## Mytilus edulis L. as Bioindicator for Organochlorine in the Baltic Sea

K. M. Lee , H. Kruse and O.Wassermann
Department of Toxicology, University of Kiel, Germany

### Introduction

For the monitoring of aquatic pollution with xenobiotics, aquatic biota are chosen rather than water analysis itself. The quantification of contaminants such as polychlorinated biphenyls, DDT, and HCH in sea-water is very inaccurate or actually impossible due to the low concentrations.
In contrast, the aquatic biota highly concentrate persistent lipophilic contaminants such as PCBs, making their analysis relatively simple and providing a direct picture of pollutant bio-availability.
International monitoring programs are carried out using mussels, particularly Mytilus edulis L because of its advantages as a bioindicator, like wide distribution, abundance, sedentary behaviour, and pronounced ability to accumulate PCBs.
Several authors reported a great seasonal fluctuation of PCB levels over an annual cycle, which creates several problems for the comparison of contamination levels between different areas. They suggested that this fluctuation is related to changes in biological or biochemical activity of the mussel(Farrington et al 1983;Capuzzo et al 1989).
We present the monitoring of the West German Baltic Sea coastal waters, and we discuss the factors influencing the concentration of PCBs in Mytilus edulis.L.

### Material and Methods

During February 1991 samples were taken between Flensburg and Travemünde. The stations Flensburg, Eckernförde, Kiel Innenförde, Neustadt, Travemünde were located in harbour areas. The other stations, Gelting, Schilksee, Kellenhüsen, and Scharbeutz were located at bathing beaches and Kiel Leuchtturm in the open sea (Fig. 1).
For time-integration of PCB contamination, samples were taken monthly from July 1990 to July 1991 at stations Kieler Innenförde, Schilksee, and Kiel Leuchtturm.
For all analyses, 8 mussels, 60±4mm in shell length, were taken at each station. 40 mussels in shell lengths of 40±4mm, 50±4mm and 60±4mm, respectively, were taken for comparison of age differences in PCB concentration in March 1992 at Kiel Innenförde.
All mussels were cleaned in the laboratory, and the soft tissue was carefully removed from the shell using a scalpel.
They were stored in aluminium foil at -20 C°. Before the analysis, the soft tissue was freeze dried and then ground with sodium sulfate in a mill. The homogenised samples were extracted with 150ml n-hexane for 6 hours in a soxhlet apparatus.
The clean up of the extracted fat containing the organochlorine compounds was carried out using gel chromatography and adsorption chromatography according to Specht and Thilkes (1980). A gas chromatograph HRGC 5160 Carlo Erba with DB 5 non polar silicon column and electron capture detector was used for identification and quantification. The identification was verified by mass spectrometry. The PCB congeners with identical retention times on the DB 5 capillary columns were separated by two-dimensional gaschromatography (Sichromat 2-8,Siemens) using columns of different polarity. The most toxic PCBs could be identified only by using this technique.

### Results

The major contributors to organochlorine contamination in the Baltic sea were PCBs. In comparison, the levels of HCH isomers, HCB, DDT and its metabolites were insignificant (Figure 1).

# PCB

In the harbour areas, Flensburg, Eckernförde, Kieler Innenförde, Neustadt and Travemünde, particularly high concentrations of PCBs were determined, in contrast to the bathing areas e.g. Gelting, Schilksee, Kellenhusen, Scharbeutz and the open sea, Kiel Leuchtturm. The average total PCB concentration in mussel from the most contaminated harbour, Kieler Innenförde, was 487 µg/kg dry weight whereas the least contaminated sample from Gelting contained only 38 µg/kg dry weight (Table 1). The major fraction in PCB were the hexa congeners 138 (22%) and 153 (28% of total PCB). The highly toxic planar PCB 77 was detected at all stations except Schilksee and Scharbeutz. The average concentration of PCB 77 in Flensburg harbour was 2,6 µg/kg dry weight, equal to 2% of the main contaminant, PCB 153. Other planar PCBs, like 126 and 129, were not detected at all stations. However, we have found relatively high levels of mono-ortho PCB at the harbour stations, which were particularly highly contaminated with PCB 118. The concentration of mono-ortho PCBs 105, 118, 156 and 167 amounts to 12% of total PCBs (Fig.2).
We measured PCB levels monthly over a complete year at individual stations, and we have found great seasonal fluctuations (Fig.3). The concentration of PCBs increases during autumn and winter and decreases in late spring and early summer. The fluctuation correlates with the fat content of the mussels. A negative correlation between PCB levels and fat content was observed in the Kieler Innenförde (most contaminated area) during winter.
By comparing bioaccumulation at different shell lengths, 40mm±4, 50mm±4, 60mm±4, low chlorinated PCBs 26 and 52 were significantly ($P<0,05$) more concentrated in young animals and highly chlorinated PCB 170 and 180 more concentrated in adults.

Table 1: Concentration of selected organochlorine compounds in mussels from the Baltic Sea (concentration in µg/kg dry weight)

| Station | ΣHCH | HCB | ΣDDT | ΣPCB*) | Coplanar PCBs | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | 77 | 105 | 118 | 156 | 167 |
| Flensburg | 6,4 | 1,6 | 42,8 | 392,2 | 2,6 | 8,2 | 20,3 | 6,4 | 3,4 |
| Gelting | 6,1 | 0,7 | 6,6 | 38,6 | 0,4 | 3,4 | 2,2 | 0,8 | nd |
| Eckernförde | 6,2 | 0,8 | 26,9 | 318,4 | 0,4 | 1,2 | 13,2 | 5,9 | 3,6 |
| Schilksee | 2,5 | 0,4 | 13,1 | 75,0 | nd | 2,4 | 5,3 | 1,4 | 1,4 |
| Kiel Leuchtt | 6,4 | 1,0 | 26,4 | 66,7 | 0,2 | 1,7 | 4,3 | 0,7 | nd |
| Kiel Innenf. | 22,3 | 2,4 | 41,0 | 487,1 | 0,5 | 4,4 | 35,6 | 5,4 | 9,8 |
| Kellenhusen | 4,0 | nd | 14,7 | 40,6 | 0,2 | 0,5 | 1,2 | 0,9 | nd |
| Scharbeutz | 6,5 | 0,2 | 16,0 | 41,5 | nd | 2.0 | 3.4 | 2.1 | nd |
| Neustadt | 6,1 | 0,9 | 59,1 | 275,5 | 0,7 | 1,1 | 14,8 | 5,2 | 4,2 |
| Travemünde | 13,6 | 2,2 | 88,3 | 266,4 | 2,2 | 4,2 | 14,5 | 3,8 | 4,9 |

*) ΣPCB = 52, 151, 149, 118, 153, 138, 187, 183, 180.

## Discussion

Some authors suggested that equilibrium partitioning is a major factor determining the bioaccumulation in gill breathing aquatic organisms (Schneider 1982; Tanabe et al. 1984). The uptake and release of organochlorines is a function of the exchanges through the gills and across the body surface between the ambient water and the body lipids. Tanabe et al. (1989) found great differences in equilibrium rates of various PCB isomers and congeners; the low chlorinated PCBs are both taken up and lost more rapidly by mussels than higher chlorinated PCBs.
The monitoring of Baltic Sea mussels revealed the concentration of contaminants in the ambient water as the major factor controlling distribution of PCBs in mussels. Therefore, mussels are suitable bioindicators. The great seasonal fluctuations in concentrations of PCBs with a marked decline during spring and early summer and with a marked increase during autumn and winter was correlated with lipid content (Fig. 3). Lipophilic PCBs primarily bind to the non-polar part of the lipids.

Fig. 1: Regional differences in the organochlorine contamination

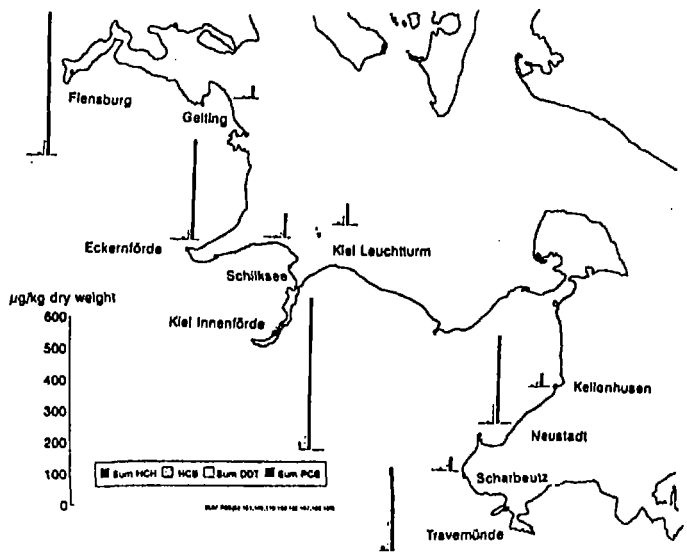



Fig. 2: Regional differences in contamination with co-planar PCBs

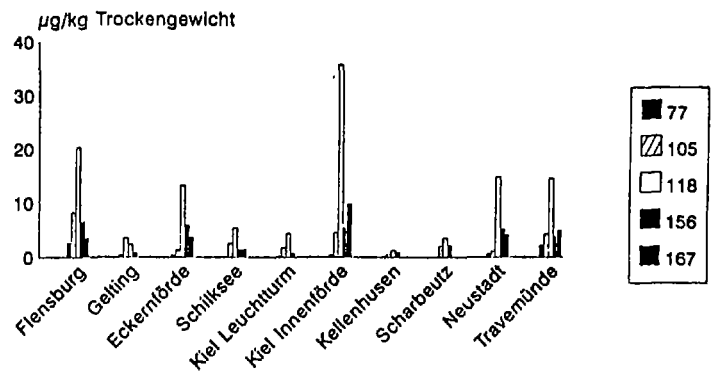
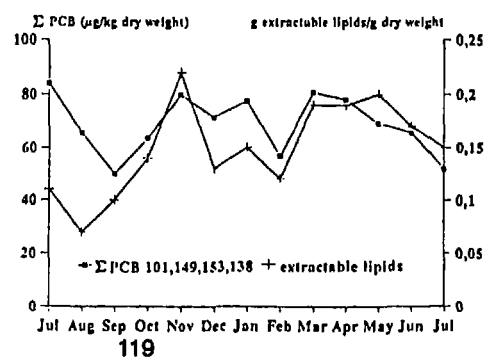



Fig. 3: Seasonal fluctuation of PCB concentration and extractable lipids in mussels from Schilksee area



Capuzzo et al. (1989) connect the fluctuation in PCB concentration with seasonal cycle of gametogenesis and spawning activity. The lipid content of mussels increases before spawning and declines during and after spawning (Phillip 1978). Together with the high fat content in gametes significant amounts of PCBs are excreted during spawning. We observed that the fluctuation in concentration of PCBs between summer and winter is more than twofold.
The negative correlation between PCB levels and lipid content in winter was observed in mussels from heavily polluted areas. This can be explained by a reduction in the condition index (CI). In mussels stressed by high pollution, a marked decrease of the CI was observed (Veldhizen-Tsoerkon et al. 1991), measured as the ratio of dry weight to shell volume. The reduction of CI correlates with lipid content. The change in CI influences the accumulation and utilisation of lipid reserves (Capuzzo et al. 1989).
By comparing size dependent accumulation we have found a significantly high accumulation ($P<0.05$) of low chlorinated PCB 26 and 52 in young animals. The adults were more contaminated with higher chlorinated PCBs 170 and 180.
Muncaster et al. (1990) suggested that the negative correlation between size and PCB levels is due to the high filtration rate of young animals. However, Kuware et al. (1986) reported a positive correlation. We have found no significant increase in lipid content with age. We assume a differential uptake route between young and adult animals. The uptake of PCBs in young animals primarily may depend on equilibrium partitioning, and the high accumulation in adult mussels can, in addition, be attributed to ingestion of seston suspended in the water, to which highly chlorinated PCBs preferentially tend to be bound.

## Conclusion

The major factor for the bioaccumulation of PCBs in mussels appears to be the concentration of PCBs in the ambient water. Mussel, like Mytilus edulis L., can be used as a suitable bioindicator. However, it is necessary to observe other factors influencing the accumulation of PCBs such as seasonal fluctuation of lipid content related to the reproductive cycles of mussels, physico-chemical properties of individual PCBs, diet determined and size dependent differential accumulation.

## References


Cappuro C.M., Farrington I.W., Rantamaki P., Clofford C.H. Lancaster B.A., Leavitt D.F. and Xiaoping Jia (1989), The relationship between lipid composition and seasonal differences in the distribution of PCBs in Mytilus edulis L., *Marine Environmental Research*, 28, 259-264.

Farrington J.W.,E.D. Goldberg, R.D. Riesbrough, J.H. Martin, and V.T. Bowen (1983), U.S. "Mussel Watch"1976-1978 :An Overview of the trace-metal, DDE,PCB, hydrocarbon and radionuclide data,Environ. Sci.Technol. 17, 490-498.

Kuware K., S. Fukushima, R. Tanaka, H. Miyata, T. Kashimoto (1986), Relationship between shell length of blue mussels and lables of residual PCBs within the body, *J. Food Hyg. Soc. Jap.*, 27, 565-568.

Muncaster B.W., N. Hebert, R. Lazar (1990), Biological and physical factors affecting the body burden of organic contaminants in freshwater mussels, *Arch. Environ. Contam. Toxicol.* 19, 25-34.

Phillip D.J.H. (1978), Use of biological indicator organismens to quantitate organochlorine pollutants in aquatic environments, a Review, Environ.Pollut. 16, 167-229 .

Schneider R. (1982), Polychlorinated biphenyls (PCBs) in cod tissues from the Western Baltic: Significance of equilibrium partitioning and lipid composition in the bioaccumulation of lipophilic pollutants in gill-breathing animals, *Meeresforschung*, 29, 69-79.

Tanabe S, H. Tanaka, R. Tatsukawa (1984), Polychlorobiphenyls, sigma-DDT;, and hexachlorocyclohexane isomers in the western North Pacific ecosystem, *Arch. Environ. Contam. Toxicol.*, 13, 731-738.

Tanabe S., R. Tatsukawa, D.J.H. Phillips (1989), Mussels as bioindicators of PCB pollution: A case study on uptake and release of PCB isomers and congeners in green-lipped mussels (Perna viridis) in Hong Kong waters, *Environmental Pollution* 47, 412-62.

Veldhuizen-Tsoerkan M.B., D.A. Holwerda, A.M.T. de Bont, A.C. Smaal, D.I. Zandee (1991), A field study on stress indices in the sea mussels, Mytilus edulis: Application of the "stress approach" in biomonitoring, *Arch. Environ. Contam. Toxicol.*, 21, 497-504.