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RESEARCH ARTICLE

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The ocean quahog *Arctica islandica* L.: a bioindicator for contaminated sediments

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Abstract The use of benthic organisms as bioindicators in the aquatic environment is a suitable method for assessing the effects of contaminants in coastal waters. The accumulation of heavy metals in body tissues due to lifestyle and feeding mechanisms makes it possible to reveal contamination rates and recovery trends within polluted areas. Comparing a polluted historical dumping site in the inner Mecklenburg Bight (western Baltic Sea) with a less-contaminated reference site at the edge of the Mecklenburg Bight, representing the background contamination of the western Baltic Sea, the present study discusses the population structure and heavy metal exposure of the ocean quahog *Arctica islandica* L. (Mollusca, Bivalvia) and evaluates this organism as a bioindicator for contaminated sediments. The organism density was higher at the reference site in comparison to the dumping site. The absence of juvenile and adult individuals at the dumping site seems to be a sign that this ecological environment has not completely regenerated since the dumping event in the late 1950s to early 1960s. Heavy metal concentrations of copper, lead, and zinc in the soft body tissue of *A. islandica* were analysed using atomic absorption spectrometry (AAS). Shell measurements were carried out using laser ablation–inductively coupled plasma–mass spectrometry. Particularly the concentrations of copper and lead were significantly higher in the soft body tissue as well as in the shell from the dumping site than from the reference site. For pollutant biomonitoring research, the shells of the ocean quahog can be used as an indicator for heavy metal accumulation. They are more suitable for reflecting historical contamination events than the soft body tissue.

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Introduction

Heavy metals in the marine environment are of considerable interest because they remain in the organism and may accumulate and magnify in body tissue (Clark 1997). Due to the substantial persistence of toxic contaminants along the food chain (Clark 1997; Donazzolo et al. 1981; Giordano et al. 1991), there is a threat to natural populations, as well as a potential human health risk. Plants and animals regulate their metal contents over a limited range. Heavy metals that are not excreted remain in the body tissue and are continuously accumulated. The transfer of heavy metals in the food chain is of major importance (Clark 1997). Toxic substances with long biological half-lives are increasingly accumulated towards the end of the food chain. The accumulation of heavy metals in marine systems is well documented for benthic living organisms, for example, *Mytilus edulis*, *Crassostera* sp., *Scrobicularia plana*, *Echinocardium cordatum*, and *Crangon crangon* (e.g. Bryan and Uysal 1978; Borchardt et al. 1988; Karbe 1990; Harms and Huschenbeth 1991; Bakker 1994; Páez-Osuna et al. 2002). Rainbow et al. (2002) discussed the important fact that the accumulation of heavy metals in organisms shows an enormous variability across the invertebrate taxa. Molluscs in particular are known for their ability to accumulate toxic metals via food uptake (e.g. Phillips 1977; Wright 1978; Prosi 1979; Franco et al. 2002).

The ocean quahog *Arctica islandica* is an arctic-boreal species occurring in the North Atlantic, North Sea, and the western Baltic Sea (Merill and Ropes 1969). They are found at water depths of 16 to 30 m in the Mecklenburg Bight (Baltic Sea) and at depths of 8 to 256 m in the Middle Atlantic (Merill and Ropes 1969; Rowell and Chaisson 1983; Zettler et al. 2001). *A. islandica* lives buried in sandy mud or mud sediment, inhabiting the boundary layer between substratum and water column, and is therefore directly exposed to heavy metals (Szefer and Szefer 1990). *A. islandica* is highly resistant to oxygen deficiency and hydrogen sulphide (Theede et al. 1969). Due to its lifestyle and feeding mechanisms (filter feeding) a high tolerance to the incorporation of metal-containing particles is possible (Winter 1970; Owen and Richardson 1996; Cargnelli et al. 1999). Therefore, this species is considered to be a good bioindicator for contaminated sediments (Steimle et al. 1986). In addition, extremely long life-spans (e.g. Baltic Sea: 80 years, Atlantic: >200 years; Murawski et al. 1982; Ropes 1985), as well as a wide distribution make this species a suitable object for tracing back historical events of contamination.

In the 1960s, various industrial waste products containing high concentrations of heavy metals and organic pollutants, including polycyclic aromatic hydrocarbons (PAHs), were deposited in the inner part of the Mecklenburg Bight (54°05′N, 11°00′E). Large-scale geochemical sediment-mapping programs of the whole bay in the mid 1980s (Leipe et al. 1998) and later in 1997 revealed that a part of the contaminated material was distributed over a large area around a "hot spot" in the first decades after the dumping. This was caused by natural sediment resuspension and transport processes (Kersten et al. 2005). Because of a general (natural) sedimentation rate of 1–2 mm/year in the whole area, a stepwise dilution and coverage of the contaminated material can also be established. Today, most of the historically dumped waste is overlaid by at least 5 cm of young sediment (Liehr 2002). However, in the area close to the dumping site the concentrations of contaminants and pollutants in the recent sediments are still much higher than in the sediment horizon below the dumped material (i.e. before the dumping activities) and in comparison to the recent sediments of the outer part of the bight.

This article evaluates the use of *A. islandica* as a bioindicator by analysing the population structure and heavy metal concentrations in the soft body tissue and shells from an industrial waste dumping site compared to a reference site representing "normal" contamination in the western Baltic Sea. Additionally, a laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS) was used to trace shell profiles for historical variability in heavy metal contamination throughout the last decades.

Materials and methods

Area of investigation

Sampling occurred in the western Baltic Sea at a contaminated historical dumping site (DS) and at a reference site (RS). Both stations are located in the Mecklenburg Bight (Fig. 1). The reference station was located about 25 nautical miles north-east of the historical dumping ground, which is far enough away not to be influenced by the dumped waste material. This station represents the "normal" situation of the western Baltic Sea. Here, the level of "contamination" by anthropogenic substances corresponds to the large-scale situation and is comparable to other sedimentary basins of the western Baltic Sea (Leipe et al. 1998, 2005). Different investigations of the heavy metal concentration in the surface sediment at the dumping and reference site between 1988 and 2003 showed that the mean value of the different sampling years of lead (Pb: 654 mg kg^{-1}), zinc (Zn: 957 mg kg^{-1}), and copper (Cu: 93 mg kg^{-1}) is

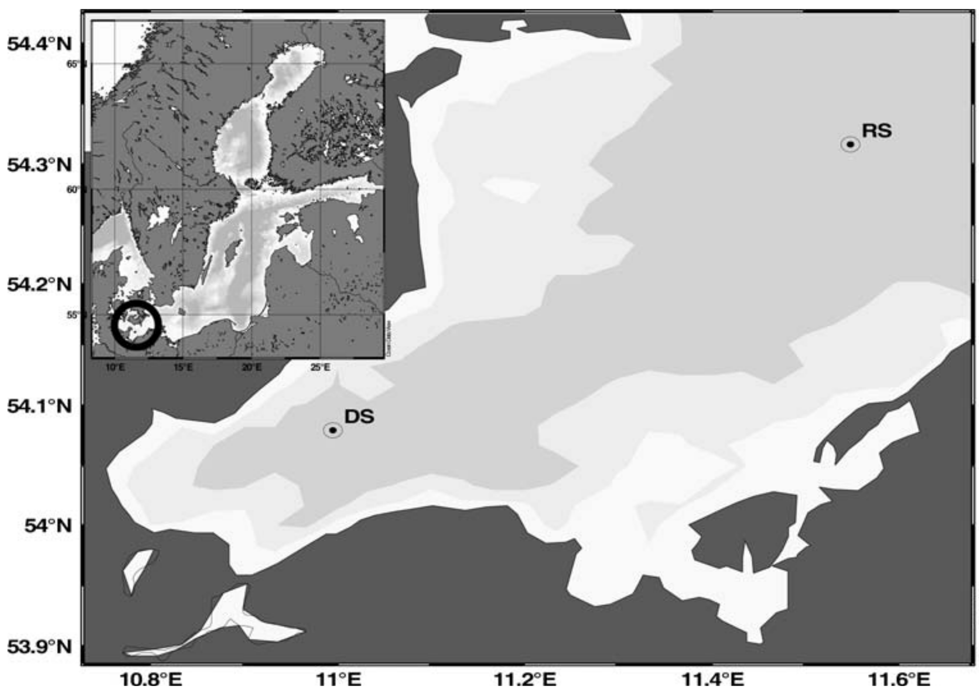

Fig. 1 Areas of investigation: historical dumping site (*DS*) and reference site (*RS*), western Baltic Sea, Mecklenburg Bight (map software from Schlitzer 2001)

much higher at the dumping site compared to the reference site (Pb: 107 mg kg^{-1}, Zn: 262 mg kg^{-1}, Cu: 62 mg kg^{-1}). Consequently, we are able to compare a historical, industrial waste-dumping site with a, for the western Baltic Sea, ''normally'' contaminated reference site.

The Baltic Sea is one of the largest semi-enclosed brackish-water regions of the world (Matthäus 1996). It is connected to the North Sea through three small channels (Danish Straits). The surface water is characterized by a low salinity and density whereas the deep layer shows high salinity and density. Mixing of these two layers is possible during thermal stratification breakdowns and strong westerly winds (Krauss and Brügge 1991). Furthermore the Baltic has a salinity gradient from the western (32‰) to eastern and northern (1–2‰) areas.

The Mecklenburg Bight is located between the North Sea and the central Baltic Sea. The currents are influenced by different processes in the two seas. The environmental conditions at the dumping site and reference site are similar. The water depth is between 25 and 30 m. Both investigation areas belong to the ''shallow water regions'' of the Baltic. The sediment at the dumping site and reference site consists of mud with an organic carbon content of approximately 5%. The grain size at both stations is silt. The salinity ranges between 13‰ and 29‰ depending on the season.

Field investigation

During an expedition with the research vessel ''Professor Albrecht Penck'' in May 2001, *Arctica islandica* was collected from both sites using a dredge three times per station (dredge time approximately 10–15 min) . The ocean quahogs were stored at –20°C after 1–2 h of depuration to eliminate sediment from the digestive tract. Soft body tissues from ten individuals and shells from three individuals per sampling site were prepared for the laboratory analysis of lead, copper, and zinc.

Analysis of population structure

All live ocean quahogs at both stations were counted. Shell length (defined as the maximum anterior to posterior distance in millimetres), shell width, and shell height were measured. The analysis of the population structure was performed according to the method of Zettler et al. (2001).

Analytical methods

For the investigation into the heavy metal accumulation in soft body tissue of *A. islandica*, ten organisms of the same size (approximately 25–30 mm shell length, age 10 years; Liehr 2002) from each station were analysed separately. The preparation and analysis of the samples were done according to the method of Kremling (1999) for the determination of trace elements. The soft body tissues were freeze-dried and homogenized. Ten milligrams of freeze-dried homogenized tissue was mixed with 1.5 ml nitric acid (HNO_3) and 100 µl perchloric acid ($HCLO_4$). Then 1 *M* HNO_3 was added and the heavy metal concentration measured with atomic absorption spectrometry (AAS) type Perkin Elmer Analyst 800. The results were calculated on a per gram dry mass basis and the individual measurements were used to calculate the mean and SD. Analysis quality assurance was checked by using internationally certified and in-house reference materials (TORT1 for heavy metals in marine biota), as well as successful performance in international intercomparison exercises such as in the framework of the QUASIMEME program.

Laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS) was used to determine the heavy metal concentrations in the internal growth bands of the cardinal teeth (Fig. 2). This allowed a chronological report of the heavy metal accumulation in the shells of *A. islandica* to be reconstructed. Measurements of heavy metals in the external growth band were not possible because of technical difficulties. The laser would shoot through a number of different growth rings. Consequently, the heavy metal accumulation over several years would be measured and no trend information would be possible. The analyses were conducted at the Johann Gutenberg Institute at the University of Mainz. The oldest individuals were taken to get an historical review (dumping site: approximately 20 years, reference site: approximately 35 years, Liehr 2002). The shell samples were positioned under a laser-permeable silica cell with three-dimensional freedom of orientation and an Nd:YAG laser was targeted at the samples. The ablation width was reduced to 10 µm diameter (Fig. 2). This enabled analyses within the different shell growth

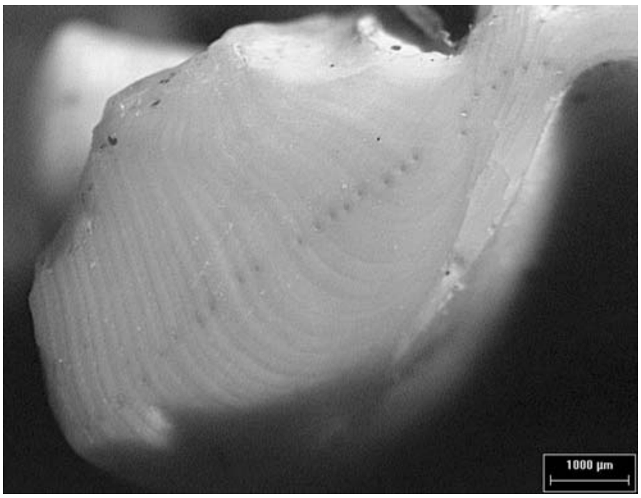

Fig. 2 Cardinal tooth of *Arctica islandica* after laser shooting (stereomicroscope photo)

bands (Fig. 2). The ablated material was passed into a stream of Argon gas directly into the plasma of the ICP-MS (Perkins et al. 1991; Pearce et al. 1992). The significance of the results for heavy metal accumulation in both the soft body and shell were investigated using a *t*-test (normality test, comparison of two mean values of two independent samples).

Results

Population structure and shell features

Population structure and shell features were estimated by measuring the following parameters: shell length, width, and height. The measured shell lengths were classified in size classes (Fig. 3). At the reference site individuals in all size classes from the 0 to 5-mm class to the >65 to 70-mm class were found. The most abundant sizes were between 25 and 35 mm. At the dumping site, the smaller and bigger sizes were not found. The most abundant sizes were between 20 and 45 mm. Differences in size classes between the two stations were not significant ($P=0.05$, *t*-test for class groups). Observed differences are due to the different numbers of individuals analysed at the respective stations. Twice as many individuals were found at the reference site ($n=236$, 66 individuals/m^2) compared to the dumping site ($n=116$, 1 individual/m^2). However, no recruits or even settled juvenile specimens could be found at the dumping site.

The shell features are plotted against each other in Fig. 4. The linear correlations in shell length with shell height, length with width, and width with height are obvious. Regression analysis on the measured parameters showed a good linear fit. No significant difference between the two stations was evident. All parameters show a close tangential correlation at both stations ($R=0.99$, $P=0.0001$).

Heavy metal accumulation in *A. islandica*

Soft body

Ocean quahogs from the dumping site contained significantly higher Pb (11 mg kg^{-1}) and Cu concentrations (28 mg kg^{-1}) than those from the reference site ($P=0.025$, one-sided *t*-test; Table 1). No significant differences in Zn contents were observed between the ocean quahogs from the two stations (one-sided *t*-test).

Shells

For analysis of Zn, Pb, and Cu concentrations in shells, three specimens each from the dumping and the reference site were measured using LA–ICP–MS. Using this method it is possible to determine the heavy metal exposure per growth year of *A. islandica*.

For Cu concentrations, significant differences ($P<0.005$, *t*-test) between the two stations were analysed (Table 2). Shells from the dumping site contained Cu values between 0.65 and 14.05 mg kg^{-1}. In shells from the reference site the Cu concentrations were lower, from not detectable (ND) to 7.89 mg kg^{-1}. Lead was also measured at significantly higher concentrations in shells from the dumping site ($P<0.0001$, *t*-test; Table 2). The maximum concentration (14.68 mg kg^{-1}) was up to 10 mg kg^{-1} higher than in shells from the reference site (from ND to 3.48 mg kg^{-1}). Zinc showed a similar tendency to the Cu and Pb concentrations (Table 2). Shells from the dumping site had maximum values up to 107 mg kg^{-1}, whereas for shells from the reference site concentrations between 5.66 and 68 mg kg^{-1} were measured. The Zn concentrations in shells from the dumping site and reference site were significantly different ($P<0.0001$, *t*-test).

When comparing the mean values of the metal concentrations, higher values in the shells from the dumping

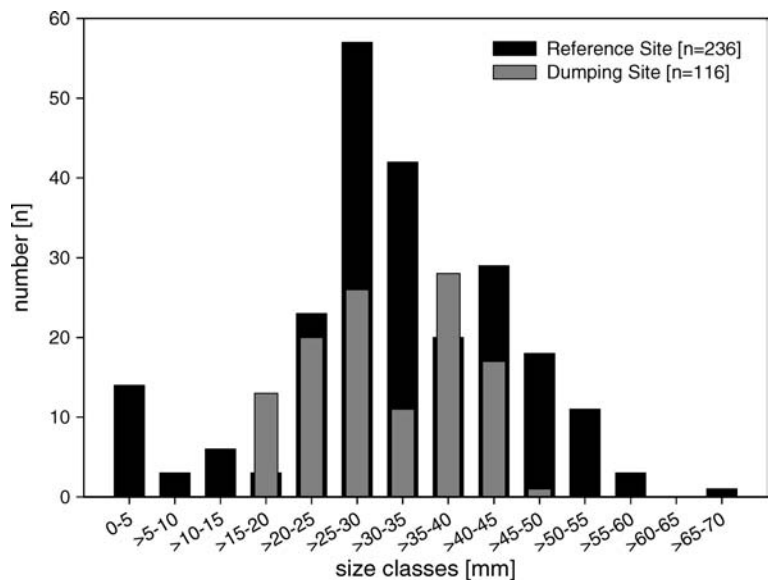

Fig. 3 Comparison of number of individuals (*A. islandica*) per size class from the dumping site (inner Mecklenburg Bight) and the reference site (outer Mecklenburg Bight) in August 2001

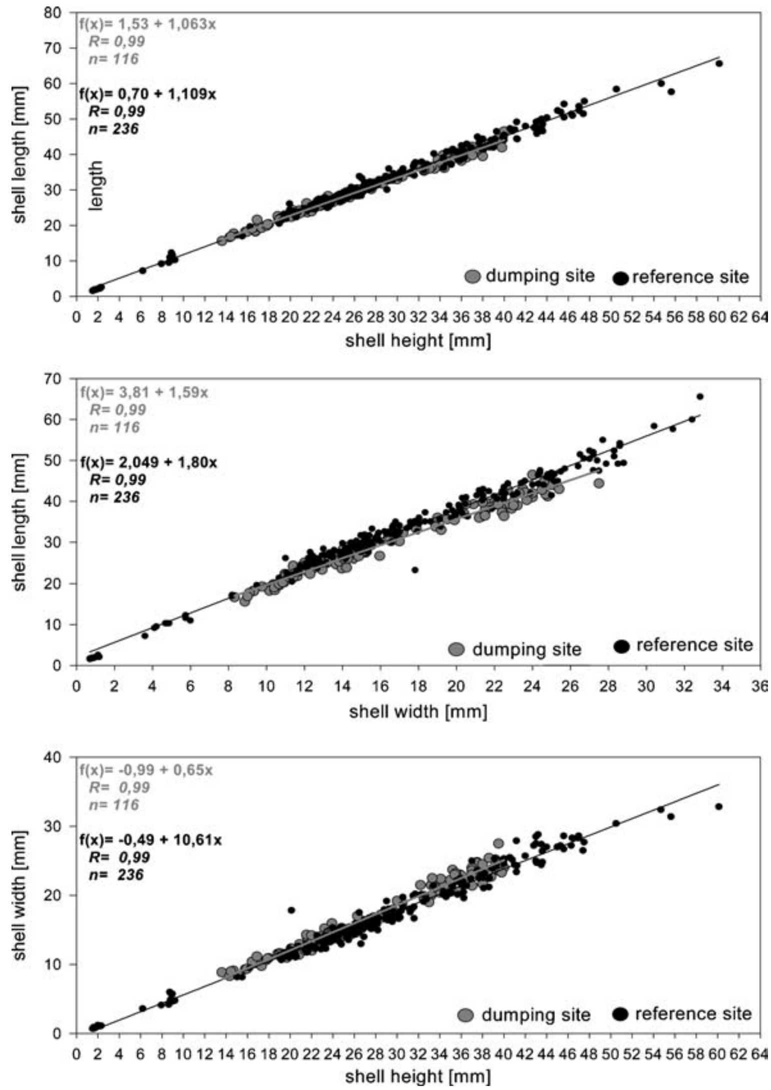

Fig. 4 A direct comparison of shell parameters of *A. islandica* from the dumping site and the reference site in August 2001

Table 1 Mean metal concentrations in soft body tissue of *Arctica islandica* from the outer Mecklenburg Bight (*reference site*) and inner Mecklenburg Bight (*dumping site*) and some metal concentration in *A. islandica*from the western Baltic (*WB*) and the north-west Atlantic (*NWA*)

Locality		Reference	Cu (mg kg^{-1})	Pb (mg kg^{-1})	Zn (mg kg^{-1})
Georges Bank–Nantucket	NWA	Stick (1978)	3.5	0.35	252
		Steimle et al. (1986)	10.3	4.1	62
Block Island	NWA	Steimle et al. (1986)	10	10.2	102
New York Bight	NWA	Steimle et al. (1986)	11.3	5.7	95
Chesapeake Bight	NWA	Steimle et al. (1986)	5.4	4.7	71
Montauk Point, N.Y. to Cape Hatteras, N.C.	NWA	Wenzloff et al. (1979)	5.04	< 1.1	13.2
Süderfahrt	WB	Swaileh and Adelung (1994)	14.9	0.84	226
		Swaileh (1995)	13.9	1.44	166.5
Millionenviertel	WB	Swaileh and Adelung (1994)	15.3	1.55	188
		Swaileh (1995)	13.8	1.76	167.2
Dorschmulde	WB	Swaileh and Adelung (1994)	13.3	1.08	144
		Swaileh (1995)	15.8	1.75	148.2
Vejsnes Kanal	WB	Swaileh and Adelung (1994)	13.8	1.25	113
		Swaileh (1995)	13.8	1.75	123.1
Reference site, mean (± SD)		This study	17.73 (± 3.8)	4.2 (± 1.96)	185.53 (± 33.1)
Dumping site, mean (± SD)		This study	27.44 (± 10.3)	10.78 (± 6.6)	174.73 (± 33.0)

Table 2 Levels of the elements Cu, Pb, and Zn in the shells of *A. islandica* (western Baltic Sea, this study, 2001) and *Modiolus modiolus* (North Sea, Richardson et al. 2001). *ND* Not detectable

	Dumping site	Reference site
Cu concentration (mg kg^{-1})		
Max.	14.05	7.89
Min.	0.65	ND
Mean ± SD	2.59 ± 3.37	1.43 ± 1.96
Richardson et al. 2001	5.04 ± 7.42	1.51 ± 0.88
Pb concentration (mg kg^{-1})		
Max.	14.68	3.48
Min.	0.59	ND
Mean ± SD	4.92 ± 3.65	0.97 ± 3.81
Richardson et al. 2001	3.65 ± 1.55	1.67 ± 0.60
Zn concentration (mg kg^{-1})		
Max.	107	68
Min.	6.21	5.66
Mean ± SD	37.86 ± 28.21	19.93 ± 14.66
Richardson et al. 2001	9.52 ± 7.29	4.04 ± 2.69

site were obtained (Table 2). The high SDs are a result of the single measurements in the shells of *A. islandica*, which induce high variabilities. The heavy metal accumulation in the shells is different per life year and produces mean values with higher SDs.

Historical review of metal concentrations

The internal growth bands in the shells of *A. islandica* were used for analysing the historical Cu, Pb, and Zn concentration trends. The profiles, that is, the concentration of an element in relation to a single life year, showed different variations between the two stations (Fig. 5). An obvious trend was not visible. However, the Cu, Zn, and Pb contents were higher in the shells from the dumping site (Fig. 5, Table 2).

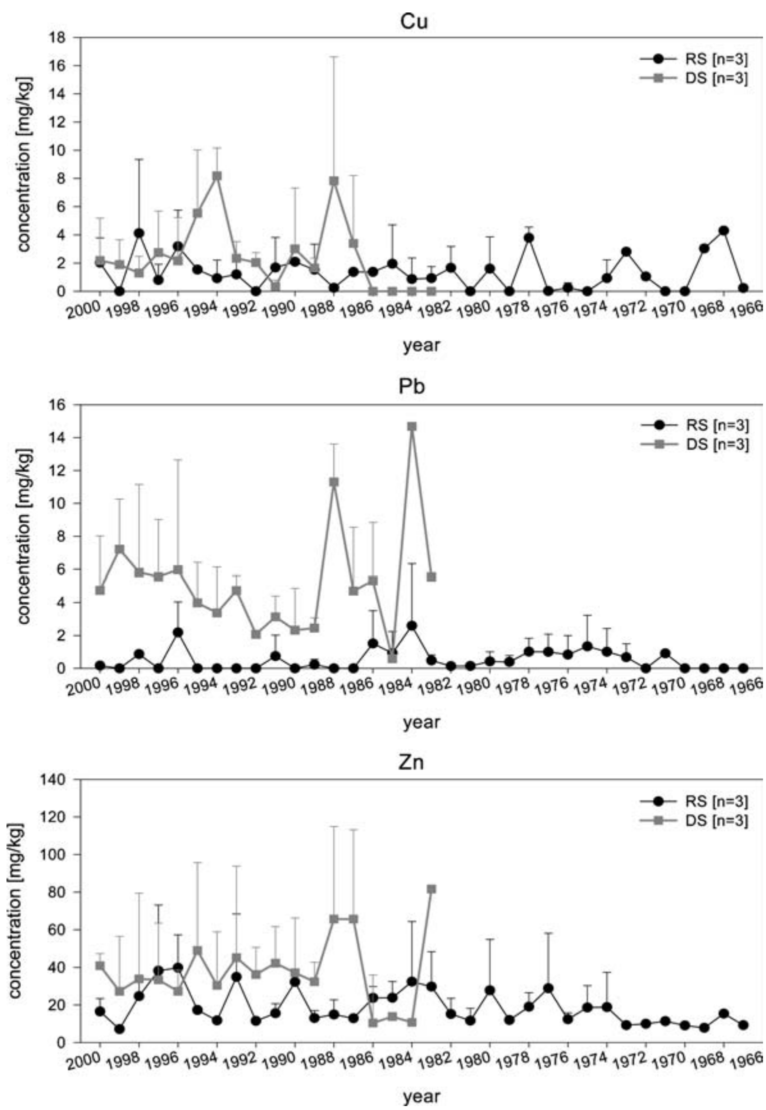

Fig. 5 Chronological review of heavy metal concentration in the shells of *A. islandica* from the dumping site (*DS*) and reference site (*RS*) in August 2001 using laser ISP–MS; measured in the cardinal tooth

Discussion

Bioindicators react to particular environmental pollutants. On the one hand they accumulate the pollutants, and on the other hand these pollutants affect, for example, the growth, reproduction, and population structure of indicator individuals (Fent 1998). Consequently, it is important to analyse these parameters in terms of size and age.

Population structure and shell features of the ocean quahog

There are similarities in the shell features of *A. islandica* of this study with research from north-west Iceland (Thorarinsdottir and Einarsson 1996), the middle Atlantic Bight (Thompson et al. 1980a, 1980b), and other Baltic localities (Zettler et al. 2001). At both sites in this study a significant difference in shell size was not evident. It is often stated that any growth differences could be the result of sex-specific (Rowell et al. 1990; Fritz 1991) and geographical differences (Witbaard et al. 1999).

In addition, the linear regression between the different shell features (shell length, height, and weight) showed no differences between the dumping and reference sites (Fig. 4). Both populations showed similar growth. Because the shell features (i.e. width, height, length) were similar at the two sites, it is likely that the sediment contamination at the dumping site had no influence on the latter.

At the dumping site smaller and bigger quahogs were not found (Fig. 3). This is a sign of missing recruitment. Perhaps the parent generation was not able to produce recruits or the environmental conditions were not favourable for larval survival. In this study the population structure is indicative of a disturbance or stress resulting from pollution at the dumping site. Another factor could be occasional oxygen depletion. However, both stations would probably be affected in similar ways.

Heavy metal content in *A. islandica*

Bioindicators react to possible environmental contamination and accumulate pollutants (Fent 1998). Molluscs accumulate heavy metals via food (Phillips 1977; Wright 1978; Prosi 1979). High accumulation in the soft body tissue or shell could be a result of the filtration of metal-containing particles (Bryan and Uysal 1978). The ocean quahog lives in the upper 5 cm of the sediment. It belongs to the group suspension feeders and absorbs particles present in its respiring water flow. It inhabits the boundary layer between the sediment and water and is thus directly exposed to heavy metals (Szefer and Szefer 1990).

In this study, the individuals from the dumping site had higher concentrations of lead and copper in the soft body tissue (Table 1). The concentrations of zinc showed no significant difference. Nevertheless, the heavy metal concentrations must be in the tolerance range, since in both research areas no differences in the shell features of the ocean quahogs were determined. However, heavy metals exhibit toxic effects that affect all life stages of shellfish, especially development stages (Calabrese et al. 1973; Calabrese and Nelson 1974; Thurberg et al. 1975). Consequently, the heavy metals could have an influence on reproduction because at the dumping site no young individuals were found (Fig. 3). This would indicate that at least the adults were not able to continuously reproduce recruits. A comparison with other studies shows that the copper and lead contents in the soft body tissues from the dumping site in this study were much higher (Table 1). Whereas the soft body tissue of *A. islandica* is not suitable for biomonitoring in terms of quantifying contamination along a time gradient, the shells of the ocean quahogs are indeed suitable.

The shell analyses were made to obtain an insight into the exposure to contamination and also to obtain a chronological review for investigating trends. Fuge et al. (1993) analysed copper concentrations (1 to 10 mg kg^{-1}), lead concentrations (0.49 to <6 mg kg^{-1}) and zinc concentrations (2 to 35 mg kg^{-1}) in the shells of *Patella* spp. In this study, higher copper, lead and zinc concentrations at the dumping site were found compared to the above study (Table 2). In shells from the Kiel Bight, the lead and copper concentrations were also lower than those from the dumping site of this study (Swaileh 1995). Consequently, high copper, lead and zinc accumulation in the shells is indicative of contamination at the dumping site.

The shell of the ocean quahog has internal growth bands, which are formed annually (e.g. Murawski et al. 1982; Ropes 1994). By carrying out measurements in several internal growth bands it is possible to get a chronological reconstruction of the metal content. Studies by Raith et al. (1996) for heavy metal accumulation in *A. islandica* shells with a chronological reconstruction suggested that it is possible to use the shells for environmental research. Their research showed a decreasing trend of lead and strontium up to the recently formed growth rings. Until now just a few studies have been available for a discussion of the historic concentration trends for cadmium, copper and zinc in shells (e.g. Westermark et al. 1996; Richardson et al. 2001).

In this study the profile of heavy metal content in the shells showed no unique trend at either station (Fig. 5). The waste material dumping of the 1950s/1960s in the inner Mecklenburg Bight is not reflected in the chronological review. One reason is the young age of the quahogs. To investigate the primary contamination the ocean quahogs would have to be 40 or 45 years old. In the dumping site quahogs with maximum 50 mm shell length were found. According to Zettler et al. (2001), a quahog with 50 mm shell length from this area is not older than 35 years. In addition, ocean quahogs live at up to 5 cm sediment depth. Bioturbation and other biological, chemical or hydrographical processes are possible reasons why the contaminated material may shift to different deeper horizons, thus reducing exposure. Conversely, these processes could also be reasons for the churning up of the deeper contaminated material so that in different years higher metal concentrations are once again found in the shells.

Studies by Richardson et al. (2001) with the shells of *Modiolus modiolus* from two different areas (a dumping and a reference site) also showed no trend. The concentrations of his dumping site varied between different individuals. Lead was the only element showing a decreasing trend. This trend was also seen in shells from his reference site. The concentrations of lead and copper were not different between the two research areas. In comparison to the studies of Richardson et al. (2001), in this study the lead and copper concentrations were always higher in shells from the dumping site compared to shells from the reference site (Fig. 5, Table 2).

The concentrations in the soft body tissue and in shells of the ocean quahog from the dumping and reference sites were compared with the results of different investigations on *A. islandica* from Atlantic and Baltic localities (Tables 1, 2). Mainly the concentrations of copper and lead were essentially higher at the dumping site than known literature values.

In summary, in sediments from the dumping site the heavy metal contamination was much higher compared to the reference site and other areas (Leipe et al. 2005). In this study the higher contamination of some metals was found in both the soft body tissue and in the shells of *A. islandica*. Although the ocean quahog accumulates heavy metals, it is not qualified for bioindicator investigations in terms of the impact of pollutants on the shell features only. For biomonitoring research the shell analysis is preferable for heavy metal accumulation studies.

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