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# Comparing measured and predicted PCB concentrations in Arctic seawater and marine biota

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## Abstract

When a mechanistic food web model was parameterized for the Arctic marine ecosystem, it predicted PCB concentrations in zooplankton and fish that were two orders of magnitude lower than measured. PCB concentrations measured in zooplankton and fish were within the laboratory's accredited quality assured criteria, and were comparable to other Arctic regions. Although on a different scale, the predicted and measured PCB concentrations were highly correlated. As sensitivity analyses indicated water concentrations as the most sensitive parameter for the model output, dissolved water concentrations were predicted using measured zooplankton and air PCB concentrations, and empirical and mechanistic models. The food web model and the empirical relationship between bioaccumulation factor and octanol–water partitioning coefficient predicted mean dissolved water concentrations of 28 and 29 pg/L  $\sum$ PCB<sub>6</sub> (PCB-28, -52, -105, -118, -138, -153), respectively. Mean dissolved water concentration predicted from measured air concentrations in 1996 was 7.6 pg/L  $\sum$ PCB<sub>5</sub> (PCB-28 was not analysed). Mean dissolved water concentration measured in Barents Sea water sampled simultaneously as the biota in 1999 was 0.3 pg/L  $\sum$ PCB<sub>6</sub>. The dissolved water concentrations predicted from zooplankton PCB concentrations were comparable to water concentrations measured in 1996, whereas the dissolved water concentrations measured in 1999 were comparable to measurements from 2001. If the present high empirically derived bioaccumulation factors (log BAF 7.3–9.0) were realistic, this suggests that bioaccumulation in Arctic zooplankton is more efficient than previously assumed. The present study illustrates and discusses some of the difficulties encountered when different approaches to study environmental distribution of contaminants are compared.

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## 1. Introduction

A number of different approaches are used to study the fate of anthropogenic chemicals in the environment. The main purpose is to understand contaminant concentrations and fluxes in a variety of environ-

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mental scenarios. Traditionally, monitoring tools, such as measurements of chemical concentrations, are employed to verify the level of contaminants in a given environmental compartment (air, water, soil, sediment, biota). Comparison of chemical concentrations measured in “coupled” compartments, such as relatively hydrophobic compounds (e.g. polychlorinated biphenyls, PCBs) in water and suspended sediment, by the use of partition coefficients such as the organic carbon–water partitioning coefficient ( $K_{OC}$ ), allows verifying the consistency of analytical data. The comparison assumes that the concentrations are in equilibrium, and that they are reflected by the partition coefficient.

Another approach to reconstruct contaminant distribution in the environment is the use of multimedia models. These are mathematical tools relating physicochemical properties of a chemical to its partition and transport in the environment. Models can be purely empirical, which correlate measured concentrations to descriptors (such as  $K_{OC}$ ), or mechanistic, which generally implement a mass balance approach in which the various transfer processes between compartments are quantified (Gobas and Morrison, 2000; Mackay and Fraser, 2000). Among the empirical and mechanistic models available, recent emphasis has been on bioaccumulation models that predict enrichment of chemicals in biota due to exposure from respiration and diet. An advantage of mechanistic models is that they quantify different processes of varying importance for bioaccumulation, such as respiration and feeding rates, growth dilution and biotransformation. A disadvantage of mechanistic models is the higher demand for parameterization. Mechanistic models are often based upon empirical relationships (such as allometric relationships) that are used to extrapolate from one species (or chemical or compartment) to the one under study. The increased need for environmental information and extrapolation between systems and species might lead to increased uncertainty in terms of model predictability.

A number of aquatic bioaccumulation models have been developed (see review by Gobas and Morrison, 2000). Many of these models are available in steady-state, assuming that the uptake and loss processes of a contaminant by a single organism or in the food web are constant over time. Comparison of a thermodynamic model (Connolly and Pedersen, 1988) and a

steady-state model (Campfens and Mackay, 1997) for a pelagic food chain yielded different contaminant behaviour, of which the steady-state model corresponded best with the field data (Binelli and Provini, 2003). Although being developed and evaluated for temperate lakes (e.g. Campfens and Mackay, 1997; Arcuri, 1998), few attempts have been made to employ these steady-state food web bioaccumulation models to other aquatic systems, such as the Arctic marine ecosystem.

Persistent organic pollutants (POPs), which have been heavily used globally in agriculture and industry, are found in the Arctic predominantly due to long-range transport from areas of use with the atmosphere, oceanic currents, and large rivers (de March et al., 1998). In addition, settlements, harbours and military sites within the Arctic contribute to local contamination, as demonstrated by locally elevated POP concentrations (de March et al., 1998; de Wit et al., 2004). Despite its remoteness and difficult logistics, the Arctic marine ecosystem has been extensively studied during the late 1990s, resulting in detailed species-specific measurements of food web distribution of organic contaminants (e.g. Borgå et al., 2001; Fisk et al., 2001a, Hoekstra et al., 2003; Muir et al., 2003). Within the Arctic, the Barents Sea food web is important in energy transfer due to local high production prompted by front systems of converging Atlantic and Polar water, and by an extensive marginal ice zone (Sakshaug et al., 1992). Lipid-rich zooplankton are attractive prey for fish, seabirds, seals and whales that migrate to the Barents Sea to feed, and some of the largest fisheries in the northern-European region are located in this area. As costly POP data are now available for the Arctic marine ecosystem, it is of interest to adapt a bioaccumulation food web model to predict the flux of contaminants in the Arctic marine ecosystem.

The initial objective of the present study was to apply a mechanistic steady-state food web bioaccumulation model (Campfens and Mackay, 1997) to predict PCBs concentrations in Arctic marine zooplankton and fish, and to identify explanations for possible discrepancy between predicted and measured PCB concentrations in zooplankton and fish. A following objective was to use empirical and mechanistic models to predict the dissolved water POP concentrations based on the measured PCB and

chlorinated pesticide concentrations in biota and/or in air.

## 2. Materials, methods and models

### 2.1. Zooplankton and fish collection and PCB quantification

Zooplankton and fish were collected in the Barents Sea marginal ice zone (Fig. 1) in May 1999 with net hauls or benthic trawls, respectively (Borgå et al., 2003; in preparation). The zooplankton included species- and stage (or size)-specific samples of herbivorous copepods and euphausiids (*Calanus glacialis* ( $n=12$ ) and *C. hyperboreus* ( $n=3$ ), and *Thysanoessa inermis* ( $n=9$ ), respectively), and predatory amphipods (*Themisto libellula* ( $n=3$ )). The fish were benthic-pelagic polar cod (*Boreogadus saida* ( $n=12$ )), collected at about 300 m depth and were in the size range of 120–182 mm (2–5 years, Falk-Petersen et al., 1986). The samples of copepods were 580–1135 pooled individuals, depending on species and stage, of euphausiids were 100–115 pooled individuals and amphipods were 7–16 pooled individuals. Polar cod were collected and analysed whole, individually.

PCBs in zooplankton and fish were analysed at the Environmental Toxicology Laboratory at the Norwegian School of Veterinary Science, Oslo, Norway, by high resolution gas chromatography with electron capture detector as described in details elsewhere (Borgå et al., 2002a,b). All reported concentrations were quantified within the accredited quality assured requirements (Borgå et al., 2003).

### 2.2. Mechanistic mass-balance food web model

The Campfens and Mackay (1997) bioaccumulation food web model (hereafter called FWM), originally available as a DOS-based GWBASIC program, was initially integrated in Visual Basic (Arcuri, 1998). The latter version was updated in the present study to better implement new scenarios and to perform sensitivity analysis. A comparison of runs between the original GWBASIC version and the current Visual Basic version was performed on selected scenarios to verify the results. The FWM is

a fugacity-based model (Mackay, 2001), based on steady-state assumptions (constant input or process rates with time), and was originally developed for temperate regions and tested for PCB in a Lake Ontario food web (data set from Oliver and Niimi, 1988). The propagation of uncertainty in the FWM has been studied (MacLeod et al., 2002), and the FWM has been modified to calculate bioaccumulation in Arctic seals (Fraser et al., 2002), and food web accumulation of other POPs and temperate lakes (e.g. Arcuri, 1998).

The general mass balance equation to describe accumulation in an organism exposed to contaminated food and water at steady state is:

$$f_W D_W + f_A D_A = f_F (D_W + D_G + D_M + D_E) \quad (1)$$

where  $f$  is the fugacity (in Pa, subscript W for water, A for diet, F for organism) (Campfens and Mackay, 1997). Fugacity is the partial pressure of a chemical in a phase, and at equilibrium a chemical has equal fugacities in connected phases (Mackay, 2001). Multiplied by the fugacity capacity of a phase ( $Z$ ), fugacity gives the concentration ( $C$ ) of a chemical in a given phase ( $C=f \cdot Z$ ) (Mackay, 2001).  $D$  is the transport value (analogous to rate constant, in mol/Pa h) expressing the rates of various uptake and loss processes (subscripts W for respiration, A for dietary uptake, G for growth dilution, M for metabolism, E for egestion). Each transport value  $D$  is solved by algebraic functions (Table 1), and follows the fugacity description. The mass balance above (Eq. (1)) refers to a single organism and must be solved for other organisms and combined using a matrix approach ( $N \times N$ , where  $N$  is the number of species) to relate contaminant distribution among different species throughout a food web (Campfens and Mackay, 1997). More information and details about the model are available in Campfens and Mackay (1997), Sharpe and Mackay (2000) and Mackay (2001).

The FWM was parameterized for the Barents Sea pelagic ecosystem in a spring (May) situation, based on literature data (Table 2). Water was sampled for dissolved and particulate PCBs concentrations (Olsson et al., 2002) simultaneously as the zooplankton and fish (Borgå et al., 2003; in preparation). Water concentrations were assumed to vary 40% to investigate the variation in model predicted biotic PCB concentration. Henry's law constant ( $H'$ , in Pa m<sup>3</sup>/



Fig. 1. Sampling sites for water (W) (Olsson et al., 2002), zooplankton (Z) and fish (F) in the Barents Sea (May) and north of Svalbard (September) in 1999. Numbers refer to station numbers as given in the cruise report (Hop and Falk-Petersen, 2003), and roman numbers to station numbers in Borgå et al. (2002a).

Table 1

(A) Equations in the model used to solve the uptake and loss processes for the different organisms by a matrix solution (reproduced from [Campfens and Mackay, 1997](#)). (B) Fugacity capacity ( $Z$ ) in the different compartments

(A) Transport values $D$ (mol/Pa · h)				
Equation		Parameter definition		
$D_A = E_A G_A Z_A$	$D_A$	Net chemical uptake from diet		
	$G_A$	Gross food ingestion rate (m <sup>3</sup> /h)		
	$E_A$	Gut absorption efficiency		
$D_E = D_A / Q$	$D_E$	Chemical loss by egestion		
	$Q$	Limiting biomagnification factor		
$D_M = V_F Z_F k_M$	$D_M$	Chemical loss by metabolism		
	$V_F$	Volume of organism (m <sup>3</sup> )		
	$k_M$	Metabolic rate constant (1/h)		
$D_G = Z_F (dV_F/dt)$	$D_G$	Growth dilution term, $t$ is time (h)		
$D_W = k_1 V_F Z_W$	$k_1$	Gill ventilation rate constant (1/h)		
(B) Fugacity capacity $Z$ (mol/Pa · m <sup>3</sup> )				
Compartment	$Z$		Definitions	Unit
Water	$Z_W = 1 / (H = C^s / P^s)$	$C^s$	Water solubility (mol/m <sup>3</sup> )	mol/m <sup>3</sup>
		$P^s$	Vapour pressure (Pa)	Pa
		$H$	Henry's law constant	Pa · m <sup>3</sup> /mol
Particles	$Z_S = K_P \rho_S / H$	$K_P$	Particle–water partition coefficient	L/kg
		$\rho_S$	Particle density	kg/L
Lipid	$Z_O = K_{OW} / H$	$K_{OW}$	Octanol–water partition coefficient	–
Organism	$Z_F = K_B \rho_B / (H = L_B Z_O)$	$K_B$	Equilibrium bioconcentration factor	– <sup>a</sup>
		$\rho_B$	Organism density	kg/L
		$L_B$	Lipid fraction of organism	–
Food	$Z_A = L_A Z_O$	$L_A$	Lipid fraction of diet	–

See [Table 2](#) for parameterization.

<sup>a</sup> Assuming biota density of 1 kg/L.

mol) was temperature corrected from those of 25 °C based on the Clausius–Clapeyron equation ([Mackay et al., 2000](#)). Enthalpies of vapourization and solution were assumed to be 50,000 and 10,000 J/mol, respectively, as suggested in the ChemCAN model ([Mackay et al., 1996](#)). A mean water temperature of –1 °C was assumed for the time of investigation, based on measurements during the cruise ([Hop and Falk-Petersen, 2003](#)).

Given that the FWM has many input variables ([Table 2](#)), all collected from literature for the system and species involved or comparable systems and species, some might be associated with uncertainties in measurements in addition to natural variation in populations or within the year. However, the FWM is not equally sensitive to all state and input variables, thus sensitivity analyses were performed for each congener and each species to identify which state and input variables it was most important to reduce the associated uncertainty or variability. The sensitivity analysis was performed by perturbing state variables by varying them one by one, according to the method used by [Burkhard \(1998\)](#).

As few data were available, no assumptions were made on the data distribution, and Spearman's rank order correlation was chosen for comparison of predicted and measured zooplankton and fish PCB concentrations. All biotic POP concentrations are compared adjusted for the organisms' lipid content.

### 2.3. Estimation of dissolved water POP concentrations

The FWM was used to back-calculate the dissolved PCB water concentration, using the measured PCB concentrations in calanoid copepods. Dissolved PCB concentration in water can also be calculated based on bioaccumulation factors (BAF). BAFs describe the PCB concentrations ([PCB]) in an organism relative to that dissolved in water ([Gobas and Morrison, 2000](#)):

$$\text{BAF} = [\text{PCB}]_{\text{organism}} / [\text{PCB}]_{\text{dissolved water}} \quad (2)$$

where  $[\text{PCB}]_{\text{organism}}$  is on a lipid weight basis, and  $[\text{PCB}]_{\text{dissolved water}}$  is the dissolved PCB concentration in water. The dissolved PCB concentration in water can thus be calculated since BAF can be predicted from its relationship with octanol water partitioning coefficient ( $K_{OW}$ ) reported for calanoid copepods or zooplankton in Canadian Arctic areas ([Hargrave et al., 2000](#); [Fisk et al., 2001b](#); [Hoekstra et al., 2002](#)). For *Calanus hyperboreus* from the North Water Polynya in May, the curvilinear relationship between BAF and  $K_{OW}$  ([Hickie et al., 2001](#) based on [Fisk et al., 2001b](#)) was:

$$\log \text{BAF} = -0.513 + 1.7341(\log K_{OW}) - 0.1143(\log K_{OW})^2 \quad (3)$$

By replacing BAF into Eq. (2) and solving with respect to dissolved water concentration predicts the dissolved water concentration needed to obtain the observed PCB concentrations in biota. For both the FWM and the BAF- $K_{OW}$  model, the predicted dissolved water concentrations were based on mean PCB concentrations in copepods *C. hyperboreus* on a lipid weight basis (mean of copepodit stages IV, V and VI).

Variation and uncertainty in predicted dissolved water concentrations were calculated assuming 30% uncertainty in  $K_{OW}$  and 95% confidence interval (CI) of zooplankton PCB concentrations. Thus, to maximize predicted water concentration, high PCB concentration in *C. hyperboreus* (mean+95% CI) and low  $K_{OW}$  (value-30%) was used, whereas the opposite was used to predict minimum dissolved water PCB concentra-

Table 2

Barents Sea parameterization for (A) selected PCBs, (B) particle concentration, (C) organism properties, (D) diet matrix

(A) Physicochemical properties and water concentration							
Congener	Molecular mass (g/mol)	Log $K_{OW}$ (at 25 °C)	Water solubility (mg/L at 25 °C)	Vapour pressure (Pa at 25 °C)	$H$ (Pa · m <sup>3</sup> /mol at 25 °C)	$H$ (Pa · m <sup>3</sup> /mol at -1 °C)	Water concentration (pg/L)
PCB 28	257.5	5.67	0.16	0.026	41.8	8.9	0.18
PCB-31	257.5	5.67	0.17	0.015	22.7	4.8	–
PCB 52	292.0	6.10	0.03	0.0049	47.6	10.2	0.17
PCB-99	326.4	6.39	0.0353	0.00316	29.2	6.3	–
PCB-101	326.4	6.40	0.01	0.00109	35.57	7.6	0.25
PCB-105	326.4	6.65	0.0034	0.00086	84.4	17.6	0.04
PCB-110	326.4	6.48	0.0082	0.00228	90.7	19.4	0.13
PCB-118	326.4	6.74	0.0134	0.00119	29.1	5.9	0.10
PCB-138	360.9	6.73	0.0015	0.0005	120.3	25.7	0.16
PCB-149	360.9	6.67	0.00434	0.00112	95.4	19.6	0.18
PCB-153	360.9	6.90	0.001	0.000119	42.9	9.2	0.17
PCB-180	395.3	7.36	0.00031	0.00013	165.7	35.4	0.03

(B) Particulate matter			
	March	May	June
Total particulate matter, g/m <sup>3</sup>	0.6	4.8	2.2
Density of particles in water column (kg/m <sup>3</sup> )	1500	1500	1500

(C) Organism properties								
Organism	Volume ( $V_F$ ) (cm <sup>3</sup> (=g))	Lipid ( $L_B$ ) (% of BW)	Feeding rate (g/g BW · day)	Growth rate (g /g BW · day)	Resistance		Digestion factor	Metabolism half-lives (day)
					Water	Organic		
<i>Calanus glacialis</i>	0.00238	2.55	0.150	0.2	5.3e-08	3.5	4	50,000
<i>C. hyperboreus</i>	0.012	2.74	0.200	0.19	5.3e-08	4.0	4	50,000
<i>Thysanoessa inermis</i>	0.075	1.85	0.150	0.1	5.3e-08	4.0	4	50,000
<i>Themisto libellula</i>	0.300	1.91	0.015	0.1	5.3e-08	1.5	4	5000
<i>Boreogadus saida</i>	16.22	3.80	0.005	0.04	5.3e-08	1.5	4	5000

(D) Diet matrix					
Prey/Predator	Calanoid copepods		Euphausiids	Amphipods	Fish
	<i>Calanus glacialis</i>	<i>Calanus hyperboreus</i>	<i>Thysanoessa inermis</i>	<i>Themisto libellula</i>	<i>Boreogadus saida</i>
<i>C. glacialis</i>	0	0	0.3	0.25	0.35
<i>C. hyperboreus</i>	0	0	0	0.25	0.35
<i>T. inermis</i>	0	0	0	0.5	0.15
<i>T. libellula</i>	0	0	0	0	0.15
<i>B. saida</i>	0	0	0	0	0

tion. Since PCBs were measured in both zooplankton and water sampled simultaneously from the central Barents Sea (stations 31–34, Fig. 1), the actually bioaccumulation factors (BAF) were calculated as in Eq. (2).

As concentrations of PCB in air ( $[\text{PCB}]_{\text{air}}$ ) are available from the Svalbard/Barents Sea region and Eastern Arctic Ocean from the late 1990s (Harner et al., 1998; de Wit et al., 2004), the dissolved water concentrations were calculated assuming that the two phases were in, or close to, equilibrium:

$$[\text{PCB}]_{\text{dissolved water}} = [\text{PCB}]_{\text{air}}/K_{\text{AW}} \quad (4)$$

where  $K_{\text{AW}}$  is the air–water partitioning coefficient, which is determined by:

$$K_{\text{AW}} = H'/RT \quad (5)$$

where  $H'$  is the Henry's Law constant ( $\text{Pa m}^3/\text{mol}$ ) corrected for temperature as described above,  $R$  is the gas constant ( $8.314 \text{ Pa m}^3/\text{mol}$ ) and  $T$  is the water temperature in Kelvin (Mackay, 2001). Uncertainty in the estimations was investigated assuming 30% measuring variation in air concentrations, and 20% variation in water solubility and vapour pressure at 25 °C. Maximum dissolved

water PCB concentration was thus predicted by assuming high air concentration (measured+30%), high water solubility (value+20%) and low vapour pressure (value-20%), whereas the opposite was used to predict minimum dissolved water PCB concentration.

#### 2.4. Total particulate matter concentration

The amount of PCBs dissolved or associated to particles is determined by the total particulate matter (TPM) in water, its organic carbon content (POC), and the congener's organic carbon–water partitioning coefficient ( $K_{\text{OC}}$ ) (Campfens and Mackay, 1997; Mackay, 2001). As a control of the consistency between measured dissolved and particulate-associated PCB concentrations in water (Olsson et al., 2002), the ratio between measured dissolved and particulate-associated PCB concentrations was obtained using the FWM for each congener at a time by altering the TPM concentration. The two phases were assumed to be in equilibrium, and the POC content in TPM was varied from 5% to 30% to include range of POC content in TPM reported for different depths in an Arctic fjord (Reigstad, 2000).

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#### Notes to Table 2:

(A) Molecular mass, water solubility, vapour pressure at 25 °C: selected values from Mackay et al. (2000) for individual congeners if reported. For congeners and parameters where no value was selected by Mackay et al. (2000), a value was chosen from the reported list to reflect an average value. PCB-105, -118 and -149 were not listed in Mackay et al. (2000), and their respective water solubility and vapour pressure were obtained from Syracuse Research's Physical Properties database (<http://www.esc.syrres.com/interkow/physdemo.htm>).

Log  $K_{\text{OW}}$ : from Hawker and Connell (1988).

$H'$ : Henry's law constant calculated based on the selected water solubility and vapour pressure at 25 °C or temperature adjusted (Eq. (5)).

Water concentrations: Total (dissolved+particulate associated) PCB concentrations (Olsson et al., 2002). Mean value of one measurement from an ice-covered and one from an open-water station.

(B) Total particulate matter (TPM) calculated from particulate organic carbon (POC) concentrations from Olli et al. (2002), assuming that 10% of TPM is POC (Reigstad, 2000). Values for May are used, but March and June are shown to illustrate the variation depending on pre-bloom, bloom, or post-bloom situation. Density of particles in water column from Campfens and Mackay (1997).

(C) Volume: Calculated based on individual body weight (BW), assuming that 1 g=1 cm<sup>3</sup>. Individual weights: Copepods—Scott et al. (2000), Euphausiids and Amphipods—calculated from sample wet weight and number of individuals, Polar cod—present study. Lipid %: Copepods—present study, Euphausiids and Amphipods—Borgå et al. (2003), Polar cod—present study. Feeding rate: Copepods—Halvorsen et al. (2001), for small and large temperate copepods, Euphausiids—predicted from Båmstedt and Karlson (1998) and Dalpadado and Skjoldal (1996), Amphipods—Percy (1993a), Polar cod—Hop and Tonn (1997). Growth rate: Copepods and Euphausiids—Edvardsen et al. (2002), Amphipods—Percy (1993b), Polar cod—Hop et al. (1997). Water and organic resistance used to predict the gastro-intestinal POP uptake efficiency was assumed by analogy to other systems where the model has been used successfully (Lake Ontario, in Campfens and Mackay (1997)). Digestion factor ( $Q$ =limiting biomagnification factor) was set to 4 for all organisms based on Gobas et al. (1993, 1999). Contaminant's half-lives were assumed long as invertebrates and fish have restricted or negligible biotransformation of PCBs (e.g. Moisey et al., 2001).

(D) Diet composition: Copepods—Scott et al. (1999), Euphausiids—Båmstedt and Karlson (1998), Amphipods—Scott et al. (1999), Polar cod—Lønne and Gulliksen (1989).

### 3. Results and discussion

#### 3.1. Predicted biotic PCB concentrations and observed bioaccumulation factors

The mechanistic FWM, parameterized for the Barents Sea marine food web in spring, predicted mean PCB concentrations in zooplankton and fish that were 20 to 200 times lower than measured (Fig. 2). The water PCB concentrations (Olsson et al., 2002) used as model input, and the biota PCB concentrations (present study) used to compare with model predictions, were collected simultaneously in May

1999 to exclude temporal variation. Although the predicted biotic PCB concentrations were much lower than measured, there was a high significant correlation between the measured and predicted concentrations, except for PCB-28 and PCB-138 (Table 3). The non-significant correlation between predicted and measured PCB-28 and PCB-138 concentrations in biota may be due to quantification uncertainty in either water or zooplankton, respectively, and will be addressed below.

The observed BAFs were two orders of magnitude higher (Fig. 3) than reported for other Arctic studies, which reported log BAFs of about 5.5–6.5 for

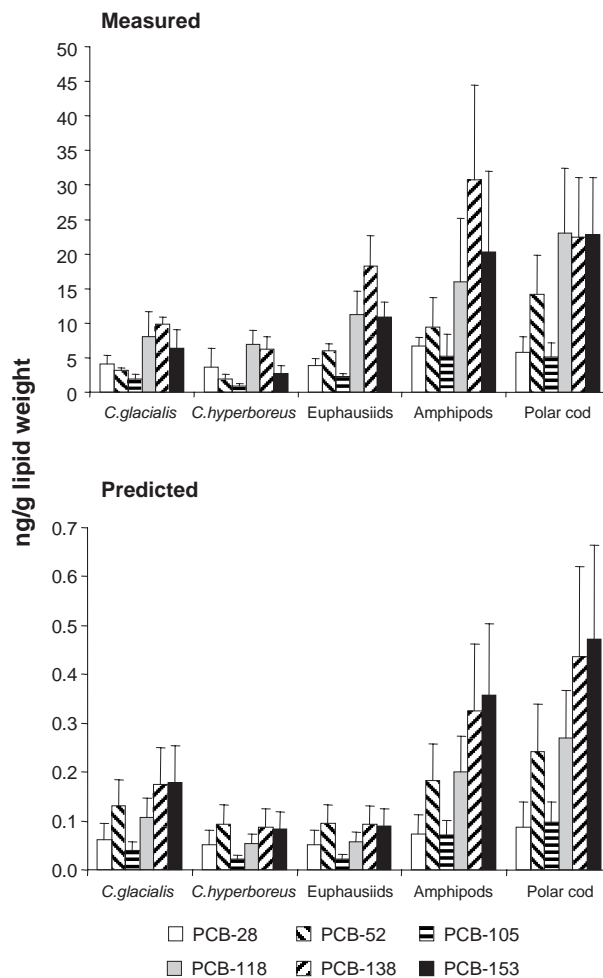


Fig. 2. Mean measured and FWM predicted PCB concentrations (ng/g lipid weight) in Barents Sea zooplankton and fish. Variation terms are 95% confidence interval in measured data, and 40% quantification uncertainty of water concentrations in the predicted data. Note that scales differ.



Table 3

Spearman's rank order correlation between measured and predicted PCB concentrations in Arctic zooplankton and fish using the Campfens and Mackay food web model (1997)

Congener	$R^2$	Adjusted $R^2$	$p$
PCB-28	0.67	0.55	0.092
<b>PCB-52</b>	<b>0.84</b>	<b>0.78</b>	<b>0.029</b>
<b>PCB-105</b>	<b>0.83</b>	<b>0.77</b>	<b>0.032</b>
<b>PCB-118</b>	<b>0.86</b>	<b>0.81</b>	<b>0.024</b>
<b>PCB-153</b>	<b>0.83</b>	<b>0.77</b>	<b>0.032</b>
PCB-138	0.49	0.32	0.189

Significant correlations are highlighted in bold.

congeners in the same  $K_{OW}$  range as the present study (Fisk et al., 2001b; Hoekstra et al., 2002). Log BAF of 4.5–6 was reported for non-Arctic freshwater zooplankton for congeners in the same  $K_{OW}$  range as the present study (e.g. Oliver and Niimi, 1988).

### 3.2. Why do predicted and measured biotic PCB concentrations differ?

#### 3.2.1. Zooplankton data

One possible explanation for high observed BAFs and for the large difference between measured and predicted zooplankton and fish PCB concentrations may be error associated to the measured PCB concentrations in biota. However, although the measured zooplankton and fish PCB concentrations were low, they were quantified within the accredited laboratory's quality assurance requirements (Borgå et al., 2003). The operating laboratory has participated and been approved in several international intercalibration tests such as those organised by ICES/IOC/OSPARCOM (Anon, 1995; de Boer et al., 1996), WHO/UNEP, IUPAC, and the National Reference Laboratories in the European Union. In addition, between-laboratory coefficient of variation for PCB quantification is about 15–20% in marine biota (de Boer and Law, 2003), and it is therefore unlikely that the observed difference between predicted and measured PCB concentrations is due to measurement errors in biota.

There are few PCB data available from the Barents Sea food web for comparison; however, the present PCB concentrations in copepods were generally comparable to levels reported for other Arctic marine areas (e.g. Fisk et al., 2001b; Borgå et al., 2002a; Hoekstra et al., 2002), and to the Barents Sea in 1995

(Borgå et al., 2001). Euphausiids and amphipod PCB levels were 2–3 times higher than reported from the Barents Sea in 1995 (Borgå et al., 2001, 2003). However, zooplankton from 1995 were not sorted to species, but were rather separated by size fractions, and the comparison is therefore not optimal. The present PCB concentrations in polar cod were comparable to or slightly lower than (depending on congener) the polar cod PCB concentrations in 1995 (Borgå et al., 2001). Although the samples may have been contaminated during the sampling procedures, precautions were taken to avoid onboard contamination (see Borgå et al., 2002a,b). In addition, contamination during sampling seems unlikely, as zooplankton PCB levels are comparable across the Arctic region.

#### 3.2.2. Predictability of the food web model

The FWM has previously been shown to satisfactorily predict the behaviour of various POPs in other aquatic food webs leading to fish (e.g. Campfens and Mackay, 1997; Arcuri, 1998; Fraser, 2002). However, the assumption of steady state in the FWM may not be realistic, both due to historical differences in contam-

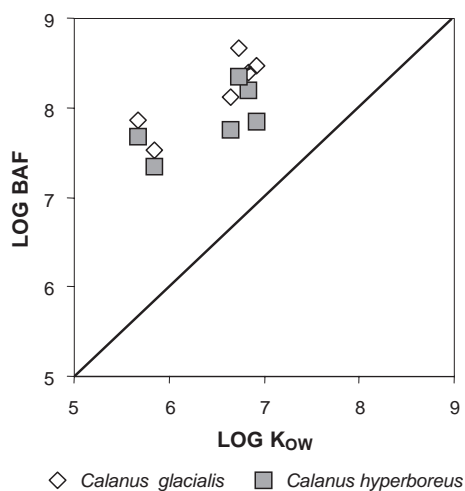


Fig. 3. Relationship between bioaccumulation factors (BAF) and octanol–water partitioning coefficients ( $K_{OW}$ ) for different PCBs in calanoid copepods from the Barents Sea in May 1999. The line represents a 1:1 relationship. Log  $K_{OW}$  are from Hawker and Connell (1988), whereas BAFs were calculated with measured dissolved water PCB concentrations (Olsson et al., 2002) and measured calanoid copepod PCB concentrations on a lipid weight basis (present study) (Eq. (2)).

ination loads, episodic or transient environmental situations, seasonal variation, and differentiated uptake and loss rates by organisms. An unsteady-state condition in environmental or organism characteristics might be of importance for environments with high seasonal amplitude (such as the Arctic), and for large and warm-blooded species at higher trophic positions in the food web (such as seabirds and marine mammals). However, although some of the assumptions made in the conceptual FWM might not be applicable to the Arctic marine ecosystem and/or large vertebrate organisms, the FWM predicted relatively well the PCBs' behaviour in the food web leading to fish. This is based on the high correlation between predicted and measured biotic PCB concentrations (Table 3), and the relatively low variation terms of predicted biota concentrations assuming 40% uncertainty in bulk water concentrations (Fig. 2).

Sensitivity analyses identified water concentration as the most sensitive input parameter for all PCBs in all biota, with a change in predicted PCB concentration as high as the change in water PCB concentration (results not shown). Dissolved water concentration is used to predict the uptake into various biota via respiration, and as the basis for dietary exposure due to absorption into phytoplankton or zooplankton. Thus, accurate data for PCBs in water are required to successfully predict the PCB propagation in the food web. Other uncertain or variable state variables of importance were organisms' growth rate and lipid content, especially for small herbivorous zooplankton.

### 3.2.3. Variance in reported PCB concentrations from northern waters

As discussed in the recent Canadian Arctic Contaminants Assessment Report, measured PCB concentrations in water vary highly both among and within the Arctic regions, depending among other things on methods used for sampling (Muir and Strachan, 2003). Dissolved PCB water concentrations from the Barents Sea in May 1999 (Olsson et al., 2002), collected simultaneously as the present zooplankton and fish, were used as original water input concentrations to the FWM. These dissolved water concentrations (0.3–0.5 pg/L  $\sum$ PCB<sub>7</sub>, including the congeners -28, -52, -105, -118, -138, -153, -180) were comparable to concentrations reported from the

Greenland Sea in August 1993 (0.1 pg/L  $\sum$ PCB<sub>6</sub>, PCB-105 not reported) (Schultz-Bull et al., 1998), and from the Barents Sea in 2001 (1.0 pg/L  $\sum$ PCB<sub>7</sub>) (Sobek and Gustafsson, 2004; Gustafsson et al., 2005). They were, however, 2 orders of magnitude lower than PCB concentrations from Barents Sea whole water samples in 1996 (57 pg/L  $\sum$ PCB<sub>10</sub>, including the congeners -28, -31, -52, -101, -105, -118, -138, -153, -156, -180) (Strachan et al., 2000 in Muir and Strachan, 2003). Bulk (dissolved+particulate) PCB concentrations in the Barents Sea in 1999 and in the Greenland Sea in 1993 were 0.6–1.0 pg/L  $\sum$ PCB<sub>7</sub> and 0.97 pg/L  $\sum$ PCB<sub>6</sub>, respectively (Schultz-Bull et al., 1998; Olsson et al., 2002).

All of these measurements are reported from laboratories with high analytical quality; however, the largest uncertainties of PCB quantification in seawater are usually encountered in the sampling step (IOC, 1993; Petrick et al., 1996). The above-mentioned studies differed in sampling method in terms of how seawater was collected (In situ collection with remotely deployed samplers or submersible pumping onto the ship), if and how particles were removed (by filtration or centrifugation), and how the dissolved PCBs were trapped (XAD-2 resin or polyurethane foam (PUF)) (see Muir and Strachan, 2003). Whereas Schultz-Bull et al. (1998) used an In situ pump (Kiel In Situ Pump, KISP), filtering seawater followed by extraction with XAD-2 resins submersed in the water, Sobek and Gustafsson (2004), Gustafsson et al. (2005) and Olsson et al. (2002) pumped seawater onboard for instant filtration followed by extraction with PUFs. The Barents Sea PCB concentrations from 1999 and 2001 were comparable and were analysed at the same laboratory, where additional precautions were made to prevent contamination (Olsson et al., 2002; Gustafsson et al., 2005; Sobek and Gustafsson, 2004). The studies used a stainless steel seawater intake system with filters and PUFs, however, the pumps differed and special care was taken in the 2001 study to keep the field conditions ultra-clean (Gustafsson et al., 2005; Sobek and Gustafsson, 2004). Filters in series from the KISP were analysed to control for potential sorption of dissolved PCBs to the filter, which showed to be negligible (Petrick et al., 1996). In contrary to Strachan et al. (2000), the studies demonstrating low PCB levels sampled high volumes and water that was filtered before extraction. Strachan

et al. (2000) sampled water with “Go-flo” bottles followed by extraction with XAD-2 resins. Although the Go-Flo bottles are convenient for rapid sampling, there is a risk of contamination as indicated by high field blanks (D.C.G. Muir, personal communication). The procedural blanks of the studies reporting low PCB levels were low and only congeners with signal-to-noise higher than 3 were accepted (Petrick et al., 1996; Sobek and Gustafsson, 2004). The blanks were subtracted from the Barents Sea results (Sobek et al., 2003; Sobek and Gustafsson, 2004; Gustafsson et al., 2005), but not from the Greenland Sea results, as they were considered low enough to be neglected (Petrick et al., 1996; Schultz-Bull et al., 1998).

In situ seawater sampling from the Canadian and Alaskan Arctic resulted in PCB concentrations in the range of those reported from the Barents Sea in 1996 (Muir and Strachan, 2003). This In situ pumping method differed from Schultz-Bull et al. (1998) in that a smaller water volume was sampled, that the XAD-2 columns were pre-spiked to control for breakthrough, and that the results were blank-corrected (e.g. Hoekstra et al., 2002). As the sampling of smaller water volumes resulted in higher PCB concentrations, this might suggest that In situ sampling of high volumes leads to breakthrough of the cartridge in use and loss of the dissolved phase. However, due to low water concentrations, analytical constraints and potential sampling contamination, high water volumes (500–1000 L) must be sampled to obtain reliable results (Petrick et al., 1996). In addition, the Canadian and Alaskan field and laboratory blanks were high (up to 50%), and high uncertainty may therefore be associated to these studies (D.C.G. Muir, personal communication). Using non-spiked PUFs to extract water pumped onboard, the filtering of different high volumes did not show any change in PCB concentrations, suggesting that breakthrough during high-volume sampling was negligible (Sobek et al., 2004).

To summarize, the different results obtained using different methods illustrate the need to obtain a standardization of methods to make future data comparable.

#### 3.2.4. Estimating dissolved water concentrations from zooplankton and air measurements

The measured water concentrations from 1999 were lower than the lowest predictions of dissolved

water concentrations for the Barents Sea, independent of estimation method and congener (Table 4, Fig. 4). Using measured PCB concentrations in *C. hyperboreus*, the FWM predicted mean dissolved water concentrations of 28 pg/L  $\sum\text{PCB}_6$  (Sum of PCB-28, -52, -105, -118, -138, -153) (Table 4, Fig. 4). Based on the BAF- $K_{OW}$  relationship reported for the North Water Polynya in the Canadian Arctic, the mean dissolved water concentration in the Barents Sea was 29 pg/L for  $\sum\text{PCB}_6$  (Table 4, Fig. 4). Thus, the empirical BAF- $K_{OW}$  model and the mechanistic FWM both predicted dissolved water PCB concentrations approximately 2 orders of magnitude higher than measured in the Barents Sea in May 1999 (0.3 pg/L  $\sum\text{PCB}_6$ , 30–150 times higher than measured depending on congener and sample used for comparison) (Table 4, Fig. 4).

Mean dissolved water PCB concentration predicted from air concentrations from the Barents Sea in 1996 (Harner et al., 1998) was 7.6 pg/L  $\sum\text{PCB}_5$  (PCB-28 was not analysed) (Table 4, Fig. 4). This is 3 to 82 times higher than measured in 1999 (84–303 times higher for PCB-118), depending on congener, air sample and water sample. Mean water PCB concentrations predicted from zooplankton data were 0.5 to 6.2 times the air-calculated ones, depending on congener, estimation method and air sample (Table 4, Fig. 4). The exception was PCB-138, which was 16–45 times higher in the zooplankton predicted water PCB concentrations than in the air-predicted ones, depending on model and sample compared (Table 4, Fig. 4). PCB-138 was one of the PCBs with a non-significant correlation between measured and predicted PCB concentrations in biota (Table 3), which may suggest that this congener was quantified too high in the zooplankton, maybe due to co-eluting compounds.

Water PCB concentrations predicted from air concentrations from the Eastern Arctic Ocean (Harner et al., 1998) and West-Spitsbergen (Ny-Ålesund) (de Wit et al., 2004) were more comparable to the recent PCB measurements (Olsson et al., 2002; Gustafsson et al., 2005) (Table 4, Fig. 4). The Barents Sea and Eastern Arctic Ocean air-PCB data were from the same study (Harner et al., 1998). Air PCB data reported by Harner et al. (1998) from Alert in the Canadian Arctic in 1993/4 were comparable to those reported by Hung et al. (2001). No further discussion is presently included due to the additional variation

Table 4  
Measured and predicted polychlorinated biphenyl (PCB) water concentrations in the Barents Sea (pg/L)

Medium/Year	Reference	Where	Station	PCB-28	PCB-52	PCB-101	PCB-105	PCB-110	PCB-118	PCB-138	PCB-149	PCB-153
<i>Measured dissolved</i>												
1999	1	Barents Sea	Open water	0.10	0.11	0.10	0.02	0.06	0.04	0.05	0.07	0.06
1999	1	Barents Sea	Ice covered	0.06	0.07	0.07	0.01	0.04	0.02	0.03	0.05	0.02
2001	2	Barents Sea	Arctic water	0.19	0.19	0.16	0.06	0.12	0.18	0.26	0.09	0.21
2001	2	Barents Sea	Arctic water	0.24	0.24	0.17	0.03	0.10	0.08	0.11	0.08	0.10
<i>Back-calculated from air measurements</i>												
1996	3	Barents Sea	Sample 1 (min–max) <sup>6</sup>	1.18 (0.55–2.30)	0.76 (0.36–1.50)	0.49 (0.08–0.33)	0.27 (0.13–0.53)	3.46 (1.58–6.48)	0.14 (0.07–0.27)	0.36 (0.16–0.67)	0.44 (0.21–0.81)	
1996	3	Barents Sea	Sample 2 (min–max) <sup>6</sup>	2.17 (1.01–4.23)	1.41 (0.66–2.75)	0.91 (0.15–0.61)	0.49 (0.23–0.97)	6.37 (2.9–11.9)	0.26 (0.12–0.50)	0.66 (0.30–1.23)	0.80 (0.38–1.93)	
1996	3	Eastern Arctic Ocean	Mean (min–max) <sup>6</sup>	0.32 (0.15–0.63)	0.21 (0.09–0.41)	0.14 (0.02–0.09)	0.07 (0.03–0.14)	0.95 (0.43–1.78)	0.04 (0.02–0.08)	0.10 (0.04–0.18)	0.12 (0.06–0.22)	
1998	4	Ny-Ålesund, Svalbard	(min–max) <sup>6</sup>	2.03 (0.61–5.97)	0.80 (0.37–1.56)	0.51 (0.24–0.99)	0.30 (0.05–0.20)	0.46 (0.21–0.86)	0.16 (0.07–0.31)	0.39 (0.19–0.73)		
<i>Back-calculated from zooplankton measurements</i>												
BAF- <i>K</i> <sub>OW</sub> /1999	5	Barents Sea	Mean (min–max) <sup>7</sup>	8.2 (1.8–16.8)	3.7 (2.2–5.8)		0.9 (0.5–1.4)	7.2 (4.9–10.1)	6.3 (4.3–8.6)		2.7 (1.6–4.0)	
FWM/1999	5	Barents Sea	Mean (min–max) <sup>7</sup>	10.1 (2.1–23.5)	4.1 (2.2–7.0)		0.8 (0.5–1.2)	5.8 (4.0–8.5)	5.2 (3.5–7.2)		2.2 (1.3–3.2)	

Congeners are only listed if reported in more than one study (water measurements, air measurements and zooplankton measurements).

(1) Olsson et al. (2002): filtered water samples, dissolved fraction collected by PUFs.

(2) Gustafsson et al. (2005): filtered water samples, dissolved fraction collected by PUFs.

(3) Harner et al. (1998): Using High-volume air samplers equipped with GFF and PUFs to collect the particulate and gas phase, respectively. Reported values are calculated from the gaseous phase collected with PUFs.

(4) de Wit et al. (2004): High-volume samplers. Total air concentration (particulate+gas phase), average annual concentrations.

(5) Present study.

(6) Minimum water PCB concentration was predicted by assuming minimum air concentration (Sample–30%), minimum water solubility (selected value–20%) and maximum vapour pressure (selected value+20%), and vice versa for maximum estimations of PCB concentrations.

(7) Minimum water PCB concentrations predicted by the FWM and the BAF-*K*<sub>OW</sub> model was obtained assuming minimum zooplankton PCB concentration (mean–95% confidence interval) and minimum water solubility (*K*<sub>OW</sub>+30%), and vice versa for maximum estimations of PCB concentrations.

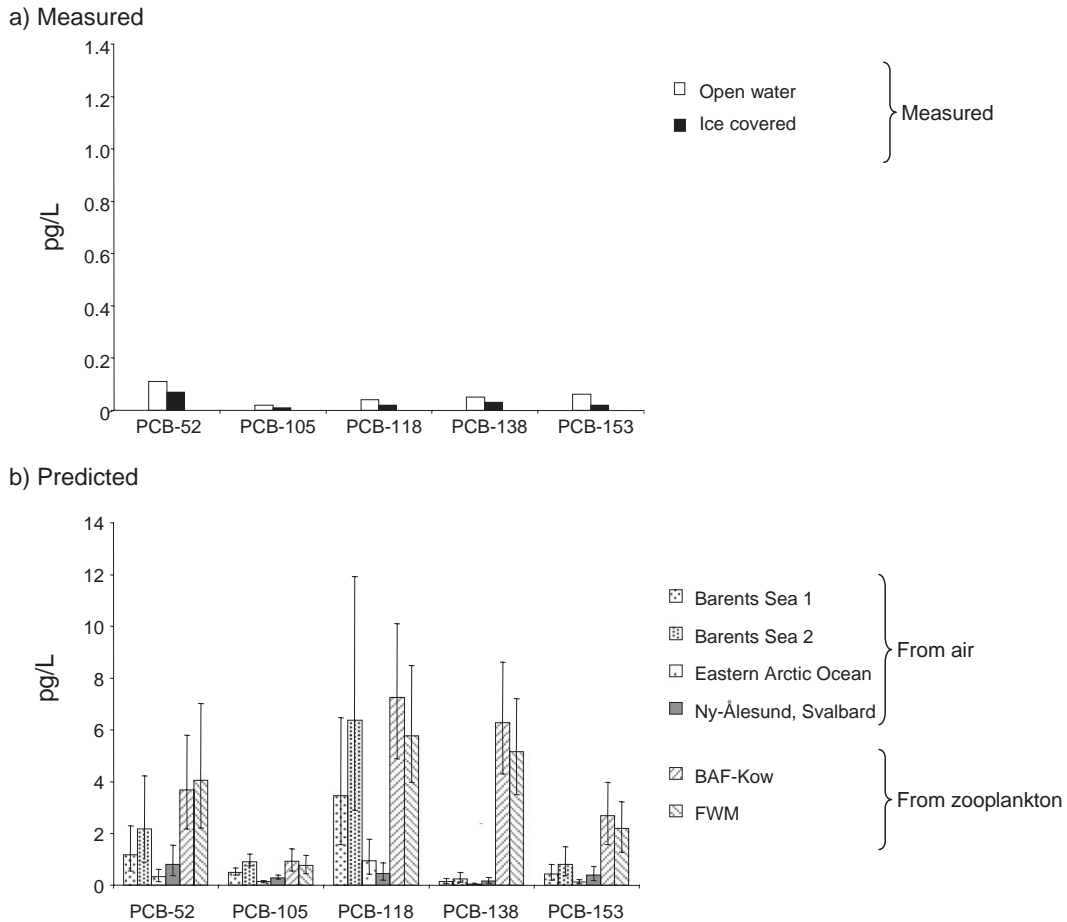


Fig. 4. Dissolved water PCB concentrations. (a) Measured by Olsson et al. (2002). (b) Predicted based on measured PCB concentrations in calanoid copepods (Present study) and air (Harner et al., 1998; de Wit et al., 2004), see text and Table 4 for explanation. Minimum and maximum estimations for the zooplankton back calculations were based on minimum and maximum zooplankton concentrations (mean  $\pm$  95% confidence interval), and maximum and minimum BAF ( $K_{OW} \pm 30\%$ ), respectively. Minimum and maximum in air-back calculations were based on minimum and maximum air concentration (measured  $\pm 30\%$ ), minimum and maximum water solubility (selected value  $\pm 20\%$ ) and maximum and minimum vapour pressure (selected value  $\pm 20\%$ ), respectively. Note that the scale in (a) is 10 times lower than in (b).

introduced by different area, year and/or sampling method in the different studies (see Table 4 footnote). Predictions of water concentrations from measurements in air must be considered as a tentative attempt to further back-calculate water concentrations. Given high variability in air concentrations high uncertainty in predictions is expected, as illustrated by minimum and maximum predictions (Table 4, Fig. 4).

As a control of the back-calculations for PCBs, hexachlorocyclohexane (HCH) concentrations in Arctic waters were predicted using the FWM and the BAF- $K_{OW}$  model. Only few problems seem to be associated to the sampling and analyses of dissolved

HCHs, as the measurements correspond relatively well among field studies (Muir and Strachan, 2003). Dissolved HCH concentrations from water are available from the Barents Sea, eastern Arctic Ocean, north of Svalbard and the Greenland Sea from 1994 and 1996 (Jantunen and Bidleman, 1996, 1998; Harner et al., 1999; Strachan et al., 2000 in Muir and Strachan, 2003) (Table 5). Unfortunately, none of the studies reporting low PCB concentrations in the Barents Sea measured HCHs. *C. hyperboreus* from the Greenland Sea and North-East of Svalbard (Fig. 1) from September and October 1999 (Borgå et al., 2002a) were used to predict dissolved water concentrations in

Table 5  
Dissolved hexachlorocyclohexane (HCH) concentrations (ng/L) in European Arctic seawater in the 1990s

Area	Reference	Year	Month	Method <sup>7</sup>	Depth (m)	α-HCH			γ-HCH		
						Mean (Range)	S.D. <sup>8</sup>	Min <sup>9</sup> –Max <sup>9</sup>	Mean (Range)	S.D. <sup>8</sup>	Min <sup>9</sup> –Max <sup>9</sup>
Barents Sea	1	1996	July and September	Pump/Filter/SPE	Surface	0.39	0.05		0.14	0.03	
	2	1999	May	BAF– $K_{OW}$	–	0.20		0.08–0.42	0.20		0.08–0.41
North of Svalbard <sup>10</sup>	2	1999	May	Food web model	–	0.85		0.31–1.82	0.85		0.35–1.80
	3	1994	September	Go-Flo/Filtered/SPE	10–109	(0.95–1.44)			–		
	4	1996	July–September	Go-Flo/Centrifuge/XAD	–	0.56			0.17		
	1	1996	July	Pump/Filter/SPE	Surface	0.76	0.12		0.21	0.04	
Greenland Sea	5	1994	September	SPE	40–60	0.95			0.22		
	2	1999	September	BAF– $K_{OW}$	–	0.25		0.18–0.38	0.17		0.07–0.35
	2	1999	September	Food web model	–	1.08		0.75–1.70	0.75		0.32–1.60
	5	1994	September	SPE	40–60	0.63			0.20		
	1	1996	July–September	Pump/Filter/SPE	Surface	1.18	0.32		0.27	0.08	
	6	2000	July	Pump/SPE (unfiltered)	11	0.32	0.18		0.14	0.05	
Greenland Sea	3	1994	September	Go-Flo/Filtered/SPE	0–50	0.87	0.22		0.20	0.03	
	2	1999	October	BAF– $K_{OW}$	–	0.53		0.38–0.78	0.14		0.11–0.21
	2	1999	October	Food web model	–	2.20		1.60–3.45	0.63		0.46–0.95

(1) Harner et al. (1999).

(2) Present study: Estimations from *Calanus hyperboreus* HCH concentrations from the Barents Sea (present study), North of Svalbard and Greenland Sea (Borgå et al., 2002a). Physicochemical data used in estimations for α- and γ-HCH were from Mackay (2001).

(3) Jantunen and Bidleman (1996).

(4) Strachan et al. (2000).

(5) Jantunen and Bidleman (1998).

(6) Lakaschus et al. (2002). The samples were unfiltered, and thus represent bulk concentration.

(7) Methods:

Go-Flo: Teflon-linen Go-Flo bottles to collect water

Pump: Teflon tube surrounded by a flexible metal mesh, water transferred to stainless steel cans using Teflon tubing

Filtered: glass fiber filter to remove particles

Centrifuge: continuous centrifugation to remove particles

SPE: Silica solid phase extraction cartridge

XAD: dissolved fraction of organochlorines absorbed to XAD-2 resin

(8) Standard deviation.

(9) Min and Max: Minimum and Maximum dissolved water concentrations were predicted as described for PCBs (text and Table 4), assuming a  $K_{OW}$  uncertainty of 30%, and 95% confidence interval of calanoid copepod PCB concentrations. *C. hyperboreus* copepodit stages IV, V and VI were used for the Barents Sea, and stage CVI for the Arctic Ocean and Greenland Sea. Default log  $K_{OW}$  used of estimations were 3.81 and 3.7 for α- and γ-HCH, respectively (Mackay, 2001).

(10) North of Svalbard also includes the area of Eastern Arctic Ocean and the Northern Barents Sea.

these areas, whereas *C. hyperboreus* from the Barents Sea in May 1999 (present study) were used for the Barents Sea comparison. The BAF– $K_{OW}$  predicted HCH concentrations were comparable to those measured in the northern Barents Sea, eastern Arctic Ocean and in the Greenland Sea (Table 5). The FWM

predicted HCH concentrations in the upper range, or 2–3 times of reported concentrations (Table 5).

### 3.2.5. Bioaccumulation factors and equilibrium

A curvilinear relationship between log BAF and log  $K_{OW}$  may suggest that the zooplankton are not in

equilibrium with water, or that one of more of the assumptions for equilibrium partitioning are incorrect (Fisk et al., 2001b). If the assumptions for equilibrium partitioning are correct, a linear 1:1 relationship is expected between log BAF and log  $K_{OW}$  (e.g. Neely et al., 1974; Veith et al., 1979). The assumptions for equilibrium partitioning are that POPs partition only into the lipid pool of the organism, that the main exchange route is respiration, that organisms have negligible biotransformation abilities, and that measured dissolved POP concentrations in water reflect the truly bioavailable fraction.

Whereas assuming negligible biotransformation in zooplankton and fish seems to be acceptable (e.g. Moisey et al., 2001), assuming that POP partition only into lipids and not other organic phases is debated (e.g. Swackhamer and Skoglund, 1993; Skoglund and Swackhamer, 1999). The importance of dietary versus direct uptake in zooplankton and small fish is also debated and depends highly on the water solubility of the contaminant (e.g. Russell et al., 1999).

In the FWM and the BAF- $K_{OW}$  models, POP concentration in biota is predicted from the bioavailable (=truly dissolved) concentration that can partition into the organisms. This is also true for predatory zooplankton and fish, since concentrations in their diet (both primary producers and consumers) are predicted from the dissolved POP concentration. Whereas truly dissolved PCBs are available for respiratory uptake, PCBs partitioned into dissolved organic carbon are not, and this fraction is not easily separated from the truly dissolved fraction (Urrestazu-Ramos et al., 1998). Thus, if measuring truly dissolved POP concentration in water is problematic this will lead to variation between predicted and measured POPs in biota.

The difference between measured and predicted PCB concentrations in biota is too large to be explained by temporal variation in water concentrations and that zooplankton might not be in equilibrium with water. The difference between predicted and measured dissolved PCB water concentration is much larger (100 times) than the annual change in dissolved water concentrations (2 times) reported from other Arctic ice covered marine areas (Hargrave et al., 1997, 2000; Fisk et al., 2001b). Although contaminants in the Arctic have decreased in the last decades, such as in air (Bidleman et al., 2002), there was in general no

decrease in the 1990s in PCB air concentrations neither in the Svalbard area nor the Canadian Arctic (Hung et al., 2001; Berg et al., 2003). Thus, the large difference between reported (or predicted) water concentration from 1996 and 1999 cannot be due to temporal decrease in ambient PCB concentration. The difference between Barents Sea PCB concentrations measured in 1996 and 1999/2001 cannot either be attributed to variation in depth, as water was sampled close to the surface (Strachan et al., 2000; Olsson et al., 2002; Gustafsson et al., 2005).

If the present high BAFs were realistic, bioaccumulation in Arctic zooplankton is more efficient than previously demonstrated. Then, BAFs previously reported for Arctic zooplankton (Fisk et al., 2001a,b; Hoekstra et al., 2003) are too low, probably as they were calculated from water PCB concentrations that might be too high (see discussion above). If so, then the empirically derived BAF- $K_{OW}$  relationship for Arctic zooplankton (Fisk et al., 2001a,b) is also incorrect, and dissolved water concentrations predicted from PCB concentrations in zooplankton, using the empirical BAF- $K_{OW}$  relationship, are too high. Further, the scaling-difference between measured and FWM-predicted PCB concentrations in biota could be related to underestimation of the copepod PCB concentration by the FWM. Reasons for potential bioaccumulation underestimation at the first trophic level are not yet clear.

### 3.3. Changing FWM parameterization to obtain the measured PCBs in water and zooplankton

#### 3.3.1. Using measured dissolved-to-particulate PCB ratio to predict total particulate matter

When the measured ratio between dissolved and particulate PCB concentrations (Olsson et al., 2002) was predicted by the FWM, the total particulate matter (TPM) concentration in the water had to be varied from 1.8 to 145 g/m<sup>3</sup>, depending on congener, ice cover, and percentage of particulate organic carbon (POC) assumed in TPM (Fig. 5). These calculations were done independently of zooplankton and fish PCB concentrations, and only involved the measured PCB concentrations in water and the chemical and environmental state variables. Theoretically, assuming equilibrium partitioning between the dissolved and the particulate phase, the predicted TPM concentrations

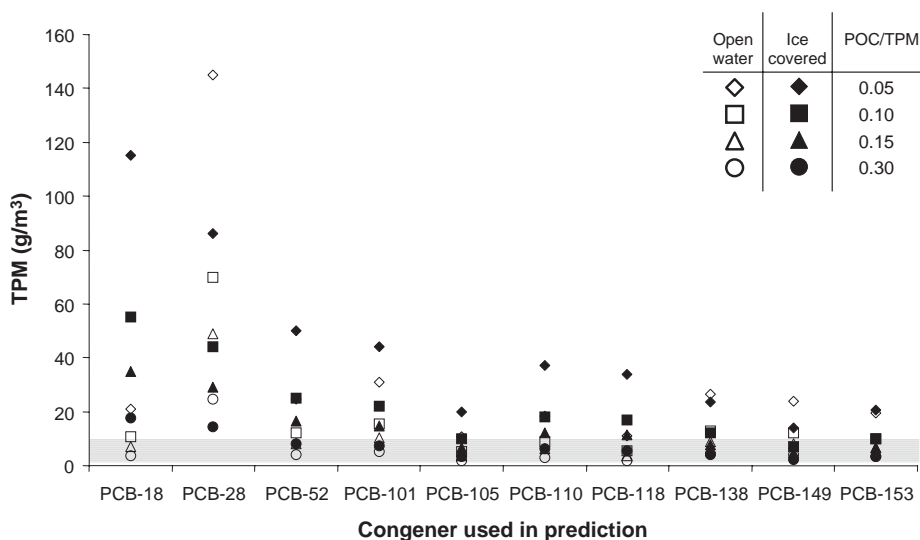


Fig. 5. Total particulate matter concentration (TPM) ( $\text{g}/\text{m}^3$ ) in the water column predicted from the measured ratio between dissolved and particulate associated PCB concentrations in the Barents Sea in May 1999 (Olsson et al., 2002). The organic carbon (POC) in total particulate matter was assumed to be 5%, 10%, 15% and 30% as indicated by the symbols, to cover the range in an Arctic fjord (Reigstad, 2000). The TPM was calculated for dissolved-to-particulate ratio from an ice-covered (filled symbols) and an open-water (open symbols) station. The grey area illustrates the range of TPM ( $1.6\text{--}9.5 \text{ g}/\text{m}^3$ ) in the Barents Sea in May calculated from measured POC (Olli et al., 2002), assuming POC content in TPM of 5 to 30%.

should be the same independently of which congener is used for back-calculation. The different physico-chemical properties important for the partitioning between the dissolved and the particulate phase are accounted for by the organic carbon–water partitioning coefficient ( $K_{OC}$ ). However, differences in TPM predictions can be expected depending on POC content in TPM and area-specific primary production. With increasing POC in TPM, the TPM needed to obtain the observed dissolved-to-particulate PCB ratio decreased (Fig. 5). TPM was generally higher for the ice-covered station than for the open-water station for all congeners and POC contents, in accordance with higher productivity and sedimentation is in ice-covered than in open-water areas (e.g. Olli et al., 2002).

Depending on the assumed POC content in TPM (5–30%), TPM concentrations in the Barents Sea marginal ice zone calculated from POC measurements (Olli et al., 2002) were  $0.2\text{--}1.2$ ,  $1.6\text{--}9.5$  and  $0.7\text{--}4.5 \text{ g}/\text{m}^3$  in March, May and June, respectively, illustrating a pre-bloom, bloom, and post-bloom situation. Compared to TPM calculated from measured POC, the FWM-predicted TPM concentrations were especially high assuming low POC content for all

congeners (Fig. 5). Even when assuming high POC content, high TPM concentrations were predicted using PCB-18 and PCB-28 in the ice-covered station, and PCB-28 in the open-water station, for the back-calculation (Fig. 5). The high TPM concentrations predicted from these congeners may indicate non-equilibrium or quantification difficulties. PCB-28 was one of the congeners with a non-significant correlation between measured and predicted PCB concentrations in zooplankton and fish (Table 3), whereas PCB-18 was not analysed in biota. PCB-28 is known to co-elute with PCB-31 under certain gas chromatographic conditions, and PCB-28 and PCB-31 have similar relatively high dissolved water concentrations in Arctic waters (Schultz-Bull et al., 1998; Fisk et al., 2001b). The TPM concentrations predicted using the other congeners were more coherent, and suggest a TPM concentration of  $2.4\text{--}25.0$  and  $1.8\text{--}15.5 \text{ g}/\text{m}^3$  in ice-covered and open-water areas, respectively, assuming POC content in TPM of 10–30% (Fig. 5).

### 3.3.2. Maximizing bioaccumulation

To understand the low prediction of PCBs in biota, one question asked was whether the predicted elimination was too rapid. Growth rate, and thus



growth dilution as a way of reducing PCBs concentrations, was one of the state variables with high influence on the output of the model. When parameterized for the Arctic ecosystem (Table 2), growth rate for copepods was assumed similar to *Calanus finmarchicus* from an Arctic fjord (Edvardsen et al., 2002). *C. finmarchicus* is an Atlantic relative that has distribution areas partly overlapping with the Arctic species of the present study, *C. glacialis* and *C. hyperboreus*. New simulations were carried out changing the growth rate of copepods and other zooplankton to 0.01 g/g body weight day. This resulted in predicted PCB concentrations that were higher than with the original parameterization, but they were still 7 to 33 times lower than the measured PCB concentrations in *C. hyperboreus* depending on congener (excluding PCB-28, results not shown).

Several studies point to the importance of trophic interactions for contaminant transfer also for organisms low in the food web (e.g. Oliver and Niimi, 1988; Evans et al., 1991; Russell et al., 1999; Fisk et al., 2001b). Could the discrepancy between measured and predicted PCB concentrations in zooplankton and fish, even after reducing the growth rate, be due to underestimation of dietary exposure of PCBs? To investigate this hypothesis, dietary PCB uptake in herbivorous zooplankton was maximised by feeding on an unrealistically lipid-rich phytoplankton (50% lipids), exposed to measured dissolved PCB concentrations. However, there was still a large difference between measured and predicted PCB concentrations in biota, with 3 to 14 times higher PCB concentrations measured than FWM predicted in *C. hyperboreus* depending on congener (excluding PCB-28, results not shown).

#### 3.4. Summary

The present study has illustrated some of the difficulties encountered when different approaches of understanding environmental distribution of contaminants are compared. The comparison between measured and model-predicted contaminant concentrations in water and biota opens a wide array of possibilities to verify the overall model ability and/or adequacy of measured concentrations. Theoretically the results should be coherent among studies, but as illustrated unfortunately they are not. Recent meas-

ured PCB concentrations in water were lower than the lowest model predicted dissolved water concentrations for the Barents Sea, independently of estimation method and congener. They were also lower than earlier and recently reported PCB concentrations from the Barents Sea and other Arctic areas using other sampling methodology. Several possible hypotheses were investigated to identify reasons for the difference among measured water concentrations and between measured water concentrations and those predicted from measured PCB concentrations in zooplankton and in air. The zooplankton PCB data from the Barents Sea fits well with other studies, whereas the water measurements differ highly among studies using different methods. The recent low PCB concentrations were reported from studies sampling high water volumes, and where the field and laboratory blanks were low. Compared to the large difference between measured water PCB concentrations and those predicted from zooplankton, there was a smaller difference between water PCB concentrations predicted from air concentrations and from zooplankton. There was a relatively good correspondence between predicted and measured HCH concentrations in seawater; however, none of the studies reporting low PCB concentrations reported HCH measurements. If the high measured BAFs (log BAF 7.3–9.0) were realistic, it would mean that the bioaccumulation in Arctic zooplankton is more efficient than previously demonstrated. The present BAFs were calculated based on the low PCB concentrations measured in water sampled simultaneously as zooplankton. The present high BAFs then imply that previously empirically derived BAF- $K_{OW}$  relationships are incorrect as their BAFs were too low due to calculation from too high water PCB concentrations. When maximizing bioaccumulation by reducing growth and including a diet of lipid-rich phytoplankton, measured PCB concentrations in copepods were 3–14 times higher than predicted. In summary, sampling and quantification of PCBs from water is an issue deserving further attention. More studies are needed on the validation of sampling and quantification of contaminants in areas where concentrations are low due to the distance from sources, and where sampling conditions are difficult due to the low temperatures. Preferentially, an inter-calibration between different sampling and analytical methods is

needed to more accurately determine PCB water concentrations in polar regions.

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