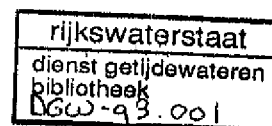




Tidal Waters Division



Concentrations of PAHs, PCBs and heavy metals in the blue mussel, *Mytilus edulis*

DGW-93.001

Project POLWAD/BEON EFFEKT

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Beleidssamenvatting

De enorme zeehondensterfte in de Waddenzee was in september 1988 aanleiding om contaminanten te meten. Hoewel al snel bekend was dat de dieren aan een virusinfectie bezweken waren, werd een verzwakte conditie onder invloed van vervuiling niet uitgesloten. Om een goed beeld te krijgen van de vervuilingstoestand in de Waddenzee werden contaminanten gemeten in mosselen, *Mytilus edulis*. Mosselen staan bekend om hun vermogen veel (giftige) stoffen te accumuleren via het water en hun voedsel. Zelf worden ze gegeten door zeesterren, vissen en vogels, waardoor ze op hun beurt eventueel giftige stoffen doorgeven in de voedselketen.

Op 29 lokaties langs de Nederlandse, Duitse en Deense kusten van de Noordzee en de Waddenzee zijn toen mosselen verzameld om de gehalten aan Polycyclische Aromatische Koolwaterstoffen (PAK's), Polychloor Bifenylen (PCB's) en zware metalen te bepalen.

Schelde sterk vervuild

De mosselen uit Hansweert (locatie 1) in het Schelde estuarium vertoonden de hoogste contaminatie. Voornaamste vervuiling betrof hier cadmium, PCB's en benzo(e)pyreen (BeP), een PAK. De mosselen van Sylt bij de Duits-Deense grens bevatten de laagste gehalten aan contaminanten.

Ten opzichte van de meeste andere Europese estuaria is de situatie voor PCB's en zware metalen in de Westerschelde ernstig.

De gehalten zijn zo hoog dat een herstel van zeehond populaties op korte termijn bemoeilijkt kan worden door effecten op de voortplanting. Ook in de Waddenzee is de vervuiling nog zo hoog, dat hier soortgelijke negatieve effecten op het ecosysteem verwacht kunnen worden. Pas bij de Duits-Deense grens en voor de Deense kust zijn de gehalten aan giftige stoffen aanmerkelijk lager.

Verschillende bronnen

De belasting met PAK's bleek van lokale oorsprong te zijn.

Uitzondering is BeP, dat duidelijk door de rivieren aangevoerd wordt. Vooral in de nabijheid van havens, zoals Den Helder, Den Oever en Lauwersoog, werden hoge concentraties van verschillende PAK's aangetroffen. De hoge concentratie bij Dagebüll, een kleine visserijhaven in Noord-Duitsland, kon niet verklaard worden uit een bekende bron en er is geadviseerd op die lokatie verder onderzoek te doen.

De PCB's zijn afkomstig van de rivieren. Uit de verdelingspatronen van de individuele PCB's blijkt, dat de invloed van de Rijn zich uitstrekt van de monding van de Oosterschelde tot het westelijk deel van de Nederlandse Waddenzee. Het oostelijk deel van de Waddenzee staat o.i.v. de Elbe.

Zowel zink als cadmium komen in de Westerschelde in hoge concentraties voor. Maar ook in het estuarium van de Eems en de uitstroom van de Elbe zijn hoge gehalten van deze metalen in de mosselen aangetroffen. De overige metalen (Pb, Cu, Cr en Ni) kwamen niet in uitzonderlijke concentraties voor.

Summary

The catastrophic reduction of the Wadden Sea seal populations in 1988 was a first motive to investigate the contamination in that area. Although it was established that the animals suffered from a virus infection, a weakened condition caused by pollutants could not be excluded. In order to obtain a good picture of the amount of pollution in the Wadden Sea contaminants were measured in mussels, *Mytilus edulis*. This species is known for its ability to concentrate pollutants and its important role in the ecosystem. Polycyclic Aromatic Hydrocarbons (PAHs), Polychlorinated Biphenyls (PCBs) and heavy metals were analysed in samples, which were collected at twenty-nine locations along the North Sea and Wadden Sea coasts of The Netherlands, Germany and Denmark.

The Scheldt heavily polluted

The samples from location 1, Hansweert in the Scheldt estuary were contaminated the most. The largest contributors to the pollution here were PCBs, cadmium and benz(e)pyrene (BeP). Compared with most other European estuaries the condition in the Scheldt estuary concerning PCB and heavy metal contamination is alarming. The levels are so high that the recovery of seal populations on a short term can be hindered since effects on reproduction can occur. The pollution in the Wadden Sea is still of such a level that similar effects in this ecosystem can be expected.

Different sources

Contamination with PAHs appeared to originate locally. Benz(e)pyrene is an exception. This compound has a riverine origin. High concentrations of different PAHs were found especially in the neighbourhood of (small) harbours like Den Helder, Den Oever and Lauwersoog. It is difficult to explain the extremely high concentration of PAHs at Dagebüll, a small fishery harbour in the north of Germany. Further monitoring at this location is advisable.

The PCBs have a riverine origin. The distribution patterns of the individual PCBs indicate that the influence of the river Rhine stretches from the mouth of the eastern Scheldt to the western part of the Dutch Wadden Sea. The eastern part of the Wadden Sea is contaminated by the river Elbe.

Cadmium as well as zinc occurred in high concentrations in the Scheldt estuary. Also the rivers Elbe and Ems can be recognized as major sources of these metals.

1. Introduction

About 6,000 seals died in the Wadden Sea between April and October 1988, approximately 60% of the population at that time (Reijnders, 1992). Soon, it was established that a virus infection and not an undetected toxic spill, had caused this catastrophic reduction in number (Osterhaus, 1988a, 1988b). However, the reason for the death of this extraordinarily large number was probably due to several factors. Brouwer et al. (1989) pointed out that pollution, by causing immunosuppression could have played a role in the sudden outbreak of a new disease.

A large body information and data on contamination in organisms in the Wadden Sea is available in the literature, but it is difficult to compare the different reports. This is due to the different fields of interest of scientists, different sampling methods, the use of different organisms or fractions of organisms and differences in analytical methods. A sampling program was carried out to obtain a consistent set of data as a part of an attempt to explain the near extinction of the Wadden Sea seals.

In September 1988, samples of mussels, *Mytilus edulis*, were collected along the North Sea and Wadden Sea coasts of Denmark, Germany and The Netherlands, the purpose being to determine the amount of contaminants accumulated in these animals. Biomagnifying pollutants in the Wadden Sea is considered to be a threat for the diversity of the ecosystem and, in particular, for the survival of the common seal, *Phoca vitulina*, the top predator (Reijnders, 1986; Marquenie & Simmers, 1988).

There were several reasons for choosing *M. edulis* to monitor the pollution.

The usefulness of this species satisfying the criteria for biomonitoring has been described many times (Phillips, 1977; Goldberg et al., 1978; Farrington et al., 1983; de Kock, 1986). The most characteristic features of this species are its wide distribution, its ability to concentrate pollutants, its sedentary life-style, its long life span and its ease of collection.

The ecological value of the mussel makes this organism even more relevant as a sample. They feed on algae and so contribute to the clarity of the water. They are predated on by a number of other species and therefore an important step in transferring the algal biomass into higher food chains.

Mussels feed by filtering microscopic particles from the water. To this end, they pump large volumes of water over their gills, and thus are exposed to contaminants dissolved in the water, either bound to particles or present in algae. The biotransformation activity in mussels is low, resulting in accumulation (Livingstone & Farrar, 1984; Stegeman, 1985).

By concentrating contaminants, mussels can be a threat to their predators (eiderducks, seagulls, oystercatchers, flatfish, sea stars, crabs and several mollusca) and contribute to the transfer of pollutants through the ecosystem.

For example, den Besten (1991) demonstrated the effects of PCBs and cadmium contaminated *M. edulis* on the reproduction of one of their predators, the sea star *Asterias rubens*.

The results from this unique inventory were used to make a risk evaluation for mussel predators by comparing the data with recent calculations on maximum permissible risk levels.

2. Material & methods

2.1 Sampling

In September 1988, 29 samples of mussels (*M. edulis*) were collected along the Dutch, German and Danish Wadden Sea and North Sea coasts. The sampling was carried out within two weeks by one team of technicians from the Tidal Waters Division (TWD), using the same protocol and selection criteria. At each location at least 100 mussels (length between 30 and 50 mm) were collected in polythene bags. Immediately after, they were frozen using a transportable freezer (-20°C).

In the laboratory the soft tissue was dissected with titanium knives, homogenized by an Ultra-Thurrax with titanium rotor and frozen in washed glass bottles at -20°C. The samples were freeze-dried and stored at -20°C until further analysis. The analyses were carried out by the same technician, in one batch, to avoid analytical differences between runs.

2.2 Analysis

2.2.1 PAHs

Two grams of freeze-dried homogenate per sample was used in the analysis of PAHs. PAHs were extracted over 4 hours with a hexane/acetone mixture (1:3).

The extracts were evaporated down to 1-2 ml, underwent a clean-up procedure using a column with Florisil and eluted with a hexane/acetone mixture (1:3).

PAHs were further separated on a dried (180°C) SiO₂ column by elution with hexane/diethylether, and the extract was brought quantitatively on a column with 1 g AlO₃ (dried at 180°C). The final extract was injected into a HP 1090 High Performance Liquid Chromatography (HPLC) system consisting of a reversed phase column, type Vydac 201 PB-5 (250 x 4.6 mm) with a methanol/water gradient elution program, coupled to two HP 1046A fluorescence detectors used in series.

The following PAH compounds were measured: Phenanthrene (Fen), Anthracene (Ant), Fluoranthene (Fla), Pyrene (Pyr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(e)pyrene (BeP), Benzo(a)pyrene (BaP), Benzo(g,h,i)perylene (BPe), Dibenzo(a,h)anthracene (DBA), Indeno(1,2,3-c,d)pyrene (InP).

2.2.2 PCBs

The analysis of PCBs was carried out on 1 g of the freeze-dried homogenate. PCBs were extracted over 4 hours with a hexane/acetone mixture (1:3). The extracts were evaporated down to 1-2 ml, underwent a clean-up procedure using a column with Florisil and eluted with a hexane/acetone mixture (1:3). PCBs were further separated on a dried (180°C) SiO₂ column by elution with hexane.

PCBs were measured with a HP 5880 gas chromatograph with two capillary columns and two ECD detectors. The two columns were coated with SE 54 and CP Sil 19CB respectively.

The PCB congeners determined were: CB 18, 31, 28, 52, 49, 44, 101, 118, 153, 105, 138, 187, 180 and 170.

2.2.3 Heavy metals

Heavy metals were analysed according to the standard procedures of the TWD laboratories (Siderius & Koning, 1989). First, the freeze-dried homogenates were broken down with concentrated nitric acid.

Cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni) and lead (Pb) were analysed using Graphite Furnace Atomic Absorption Spectrometry (GF-AAS). Zinc (Zn) was measured by Flame AAS and mercury (Hg) by Cold Vapour AAS.

3. Results

For clarity, the results of PAH, PCB and heavy metal analysis will be treated of separately. The concentrations of the PCBs and PAHs are presented both on an ash-free dry weight basis and a lipid basis. Σ PAH and Σ PCB mean the sum of the compounds measured in this survey (14 PAHs and 13 PCBs, respectively).

Σ PCB is the sum of 7 congeners: 28, 52, 101, 118, 138, 153 and 180. The heavy metal concentrations are expressed as dry weights. The results of the analyses of PAHs, PCBs and heavy metals are summarized in Appendix 1. These also include results expressed as freeze dried weight, dry weight, ash-free dry weight and lipid weight. When data are expressed as wet weights, it is assumed that the fresh weight of the average mussel consisted of 17% dry weight (Smedes pers.comm.).

On the foliate (Appendix 3), a list of location numbers, names of the locations, kilometre distance between the sampling locations and a list of the main points of river outflow is given. All the numbers are indicated on a map (Appendix 2).

3.1 PAHs

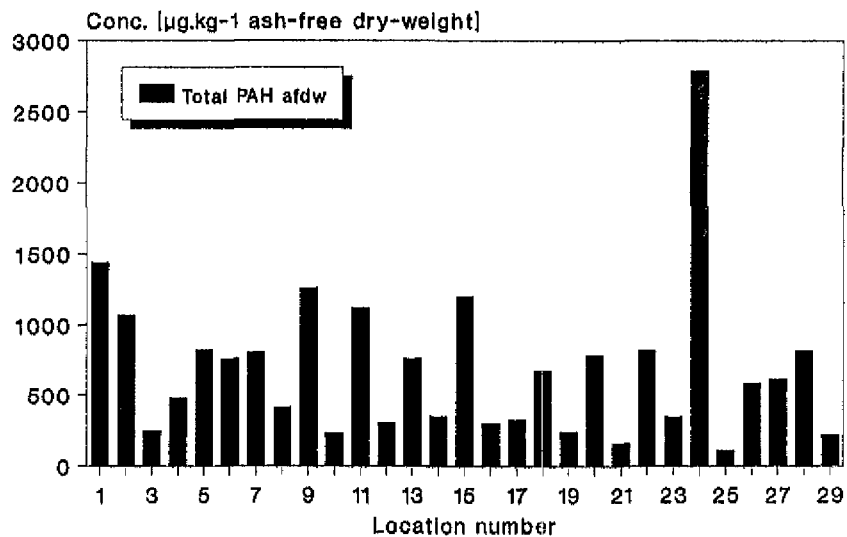
The highest PAH concentration, on an ash-free dry weight basis, is found at Dagebüll (loc.24), which is only 22 kilometres from the location with the lowest concentration, Sylt (loc.25) (Figure 1). Relatively high concentrations are also found at Hansweert (loc.1) and Den Helder (loc.9).

On a lipid basis, the pattern is similar (Figure 2). No value is given for the location Esbjerg (28), because the lipid analysis of that particular sample failed.

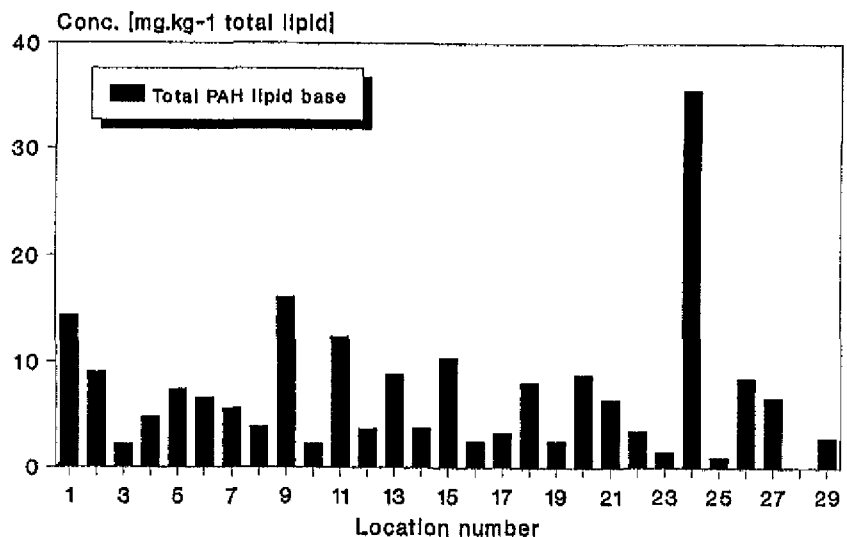
The relative contribution of the individual PAH compounds to the total PAH concentration can differ widely between the various sampling stations, as illustrated, for example, by an analysis of the so-called six of Borneff, (fluoranthene (Fla), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), benzo(g,h,i)perylene (BPe), indeno(1,2,3-c,d)pyrene (InP), and anthracene (Ant) and benzo(e)pyrene (BeP) (Figures 3 to 6). Isomers are placed together where ever possible. The contributions of Fla, Ant and BbF at Dagebüll (loc.24) are high. On the other hand, their contribution to the total at the second highest location, Hansweert (loc.1), is rather low. Here the contributions of BeP and BPe to the total are relatively high. The high levels of Bpe and InP at Kornwernerzand (loc.13) are significant.

Figure 1.

PAH concentrations in *M. edulis* vary over a wide range (two orders of magnitude). The overall PAH contamination does not seem to be related to the output from major rivers (A = Scheldt; B = Rhine; C = Lake IJssel; D = Ems; E = Weser; F = Elbe).

**Figure 2.**

The pattern of PAH concentrations expressed on a lipid base is similar to that on an ash free dry weight base (above). Local contamination and various discharges from shipping activities mask the river output.

**Figure 3**

The pattern of distribution of PAHs varies widely between the locations, mainly according to the variety of local sources. BeP seems to originate from the rivers. An increased BaP level is found at a few locations.

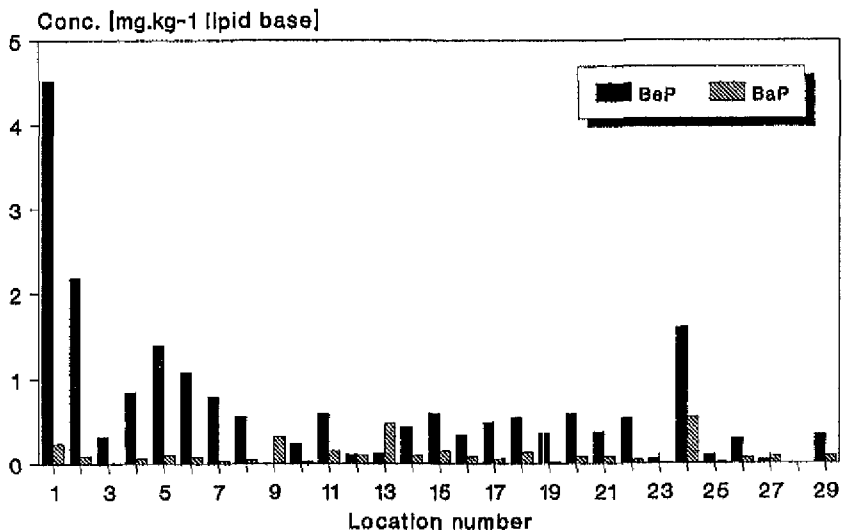
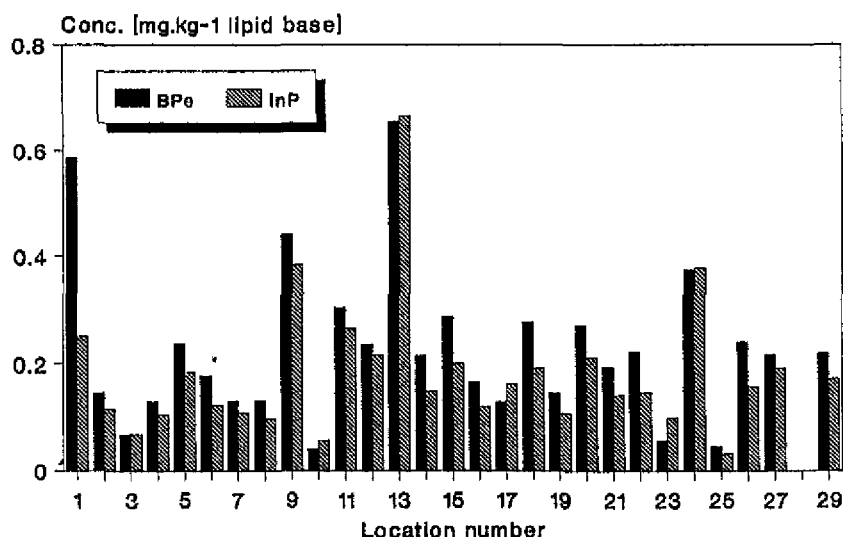
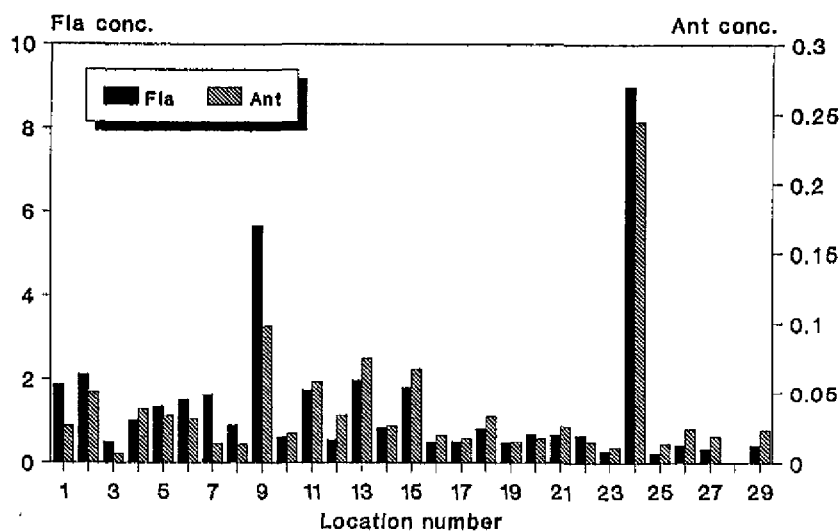


Figure 4

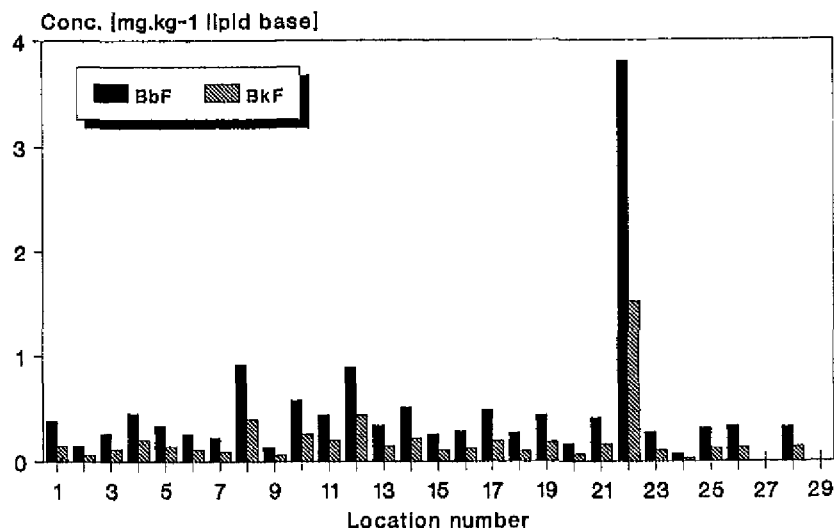
The pattern of distribution of PAHs varies widely between the locations, mainly according to the variety of local sources. The patterns of BPe and InP are similar.

**Figure 5**

The pattern of distribution of PAHs varies widely between the locations, mainly according to the variety of local sources. Fla and Ant show high concentrations at Dagebüll, a small harbour in Germany.

**Figure 6**

The pattern of distribution of PAHs varies widely between the locations, mainly according to the variety of local sources. The concentrations of BbF and BkF in the Elbe outflow at Büsum are much higher than elsewhere.



3.2 PCBs

The total PCB concentration, either on an ash-free dry weight basis or on a lipid basis, shows a regularly decreasing pattern from south to north (Fig 7 & 8). The input from different rivers is clearly visible, especially from the Scheldt at Hansweert (loc.1), the Rhine at Hoek van Holland (loc.5), Lake IJssel at Kornwernerzand (loc.13) and the Elbe at Büsum (loc.22).

Figure 7

In contrast to the PAHs, the PCB pattern in *M. edulis* shows a clear relation with the outflow of major rivers, in particular from the Scheldt (A), the Rhine (B) and the Elbe (F; on lipid basis). In general, the trend decreases from the south to the north.

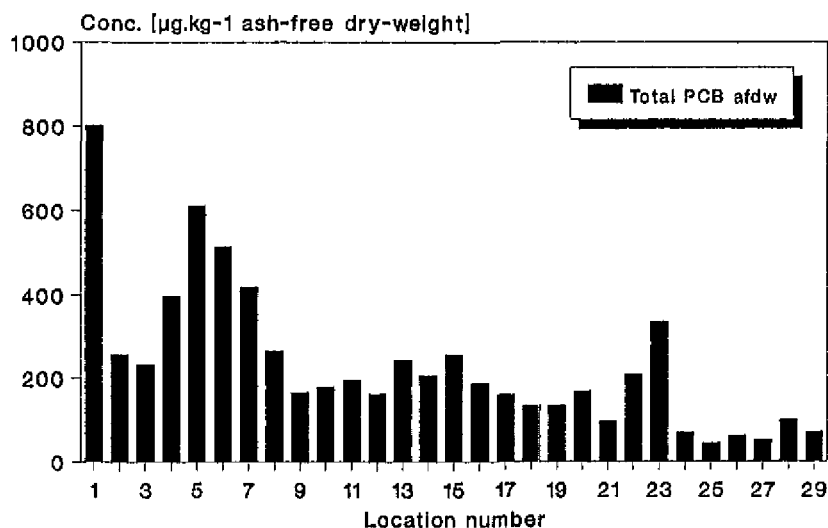


Figure 8

The PCB patterns are almost identical either expressed on an ash-free dry-weight (above) or on a lipid basis (below).

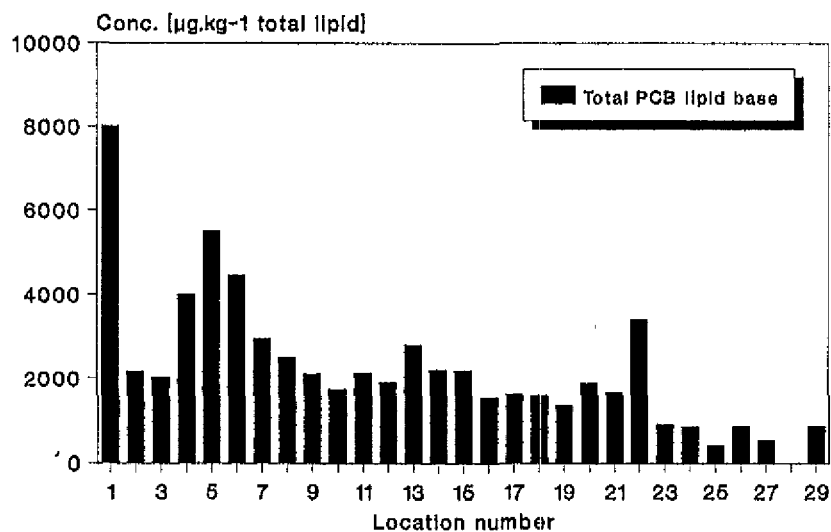
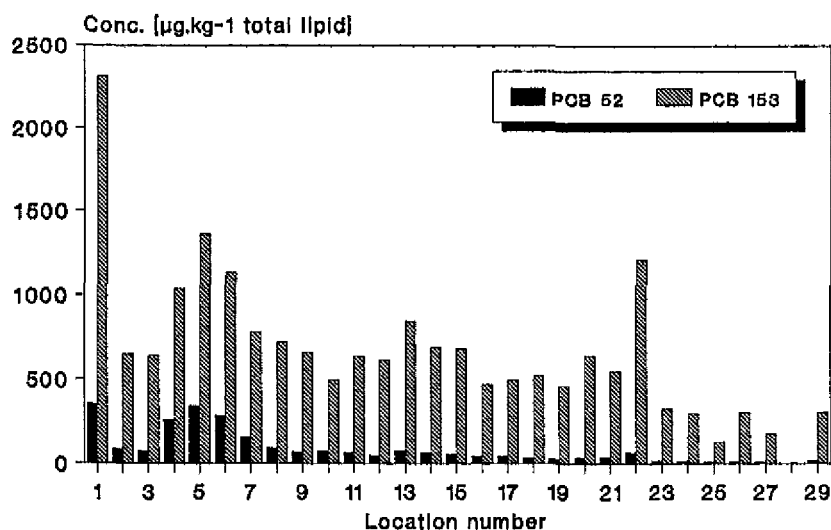
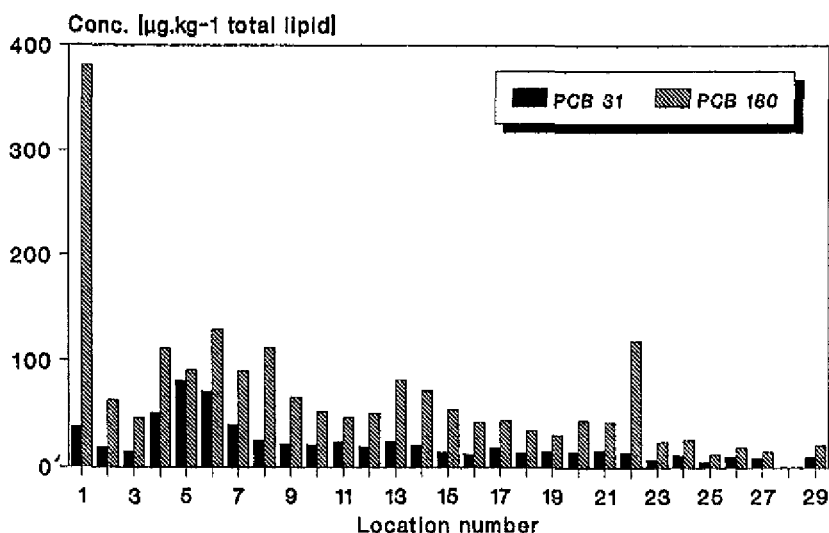


Figure 9

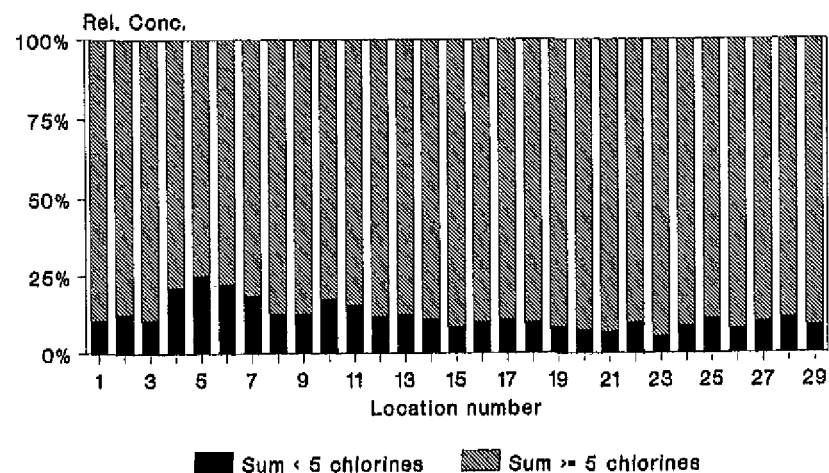
The distribution patterns of the separate congeners are similar to the total PCB pattern and show that there are no major local sources along the North Sea and Wadden Sea coasts. (See also figure 10).

**Figure 10**

A difference in contribution of lower or higher chlorinated congeners to the total PCB concentration can be recognized when comparing the Rhine outflow (loc.5; B) with the others. (See also figure 9).

**Figure 11**

The relation concentrations of lower and higher chlorinated PCBs differ along the sampling stretch. The contribution of lower chlorinated PCBs to the Σ PCBs seems higher in the Rhine outflow. (loc.5; B)



3.3 Heavy metals

The concentration of heavy metals in the mussel, on a dry-weight basis, is illustrated in Figures 12 and 13. Figure 12 shows the concentrations of Cd and Zn. Zn is expressed as 1% of the actual concentration to enable comparison with the Cd concentration. Figure 13 shows the concentrations of Hg and Cu. Cr, Ni and Pb are not presented here. The concentrations of these metals were very close to the detection limits, although higher levels were found at some locations (see below). The highest concentration of heavy metals was found at Hansweert (especially Cd and Zn) (loc.1). At other locations, elevated levels of only one or two metals were found, e.g. a high concentration of Ni at Callantsoog (loc.8), a high concentration of Hg at Breezanddijk (loc.12), high levels of Zn and Cd at Bocht van Watum (loc.18) and Wilhelmshaven (loc.21). Relatively high concentrations of Cr and Ni were present at the western part of the Wadden Sea, from Breezanddijk (loc.12) moving east. The lowest concentrations for all metals were found at Sylt (loc.25).

Figure 12

In general, the concentration of Cd is higher in the eastern part of the Wadden Sea (from Bocht van Watum (loc.18) to the north) than in the western part, probably due to the influence of the Ems (D) and Weser (E). The highest Cd concentration however, is found at Hansweert (loc.1) in the Scheldt estuary (A). The concentration at the end of the estuary, at Vlissingen, (loc.2) is not higher. Most of the Cd seems to accumulate inside the estuary.

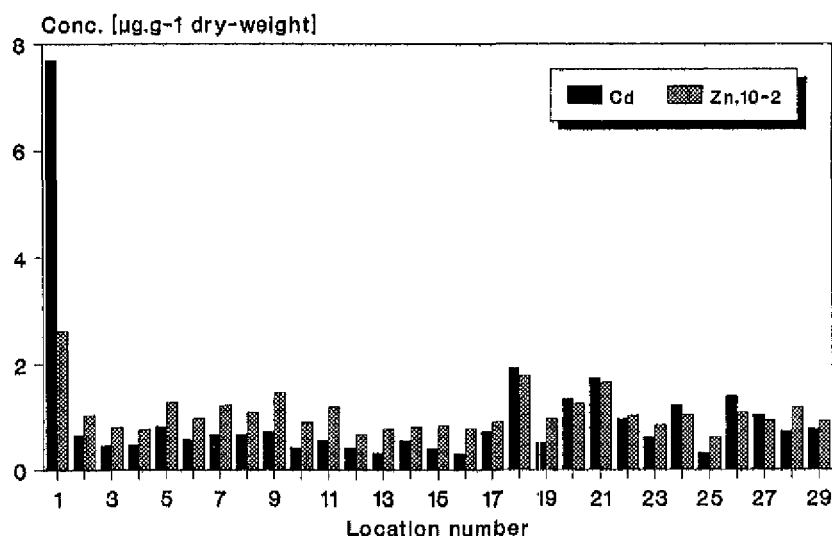
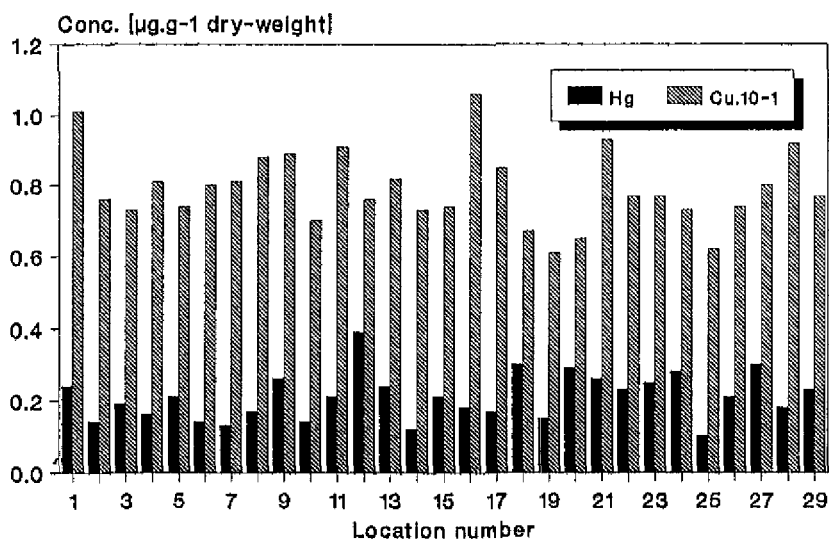


Figure 13

The concentrations of Hg and Cu in *M. edulis* do not vary much along the North Sea and Wadden Sea coasts. Hg exceeds its background level (<0.2 µg/g dry weight) in mussels at a number of locations.



4. Discussion

4.1 PAHs

In general, the PAH concentrations are not clearly influenced by river-related pollution sources. Some compounds, however, for example BeP, show the same distribution pattern as the PCBs and one presumes a riverine influence.

The contribution of the single compounds to the total amount per location differs significantly. Local contamination with specific compounds can be recognized and is probably due to specific wastes.

4.1.1 Mainly local contamination

Klamer et al. (1990a) proposed a procedure to analyse possible sources and the fate of single PAH compounds, based on the method of Duinker et al. (1988) used to analyse characteristics of PCB mixtures in different samples. This method analyses the ratio between the sum of the concentrations of the compounds c.q. congeners determined and their percentage distribution anchored at the overall average value of the single compounds (Figures 14a-d).

The distribution of BeP along the coast (Fig.14a) suggests a riverine source, both from the river Scheldt and the river Rhine. At Hansweert (loc.1), the BeP concentration is relatively high and, taking the percentage distribution into consideration, the river Scheldt is the probable source. Also the river Rhine contributes to the BeP distribution along the Dutch coast.

The percentage distribution for BaP (Fig.14b) shows a totally different picture. One clear BaP pollution can be recognized at Kornwerderzand (loc.13), at the outlet of Lake IJssel, which could be the polluting source. The water from Lake IJssel originates from the river Rhine (B); however, no increase of BaP level is detected at Hoek van Holland (loc.5). The major source for BaP pollution to the water are shipyards, the leaking of tarred ship's coatings and the exhaust of ships (Van Campen et al., 1991). The sluices form the corridor between Lake IJssel and the Wadden Sea. They are used frequently, especially during the summer season by recreational vessels. Increased BaP levels were observed at two other locations, Rottumerplaat and Ho Bugt. There is no indication of polluting sources present near these locations. Rottumerplaat is a nature reserve and access is allowed only with special permission. Closer investigation is recommended here.

The percentage distribution of BPe (Fig.14c) shows a remarkable resemblance to BaP. Except for the three locations mentioned previously, increased levels of BPe are also found at Harlesiel (loc.19) and Nordstrand (loc.23). The high concentration of BPe may be due to the fact that the mussels were collected in a sedimentation area. Under anaerobic conditions, BPe may be formed through the reduction of chinones, sedimented residues from dead algae and bacteria (Neff, 1979). The high concentration of BPe at Kornwerderzand (loc.13) may be explained by the same mechanism. Drainage sluices are situated in this area.

The draining-off of freshwater from Lake IJssel kills algae, bacteria and several other organisms and causes heavy sedimentation of dead organic matter in the vicinity of the sluices (Vethaak, 1990).

The main sources of Fla (fig.14d) seem to be located at the harbours of IJmuiden (loc.7), Den Helder (loc.9) and Dagebüll (loc.24). The riverine influence for this compound is negligible. The increase of Fla seems to be related to specific harbour activities which are not found in the harbours connected to the rivers Scheldt, Rhine, Ems and Elbe. Intensive use of creosoted material could play an important role here.

4.1.2 Unexplained high levels

The extremely high concentrations of PAHs at Dagebüll (loc.24) cannot be explained easily. It is a small harbour with low activity. The mussels were sampled at the south-west point of the harbour, 1.5m below the high-water mark. The sediment at the foot of the dyke consisted of medium-grained sand, which is generally not known for a great binding capacity of organic compounds, although Klamer et al. (1990b) reported that up to 58% of the total PAH can be present in the $>63\mu\text{m}$ fraction. However, this still cannot account for the very high PAH concentration in the mussels from that location. It is most probably due to a local source for creosoted wood used in the harbour.

The high concentrations of fluoranthene and anthracene also makes this more likely (van Campen et al., 1991). The relatively high concentrations of the heavier PAHs (InP, BPE and BeP) indicate that leakage from ship coatings could play a role. Additional research at this location is required.

Figure 14a

Percentage distribution of benz(e)pyrene (BeP) in *M. edulis* samples per location. A riverine influence of the Scheldt (A) and the Rhine (B) is clearly visible. This is not easily recognizable in the outflows of the other big rivers.

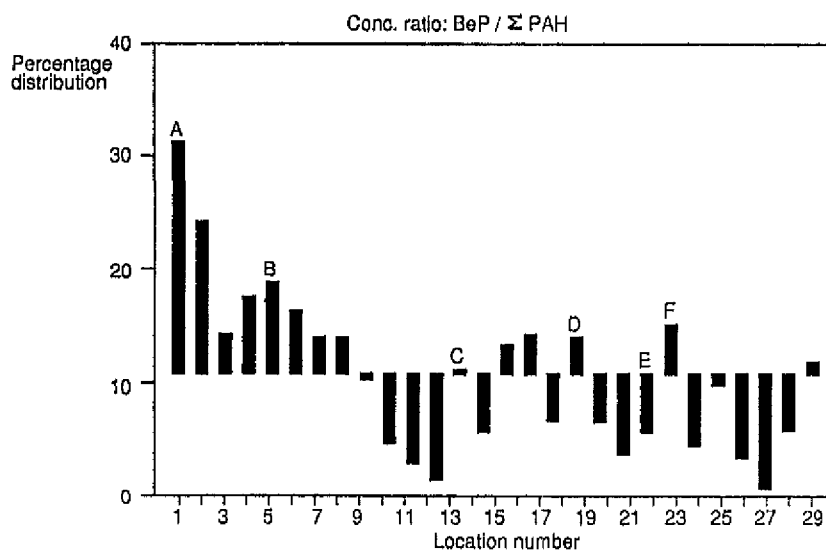
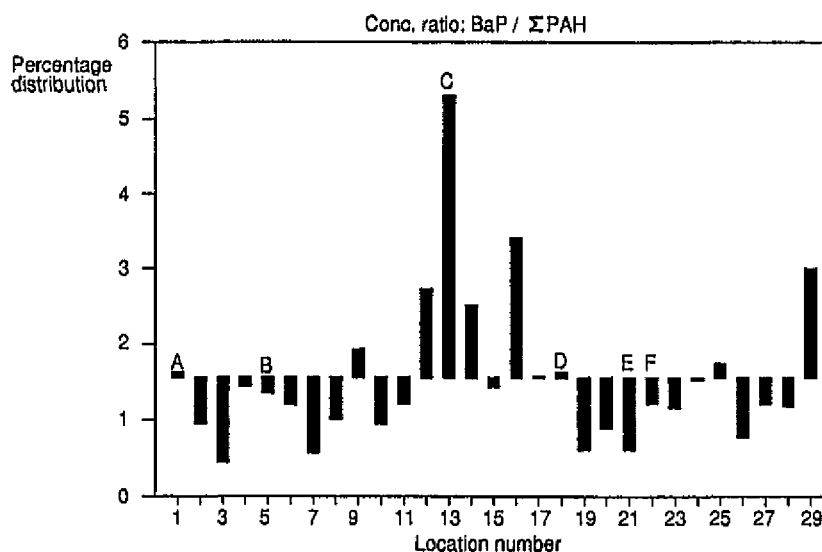
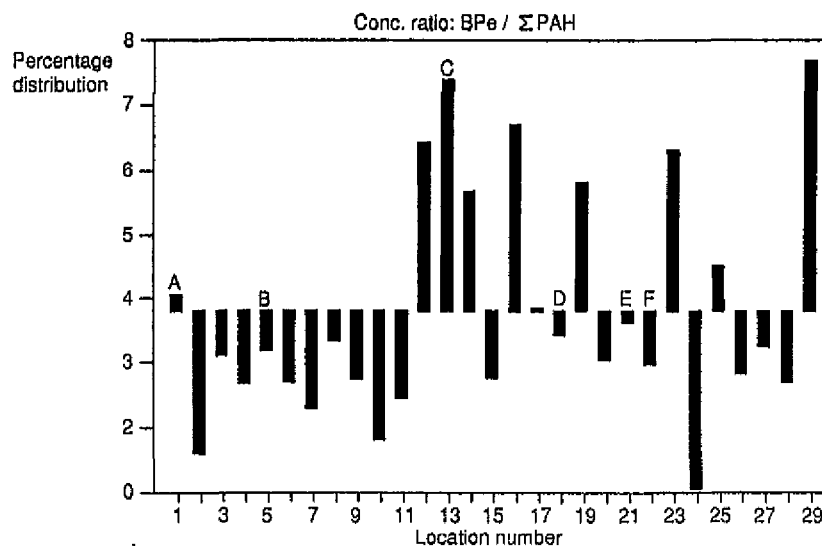


Figure 14b

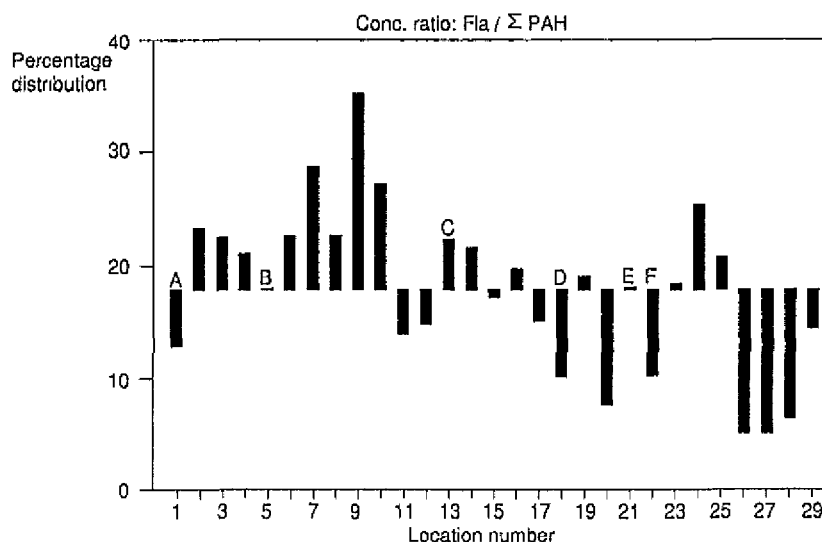
The percentage distribution of benz(a)pyrene (BaP) in *M. edulis* shows a remarkable threefold increase at Kornwerderzand (loc.13), the outflow of Lake IJssel (C). The use of the sluices present here is high, which could contribute to the relatively high BaP concentration.

**Figure 14c**

The percentage distribution of Benzo(g,h,i)perylene (BPe) in *M. edulis* samples has a very local character. The increased occurrence at some of the locations could be explained by BPe formation through reduction of chinones.

**Figure 14d**

There is no clear explanation for the increased levels of fluoranthene (Fla) in *M. edulis* samples. It could be due the use of creosoted material.



4.2 PCBs

4.2.1 Streams of PCBs

The distribution of the Σ PCBs in *M. edulis* along the sampling points shows a clear riverine influence. At Hansweert (loc.1), Hoek van Holland (loc.5), Korwernerzand (loc.13) and Büsum (loc.22) we find peaks, which differ by a factor 5 to 17 from the lowest values at Sylt (loc.25). These locations are situated in the estuaries of the rivers Scheldt, Rhine, Lake IJssel and Elbe, respectively. The output of contaminated freshwater seems to be the major contribution to PCB exposure to marine organisms in the coastal zone of the North Sea and Wadden Sea.

Compared with other data from the coastal waters in Europe, Hansweert (loc.1) in the Scheldt estuary shows a high Σ_7 PCB concentration. The Σ_7 PCB concentration found in this survey lies in the range from 330-6560 $\mu\text{g/kg}$ (Sylt-Hansweert) on a lipid basis. Granby (1987) reported PCB concentrations in *M. edulis* from 180-1190 $\mu\text{g/kg}$ (lipid basis) along the East coast of Denmark. Only one sampling point near a refuse tip showed a high concentration of 16,000 $\mu\text{g/kg}$. In the estuary of the rivers Weser and Jade, ICES (1985/1990) reported concentrations of 1167-3900 $\mu\text{g/kg}$ (lipid basis) (Σ_7 PCB).

4.2.2 Different sources, different effects?

A comparison of the distribution of congeners originating from the various rivers makes clear that there is a difference between the sources.

In the outflow of the river Rhine (Hoek van Holland: loc.5), 25% of the measured PCBs belongs to the lower chlorinated PCBs, while in the Scheldt estuary (Hansweert: loc.1) as well as in the Weser and Elbe estuaries only 11% of the Σ PCB belongs to that group. These values may be compared with concentrations in sediment samples from more or less the same locations (1 and 5) in the Scheldt and Rhine estuaries (Akkerman et al., 1992) (Table 1, Figure 11).

Table 1

There is a clear difference in the percentage of lower chlorinated PCBs between the estuaries, while there is no difference between the sediment and *M. edulis* in the same estuary. The latter is in accordance with the low metabolic rate of mussels.

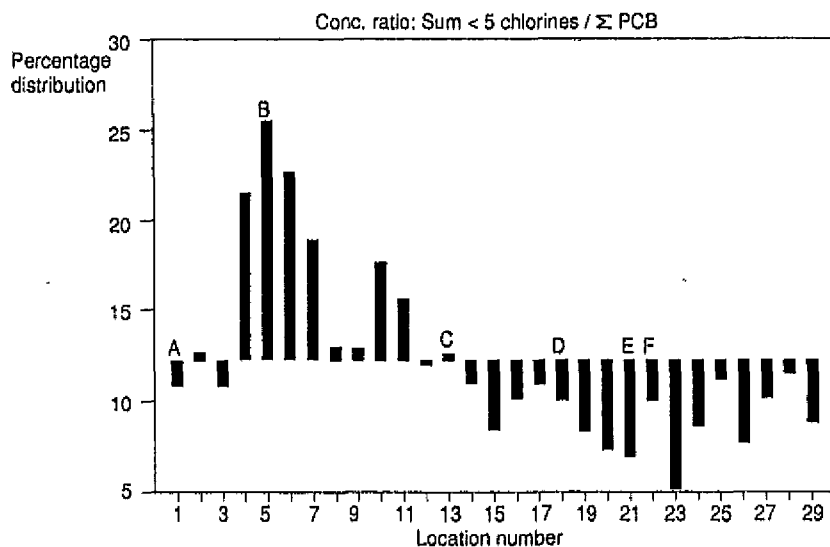
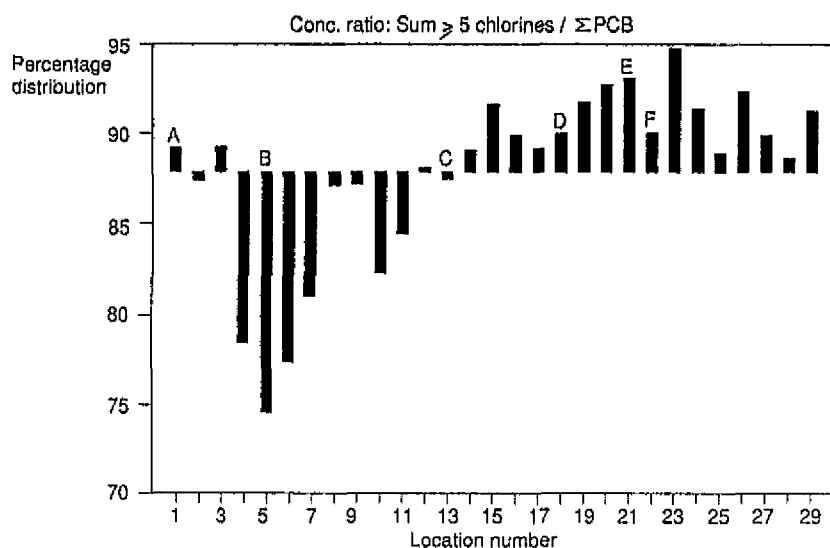
	Hansweert	Hoek van Holland	
Cl<5	11%	22%	sediment
	11%	25%	<i>M. edulis</i>
Cl%	89%	78%	sediment
	89%	75%	<i>M. edulis</i>

It can be concluded that there is a difference in composition of the PCB-mixtures coming from these rivers.

This difference in composition is illustrated clearly by the same method as mentioned above (Duinker et al., 1988; Klammer et al., 1990a) (Figures 15a and 15b). A percentage distribution is given for PCB congeners with less than 5 chlorine atoms (Fig.15a) and for PCB congeners with 5 or more chlorine atoms (Fig.15b). It is shown that the River Rhine is a major source of lower CBs.

Figure 15a-b

The percentage distribution of the lower chlorinated PCBs shows that the Rhine is a definite source, while the Scheldt, Ems and Elbe carry much more higher chlorinated PCBs.

**Figuur 15b**

Van Zoest & van Eck (1990) have reported differences for PCB content in particulate matter between the upper Scheldt estuary (loc.1) and the lower estuary, Vlissingen (loc.2). They suggest that the most plausible reason for this may be the occurrence of discharges of PCB mixtures enriched in lesschlorinated congeners in the lower estuary. It could be that the shift is partly due to the influence of the Rhine outflow (Fig.15a).

The source for the higher CBs is also clear. Relatively more higher chlorinated PCBs originate from the Elbe and Weser estuary, although the sample taken nearest to the outflow of the river Elbe does not show the highest value. In general, higher chlorinated congeners are more toxic than the lower ones (de Voogt et al., 1990). The differences between the sources could result in a relatively bigger effect in the Scheldt (A) estuary and the eastern part of the Wadden Sea (D, E, F).

4.2.3 No improvement after the ban

The concentrations of PCBs found in this survey do not differ from concentrations measured the last decennium in Dutch coastal waters (Anonymous, 1987; Stutterheim & Zevenboom, 1988; Akkerman et al., 1989; Akkerman et al., 1992).

This is in accordance with conclusions of Stronkhorst (1992). He presented a statistical analysis of temporal trends of PCB data in the Scheldt estuary and the Ems Dollard estuary over the period 1985-1990. He finds, in spite of the international agreements on the banning of PCBs, no indications of an improvement in PCB levels in *M. edulis*. This is in agreement with findings on other trophic levels (Norstrom et al., 1988) and estimations of several authors. Bletchly (1984) estimated that the disposal of PCBs used in older transformers and capacitors will reach its peak in this decade and Tanabe (1988) estimated that about 780×10^3 tons of PCBs (70% of the total world production) are still in use in older electrical equipment and other products.

4.2.4 Lack of information on most toxic PCBs

Recently a report of the Directorate-General for the Environment and the Directorate-General for Public Works and Water Management was published concerning derivations of micropollutant risk levels for the North Sea and Wadden Sea (Jonkers & Everts, 1992). Here Maximum permissible Risk Levels (MRL) of several compounds have been calculated for seabirds, based on bird diet data, via the water-mussel-seabird route (Everts et al., 1992). The MRL of a substance is defined as the concentration at which 95% of all the species potentially present in the ecosystem are afforded complete protection, in the sense of their chronic No Observed Effect Concentration (NOEC) not being exceeded. For PCB153 the authors find a MRL of $146 \mu\text{g/kg}$ wet weight in the mussel. A comparison is made with the concentrations of PCB153 found in *M. edulis* at locations in different estuaries sampled in this survey (Table 2).

Table 2

The proposed MRL for PCB153 is not exceeded at any location. That effects of PCBs in the ecosystem are still being reported implies an underestimation of PCB toxicity. This is probably caused by not taking into account the toxicity of coplanar PCBs and the joint toxic action of micropollutants in general.

Location	Conc.PCB153	MRL(mussel)	Estuary
Hansweert (1)	34 $\mu\text{g/kg w.w.}$	146 $\mu\text{g/kg w.w.}$	Scheldt
Hoek v. Holland (5)	22	146	Rhine
Bocht v. Watum (18)	6	146	Ems
Wilhelmshaven (21)	10	146	Weser
Büsum (22)	17	146	Elbe

The concentration of PCB153 in *M. edulis* found in this survey does not exceed the MRL based on secondary poisoning for sea birds. In contrast with this Reijnders & Duinker (1991, in Jonkers & Everts 1992) conclude based on extensive research that PCB levels in the Wadden Sea should be reduced to one fifth of their present concentration, to ensure normal reproduction in seals.

Taking the following into account they are probably have a point. There have been no real changes in PCB concentrations the last decennium (Stronkhorst, 1992), but effects of pollution have been reported (Reijnders, 1986; Marquenie & Simmers, 1988; Brouwer et al., 1989). The MLR of PCBs, which is not exceeded at any location, is based on congener PCB153. This congener, or any other, is not representative the toxicity of all PCBs, since all 209 possible congeners

have different toxic and biologic responses. The intrinsic toxicity of PCBs results from the planar PCBs (the non-ortho PCBs) and some of their mono-ortho substituted congeners (Tanabe, 1988; De Voogt et al., 1990). PCB153 does not belong to this group, this leading to a major underestimation of the possible toxicity. More research is required into the reasons concerning the toxicity of selective PCB congeners. Joint toxic action of PCBs and other micropollutants is also a reason for underestimation in the MRL approach (Everts et al., 1992). Joint toxic action should also be an important part of future research.

4.3 Heavy metals

4.3.1 Cadmium: the Scheldt is a major source

Mussels at Hansweert (loc.1) show high concentrations of all metals measured compared to the other locations, while mussels at Sylt (loc.25) show very low concentrations for all metals. The concentration of Cd at Hansweert is a factor 25 higher on a dry weight base. The Cd concentration at Hansweert (7.69 mg/kg dry wt.) appeared to be about one order of magnitude higher than that in other areas of Europe. Coimbra et al. (1991) noted concentrations one order of magnitude lower (0.65-0.86 mg/kg dry wt.) in *M. edulis* from the urban and industrial area at the northern coast of Portugal. In *Mytilus galloprovincialis* from the Italian coastal waters, the highest concentration found was 2 mg/kg dry wt. (Giordano et al., 1991). Broman et al. (1991), collecting *M. edulis* on the coast of the southern Bothnian Sea, showed a similar Cd concentration range to those in Hansweert (loc.1) (5.1-9.9 mg/kg dry wt.). In an ecotoxicological study focusing on seasonal variation of metal content in *M. edulis*, Amiard et al. (1986) reported Cd concentrations one order of magnitude lower than the highest concentration found in this study. Their study was carried out in a relatively clean area devoted mainly to oyster culture. Their results can be compared with almost the whole range of locations from this study. Besides Hansweert, only two other locations, Bocht van Watum (loc.18) and Wilhelmshaven (loc.21), show slightly increased levels of Cd. These three locations are situated in the (neighbourhood of) the estuaries of the rivers Scheldt, Ems and Weser/Elbe, respectively. In general, the concentrations of Zn showed little variation (factor 2) between the sampling locations. However, the sample from Hansweert (loc.1) was again slightly higher, as were also the mussels from Bocht van Watum (loc.18) and Wilhelmshaven (loc.21). The similarity between the occurrence of Zn and Cd is obvious and probably due to their chemical relationship and related industrial disposal (Phillips, 1976). The big European rivers are the main source for these two metals.

4.3.2 Background levels

The other metals analysed in this study (Cu, Cr, Ni and Pb) hardly varied between the sample points, except Hg which differs a factor 3 between sampling points. A comparison of all metal concentrations found in this survey with recently published background values of heavy metals in mussels (Knutzen & Skei, 1992; Laane et al., 1992) shows that the proposed background values are only exceeded slightly at a few locations (Table 3). The concentrations found are significantly ($p < 0.05$) higher than the background levels, when the difference of

both is bigger than 3x the standard deviation of the analyses. This could either mean that heavy metal contamination does not exist along the North Sea and Wadden Sea coasts of The Netherlands, Germany and Denmark or that the proposed background levels have been set at too high a level. Heavy metal concentrations in *M. edulis*, measured as a routine in the Dutch 'Musselwatch' 1989-1990 point in that direction (Smedes, 1991). A reconsideration of the proposed background levels based on these measurements would be advisable. It is a remarkable fact that the metal concentrations at Hansweert almost all exceed the background values.

Table 3

The number of locations where the metal concentrations are significantly ($p < 0.05$) higher than the proposed background levels is small. Five out of ten exceedings concern Hansweert (loc.1).

background in $\mu\text{g/g}$ d.w.	Cd <2	Zn <200	Hg <0,2	Pb <5	Cu <10	Cr <3	Ni <5
number of locations exceeded	1	1	5	1	0	0	2

4.3.3 Metals weighed in risk evaluation

Comparing the data from this study with MRLs of Cd and Hg for sea birds proposed by Jonkers & Everts (1992), it is evident that the heavy metal content in *M. edulis* in the estuaries is higher than those considered to be safe for predators of mussels (Table 4). Cd is a factor 10-100 higher than the MRL and Hg is a factor 4-7 higher. Even the lowest concentrations found at Sylt (loc.25) are higher than the MRLs based on secondary poisoning. This is in contradiction with the proposed background levels, which either are not or slightly exceeded. A discussion on this subject would be appropriate.

Table 4

At all locations the proposed MRLs for Cd and Hg are exceeded. Only the locations near the estuaries are shown. The assumption was made that on the average mussel fresh weight consisted of 17% dry weight.

Location	Conc.Cd	MRL (mussel)	Estuary
Hansweert (1)	1307 $\mu\text{g/kg}$ w.w.	12.0 $\mu\text{g/kg}$ w.w.	Scheldt
Hoek v. Holland (5)	139	12.0	Rhine
Bocht v. Watum (18)	326	12.0	Ems
Wilhelmshaven (21)	296	12.0	Weser
Büsum (22)	162	12.0	Elbe
Location	Conc.Hg	MRL (mussel)	Estuary
Hansweert (1)	40.8 $\mu\text{g/kg}$ w.w.	7.0 $\mu\text{g/kg}$ w.w.	Scheldt
Hoek v. Holland (5)	35.7	7.0	Rhine
Bocht v. Watum (18)	51.0	7.0	Ems
Wilhelmshaven (21)	44.2	7.0	Weser
Büsum (22)	39.1	7.0	Elbe

The level of contamination in the estuaries give cause for concern since they are important feeding areas for a large number of bird species, especially the very high concentration of Cd in the Scheldt estuary. A known predator of the mussel, the oyster catcher *Haematopus ostralegus*, is declining in numbers at this location since 1979 (Stuart et al. 1990). The contamination with organic pollutants and heavy metals having been suggested as an important factor (Werkgroep Waterbeheer Westerschelde 1989). A better waste water control in this area is required, to ensure better conditions for a healthy ecosystem.

5. Conclusions & Recommendations

- The highest degree of contamination (with PCBs, heavy metals and several PAH compounds) in mussels between the Scheldt and Esbjerg was observed in the Scheldt estuary.
- The western and middle part of the Wadden Sea (Dutch and German Wadden Sea) is highly contaminated compared to the north-eastern part (Danish Wadden Sea).
The contamination originates from the rivers Rhine, Ems, Weser and Elbe.
- Most of the contamination with PAHs along the coast from the south of The Netherlands to the south of Denmark is local in origin. BeP is an exception and seems to arise mainly from the Rhine.
- Additional confirmation and tracing of the source regarding the extremely high PAH concentration in the harbour of Dagebüll (northern Germany) is required.
- PCB contamination has a riverine origin. The outflow of the river Rhine contains relatively more lower-chlorinated biphenyls than the other rivers.
- PCB data from this survey do not exceed recently proposed Maximum permissible Risk Level (MRL) for PCB153. Regarding the present knowledge of PCB-toxicity, the MRL for PCBs should be reconsidered, taking planar PCB-toxicity and the joint toxic action of micropollutants into account.
- Heavy metal contamination is found mostly in the estuaries of the rivers Scheldt, Elbe, Ems and Rhine. The high concentration of cadmium in the Scheldt estuary is a special reason for concern. Measures for the disposal of waste water containing heavy metals should be enforced.
- The heavy metal concentrations in this study exceed recently proposed background levels slightly at a few locations. Reconsideration of the background levels based on this study and other data is advised.
- Cadmium and mercury concentrations exceed recently proposed Maximum permissible Risk Levels (MRLs) at all locations. As opposed to this, recently proposed background levels in mussels are either not or only slightly exceeded.

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Appendix 1

Table 5

PAHs in $\mu\text{g/kg}$ on a freeze-dried base

Location	KM(0 = Vlis)	Fen	Ant	Flu	Pyr	BaA	Chr	BeP	BbF	BkF	Bap	BghiPe	DBahA	InP
Hansweert	-30	70	2	155	210	67	107	376	87	34	20	49	6	21
Vlissingen	0	97	5	217	166	49	80	224	39	15	9	15	0	12
Colynsmaat	29.9	48	1	48	32	6	14	30	14	6	1	7	0	7
Brouwersdam	54.05	47	3	88	89	21	35	73	22	10	6	11	0	9
Hoek v. Holland	83.7	83	3	123	126	40	70	128	42	19	9	22	0	17
Delfl. Hoofden	97.2	85	3	141	124	30	56	102	32	13	8	17	0	12
Ijmuiden	144.9	92	2	192	136	26	52	93	30	13	4	15	0	13
Callantsoog	187.2	62	1	74	57	13	25	46	19	8	3	11	0	8
Den Helder	201.5	171	6	361	224	50	50	0	59	25	20	28	6	25
Texel	208.5	31	2	52	33	13	13	20	12	5	2	3	1	5
Den Oever	220.4	441	4	121	81	30	39	40	40	18	11	21	3	19
Breezanddijk	235.6	44	2	38	31	29	20	8	31	14	7	17	3	15
Kornwernerzand	245.7	62	5	144	106	20	61	9	65	32	34	48	8	48
Piet Scheveplaat	300	33	2	62	49	18	20	32	25	11	7	16	0	11
Lauwersoog	318.3	409	6	161	106	33	39	53	46	19	13	26	3	18
Rottumerplaat	339	25	2	49	38	11	16	33	25	10	8	17	2	12
Blind Ranselgat	361.5	64	1	42	40	13	17	40	23	10	4	11	0	14
Bocht van Watum	373.5	267	2	54	44	25	21	36	32	13	9	18	0	13
Harlesiel	423	30	1	38	31	7	11	28	21	8	1	12	3	9
Mundahn	440.6	365	1	47	42	20	17	41	30	13	6	19	1	15
Wilhelmhaven	454	350	2	63	52	18	17	35	26	10	8	18	1	13
Büsum	506	32	1	52	48	10	17	42	32	13	3	18	0	12
Nordstrand	536	30	1	23	21	6	7	5	13	5	1	5	1	8
Dagebüll	568	349	15	544	221	302	215	97	230	92	33	23	0	23
Sylt	590	18	1	19	14	9	5	9	7	3	2	4	0	3
Koldby	608.8	336	1	23	19	12	11	16	17	7	4	13	0	9
Rømø	620	314	1	23	18	18	11	3	23	9	6	15	0	13
Esbjerg	655.2	391	3	40	43	17	22	36	26	10	7	17	0	11
Ho Bugt	669.6	21	1	27	25	14	13	22	21	9	6	14	0	11

Tabel 6

PAHs in µg/kg on a dry-weight base

Location	KM(O = Vlis)	Fen	Ant	Flu	Pyr	BaA	Chr	BeP	BbF	BkF	Bap	BghiPe	DBahA	InP
Hansweert	-30	72	2.2	160	217	69.3	110.0	388.3	90.0	35.2	20.2	50.4	5.8	21.6
Vlissingen	0	100	5.3	222	170	50.3	81.5	229.5	39.9	15.5	9.1	15.3	0.0	12.2
Colynsplaats	29.9	49	0.6	49	32	5.7	14.5	30.6	14.2	5.8	1.0	6.7	0.0	6.7
Brouwersdam	54.05	48	3.4	90	91	21.8	36.3	74.4	23.0	10.6	6.2	11.4	0.0	9.3
Hoek v. Holland	83.7	86	3.2	127	131	41.1	73.2	132.7	43.3	19.3	9.6	22.6	0.0	17.7
Deffl. Hoofden	97.2	89	3.0	147	130	31.7	58.3	106.0	33.3	14.1	7.9	17.6	0.0	12.1
IJmuiden	144.9	97	1.7	203	144	27.7	55.6	99.1	31.9	14.2	4.0	16.2	0.0	13.6
Callantsoog	187.2	64	1.2	77	60	13.3	25.8	47.6	19.6	8.2	3.4	11.4	0.0	8.6
Den Helder	201.5	180	6.6	380	236	52.4	52.7		62.0	26.8	20.8	29.7	5.8	25.9
Texel	208.5	33	1.9	55	35	13.5	13.7	20.8	12.2	5.2	1.9	3.7	1.2	5.1
Den Oever	220.4	472	4.3	129	86	31.8	41.4	43.3	43.0	19.0	11.3	22.8	3.0	19.8
Breezanddijk	235.6	46	2.5	40	32	30.1	20.5	7.9	32.3	14.4	7.4	17.3	2.9	15.8
Kornwerperzand	245.7	64	5.6	148	110	20.2	63.1	9.4	66.7	32.7	35.2	49.0	8.7	49.8
Piet Scheveplaat	300	35	2.1	65	52	19.2	20.4	33.4	26.4	11.6	7.6	17.0	0.0	11.8
Lauwersoog	318.3	433	6.4	171	112	34.6	41.5	55.8	48.7	20.5	14.3	27.3	3.5	19.1
Rottumerplaat	339	26	2.1	52	41	11.6	16.8	35.0	26.3	11.1	8.9	17.6	1.8	12.7
Blind Ranselgat	361.5	66	1.5	44	41	13.5	17.8	41.2	24.2	10.6	4.6	11.1	0.0	14.0
Bocht van Watum	373.5	277	2.2	56	46	26.1	21.8	36.8	33.1	13.1	9.0	19.0	0.0	13.2
Harlesiel	423	32	1.3	40	32	6.9	11.9	29.0	22.0	8.0	1.3	12.0	2.9	8.9
Mundahn	440.6	382	1.2	49	44	21.0	18.1	42.5	31.5	13.3	5.8	19.6	1.3	15.4
Wilhelmhaven	454	378	2.6	68	56	19.8	18.5	37.5	27.8	10.8	8.2	19.7	1.3	14.3
Büsum	506	34	1.2	54	50	10.6	17.4	44.1	33.8	13.3	3.4	18.4	0.0	12.2
Nordstrand	536	32	1.0	24	22	6.1	7.7	4.9	13.5	5.4	0.8	4.8	1.4	8.4
Dagebüll	568	368	15.6	574	233	318.9	226.9	102.1	242.9	96.8	34.6	24.0	0.0	24.2
Sylt	590	19	1.3	20	14	9.0	4.7	9.5	6.8	2.7	1.7	4.3	0.0	3.1
Koldby	608.8	352	1.4	24	19	12.4	11.7	16.7	18.0	7.4	3.7	13.9	0.0	9.1
Rømø	620	339	1.4	25	20	19.4	12.0	3.7	24.5	9.9	6.0	15.9	0.0	14.1
Esbjerg	655.2	422	3.6	43	47	18.8	23.9	39.3	28.0	11.1	8.0	18.2	0.0	11.8
Ho Bugt	669.6	22	1.6	28	26	14.8	13.2	22.6	21.6	9.2	5.7	14.6	0.0	11.5

Tabel 7

PAHs in $\mu\text{g/kg}$ on an ash-free
dry-weight base

Location	KM(O = Vlis)	Fen	Ant	Flu	Pyr	BaA	Chr	BeP	BbF	BkF	Bap	BghiPe	DBaH	INP
Hansweert	-30	84	2.6	186	252	80.5	127.9	451.2	104.6	40.9	23.5	58.6	6.7	25.1
Vlissingen	0	112	6.0	249	191	56.4	91.5	257.5	44.7	17.4	10.2	17.2	0.0	13.7
Colynsplaet	29.9	56	0.7	56	37	6.6	16.7	35.4	16.5	6.7	1.1	7.8	0.0	7.8
Brouwersdam	54.05	54	3.8	101	102	24.6	40.8	83.7	25.9	11.9	6.9	12.8	0.0	10.5
Hoek v. Holland	83.7	100	3.7	148	152	47.9	85.2	154.5	50.5	22.5	11.2	26.3	0.0	20.6
Delfl. Hoofden	97.2	104	3.5	172	151	37.0	68.0	123.6	38.9	16.4	9.2	20.5	0.0	14.1
IJmuiden	144.9	110	2.0	231	163	31.4	63.0	112.3	36.2	16.1	4.6	18.4	0.0	15.4
Callantsoog	187.2	78	1.4	93	72	16.1	31.1	57.4	23.6	9.9	4.2	13.7	0.0	10.3
Den Helder	201.5	210	7.7	443	276	61.1	61.5		72.3	31.2	24.2	34.6	6.8	30.2
Texel	208.5	37	2.1	63	40	15.5	15.7	23.8	14.1	5.9	2.2	4.2	1.3	5.9
Den Oever	220.4	570	5.2	156	104	38.4	50.0	52.3	52.0	23.0	13.7	27.5	3.7	24.0
Breezanddijk	235.6	52	2.8	46	37	34.6	23.6	9.1	37.0	16.6	8.4	19.9	3.4	18.1
Kornwerfzand	245.7	73	6.4	170	126	23.1	72.3	10.8	76.5	37.4	40.3	56.2	9.9	57.1
Piet Scheveplaat	300	40	2.4	76	60	22.4	23.8	38.9	30.8	13.5	8.8	19.9	0.0	13.7
Lauwersoog	318.3	525	7.7	207	136	42.0	50.2	67.7	59.0	24.9	17.3	33.1	4.3	23.1
Rottumerplaat	339	30	2.3	59	46	13.3	19.1	40.0	30.0	12.6	10.2	20.1	2.1	14.5
Blind Ranselgat	361.5	75	1.7	50	47	15.4	20.2	46.9	27.6	12.0	5.2	12.6	0.0	15.9
Bocht van Watum	373.5	338	2.7	68	56	31.8	26.6	45.0	40.5	16.0	11.0	23.2	0.0	16.1
Harlesiel	423	37	1.5	46	37	8.0	13.9	34.0	25.7	9.4	1.5	14.1	3.4	10.5
Mundahn	440.6	467	1.5	60	54	25.7	22.2	52.0	38.5	16.3	7.0	24.0	1.6	18.8
Wilhelmhaven	454	468	3.2	84	70	24.5	22.9	46.4	34.4	13.4	10.1	24.4	1.6	17.7
Büsum	506	40	1.5	64	59	12.5	20.7	52.4	40.2	15.8	4.1	21.9	0.0	14.5
Nordstrand	446	39	1.2	29	27	7.5	9.4	5.9	16.5	6.6	1.0	5.8	1.8	10.3
Dagebüll	568	454	19.3	708	288	393.6	280.0	126.0	299.7	119.5	42.8	29.6	0.0	29.9
Sylt	590	21	1.4	22	16	10.0	5.3	10.5	7.5	3.0	1.9	4.8	0.0	3.4
Koldby	608.8	420	1.6	29	23	14.8	14.0	20.0	21.5	8.8	4.5	16.6	0.0	10.9
Rømø	620	424	1.7	31	25	24.3	15.0	4.6	30.6	12.4	7.5	19.9	0.0	17.6
Esbjerg	655.2	507	4.4	52	56	22.6	28.7	47.2	33.6	13.4	9.7	21.9	0.0	14.2
Ho Bugt	669.6	26	1.8	32	30	17.3	15.4	26.3	25.2	10.8	6.7	17.1	0.0	13.4

Table 8

PAHs in mg/kg on a lipid base

Location	KM(O = Vlis)	Fen	Ant	Flu	Pyr	BaA	Chr	BeP	BbF	BkF	Bap	BghiPe	DBahA	InP
Hansweert	-30	0.84	0.03	1.86	2.52	0.81	1.28	4.51	1.05	0.41	0.23	0.59	0.07	0.25
Vlissingen	0	0.95	0.05	2.11	1.62	0.48	0.77	2.18	0.38	0.15	0.09	0.15	0.00	0.12
Colynsplaet	29.9	0.49	0.01	0.50	0.33	0.06	0.15	0.31	0.14	0.06	0.01	0.07	0.00	0.07
Brouwersdam	54.05	0.55	0.04	1.02	1.03	0.25	0.41	0.84	0.26	0.12	0.07	0.13	0.00	0.11
Hoek v. Holland	83.7	0.90	0.03	1.34	1.37	0.43	0.77	1.39	0.45	0.20	0.10	0.24	0.00	0.19
Delfl. Hoofden	97.2	0.91	0.03	1.50	1.32	0.32	0.59	1.08	0.34	0.14	0.08	0.18	0.00	0.12
IJmuiden	144.9	0.78	0.01	1.63	1.15	0.22	0.44	0.79	0.26	0.11	0.03	0.13	0.00	0.11
Callantsoog	187.2	0.74	0.01	0.89	0.68	0.15	0.30	0.55	0.23	0.09	0.04	0.13	0.00	0.10
Den Helder	201.5	2.67	0.10	5.64	3.51	0.78	0.78	0.00	0.92	0.40	0.31	0.44	0.09	0.38
Texel	208.5	0.37	0.02	0.61	0.39	0.15	0.15	0.23	0.14	0.06	0.02	0.04	0.01	0.06
Den Oever	220.4	6.30	0.06	1.73	1.15	0.42	0.55	0.58	0.57	0.25	0.15	0.30	0.04	0.27
Breezanddijk	235.6	0.62	0.03	0.55	0.44	0.41	0.28	0.11	0.44	0.20	0.10	0.24	0.04	0.22
Kornwerperstrand	245.7	0.85	0.07	1.97	1.46	0.27	0.84	0.13	0.89	0.44	0.47	0.65	0.12	0.66
Piet Scheveplaat	300	0.44	0.03	0.82	0.65	0.24	0.26	0.42	0.33	0.15	0.10	0.22	0.00	0.15
Lauwersoog	318.3	4.57	0.07	1.81	1.18	0.37	0.44	0.59	0.51	0.22	0.15	0.29	0.04	0.20
Rottumerplaat	339	0.25	0.02	0.49	0.38	0.11	0.16	0.33	0.25	0.10	0.08	0.17	0.02	0.12
Blind Ranselgat	361.5	0.77	0.02	0.51	0.48	0.16	0.21	0.48	0.28	0.12	0.05	0.13	0.00	0.16
Bocht van Watum	373.5	4.06	0.03	0.82	0.67	0.38	0.32	0.54	0.49	0.19	0.13	0.28	0.00	0.19
Harlesiel	423	0.39	0.02	0.48	0.39	0.08	0.14	0.35	0.27	0.10	0.02	0.15	0.04	0.11
Mundahn	440.6	5.29	0.02	0.68	0.61	0.29	0.25	0.59	0.44	0.18	0.08	0.27	0.02	0.21
Wilhelmhaven	454	3.74	0.03	0.67	0.56	0.20	0.18	0.37	0.27	0.11	0.08	0.19	0.01	0.14
Büsum	506	0.41	0.01	0.65	0.60	0.13	0.21	0.53	0.41	0.16	0.04	0.22	0.00	0.15
Nordstrand	446	0.38	0.01	0.28	0.26	0.07	0.09	0.06	0.16	0.06	0.01	0.06	0.02	0.10
Dagebüll	568	5.77	0.24	8.99	3.65	5.00	3.56	1.60	3.81	1.52	0.54	0.38	0.00	0.38
Sylt	590	0.20	0.01	0.22	0.16	0.10	0.05	0.10	0.07	0.03	0.02	0.05	0.00	0.03
Koldby	608.8	6.11	0.02	0.42	0.34	0.21	0.20	0.29	0.31	0.13	0.07	0.24	0.00	0.16
Rømø	620	4.63	0.02	0.34	0.27	0.27	0.16	0.05	0.33	0.14	0.08	0.22	0.00	0.19
Esbjerg	655.2													
Ho Bugt	669.6	0.33	0.02	0.41	0.38	0.22	0.20	0.34	0.32	0.14	0.09	0.22	0.00	0.17

Table 9PCBs in $\mu\text{g/kg}$ on a freeze dried
base

Location	KM 0 = Vlis	se 18	se 31	se 28	se 52	se 49	se 44	se 101	se 118	se 153	cp 105	se 138	se 187	se 180	cp 170
Hansweert	-30	3.5	3.2	5.8	29.8	17.3	12.9	97.5	55.2	193.0	20.0	133.2	56.8	31.7	8.7
Vlissingen	0	1.04	1.80	2.32	9.25	9.61	4.21	31.54	20.06	66.96	6.30	39.31	21.60	6.51	2.46
Colynsplaet	29.9	0.83	1.42	1.92	7.33	6.39	3.30	27.74	18.31	62.18	4.94	35.49	19.86	4.47	1.89
Brouwersdam	54.05	2.66	4.41	5.42	22.80	27.17	11.50	54.87	27.93	89.02	7.64	51.05	26.68	9.62	3.55
Hoek v. Holland	83.7	4.82	7.47	8.81	31.95	58.84	16.65	76.15	42.96	125.20	11.00	74.18	36.34	8.41	3.12
Delfl. Hoofden	97.2	4.04	6.75	78.81	27.08	35.15	14.49	61.52	35.80	107.11	10.30	62.82	32.04	12.20	4.51
IJmuiden	144.9	2.83	4.73	6.04	18.93	22.47	10.64	48.86	31.41	92.36	9.67	56.44	28.59	10.67	4.11
Callantsoog	187.2	1.10	2.02	2.45	7.73	9.38	4.53	26.91	18.60	60.37	5.94	37.40	20.35	9.38	3.76
Den Helder	201.5	0.65	1.33	1.64	4.47	6.80	2.49	17.11	11.30	42.28	3.67	24.54	12.74	4.21	1.79
Texel	208.5	0.93	1.72	2.29	6.20	11.38	3.44	19.67	12.31	41.84	4.19	24.32	12.56	4.42	1.83
Den Oever	220.4	0.90	1.61	2.06	4.71	11.28	2.85	17.86	13.17	44.81	4.29	26.97	14.98	3.30	1.59
Breezanddijk	235.6	0.71	1.34	1.74	3.43	6.76	2.20	16.67	11.58	43.44	3.83	25.47	13.15	3.56	1.55
Kornwerperzand	245.7	0.78	1.70	2.14	5.61	12.21	3.32	26.02	19.96	61.57	6.11	37.65	19.20	6.03	2.36
Piet Scheveplaat	300	0.64	1.49	1.90	4.72	6.92	2.76	21.50	15.72	52.34	4.92	31.44	16.12	5.48	2.16
Lauwersoog	318.3	0.56	1.27	1.60	5.06	5.52	2.68	24.58	20.14	61.34	6.69	40.12	21.39	4.88	2.09
Rottumerplaat	339	0.80	1.26	1.83	4.47	5.09	2.40	20.14	16.26	46.93	5.22	30.57	15.18	4.27	1.81
Blind Ranselgat	361.5	0.96	1.48	1.79	3.64	4.90	2.03	16.36	12.79	40.96	4.52	26.85	13.98	3.72	1.66
Bocht v. Watum	373.5	0.53	0.89	1.15	2.27	4.38	1.39	11.32	9.36	34.48	3.17	21.39	11.95	2.24	1.14
Harlesiel	423	0.59	1.13	1.39	2.34	2.20	1.48	11.50	9.70	36.42	3.34	23.48	12.55	2.35	1.08
Mundahn	440.6	0.59	0.99	1.22	2.48	2.86	1.46	13.13	10.71	44.26	3.71	28.64	16.33	3.06	1.42
Wilhelmhaven	454	0.95	1.47	1.78	3.79	5.35	2.26	17.23	12.60	51.42	4.59	32.64	15.88	4.01	1.76
Büsum	506	0.62	1.08	1.32	5.29	3.87	1.93	39.12	16.03	96.33	5.24	57.96	27.40	9.49	4.18
Nordstrand	536	0.71	0.60	0.72	1.26	1.14	0.65	6.64	5.28	25.85	1.90	17.21	8.82	1.85	0.91
Dagebüll	568	0.41	0.69	0.76	0.96	1.10	0.63	4.37	3.08	18.20	1.24	11.93	6.89	1.53	0.88
Sylt	590	0.44	0.47	0.59	1.05	1.02	0.63	3.94	3.12	11.51	1.16	8.18	3.91	1.10	0.54
Koldby	608.8	0.47	0.55	0.60	0.73	0.89	0.49	3.29	3.11	16.96	1.18	11.53	7.17	1.01	0.54
Rømø	620	0.61	0.63	0.66	0.70	0.78	0.41	2.64	2.69	12.30	1.07	8.51	4.78	0.97	0.54
Esbjerg	655.2	0.84	1.01	1.08	2.20	2.13	1.41	8.69	7.04	22.98	3.06	15.67	6.79	1.92	0.91
Ho Bugt	669.6	0.47	0.62	0.74	1.17	1.34	0.70	4.76	4.86	19.58	1.81	12.96	6.48	1.31	0.62

Table 10

PCBs in µg/kg on a dry-weight base

Location	KM 0 = Vlis	se 18	se 31	se 28	se 52	se 49	se 44	se 101	se 118	se 153	cp 105	se 138	se 187	se 180	cp 170
Hansweert	-30	3.7	3.3	6.0	30.8	17.8	13.3	100.6	56.9	199.1	20.6	137.4	58.6	32.7	9.0
Vlissingen	0	1.1	1.8	2.4	9.5	9.8	4.3	32	21	69	6.5	40	22	6.7	2.5
Colynsplaet	29.9	0.84	1.4	2.0	7.5	6.5	3.4	28	19	63	5.0	36	20	4.5	1.9
Brouwersdam	54.05	2.7	4.5	5.6	23	28	12	56	29	91	7.8	52	27	9.9	3.6
Hoek v. Holland	83.7	5.0	7.8	9.1	33	61	17	79	45	130	11	77	38	8.7	3.2
Delfl. Hoofden	97.2	4.2	7.0	8.1	28	37	15	64	37	112	11	66	33	13	4.7
IJmuiden	144.9	3.0	5.0	6.4	20	24	11	52	33	98	10	60	30	11	4.4
Callantsoog	187.2	1.1	2.1	2.5	8.0	9.8	4.7	28	19	63	6.2	39	21	9.8	3.9
Den Helder	201.5	0.68	1.4	1.7	4.7	7.2	2.6	18	12	45	3.9	26	13	4.4	1.9
Texel	208.5	0.98	1.8	2.4	6.6	12	3.6	21	13	44	4.4	26	13	4.7	1.9
Den Oever	220.4	0.97	1.7	2.2	5.0	12	3.1	19	14	48	4.6	29	16	3.5	1.7
Breezanddijk	235.6	0.74	1.4	1.8	3.6	7.0	2.3	17	12	45	4.0	27	14	3.7	1.6
Kornwerperzand	245.7	0.81	1.7	2.2	5.8	13	3.4	27	21	64	6.3	39	20	6.2	2.4
Plet Scheveplaat	300	0.67	1.6	2.0	4.9	7.2	2.9	22	16	55	5.1	33	17	5.7	2.3
Lauwersoog	318.3	0.59	1.3	1.7	5.4	5.9	2.8	26	21	65	7.1	43	23	5.2	2.2
Rottumerplaat	339	0.85	1.3	1.9	4.7	5.4	2.5	21	17	49	5.5	32	16	4.5	1.9
Blind Ranselgat	361.5	0.99	1.5	1.9	3.8	5.1	2.1	17	13	42	4.7	28	14	3.9	1.7
Bocht v. Watum	73.5	0.55	0.9	1.2	2.3	4.5	1.4	12	10	36	3.3	22	12	2.3	1.2
Harlesiel	423	0.62	1.2	1.5	2.4	2.3	1.5	12	10	38	3.5	25	13	2.5	1.1
Mundahn	440.6	0.62	1.0	1.3	2.6	3.0	1.5	14	11	46	3.9	30	17	3.2	1.5
Wilhelmhaven	454	1.03	1.6	1.9	4.1	5.8	2.4	19	14	56	5.0	35	17	4.3	1.9
Büsum	506	0.65	1.1	1.4	5.5	4.0	2.0	41	17	100	5.5	60	29	9.9	4.4
Nordstrand	536	0.75	0.63	0.77	1.3	1.2	0.69	7.0	5.6	27	2.0	18	9.3	2.0	1.0
Dagebüll	568	0.43	0.73	0.81	1.0	1.2	0.66	4.6	3.3	19	1.3	13	7.3	1.6	0.9
Sylt	590	0.46	0.49	0.62	1.1	1.1	0.66	4.1	3.2	12	1.2	8.5	4.1	1.1	0.6
Koldby	608.8	0.50	0.58	0.63	0.77	0.93	0.51	3.4	3.2	18	1.2	12	7.5	1.1	0.6
Rømø	620	0.66	0.68	0.71	0.76	0.84	0.44	2.9	2.9	13	1.2	9.2	5.2	1.1	0.6
Esbjerg	655.2	0.91	1.1	1.2	2.4	2.3	1.5	9.4	7.6	25	3.3	17	7.3	2.1	1.0
Ho Bugt	669.6	0.49	0.65	0.77	1.2	1.4	0.73	5.0	5.1	20	1.9	13	6.7	1.4	0.6

Table 11

PCBs in $\mu\text{g/kg}$ on an ash-free
dry-weight base

Location	KM 0 = Vlis	se 18	se 31	se 28	se 52	se 49	se 44	se 101	se 118	se 153	cp 105	se 138	se 187	se 180	cp 170
Hansweert	-30	4.2	3.8	6.9	35.8	20.7	15.5	116.9	66.2	231.4	24.0	159.7	68.1	38.1	10.4
Vlissingen	0	1.2	2.1	2.7	11	11	4.8	36	23	77	7.2	45	25	7.5	2.8
Colynsmaat	29.9	0.98	1.7	2.3	8.6	7.5	3.9	33	22	73	5.8	42	23	5.3	2.2
Brouwersdam	54.05	3.1	5.1	6.2	26	31	13	63	32	103	8.8	59	31	11	4.1
Hoek v. Holland	83.7	5.8	9.0	11	39	71	20	92	52	151	13.3	90	44	10	3.8
Delft. Hoofden	97.2	4.9	8.2	9.5	33	43	18	75	44	130	12.5	76	39	15	5.5
IJmuiden	144.9	3.4	5.7	7.3	23	27	13	59	38	111	11.6	68	34	13	4.9
Callantsoog	187.2	1.4	2.5	3.1	9.7	12	5.7	34	23	76	7.4	47	26	12	4.7
Den Helder	201.5	0.80	1.6	2.0	5.5	8.4	3.1	21	14	52	4.5	30	16	5.2	2.2
Texel	208.5	1.1	2.1	2.8	7.5	14	4.2	24	15	51	5.1	30	15	5.4	2.2
Den Oever	220.4	1.2	2.1	2.7	6.1	15	3.7	23	17	58	5.5	35	19	4.3	2.1
Breezanddijk	235.6	0.85	1.6	2.1	4.1	8.1	2.6	20	14	52	4.6	30	16	4.3	1.9
Kornwerderzand	245.7	0.92	2.0	2.5	6.6	14	3.9	31	24	73	7.2	45	23	7.1	2.8
Piet Scheveplaat	300	0.79	1.8	2.3	5.8	8.4	3.4	26	19	64	6.0	38	20	6.7	2.6
Lauwersoog	318.3	0.71	1.6	2.1	6.5	7.1	3.4	32	26	79	8.6	52	27	6.3	2.7
Rottumerplaat	339	0.97	1.5	2.2	5.4	6.1	2.9	24	20	57	6.3	37	18	5.1	2.2
Blind Ranselgat	361.5	1.1	1.7	2.1	4.3	5.8	2.4	19	15	48	5.3	32	16	4.4	2.0
Bocht v. Watum	373.5	0.67	1.1	1.5	2.9	5.5	1.8	14	12	44	4.0	27	15	2.8	1.4
Harlesiel	423	0.72	1.4	1.7	2.9	2.7	1.8	14	12	45	4.1	29	15	2.9	1.3
Mundahn	440.6	0.76	1.3	1.6	3.2	3.7	1.9	17	14	57	4.8	37	21	3.9	1.8
Wilhelmhaven	454	1.3	2.0	2.4	5.1	7.2	3.0	23	17	69	6.1	44	21	5.4	2.4
Büsum	506	0.77	1.3	1.6	6.5	4.8	2.4	48	20	119	6.5	72	34	12	5.2
Nordstrand	536	0.92	0.77	0.94	1.6	1.5	0.84	8.6	6.8	33	2.5	22	11	2.4	1.2
Dagebüll	568	0.53	0.90	0.99	1.2	1.4	0.82	5.7	4.0	24	1.6	16	9.0	2.0	1.1
Sylt	590	0.51	0.54	0.68	1.2	1.2	0.73	4.6	3.6	13	1.3	9.4	4.5	1.3	0.62
Koldby	608.8	0.59	0.69	0.76	0.92	1.1	0.61	4.1	3.9	21	1.5	14	9.0	1.3	0.67
Rømø	620	0.82	0.85	0.89	1.0	1.1	0.55	3.6	3.6	17	1.5	11	6.5	1.3	0.73
Esbjerg	655.2	1.1	1.3	1.4	2.9	2.8	1.8	11	9.1	30	4.0	20	8.8	2.5	1.2
Ho Bugt	669.6	0.57	0.76	0.90	1.4	1.6	0.85	5.8	5.9	24	2.2	16	7.9	1.6	0.76

Table 12

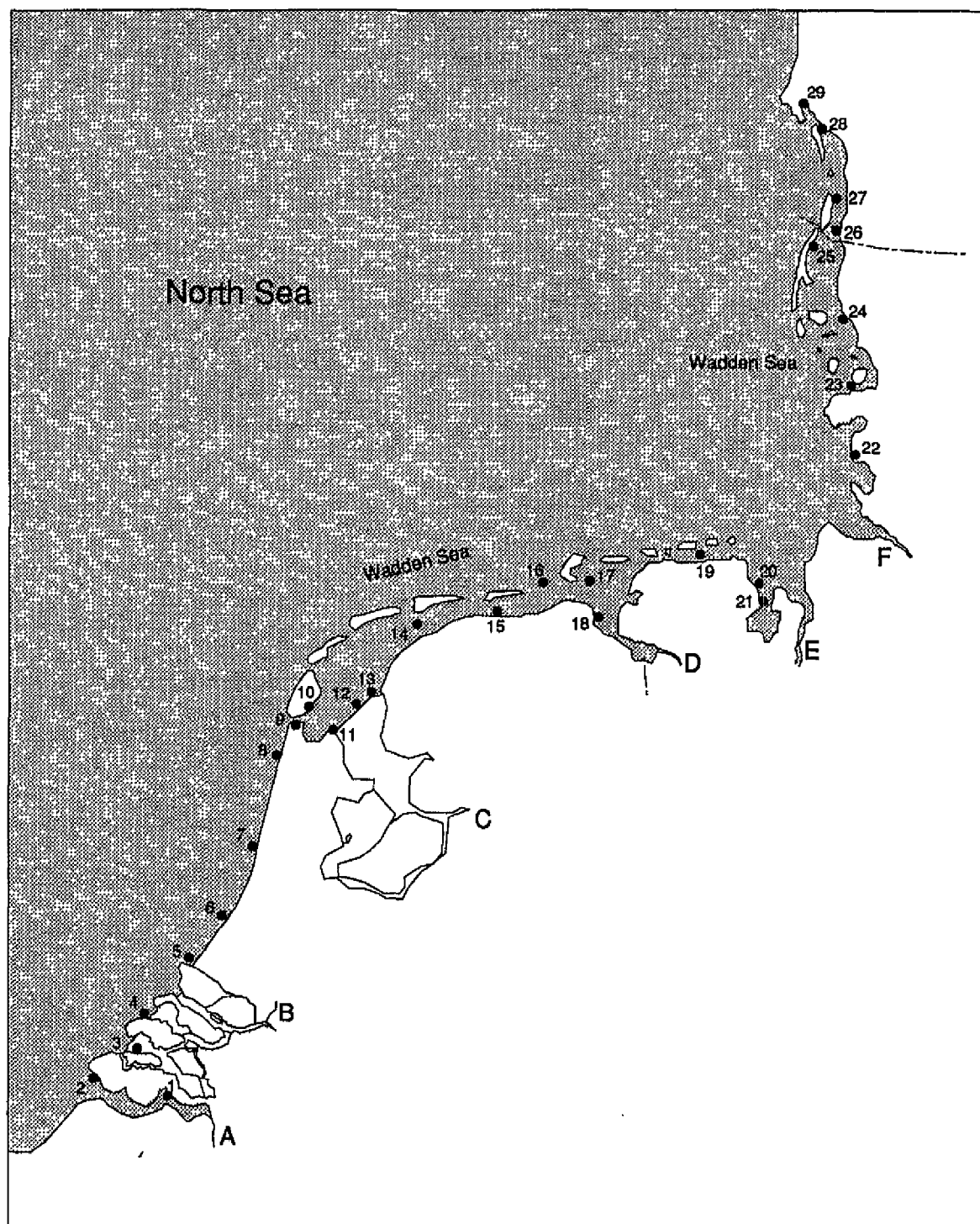
PCBs in mg/kg on a lipid base

Location	KM 0 = Vlis	se 18	se 31	se 28	se 52	se 49	se 44	se 101	se 118	se 153	cp 105	se 138	se 187	se 180	cp 170
Hansweert	-30	42	38	69	358	207	155	1169	662	2315	240	1598	681	381	104
Vlissingen	0	10	18	23	90	93	41	307	195	651	61	382	210	63	24
Colynsplaet	29.9	8.6	15	20	76	66	34	287	190	644	51	367	206	46	20
Brouwersdam	54.05	31	51	63	265	316	134	637	324	1034	89	593	310	112	41
Hoek v. Holland	83.7	52	81	96	348	641	181	829	468	1364	120	808	396	92	34
Delfl. Hoofden	97.2	43	71	83	287	372	153	652	379	1135	109	665	339	129	48
IJmuiden	144.9	24	40	51	161	191	90	415	267	784	82	479	243	91	35
Callantsoog	187.2	13	24	29	92	112	54	322	223	722	71	447	243	112	45
Den Helder	201.5	10	21	26	70	106	39	268	177	662	57	384	199	66	28
Texel	208.5	11	21	27	74	135	41	234	146	498	50	289	149	53	22
Den Oever	220.4	13	23	29	67	161	41	256	188	641	61	386	214	47	23
Breezanddijk	235.6	10	19	25	49	96	31	236	164	616	54	361	187	51	22
Kornwernerzand	245.7	11	23	29	77	168	46	358	275	847	84	518	264	83	32
Piet Scheveplaat	300	8.5	20	25	63	92	37	285	208	693	65	416	214	73	29
Lauwersoog	318.3	6.2	14	18	57	62	30	275	225	686	75	449	239	55	23
Rottumerplaat	339	8.0	13	18	45	51	24	201	162	469	52	305	152	43	18
Blind Ranselgat	361.5	12	18	22	44	59	25	198	155	495	55	325	169	45	20
Bocht v. Watum	373.5	8.1	13	17	34	67	21	172	142	524	48	325	182	34	17
Harlesiel	423	7.5	14	18	30	28	19	146	123	463	43	299	160	30	14
Mundahn	440.6	8.6	14	18	36	41	21	190	155	641	54	415	237	44	21
Wilhelmhaven	454	10	16	19	41	57	24	184	135	549	49	349	170	43	19
Büsum	506	7.8	14	17	67	49	24	493	202	1213	66	730	345	119	53
Nordstrand	536	8.9	7.5	9.1	16	14	8.2	84	66	326	24	217	111	23	12
Dagebüll	568	6.7	11	13	16	18	10	72	51	301	21	197	114	25	14
Sylt	590	4.9	5.3	6.7	12	12	7.1	44	35	129	13	92	44	12	6.1
Koldby	608.8	8.6	10	11	13	16	8.9	60	56	308	21	209	130	18	10
Rømø	620	9.0	9.3	10	10	11	6.0	39	40	181	16	125	71	14	8.0
Esbjerg	655.2														
Ho Bugt	669.6	7.3	10	12	18	21	11	74	76	305	28	202	101	20	10

Table 13Metals in mg/kg on a dry-weight
base

Location	Km	Gloelverl	Gloeirest	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Hansweert	-30.0	86.47	13.53	7.69	5.3	10.1	0.24	10	6.3	263
Vlissingen	0.0	89.40	10.60	0.65	2.3	7.6	0.14	4	3.0	103
Colynsmaat	29.9	86.59	13.41	0.45	1.5	7.3	0.19	3	2.2	80
Brouwersdam	54.1	89.17	10.83	0.46	1.5	8.1	0.16	4	2.2	75
Hoek v. Holland	83.7	86.42	13.58	0.82	2.2	7.4	0.21	6	4.0	128
Delfl. Hoofden	97.2	86.31	13.69	0.57	1.9	8.0	0.14	3	2.8	96
IJmuiden	144.9	88.91	11.09	0.66	2.3	8.1	0.13	4	2.9	121
Callantsoog	187.2	83.66	16.34	0.66	2.6	8.8	0.17	10	2.5	108
Den Helder	201.5	86.47	13.53	0.71	2.4	8.9	0.26	3	3.8	146
Texel	208.5	87.80	12.20	0.41	1.5	7.0	0.14	<d.l.	2.2	89
Den Oever	220.4	83.96	16.04	0.55	2.4	9.1	0.21	4	3.0	118
Breezanddijk	235.6	87.65	12.35	0.40	4.8	7.6	0.39	5	2.2	66
Kornwerderzand	245.7	87.66	12.34	0.31	3.7	8.2	0.24	7	2.4	77
Piet Scheveplaat	300.0	86.40	13.60	0.55	3.0	7.3	0.12	5	1.9	81
Lauwersoog	318.3	83.52	16.48	0.39	3.7	7.4	0.21	6	3.2	83
Rottumerplaat	339.0	88.14	11.86	0.30	3.1	10.6	0.18	4	3.2	78
Blind Ranselgat	361.5	88.41	11.59	0.72	3.6	8.5	0.17	5	1.8	90
Bocht v. Watum	373.5	82.48	17.52	1.92	3.0	6.7	0.30	6	3.4	178
Harlesiel	423.0	86.02	13.98	0.50	1.8	6.1	0.15	<d.l.	2.2	95
Mundahn	440.6	82.65	17.35	1.34	3.1	6.5	0.29	3	3.5	125
Wilhelmshaven	454.0	82.23	17.77	1.74	3.2	9.3	0.26	3	2.8	165
Büsum	506.0	84.97	15.03	0.95	3.2	7.7	0.23	4	2.2	102
Nordstrand	536.0	82.84	17.16	0.61	2.8	7.7	0.25	<d.l.	1.8	84
Dagebüll	568.0	82.02	17.98	1.21	3.2	7.3	0.28	4	2.4	103
Sylt	590.0	90.39	9.61	0.31	1.2	6.2	0.10	<d.l.	<d.l.	61
Koldby	608.8	84.33	15.67	1.39	3.1	7.4	0.21	3	2.1	107
Rømø	620.0	81.52	18.48	1.03	3.0	8.0	0.30	4	2.6	93
Esbjerg	655.2	84.51	15.49	0.72	2.9	9.2	0.18	4	2.9	117
Ho Bugt	669.6	86.20	13.80	0.77	3.5	7.7	0.23	4	2.2	91

Appendix 2



Appendix 3

Location	Number	Kilometre	River
Hansweert	1	- 30	A: Scheldt
Vlissingen	2	0	
Colynsplaet	3	30	
Brouwersdam	4	54	
Hoek van Holland	5	84	B: Rhine
Delftlandse hoofden	6	97	
IJmuiden	7	145	
Callantsoog	8	187	
Den Helder	9	202	
Texel	10	209	
Den Oever	11	220	
Breezanddijk	12	236	
Kornwerderzand	13	246	C: IJssel
Piet Scheveplaat	14	300	
Lauwersoog	15	318	
Rottumerplaat	16	339	
Blind Ranselgat	17	362	
Bocht van Watum	18	374	D: Ems
Harlesiel	19	423	
Mundahn	20	441	
Wilhelmshaven	21	454	E: Weser
Büsum	22	506	F: Elbe
Nordstrand	23	536	
Dagebüll	24	568	
Sylt	25	590	
Kolby	26	609	
Rømø	27	620	
Esbjerg	28	655	Vardø å
Ho Bugt	29	670	

Grafische verzorging:
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