

## **Bilag 61**

Note

## PCB-containing paint and plaster caused extreme PCB-concentrations in biota from the Sørffjord (Western Norway)—A case study

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### Abstract

The Sørffjord (Western Norway) has a long history of industry and pollution, and has been monitored for several decades. The environmental monitoring has comprised analyses of different contaminants in sea water, mussel, fish, seaweed and sediments. Measurements of polychlorinated biphenyls (PCBs) in blue mussels (*Mytilus edulis*) some 17 years ago indicated a local source. In 2001 severe concentrations were measured in blue mussels and further investigations disclosed the primary source of PCBs in the Sørffjord. In 2002, extreme PCB-concentrations were found in cod (*Gadus morhua*) from the same area. However, no induction of EROD activity was detected, indicating the limitations of this assay as marker for PCB contamination. A future scientific challenge will be to understand the specific mechanisms and effects of such PCB accumulation in fish.

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### 1. Introduction

The unique nature in the area of Odda and the Sørffjord (Western Norway) has rendered this area a tourist attraction since the end of the 19th century. The waterfalls from the mountain plateau Hardangervidda in combination with the deep and ice-free fjord have also made it possible for profitable industry in Odda. The hydro-electric power company A/S Tyssefaldene was established in 1906, and soon new industries were founded in the vicinity of Odda, producing calcium carbide (since 1908), calcium cyanamide (since 1909), aluminium (1916–1982), zinc (since 1929) and titanium oxide (since 1986). Environment concerns were to follow and the Sørffjord was at one time considered as one of the most metal polluted fjords in the world (Skei et al., 1972). Remedial actions in the area have included

disposal of metal containing sludge in mountain caverns and containment of contaminated sediment by the use of sheet pilings or capping.

The Norwegian State Pollution Monitoring Programme (NSPMP) in the Sørffjord and Hardangerfjord has been continuous since 1979, with the objective of monitoring the environmental changes following remedial actions, to support the environmental authorities in their assessment of potential needs for further remedial actions and to produce a foundation for the food safety authorities in their evaluation of the edibility of fish and shellfish. The programme has comprised analyses of different contaminants in sea water, mussels, fish, seaweed and sediments. The Sørffjord has also been monitored since 1987, through the international *Joint Assessment and Monitoring Programme* (JAMP) administrated by the Oslo–Paris Convention (OSPAR). Through this programme, some biological effect parameters (biomarkers) are also monitored (since 1997; Ruus et al., 2003).

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Industrial activities in the Odda area have been the major source of contamination of metals (mercury, cadmium, lead, zinc and copper), polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs). The point sources of the contaminants have been well known, except for PCBs.

## 2. Materials and methods

### 2.1. Study area

The Sør fjord is a southern branch of the Hardanger fjord in Western Norway. The municipality of Odda is located at the head of the fjord (N 60° 05'; E 06° 31').

### 2.2. Sampling

Blue mussels (*Mytilus edulis*) were sampled annually in shallow waters at several stations in the Sør fjord (through NSPMP and JAMP; Ruus and Green, 2002; Green et al., 2003). Pooled samples (of usually 50 individuals) were prepared for each station. Cod (*Gadus morhua*) were sampled annually (by trawling) in the inner Sør fjord (near Tyssedal, N 60° 07'; E 06° 33'; Green et al., 2003). Individual livers were sampled for analysis of PCBs and hepatic ethoxyresorufin-*O*-deethylase (EROD) activity (frozen at –20 °C and in liquid nitrogen, respectively).

### 2.3. Analyses

Analyses of PCBs were performed at NIVA's laboratory according to the method described by Brevik (1978), with some modifications. EROD activity was assayed fluorimetrically (Burke and Mayer, 1974) and normalised to protein content in the microsomal fraction, determined according to Lowry et al. (1951).

## 3. Results and discussion

The results of the monitoring of the Sør fjord early suggested the presence of a local source of PCB, as blue mussels near one site (Tyssedal) accumulated high concentrations ( $\sum\text{PCB} = 86 \text{ ng/g wet wt.}$ ; Skei et al., 1989). In 1990, a sediment survey and analyses of oil from electrical transformers previously used in the area were conducted to identify the potential source of the PCB. No specific PCB source was, however, found (Skei and Klungsoyr, 1990). Then, in 2000, semi-permeable membrane devices (SPMDs) were deployed at different localities in the Sør fjord and the highest amounts of PCBs were found at the station near Tyssedal (Skei and Tellefsen, 2000).

The PCB-concentrations in blue mussels near Tyssedal showed large variability through the years, and in 2001 a severe contamination was measured ( $\sum\text{PCB}_7 = 1132 \text{ ng/g wet wt.}$ ;  $59584 \text{ ng/g lipid wt.}$ ; Ruus and Green, 2002). For comparison, a suggested Norwegian reference “high

background” concentration for blue mussels is  $\sum\text{PCB}_7 = 3 \text{ ng/g wet wt.}$  (Green and Knutzen, 2003). The time of sampling corresponded with the renovation of the old power station of Tyssefaldene A/S, which was designated a national historical monument the year before. Old paint and plaster were removed from  $\approx 1500 \text{ m}^2$  of the facade of the 175 m long building, which is located on the shore of the fjord. A fibre sheet was applied to prevent release of waste material to the environment, however, some paint and plaster waste still entered the fjord.

Analysis of paint/plaster from the power station showed high concentrations of PCB ( $\sum\text{PCB}_7 \geq 336030 \text{ ng/g}$ ). During the 1960s the power station was painted with a product containing an unknown PCB-mixture. The PCB-profiles (relative concentrations of the congeners) of blue mussels sampled in 2001 and 2002 were similar to the PCB-profile of the paint/plaster, thus it was concluded that the paint/plaster was likely the main source of PCB in the Sør fjord biota (Ruus and Green, 2002).

In 2002, four (out of 25) cod sampled near Tyssedal had extremely high hepatic PCB-concentrations (Table 1). The individual with the highest PCB content ( $\sum\text{PCB}_7 = 427100 \text{ ng/g wet wt.}$ ) had an amount of PCB in the liver corresponding to 3.6‰ of the weight of the hepatic lipid. For comparison, a suggested Norwegian reference “high background” concentration for cod-liver is  $\sum\text{PCB}_7 = 500 \text{ ng/g wet wt.}$  (Green and Knutzen, 2003). Interestingly, the other 21 individuals did not show PCB-concentrations deviating from the concentrations common in this area (Green et al., 2003). Furthermore, these 21 individuals showed different PCB-profiles than the cod with the extreme PCB-concentrations, which were more similar to the profiles in the paint/plaster from the power station. Higher percentages of PCB-101 and –118, and lower percentages of –138, –153, and –180 were measured in the most contaminated cod, as compared to common PCB-mixtures (Frame et al., 1996) and to profiles in organisms at different trophic levels (Ruus et al., 2002; Fig. 1). A review of the profiles in cod-liver samples from Tyssedal 1999 to 2001 did not indicate such similarity to the paint/plaster profile (Green et al., 2003).

EROD is a measure of cytochrome P450 1A activity, which is induced by planar compounds, such as specific PCBs (primarily non-*ortho* substituted congeners), PAHs and polychlorinated dibenzo-*p*-dioxins (PCDDs) (Goksøyr and Förlin, 1992). Cod accumulating extreme PCB-concentrations generally did not show any elevated EROD activity in the present study (Table 1). This finding indicates the limitations of this assay as a biomarker of PCB contamination (at least for this combination of congeners), as discussed by Kennedy et al. (2003). The results are also in correspondence with findings of Besselink et al. (1998), who showed inhibition of TCDD-induced EROD activity by PCBs in flounder (*Platichthys flesus*).

A challenge will be to understand the specific mechanisms leading to the accumulation of such extreme PCB-concentrations in fish, as presented here, and effects on these indi-

Table 1  
Length, weight, liver lipid content, liver-PCB-concentrations and hepatic ethoxyresorufin-*O*-deethylase (EROD) activity in 25 cod caught in Sør fjorden (Western Norway) October 2002 (sorted with increasing PCB-concentrations)

Length (mm)	Weight (g)	Liver lipid (%)	$\Sigma$ PCB <sup>a</sup> (ng/g wet wt.)	$\Sigma$ PCB <sub>7</sub> <sup>b</sup> (ng/g wet wt.)	EROD (pg/min/mg prot.)
320	228	2.9	51.1	47.3	196.0
320	275	3.0	160.2	153.6	95.1
370	516	3.1	226.9	202.2	20.6
360	513	26.0	252.1	235.9	112.5
330	324	5.1	346.5	313.2	102.2
390	358	2.5	370.1	344.8	16.0
410	536	4.4	383.5	346.2	27.8
360	384	6.6	460.0	417.7	127.6
310	329	10.0	540.3	521.6	89.8
435	916	61.0	596.0	546.5	278.1
410	700	33.0	770.9	710.9	87.3
420	694	26.0	812.7	749.7	12.1
340	330	17.0	1052.7	955.7	155.7
390	536	54.0	1106.6	1006.0	223.3
330	360	23.0	1545.0	1470.0	32.3
670	2890	25.0	1965.1	1813.1	117.2
470	900	17.0	2148.2	1972.6	83.2
480	1095	30.0	2519.5	2421.5	174.0
350	459	38.0	2695.2	2414.7	93.9
490	1044	3.5	5989.3	5318.9	67.5
380	502	15.0	8860.7	7940.0	9.0
510	1058	12.0	16040.4	14270.4	41.1
440	797	8.6	31630.6	28130.3	60.3
410	679	8.0	37370.8	33270.3	460.1
440	796	12.0	47810.9	42710.0	121.6

<sup>a</sup> Sum of 10 congeners: PCB-28, -52, -101, -105, -118, -138, -153, -156, -180, -209.

<sup>b</sup> Sum of 7 congeners: PCB-28, -52, -101, -118, -138, -153, -180.

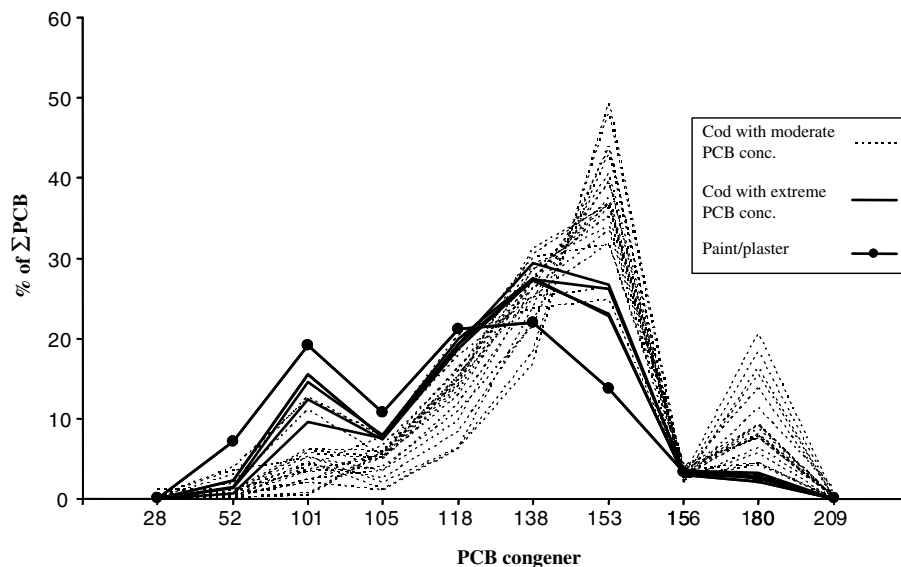


Fig. 1. PCB-profiles (% of sum concentration of 10 congeners) in paint/plaster from the old power station in Tyssedal (Western Norway), and in 25 cod caught in the Sør fjord (near Tyssedal) 2002. The PCB-concentrations in the cod-liver samples are presented in Table 1.

viduals. Dietary accumulation is a most likely mechanism (Thomann and Connolly, 1984). However, in the Sør fjord it is uncertain if prey organisms (or which prey organisms) have accumulated exceptionally high PCB-concentrations before being consumed by cod. There is also a possibility that some cod (feeding opportunistically) have consumed paint/plaster particles when it was in the water column.

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## References

- Besselink, H.T., Denison, M.S., Hahn, M.E., Karchner, S.I., Vethaak, A.D., Koeman, J.H., Brouwer, A., 1998. low inducibility of CYP1A activity by polychlorinated biphenyls (PCBs) in flounder (*Platichthys flesus*): Characterization of the Ah receptor and the role of CYP1A inhibition. *Toxicol. Sci.* 43, 161–171.
- Brevik, E.M., 1978. Gas chromatographic method for the determination of organochlorine pesticides in human milk. *Bull. Environ. Contam. Toxicol.* 19, 281–286.
- Burke, M.D., Mayer, R.T., 1974. Ethoxyresorufin: Direct fluorimetric assay of a microsomal *O*-dealkylation which is preferentially inducible by 3-methylcholanthrene. *Drug Metab. Dispos.* 2, 583–588.
- Frame, G.M., Cochran, J.W., Bøwadt, S.S., 1996. Complete PCB congener distributions for 17 Aroclor mixtures determined by 3 HRGC systems optimized for comprehensive, quantitative, congener-specific analysis. *J. High Resolut. Chromatogr.* 19, 657–668.
- Goksøyr, A., Förlin, L., 1992. The cytochrome *P*-450 system in fish, aquatic toxicology and environmental monitoring. *Aquat. Toxicol.* 22, 287–312.
- Green, N.W., Knutzen, J., 2003. Organohalogen and metals in marine fish and mussels and some relationships to biological variables at reference localities in Norway. *Mar. Pollut. Bull.* 46, 362–377.
- Green, N.W., Hylland, K., Ruus, A., Walday, M., 2003. Joint Assessment and Monitoring Programme (JAMP). National Comments regarding the Norwegian Data for 2002. Norwegian State Pollution Monitoring Programme Report no. 894/2003. TA-No. 2003/2003.
- Kennedy, S.W., Fox, G.A., Jones, S.P., Trudeau, S.F., 2003. Hepatic EROD activity is not a useful biomarker of polychlorinated biphenyl exposure in the adult herring gull (*Larus argentatus*). *Ecotoxicology* 12, 153–161.
- Lowry, O.H., Rosebrough, N.J., Farr, A.L., Randall, R.J., 1951. Protein measurement with the folin phenol reagent. *J. Biol. Chem.* 193, 265–275.
- Ruus, A., Green, N.W., 2002. Measure oriented environmental monitoring of the Sør fjord and Hardanger fjord 2002. Report component 2, Contaminants in organisms. Norwegian State Pollution Monitoring Programme Report no. 865/02. TA-No. 1922/2002 (in Norwegian).
- Ruus, A., Uglund, K.I., Skaare, J.U., 2002. Influence of trophic position on organochlorine concentrations and compositional patterns in a marine food web. *Environ. Toxicol. Chem.* 21, 2356–2364.
- Ruus, A., Hylland, K., Green, N.W., 2003. Joint Assessment and Monitoring Programme (JAMP). Biological effects methods, Norwegian Monitoring 1997–2001. Norwegian State Pollution Monitoring Programme, Report No. 869/03. TA-No. 1948/2003.
- Skei, J., Klungsoyr, J., 1990. Mapping of PCBs in sediments from the inner Sør fjord. NIVA-Report No. 2528-1990, NIVA, Oslo (in Norwegian).
- Skei, J., Tellefsen, T., 2000. Measure oriented environmental monitoring of the Sør fjord and Hardanger fjord 2000. Mapping of PCB in the inner Sør fjord using semi-permeable low density polyethylene membranes (LDPE-SPMDs). Norwegian State Pollution Monitoring Programme, Report No. 809/00. TA-No. 1769/2000 (in Norwegian).
- Skei, J.M., Brice, N.B., Calvert, S.E., Høltedahl, H., 1972. The distribution of heavy metals in sediments of the Sør fjord, West Norway. *Water Air Soil Pollut.* 1, 452–461.
- Skei, J., Knutzen, J., Næs, K., 1989. Measure oriented environmental monitoring of the Sør fjord and Hardanger fjord 1987–1988. Norwegian State Pollution Monitoring Programme, Report No. 346/89 (in Norwegian).
- Thomann, R.V., Connolly, J.P., 1984. Model of PCB in the Lake Michigan lake trout food chain. *Environ. Sci. Technol.* 18, 65–71.