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Concentrations of mercury (Hg) and cadmium (Cd), and the condition of some coastal Baltic fishes

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Academic Dissertation in Environmental Science

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Cover pictures: Anonymous drawings of smelt, flounder and eelpout

To the memory of my son Franz-Philip Rudolf Voigt (1976 – 1995)

**"Was is das nit gift ist?
Alle ding sind gift und nichts ohn gift –
Allein dosis macht das ein ding kein gift ist"** *

*
Paracelsus´ (Philippus Theophrastus Bombastus von Hohenheim) definition of toxic/harmful substances: "What is not poisonous? All matter is toxic and (there is) no matter without poison – the dose only makes the matter harmless" 1564. *Drey Bűcher.* Arnold Byrckmann Nachfolger, Coelln.

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Growing up as child in the SW Finnish archipelago by foster-parents, an elderly fisher-couple, origin from the ceded Porkala peninsula, at the southern coast of Finland, I obey them, the late Sigurd Wilhelm Ollonquist, and his wife, the late Fanny Irene, all my knowledge in practical coastal fishing, including reasonable care for the nature, and natural resources, in that particular coastal biotope of the Baltic Sea.
Later on as a student at the university, and especially at my work, as editor and producer of the Scientific Programmes at the Finnish Broadcasting Co Ltd, and, as teacher, in fish biology, limnology, and environmental protection at the State Fishery School, and at the university, I also had several opportunities, and even duties, to visit other universities, research centres, and stations, fishing harbours, and fishing grounds all over the Baltic Sea, leading to a better understanding of e.g. the environmental problems in this sensible semi-closed brackish-water basin of estuary character.
Professor Pekka Nuorteva, when still active, and his immediate successor, acting professor Dr Sc. Martin Lodenius, the supervisor of this study, and finally professor Pekka Kauppi, professor in environmental science (Department of Limnology and Environmental Protection, since 1.1.2004 Department of Biological and Environmental Sciences), they all three, accepted and remarkably supported my intensions for a thesis on harmful substances, e.g. heavy metals, as mercury and cadmium, and their possible effects upon some coastal fishes from various parts of the Baltic Sea. For this I am deeply indebted.

Furthermore I like to express my gratitude to all my friends and colleagues at the Department of Limnology and Environment Protection, for providing me excellent working and laboratory facilities, besides good advice and encouraging support, especially to my supervisor Martin Lodenius, and to the chief of our laboratory, Esa Tulisalo, but also to Sirpa Braunschweiler, Pertti Eloranta, Hannu Lehtonen, Pekka Kauppi, Arun Mukherjee, Kari Nyberg, Ali Soltanpour-Gargari, Hannele Pulkkinen, Paavo Tamminen, Heikki Tervahattu, and Pertti Venäläinen.

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An expression of greatest gratitude goes to both reviewers of this thesis, professor, John P. Giesy (Institute of Environmental Toxicology, Michigan State University, U.S.A.), and professor, Karel Pivnička (Institute of Environmental Studies, Charles University, Prague, Chech Republik), for their valuable criticism and comments on the manuscript.

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Last but not least I want to express my gratitude to my beloved children; Maximilian, Erika, and Heidi, who all, in their own way, supported and assisted me during the work – but most of all, to their brother, my late son Franz-Philip, to the memory of whom, this thesis is dedicated.

Original publications

The thesis is based on the following papers, which are referred to in the text by their Roman numerals:

I. Voigt, H.-R., Wiklund, T. & Bylund, G. 1994. Mercury in flounder (*Platichthys flesus* L.) from Finnish coastal waters. *BMB (Baltic Marine Biologists) Publication* 15:117-119.*

II. Voigt, H.-R. 1999. Concentrations of heavy metals in fish from coastal waters around the Baltic Sea (Extended abstract). *ICES Journal of Marine Science* 56 Suppl.:140-141.

III. Voigt, H.-R. 2000. Heavy metal and organochlorine levels in coastal fishes from the Väike Väin Strait, western Estonia, in high summers of 1993-94. *Proceedings Estonian Acad. Sci. Biol. Ecol*. 49(4):335-343.

IV. Voigt, H.-R. 2001. High summer concentrations of mercury in big perch (*Perca fluviatilis* L.) from the Tvärminne archipelago (SW Finland), and Nåtö (Åland Islands) Baltic Sea. *Nahrung-Food* 45(2):109-113.

V. Voigt, H.-R. 2002. Concentrations of mercury (Hg), and cadmium (Cd) in the Baltic eelpout (*Zoarces viviparous* L.) from the Gulf of Riga (Latvia), and the Archipelago Sea (SW Finland), including a parasitological remark. *Proceedings Latvian Acad. Sci. B* 56(1-2):64-68.

VI. Voigt, H.-R. 2002. Kadmiumkoncentrationer i muskulatur, lever, njure, mjälte, galla och gonader hos åländsk flundra (*Platichthys flesus* L.) – Concentrations of cadmium in flounder (*Platichthys flesus* L.) in the Åland Islands, SW Finland. *Memoranda Soc. Fauna Flora Fennica* 77(1-2):5-11.

VII. 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.

VIII. Voigt, H-R. 2003. Concentrations of mercury and cadmium in some coastal fishes from the Finnish and Estonian parts of the Gulf of Finland. *Proceedings Estonian Acad. Sci. Biol. Ecol.* 52(3):305-318.

* Tom Wiklund provided the material for the study, and Göran Bylund edited the manuscript. Heinz-Rudolf Voigt made the analyses, and wrote the article.

Abstract: Of eight main selected coastal fish species; Baltic sprat, herring, smelt, eelpout, four-horn sculpin, perch, flounder and turbot, five; herring, smelt, eelpout, four-horn sculpin, flounder, seem most suitable for heavy metal monitoring in coastal waters in the Norhern Baltic Sea. In general low concentrations of Hg and Cd were measured from the edible parts (muscle tissue) of the fishes. Predatory species, as bull-rout, pike, perch, turbot, however were significantly more contaminated than non-predatory fishes. In a few cases the concentrations of Hg in muscle tissue of the investigated predatory fishes exceeded the stipulated safety levels for fish as food for human consumption. The concentrations of Hg were higher in inshore fishes compared to open-sea fishes. The concentrations of Cd were elevated in eelpouts from the Finnish Archipelago Sea, and the Gulf of Finland compared to eelpouts from the Gulf of Riga, and the Firth of Kiel. The same distinction also was valid for flounder from the waters around the Åland Islands, and the Gulf of Finland, compared to flounder from the Firth of Kiel.

Male fishes appeared to be slightly more contaminated than female fishes, for both metals.
The obtained significant correlations between condition factor (CF) and the metal concentration in different tissues and organs of eelpout and flounder require more attention in future studies.

Key words: Heavy metals (Hg, Cd), coastal fish (Baltic sprat, herring, smelt, eelpout, four-horn sculpin, perch, flounder, turbot), Baltic Sea (Archipelago Sea, Gulf of Finland, Western Estonian coast, Gulf of Riga, Firth of Kiel).

Introduction:

The multiplicity of results from heavy metal monitoring of Baltic fishes, as of mercury (Hg) and cadmium (Cd), includes, besides comparisons between more or less contaminated areas, aspects of both changes by time, and aspects of human health respectively (ICES 1974, 1979, 1980, 1984, 1988, 1989, 1991, and HELCOM 1981, 1987, 1988, 1996, 2001, 2002a, 2003).

Studies concerning the concentrations of these two harmful metals, in fish, however mainly focus on various toxicological tests, in finding out limits of tolerance of the fish to the metals in question (e.g. Carpenter 1930, Jones 1938, Hoffmann 1950, Schweiger 1957, Boëtius 1960, Lloyd 1961, Ishio 1965, Ball 1967, Matida et al. 1971, Eaton 1973, Haider 1977, Pentreath 1977, Fowler et al. 1981, Duncan and Klaverkamp 1983, Larsson et al. 1985, 1986, Abel and Papoutsoglu 1986, Hofer and Lackner 1995).

Studies regarding the contamination of the fish itself, by observing the distribution and the concentrations of the harmful metals in various tissues, and organs, are not as numerous (VI-VIII, and e.g. Julshamn and Brækkan 1976, Vogt and Kittelberger 1976, Topping and Graham 1977, Eagle 1981, Krüger 1981, Julsham et al. 1982, Salanki et al. 1982, Amiard et al. 1983, Protasowicki 1986, 1989, Zarski et al. 1997).

Studies concerning possible sub-lethal effects of Hg and Cd, and the estimation of these effects upon the fishes, are actually scarce (e.g. Sprague 1976, Engel et al. 1981, George 1982, Miklovics, et al. 1985, Atchison et al. 1987, Langston 1990, Baldwin et al 2003).

Thus the aim of this study is to:
1. survey and, to compare the concentrations of Hg and Cd, in edible parts (e.g. dorsal muscle tissue) of some selected common coastal Baltic fish species,
2. describe differences of the concentrations of Hg and Cd, between different tissues (muscle), and organs (liver, gonads),
3. investigate links between the heavy metal contamination of the fishes with the physiological condition of the fishes.

Material and methods:

The main eight abundant coastal species selected are; Baltic sprat (*Sprattus sprattus sprattus* L.), Baltic herring (*Clupea harengus membras* L.), smelt (*Osmerus eperlanus* L.), eelpout or viviparous blenny (*Zoarces* viviparus L), fourhorn-sculpin (*Myoxocephalus quadricornis* L.), perch (*Perca fluviatilis* L.), flounder (*Platichthys flesus* L.), and turbot (*Psetta maxima* L.), out of which in total 842 adult specimens have been investigated (I-VIII).

The main nine sampling stations were chosen according to possible contamination gradients, and they were located around the Åland Islands (Eckerö-Käringsund, Lemland-Nåtö), in the Archipelago Sea (Korpo-Brunskär, Peimari/Pemarfjärden), Gulf of Finland (peninsula of Hanko/Hangö-Tvärminne in Finland, Bays of Kunda, Käsmu, Muuga in Estonia), and at the Western Estonian coast (Bay of Matsalu, Väike Väin Strait), (I-VI, VIII).
The fishes were sampled in 1993-2002, by trap-nets, gill-nets, and trawl, during summer-autumn season, when the active forage may have resulted in an optimal physiological condition of the fishes (I-V, VIII). Eelpouts were additionally sampled from the Latvian part of the Gulf of Riga (Roja), and the German Firth of Kiel (Laboe, and Schwentine) V, as were also flounder, from the Firth of Kiel only (VI, VII). Fig.1.

Additional sampling of white fish (*Coregonus lavaretus* L. s.l.), pike *Esox lucius* L.), roach (*Rutilus rutilus* L.), ide (*Leuciscus idus* L.), bleak (*Alburnus alburnus* L.), bream (*Abramis brama* L.), Crusian carp (*Carassius carassius* L.), burbot (*Lota lota* L.), garfish (*Belone belone* L.), three-spined stickleback (*Gasterosteus aculeatus* L), nine-spined stickleback (*Pungitius pungitius* L.), bull-rout (*M. scorpius* L.), lumpfish (*Cyclopterus lumpus* L.), pikeperch (*Stizostedion lucioperca* L.), ruffe (*Gymnocephalus cernuus* L.), sandeel (*Ammodytes tobianus* L.), sand goby (*Pomatoschistus minutus* Pallas), mainly from Tvärminne, Matsalu, and Väike Väin, in order to increase the investigation material for verifying the results concerning the metal contamination, complete the list over examined species (III, VIII, Voigt 1994a, b, c, 1995a, b, 1998b, 2002b, c, d, 2003a, b, d).

Furthermore in total 340 specimen of adult smelt have been sampled from Tvärminne since 1968 (20 specimen pro study period) exclusively for monitoring of Hg concentrations in muscle tissue (HgM), VIII, Voigt 1995a, 1998a, b, c, 2002b, c, d, 2003a.

The methods of preparation of the material for analyses have been presented previously (V-VIII, Voigt 1994a, 2000b). All fishes were measured (total length, cm; TL), and weighed (total weight, g; TW) prior to the investigations and metal analyses.

The condition factor (CF), describing the physiological (health) condition of the fish (Suworow 1959, Baur and Rapp 1988), has been calculated according to the formula: $CF = 100 \times TW / (TL)^3$ (Rounsfell and Everhart, 1953, Busacker et al., 1990).

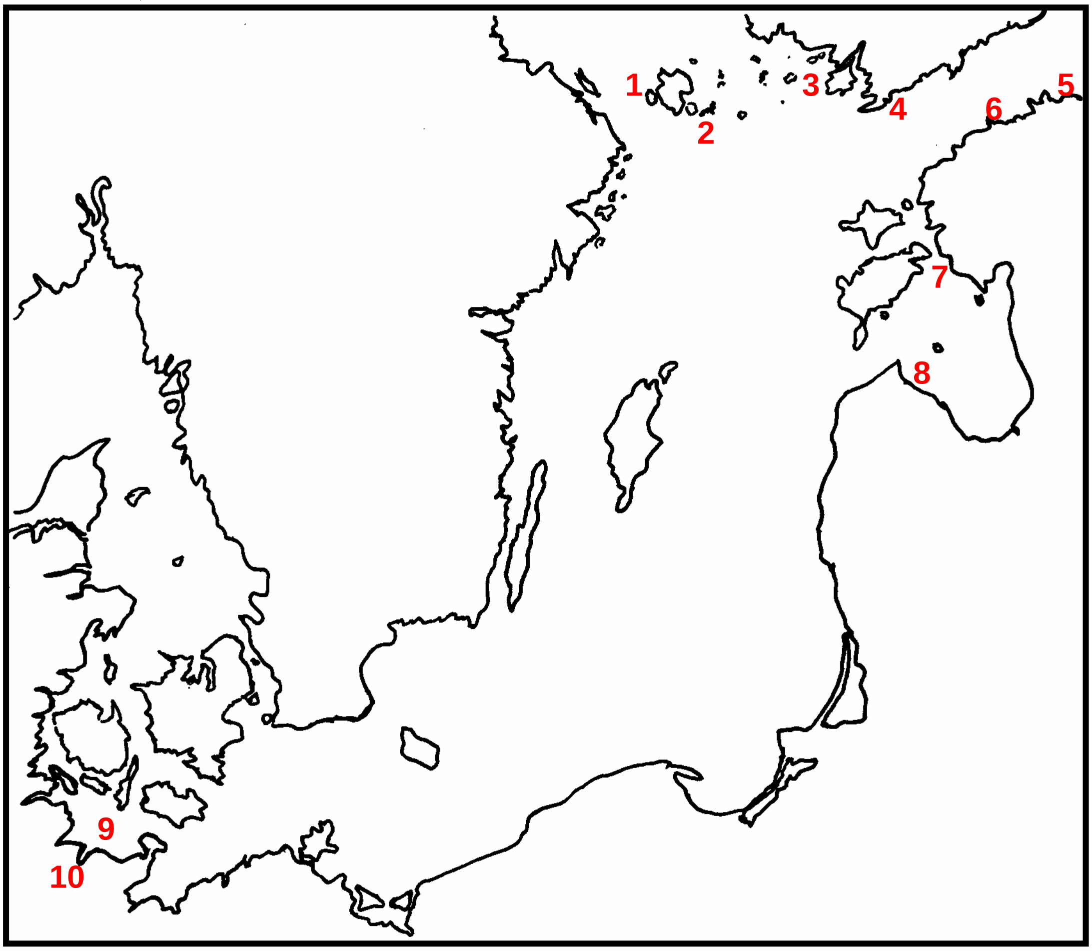

Fig. 1. Sampling stations in the Baltic Sea ; 1 = Käringsund, 2 = Nåtö, 3 = Brunskär and Peimari (Archipelago Sea), 4 = Tvärminne, 5 = Kunda, 6 = Muuga , 7 = Matsalu and Väike Väin, 8 = Roja (Gulf of Riga), 9 = Laboe and 10 = Schwentine (Firth of Kiel).

All fishes in the present investigation were analysed individually (I-VIII), in order to enable distinctions between the tissues and organs, in opposite to the procedure, used for heavy metal monitoring of fishes, as Baltic herring in the Baltic Sea, by e.g. the Finnish Institute of Marine Research, for the Baltic Marine Environment Protection Commission (HELCOM), where fishes are pooled prior to analyses (HELCOM,1984, 2002a, b)

The analytical methods used both for Hg (CVAAS by use of Coleman MAS-50B), and Cd (ETAAS by use of Varian SpectrAA-40 equipped with GTA-96), also have been presented previously (III-VIII). The determination limits for the methods used are 0.01 $mg.kg^{-1}$, fresh weight (f.wt) for Hg and 0.01 $mg.kg^{-1}$, dry weight (d.wt) for Cd, respectively.

The results of the analyses for Hg, are all expressed in $mg.kg^{-1}$ f.wt, and the results of the method used for analyses of Cd are primary given in $mg.kg^{-1}$, d.wt. For comparisons the means of Cd have been transformed into $mg.kg^{-1}$ f.wt, by using separately for each organ and tissue calculated conversion factors (V-VIII, Voigt 1998b, 2000b, 2003b).

All samples were analysed in duplicate, and the accuracy was assessed using blanks (5 per each sequence of 40 samples), and reference materials – CRM-422 cod muscle (Quevauviller et al., 1993), for which the obtained certificate values have been presented previously (IV-VIII).

The analytical data from each locality has been separately tested for homogeneity by the Shapiro-Wilk (W)/rankit plot test for normality, for correlations by the Pearson (P-test) or Spearman rank (Sr-test) correlation, and for differences between the sexes (♂♂ ♀♀), sampling seasons, and size groups by the two-sample (ts-test) t-test or the Wilcoxon rank (Wrs-test) sum test. Afterwards the significance of the obtained differences between the sampling stations, have been confirmed by separate AOV – tests (V-VIII).

Results and discussion:

The calculated mean length (TL), the condition factor (CF), and the number of the investigated fishes (N) from the various sampling stations, have been presented previously (IV-VIII, Voigt 1994a, b, c, 1997a, 1998b, 2002a, b, c, 2003a, b). Female fishes were bigger than male fishes at all main sampling areas (Åland, Archipelago Sea, Gulf of Finland, Western Estonian coast, Gulf of Riga, Firth of Kiel), although with a statistical significance, for eelpout (Tvärminne, Schwentine), ruff (Tvärminne), and flounder (Nåtö, Tvärminne) only (VI-VIII).

The numerical data of the calculated means of the various metal analyses, have been published fragmentally in a number on several occasions (I-VIII, Voigt 1994a, b, c, e, 1995a, b, 1997a, b, 1998a, b, c, 2001, 2002a, b, c, d, 2003a, b, d). They all are checked, in a few cases corrected, reproduced and presented separately in the appendix (Appendix), and thus treated mainly as figures in the present text (Figs. 2. – 5.).

The calculated conversion factors for converting d.wt into f.wt are, depending on species, for muscle tissue ca 0.20 (0.17- 0.22), liver ca 0.35 (0.25 – 0.45), and gonads ca 0.20 (ovaries 0.20 – 0.22, testes 0.18 – 0.20), III, V-VIII, Voigt 1998a, b, c, 2000b, 2003b.

As the analyses of the fishes were made individually, comparisons with e.g. the corresponding monitoring results of HELCOM are problematic, the fishes, of the Baltic monitoring programme COMBINE (HELCOM 2002b), e.g. Baltic herring, being analysed as homogenates of twenty (female) individuals (ICES 1977, 1979, HELCOM 1984, 2002a).

1. Concentrations of Hg and Cd in dorsal musculature (HgM, CdM) of some coastal fishes from various parts of the Baltic Sea.

Hg, Comparisons between sampling stations: The highest mean concentrations of Hg in dorsal musculature (HgM) of the main investigated species were calculated for perch, four-horn sculpin, and turbot. For all three species these values are increasing by the order Nåtö-Brunskär-Peimari, Tvärminne, Kunda-Käsmu (III-IV, VIII). For all the other species studied such a pattern was not detected (Fig. 2.).

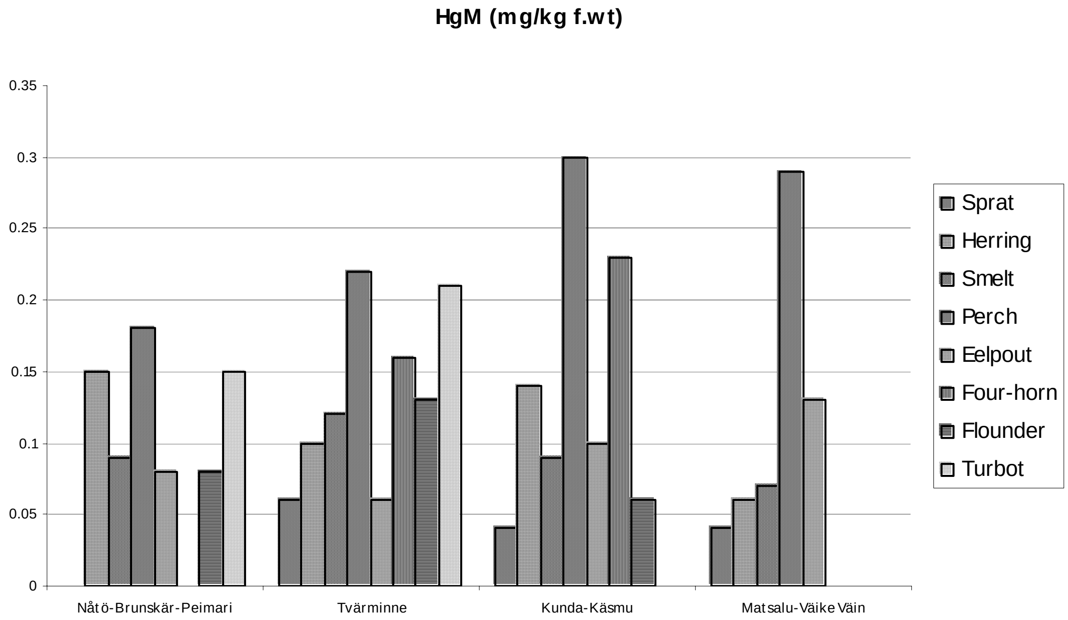

Fig. 2. Mean concentrations of mercury (mg.kg^{-1} f.wt), in muscle tissue (HgM) of sprat, herring, smelt, eelpout, perch, four-horn sculpin, flounder, and turbot from Nåtö-Brunskär-Peimari, Tvärminne, Kunda-Käsmu, Matsalu-Väike Väin.

Elevated means for HgM (0.22 mg.kg^{-1} f.wt) were additionally calculated for flounder from Käringsund (VI, Voigt 2001).

Hg, Contamination according to ecological traits: Altogether with the results for the less intensively studied species as, white-fish, pike, roach, ide, bream, Crusian carp, burbot, garfish, three-spined stickleback, nine-spined stickleback, bull-rout, lumpfish, pike-perch, ruffe, sandeel, and sand goby from the Archipelago Sea, Tvärminne, Kunda, Väike Väin, and Matsalu, the concentrations of Hg were higher in pure predatory fishes (bull-rout, pike, perch, turbot) in opposite to pure non-predatory fish (sprat, white-fish, lumpfish) III, VIII, Voigt 1998a, b, 2000a, 2002a, b, c, d, 2003a), and as previously stated for Baltic fishes (HELCOM 1987).

The concentrations of HgM also were higher in long-living fishes (roach, ide, bream, four-horn sculpin) compared to short-living fishes (bleak, sticklebacks and gobies) III, VIII, Voigt 2002f, g, 2003a. For most species the concentrations of HgM were increasing by age, as stated already in the early 1970:s (Bache et al. 1971, Westöö 1973). For adult big (female) "sea" perch (from outer coastal waters in SW Finland) this relation however, was not as obvious (IV).

Open-sea species (garfish, sprat, lumpfish) were less contaminated than inshore species (Crusian carp, pike-perch) III-IV, VIII, Voigt 1995b, 2000a, 2002b).
This later situation also is true for open-"sea" perch compared to inshore perch (IV), as for open-sea herring compared to inshore herring as well, herring from inshore

waters (Peimari, Tvärminne) having higher concentrations of Hg than herring from open-sea waters off the coast (Väike Väin) (III, VIII). A comparison of the primary monitoring results for open sea-herring from the Gulf of Finland, prepared for HELCOM by the Finnish Institute of Marine Research (Haahti, and Leivuori, respectively, personal communications), and the results of the coastal herring of this study, support the assumption. Comparisons of this kind, however are problematic, owing to the differences in the analytical procedure ("Material and methods"), and by evident local differences within the species, as previously shown for Finnish open sea-herring by Haahti (1991).

Hg, Temporal patterns: The data regarding the mean concentrations of HgM by time of smelt from Tvärminne during the period 1968 – 2002 are presented in Fig. 3.

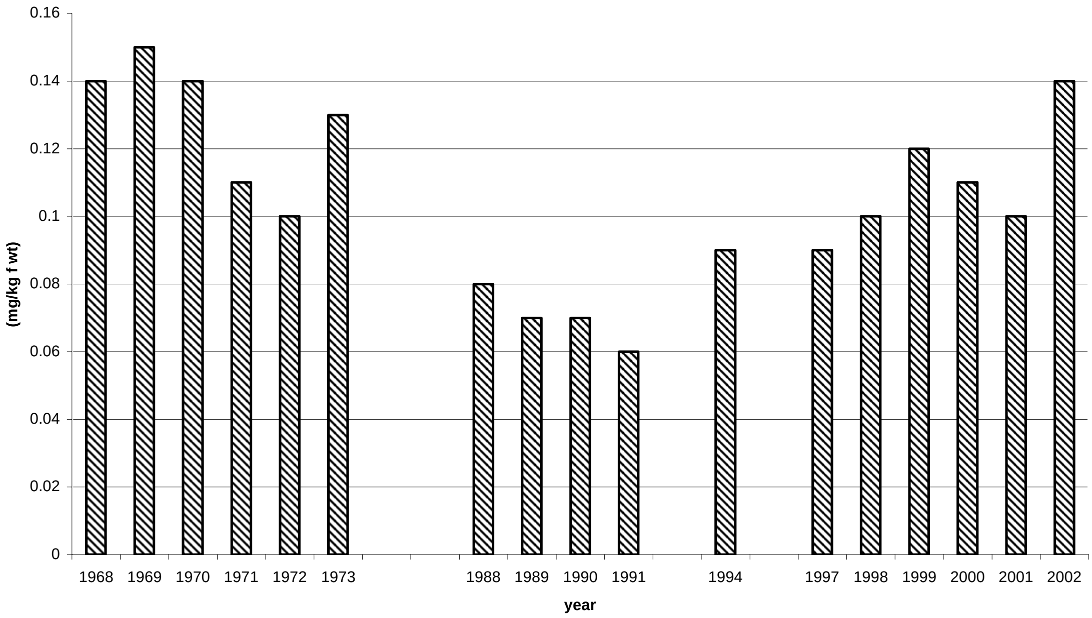

Fig. 3. Mean concentrations of mercury (Hg) in muscle tissue (mg.kg^{-1} f.wt) of smelt (*Osmerus eperlanus* L.) from Tvärminne during the period 1968 – 2002.

Jointly with the corresponding data for four-horn sculpin, and flounder in the 1970:s, and present, the obtained results show no significant increase or decrease of Hg by time for all three, more or less, non-migratory species (VIII, Voigt 2001, 2002c, d, 2003a). Additionally, however, the data for smelt show a period of decrease during the late 1980:s, and early 1990:s, compared to the situation in the late 1960:s, and early 1970:s, on one hand and to the situation in the late 1990:s, and early 2000:s, on the other (VIII, Voigt 1998b, 2002b, c, d). This latter insignificant, but still weak, increasing trend of Hg has also been recognized from the monitoring results of Hg in muscle tissue of Baltic herring in the Gulf of Finland (Perttilä et al. 2003). This trend is even significant for herring from the Baltic proper (HELCOM 2002a).

As a whole, the concentrations of Hg in muscle tissue, e.g. in the edible parts, of the investigated coastal fishes, were considerably low, thus not constituting any risk of harm by consumption or feeding. Only in a few cases for pike, bull-rout, and perch

(Tvärminne), the stipulated safety levels for mercury in fish, for human consumption (EUROPEAN COMMISSION 1993), were exceeded (IV, VIII, Voigt 1998b, c, 2002b, c, d, 2003a).

Cd, Comparisons between sampling stations: According to the corresponding data of Cd in muscle tissue (CdM) of the main studied coastal species, from Nåtö-Brunskär-Peimari, Tvärminne, Kunda-Käsmu and Matsalu-Väike Väin, there are, with exception for flounder (Nåtö and Tvärminne), and for both of which elevated means were calculated, no significant differences between the sampling stations (VI-VIII, Voigt 1998a, 2000a, 2001, 2002a, b, 2003b), Fig. 4

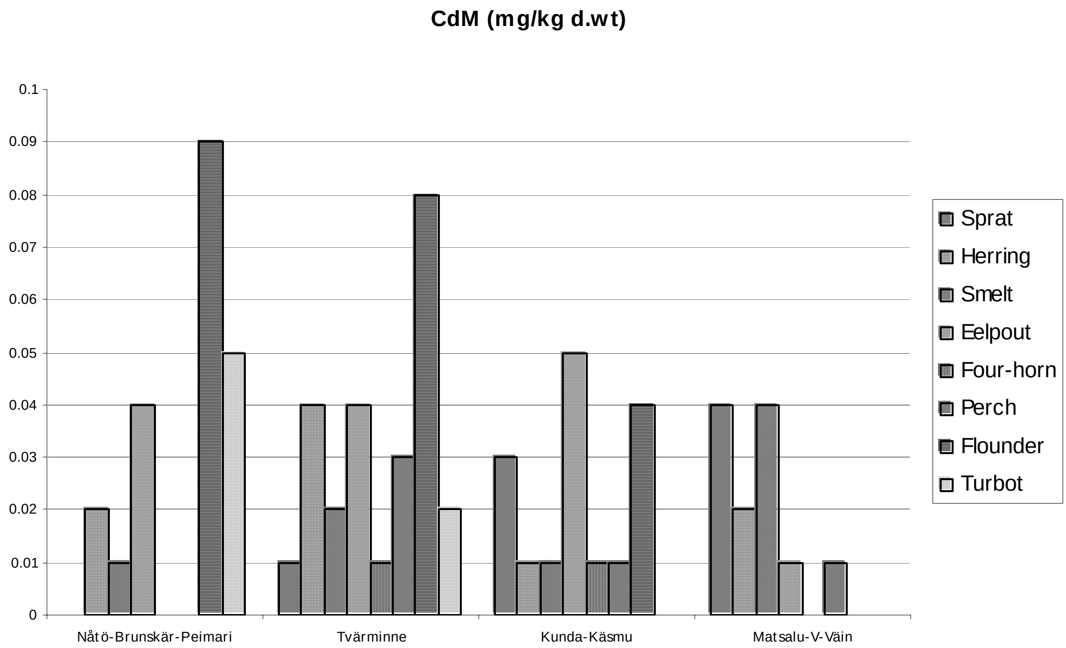

Fig. 4. Mean concentrations of Cd cadmium ($mg.kg^{-1}$ d.wt) in muscle tissue (CdM) of sprat, herring, smelt, eelpout, four-horn sculpin, and flounder from Nåtö-Brunskär-Peimari, Tvärminne, Kunda-Käsmu, Matsalu-Väike Väin (Northern Baltic Sea).

For all the other fishes investigated, there were no significant differences between the sampling stations, and the mean concentrations of CdM did not exceed 0.05 $mg.kg^{-1}$ d.wt (II-III, V-VIII, Voigt 1994a, b, c, 1995a, 1997a, b, 1998a, b, 2001, 2002a, c, 2003b, c). Equally low values (not exceeding 0.05 $mg.kg^{-1}$ d.wt) were additionally also calculated for white-fish, roach, ide, bream, Crusian carp (Tvärminne), eelpout (Roja), bull-rout (Tvärminne), lumpfish (Kunda-Käsmu), ruff (Tvärminne, Matsalu-Väike Väin), flounder (Laboe, Schwentine), and turbot (Nåtö, Tvärminne), III, V, VII-VIII, Voigt 1998a, b, 2001, 2002a, b, 2003b. As the low mean value of 0.05 $mg.kg^{-1}$ Cd d.wt., equal to ca 0.01 $mg.kg^{-1}$ Cd f.wt, constitutes only a fifth of the stipulated levels for fish as human food (0.05 $mg.kg^{-1}$ f.wt), EUROPEAN COMMISSION (2002), these fishes are, in this respect, considered completely safe.

The elevated, but still harmless, concentrations of Cd in muscle tissue of flounder from both Nåtö (0.09 $mg.kg^{-1}$ d.wt), and Tvärminne (0.08 $mg.kg^{-1}$ d.wt), require more attention in future studies, though at Tvärminne the activity of the nearby iron- and steel factory at Koverhar, as a possible source, has to be taken into consideration (Voigt 2002c, 2003b, c, d), in contrast to the situation at Nåtö, where no comparable pollution sources are known (VI-VII, Voigt 1998a, 2001, 2003b).

2. Concentrations of Hg and Cd, in dorsal musculature and liver of some selected coastal Baltic fish species.

The calculated means of the concentrations of Hg and Cd, in interior organs as liver (L), and gonads(G) of the studied fishes, are included in the appendix (Appendix).

Hg muscle-liver (HgM:HgL). For most species (e.g. smelt, pike, roach, ide, bream, Crusian carp, burbot, eelpout, four-horn sculpin, bull-rout, perch, ruff, pike-perch, flounder, turbot) the mean concentrations, of Hg in muscle tissue, exceeded the mean concentrations obtained from liver (II-III, V-VIII, Voigt 1994b, 1995a, 1997a, b, 1998a, b, c, 2001, 2002a, b). For herring (Tvärminne, Kunda-Käsmu), and lumpfish (Kunda-Käsmu) only, the mean values for HgL were higher than for HgM (VIII). Although not significant for either species, this distinction was more pronounced for herring than for lumpfish. Weather the obvious higher fat content of the liver of these two species, compared to the other studied species, has a function, as it has for cod, regarding metals other than Hg (Grimås et al. 1985), may be worth a consideration in future studies. As the metallothioneins in the liver of fishes also are known to bind Hg (besides Cd, see below), Krezoki et al. (1988), that aspect has to be considered too, when comparing fishes of different species.

Cd muscle-liver (CdM:CdL) The concentrations of Cd in the liver, by far exceeded the concentrations in the muscle tissue, for all investigated species, and at all sampling stations (Fig. 5.), and the variation of the calculated ratio CdM:CdL (0.02 – 0.05), for the investigated fishes (II, V-VIII), only underlines the fact of Cd mainly accumulating in liver, besides kidneys (Reichenbach-Klinke 1978, Hofer and Lackner 1995).

A ratio CdM:CdL, equal to, or lower than, 0.05 was calculated for herring (Kunda-Käsmu), eelpout (Kunda-Käsmu, Muuga, Roja), bull-rout (Tvärminne), perch (Tvärminne, Väike Väin), and flounder (Käringsund, Nåtö, Tvärminne), indicating considerable higher concentrations in liver compared to muscle tissue (II-III, V-VIII).

Whether the amount of fat, in the liver of these species, influences the interpretation of the analytical results (Grimås et al. 1985), has not been considered in this study. Deviating high concentrations of Cd were found, not only in muscle tissue (see above), but especially in liver, of flounder from Åland (Käringsund, Nåtö), and Tvärminne, compared to flounder from Kunda-Käsmu, Laboe, Schwentine (II, VI-VIII, Voigt 1998a, b, 2001, 2002a, b, 2003b, d). The mean concentrations of Cd in liver of flounder from these three domestic sampling stations are presented in Table 1.

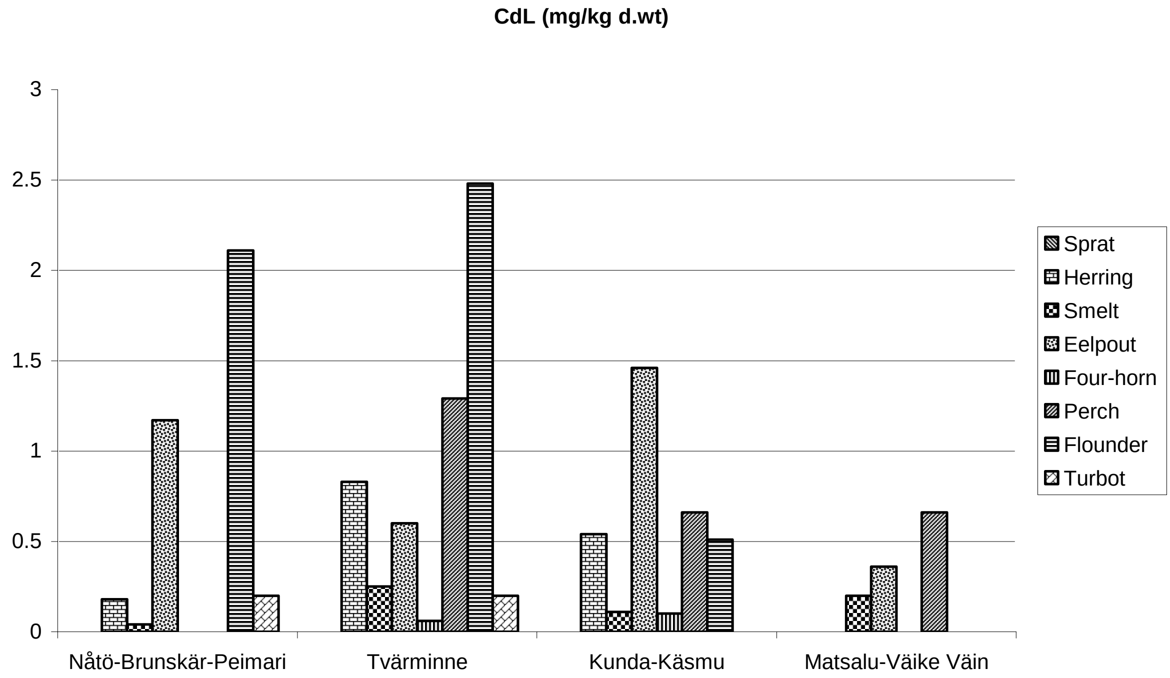

Fig. 5. Mean concentrations of Cd cadmium (mg.kg^{-1} d.wt) in liver (CdL) of herring, smelt, eelpout, four-horn sculpin, and flounder from Nåtö-Brunskär-Peimari, Tvärminne, Kunda-Käsmu, Matsalu-Väike Väin (Northern Baltic Sea).

Table 1. Mean concentrations of cadmium in muscle tissue (CdM) and liver (CdL) of flounder from Käringsund (1999), Nåtö (1997), Nåtö (1998-1999), and Tvärminne (1996-2001).

Sampling station, and time	CdM (SD)	CdL (SD)	N
Käringsund 1999	0.01 (0.01)	3.77 (3.10)	6
Nåtö 1997	0.19 (0.13)	3.12 (2.74)	21
Nåtö 1998-1999	0.01 (0.01)	2.11 (1.54)	24
Tvärminne 1996-2001	0.08 (0.22)	2.48 (2.57)	73

The contamination of the fishes by cadmium in liver, is obvious, and the above suggested explanation for the situation at Tvärminne remains (Voigt 2002a, 2003b, d), but for the findings from the waters around Åland, any assumptions cannot be made at present, as there are no known pollution sources for Cd, either at Käringsund, nor at Nåtö (VI, 2001, 2003b).

The considerably high concentrations of Cd in muscle tissue of flounder from Nåtö (1997), differ remarkably from both the findings later on (1998-1999), and from the findings at Käringsund (VI). As this concentration is even higher, than the corresponding concentration at Tvärminne, the assumption of an acute pollution situation in 1997, at Nåtö, seems even more justified than in 2002 (VI, Voigt 2003b). Equally high concentrations of Cd in flounder from other parts of the Baltic Sea are not known (VII). The corresponding concentrations of Cd in other internal organs, as kidneys, spleen, bile, and the gonads of flounder, they all contribute to the assumption (VI-VIII).

Cd in fish is absorbed both from the surrounding water by the gills, and from the food by digestion, and then transported by the blood, mainly to the liver and kidneys (Reichenbach-Klinke 1980, Halsband et al. 1984). In these both organs Cd is bound to certain proteins, metallothioneins (Eisler 1985), in flounder mainly in the liver (Kruse 1981), from which it is secreted, according to the metallothionein content of the organ in question (Hofer and Lackner 1995). Thus, as the muscle tissue of fish is not known to accumulate Cd, the concentration of Cd in muscle tissue is assumed, to reflect only the content of Cd in the transporting blood (VI).

3. Condition, in relation to the concentrations of Hg and Cd in some coastal fishes from the Baltic Sea.

Health: With the exception of liver damages in flounder (Nåtö 1997), the health condition, of the investigated fishes, appeared to be good, as no visible severe diseases, or malformations were observed (IV-VIII, Voigt 1994a, b, c, d, 1995a, 1997b, 2000a, 2001). Instead comparable gentle infestations of macroscopical intestinal helminths, as certain nematodes (smelt, eelpout), and cestods (turbot) were occasionally observed (V, Voigt 1992, 1995b 1997b).

Condition factor: Statistically significant negative correlations were calculated for; CF-HgM; male eelpout (Tvärminne), and female eelpout (Kunda-Käsmu, Schwentine), female flounder (Tvärminne, Laboe), male flounder (Laboe), CF-HgL; female eelpout (Tvärminne, Schwentine), female flounder (Tvärminne, Laboe), CF-HgG; female eelpout (Schwentine), CF-CdM; female eelpout (Brunskär, Kunda-Käsmu), CF-CdL; female flounder (Nåtö 1998-1999, Laboe), male flounder (Laboe), CF-CdG; male flounder (Schwentine), Table 2.

Whether these statistically significant negative correlations between the CF, and the both harmful metals in muscle tissue, liver and in the gonads, of the fishes, actually indicate, or reflect, a state of condition of the fishes, remains open for discussion, but contradictory to previous statements (IV-VII), these findings most certainly require more attention in future studies (VIII).

Table 2. Statistically significant negative correlations (r) between CF, and HgM, HgL, HgG (the mean concentrations of Hg in the gonads), CdM, CdL, CdG (the mean concentrations of Cd in the gonads), for both sexes separately, of eelpout and flounder from various sampling stations around the Baltic Sea.

Species	Sex	Sampling station	CF-M, L, G	r	N
Eelpout	male	Tvärminne	CF-HgM	-0.58	34
Eelpout	female	Kunda-Käsmu	CF-HgM	-0.65	10
Eelpout	female	Schwentine	CF-HgM	-0.57	15
Flounder	female	Tvärminne	CF-HgM	-0.43	55
Flounder	female	Laboe	CF-HgM	-0.72	11
Flounder	male	Laboe	CF-HgM	-0.63	12
Eelpout	female	Tvärminne	CF-HgL	-0.66	43
Eelpout	female	Schwentine	CF-HgL	-0.67	15
Flounder	female	Tvärminne	CF-HgL	-0.48	55
Flounder	female	Laboe	CF-HgL	-0.83	11
Eelpout	female	Schwentine	CF-HgG	-0.58	15
Eelpout	female	Brunskär	CF-CdM	-0.64	23
Eelpout	female	Kunda-Käsmu	CF-CdM	-0.62	10
Flounder	female	Nåtö 1998-99	CF-CdL	-0.65	20
Flounder	female	Laboe	CF-CdL	-0.91	10
Flounder	male	Laboe	CF-CdL	-0.71	12
Flounder	male	Schwentine	CF-CdG	-0.68	8

Concluding remarks:

When focusing only on changes by time, the usage of only few selected species for environmental monitoring purposes may be justified, but for a description of the environmental state in a certain area, as restricted coastal waters, several species are required.

For interpreting the environmental situation in coastal areas, in a larger scale, as for e.g. the Gulf of Finland, such data may, however be of less significance, as the fishes in the separate sub-areas mainly reflect the local situation (VIII), according to the food organisms available (Voigt 2003c, d). Thus in future studies more attention should be paid to, both consider, and to investigate the metal contamination of the food organisms of the fishes, parallel to the corresponding fish investigations (V, VI, Voigt 2003b, c, d).

According to their ecology and food habits, herring, smelt, eelpout, four-horn sculpin, and flounder seem most suitable as tools for descriptions of environmental conditions of certain sub-areas of coastal waters. Herring, which is migrating between the open sea and inshore areas (Aro 1989) may be expected, to some extent, to reflect both open sea conditions and inshore. The food habits of herring in coastal waters (Aneer 1975, Rajasilta 1997) partially remind of the habits of non-migrating smelt, as they both compete on certain macro-scopical pelagic and benthic crustaceans (Aneer 1975, Voigt 1982, 1992, Urtans 1990). The omnivorous eelpouts are truly stationary and non-migrating (Schmidt 1917), and they have a strong preference for benthic invertebrates (Aneer 1975, Urtans 1990, 1992), as has the four-horn sculpin (Westin 1970, Fonds et al. 1989). Flounder is regarded a pure consumer of bentho-fauna (Blegvad 1932, Arntz 1978), and flounder is more stationary in the coastal waters of the Northern Baltic than in the Southern Baltic Sea (Aro and Sjöblom 1983).

The concentrations of both harmful metals, Hg and Cd, were in most cases notably low, causing no risks for the potential human consumers (I-VIII). For consumers of the total fish, as predatory fishes, fish consuming birds and mammals, contaminated eelpouts and flounders, from the SW Finnish coastal waters, they may, however, constitute a potential risk.

Metal concentrations in internal organs of the fishes require more attention in future studies, as by observing the contamination of the whole fish, questions regarding e.g. detoxification, may be more understandable (Duncan and Klaverkamp 1983). For this purpose more attention should be paid to the binding of both metals by the metallothioneins in liver, as in the kidneys as well (Dallinger et al. 1997).

Whether the comparable low concentrations of Hg and Cd, in the investigated fishes, had a pronounced skewed distribution, in the sense of normality, compared to the distribution for essential metals, as e.g. for Zn (Giesy and Wiener 1977), was not object to the investigations. In the majority of the cases tested, however, the normality (Shapiro-Wilk, W) for HgM, HgL, CdL, was obvious, in opposite to the normality of the mainly, even lower corresponding, concentrations in muscle tissue (CdM), and of the gonads of both sexes (HgG, CdG).

The pronounced differences in metal-contamination of the gonads between the sexes may be linked to the weaker differences between the sexes regarding other organs and tissues of the fish, as previously suggested for herring, cod, trout, perch, bream, and roach, from the Southern Baltic (Protasowicki 1986).

The CF may be a useful tool for indicating sub-lethal effects of harmful substances, as heavy metals, but the possible influence of other metals (e.g. zinc, copper, lead), and other harmful substances (e.g. DDT:s, PCB:s, dioxins), should be considered as well, especially, as it is well known, that e.g. zinc (Zn) reacts either synergistic or antagonistic with Cd in fishes (Eisler 1993, Hofer and Lackner 1995).

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