

Heavy metals in the coastal environment around Nåtö, Lemland (Åland Islands, Baltic Sea)

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Elevated concentrations of certain heavy metals: cadmium, lead, nickel, zink, copper, manganese and iron, were recorded from surface sediments close to two fish-farms at Nåtö, as were the concentrations of cadmium in liver, kidney, spleen and bile, of lead in kidney and spleen, of nickel in kidney and spleen, of zink in liver, kidney, gonads, spleen, of copper in liver, gonads, spleen, of manganese in kidney and gonads, of iron in spleen, of flounder, from Nåtö. For turbot only the concentrations of cadmium and copper in the kidneys, and of zink in gonads were noteworthy. In the soft tissue of Baltic clay-mussel elevated concentrations of cadmium were recorded from Nåtö Biological Station, lead, zink, copper, and iron, from both the station and the mouth of the former Nåtö sewage pipeline. Elevated concentrations of cadmium were recorded from soft tissue of blue-mussel from the Föglö fairway (close to the Föglö fish-farm), and from Kobbaklintar light-house island, where also the concentrations of manganese were notable, as were lead in mussels from the Nåtö Biological Station. Also for the cockle-mussel the concentrations of cadmium were elevated at Nåtö Biological Station. All flounders, however, were safe for e.g. human consumption.

Introduction

Although the waters around the Åland Islands mainly are regarded unpolluted, and even of high quality in the Baltic Sea (Leppäkoski et al. 1986), especially concerning hazardous substances (Grimås et al. 1991, HELCOM 2010), there are, however, some indications of severe local pollution, regarding certain heavy metals, in both sediments and biota (Voigt 2002a, 2003a, 2004, 2008, 2014). This became obvious in late summer 1997, when the waters off and around the island of Nåtö (Lemland, Åland), identical to the location of the Biological Station of the Societas pro Fauna et Flora Fennica, were appointed reference area for a study of hazardous substances in flounder (*Plat-*

ichthys flesus L.) from various parts of the Baltic Sea (Voigt 2003a, 2004). Besides his own collected fish-material the author was presented "self-died" flounders, from catches in the close neighborhood, by locals from Nåtö. Self-died flounder in gill-nets are uncommon phenomena, but besides pale color of collapsed liver tissue, the flounders by superficial observation appeared undamaged (Discussion). After receiving the first results of metal-analyses of these fishes, the material (Material and methods) at first was regarded either contaminated or even otherwise unreliable. Separate control analyses and new catches in 1998–1999, however gave similar results (Voigt 2002a), as have catches afterwards. Together with corresponding analyses of bottom-sediments and

some benthic-organisms (2003–2010), the heavy metal analyses from the coastal environment off and around Nåtö, no doubt, point in the direction of a sever polluted area on Åland, thus no longer regarded "unpolluted and of high quality" (Voigt 2002a, 2003a).

Material and methods

The fish-material for the study was sampled by gill-nets for flounder, from the open waters west of the Nåtö Biological Station (NPA-NFF), where both a minor nature protection area (NPA), and a medium-size fish-farm (Nåtö Lax, NFF) for rainbow-trout (*Oncorhynchus mykiss* Walbaum) are located. Additional fish-material originates from the Nåtö-current off Rödgrund-island (NFR). Sediment cores were sampled by an Ekman-Birge box-corer from: 1) directly outside the Station (NBS = ca 5 m), 2) the mouth of an old inactive sewage pipe of the Nåtö community (1979–1991), ca 300 m towards the Nåtö current (NSP = ca 10 m), 3) the Nature protection area of Nåtö (NPA = ca 15 m), 4) Nåtö fish-farm (NFF = ca 15 m), 5) Western fairway off Nåtö (WFW = ca 20 m), 6) at Idskär-island westwards to the open se (IDW = ca 20 m), 7) Northern fairway at Rödgrund-island (NFR = ca 15 m), 8) Northern Nåtö old fish-farm (NOF = ca 10 m), 8) Bergö-Nåtö sound (BNS = ca 10 m, 9) Bergö-Nåtö fish-farm (BNF = ca 10 m), 10) Föglö fairway and fish-farm (FFF = ca 20 m), Fig.1.

Additionally some sampling for benthos took place at Kobbaklinter-lighthouse-island (Kobb), W of Nåtö.

From flounder and turbot, muscle tissue (M), liver (H), Kidney (R), spleen (L), bile (F) were removed and stored in $-20\text{ }^{\circ}\text{C}$, as were the benthic organisms (soft tissue of the Baltic clay-mussel, *Macoma balthica* L., blue mussel, *Mytilus edulis* L., cockle shell, *Cerastoderma glaucum* L.), and the sediments, until metal-analyses (of mercury by CVAAS, Coleman MAS-50B), of cadmium, lead and nickel by ETAAS by Varian-SpectrAA-equipped with GTA-96, of copper, zink, iron and manganese by Varian-SpectrAA 400 with FAAS) were performed in the laboratory of the Department of Environmental Sciences (Voigt 2000, 2003, 2004).

The treatment of the sediment samples have been described detailed previously (e.g. Voigt 2000, 2007, Lodenius et al. 2008).

The obtained results for the standard reference material (in brackets) used (NIST SRM 8704 Buffalo River sediment, NIST SRM 1573a tomato leaves, and CRM-422 cod muscle) were: obtained Cd 3.1 ± 0.28 (2.94 ± 0.29), Pb 149 ± 8.2 (150 ± 17), Ni 40 ± 1.5 (42.9 ± 3.7), Zn 390 ± 2.0 (408 ± 15), Cu 4.9 ± 0.2 (4.70 ± 0.14), Mn 220 ± 10 (246 ± 8), Fe 374 ± 8 (368 ± 7) mgkg^{-1} dry weight (dwt), and Hg 0.55 ± 0.035 (0.559 ± 0.016) mgkg^{-1} fresh weight (fwt), respectively.

The samples were analyzed in duplicate, and individually for flounder only. For turbot and the mussels they were pooled samples of 7 per sample. The accuracy of the analyzes was assessed by using blanks (5 per each sequence of 40 samples),

Results

The obtained results (mgkg^{-1} dwt), of the metal analyses of the sediments sampled in 2003–2008 from various sampling-stations in the vicinity of Nåtö, are presented in Table 1.

With the exception of cadmium (Cd) in the sediments from the mouth area of the old inactive sewage pipe of Nåtö community (NSP), the highest mean values of all metals analyzed were calculated for the sediment-samples from both fish-farms (NFF, BNF) in the study area. The mean concentrations of lead (Pb), zink (Zn), manganese (Mn) also were higher near the sewage pipe mouth (NSP) compared to the other sampling-stations (NBS, NPA, WFW, IDW, NFR, NOF, FFF), though not as pronounced as for the two fish-farms (NFF, BNF). The lowest concentrations of metals were recorded from the open sea station at Idskär (IDW), and near the large fish-farm complex at Föglö main fairway (FFF). In the sediments from the nature-protection area (NPA) the metal-concentrations were intermediate.

The mean values of metals (mgkg^{-1} for Cd, Pb, Ni, Zn, Cu, Mn, Fe, Hg) from muscle tissue (M), liver (H), and kidney (R) in flounder (PIF), and turbot (PsM), sampled from Nåtö, 1997–2008, are presented in Table 2.

The means of metal-concentrations in muscle, liver, and kidney, of flounder have been updated,

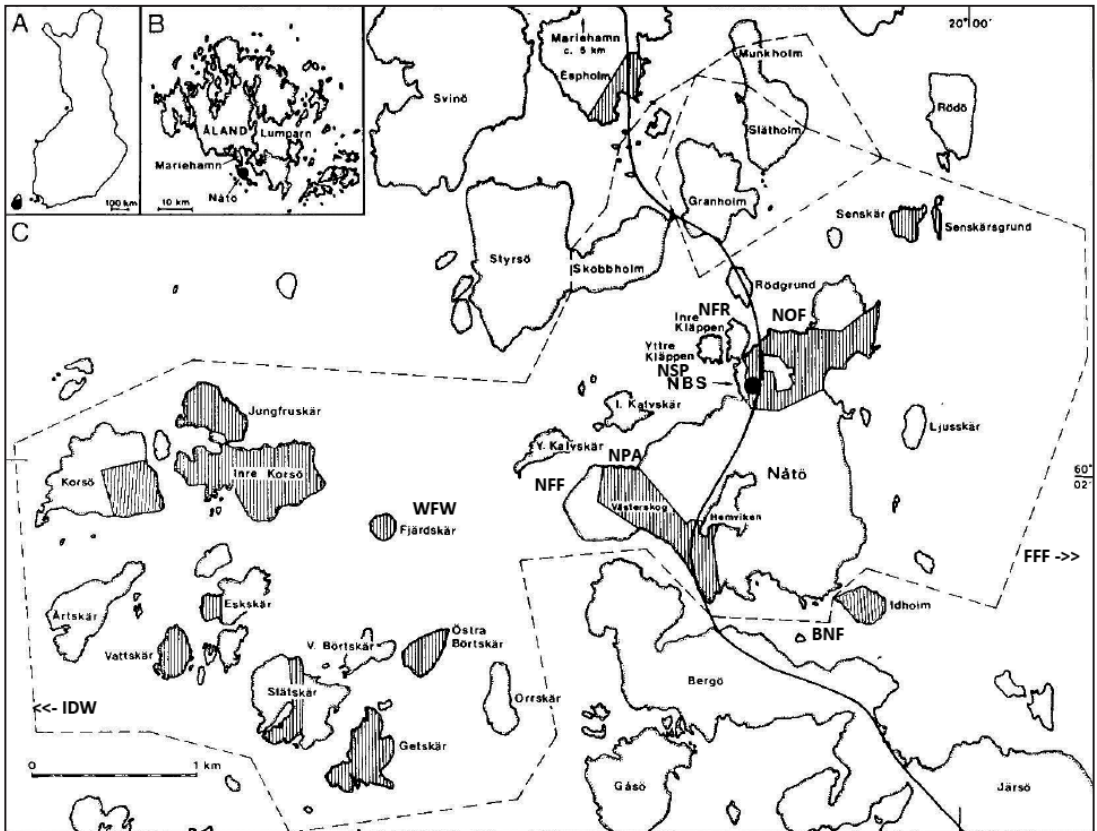


Fig.1. Nåtö island, Lemland (Åland), and sampling-stations in the vicinity: IDW (Idskär, westwards off the map), WFW (western fair-way), NFF (Nåtö fish-farm), NPA (nature protection area), NSP (sewage pipe), NBS (Nåtö Biological Station), NFR (Rödgrund island), NOF (old fish-farm), BNF (Bergö-Nåtö fish-farm), FFF (Föglö fairway and fish-farm, eastwards and off the map) – for further information read the text (Material and methods). Map, by permission modified, from C.-A. Haeggström, 1996: Nåtö Biological Station. *Memoranda Soc. Fauna Flora Fennica* 72: 227–235.

checked, and recalculated from values, some of which being partly published previously (Voigt 2002a, 2003a, 2004).

The concentrations of cadmium are notable high in liver of flounder for which also the concentrations in kidney, spleen and bile are pronounced – especially compared to the corresponding concentrations in turbot, for which only the cadmium-concentrations in kidneys are slightly pronounced (Voigt 2002b). With the exception of the notable high concentrations of lead in spleen of flounder, the concentrations of lead in both species appear modest, especially for turbot.

Nickel-concentrations in all analyzed tissues and organs of flounder are notable high – especially compared to the almost non-contaminat-

ed turbot. For zink and copper contradictory results hold good for the concentrations in kidneys of both species, though not as pronounced for gonads of turbot. In muscle tissue of flounder, however, the concentrations of copper are notable compared to turbot. For manganese the concentrations in the organs and the muscle tissue are higher in flounder than in turbot as they also are for iron though with bigger differences. Regarding mercury the concentrations are higher in the muscle tissue of predatory turbot compared to mainly mussel-feeding flounder.

The concentrations of cadmium, lead, nickel, zink, copper, manganese, iron and mercury (mg kg⁻¹), in soft tissue of the Baltic clay-mussel (*Macoma balthica* L.), sampled in 1999–2008 from

Table 1. Mean values of metals (mgkg⁻¹ dwt) in surface-sediments (5–10 cm) from various sampling-stations around Nåtö (Åland). For identifying the sampling-stations, see above (Material and methods).

Metal	NBS	NSP	NPA	NFF	WFW	IDW	NFR	NOF	BNF	FFF
Cd	0.65	1.38	0.24	1.67	0.17	0.10	0.44	0.28	0.65	0.05
Pb	19	24	–	32		3	18	15	41	4
Ni	15	21	–	56	–	–	26	–	70	–
Zn	56	100	54	356	64	17	64	62	154	26
Cu	20	43	33	132	40	4	17	16	70	5
Mn	121	174	101	253	91	66	136	135	256	94
Fe	13000	20300	7600	25000	7000	5000	13500	14500	30900	7100
Hg	0.01	0.02	0.01	0.02	0.01	–	0.02	0.02	0.03	0.02

Table 2. The mean concentration of metals (mg kg⁻¹), in muscle tissue (M), liver (H), and kidney (R), gonads (G), spleen (L), bile (F) of flounder (*Platichthys flesus* L.), individual samples, and turbot (*Psetta maxima* L.), pooled samples. NB the concentrations of all metals, are given in mgkg⁻¹dwt, except for mercury (Hg) with concentrations given in mg·kg⁻¹fwt.

Metal	Cd	Pb	Ni	Zn	Cu	Mn	Fe	Hg	N
PIF-M	0.09	0.1	0.3	35	4.2	1.5	60	0.08	43
PIF-H	2.57	0.2	0.8	126	52	2.8	850	0.05	43
PIF-R	0.91	0.5	2.9	152	7.9	3.3	1400	0.06	43
PIF-G	0.26	0.3	1	224	15.8	5.1	280	0.02	43
PIF-L	0.76	1.7	2.8	111	10.3	2.2	5300	0.05	14
PIF-F	0.61	–	–	–	–	–	–	0.04	7
PsM-M	0.05	0.1	0.05	18	0.6	1.2	4	0.15	7
PsM-H	0.10	0.1	0.05	43	4.8	1.9	100	0.04	7
PsM-R	0.14	0.1	0.1	160	58	1.8	340	0.03	7
PsM-G	0.03	0.1	0.1	310	15.5	2.8	66	0.02	7
PsM-L	0.05	0.1	0.1	96	4.4	2	590	0.05	7
PsM-F	0.04	0.1	0.1	25	2.4	0.1	16	0.02	7

Table 3. Mean concentrations of heavy metals: cadmium, lead, nickel, zink, copper, manganese, iron (mg kg⁻¹dwt), and mercury (mg kg⁻¹fwt), in pooled samples of soft tissue of the Baltic clay-mussel (Mac = *Macoma balthica* L.), blue mussel (Myt = *Mytilus edulis* L.), cockle shell (Cer = *Cerastoderma glaucum* L.) in 1999–2010. For definition of sampling-stations see Material and methods.

	Cd	Pb	Zn	Cu	Mn	Fe	Hg	Size (mm)
Mac-NBS	2.15	4.6	400	120	39	4900	0.03	10–12
Mac-NSP	0.57	4.5	580	220	47	4400	0.02	8–9
Mac-NFF	0.39	1.8	210	207	43	3400	0.04	8–11
Myt-NBS	2.42	4.3	80	13	34	1500	0.12	20–24
Myt-Kobb	4.1	3	60	10	59	1500	0.05	22–25
Myt-FFF	5.6	2.4	135	14	81	2200	0.01	20–24
Cer-NBS	1.9	2	110	26	31	2700	0.06	14–17

Nåtö, and adjacent areas, are presented in Table 3., as are the corresponding data for the blue mussel (*Mytilus edulis* L.), and the cockle shell (*Cerastoderma glaucum* L.), sampled in 2008–2010. NB the concentrations of the metals, except for mercury (Hg) are given in mgkg⁻¹dwt, but for Hg they are in mg kg⁻¹fwt.

In the soft tissue of Baltic clay-mussel the concentrations of cadmium, lead and iron were highest close to the shore at Nåtö Biological Station, followed by mouth area of the Nåtö sewage pipe-line, regarding cadmium, lead, iron, where highest concentrations of zink, copper and manganese were recorded. With the exception of mercury the concentrations of all metals analyzed were the lowest close to the Nåtö fish-farm.

In the soft tissue of blue mussel high concentrations of cadmium were recorded from all four sampling-stations: Föglö fairway, Nåtö fish-farm, Kobbaklinter and Nåtö Biological Station, and they were even higher compared to the two other mussel species, but for zink and copper the situation was the opposite. At Föglö fairway besides cadmium, also zink and manganese concentrations were the highest.

Mercury concentrations were highest in blue-mussel from the Nåtö Biological Station.

The metal-concentrations in cockle-shell from Nåtö Biological Station were more or less intermediate compared to the two other mussel-species.

Discussion

Because of the shallow and few depths, in the area around Nåtö-island, containing sediments suitable for reliable analyses of e.g. metals, the presented results are mainly guiding, though still reliable for comparisons between the various sampling-stations and other comparable data from depths in e.g. the Finnish Archipelago Sea and some depths in the Gulf of Finland.

The surface sediments at Nåtö and around Nåtö however appear contaminated by especially cadmium (NFF, NSP, NBS, BNF), at some sampling-stations also by nickel (BNF, NFF), zink (BNF, NFF), copper (BNF, NNF), manganese (BNF, NFF), and iron (BNF, NFF, NSP), when compared to the:

- 1) "back-ground data" for surface sediments of the whole Baltic Sea: Cd = 0.2, Pb = 25, Zn = 100, Cu = 20, Hg = 0.03 mg kg⁻¹dwt (Pertilä & Brüggmann 1993),
- 2) two representative (Nuorteva 1994) deep (40–47 m) sampling-stations in the Finnish Archipelago sea: Själo-Seili, Nagu-Nauvo: Cd = 0.3, Pb = 50–55, Ni = 50–55, Zn = 210–240, Cu = 55, Mn = 860–1380, Fe = 42500–54500 mg kg⁻¹dwt (Müller 1999),
- 3) representative (Nuorteva 1994) depth (60 m) of open sea Western Gulf of Finland: Cd = 0.36, Zn = 180, Cu = 33, Mn = 510, Fe = 43000, Hg = 0.06 mg kg⁻¹dwt (Leivuori 1998),
- 4) Långskär depth (40 m), open sea at Tvärminne, Western Gulf of Finland: Cd = 0.64, Pb = 15, Zn = 93, Cu = 28, Mn = 390, Fe = 24000, Hg = 0.04 mg kg⁻¹dwt (Voigt 2010),
- 5) Tvärminne Storfjärden (35 m), off Koverhar steel- and iron-factory: Cd = 0.84, Pb = 53, Ni = 35, Zn = 175, Cu = 44, Mn = 360, Fe = 40600, Hg = 0.07 mg kg⁻¹dwt (Voigt 2007),
- 6) joint mean calculated for several representative (Nuorteva 1994) depths (ca 60 m) of open sea Eastern Gulf of Finland: Cd = 1.23, Pb = 52, Zn = 209, Cu = 43, Mn = 507, Fe = 45000, Hg = 0.13 mg kg⁻¹dwt (Vallius & Leivuori 1999).

As the optimal depth for undisturbed sedimentation in the Gulf of Finland exceeds 40 m, and as the reliability of using the sediments for analyses of e.g. temporal contamination, begins at ca 60 m (Nuorteva 1994), the data from most of the quoted studies, are in this respect considered uncertain, though not as uncertain and unreliable, as the data obtained from the considerably shallower depths in the present study from Nåtö. Still the provided results are guiding and reliable in order of magnitude, and therefore useful for comparisons between the sampling-stations.

The remarkable high concentrations of all metals, except for mercury, analyzed from the surface sediments close to both fish-farms in the investigation area around Nåtö, no doubt point towards the farms as suspect-table sources for the contamination.

The strong currents around Nåtö (Voigt 1991), especially close to the Nåtö fish-farm (NFF) and the western fairway (WFF), at the open water of Idskär (IDW), and the even pronounced strong

current at Föglö (FFF), all contributed by the secondary currents from the intensive and regularly run shipping in the area, certainly disturb the sedimentation in the shallow depths by spreading the sediments over vast areas, even towards the Nåtö Biological Station. Thus some of the surprising high concentrations of the heavy metals in the area may be explained, although not revealing the source, or the sources, of the heavy metals in the environment.

Studies concerning heavy metals in sediments beneath fish-farm cages still are scarce but according to the results from a shallow (mean depth ca 5 m) inlet in the Finnish Archipelago Sea the mean concentrations of cadmium were ca 0.4, lead = ca 21, nickel = ca 25, zink = ca 180, copper = ca 43, manganese = ca 372, and iron = ca 13600 mg kg⁻¹dwt, respectively (Uotila 1987). For cadmium, lead, nickel and iron, they are of the same order of magnitude as for the concentrations of metals at Nåtö Biological Station, but far below the corresponding values from the fish-farms at Nåtö and Bergö. For zink and copper they are intermediate to the values from Nåtö Biological Station and the both fish-farms on Nåtö, and for manganese they exceed the values from Nåtö. As the inlet in question is not only shallow, but also partly separated from the sea, by thresholds, further meaningful comparisons are made difficult.

Among the first catches of flounder from Nåtö, in late summer of 1997, a few fishes with pale liver occurred, as had also the "self-died" flounders, offered for investigations to the author, by anxious neighbors (Introduction). Besides a "collapsed" liver, constituting mainly of floating tissue than of a compact organ, there were no other visible damages of the fishes, when comparing to descriptions of damaged liver of fishes (e.g. Reichenbach-Klinke 1980), wherefore further observations were neglected. As the results of analyzes of cadmium in these flounders were exceptionally high, in both liver but also in muscle tissue, repeated analyzes were performed several times, only to confirm the previous results (Voigt 2002a). Similar damages of liver, and equally high concentrations of cadmium in muscle tissue, were not observed in flounders sampled from Nåtö afterwards (Voigt 2002a, 2004).

Compared to corresponding data from flounder from Tvärminne, an area continuously load-

ed by a spectrum heavy metals, since the early 1960s, and therefore considered polluted in this respect (Voigt 2003b, 2007, 2010), the results from Nåtö differ only slightly: CdM = 0.08, CdH = 2.79, CdR = 0.69, CdG = 0.18, CdL = 0.38, CdF = 0.75, PbH = 0.20, NiH = 0.76, ZnL = 134, ZnR = 166, ZnG = 236, CuH = 70, CuR = 9, CuG = 11 mg kg⁻¹dwt (Voigt 2007), and of which, the order of magnitude is similar to the corresponding values of flounder from Nåtö, At Nåtö, however, higher concentrations of CdH and CdL than at Tvärminne were recorded – in all indicating a state of pollution in both areas (Voigt 2004).

With the exception of mercury, the concentrations of other heavy metals (cadmium, lead, zink, copper), analyzed from flounder appear elevated at Nåtö, when compared to samples from other parts of the Baltic Sea, e.g. from Estonia: Kunda, Kakumäe, Gulf of Finland, Pärnu, Gulf of Riga (Jankovski et al. 1996, Voigt 2003c), Poland: Bay of Gdańsk, Slupsk Furrow, Pomeranian Bay (Protasowicki 1986, Polak-Juszczak 2000), Germany: Bays of Wismar, Lübeck, Kiel (Senocak 1995, Voigt 2004), Denmark: Bay of Køge (Engberg 1976), especially in liver, kidney, and in some cases in the gonads also (Voigt 2002a, 2003a, 2004).

The metal concentrations in soft tissue of the mussels analyzed are either, of the same order of magnitude, or in some cases even higher at Nåtö comparing to e.g. Tvärminne, where they for the Baltic clay-mussel, regarding cadmium are 0.79, lead 3.7, zink = 799, copper = 234, manganese = 77, and iron = 2500 mg kg⁻¹dwt, respectively. For blue mussel some corresponding data are: cadmium = 1.4, zink = 180, copper = 25, manganese = 113, and iron = 1130 mg kg⁻¹dwt, respectively, and for cockle-shell: cadmium = 0.90 mg kg⁻¹dwt, respectively (Voigt 2006a,b, 2007).

At the Estonian coast of Gulf of Finland (Osmussaar Island, Bays of Tallinn, Muuga, Käsnu), with the exception of zink, the concentrations, of cadmium, lead, copper, and mercury, of the Baltic clay-mussel, at all sampling-stations, exceeded the values calculated for both Nåtö and Tvärminne (Jankovski et al. 1996, Roots & Simm 2002).

The source – or the sources – for the occurrence of these heavy metals in the environment around Nåtö still is – or are – open for discus-

sion, but no doubt the both fish-farms cannot be neglected in the search after the source(s) – especially as the analyzes of an sample of the corrosion protective paint, used for the fish-cages by the fish-farms, gave the result: copper (32 %), zink (7.7 %), iron (7.6 %), manganese (ca 0.01 %), cadmium (ca 0.0004 % = ca 3.7 mg kg⁻¹dwt), lead and nickel, in close to detectable concentrations. This analyze of the paint was performed, by the same Helsinki University Environment laboratory, where the author has analyzed all other samples in this investigation.

Conclusion

As the analytical results of both sediments and flounder, from Nåtö, all mainly point towards the fish-farms around Nåtö, as potential polluters regarding emissions of heavy metals (mainly copper, zink, cadmium), there is a need for further investigations, especially as the toxic metal cadmium may cause severe damages and harm, not only to flounder, besides other fishes in the area, but also to fish-consuming species, as birds and mammals of pray, man included. The area around Nåtö, thus is no longer considered "clean and unpolluted". As liver of flounder is not consumed by man, the flounder from Nåtö still are regarded acceptable for human consumption (EUROPEAN COMMISSION 2006).

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