 |
| *Scorpaena porcus* | | | | 3.77865 |
| Fish invertivourus 1" | | | | 3.62597 |
| Fish invertivourus 2 | | | | 3.62597 |
| Fish invertivourus 3 | | | | 3.62597 |
| Fish pred. 1 | | | | 3.72602 |
| Fish pred. 2 | | | | 2.98442 |
| *Conger conger* | | | | 4.0845 |
| *Belone belone* | | | | 4.40014 |

It is clear that the structure of the generated food web depends on the quality of the dataset employed, but this is a common problem in the generation of food web networks. However, with this approach we can have a better idea of the trophic levels, than using the simplified scheme proposed by the TDG (EC, 2003) for freshwater and marine ecosystems. In our case, it seems that there are similar differences, in terms of TLs, between freshwater and coastal ecosystem for the common species.

In this case the maximum difference between calculated trophic levels for the four ecosystem is around 0.6-0.5 (0.51 European Otter, *Lutra lutra*; 0.56 Birds insectiv./graniv. 3; 0.56 Black Kite, *Milvus migrans*; 0.57 Montagu's harrier, *Circus pyragus*). Assuming a linear correlation similar to Eq. (7) between the TL and the concentration of chemicals this would imply that there will be concentrations differences around half an order of magnitude between the same species at several ecosystems. Of course, in this case we assume that the diet of each species is the same at each ecosystem which is normally not the case (Keough et al., 1996).

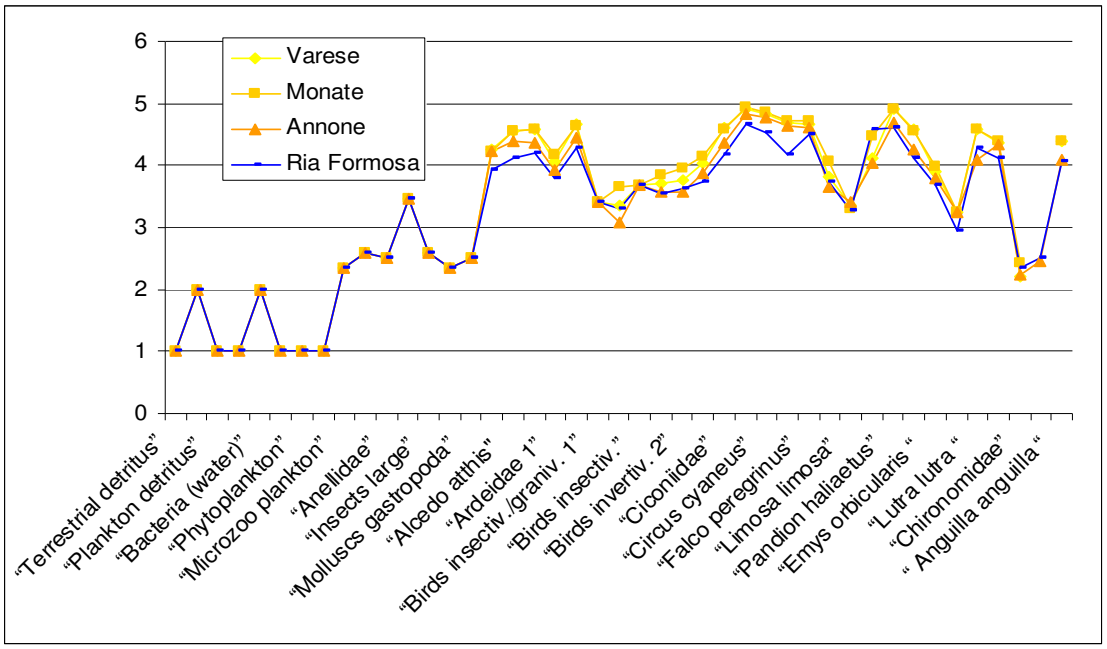



Figure 8. Trophic levels for the common species in all four ecosystems.

## 3.2. INFLUENCE OF THE DIET ON BIOACCUMULATION POTENTIAL

To study not only the influences of the food web structure on the bioaccumulation potential, but also to consider the diet, we have used the simplified ecological plus bioaccumulation model developed in Section 2. In this case we have performed simulations considering different chemicals and two different food preferences. In the first case, we consider that zooplankton feeds mostly on phytoplankton ($Y_{P,0}$ =0.85, $Y_{B,0}$ =0.14, $Y_{D,0}$ =0.01) and fish mostly on zooplankton ($X_{Z,0}$=0.98, $X_{P,0}$=0.01, $X_{D,0}$=0.01), whereas in the second case, we assume that zooplankton feeds on detritus ($Y_{P,0}$ =0.10, $Y_{B,0}$ =0.10, $Y_{D,0}$ =0.80) and fish on phytoplankton ($X_{Z,0}$=0.10, $X_{P,0}$=0.80, $X_{D,0}$=0.10). With this approach we tend to increase the TL of fish in the first case and decrease in the second, since a complete diet on zooplankton would imply a TL =3 whereas a complete diet on phytoplankton would produce a TL =2. However, it is necessary to consider that these are food preferences, but that the population adapts depending on food availability. This approach gives stability to the system decreasing the probability of species extinction.

As a consequence of the differences in diet preferences, the dynamics of the ecosystem changes for the two cases. Figures 9 and 10 show the last year after 10 years of simulation starting from the same initial conditions but applying the two types of food preferences.

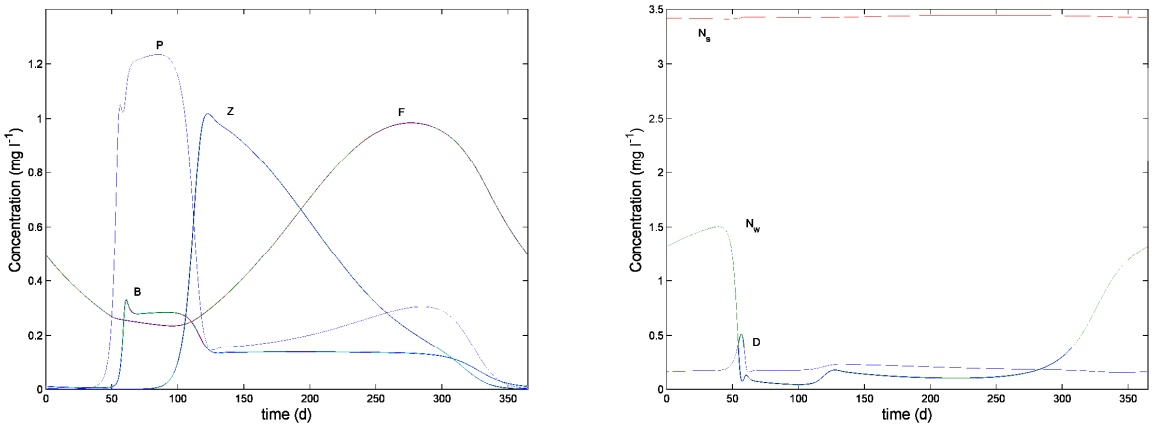



Figure 9. Ecological system with food preferences that increase TL.

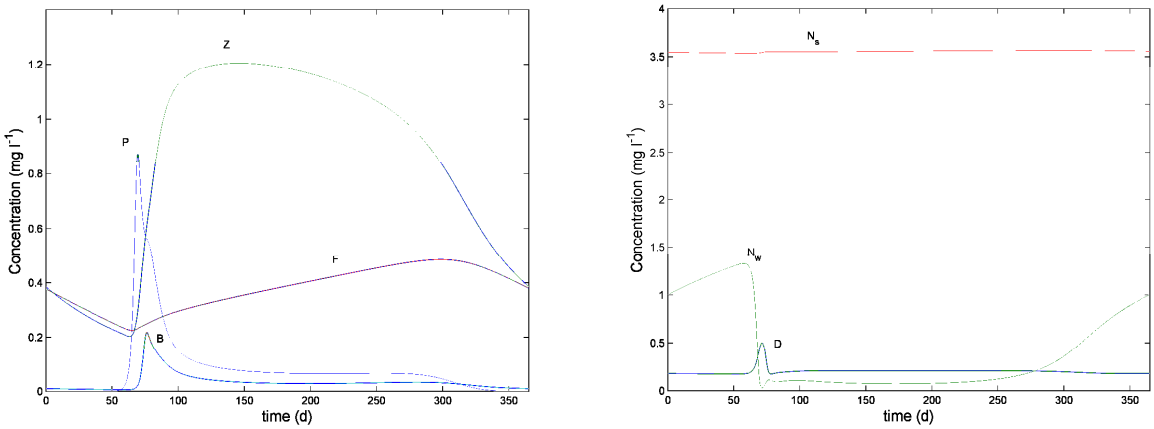



Figure 10. Ecological system with food preferences that decrease TL.

The concentrations in the different compartments of the ecological system are shown in Fig. 11. Surprisingly, fish and zooplankton reach higher concentrations in the case of lower TL. This is due to the partitioning of the chemical. In fact, practically the chemical is on the particulate phase (detritus) and then feeding on detritus provides higher fluxes of contaminant than in the case of the standard food web, i.e. phytoplankton→zooplankton →fish. This aspect is another element that should be assessed for hydrophobic chemicals which tend to attach to the particulate phase. In addition, concentrations are changing with time and therefore the bioaccumulation will also vary during the year. The fact that particulate organic matter (detritus in the model) plays an important role in the bioaccumulation of hydrophobic chemicals should be carefully considered. In our model, we do not have included the role of the sediments and benthic species which would recycle in the ecosystem the contaminants reach the sediments by settling.

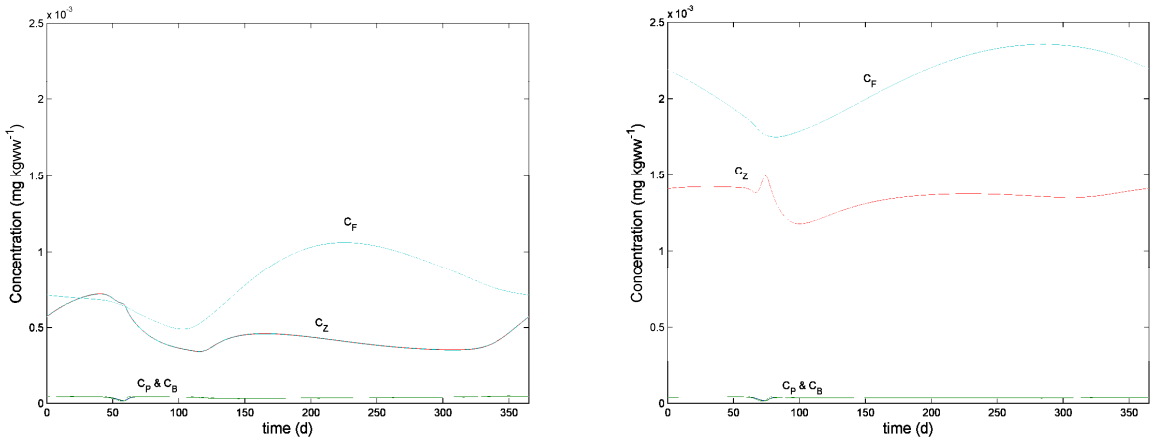



Figure 11. Species concentrations for the two food preferences.

To compare the influence of diet on bioaccumulation potential, we have run the model for the list of chemicals in Table 3 for the two different diets and calculated and average standard deviation concerning the $BCF_{fish}$, i.e. $BCF_{fish}=C_F/C_w$. Table 5 summarizes the results obtained; as it can be observed there are important differences between PAHs, and PCBs and PCDD/Fs for similar values of log $K_{ow}$, because for the first family of compounds we consider metabolism. In addition, also for PAHs, due to metabolism, the differences are smaller than for PCBs and PCDD/Fs that can be around half an order of magnitude, but if we consider the fluctuations around the year due to several effects like temperature, particulate organic matter, etc., then they can easily reach one order of magnitude in the calculation of BCF.

Figure 12 compares the simulated results with know *BCF* correlations in literature. As it can be observed, the simulated results are in good agreement with correlations developed for PCBs and PCDD/Fs, but disagree with the correlations developed by Arnot and Gobas (2006). In addition, the values for PAHs in which we consider metabolism are always lower than existing correlations. This points out an important factor that should be considered when developing EQS for biota. If in the food web structure there are species able to metabolize the compound the *BC*F will tend to decrease when compared with another system in which there is no species able to perform this function. This calls for an integrated evaluation of the aquatic ecosystem.

Table 5. Calculated annual averages of fish bioconcentration factors (l $kg^{-1}$)for the two diets and for the chemical families considered in the model (Notice that metabolism is considered for the PAHs).

| **Compound** | ***log $K_{ow}$*** | **Diet 1** | **Diet 2** |
|---|---|---|---|
| **PAHs** | | ***$BCF_{fish}$*** | ***$BCF_{fish}$*** |
| Naphthalene | 3.37 | 0.41 | 0.44 |
| Fluorene | 4.12 | 1.07 | 1.33 |
| Antracene | 4.54 | 2.42 | 3.17 |
| Phenanthrene | 4.57 | 2.58 | 3.38 |
| Pyrene | 5.17 | 9.61 | 12.99 |

| | | | |
|---|---|---|---|
| Fluoranthene | 5.22 | 11.32 | 14.63 |
| Benzo[a]anthrecene | 5.84 | 44.75 | 60.23 |
| Chrysene | 5.84 | 44.75 | 60.23 |
| Benzo [a]pyrene | 6.04 | 71.10 | 94.64 |
| Benzo[b]fluoranthene | 6.44 | 178.70 | 241.38 |
| Benzo[k]fluoranthene | 6.44 | 178.70 | 241.38 |
| Indeno[1,2,3-cd]pyrene | 6.58 | 241.81 | 332.27 |
| **PCBs** | | | |
| PCB28 | 5.67 | $5.02 \cdot 10^3$ | $1.56 \cdot 10^4$ |
| PCB52 | 5.80 | $6.81 \cdot 10^3$ | $2.07 \cdot 10^4$ |
| PCB101 | 6.40 | $2.70 \cdot 10^4$ | $8.30 \cdot 10^4$ |
| PCB118 | 6.70 | $4.98 \cdot 10^4$ | $1.65 \cdot 10^5$ |
| PCB138 | 6.83 | $6.58 \cdot 10^4$ | $2.25 \cdot 10^5$ |
| PCB153 | 6.92 | $8.05 \cdot 10^4$ | $2.78 \cdot 10^5$ |
| PCB180 | 7.40 | $2.35 \cdot 10^5$ | $8.40 \cdot 10^5$ |
| **PCDD/Fs** | | | |
| TCDD | 6.9 | $7.91 \cdot 10^4$ | $2.67 \cdot 10^5$ |
| PeCDD | 7.4 | $2.35 \cdot 10^5$ | $8.40 \cdot 10^5$ |
| HxCDD | 7.8 | $5.80 \cdot 10^5$ | $2.10 \cdot 10^6$ |
| HpCDD | 8.0 | $9.14 \cdot 10^5$ | $3.27 \cdot 10^6$ |
| OCDD | 8.2 | $1.48 \cdot 10^6$ | $5.29 \cdot 10^6$ |
| TCDF | 7.7 | $4.51 \cdot 10^5$ | $1.64 \cdot 10^6$ |
| PeCDF | 7.6 | $3.65 \cdot 10^5$ | $1.30 \cdot 10^6$ |
| HxCDF | 7.7 | $4.51 \cdot 10^5$ | $1.64 \cdot 10^6$ |
| HpCDF | 7.5 | $2.91 \cdot 10^5$ | $1.05 \cdot 10^6$ |
| OCDF | 7.6 | $3.65 \cdot 10^5$ | $1.30 \cdot 10^6$ |

# 4. CONCLUSIONS

To analyze the effects of ecosystem food web and trophic level on the bioaccumulation-biomagnification potential, we have studied four terrestrial and aquatic trophic network models generated previously for three similar lakes and a costal lagoon. The results point out those even small differences in trophic level, could provoke variations in the concentrations of hydrophobic compounds and that these effects should be taken into account when developing EU wide EQS values for biota. In addition, they call for the development of better methods and tools for analyzing the ecosystem structure to be able to perform intercomparison exercises between different ecosystems.

The development of a food web is the first step for comparing ecosystems, but diet preferences need also to be evaluated since they would modify the TLs values. For this reason, we have developed a simplified food web model and assessed the effects that different diets may have on the bioaccumulation potential. Also in this case we have found that differences in *BCF* are possible. In addition, this simple model points out also the influence of other parameters such as temperature, organic matter and seasonal variations in the food web on the *BCF* value.

Both analysis suggest that a greater understanding on the food web structure and the relative contribution of the linkages between species is necessary for tailoring the developed Environmental Quality Standards to a particular ecosystem, since there is the possibility, that the same concentration of a hydrophobic and bioaccumulative chemical in the water column, could give between one or two

orders of magnitude differences on the concentrations of the same top or pelagic predator when compared to even a similar ecosystem.

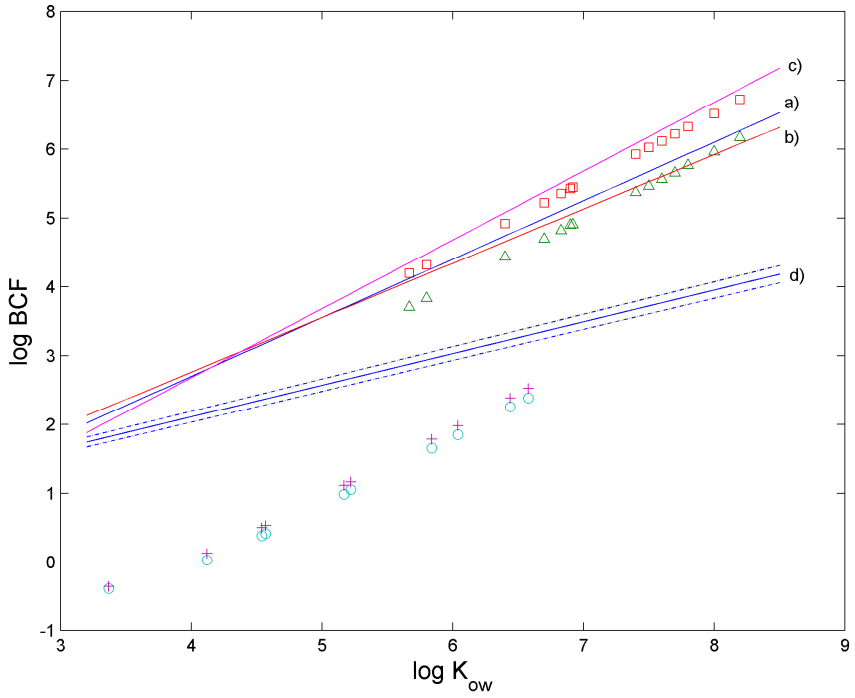



Figure 12. Comparison between simulated BCFs (squares and triangles correspond to PCBs and PCDD/Fs whereas sign plus and circles correspond to PAHs –considering metabolism- for Diets 2 and 1, respectively) and several correlations for BCF for fish: a) log $BCF$=0.85·log$K_{ow}$ (Veith et al. and EC 2003); b) log $BCF$=0.79·log$K_{ow}$-0.40 (Veith and Kosian,1983); c)log$BCF$=log$K_{ow}$-1.32 (Mackay, 1982); d)log $BCF$=0.27(0.04)+0.46(0.01)·log$K_{ow}$ (Arnot and Gobas, 2006).

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**Abstract.** With the implementation of the Water Framework Directive and the daughter directive on Environmental Quality Standards (EQS), the development of EQS values for biota has become an important issue. However, for certain hydrophobic compounds, bioaccumulation and biomagnification in the food chain may occur. In this case, it is important to analyze, if depending on the food web and diet preferences, bioaccumulation values could be different for the same species in different ecosystems. To analyze this effect, we have studied four terrestrial and aquatic trophic network models generated previously for three similar lakes and a costal lagoon. In addition a simple ecological plus bioaccumulation model has been developed for analyzing diet preferences which could not be assessed using the developed trophic food webs. The objective was to assess if the similarities and differences between calculated trophic level could have consequences on the bioaccumulation and biomagnification potential of contaminants. The results indicate that differences of around two orders of magnitude can be expected for similar ecosystems. These results are important since they indicate that the ecosystem structure should be first assessed for the development of EQS for biota and that without this information intercomparison exercises between EU ecosystems are not possible.

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