

Congener profiles in European eel (*Anguilla anguilla* L.) as a method of tracing the origin of PCB contamination

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Introduction

Since 1994 the Institute for Forestry and Game Management (IBW) has built out a pollutant monitoring network for public water bodies in Flanders (Belgium) using eel (*Anguilla anguilla* (L.)) as a biomonitor. Eel is used for biomonitoring because it is a very fatty fish that bioaccumulates lipophilic organochlorine (OC) pesticides and PCBs, feeds in the benthic layer, and is sedentary during part of its life cycle (the yellow eel or immature adult phase, during which eels remain in brackish or fresh waters for five to twenty years). Eels are long-lived and widespread, occurring in very diverse habitats and even in polluted waters. Their position on the trophic ladder and the absence of an annual reproductive cycle, affecting lipid metabolism, are additional advantages for their use as a sentinel organism.

Contaminants analyzed were heavy metals, PCBs, OC pesticides, brominated flame retardants and volatile organic compounds. At present, the dataset includes results from approximately 2000 individually analyzed eels from more than 300 different localities in Flanders.

The results have been communicated to national managers as the high PCB values measured in eels from most of the locations are of great concern. Hence, immediate action has been undertaken to protect the local consumer health. A catch and release obligation for every eel caught in Flanders was set by ministerial decree. In some eel samples, PCB levels as high as 7,000 ng/g BW (measured as the sum of the 7 indicator PCBs) were measured, nearly exceeding the national PCB standard (75 ng/g BW) by two orders of magnitude.

Water managers were concerned about the origin of the high contamination. In this respect it was worthwhile to analyze the PCB profiles to evaluate if spatial or temporal variations exist and, if fingerprinting of contaminants such as PCBs could be a useful method to trace the contamination source.

Materials and Methods

PCB-analyses were carried out on 1968 individual eels which were collected from 325 different locations in Flanders over the nine year period from 1994 to 2003. The eels were collected by electrofishing, fykenets or a combination of both. Locations included rivers, canals, polder waters and closed water bodies. Some locations were sampled more than once during the study period.

Fish fillets were wrapped in aluminum paper (cleaned with hexane 99 %) and stored at -20 °C. All fish were analyzed individually. For this paper only the eels with a length between 30 and 50 cm are considered for standardization and comparison reasons. This final standardized dataset contained 1587 eels from 305 different locations. Chemical analyses were carried out by the Sea Fisheries Department in Ostend.

10 grams of fish fillet was extracted using the Bligh & Dyer method¹. The extract is evaporated (rotavapor) and at the most 100 mg lipid is dissolved in hexane and applied on an aluminum oxide chromatography column. After elution with hexane, the lipid free eluate is evaporated and applied on a silica gel chromatography column. Ten PCBs (CB 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180), *p,p'*-DDE and HCB are isolated after elution with hexane.

This fraction is evaporated to 1 ml, after addition of an external standard (tetrachloronaphtalene) and separated by GC using a Rtx-5ms capillary column (60 m x 0.25 mm x 0.25 µm), with helium as a carrier gas and an electron capture detector (ECD).

Results and Discussion

In Flanders there exists a clear spatial variation in contamination which can be linked to human interactions and/or land use. This variation is clearly reflected by the contamination levels in eel. ²⁻⁶ In both heavily as in mildly polluted waters, PCB profiles were used to evaluate spatial and temporal trends and to determine if fingerprinting could be a useful method to trace the origin of the contamination.

Spatial variation. We observed a high similarity in the fraction that PCB 118 accounted for compared to the total of the other measured (non-dioxin like) PCBs for eel from one and the same location, but at the same time there was a striking inter-location variation. For none of the other congeners there was a visible pattern if compared to the sum of the indicator PCBs (Figure 1).

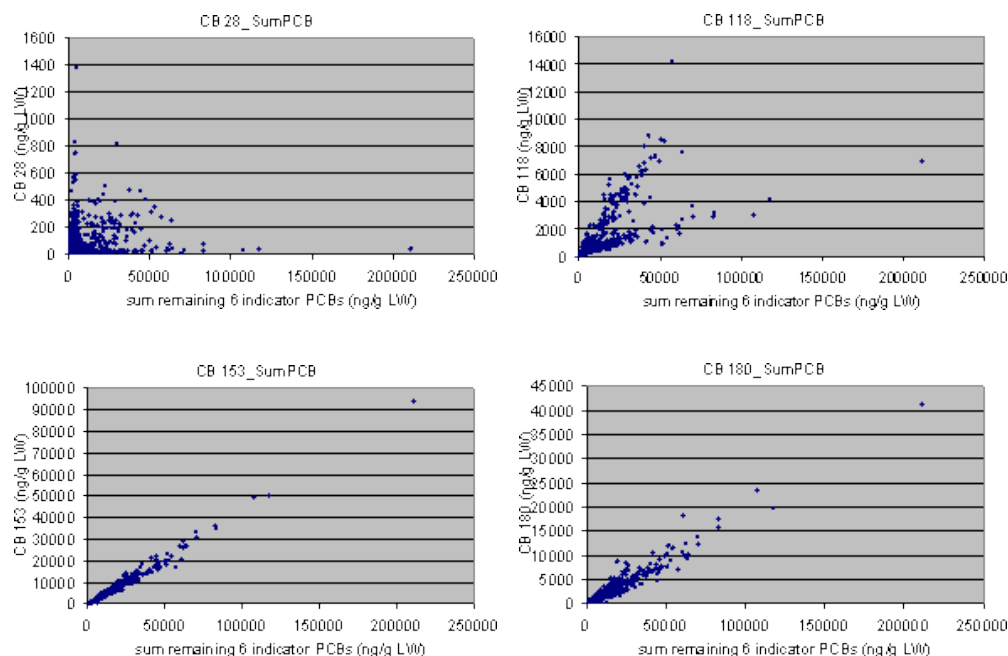


Figure 1: Scatter plots of different indicator PCB-congeners to the sum of the 6 remaining indicator PCBs. Only when we plotted CB 118 we got a pattern dividing our data in more or less 2 main groups. Individual results of 1587 eels from 305 locations in Flanders, Belgium.

After analyzing these results in more detail we found that for eels from a specific location the ratio of CB 118 to the sum of the remaining indicator PCBs was almost constant. At the same time this ratio varied considerably amongst different locations (Figure 2).

This variation could be depending on which commercial PCB-mixtures were used (probably related to specific industrial activities). On the other hand the PCB profile might be indicative for the “age” of the contamination. As PCB 153 is thought to be more persistent than PCB 118, high PCB 153 ratios could indicate old/older contaminations. But, probably the spatial variation is due to a combination of the commercial mixtures used and the “age” of the contamination.

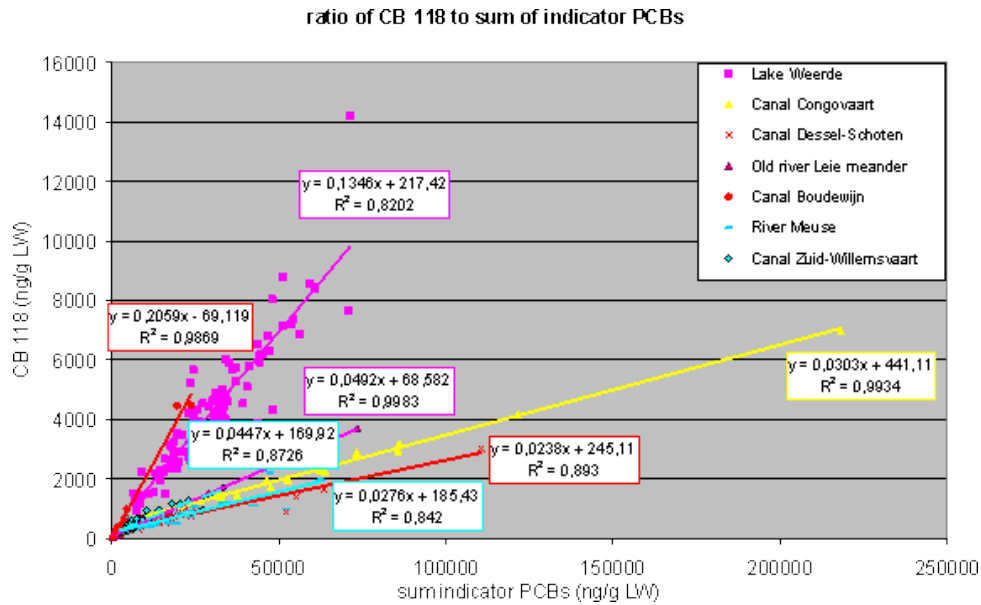


Figure 2: Variation in location specific ratio of CB 118 to the sum of the indicator PCBs. Selection of 337 eels from 7 waterbodies in Flanders, Belgium.

Temporal trend. Repeated measurements over time are only available for a limited series of waters. For these waters, we mostly observed a shift over time for the fraction of PCB 118 compared to the total of the other measured (non-dioxin like) PCBs. In most cases there was an increase in time for the importance of PCB 118, while the fraction of PCB 153 stayed more or less stable. These findings are in contrast with the common thought of PCB 153 being more persistent than PCB 118.

In Figure 3 a time trend is given for Lake Weerde, with exception of the year 2001 we notice a clear upwards shift in time. Before the year 2000 CB 118 accounted for +/- 10 % of the indicator PCBs, in 2001 about 15 % of the concentration of indicator PCBs was CB 118, in 2002 it accounted for almost 20 % of the indicator PCBs.

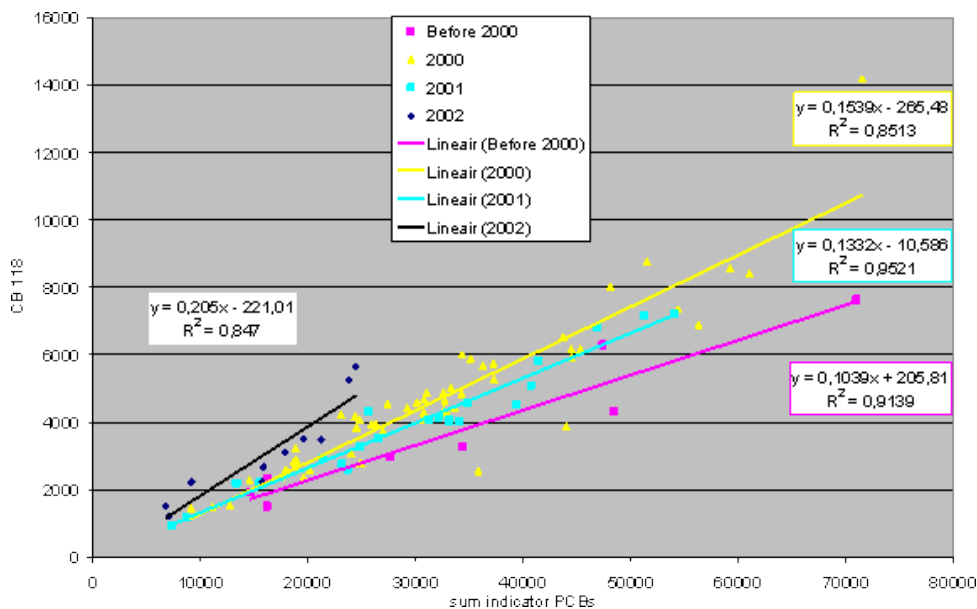


Figure 3: Temporal shift in relative fraction of PCB 118 compared to the sum of the indicator PCBs. Before 2000 CB 118 accounted for +/- 10 % of the indicator PCBs, by 2002 it accounted for almost 20 % of the indicator PCB's.

Conclusions

At this moment it is not very clear to us whether these spatial and temporal trends are due to the origin of the PCB-pollution or that it might be due to the higher persistence of CB 118, compared to the other indicator PCBs in eel (biota). It would be very interesting to do some controlled experiments to find out if CB 118 indeed is more persistent in biota than CB 153.

Acknowledgements

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References

1. Bligh E.G. & Dyer W.J. (1959). A rapid method of total lipid extraction and purification. *Can. J. Biochem. Physiol.*, 37: 911-917.
2. Goemans G., Belpaire C., Raemaekers M., Guns M., 2003. The Flemish eel pollution monitoring network 1994–2001: polychlorine biphenyls, organochlorine pesticides and heavy metals in eel. Institute for Forestry and Game Management, report. IBW.Wb.V.R.2003.99. (in Dutch) 169 p.
3. Goemans G., Belpaire C. (2004) The eel pollutant monitoring network in Flanders, Belgium. Results of 10 years monitoring. *Organohalogen Compounds*. 66: 1834 – 1840.
4. Morris S., Allchin C.R., Zegers B.N., Haftka J.J.H., Boon J.P., Belpaire C., Leonards P.E.G., Van Leeuwen S.P.J. and Boer, J. de (2004). Distribution and fate of HBCD and TBBPA brominated flame retardants in North Sea estuaries and aquatic food webs. *Environ. Sci. and Technol.* 38: 5497-5504.
5. Belpaire C., Goemans G., de Boer J. and Van Hooste H. (2003) Distribution of Brominated Flame Retardants. In: MIRA-T 2003: Report of the Environment and Nature in Flanders, Flemish Environmental Agency and Lannoo publishing, Heverlee, Belgium, pp 387-395. (in Dutch)
6. Roose P., Van Thuyne G., Belpaire C., Raemaekers M. en Brinkman U. (2003) Determination of VOCs in yellow eel from various inland water bodies in Flanders (Belgium). *J. Environ. Monit.* 5: 876-884.