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Pollutant Concentrations and Toxic Effects on the Red Alga *Ceramium tenuicorne* of Sediments from Natural Harbors and Small Boat Harbors on the West Coast of Sweden

Britta Eklund[1] · Tomas Hansson[1] · Henrik Bengtsson[2] · Ann-Kristin Eriksson Wiklund[1]

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Abstract This investigation set out to analyze the toxicity of surface sediments in a number of natural harbors and small boat harbors on the west coast of Sweden. This was done with the growth inhibition method with *Ceramium tenuicorne*. Also, concentrations of copper (Cu), lead (Pb), zinc (Zn), irgarol, organotin compounds, and polycyclic aromatic hydrocarbons (PAHs) in the sediments were analyzed. The small boat harbors were heavily polluted by Cu, Zn, butyltins, and PAHs, and to a lesser extent by Pb. The Cu, Pb, Zn, and butyltins probably originated from their past and/or present use in antifouling paints, whereas the PAHs probably had multiple sources, including boat motor exhausts. The measured toxicity of the sediment was generally related to their Cu, Zn, and butyltin content, although other toxic substances than those analyzed here probably contributed to the toxicity in some of the harbors. The natural harbor sediments contained less pollutants and were less toxic than the small boat harbor sediments. Nevertheless, our data indicate that the boating pressure today may be high enough to produce toxic effects even in natural harbors in pristine areas. The strongest relationship between toxicity and the major pollutants was obtained when the sediment toxicity was expressed as gram wet weight per liter compared with gram dry weight per liter and gram total organic carbon per liter. Hence, for pollutants that can be elutriated with natural sea water, sediment toxicity expressed as gram wet weight per liter appears preferable.

✉ Britta Eklund
britta.eklund@aces.su.se

✉ Tomas Hansson
tomas.hansson@aces.su.se

[1] Department of Environmental Science and Analytical Chemistry (ACES), Stockholm University, 10691 Stockholm, Sweden

[2] Environmental Protection Department, County Administrative Board of Västra Götaland, 40340 Göteborg, Sweden

Sweden has a particularly rich public boat life, probably because of the extensive coastal archipelagos, which stimulate the use of pleasure boats. It has been estimated that almost 2.5 million Swedes in the age interval 20–74 years, distributed over more than 1.5 million households, used pleasure boats in 2010 (Anon 2010). The total number of pleasure boats in Sweden has been estimated to approximately 0.86–1.03 millions (Anon 2010). Within the European Union (EU), a pleasure boat is every boat, propelled in any way, with a hull length of 2.5–24 m intended for sport or leisure purposes. In our investigation, a small boat is defined as this kind of pleasure boat with a hull length of less than 15 m. Among the Swedish pleasure boats with known hull length, 65 % were shorter than 5 m, 32 % were in the interval 6–10 m, and 3 % were in the interval 11–15 m (Anon 2010). Numerous public small boat harbors have been established for the specific purpose to host these small boats, i.e., these harbors are not used by larger boats or other boats than pleasure boats.

The boating described involves the release of certain environmental pollutants, which often accumulate in the sediments. Some pollutants originate from antifouling paints, which are designed to release toxic compounds in order to prevent attachment of fouling organisms to the boat hull. Other pollutants, such as polycyclic aromatic hydrocarbons (PAHs), are present i.a. in boat motor exhausts.

Formerly, lead (Pb) was used extensively as the active substance in red lead paint, which was especially effective

on wooden boats to protect them from ship worm (Lunn 1974). The use of Pb in various products started to be restricted in 1976 (Council Directive 76/769/EEC), when Pb was shown to have retarding effects on children. Today, antifouling paint containing Pb may only be used by professionals for protection of old wooden boats of high cultural value. The organotin compounds tributyltin (TBT), dibutyltin (DBT), and triphenyltin (TPT) have been used extensively as active substances in antifouling paints from around 1960 until they were forbidden in EU in 1989 for use on pleasure boats shorter than 25 m (Council Directive 89/677/EEC) because of their harmful effects on biota (Alzieu et al. 1986; Bryan et al. 1986; Fent 2006). Despite the prohibition more than 25 years ago, organotin compounds are still found in high concentrations in pleasure boat harbors (Maguire 2000; Cato et al. 2007; Eklund et al. 2008, 2010; Bengtsson and Cato 2011), and the reason has been suggested to be old paint layers containing these compounds (Eklund et al. 2008). A well-known effect of organotin compounds is imposex in molluscs, which has been observed in pleasure boat harbors along the Swedish coast (Magnusson et al. 2005), indicating that aquatic organisms are still exposed to these substances. After the prohibition of organotin compounds, copper (Cu) became the most extensively used active substance in antifouling paints (Brooks and Waldock 2009). The preventive effect of various copper compounds is enhanced by so-called booster biocides, e.g., chlorothalonil, dichlofluanid, diuron, irgarol, 2,3,3,6 tetrachloro-4-methylsulfonyl (TCMS) pyridine, 2-(thiocyanomethylthio) benzothiazole (TCMTB), zinc pyrithione, and zineb (Voulvoulis et al. 2002, Konstantinou and Albanis 2004). Zinc (Zn) also is commonly included in the paint as a binder and/or pigment (Yebra et al. 2004). Because of the widespread use of antifouling paints, elevated concentrations of both Cu and Zn have been observed worldwide in sediments, e.g., in marinas (Turner 2010), ship lanes (Strand et al. 2003), pleasure boat harbors (Eklund et al. 2010; Bengtsson and Cato 2011), and estuaries (Matthiessen et al. 1999).

Along the Swedish coast, sheltered locations in the archipelago often are used as natural harbors for pleasure boats staying one or a few nights, especially during the summer months. These natural harbors are usually situated in remote areas only reachable by boat, and they contain no man-made constructions. To the best of our knowledge, environmental pollutants and their toxic effects have not been analyzed in such natural harbors previously. We investigated the potential sediment toxicity in four natural harbors and eleven traditional small boat harbors along the Swedish west coast. The manifestation of toxic effects in the field depends not only on the pollutant concentrations, but also on their physicochemical properties and bioavailability to living organisms. Hence, we performed both chemical analysis and eco-toxicological testing of surface sediments sampled in the investigated natural harbors and small boat harbors. For the eco-toxicological testing we used the growth inhibition method with the red alga *Ceramium tenuicorne* (ISO 10710:2010), which was exposed at the laboratory to surface sediment elutriates in different concentrations. The utility of expressing sediment toxicity as gram wet weight (ww) per liter, gram dry weight (dw) per liter, or gram total organic carbon (TOC) per liter also was evaluated. The analysis of environmental pollutants in the sediments was part of a larger investigation of natural harbors and small boat harbors along the Swedish west coast (Bengtsson and Cato 2011).

Materials and Methods

Investigated Harbors

The investigation included four natural harbors (NH) and eleven small boat harbors (SBH) in the County of Västra Götaland on the west coast of Sweden (Fig. 1; Table 1). Three of the natural harbors, Gråa holmarna, Store Bror, and Tjälleskär, were situated in the Kosterhavet Marine National Park, which may be considered as a pristine area. Even though they were situated relatively close to each other, they were separated by land in a way that they belonged to different water basins. The fourth natural harbor, Gluppö-Fläskön, was situated approximately 1 km west of the Hjärterö Archipelago Nature Reserve in a relatively remote area. Estimated maximum number of contemporaneously visiting boats was 10 for Gråa holmarna, 5 for Store Bror and Tjälleskär, respectively, and 30 for Gluppö-Fläskön. All small boat harbors, except Vallhamn, were situated in, or close to, developed areas. The number of boat places in these small boat harbors varied from 30 to 2500 (Table 1).

Sediment Sampling

One sediment core was collected from each harbor in September and October 2010. The only exception was Hinsholmskilen, where two sediment cores were collected: one from the inner part and one from the outer part of the harbor. The samples were taken with an Ekman box sampler with a sampling area of 20 × 20 cm (Ekman 1911). The top 0–2 cm of each sediment core were sliced off and kept in sealed plastic jars at 4 °C until further processing. Each sediment sample was mixed to homogeneity before it was split between chemical analysis and eco-toxicological testing.

Fig. 1 Investigated natural harbors and small boat harbors on the Swedish west coast (Color figure online)

Chemical Analysis

The chemical analysis of the surface sediments included: the three metals copper (Cu), lead (Pb), and zinc (Zn); irgarol; the six organotin compounds tributyltin (TBT), dibutyltin (DBT), monobutyltin (MBT), triphenyltin (TPT), diphenyltin (DPT), and monophenyltin (MPT); and the 16 polycyclic aromatic hydrocarbons (PAHs) naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, dibenz[*a*,*h*]anthracene, benzo[*ghi*]perylene, pyrene, and indeno[1,2,3-*cd*]pyrene. ΣBT was defined as the sum of TBT, DBT, and MBT. ΣPT was defined as the sum of TPT, DPT, and MPT. Σ6OrgT was defined as the sum of TBT, DBT, MBT, TPT, DPT, and MPT. Σ16PAH was defined as the sum of the 16 analyzed PAHs. Σ7PAHcarc was defined as the sum of the seven analyzed PAHs that are classified as probable human carcinogens by the U.S. Environmental Protection Agency (Anon 1995): benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, dibenz[*a*,*h*]anthracene, and indeno[1,2,3-*cd*]pyrene. The chemical analyses were performed by ALS Scandinavia AB (Täby, Sweden), which was accredited for quantitation of all substances except irgarol. Metals were quantitated with inductively coupled plasma sector field mass spectrometry (ICP-SFMS) according to modified EPA methods (Method 200.7; Method 200.8). Organotin compounds were extracted with a mixture of MeOH and hexane and quantitated with gas chromatography with flame photometric detector (GC-FPD). PAHs were extracted with a mixture of acetone, hexane, and cyclohexane and quantitated with gas chromatography and mass spectrometry (GC–MS). Irgarol was also quantitated with GC–MS. Sediment dry weight and TOC per g dw were determined with standardized procedures (SS 28113; DIN ISO 10694).

Eco-toxicological Testing

The toxicity of the surface sediments was tested with the growth inhibition method (ISO 10710:2010) with the red alga *C. tenuicorne* (Rueness 1978; Gabrielsen et al. 2003). The global genus *Ceramium* is present in all oceans and is particularly abundant at temperate latitudes (Lüning 1990). *C. tenuicorne* is commonly growing in both marine and brackish waters. In this investigation, we used the marine clone isolated from the Oslo fjord in 1978 (Rueness 1978). *C. tenuicorne* may be used for routine eco-toxicological testing and its performance in different light intensities, temperatures, and salinities has been investigated by Eklund (2005). The test principle is to measure growth inhibition by various treatments during a seven day exposure period (Bruno and Eklund 2003; Eklund 2005; ISO 10710:2010). An advantage of the method is that *C. tenuicorne* can be adapted to salinities between 4 and 32 ‰. In this investigation, we used specimens adapted to 20 ‰, because this was the prevailing salinity in most harbors. The method was modified for testing of sediments as described by Eklund et al. (2010), who also compared this method with exposure to whole sediment. In the latter method, weighed aliquots of the sediment were placed in small plastic containers, and the algal tips were exposed to the sediment on a net directly above the sediment surface (Eklund et al. 2010). Because the two methods gave equivalent results, we chose the less time consuming method with sediment elutriates in the present investigation.

Test elutriates from each surface sediment were prepared by the following procedure. In an E-flask, 19.2 g of wet sediment was suspended in 120 mL of natural

Table 1 Harbors, positions, and sediment properties

Harbor	Number of boat places	Harbour area (m^2)	Latitude	Longitude	Fraction dry weight (%)	Fraction TOC dw (%)
Natural harbors (NH)						
Gluppö-Fläskön	–	150,000	58°34.969′N	11°12.895′E	17.2	5.0
Gråa holmarna	–	15,000	58°51.255′N	11°02.742′E	23.8	5.4
Store Bror	–	3000	58°50.780′N	11°01.630′E	42.2	2.8
Tjälleskär	–	6500	58°51.135′N	11°02.418′E	60.2	1.0
Small boat harbors (SBH)						
Björlanda kile	2500	200,000	57°45.687′N	11°48.365′E	38.9	3.7
Eriksberg	30	50,000	57°41.999′N	11°54.938′E	37.1	3.1
Hinsholmskilen inner	1500	300,000	57°39.850′N	11°51.526′E	25.2	4.1
Hinsholmskilen outer	1500	300,000	57°39.522′N	11°51.022′E	25.8	3.6
Hjuvik	200	17,500	57°42.703′N	11°42.704′E	29.9	2.8
Hålkedalskilen	200	17,000	58°55.706′N	11°11.134′E	23.3	5.0
Kebal	350	30,000	58°56.566′N	11°09.684′E	28.8	4.3
Kungshamn	400	65,000	58°21.426′N	11°16.173′E	18.5	8.1
Marstrand	250	30,000	57°53.254′N	11°35.792′E	26.1	5.9
Nödinge	30	2000	57°53.415′N	12°02.363′E	32.3	2.7
Vallhamn	250	45,000	58°00.334′N	11°42.045′E	23.6	3.8
Önnered	300	60,000	57°38.205′N	11°52.263′E	21.0	5.8

(uncontaminated) seawater (20 ‰) from the Swedish west coast. This water was collected at the Sven Lovén Centre for Marine Sciences at Tjärnö (Strömstad, Sweden). The resulting suspension corresponded to 160 g of wet sediment/L. The E-flask was shaken by hand for 10–20 s, and half the volume was directly decanted into a second E-flask. Another 60 mL of natural seawater was added to the second E-flask. After shaking by hand for 10–20 s, half of the volume of the second E-flask was transferred to a third E-flask and mixed with another 60 mL of natural seawater. This procedure was continued until a series of six concentrations (160, 80, 40, 20, 10, and 5 g of wet sediment/L) was obtained. Also from the last E-flask, half the volume was discarded so that all E-flasks contained 60 mL of sediment suspension. As control, an E-flask containing only 60 mL of natural seawater was included. The whole dilution series was completed in ca 5 min, so there was no settling of the sediment before decantation. In this way, we produced one dilution series per sediment sample. All E-flasks were placed in darkness at room temperature during 24 h for elutriation. The rationale of this elutriation method was to mimic possible release of pollutants from sediments in the field. It should be noted that the efficiency of the elutriation was related to the degree of hydrophilicity. Metal ions are strongly hydrophilic, whereas organotin compounds are less hydrophilic, and larger PAHs (like the ones analyzed here) are strongly hydrophobic. Mean partitioning coefficients between sediment organic carbon and water (log K_{OC} values) have been estimated to ca 4 for TBT (Bangkedphol et al. 2009) and ca 6–8 for many of the PAHs analyzed here (Hawthorne et al. 2006). At the end of the elutriation, 40 mL of the superficial water from each E-flask was filtered through a 0.45-μm Whatman polyamide membrane filter (Sigma-Aldrich, Stockholm, Sweden). Nutrients were added to a concentration of 3.5 μg of nitrogen per liter, 0.8 μg of phosphorus per liter, and 0.1 μg of iron per liter. Each 40-mL elutriate was then split, in equal parts, into four Petri dishes, and two algal tips with a length of ca 1 mm were added to each Petri dish. The exposure was performed during 7 days at a temperature of 22 ± 2 °C, a photosynthetic photon flux of 70 ± 7 μmol/m^2/s (ca 3600 lux), and a light/dark regime of 14 h light and 10 h darkness. At the end of the exposure, the length of the specimens was measured and growth inhibition was determined compared with the control.

Statistics

The statistical analysis included the Pearson correlation, the exact Wilcoxon–Mann–Whitney test, and simple linear regression. A pollution index was created for the Cu, Zn, and ΣBT concentrations in the sediments from the small boat harbors. This index was based on the concentrations in the sediment in the harbor Hålkedalskilen, which had an intermediate degree of pollution among the small boat harbors. The index value was set to 1 for each of the three pollutants in Hålkedalskilen, and all other values were expressed relative to the concentrations in this harbor. The

softwares Intercooled Stata 12.1 (StataCorp LP, College Station, TX) and R 3.1.0 (The R Foundation for Statistical Computing, Vienna, Austria) were used for the analyses. The software RegTox V6.4 (Vindimian 2001) was used to calculate the sediment concentrations where 50 % growth inhibition occurred, i.e., EC50 values, with 95 % confidence intervals. The EC50 values were expressed in three ways: g ww/L, g dw/L, and g TOC/L.

Results

Sediment Properties

The dry substance fraction of the sediments varied from 17.2 to 60.2 % in the natural harbors and from 18.5 to 38.9 % in the small boat harbors (Table 1). The TOC fraction of dry substance varied from 1.0 to 5.4 % in the natural harbors and from 2.7 to 8.1 % in the small boat harbors (Table 1).

Pollutant Concentrations

The analyzed concentrations of Cu, Pb, Zn, TBT, DBT, MBT, and Σ16PAH in the sediments were classified according to the criteria for evaluation of sediment status (Anon 1999, 2011) established by the Swedish Environmental Protection Agency (SEPA). These criteria comprise five classes of deviation from preindustrial concentrations (Table 2). From here on, "elevated concentrations" means all concentrations that are higher than those of the lowest class (no or insignificant deviation). Elevated concentrations of Cu, Pb, Zn, TBT, DBT, and MBT were found in some of the natural harbors, whereas Σ16PAH was elevated in all natural harbors (Table 3). In the small boat harbors, the concentrations of Cu, Zn, TBT, DBT, MBT, and Σ16PAH were strongly elevated in most cases, whereas the Pb concentration fell within the three lower deviation classes (Table 3). Irgarol was below the detection limit (0.02 mg/kg dw) in all harbors (Table 4) and thus was not further considered. The TBT concentration ranged from below the detection limit (1.0 µg/kg dw) to 2.5 µg/kg dw in the natural harbors, whereas in half of the small boat harbors, the TBT concentration was higher than 100 µg/kg dw (Table 3). The TBT fraction of ΣBT varied from 31 to 44 % in the natural harbors and from 28 to 62 % in the small boat harbors (Table 4). In the small boat harbors, Σ6OrgT was dominated (92–97 %) by ΣBT (Table 4), whereas ΣPT constituted only a minor part (Table 4) and thus was not further considered. The Σ7PAHcarc fraction was 42–55 % in those harbors where it was analyzed (Table 4). The overall pattern was lower concentrations of Cu, Pb, Zn, ΣBT, and Σ16PAH in the natural harbors than in the small boat harbors (Table 5). In the small boat harbors, the concentrations of Cu, Pb, Zn, and ΣBT were pairwise correlated ($r > 0.7$, $p < 0.05$), but uncorrelated with the concentrations of Σ16PAH (not shown). The pollution indices for Cu, Zn, and ΣBT are shown in Fig. 2. Moreover, the respective concentrations of Cu, Zn, and ΣBT were correlated with the number of boat places in the small boat harbors ($r > 0.7$, $p < 0.05$, not shown).

Sediment Toxicity

There was no growth inhibition by the natural harbor sediments, except the sediment from Gluppö-Fläskön, which had an EC50 value of 152 g ww/L (Table 6) corresponding to 15.2 % wet sediment (Fig. 3). The absence of a toxic effect at three natural harbors proves that there was no toxicity of sediment alone (beyond anthropogenic substances). Hence, these sediments may serve as a sediment control. Among the small boat harbors, the most toxic sediment was found in Hjuvik, where only 2 % wet sediment was sufficient to produce 50 % growth inhibition (Fig. 3). Linear regression of the EC50 values on the combined pollution indices for Cu, Zn, and ΣBT showed

Table 2 Classes of deviation from preindustrial concentrations of Cu, Pb, Zn, butyltins, and Σ16PAH in Swedish sediments (Color table online)

Pollutant	Class 1 No or insignificant deviation	Class 2 Small deviation	Class 3 Clear deviation	Class 4 Large deviation	Class 5 Very large deviation
Cu (mg/kg dw)[a]	<15	15–30	30–50	50–80	>80
Pb (mg/kg dw)[a]	<25	25–40	40–65	65–110	>110
Zn (mg/kg dw)[a]	<85	85–128	128–204	204–357	>357
TBT (µg/kg dw)[b]	<1	1–6	6–24	24–100	>100
DBT (µg/kg dw)[b]	<1	1–3	3–12	12–47	>47
MBT (µg/kg dw)[b]	<1	1–4	4–10	10–25	>25
Σ16PAH (mg/kg dw)[b]	<0.001	0.001–0.2	0.2–1.1	1.1–4.8	>4.8

[a] Calculated from Tables 34 and 36 in SEPA Report no. 4914 (Anon 1999)

[b] According to Anon (2011), which is an update of Anon (1999)

Table 3 Concentrations of Cu, Pb, Zn, butyltins, and Σ16PAH in the sediments. Concentrations classified according to Table 2 (Color table online)

Harbor	Cu (mg/kg dw)	Pb (mg/kg dw)	Zn (mg/kg dw)	TBT (µg/kg dw)	DBT (µg/kg dw)	MBT (µg/kg dw)	Σ16PAH (mg/kg dw)
Natural harbors (NH)							
Gluppö-Fläskön	34.4	27.4	129	2.5	3.5	2.0	0.42
Gråa holmarna	42.1	12.3	75.8	<1.0	<1.0	<1.0	0.01
Store Bror	11.2	7.79	47.1	<1.0	<1.0	<1.0	0.22
Tjälleskär	6.68	4.09	37.4	1.2	1.5	<1.0	0.02
Small boat harbors (SBH)							
Björlanda kile	253	50.2	400	560	340	200	0.64
Eriksberg	86.3	51.6	235	160	82	16	1.22
Hinsholmskilen inner	301	47.1	299	510	400	160	–
Hinsholmskilen outer	235	50.0	415	180	180	49	0.83
Hjuvik	46.9	22.5	140	67	66	25	1.13
Hålkedalskilen	121	36.4	265	93	66	18	–
Kebal	59.7	29.7	147	10	18	7.7	7.47
Kungshamn	252	47.4	332	270	330	88	2.31
Marstrand	136	43.7	256	250	120	41	1.49
Nödinge	33.3	22.2	193	71	35	22	0.60
Vallhamn	39.7	16.1	131	8.6	15	5.1	0.29
Önnered	115	28.6	253	42	64	25	–

– Not analyzed

Table 4 Concentrations and percentages of irgarol, organotin compounds, and carcinogenic PAHs in the sediments

Harbor	Irgarol (mg/kg dw)	ΣBT (µg/kg dw)	TBT as percent of ΣBT	ΣPT (µg/kg dw)	Σ6OrgT (µg/kg dw)	ΣBT as percent of Σ6OrgT	Σ7PAHcarc (mg/kg dw)	Σ7PAHcarc as percent of Σ16PAH
Natural harbors (NH)								
Gluppö-Fläskön	<0.02	8	31	<2.0	[a]	[a]	0.23	55
Gråa holmarna	<0.02	<1.5	[a]	<1.5	[a]	[a]	<0.04	[a]
Store Bror	<0.02	<1.5	[a]	<1.5	[a]	[a]	0.11	50
Tjälleskär	<0.02	2.7	44	<1.5	[a]	[a]	0.01	50
Small boat harbors (SBH)								
Björlanda kile	<0.02	1100	51	63	1163	95	0.35	55
Eriksberg	<0.02	258	62	<4.5	[a]	[a]	0.60	49
Hinsholmskilen inner	–	1070	48	78	1148	93	–	–
Hinsholmskilen outer	<0.02	409	44	24	433	94	0.44	53
Hjuvik	<0.02	158	42	5.5	164	97	0.48	42
Hålkedalskilen	–	177	53	15	192	92	–	–
Kebal	<0.02	35.7	28	<2.0	[a]	[a]	3.37	45
Kungshamn	<0.02	688	39	43	731	94	1.14	49
Marstrand	<0.02	411	61	13	424	97	0.72	48
Nödinge	<0.02	128	55	<1.5	[a]	[a]	0.28	47
Vallhamn	<0.02	28.7	30	<1.5	[a]	[a]	0.14	48
Önnered	–	131	32	12	143	92	–	–

– Not analyzed

[a] Not possible to calculate

Table 5 Comparison of concentrations of Cu, Pb, Zn, ΣBT, and Σ16PAH in the sediments in natural harbors (NH) and small boat harbors (SBH)

Pollutant	NH median	NH range	SBH median	SBH range	NH–SBH difference[a]
Cu (mg/kg dw)	23	7–42	118	33–301	$p = 0.0077$
Pb (mg/kg dw)	10	4–27	40	16–52	$p = 0.0077$
Zn (mg/kg dw)	61	37–129	255	131–415	$p = 0.0011$
ΣBT (μg/kg dw)	2.1	<1.5–8.0	218	29–1100	$p = 0.0011$
Σ16PAH (mg/kg dw)	0.12	0.01–0.42	1.13	0.29–7.47	$p = 0.0056$

[a] Exact Wilcoxon–Mann–Whitney test

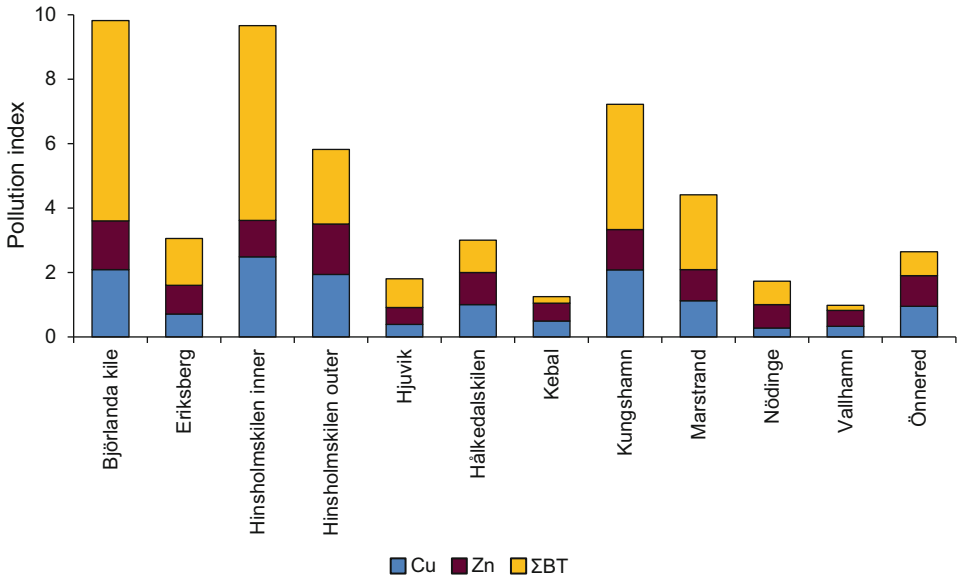

Fig. 2 Pollution indices for copper (Cu), zinc (Zn), and butyltins (ΣBT) in the small boat harbor sediments. The index value was set to 1 for each of the three pollutants in Hålkedalskilen, and all other values were expressed relative to the concentrations in this harbor (Color figure online)

that EC50 decreased with increasing pollutant concentrations in most harbors (Fig. 4). The strongest relationship was obtained when the EC50 values were expressed as g ww/L ($p = 0.00061$, $R^2 = 0.83$). Weaker relationships were obtained when the EC50 values were expressed as g dw/L ($p = 0.016$, $R^2 = 0.59$) and g TOC/L ($p = 0.0084$, $R^2 = 0.65$). The bivariate plots also revealed three outliers (Hjuvik, Marstrand, and Nödinge), with more toxic sediments than the Cu, Zn, and ΣBT pollution indices alone would predict (Fig. 4).

Discussion

Pollutant Concentrations

TBT was found in high concentrations in the small boat harbor sediments, despite the fact that its use in antifouling paints for pleasure boats shorter than 25 m has been forbidden in EU since 1989 (Council Directive 89/677/EEC). Eklund et al. (2008, 2010) have suggested that this phenomenon may be due to release from old paint layers in connection with high pressure water hosing of the boat hulls. Another likely source of TBT is run-off from adjacent boat yards, which often are heavily polluted by organotin compounds (Bengtsson and Wernersson 2012; Eklund and Eklund 2014; Eklund et al. 2014). Support for antifouling paints as the predominant source of ΣBT in the sediments of the investigated small boat harbors comes from the correlation between these pollutants and the number of boat places in each harbor. Although organotin compounds also have been used as plastic stabilizers, pesticides, and biocides in wood preservatives (EFSA 2004), such contribution to the observed sediment content is probably low. Several investigations have shown that background concentrations of organotin compounds often are at least one order of magnitude lower than those observed in many of the small boat harbors investigated here (Thomas et al. 2000; Cato 2003; Sternbeck et al. 2006; Eklund et al. 2010). High TBT concentrations (up to 1330 μg/kg dw) in the sediment have also been observed in coastal marinas in other parts of Sweden (Eklund et al. 2008) and northern Europe. Biselli et al. (2000) reported TBT concentrations of up to 220 μg/kg dw in marinas along the German Baltic Sea coast, and Thomas et al. (2000) reported TBT concentrations of 40–1400 μg/kg dw

Table 6 EC50 values with 95 % confidence intervals (CI) for growth inhibition of *Ceramium tenuicorne* expressed as sediment wet weight, dry weight, and total organic carbon (TOC) per liter

Harbor	EC50 (g ww/L)	95 % CI (g ww/L)	EC50 (g dw/L)	95 % CI (g dw/L)	EC50 (g TOC/L)	95 % CI (g TOC/L)
Natural harbors (NH)						
Gluppö-Fläskön	152	126–186	26	22–32	1.3	1.1–1.6
Gråa holmarna	>160	[a]	>38	[a]	>2.1	[a]
Store Bror	>160	[a]	>68	[a]	>1.9	[a]
Tjälleskär	>160	[a]	>96	[a]	>0.96	[a]
Small boat harbors (SBH)						
Björlanda kile	61	45–87	24	18–34	0.88	0.65–1.3
Eriksberg	104	89–127	39	33–47	1.2	1.0–1.5
Hinsholmskilen inner	52	43–60	13	11–15	0.54	0.44–0.62
Hinsholmskilen outer	72	59–83	19	15–21	0.67	0.55–0.77
Hjuvik	23	16–35	6.9	4.8–10	0.19	0.13–0.29
Hålkedalskilen	145	102–231	34	24–54	1.7	1.2–2.7
Kebal	166	163–191	48	47–55	2.1	2.0–2.4
Kungshamn	63	56–70	12	10–13	0.94	0.84–1.1
Marstrand	43	30–61	11	7.8–16	0.66	0.46–0.94
Nödinge	63	57–70	20	18–23	0.55	0.50–0.61
Vallhamn	155	130–157	37	31–37	1.4	1.2–1.4
Önnered	107	94–121	22	20–25	1.3	1.1–1.5

[a] Not possible to calculate

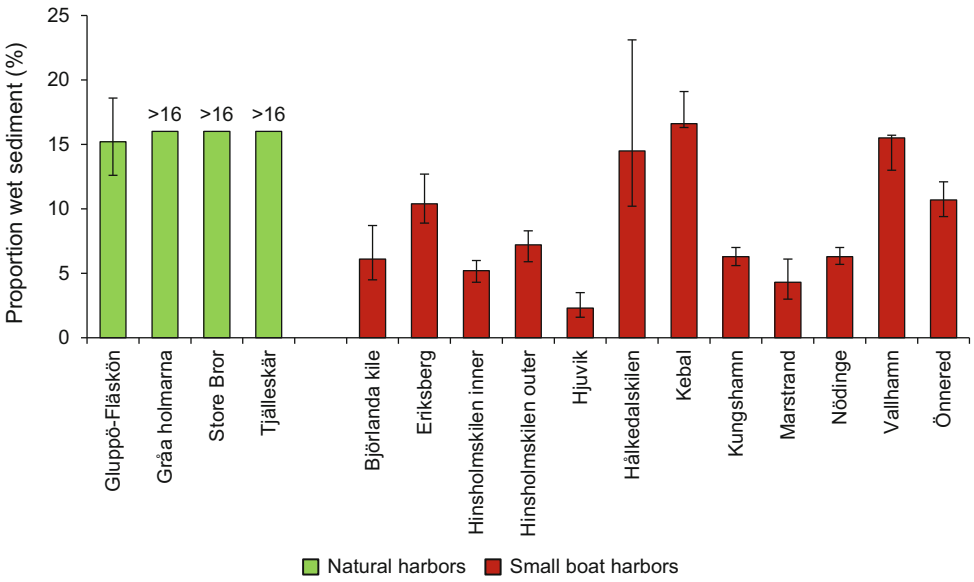

Fig. 3 Proportion of wet sediment from the natural harbors and small boat harbors that produced a 50 % growth inhibition in *Ceramium tenuicorne*. A toxic effect of the sediment was observed in all eleven small boat harbors and in one of four natural harbors. Error bars indicate 95 % confidence intervals (Color figure online)

in estuaries and marinas along the coast of UK. For comparison, TBT concentrations in the range 200–5000 µg/kg dw have been reported in larger harbors for ships (e.g., Harino et al. 1998; Diez et al. 2002, 2006; Cornelissen et al. 2008). It also should be noted that the degradation of TBT in sediments is slow at Swedish latitudes. An investigation of contaminants in the Oslo harbor area (not far from the Swedish west coast) estimated the half-life of TBT in the sediment to more than 10–20 years (Cornelissen et al. 2008). The observation that ΣBT dominated largely over ΣPT is supported by the fact that TBT has been commonly used in antifouling paints for

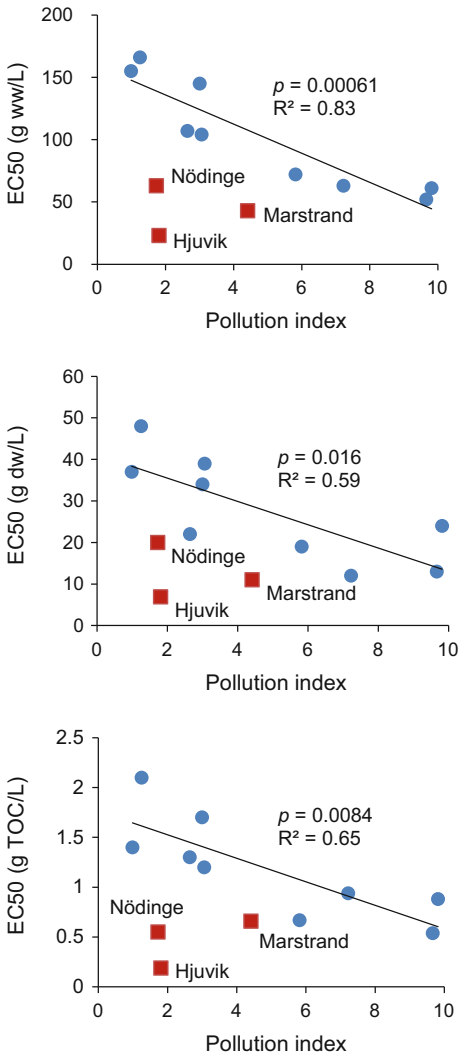

Fig. 4 Linear regression of EC50 values for growth inhibition in *Ceramium tenuicorne* on the combined pollution indices for copper (Cu), zink (Zn), and butyltins (ΣBT) in the small boat harbors. *From the top* EC50 values expressed as gram wet weight per liter; EC50 values expressed as gram dry weight per liter; and EC50 values expressed as total organic carbon (TOC) per liter. In each regression, there were three outliers (Hjuvik, Marstrand, Nödinge), which had a more toxic sediment than predicted by the pollution indices. The best linear regression was obtained for EC50 values expressed as gram wet weight per liter (Color figure online)

pleasure boats, whereas antifouling paints containing TPT have been used only in small amounts on pleasure boats in most countries except Japan (Horiguchi et al. 1997).

In the implementation of the water framework directive, EU has established a predicted no effect concentration (PNEC) value for TBT in sediment of 0.02 µg/kg dw (Anon 2005). A corresponding value established by the Norwegian Pollution Control Authority is 0.002 µg/kg dw (Anon 2007). It should be remembered, however, that such low concentrations are difficult to quantify. They may rather be taken as indications that biological effects start to occur already at very low concentrations. The highest sediment TBT concentration in the present study, 560 µg/kg dw, is 28,000 times higher than the EU threshold and 280,000 times higher than the Norwegian threshold. The threshold for good chemical sediment status established by the Swedish Agency for Marine and Water Management is 1.6 µg/kg dw (Anon 2015). The highest sediment TBT concentration in the present study exceeds this threshold by a factor of 350. Also the Norwegian Pollution Control Authority has established management related thresholds, and here the lower limit for the class of highest pollution is 100 µg/kg dw (Anon 2007). The highest sediment TBT concentrations in the present study are still higher than this threshold. This means that TBT in the sediment occurs in toxic concentrations in all small boat harbors investigated here, and at least five of them must be considered as severely polluted by TBT.

Support for antifouling paints as the predominant source of Cu, Pb, and Zn in the sediments of the investigated small boat harbors comes from their pairwise correlations with each other and with ΣBT. Cu and Zn also were correlated with the number of boat places in each harbor. Moreover, sediment concentrations of these metals have been demonstrated to be much lower in background areas (Eklund et al. 2010). Similar to the harbors on the Swedish west coast, high concentrations of Cu (40–800 mg/kg dw) in sediments have also been observed in coastal marinas along the Baltic Sea coast (Eklund et al. 2008, 2010) and in other parts of Europe (e.g., Warnken et al. 2004; Jones and Bolam 2007; Singh and Turner 2009; Takahashi et al. 2012; Turner 2013). This was expected, since Cu has been the most commonly used active substance in antifouling paints during the last decades (e.g., Yebra et al. 2004; Jones and Bolam 2007). The concentrations of Pb in the sediments in the present investigation deviated notably less from preindustrial concentrations than those of the other analyzed pollutants. In 6 of 15 harbors, there was no deviation at all. This observation may be related to the fact that restrictions to the use of Pb were introduced already in 1976 (Council Directive 76/769/EEC). Moreover, the need to protect wooden boats from ship worm has declined, as most pleasure boats today are made of plastic. Zinc is currently not classified as a biocide, but because of its potential toxic properties it has been considered as a substance of concern in risk assessment (personal communication, Anneli Rudström, Swedish Chemicals Agency).

The sometimes rather high concentrations of Σ16PAH and their lack of correlation with Cu, Pb, Zn, and ΣBT indicate that boat motor exhausts are not the only source of PAHs in the investigated harbors. Support for this hypothesis comes from an investigation of PAH concentrations in surface sediments (top 0–5 cm) in the Stockholm archipelago along a pollution gradient from central

Stockholm to the outer archipelago in the Baltic Sea (Hansson et al. 2007). The comparison was based on those ten PAHs that were measured in both investigations: phenanthrene, fluoranthene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*ghi*]perylene, pyrene, and indeno[1,2,3-*cd*]pyrene. Kebal had very high PAH concentrations, corresponding to ca half of those in central Stockholm. A likely explanation is that this area was drained and used as industrial premises for the building of an oil rig in the 1970s. When the oil rig was finished, the area was submerged again, and later the small boat harbor was established. The PAH concentrations in Eriksberg, Hjuvik, Kungshamn, and Marstrand were similar to those in the moderately polluted inner Stockholm archipelago ca 17–26 km downstream from central Stockholm, whereas the PAH concentrations in Gluppö-Fläskön, Store Bror, Björlanda kile, Hinsholmskilen outer, Nödinge, and Vallhamn were more similar to those in the less polluted middle and outer Stockholm archipelago ca 52–84 km downstream from central Stockholm, i.e. the Baltic Sea background. Even lower PAH concentrations were found in Gråa holmarna and Tjälleskär. It is reasonable to assume that the sediment content of Σ16PAH in the small boat harbors originate not only from boating, but also from local sources in the surrounding municipalities. The high fraction of carcinogenic PAHs in the sediments calls for attention.

The generally lower pollutant concentrations in the natural harbors than in the small boat harbors were expected. Still, elevated pollutant concentrations and toxic effects were detected in some of the natural harbor sediments. This is especially noteworthy, since pleasure boats in Sweden, on average, are away from their home harbors only 29 days of the 5 months long boating season (Anon 2010). This also implies that most of the pollutant release occurs in the home harbors (Hoch 2001).

Sediment Toxicity

The difference in pollutant concentrations between the natural harbors and the small boat harbors was reflected by the toxicity of the sediments. All small boat harbor sediments caused various degree of growth inhibition of *C. tenuicorne*, whereas only one natural harbor sediment caused such growth inhibition. The latter observation indicates that the boating pressure today may be high enough to produce toxic effects even in natural harbors in pristine areas. The toxicity was generally well predicted by the combined Cu, Zn, and ΣBT pollution indices, although the sediments from Hjuvik, Marstrand, and Nödinge where more toxic than predicted by these pollution indices. Apparently, other toxic substances than those analyzed here occur in the sediments. Still, the general relationship between the combined pollution indices and growth inhibition indicated that Cu, Zn, and ΣBT were responsible for a significant part of the sediment toxicity.

As also demonstrated previously (e.g., Eklund et al. 2010), the present investigation shows that the growth inhibition method with *C. tenuicorne* is useful for characterization of sediments and a good complement to chemical analyses. The procedure to use natural water elutriates from fresh sediments involves minimal handling of the sediment and mimics the way water-dwelling organisms are exposed to water soluble pollutants in the field. An advantage is also that organic solvents are avoided. The degree of toxicity cannot, however, be extrapolated directly to other types of organisms. For example, *C. tenuicorne* has been demonstrated to be more sensitive to this type of toxicity than other indicator species (Eklund et al. 2010). Other ways to analyze sediment toxicity involve sediment suspension, pore water, extract or whole sediment, whereby the effect on various endpoints is measured (e.g., Nendza 2002; Castro et al. 2006; McPherson et al. 2008; Eriksson Wiklund et al. 2009). So far, there is no customary way to express the toxicity of sediments in such experiments. We found that the strongest relationship between toxicity and the major pollutants was obtained when the EC50 values were expressed as g ww/L. In this respect, g ww/L appears to be preferable to g dw/L and g TOC/L, at least for the toxicity of pollutants that can be elutriated with natural sea water. It should be remembered that the present investigation was not designed to extract lipophilic pollutants, and accordingly, their toxicity may still be more strongly related to dry weight or TOC of the sediment. It also should be remembered that the sediment may be a significant source of lipophilic pollutants in the field, because lipophilic pollutants accumulate in biota even at low concentrations in the water. Hence, the total toxicity of the sediments in the field may be even stronger than that of the water elutriates analyzed here.

Conclusions

The growth inhibition method with *C. tenuicorne* used here indicates that the boating pressure today may be high enough to produce toxic effects even in natural harbors in pristine areas. Because the sensitivity differs between organisms, the degree of toxicity cannot be extrapolated directly to other organisms. TBT still constitutes a serious pollution problem in small boat harbors, even though it was prohibited more than 25 years ago. The strongest relationship between the toxic effect on *C. tenuicorne* and the major pollutants in the small boat harbors was obtained when the sediment toxicity was expressed as gram wet weight per liter compared with gram dry weight per liter

and gram TOC per liter. Hence, for pollutants that can be elutriated with natural sea water, sediment toxicity expressed as gram wet weight per liter appears preferable.

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