

## Concentrations of lead in some coastal fishes from the Baltic Sea

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**Abstract.** Although the present mean concentrations of lead in various tissues and organs of the investigated coastal Baltic fishes are fairly low (0.05–1.04 mg kg<sup>-1</sup> dwt in muscle tissue, 0.05–1.29 mg kg<sup>-1</sup> dwt in liver, 0.05–0.95 mg kg<sup>-1</sup> dwt in gonads), i.e. lower than in the 1970s, they still indicate a state of pollution in the Baltic Sea. This justifies the monitoring of a selection of species, thus reflecting the contamination of their environment. In the sampling areas chosen for the study, fishes from the coastal waters of both sides of the Gulf of Finland and at the western Estonian coast appeared to be more contaminated with lead than their counterparts from the Finnish Archipelago Sea, the Gulf of Riga, and the Kiel Fjord. For flounder, however, such pattern was not so obvious. The highest concentrations of lead were analysed from perch (muscle 1.04 and liver 1.29 mg kg<sup>-1</sup> dwt) from the isolated coastal freshwater reservoir at Pargas-Parainen. Additionally, noteworthy concentrations of lead were calculated for the liver (0.97 mg kg<sup>-1</sup> dwt) of eelpout from Muuga and the gonads (0.95 mg kg<sup>-1</sup> dwt) of eelpout from Tvärminne, the kidneys (0.58 mg kg<sup>-1</sup> dwt) and bile (1.83 mg kg<sup>-1</sup> dwt) of flounder from the Kiel Fjord, and the bile (1.54 mg kg<sup>-1</sup> dwt) of flounder from the Åland Islands. Nevertheless, all investigated fishes were safe for human consumption.

**Key words:** lead, coastal fishes, Baltic Sea.

### INTRODUCTION

In contrast to other harmful metals, as e.g. mercury and cadmium, the concentrations of lead in coastal fishes from the Baltic Sea have attracted far less attention (e.g. von Westernhagen and Bignert, 1996; Jezierska and Witeska, 2001; Szefer, 2002; Pokorska et al., 2012; Nyberg et al., 2013; Boalt et al., 2014) although effects of this toxic metal on humans (e.g. Guinee, 1972; Bremner, 1974; Brian et al., 1980; IPCS, 1989; ATSDR, 2005), fishes, mammals, and birds (e.g. Haider, 1964, 1977; NRCC, 1979; Atchison et al., 1987; Eisler, 1988; Hofer and Lackner, 1995; Pain, 1996; Dietz et al., 1998) have been documented. Considering the significant inflow of lead into the sea and over its vast drainage area (e.g. Rühling et al., 1992; HELCOM, 1997, 2003, 2010; Bartnicki et al., 2000; Buse et al., 2003) this is rather surprising.

In the 1970s some investigations on the concentrations of lead in certain species of Baltic fish, mainly Baltic herring (*Clupea harengus membras* L.) and cod (*Gadus morhua callaris* L.), were performed (e.g. Lehtonen, 1973; Voipio

et al., 1977; Tervo et al., 1980; Perttilä et al., 1982), followed by attempts at its monitoring, mainly in the muscle tissue of herring and in the liver of cod, for the Baltic Marine Environment Protection Commission (Helsinki Commission, HELCOM) by some Baltic national research institutes, e.g. the Finnish Institute of Marine Research (Haahti, 1991), the Swedish Museum of Natural History (Jorhem and Sundström, 1993), and the Estonian Marine Institute (Jankovski et al., 1996). In the southern Baltic Sea similar investigations additionally included sprat (*Sprattus sprattus balticus* Schn.), flounder (*Platichthys flesus* L.), perch (*Perca fluviatilis* L.), and some other species (Falandysz and Lorenc-Biała, 1984). After confirmation that lead mainly concentrates in internal organs, such as kidney and liver, and bone tissue of fish (e.g. Reichenbach-Klinke, 1980), and not in muscle tissue like mercury, the interest in such monitoring declined significantly, especially because the observed concentrations of lead in the muscle tissue of fish were far below the accepted security levels for fish as food for human consumption (e.g. Nuutamo et al., 1980; Haahti, 1991; Tahvonen and Kumpulainen, 1996; Leivuori, 2007). Starting again in the 1980s, monitoring of the concentrations of lead has been continued, with mainly liver and muscle tissue of herring studied (e.g. Jankovski et al., 1996; Roots and Simm, 2002; Lind et al., 2006; Leivuori, 2007).

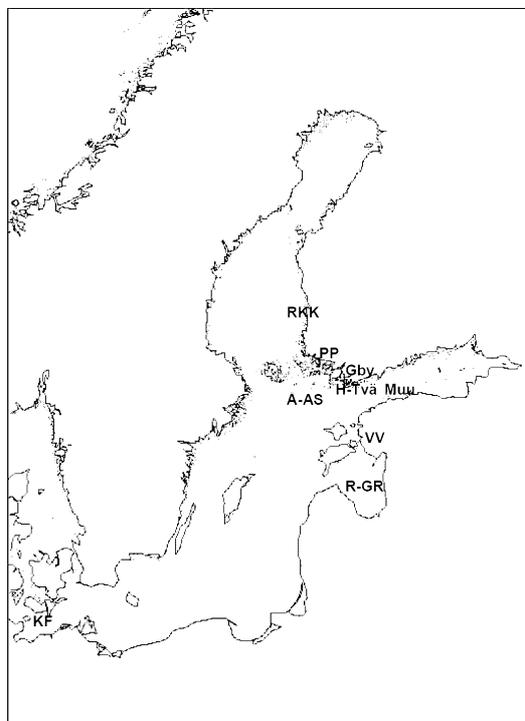
The present study focuses on the concentrations of lead in the muscle tissue and some internal organs, mainly liver, but in some cases also in gonads, kidneys, spleen, and bile, of some representative coastal fish species from various parts of the Baltic Sea, including two coastal inlets in SW Finland (isolated in the late 1950s from the Baltic Sea). The aim is to survey and to compare the concentrations of lead in the similar fish species as in previous studies from various parts of the Baltic Sea (Voigt, 1999, 2000a, 2000b, 2003, 2004, 2008a, 2008b, 2013).

## MATERIAL AND METHODS

Some abundant coastal fish species (adults and mainly of comparable size for each species separately, with the exception for the fishes from two isolated freshwater reservoirs, see below) from various parts of the Baltic Sea (Fig. 1) were sampled in the autumn season during the years 1997–2006 for analysis of concentrations of lead in muscle tissue and some internal organs, mainly liver and gonads. Besides bow-nets and gillnets trawling was used for sampling.

The following five fish species were focused upon:

- Baltic herring, sampled from Peimari-Pemarn in the Finnish Archipelago Sea (Å-AS), Tvärminne area at the Finnish SW coast of the Gulf of Finland (H-Tvä), and Väike Vain Strait at the western Estonian coast (VV);
- smelt (*Osmerus eperlanus* L.) from the rivermouth area of the Kokemäenjoki-Kumoälv in the Bothnian Sea (RKK), Peimari-Pemarn (Å-AS), Tvärminne (H-Tvä), Väike Vain (VV), and south-western coastal freshwater basin of Pargas-Parainen (PP);



**Fig. 1.** Sampling stations around the Baltic Sea: RKK = rivermouth area of the Kokemäenjoki-Kumoälvi, PP = freshwater reservoir at Pargas-Parainen, Gby = freshwater basin of Gennarbyviken, Å-AS = Åland Islands and the Archipelago Sea, H-Tvä = Hanko-Hangö Peninsula including Tvärminne, Muu = Muuga Bay, VV = Väike Väin Strait, R-GR = Roja area of the Gulf of Riga, KF = Kiel Fjord.

- perch from Tvärminne (H-Tvä), Väike Väin (VV), Pargas-Parainen (PP), and the southern coastal freshwater basin of Gennarby (Gby);
- eelpout (*Zoarces viviparus* L.) from Brunskär in the Archipelago Sea (Å-AS), Tvärminne (H-Tvä), Bay of Muuga on the northern Estonian coast of the Gulf of Finland (Muu), the Roja area of the Latvian Gulf of Riga (R-GR), and the German Kiel Fjord at the southern Baltic coast (KF);
- flounder from Kokemäenjoki-Kumo (RKK), Nåtö and Åland Islands in the Archipelago Sea (Å-AS), Tvärminne (H-Tvä), and the Kiel Fjord (KF).

All these species have been considered acceptable bioindicators for heavy metals in the environment (e.g. Voigt, 2004).

Occasional by-catches of the following 12 fish species were also analysed:

- sprat from Tvärminne (H-Tvä) and Väike Väin (VV);
- pike (*Esox lucius* L.) from Pargas-Parainen (PP) and Gennarby (Gby);
- roach (*Rutilus rutilus* L.) from Tvärminne (H-Tvä) and Pargas-Parainen (PP);
- ide (*Leuciscus idus* L.) from Tvärminne (H-Tvä);
- bleak (*Alburnus lucidus* L.) from Tvärminne (H-Tvä) and Väike Väin (VV);

- bream (*Abramis brama* L.) from Tvärminne (H-Tvä) and Pargas-Parainen (PP);
- Crucian carp (*Carassius carassius* L.) from Tvärminne (H-Tvä);
- ruffe (*Gymnocephalus cernuus* L.) from Tvärminne (H-Tvä), Väike Väin (VV), and Pargas-Parainen (PP);
- pikeperch (*Stizostedion lucioperca* L.) from Tvärminne (H-Tvä) and Pargas-Parainen (PP);
- burbot (*Lota lota* L.) from Tvärminne (H-Tvä), Pargas-Parainen (PP), and Gennarby (Gby);
- bull-rout (*Myoxocephalus scorpius* L.) from Tvärminne (H-Tvä);
- turbot (*Psetta maxima* L.) from Nätö (Å-AS) and Tvärminne (H-Tvä).

These additional species were included in the study in order to verify (i.e. to contradict or support) the obtained results for the five main species.

Prior to analyses of the abdominal muscle tissue and the removed internal organs, mainly liver and gonads and in some cases also kidneys, spleen and bile (flounder and turbot), the fish were stored at  $-20^{\circ}\text{C}$ . After treatment with acids ( $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$ ), the samples were analysed individually (herring, smelt, perch, eelpout, and flounder from the sea). The other 12 species from the reservoirs together with the gonads of herring and smelt from the sea were analysed as pooled samples (homogenized tissues and organs, separately treated as individual samples described above). The AAS-technique was used: Varian-SpectrAA 400, equipped with a graphite furnace; GTA-96, ETAAS (Voigt, 2000a, 2003).

The certified and obtained values for the reference materials were Pb certified  $0.08 \pm 0.015$  and Pb obtained  $0.09 \pm 0.06 \text{ mg kg}^{-1} \text{ dwt}$ .

All samples were analysed in duplicate. The accuracy was assessed by using blanks (5 per each sequence of 40 samples) and reference material BCR CRM-422 (cod muscle) (Quevauviller et al., 1993). All results are expressed in  $\text{mg Pb kg}^{-1} \text{ dwt}$  (dry weight).

## RESULTS

The mean concentrations of lead ( $\text{mg kg}^{-1} \text{ dwt}$ ) in the muscle tissue, liver, and gonads of the five selected fishes are presented in Tables 1, 2, and 3, respectively. For flounder in some cases additionally also other internal organs, such as kidneys, spleen, and bile, were analysed for the concentrations of lead (Table 4). The concentrations of lead in the muscle tissue and liver of the analysed additional species are given in Tables 1a and 2a, respectively.

The mean concentrations of lead in the muscle tissue of the investigated fishes decreased in the order perch (Pargas-Parainen), Crucian carp (Tvärminne), eelpout (Muuga), bleak (Väike Väin), burbot (Gennarby), smelt (Väike Väin), perch (Väike Väin), Baltic herring (Väike Väin), ide (Tvärminne), burbot (Pargas-Parainen) (Tables 1 and 1a). All these concentrations exceeded the value of  $0.20 \text{ mg kg}^{-1} \text{ dwt}$ .

For the mean concentrations of lead in the liver of the investigated fishes the decreasing order is: perch (Pargas-Parainen), eelpout (Muuga), Crucian carp (Tvärminne), roach (Tvärminne), burbot (Pargas-Parainen), burbot (Gennarby),

**Table 1.** Mean concentrations of lead ( $\text{mg kg}^{-1}$  dwt) in the muscle tissue of the sampled five main species (a) and 12 additionally analysed species (b). For sampling areas see Material and Methods. SD = standard deviation,  $N$  = number of fish analysed, – not sampled

(a)

Area	Herring (SD, $N$ )	Smelt (SD, $N$ )	Perch (SD, $N$ )	Eelpout (SD, $N$ )	Flounder (SD, $N$ )
RKK	–	0.16 (0.08, 20)	–	–	0.10 (0.12, 28)
Å-AS	0.12 (0.06, 6)	0.12 (0.04, 7)	–	0.18 (0.11, 21)	0.11 (0.08, 29)
H-Tvä	0.10 (0.09, 15)	0.16 (0.16, 14)	0.10 (0.15, 5)	0.18 (0.22, 23)	0.08 (0.03, 13)
Muu	–	–	–	0.37 (0.26, 27)	–
VV	0.25 (0.06, 10)	0.32 (0.03, 12)	0.28 (0.36, 5)	–	–
R-GR	–	–	–	0.11 (0.09, 16)	–
KF	–	–	–	0.19 (0.30, 41)	0.18 (0.21, 24)
PP	–	0.12 (–, 30)	1.04 (–, 8)	–	–
Gby	–	–	0.05 (–, 5)	–	–

(b)

Area	Sprat ( $N$ )	Pike ( $N$ )	Roach ( $N$ )	Ide ( $N$ )	Bleak ( $N$ )	Bream ( $N$ )	Crucian carp ( $N$ )	Ruffe ( $N$ )	Pike- perch ( $N$ )	Burbot ( $N$ )	Bull- rout ( $N$ )	Turbot ( $N$ )
H-Tvä	0.05 (5)	–	0.08 (6)	0.22 (7)	0.05 (6)	0.05 (14)	0.40 (3)	0.05 (5)	0.10 (4)	0.05 (2)	0.10 (5)	0.07 (6)
VV	0.10 (5)	–	0.05 (5)	–	0.35 (5)	–	–	0.11 (7)	–	–	–	–
PP	–	0.09 (9)	0.18 (5)	–	–	0.18 (6)	–	0.05 (10)	0.05 (4)	0.20 (2)	–	–
GBy	–	0.05 (7)	–	–	–	–	–	–	–	0.34 (2)	–	–
Å-As	–	–	–	–	–	–	–	–	–	–	–	0.05 (7)

eelpout (Roja), smelt (Tvärminne), ide (Tvärminne), eelpout (Tvärminne), smelt (Väike Väin), smelt (Kokemäenjoki-Kumoälv) (Tables 2 and 2a). These concentrations were also all higher than  $0.20 \text{ mg kg}^{-1}$  dwt.

The mean concentrations of lead exceeding the value of  $0.20 \text{ mg kg}^{-1}$  dwt in the gonads were all determined in four main species sampled. The decreasing order is: eelpout (Tvärminne), perch (Pargas-Parainen), eelpout (Muuga), eelpout (Roja), eelpout (Kiel Fjord), smelt (Tvärminne), flounder (Kiel Fjord) (Table 3).

The mean concentrations of lead in the gonads of the additionally analysed species were low: for pike Gennarby =  $0.05$  ( $N = 7$ ) and turbot Tvärminne =  $0.05$  ( $N = 7$ )  $\text{mg kg}^{-1}$  dwt.

The mean concentrations of lead in other inner organs (kidneys, spleen, and bile) for flounder were from  $0.20$  to  $1.83 \text{ mg kg}^{-1}$  dwt (Table 4). The mean concentration of lead in the kidneys of turbot from Nätö was  $0.10$  ( $N = 7$ ) and from Tvärminne  $0.20$  ( $N = 7$ )  $\text{mg kg}^{-1}$  dwt.

**Table 2.** Mean concentrations of lead ( $\text{mg kg}^{-1}$  dwt) in the liver of the sampled five main species (a) and some additionally analysed species (b). For sampling areas see Material and Methods. SD = standard deviation,  $N$  = number of fish analysed, – not sampled

(a)

Area	Herring (SD, $N$ )	Smelt (SD, $N$ )	Perch (SD, $N$ )	Eelpout (SD, $N$ )	Flounder (SD, $N$ )
RKK	–	0.23 (0.14, 18)	–	–	–
Å-AS	0.10 (0.12, 6)	0.10 (0.12, 7)	–	0.20 (0.11, 20)	0.16 (0.08, 29)
H-Tvä	0.20 (0.21, 10)	0.28 (0.24, 14)	–	0.24 (0.19, 20)	0.14 (0.08, 13)
Muu	–	–	–	0.97 (0.29, 27)	–
VV	0.20 (0.23, 10)	0.24 (0.18, 12)	0.12 (0.08, 5)	–	–
R-GR	–	–	–	0.29 (0.16, 26)	–
KF	–	–	–	0.19 (0.23, 39)	0.20 (0.35, 24)
PP	–	0.05 (–, 20)	1.29 (–, 8)	–	–
Gby	–	–	0.05 (–, 5)	–	–

(b)

Area	Pike ( $N$ )	Roach ( $N$ )	Ide ( $N$ )	Bream ( $N$ )	Crucian carp ( $N$ )	Ruffe ( $N$ )	Burbot ( $N$ )	Bull- rout ( $N$ )	Turbot ( $N$ )
H-Tvä	–	0.55 (6)	0.25 (7)	–	0.70 (3)	–	0.10 (2)	0.20 (5)	0.20 (7)
PP	0.07 (5)	–	0.19 (5)	0.18 (6)	–	0.05 (6)	0.50 (2)	–	–
VV	–	0.20 (5)	–	–	–	–	–	–	–
Å-As	–	–	–	–	–	–	–	–	0.05 (7)
GBy	0.06 (7)	–	–	–	–	–	0.50 (2)	–	–

**Table 3.** Mean concentrations of lead ( $\text{mg kg}^{-1}$  dwt) in the gonads of the sampled five main species. For sampling areas see Material and Methods. SD = standard deviation,  $N$  = number of fish analysed, – not sampled

Area	Herring ( $N$ )	Smelt ( $N$ )	Perch ( $N$ )	Eelpout (SD, $N$ )	Flounder (SD, $N$ )
RKK	–	0.15 (8)	–	–	–
Å-AS	0.07 (6)	0.05 (7)	–	–	0.15 (0.16, 14)
H-Tvä	0.10 (10)	0.25 (14)	–	0.95 (0.06, 15)	0.10 (0.06, 13)
Muu	–	–	–	0.44 (0.67, 15)	–
VV	0.11 (10)	0.19 (10)	–	–	–
R-GR	–	–	–	0.33 (0.30, 6)	–
KF	–	–	–	0.27 (0.22, 32)	0.25 (0.37, 24)
PP	–	0.05 (12)	0.70 (4)	–	–
Gby	–	–	0.05 (5)	–	–

**Table 4.** Mean concentrations of lead ( $\text{mg kg}^{-1}$  dwt) in kidneys (R), spleen (S), and bile (B), of flounder. For sampling areas see Material and Methods. SD = standard deviation,  $N$  = number of fish analysed

Area	PbR (SD, $N$ )	PbS (SD, $N$ )	PbB (SD, $N$ )
ÅA-S	0.50 (0.34, 22)	0.50 (0.55, 20)	1.54 (0.89, 6)
H-Tvä	0.20 (0.23, 13)	0.28 (0.63, 10)	0.92 (0.79, 5)
KF	0.61 (0.55, 24)	0.41 (0.23, 14)	1.83 (1.71, 6)

Comparison of the mean concentrations of lead in the muscle tissue, liver, and gonads of fishes analysed revealed differences between the sampling areas.

1. Lead in muscle tissue (PbM): Most fishes from the Väike Väin (herring, smelt, perch, sprat, ruffe) apparently were, with the exception of the two freshwater reservoirs (Pargas-Parainen, Gennarby), more contaminated with lead in muscle tissue than the corresponding species at the other two coastal sampling stations (Peimari-Pemarn, Tvärminne). For roach, however, the opposite seems to be the case. In addition notable concentrations of PbM were calculated for perch and burbot from the Pargas-Parainen reservoir in contrast to the comparably low concentrations of smelt, pike, ruffe, pikeperch, roach, and bream from the same basin as well as pike and perch from the Gennarby reservoir. Notable concentrations of PbM were also calculated for ide and Crucian carp, both from Tvärminne, and eelpout from Muuga. For flounder the means of PbM were of the same modest order of magnitude from all sampling stations. Even lower mean values were calculated for PbM of sprat, herring, smelt, perch, flounder, ruffe, pikeperch, roach, burbot, bull-rout, and turbot from Tvärminne.
2. Lead in liver (PbL): The calculated mean concentrations of lead in liver were of the same low order of magnitude for herring, smelt, ide, bream, and bull-rout from Tvärminne as for herring, smelt, and roach from the Väike Väin, smelt from Kokemäenjoki-Kumoälvi, and eelpout from Brunskär. For flounder the values for PbL were of the same low order of magnitude from all sampling stations (Nätö, Tvärminne, Kiel Fjord). Moreover, they all were considerably lower than the comparably higher means for roach, eelpout, and Crucian carp from Tvärminne, eelpout from Muuga, perch and burbot from Pargas-Parainen. All these contrasted the obviously low values for PbL in herring, smelt, eelpout, flounder, and turbot from Åland and the Archipelago Sea; perch and roach from the Väike-Väin; burbot from Tvärminne; smelt, pike, ruffe, and pikeperch from Pargas-Parainen; and pike and perch from the Gennarby basin.
3. Lead in gonads (PbG): Notably high concentrations of PbG were calculated for smelt and eelpout from Tvärminne, eelpout from Muuga and the Kiel Fjord as well as for flounder. The concentrations of PbG were also high for perch from Pargas-Parainen. Fairly modest concentrations were calculated for flounder from Nätö, smelt from the Väike Väin and from Kumo-Kokemäenjoki. The values were even lower for herring and smelt from Tvärminne, smelt from Pargas-Parainen, perch from Gennarby, and flounder and turbot from Tvärminne.

As the concentrations of lead equal to or below  $0.05 \text{ mg kg}^{-1}$  dwt are close to the detection limit of the technical equipment used, they are considered as 'uncertain values'. Consequently, the mean values of and below  $0.5 \text{ mg kg}^{-1}$  dwt are treated only as 'orders of magnitude' when comparing the results from various sampling stations. For this reason no statistical treatment of the results was performed.

## DISCUSSION

Bearing in mind the limitations regarding the obtained results and the recognized difficulties involved in analyses of low concentrations of lead in biotic materials (e.g. Martin and Coughtrey, 1982), there still are noteworthy distinctions in the calculated mean concentrations between the sampling stations.

The most contaminated species was perch from the Pargas-Parainen reservoir with high concentrations calculated for the whole fish (PbM, PbL, PbG). It was followed by eelpout from Muuga and Tvärminne, then by eelpout from Roja and the Kiel Fjord. The eelpout was least contaminated at Brunskär. Flounder in general were less contaminated with lead than eelpout at all sampling stations.

The highest concentrations of Pb in flounder were calculated from the bile (PbB) at all its three sampling stations the Kiel Fjord, Nåtö, and Tvärminne; followed by kidney (PbR) at the Kiel Fjord, Nåtö, and Tvärminne; and spleen (PbS) at Nåtö, the Kiel Fjord, and Tvärminne.

With the exception of the considerably high mean values calculated for lead in the muscle tissue, liver, and gonads of perch and in the liver of burbot from the freshwater Pargas-Parainen reservoir and the corresponding lower although still notable values for the liver and gonads of eelpout from Muuga and Tvärminne, the other results mainly are of such an uncertain character as stated before (Results). This probably justifies the decision not to treat the material statistically.

Thus, comparison of the obtained data shows that the pelagic and semi-pelagic species such as sprat, herring, and smelt apparently are more contaminated with lead in the West Estonian Väike Väin and at the inlet to the Gulf of Finland at Tvärminne than in the Finnish Archipelago Sea. The mean concentrations of lead in the eelpout (muscle, liver, and gonads) from Muuga and the eelpout and smelt (muscle, liver, and gonads) from the Tvärminne area were also distinct from most corresponding results from the other sampling stations (Finnish Archipelago Sea, Gulf of Riga, and Kiel Fjord).

For flounder, only the calculated mean concentrations of lead in liver were higher at Tvärminne compared to the mean concentrations in muscle, gonads, kidneys, spleen, and bile of flounder from the Åland Islands and the Kiel Fjord. Such a pattern is different from the results for the other fishes studied.

As heavy metal accumulation in fish mainly takes place via consumption (e.g. Reichenbach-Klinke, 1980), the contradictory results regarding the concentrations of lead may be due to different food and migration habits of both flounder and eelpout in the sampling areas of this study. Flounder and eelpout in the Kiel area and in the Gulf of Riga, respectively, consume a broad spectrum of benthos including

several species of polychaete worms and mussels (Arntz, 1978; Urtans, 1990) in contrast to feeding on mainly mussels (*Macoma balthica*, *Mytilus edulis*) at both Nåtö, Åland, and at Tvärminne, and including mainly benthic crustaceans for eelpout from the northern sampling areas (Urtans, 1990; and the author's unpublished data). Besides, in the southern Baltic Sea flounder is migrating widely when feeding and spawning, in contrast to its mainly stationary behaviour in the northern Baltic Sea (Aro, 1989). For eelpout the extremely stationary behaviour has been well known for a long time (Schmidt, 1917).

The high mean values of lead in the gonads of smelt and eelpout from Tvärminne and in the gonads of perch from Pargas-Parainen support the assumption that both these sampling areas are contaminated with lead. The fishes from both freshwater reservoirs were also considerably more contaminated with other heavy metals analysed previously (e.g. mercury, copper, cadmium, nickel) compared to the same species outside the reservoirs (Voigt, 2000a, 2008b).

Assuming that fish reflect the contamination with e.g. heavy metals in their environment (e.g. Vogt and Kittelberger, 1976; Haider, 1980; Polak-Juszczak, 1996; Voigt, 2004), the modest and comparably low concentrations of lead in the analysed organs and tissues of fishes from the Gulf of Riga (eelpout) and the Kiel Fjord (eelpout and flounder) may indicate a lower contamination with lead of these sampling areas compared to the other sampling areas.

The available data on mean concentrations of lead in the surface sediments from the various sampling stations (Nåtö, Åland = 25 mg kg<sup>-1</sup> dwt (author's unpublished data), Seili-Själö, Archipelago Sea = 30 mg kg<sup>-1</sup> dwt (Müller, 1999), Tvärminne = 53 mg kg<sup>-1</sup> dwt (Voigt, 2007), Gulf of Riga = 31 mg kg<sup>-1</sup> dwt (Leivuori et al., 2000), Pargas-Parainen reservoir = 45 mg kg<sup>-1</sup> dwt (Voigt, 2000a)), however, with the exception of Tvärminne, do not support verdicts in any direction. The Tvärminne area has been influenced by various metal emissions from a minor local iron and steel factory since the 1960s, which is reflected both in sediments and biota, mainly regarding cadmium (e.g. Voigt, 2004, 2007).

For coastal fishes off Hanko-Hangö, SW Finland, sampled in the 1970s, the mean concentrations of lead in muscle tissue were varying: for herring ca 0.7–1.25, smelt ca 0.75, flounder ca 0.4–0.60 mg kg<sup>-1</sup> dwt (Karppanen and Stabel-Taucher, 1976; Voipio et al., 1977; Tervo et al., 1980).

In the same period, for smelt from Tvärminne, the mean concentrations of lead were calculated as 0.22 (muscle), 0.89 (liver), and 0.90 (gonads) mg kg<sup>-1</sup> dwt (Voigt, 1999). The corresponding mean calculated for muscle tissue of herring from the Helsinki region varied between ca 1.9 and 2.4 mg kg<sup>-1</sup> dwt (Lehtonen, 1973). For flounder from the Archipelago Sea the mean concentration of lead in muscle tissue was ca 0.45 mg kg<sup>-1</sup> dwt (Miettinen and Verta, 1978). Consequently, all the calculated mean values of concentrations of lead in the muscle tissue of the analysed fishes were considerably higher in the 1970s than at present (Tables 1–4).

Regarding herring from the open waters between Finland and Estonia the present mean concentrations of lead in both muscle tissue and liver are even notably lower: in muscle tissue ca 0.01–0.05 and in liver ca 0.07 mg kg<sup>-1</sup> dwt, which are close to the detection limit of ca 0.01 mg kg<sup>-1</sup> dwt (Leivuori, 2007).

In herring and flounder from the Estonian coastal waters of the Gulf of Finland (1973–1999) the mean concentrations of lead in both muscle tissue and liver are in some cases higher than reported in the material from Finland, although still mainly of the same order of magnitude, i.e. herring PbM = ca 0.2–0.45 and PbL ca 0.3–0.51 mg kg<sup>-1</sup> dwt, and flounder PbM = ca 0.15–0.60 and PbL 0.47–0.58 mg kg<sup>-1</sup> dwt, respectively. However, for gonads the values differ remarkably: in herring PbG = ca 0.25–3.1 and in flounder PbG = ca 0.40 mg kg<sup>-1</sup> dwt (Jankovski et al., 1996). With the exception of the year 1998 (when an extremely high mean value of ca 3.0 mg kg<sup>-1</sup> dwt was calculated), the mean concentration of lead in the liver of herring from the Bay of Tallinn varied between 0.25 and 0.75 mg kg<sup>-1</sup> dwt in the years 1994–2001; the highest value was calculated for 1994. Also in the Gulf of Finland off Kunda, eastwards from Tallinn, the value of lead in the liver of herring varied between ca 0.25 and 0.75 mg kg<sup>-1</sup> dwt during the period of 1994–2001 (Roots and Simm, 2002). In Pärnu Bay, northern Gulf of Riga, the corresponding values are for herring PbM = ca 0.35, PbL = ca 0.47, and PbG = ca 2.25 mg kg<sup>-1</sup> dwt, respectively (Jankovski et al., 1996).

In herring from the Polish southern coast of the Baltic Sea (1991–1997) the mean value of lead in the muscle tissue of herring was ca 0.05–0.60 and in the muscle tissue of flounder ca 0.05–0.50 mg kg<sup>-1</sup> dwt (e.g. Pempkowiak and Szefer, 1992; Polak-Juszczak, 2000), in the liver and kidneys of flounder (1984–1993) ca 1.1–2.5 and ca 4.0 mg kg<sup>-1</sup> dwt, respectively (Protasowicki, 1989; Polak-Juszczak and Domagała, 1994), i.e. in several cases higher than in the present material (Table 1), although with a tendency towards decreasing in time. The same tendency has been observed for most analysed fishes from all over the Baltic Sea (e.g. Roots and Simm, 2002; Lind et al., 2006; Leivuori, 2007; HELCOM, 2010; Nyberg et al., 2013; Boalt et al., 2014). In a recent study of herring and flounder from the southern Polish Baltic Sea in 2009, this decline of the concentrations of lead in both species was even more pronounced: for the muscle tissue of herring the mean value was calculated to 0.7 mg kg<sup>-1</sup> dwt and for flounder to ca 0.11 mg kg<sup>-1</sup> dwt (Pokorska et al., 2012). The corresponding mean values for the lead concentration in the liver of these species were 0.06 and 0.03 mg kg<sup>-1</sup> dwt, respectively (Pokorska et al., 2012). All these concentrations are remarkably lower than the previously published values, but also the relevant data given in Tables 1–4.

## CONCLUSIONS

The obtained concentrations of lead in muscle tissue, liver, and gonads from several abundant coastal Baltic fish species are, compared to previous investigations of the corresponding fish species from adjacent areas, notably lower than in the 1970s. This finding supports the earlier statements of decreasing temporal trends by the Swedish Museum of Natural History for the period 1969–2011 (Lind et al., 2006; Boalt et al., 2014) and by HELCOM (e.g. HELCOM Secretariat, 2008; Nyberg et al., 2013).

Fish from large reservoirs isolated from the Baltic Sea may be more contaminated with lead than fish sampled outside the basins. The same was previously shown for some other heavy metals (e.g. Voigt, 2000a, 2008a).

The stipulated safety level,  $0.30 \text{ mg kg}^{-1}$  fwt, corresponding to ca  $1.5 \text{ mg kg}^{-1}$  dwt, in muscle tissue, for fish as food for humans (European Commission, 2006) was not exceeded in any sample.

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### REFERENCES

- Arntz, W. E. 1978. Predation on benthos by flounders (*Platichthys flesus* L.) in the deeper parts of Kiel Bay. *Meeresforschung*, **26**(1), 70–78.
- Aro, E. 1989. A review of fish migration patterns in the Baltic. *Rapports et procès-verbaux des réunions/Conseil permanent international pour l'exploration de la mer*, **190**, 72–96.
- Atchison, G. J., Henry, M. G., and Sandheinrich, M. B. 1987. Effects of metals on fish behavior: a review. *Environmental Biology of Fishes*, **18**(1), 11–35.
- [ATSDR] Agency for Toxic Substances and Disease Registry. 2005. *Toxicological Profile for Lead*. US Department for Health and Human Services, Public Health Service, Atlanta, Georgia.
- Bartnicki, J., Gusev, A., Pavlova, N., Ilyin, I., Lükewille, A., and Barrett, K. 2000. *Atmospheric Supply of Nitrogen, Lead, Cadmium, Mercury and Lindane to the Baltic Sea in 1997*. EMEP (European Monitoring and Evaluation Programme) Centres Joint Note for HELCOM. Research Report No. 97. Norwegian Meteorological Institute, Oslo, Norway.
- Boalt, E., Miller, A., and Dahlgren, H. 2014. Distribution of cadmium, mercury, and lead in different body parts of Baltic herring (*Clupea harengus*) and perch (*Perca fluviatilis*): Implications for environmental status assessments. *Marine Pollution Bulletin*, **78**, 130–136.
- Bremner, I. 1974. Heavy metal toxicities. *Quarterly Review of Biosphysics*, **7**(1), 75–124.
- Brian, B. J., Smith, S., and Coleman, D. O. 1980. *Lead Pollution of the Global Environment*. A Technical Report. MARC (Monitoring and Assessment Research Centre) Reports, **16–18**(1), 1–31.
- Buse, A., Norris, D., Harmens, H., Büker, P., Ashenden, T., and Mills, G. 2003. *Heavy Metals in European Mosses: 2000/2001 Survey – UNECE ICP Vegetation*. CEH (Centre for Ecology and Hydrology) – Natural Environment Research Council, University of Wales Bangor, Bangor Gwynedd LL57 2UP, 1–45.
- Dietz, R., Pacyna, J., and Thomas, D. J. 1998. Heavy metals. In *AMAP Assessment Report: Arctic Pollution Issues*. Arctic Monitoring and Assessment Program (AMAP), Oslo, pp. 373–527.
- Eisler, R. 1988. *Lead Hazards to Fish, Wildlife, and Invertebrates: A Synoptic Review*. Contaminant Hazard Reviews, Report No. 14. Biological Report 85(1.14). Fish and Wildlife Service, U.S. Department of the Interior, Laurel, MD.

- European Commission. 2006. Commission Regulation (EC) 1881/2006, 20.12.2006. *Official Journal of the European Union L*, 345, 5–24.
- Falandysz, J. and Lorenc-Biała, H. 1984. Trace metals in fish from the southern Baltic. *Meeresforschung*, **30**(29), 111–119.
- Guinnee, V. F. 1972. Lead poisoning. *The American Journal of Medicine*, **52**, 283–288.
- Haahti, H. 1991. Concentration of harmful substances in fish in the northern Baltic. *Memoranda Societatis pro Fauna et Flora Fennica*, **67**(1), 15–20.
- Haider, G. 1964. Zur Kenntnis von Schwermetallvergiftungen bei Fischen. *Zeitschrift für angewandte Zoologie*, **51**, 347–368.
- Haider, G. 1977. Neue Befunde über die Änderung des Blutbildes beim Fisch nach der Einwirkung verschiedener Schwermetalle. *Fisch und Umwelt*, **4**, 25–36.
- Haider, G. 1980. Der Fisch als Indikator für die Schwermetallbelastung. *Fisch und Umwelt*, **8**, 105–121.
- HELCOM. 1997. Airborne pollution load to the Baltic Sea 1991–1995. *Baltic Sea Environment Proceedings*, 69.
- HELCOM. 2003. The Baltic marine environment 1999–2002. *Baltic Sea Environment Proceedings*, 87.
- HELCOM. 2010. Hazardous substances in the Baltic Sea. *Baltic Sea Environment Proceedings*, 120B.
- HELCOM Secretariat. 2008. Press release 3.1.2008. The Baltic Sea fish are becoming less contaminated with lead and PCB's.
- Hofer, R. and Lackner, R. 1995. *Fischtoxikologie – Theorie und Praxis*. Gustav Fischer, Stuttgart.
- [IPCS] International Programme on Chemical Safety. 1989. *Lead – Environmental Aspects*. Environmental Health Criteria, 85. WHO (World Health Organization), Geneva.
- Jankovski, H., Simm, M., and Roots, O. 1996. Harmful substances in the ecosystem of the Gulf of Finland. *EMI (Estonian Marine Institute) Report Series*, 4.
- Jezierska, B. and Witeska, M. 2001. *Metal Toxicity to Fish*. University of Podlasie, Monografie 42. Wydawnictwo Akademii Podlaskiej, Siedlce.
- Jorhem, L. and Sundström, B. 1993. Levels of lead, cadmium, zinc, copper, nickel, chromium, manganese and cobalt in foods on the Swedish market, 1983–1990. *Journal of Food Composition and Analysis*, **6**, 223–241.
- Karppanen, E. and Stabel-Taucher, R. 1976. Suomen vesistöjen kalojen ja kaupan olevien kalasäilykkeiden metallipitoisuuksia [Metal concentrations in fish from Finnish waters and tinned goods]. *Suomen Eläinlääkärilehti-Finsk Veterinärtidskrift*, **82**(6), 277–284 (in Finnish).
- Lehtonen, H. 1973. Kalojen jäämäainepitoisuuksia Helsingin merialueella ja muualla Itämeressä [Harmful substances in fish from Helsinki sea-area and other parts of the Baltic Sea]. *Ympäristö ja Terveys-lehti*, **4**(9–10), 847–851 (in Finnish).
- Leivuori, M. 2007. Heavy metals in the Baltic herring. *Meri*, **59**, 87–91.
- Leivuori, M., Kestutis, J., Seisuma, Z., Kulikova, I., Petersell, V., Larsen, B., et al. 2000. Distribution of heavy metals in sediments of the Gulf of Riga, Baltic Sea. *Boreal Environment Research*, **5**(2), 165–185.
- Lind, Y., Bignert, A., and Odsjö, T. 2006. Decreasing lead levels in Swedish biota revealed by 36 years (1969–2004) of environmental monitoring. *Journal of Environmental Monitoring*, **8**, 824–834.
- Martin, M. H. and Coughtrey, P. J. 1982. *Biological Monitoring of Heavy Metal Pollution*. Applied Science Publishers, London, New York.
- Miettinen, V. and Verta, M. 1978. On the heavy metals and chlorinated hydrocarbons in the Gulf of Bothnia in Finland. *Finnish Marine Research*, **244**, 219–226.
- Müller, A. 1999. Distribution of heavy metals in recent sediments in the Archipelago Sea of southwestern Finland. *Boreal Environment Research*, **4**(4), 319–330.
- [NRCC] National Research Council Canada. 1979. *Effects of Lead in the Environment – 1978*. Publication 16736, Ottawa.
- Nuutamo, M., Varo, P., Saari, E., and Koivistoinen, P. 1980. Mineral element composition of Finnish foods – fish and fish products. *Acta Agriculturae Scandinavica*, Supplement 22, 77–87.
- Nyberg, E., Larsen, M. M., Bignert, A., Boalt, E., and Danielson, S. 2013. Metals (lead, cadmium and mercury). HELCOM Core Indicator Report. Online (accessed 09.01.2014).

- Pain, D. J. 1996. Lead in waterfowl. In *Environmental Contaminants in Wildlife* (Beyer, W. N., Heinz, G. H., and Redmon-Norwood, A. W., eds), pp. 251–264. Lewis Publishers, Boca Raton, Florida.
- Pempkowiak, J. and Szefer, P. 1992. Origin, sources and concentrations of selected heavy metals in southern Baltic biota. *Bulletin of the Sea Fisheries Institute*, **125**(1), 29–32.
- Perttilä, M., Tervo, V., and Parmanne, R. 1982. Heavy metals in Baltic herring and cod. *Marine Pollution Bulletin*, **13**(11), 391–393.
- Pokorska, K., Protasowicki, M., Bernat, K., and Kucharczyk, M. 2012. Content of metals in flounder, *Platichthys flesus* L., and Baltic herring, *Clupea harengus membras* L., from the southern Baltic Sea. *Archives of Polish Fisheries*, **20**, 51–53.
- Polak-Juszczak, L. 1996. The correlation between the concentration of heavy metals in the muscle tissue of fish and their habitat. *Biuletyn Morskiego Instytutu Rybackiego*, **137**(1), 35–39.
- Polak-Juszczak, L. 2000. Levels and trends of changes in heavy metal concentrations in Baltic fish, 1991 to 1997. *Bulletin of the Sea Fisheries Institute*, **149**(1), 27–33.
- Polak-Juszczak, L. and Domagała, M. 1994. Levels of heavy metals in Baltic fish in 1991–1993. *Biuletyn Morskiego Instytutu Rybackiego*, **133**(3), 27–33.
- Protasowicki, M. 1989. Bioaccumulation and distribution of heavy metals in fish organs. In *Proceedings 21st EMBS* (European Marine Biology Symposium), pp. 609–614.
- Quevauviller, P., Imbert, J. L., Wagstaffe, P. J., Kramer, G. N., and Griepnik, B. 1993. Reference materials, ESC-EEC-EAEC. Report EUR 14557 EN. Commission of the European Communities BCR Information, Brussels.
- Reichenbach-Klinke, H.-H. 1980. *Krankheiten und Schädigungen der Fische*. Gustav Fischer Verlag, Stuttgart.
- Roots, O. and Simm, M. 2002. Heavy metals in the Baltic fish and molluscs. In *7th Regional Meeting of the Central and Eastern European Section – Trends and Advances in Environmental Chemistry and Ecotoxicology (SECOTOX 2002) October 14–16, Book of Abstracts*, pp. 58–60. Brno, Czech Republic.
- Rühling, Å., Brumelis, G., Goltsova, N., Kvietskus, K., Kubin, E., Liiv, S., et al. 1992. Atmospheric heavy metal deposition in northern Europe 1990. *Nord*, **1992**(12), 1–41.
- Schmidt, J. 1917. *Zoarces viviparus* L. og dens locale racer [*Zoarces viviparus* L. and its local races]. *Meddelelser fra Carlsberg Laboratoriet*, **13**(3), 271–386 (in Danish).
- Szefer, P. 2002. *Metals, Metalloids and Radionuclides in the Baltic Sea Ecosystem*. Trace Metals in the Environment 5. Elsevier, Amsterdam.
- Tahvonen, R. and Kumpulainen, J. 1996. Contents of lead and cadmium in selected fish species consumed in Finland in 1993–1994. *Food Additives and Contaminants*, **13**(6), 647–654.
- Tervo, V., Erkomaa, K., Sandler, H., Miettinen, V., Parmanne, R., and Aro, E. 1980. Contents of metals and hydrocarbons in fish and benthic invertebrates in the Gulf of Bothnia and in the Gulf of Finland. *Aqua Fennica*, **10**, 42–57.
- Urtans, E. 1990. Kharakteristika pitaniya koryushki (*Osmerus eperlanus* L.) i bel'dyugi (*Zoarces viviparus* L.) v Rizhskom zalive [Characteristics of food of smelt, *Osmerus eperlanus* L., and eelpout (*Zoarces viviparus* L.) in the Gulf of Riga]. *Fischerei-Forschung*, **28**(2), 34–38 (in Russian).
- Vogt, G. and Kittelberger, F. 1976. Schwermetallanreicherung in Fischen des Rheines. *Fisch und Umwelt*, **2**, 9–12.
- Voigt, H.-R. 1999. Concentrations of heavy metals in fish from coastal waters around the Baltic Sea. *ICES Journal of Marine Science*, Supplement 56, 140–141.
- Voigt, H.-R. 2000a. Water quality and fish in two freshwater reservoirs (Gennarby and Sysilax) on the SW coast of Finland. *Acta Universitatis Carolinae Environmentalica*, **14**, 31–59.
- Voigt, H.-R. 2000b. Heavy metal and organochlorine levels in coastal fishes from the Väike Väin strait, western Estonia, in high summers of 1993–94. *Proceedings of the Estonian Academy of Sciences, Biology, Ecology*, **49**, 335–343.
- Voigt, H.-R. 2003. Schwermetallkonzentrationen (Hg, Fe, Mn, Zn, Cd, Pb und Ni) in Flundern (*Platichthys flesus* L.) aus der Kieler Förde. *Umweltwissenschaften und Schadstoff-Forschung*, **15**(4), 234–239.

- Voigt, H.-R. 2004. Concentrations of mercury (Hg) and cadmium (Cd), and the condition of some coastal Baltic fishes. *Environmentalica Fennica*, **21**, 1–21.
- Voigt, H.-R. 2007. Concentrations of heavy metals in sediments and benthos of Tvärminne Storfjärd, Finnish western Gulf of Finland (Baltic Sea). *Memoranda Societatis pro Fauna et Flora Fennica*, **83**(1), 9–16.
- Voigt, H.-R. 2008a. Concentrations of nickel (Ni) in some coastal Baltic fishes. *Memoranda Societatis pro Fauna et Flora Fennica*, **84**(2), 41–48.
- Voigt, H.-R. 2008b. Kaloissamme todettuja lyijypitoisuuksia [Concentrations of lead in our fishes]. *Ympäristö ja Terveys-lehti*, **39**(7–8), 104–106 (in Finnish).
- Voigt, H.-R. 2013. Concentrations of lead (Pb) in some coastal Baltic fishes. In *Gulf of Finland Trilateral Cooperation Forum, Abstract October 16th Tallinn, Estonia*, [http://www.gof2014.fi/wp-content/uploads/2013/11/24-Concentrations-of-Pb\\_HRVoigt.pdf](http://www.gof2014.fi/wp-content/uploads/2013/11/24-Concentrations-of-Pb_HRVoigt.pdf)
- Voipio, A., Erkomaa, K., Karppanen, E., Mäkinen, I., and Tervo, V. 1977. Eräiden raskaiden metallien ja orgaanoklooriyhdisteiden pitoisuudet Itämeren kaloissa ja pohjaeläimissä [Concentrations of some heavy-metals and chlor-organic compounds in Baltic fish and benthic animals]. *Ympäristö ja Terveys-lehti*, **8**(2), 127–143 (in Finnish).
- von Westernhagen, H. and Bignert, A. 1996. Schadstoffe in Fischen. In *Warnsignale aus der Ostsee* (Lozán, J. L., Lampe, R., Matthäus, W., Rachor, E., Rumohr, H., and von Westernhagen, H., eds), pp. 212–217. Parey Buchverlag, Berlin.