PCB's in the sediment discharge test

The relationship between sediment and biota

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1220101-005

Deltares

Title
PCB's in the sediment discharge test

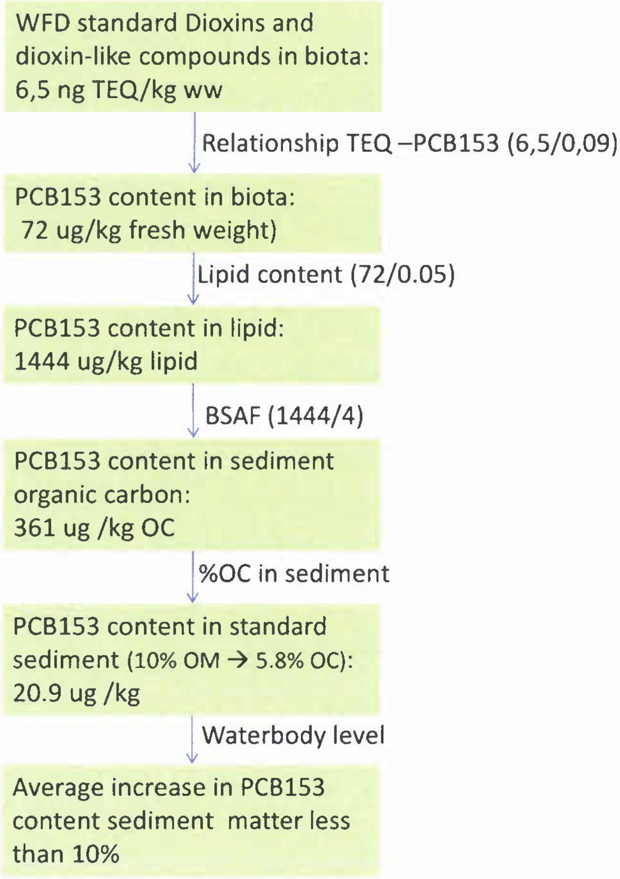
Client	Project	Reference	Pages
Rijkswaterstaat Water, Verkeer en Leefomgeving	1220101-005	1220101-005-ZKS-0003	15

Keywords
Sediment discharge test, PCB, dioxins, biota, fish, eel, bream

Summary

The Sediment discharge test is updated by implementing the new WFD-standards. The standards for the 7 so-called indicator PCBs (numbers: 28, 52, 101, 118, 138, 153 and 180) in suspended matter expired and no new PCB-standards have been proposed although it is generally acknowledged that contaminants in sediment are very relevant for PCBs. On the other hand, dioxins and dioxin-like PCBs are new priority substances which have standards in biota (fish, crustaceans and molluscs) instead of in water.

In this report a new standard has been derived for PCB153 in suspended matter which is comparable to the WFD-standard on dioxins and dioxin-like PCBs in biota. Four calculation steps are included as presented in the figure resulting in a PCB153 standard depending on the organic carbon fraction in the sediment: ($361*f_{OC}$) ug PCB153 /kg. If that is the case, the overall increase of PCB in sediment of the whole water body should not exceed 10%.



Versie	Datum	Auteur	Paraaf	Review	Paraaf	Goedkeuring	Paraaf
2	jan. 2016	Leonard Osté		Bert van Hattum*		Frank Hoozemans	

* Special thanks to reviewer Bert van Hattum for his valuable contribution to this report.

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Content

1 Introduction

The Sediment discharge test is updated by implementing the new WFD-standards. The Dutch standards for the 7 so-called indicator PCBs (numbers: 28, 52, 101, 118, 138, 153 and 180) in suspended matter expired and no new PCB-standards have been proposed although it is generally acknowledged that contaminants in sediment are very relevant for PCBs. On the other hand, dioxins and dioxin-like PCBs[1] (27 substances summated to one TEQ-value) are new priority substances in the WFD which have standards in biota (fish, crustaceans and molluscs) instead of in water.

Due to the fact that PCBs and dioxins cause extensive problems in the Dutch rivers and the fact that the mobilisation and emission of these substances from sediment to surface water is the dominating remaining source, RWS requested to replace the PCB standards in suspended matter in the Sediment discharge test (SDT) by dioxins in biota. Next question is how to link sediment quality to the standards for dioxins in biota. The Ministry of Infrastructure and Environment is still elaborating the Guidance on Monitoring of Biota (EC, 2014), but the Netherlands will probably monitor in bream, and occasionally in other biota, e.g. mussels (pers. communication Ten Hulscher).

The availability of congener-specific dioxin data in sediment is very limited and analysis is quite expensive. PCB153 in biota appears to be a reasonable indicator for dioxins in biota in the Dutch large rivers, but also in other countries. Biota to sediment accumulation factors (BSAFs) for PCB153 appear to be relatively constant in studies in different countries. Consequently, PCB153 in biota can be converted into PCB153 in sediments and/or suspended matter. This knowledge opens opportunities to use PCB153 data in sediments as an indicator for dioxins in biota. A similar approach to derive sediment quality objectives for dioxin-like compounds has been followed in a study of Traas et al. (2001).

This report will:

- Develop a calculation method to convert dioxin concentrations in biota to PCB153 in suspended matter.
- describe a short literature and (Dutch) data-scan on the relationship between PCB153 in sediment and PCB153 in fish
- describe a short literature and (Dutch) data-scan on the relationship between PCB153 in fish and dioxins in fish
- report a standard for PCB153 in suspended matter which is comparable to the WFD-standard on dioxins and dioxin-like PCBs in biota.

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[1] *This is the full name of the group of compounds. Where 'dioxins' is written in the remainder of this report, the full group of compounds (including dioxin--like substances) is meant.*

2 Conceptual framework

Figure 2.1 shows the steps that are needed to calculate dioxins in biota into PCB153 in suspended sediment.

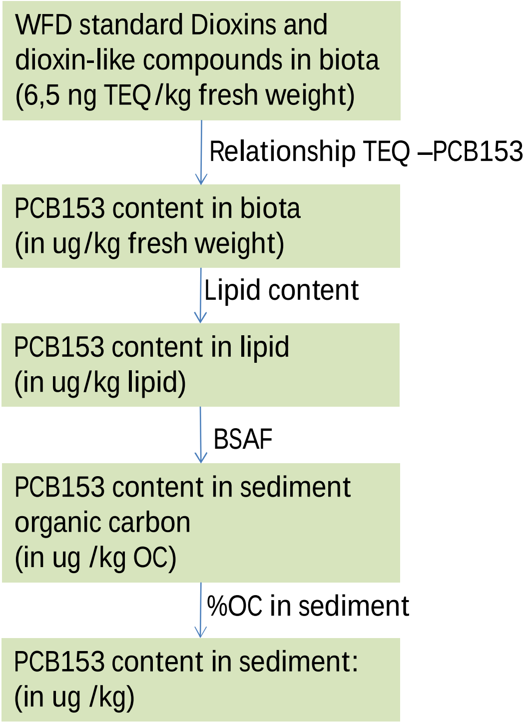


Figure 2.1 Calculation steps to convert the WFD standard for dioxins in biota to proportional standard in sediment

Starting point is the WFD standard as defined in biota. Four calculation steps are required:

1. Convert dioxins in biota into PCB153 in biota

In many studies on contaminants in biota, they have measured both indicator PCBs as well as dioxins. From these studies it is possible to derive a relationship (Kotterman and Glorius, 2011; van Hattum *et al.* 1998, 2013). Ten Hulscher (unpublished) listed a number of studies and derived relationships between sum7PCB and dioxins in fish, shell fish and cormorant eggs. For the conversion to sediment, more information is available on individual substances, so we prefer to find a relationship between PCB153 and dioxins which can be also found in most studies analysed by Ten Hulscher. The relationship between dioxins in biota and PCB153 in biota will be elaborated in Chapter 3.

2. Convert PCB153 in biota into PCB153 in lipid

Before the step to sediment, it is necessary to calculate the PCB153 in lipid instead of the full fresh weight. PCBs tend to accumulate in lipids, so the link to sediment is more reliable if the content in lipid is used. Lipid and sediment organic carbon normalised BSAF (biota to sediment accumulation factor) data are well available in the literature or on-line databases (USACE, 2015). The lipid content can vary a lot between species and between individuals, depending on e.g. sex, age, and nutritional state. Variation between individuals should be covered by competent sampling, but the variation between species needs to be implemented in the calculation. In the human risk model Sedisoil a lipid fraction of of 15% is chosen for fat fish (eel in particular) and 5% for other fish (e.g. bream, pike, perch pike, roach). Average lipid contents as found in several studies are presented in Table 2.1.

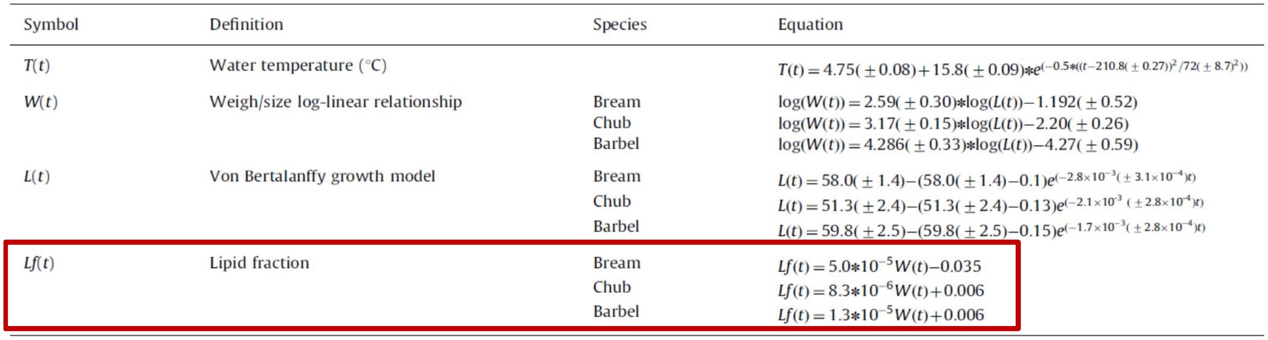
Table 2.1 Median lipid contents in various species as measured in various studies.

Species	**Number of locations or samples**	**Lipid content (%)**
Eel (NL)	74	13
Eel (Sweden; Jahnke, 2014)	5	23
‘low-fat’ fish (NL; Harezlak en Osté 2011, bijl. K)	≈30	≈3
Roach/carp/bream/chub/pike/perch (Latvia; Zacs, 2012)	9	3
Common Sole (Scheldt; Van Ael 2012)	24	0.69
European Flounder (Scheldt; Van Ael 2012)	35	0.81
Chinese Mitten Crab (Scheldt; Van Ael 2012)	13	0.7
Chinese Mitten Crab (body; NL)	6	10
Chinese Mitten Crab (legs; NL)	6	0.4

The lipid content in eel in Dutch rivers and lakes had gradually decreased since 1980 to current values in the range of 10 to 15% (De Boer et al., 2010). Lipid content data for bream, roach, perch and pike-perch from Amer and Haringvliet (1995-1997) varied between species and length classes; observed values were in the range of 0.7% to 6.5% (Van Hattum et al., 1998).

Lopez et al. (2012) developed a bioaccumulation model, using the formulas in Table 2.2 to calculate the lipid content from bodyweight data.

Table 2.2 Formulas to calculate the lipid fraction of bream, Chub and barbell, based on growth and size of the fish



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

The biota monitoring network will probably include bream or a comparable species, which is a low-fat fish. The current data-search reveals that the lipid fraction chosen for low-fat fish in SediSoil (5%) appears to be slightly high. Though 3% might even be a better value, the dataset is not large enough to deviate from the existing value of 5% for low-fat fish.

3. Convert PCB153 in lipid into PCB153 in sediment organic carbon (SOC)

A Biota-Sediment-Accumulation-Factor (BSAF) is necessary for this step. The BSAF is described by equation 1.

$$BSAF = C_{biota,lipid} / C_{sediment,SOC} \qquad [1]$$

In which:

$C_{biota,lipid}$ = contaminant concentration in lipids in biota

$C_{sediment,SOC}$ = contaminant concentration in sediment organic carbon

Variability in field-derived BSAFs can be caused by:

- The (type of) species and compound-specific metabolism. PAHs are metabolised by many fish species, and field observed BSAFs for PAHs in fish and sediment are much lower than BSAF data for PCBs, which are hardly metabolised (Burgess et al., 2002)
- Size, age or sex of the organism. Size does matter as concluded by Hendriks et al. (2001). Mundel et al. (2003) also observed an increased PCB content in lipids of fish if age and length increased. Also the lipid fraction increased with age. Kotterman (2015) found that the male eels (between 30-40 cm) contain significantly more dioxins compared to the females.
- Temperature. Opperhuizen et al. (1988) have illustrated a general trend for increasing BCF values for chlorinated benzenes with increased water temperature. Their data suggest an approximate 35%–40% increase in the BCF over a range of about 20°C.)
- Bioavailability in sediment. Recently, a few papers on passive sampling and uptake by fish have been published (Jahnke et al., 2014; Schäfer et al., 2015). They do not correlate the contaminants in biota to the total content in sediment, but to the pore water concentration. Schafer et al (2015) concluded that this approach gave clearer and more consistent results compared to conventional approaches that are based on total concentrations in sediment and BSAFs. They proposed to apply equilibrium sampling for determining bioavailability and bioaccumulation potential of HOCs, since this technique can provide a thermodynamic basis for the risk assessment and management of contaminated sediments.

Despite the variability, the use of BSAFs is most suitable for a simple tool like the Sediment Discharge Test. Detailed information on BSAFs is presented in Chapter **Error! Reference source not found.**.

4. The PCB153 content in sediment as a standard in the Sediment discharge test.

The direct relationship between PCB153 and dioxins in fish does not fit very in the Sdt, because the Sdt converts a concentration in sediment into a concentration in surface water after mixing and the resulting concentration is checked with the water standard. If there is no water standard but only a standard in sediment (and in biota), a mixing calculation in surface water is impossible. To prevent the situation that a relatively small intervention in contaminated sediment is not allowed, an additional rule is added.

Generally, the Sdt tool is only needed if the “weighed” average standardized substance content in the new sediment has a 10% higher substance than the content in the old sediment. This rule is applied on the surface of intervention only.

In case of sediment standards, this rule is extended to a water body level. If the new sediment exceeds the sediment standard, this is accepted as long as the concentration increase averaged over the water body surface is less than 10% higher of the concentration in the old sediment (equation 2)

$$\left[\frac{(C_{after} - C_{before})}{C_{before}}\right] \times \frac{A_{intervention}}{A_{Waterbody}} \quad \text{should be smaller than 0.1} \qquad [2]$$

In which

C_{after} = the concentration in sediment after intervention (the new sediment)

C_{before} = the concentration in sediment before intervention (the old sediment)
$A_{intervention}$ = the surface area of the intervention
$A_{water\ body}$ = the surface area of the whole water body

An example for standard sediment (5.8% OC) is shown in Table 2.3.

Table 2.3 Effect of the size of the intervention on the acceptability of a physical intervention in the sediment with respect to PCB153.

Parameter	**Scenario 1**	**Scenario 2**
Surface of the water body (m2)	50,000	50,000
Surface of the intervention (m2)	500	10,000
Concentration of PCB153 before intervention (ug/kg)	55	55
Concentration of PCB153 after intervention (ug/kg)	100	100
Increase	+((100-55)/55)*500/50000 = 0.0082	+((100-55)/55)*10000/50000 = 0.16
Increase in %	🟩 0.82%	🟥 16%

3 The relationship between dioxins and PCB153 in fish

Most detailed data is available from Dutch monitoring studies. Van Hattum, et al, (2013) monitored eel in the large Dutch river delta's and obtained the relationship as shown in Figure 3.1.

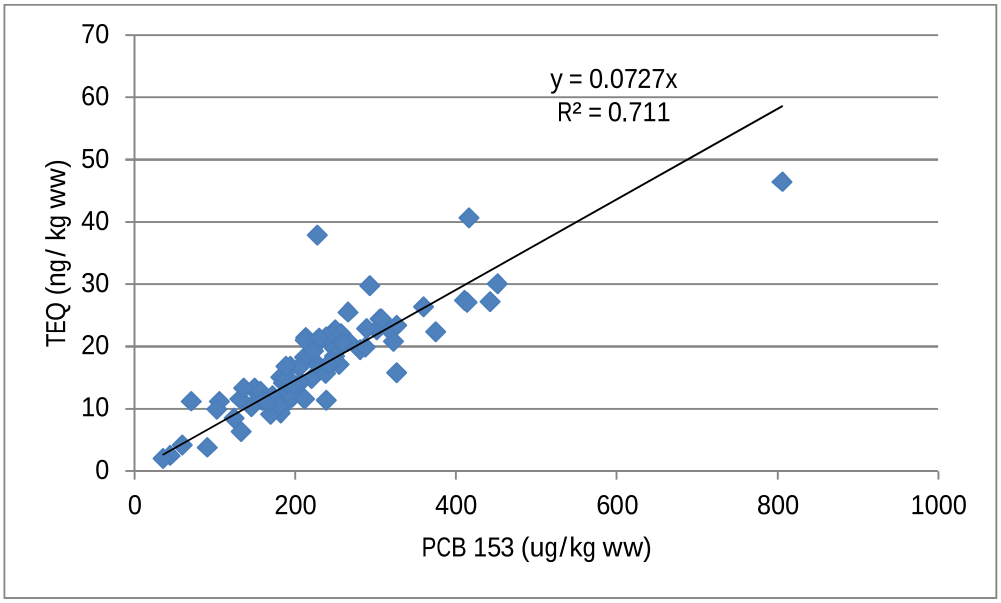


Figure 3.1 Relationship between PCB 153 and total TEQ-concentration in fresh eel as derived by Van Hattum et al. (2013).

Kotterman en Glorius (2011) presented both PCB153 en TEQ contents in eels in a lot of Dutch waters and observed a relationship close to the one found by Van Hattum (2013).

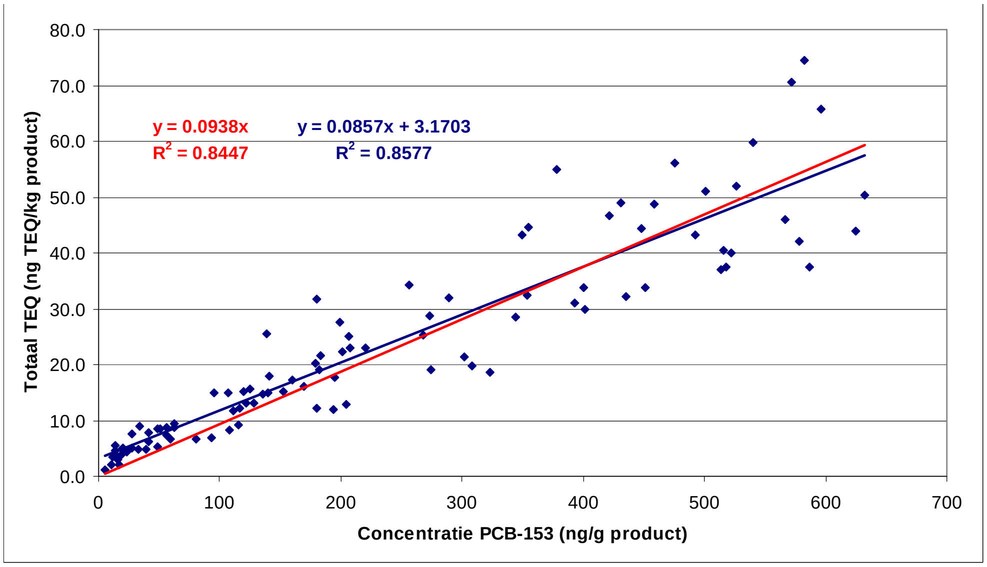


Figure 3.2 Relationship between PCB-153 en total TEQ total TEQ-concentration in fresh eel > 30 cm as derived by Glorius and Kotterman (2011).

If Glorius and Kotterman (2011) also incorporated PCB153 concentrations PCB > 700 ng/g the equation changed into: $y=0.084x$.

Similar observations for the relationship between PCB-153 and total TEQ concentrations in fish were reported for Amer and Haringvliet (Van Hattum et al., 1998) and for the Limfjord area in Denmark (Smit et al., 1996) based on old TEF values derived from literature.

Lopes et al (2012) modelled the uptake of PCB153 by fish and conversion tot TEQ's in the French river Rhone. They modelled a slightly higher uptake for eel: $y=0.132x$. The equation for barbel shows that the equations for fat and low-fat fish does not deviate much: $y=0.114x$.

If we consider all information, we will use one generic equation for all fish based on recent data describing the relationship between PCB153 in fish and total TEQ in fish:

$$\text{total TEQ in fish (ng/kg)} = 0.09 \times \text{PCB153 in fish (ug/kg)} \qquad [3]$$

4 BSAFs

In step number 3 the concentration of PCB153 is converted into the concentration of PCB153 in the sediment organic carbon by using a Biota Sediment Accumulation Factor (BSAF; see equation 1). A literature review revealed a number of studies that measured both PCB153 in fish tissues and sediment. Moreover, for a BASF also the analysis of the lipid fraction in fish and the organic carbon fraction in sediment are needed.

4.1 International studies

Babut et al., (2012) monitored a variety of fish in the River Rhone (FR). They derived BASF values for PCB153 based on a selection of paired samples (fish and sediment) of 10 fish species at 40 sites. The used a statistical method (bootstrapping) to derive a distribution of BSAFs for each site and each fish species. Consequently, they chose the 3rd quartile to derive standards. The median value will be lower (less accumulation in fish), but was not presented.

Roughly 3 groups could be distinguished as shown in Table 4.1. Though they discuss the differences, they cannot explain them very easily. Particularly the low BSAF of carp compared to bream is difficult to explain. They have some evidence that carp does not accumulate in filet fat but more in brain, viscera and mesenteric fat. However, it is questionable whether this physiological property can explain the huge differences.

Table 4.1 3rd quartile BSAFs as determined by Babut et al. (2012) at 40 sites.

Species	BSAF
Carp and chub	<0,01
Roach, Pike perch, and Giant catfish	0.3-0.7
Eel, Barel, Bream, Tench and Trout	1-10

Additional studies, partly mentioned by Babut et al. (2012) also reported BSAF for specific species. Table 4.2 shows the values. They are generally between 1 and 10, for eel, bream and trout more ore les comparable to Babut et al. (2012). The salt water fishes sole and flounder deviate extremely. This has to be clarified. Most of the BSAF data for PCB-153 in fish from the USACE (2015) database are between 1 to 10.

Table 4.2 BSAFs found in other (smaller) studies.

species	Country	BSAF	Original source
Trout	USA (Lake Michigan)	4.5	Burkhard et al., 2004
Largemouth bass	USA (Rhode Island Superfund)	1.8	US EPA, 2007
Eel	Sweden (lake)	12.7	Jahnke et al., 2014
Bream	Germany (3 sites Elbe)	5.7	Schafer et al., 2015
Sole	Belgium (Scheldt)	11368	Van Ael, 2012
Flounder	Belgium (Scheldt)	6761	Van Ael, 2012

4.2 Dutch data

PCBs in biota's have been monitored in de the Netherlands predominantly in eel and mussels. Because the WFD-standard will probably be monitored in fish, we focused on fish. IMARES reported the eel monitoring and also produced a number of trend reports. However they do not monitor sediment quality. Based on Waterbase and available data at Deltares, we searched for PCB153 and organic carbon[2] concentrations in sediment in the relevant area. Appendix A shows the average concentrations in fish for all monitoring locations and the complementary concentrations in sediment if available.

Figure 4.1 shows the calculated BSAFs if there is any data available. However, locations at the left side of the graph have a very limited number of data, locations at the right side have problems with reporting limits in sediment. The locations in the middle are most reliable. We distinguished the contaminated delta rivers from the cleaner areas, resulting in a BSAF of 5 for the river delta, and 2.5 for other area's, like IJsselmeer, and the upper part of the Meuse. This is a rather small difference compared to the variation and uncertainty. Therefore we concluded that we can use one BSAF of 4 for Dutch water.

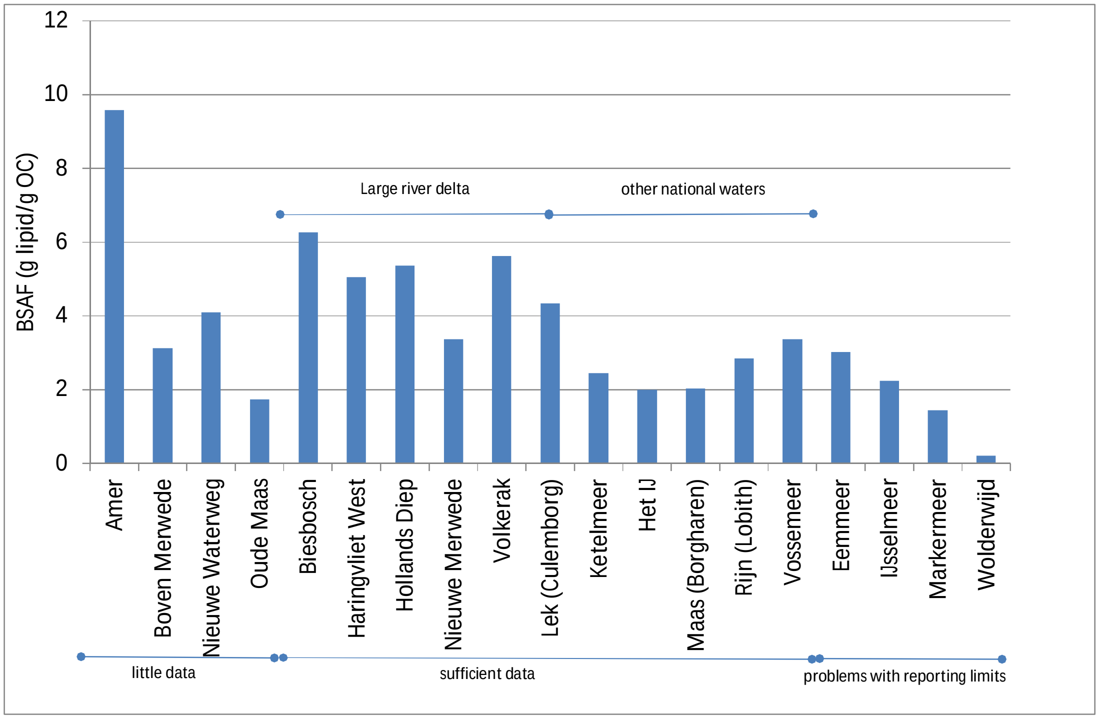


Figure 4.1 Calculated BSAFs in eel in the Dutch national waters.

4.3 Conclusion on BSAFs

The BSAFs found in several studies provide BSAFs in the same range, although there is a large variation within samples. If the samples are averaged, the BSAF mostly results in a value between 1 and 10. The average BSAF of 4 found for eels in the Dutch national waters seems to be an acceptable value. This value will be used in the Sediment discharge test, according to equation 4:

$$BSAF_{SDT} = C_{biota,lipid} / C_{sediment,SOC} = 4 \qquad [4]$$

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[2] *First option: OC. Second option: organic matter: OM / 1.724. Third option: Loss on ignition: (1 - LOI) / 1.724*

5 Conclusions and recommendations

5.1 Conclusions

All calculation steps have been quantified in this report. Figure 2.1 can be replaced by Figure 5.1.

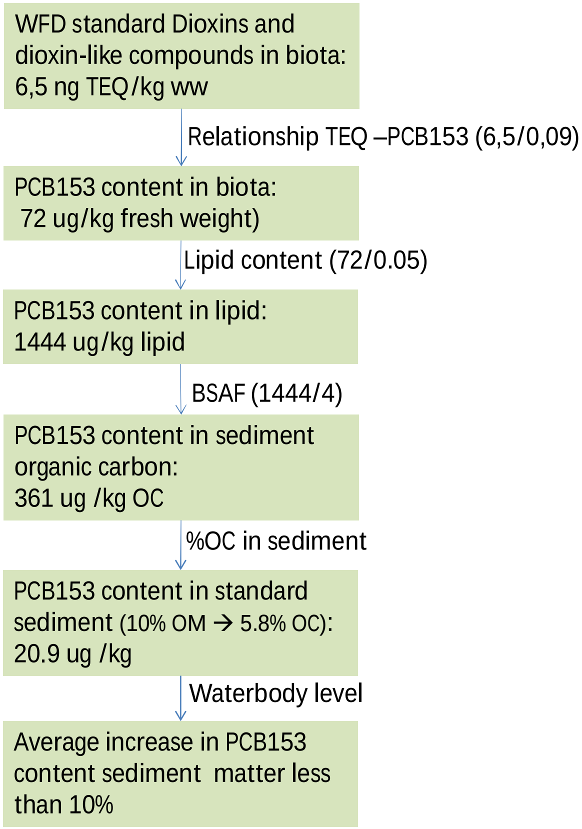

*Figure 5.1 Calculation of the WFD standard for dioxins in biota (6.5 ng TEQ/kg ww) into a standard in sediment (361*f_{OC} ug PCB153/kg dw)*

The last step in Figure 5.1 is only relevant if the standard (20.9 ug/kg) is exceeded. If the increase in PCB153 concentration of the intervention has little effect on the whole water body, it is allowed to exceed the standard. This is tested by equation 2.

5.2 Recommendations

Though the approach of using a BSAF has some fundamental limitations, the results as shown in Figure 5.1 can be improved:

- The pairing of fish and sediment data can be improved by using exact locations where eels were caught and exploring additional databases with respect to sediment data to fill the gaps (see Appendix A). This needs to be done together with Imares. They know the details on the fish samples; Deltares has a lot of knowledge on sediment quality. Data from other studies (e.g. IVM, Alterra, RWS) should be included.
- More experiences with other fish species is needed, particularly if the WFD biota monitoring will focus on other species, probably common roach.
- More information is needed on marine species.

- The TEF system used for calculate the total TEQ concentration from congener-specific PCDD/F and PCB data has shown some changes over time, which may have had an effect of reported PCB-153 to total-TEQ ratios.
- Recent studies investigate the relationship between contaminant concentrations in sediments and in biota by measuring pore water concentrations. They use passive sampling to obtain pore water concentrations. As this approach may become more important in future monitoring, it is recommended to investigate the relationship between PCB-153 form passive sampler data and total TEQ dioxin concentrations in fish.

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A Paired concentrations of PCB 153 in eel and sediment

Vislocatie	No. of eel samples	sed No. of sediment samples	PCB153 in eel (ug/kg lipid)	PCB153 in sediment (ug/kg OC)
Aarkanaal,Ter Aar	2		593	
Amer	3	1	3348	349
Amstel	1		242	
AmsterdamRijnkanaal	2		971	
Biesbosch	5	4	3541	565
Boven Merwede	1	1	1726	552
Dordtsche Kil	1		1580	
Eemmeer	6	7	152	50
Haringvliet Oost	3		2903	
Haringvliet West	18	13	3164	625
Haringvliet-midden	4		2139	
Het IJ	12	10	1401	699
Hollands Diep	19	13	3007	560
Hollandse Ijssel		5		694
Hollandse IJssel t.h.v. Gouderak	1		2081	
IJsselmeer	24	12	319	143
Jan van Riebeeckhaven, Amsterdam	2		810	
Kanaal Gent Terneuzen		9		490
Ketelmeer	21	12	934	381
Lek (Culemborg)	19	5	1945	448
Maas (Borgharen)	12	9	2258	1110
Maas, Eijsden	6		2677	
Maas,Keizersveer	4		2425	
Maasvlakte	1		1374	
Maas-Waal kanaal,Malden	2		3811	
Maas-Waalkanaal,Malden	1		4597	
Markermeer	18	18	234	163
Nieuwe Maas	1		1658	
Nieuwe Merwede	4	3	2668	793
Nieuwe Waterweg	1	1	1243	303
Noord	1		2009	
Noordhollands kanaal,Akersloot	2		187	
Noordzeekanaal	3		1377	
Oosterschelde	2		171	

Oude Maas	1	1	1793	1029
Prinses Margrietkanaal,Suawoude	3		271	
Ramsdiep	3		417	
Rijn (Lobith)	21	7	2332	819
Roer,Vlodrop	3		2553	
Twentekanaal Wiene-Goor	1		484	
Twentekanaal,Hengelo	3		1203	
Vecht,Ommen	3		277	
Volkerak	22	14	1073	190
Vossemeer	2	1	924	273
Waal,Tiel	6		1773	
Wantij		1		326
Wolderwijd	13	11	93	420
Zoommeer		1		47
Zoommeer	3		292	
Gooimeer	1		129	
IJssel, Deventer	7		1642	
Lauwersmeer	2		69	
Loosdrechtse Plassen	1		62	
Maas t.h.v. Roermond	1		3594	
Maas, boven Roermond	1		4549	
Maas, t.h.v. Maasbommel	1		2466	
Sneeker Meer	1		123	
Zwarte Meer, Zwartsluis	1		409	