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Comparison of trace metal bioavailabilities in European coastal waters using mussels from *Mytilus edulis* complex as biomonitors

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Abstract Mussels from *Mytilus edulis* complex were used as biomonitors of the trace metals Fe, Mn, Pb, Zn, and Cu at 17 sampling sites to assess the relative bioavailability of metals in coastal waters around the European continent. Because accumulated metal concentrations in a given area can differ temporally, data were corrected for the effect of season before large-scale spatial comparisons were made. The highest concentration of Fe was noted in the North Sea and of Mn in the Baltic. Increased tissue concentrations of Pb were recorded in the mussels from the Bay of Biscay and the Baltic Sea. Low concentrations of metals were determined in the mussels from the Mediterranean Sea and the Northern Baltic. Relatively low geographic variations of Cu and Zn indicate that mussels are able to partially regulate accumulated body concentrations, which means Cu and Zn are, to some extent, independent of environmental concentrations.

Keywords *Mytilus* · Trace metals · European marginal seas · Biomonitoring · Pollution · Seasonal variation coefficient

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Introduction

Marine pollution has been a serious environmental concern on local and global scales for many years. Anthropogenic activities on land, in the water, and in the air contribute to trace metals contamination of sea water, sediments, and biota (Szefer et al. 2002). Living organisms can accumulate both physiologically essential and nonessential trace elements in their tissues, which can cause deleterious effects when they exceed metabolic requirements (Rainbow 1999).

The species of the family *Mytilidae* have been used widely as efficient biomonitors of organic and inorganic contaminants in coastal and estuarine ecosystems for more than 60 years (Lobel et al. 1991), often within the frameworks of national and international monitoring programs. Mussels are useful for monitoring various marine pollutants, because contaminants, including metals, concentrate in soft tissues proportionally to external concentrations, thus, reflecting levels in the ambient environment (Storelli et al. 2000; Andral et al. 2004). Mussels live attached to shallow-water hard substrates; thus, they are exposed to environmental factors on a local scale (Green et al. 2003). The mytilids are widespread and are common on both sides of the Atlantic and Pacific oceans, with a range of occurrence that reaches from the White Sea to the Mediterranean Sea in Europe on the Eastern Atlantic coast, from the Canadian Arctic to North Carolina in the Western Atlantic, from the Arctic to California on the Eastern Pacific coast, and from the Arctic to Japan in the Western Pacific (Zatsepin et al. 1988; Gosling 1992; Seed 1992; Szefer et al. 2002, 2006). Mussels also exhibit high tolerance to fluctuations in hydrological parameters such as temperature, salinity, and dissolved oxygen in the water, which enables them to inhabit a variety of environments (Casas et al. 2004; Szefer et al. 2006). Other features which make the mussels suitable for biomonitoring include its sedentary, epibenthic life strategy, its large size, and its usually high abundance, which means they are easy to collect in sufficient quantities. Additionally, the biology of mussels is well known, and the relevant chemical data can be interpreted in an ecotoxicological context (Walker et al. 2001; Conti et al. 2002; Conti and Cecchetti 2003).

It has been suggested that accumulated trace metals concentrations in bivalves vary significantly with season and body size (Casas et al. 2004; Lobel and Wright 1982; Wallace et al. 2003; Mubiana et al. 2005), which occasionally requires correcting these effects before spatial comparisons are made on large scales. However, the influence of an animal's size (age) on soft tissue metal concentration appears to be ambiguous and can depend on local environmental conditions. For example, according to Boyden (1974), it is usual for element concentration in mollusks to decrease or remain constant with increasing body weight. Casas et al. (2004) showed that smaller mussels were richer in trace metals then larger ones, while Hummel et al. (1997) and Saavedra et al. (2004) reported no major size-related differences in metal concentrations in the soft tissues of *Mytilus e. edulis* or in cultivated mussels *Mytilus e. galloprovincialis*, respectively. Riget et al. (1996) suggested that the relationship between size and element concentrations should be taken into account particularly in areas not affected by human impact, demonstrating that some metal concentrations could increase (Pb) or decrease (Cu) with body size or that metal concentrations could be independent of mussel length (Zn). In contrast, differences in accumulated trace metal concentrations in bivalves between seasons due to factors related to the biology of animals such as the gametogenic cycle and physiology (Rainbow 1999; Rainbow et al. 2000) are well documented. Temporal variations in soft-tissue metal concentrations in mussels rather result from changes in animal physiology than in metal-exposure conditions. This means that the effect of seasonal physiological changes needs to be allowed before geographical comparisons can be made (Mubiana et al. 2005). Changes in metal levels over time can be attributed to the reproductive cycle and the resultant alterations in the relative proportion of biochemical compounds (i.e., protein, lipid, carbohydrate) which fluctuate throughout the year (Casas et al. 2004). The higher concentrations that are often observed in winter decrease in spring (Soto et al. 1995; Regoli 1998) presumably due to a loss of metals stored in gonad tissues during spawning (O'Leary and Breen 1998). The dilution or concentration effects of changing body weight on soft tissue metal concentration provide an alternative explanation for temporal shifts (Borchardt et al. 1988; Sokolowski et al. 2004). Although it is usually impossible to make interspecific (intertaxon) comparisons in accumulated metal concentrations (Lobel et al. 1990), the mussel species along the European coast are so closely related that potential interspecific differences in accumulation patterns are insignificant in comparison to the geographical differences seen in soft tissue concentrations (Blackmore and Wang 2003).

The use of mussels in the environmental monitoring of trace metals has many important advantages relative to the abiotic compartments of the marine environment (Walker et al. 2001). Metal concentrations in bivalves, in contrast to water or bottom sediments, provide information of direct ecological significance and potential relevance to fisheries and human health. What is more, dissolved and particulate-bound metals often vary spatially and temporally and are subjected to strong geochemical influences (Morrisey et al. 1994) whereas soft tissue trace metal concentrations can be compared on a wide geographical scale within a similar time period and in a historical context for the same location (Rainbow 1995). Mussels of the genus *Mytilus* have been employed extensively in Mussels Watch Programs to establish current trace metal levels in coastal environments, e.g., in the USA, UK, Australia, Hong Kong, and France (Cantillo 1998; Rainbow et al. 2000; Blackmore and Wang 2003). Mussels Watch Programs are considered efficient tools to study environmental trace metal levels, and they have been adopted in many countries (Hellou and Law 2003).

Several studies have examined the distribution of trace metals in mussels in coastal and estuarine waters, including bays, estuaries, and large semi-enclosed areas (O'Leary and Breen 1998; Regoli et al. 1998; Szefer et al. 2002; Conti and Cecchetti 2003; Rainbow et al. 2004; Saavedra et al. 2004; Mubiana et al. 2005), but few data review metal concentrations on a large, European scale. The present study was designed to determine the relative bioavailability of trace metals (Fe, Zn, Mn, Cu, Pb) in coastal waters along the European continent using mussels of the genus *Mytilus* as biomonitoring organisms. Existing pollution levels (strictly pollutant bioavailability) were measured to provide a baseline against which future changes in such levels in the coastal environment can be assessed.

Materials and methods

The blue mussels *Mytilus* spp. were sampled between January 2002 and December 2004 at 17 sampling sites located in the coastal waters around the European continent; namely, the Bay of Biscay (three sites); the English Channel (two sites); the Bay of Faxaflói (Iceland; two sites); the North Sea (three sites); the Baltic Sea (four sites); the Mediterranean Sea (one site); the Black Sea (two sites; Fig. 1). In tidal areas, bivalves were collected by hand from rocky or sandy bottoms near the low water level in the intertidal zone during emersion. In tideless basins, mussels were dredged from research vessels or collected by scuba divers in the sublittoral zone at depths of 1 to 38 m (Table 1). Immediately after sampling, the bivalves were held under wet, cool conditions while being transported to the laboratory. Here, they were kept in water taken in situ at temperatures and salinities that corresponded to the actual environmental situation (Table 1) for 24 h to purge gut contents.

In most cases, only animals of a limited shell length (20–30 mm) were selected since size can sometimes affect the trace metal concentrations in bivalve soft tissues (Wallace et al. 2003). The soft tissues were dissected from 30 to 55 mussels from each site with polypropylene or Teflon instruments, and then air-dried individually at 55°C to a constant weight in preweighed polythene vials that had been washed with 1M HNO_3 to obtain the individual soft tissue dry weight (DW). Dry samples were subsequently grouped into four to six pools of five to thirteen individuals each and homogenized in a standard ceramic mortar. The powdered tissue was then digested in a mixture of nitric/perchloric acids (14N/22N, Suprapur, Merck®) at 150°C for 24 h in closed beakers. The samples were then uncovered and dried by evaporation in acid digestion fumehood. The dry residues were redissolved in 5 ml 0.3 N nitric acid (Merck®) and stored at 4°C until analysis. The concentrations of trace metals Fe, Zn, Mn, Cu, and Pb were determined by flame atomic absorption spectrophotometry (F-AAS Shimadzu 6501 in background correction mode for all metals except Cu). The quality of the results was controlled by the simultaneous analysis of biological standard reference material (TORT-1 Lobster Hepatopancreas, National Research Council, Canada) according to the same procedures. Recovery and precision ranged from 77.1% to 134.3% and from 1.5% to 10.7%, respectively.

Because of the convincing presence of seasonal effects on metal concentrations in the tissues, the measured metal concentrations were adjusted separately for each metal using seasonal variation coefficients (SVC). The seasonal variation coefficient was calculated based on literature data as the ratio of metal concentration in mussels collected in a particular area in a given season and metal concentration in mussels from the same area in autumn, which was selected as the reference season

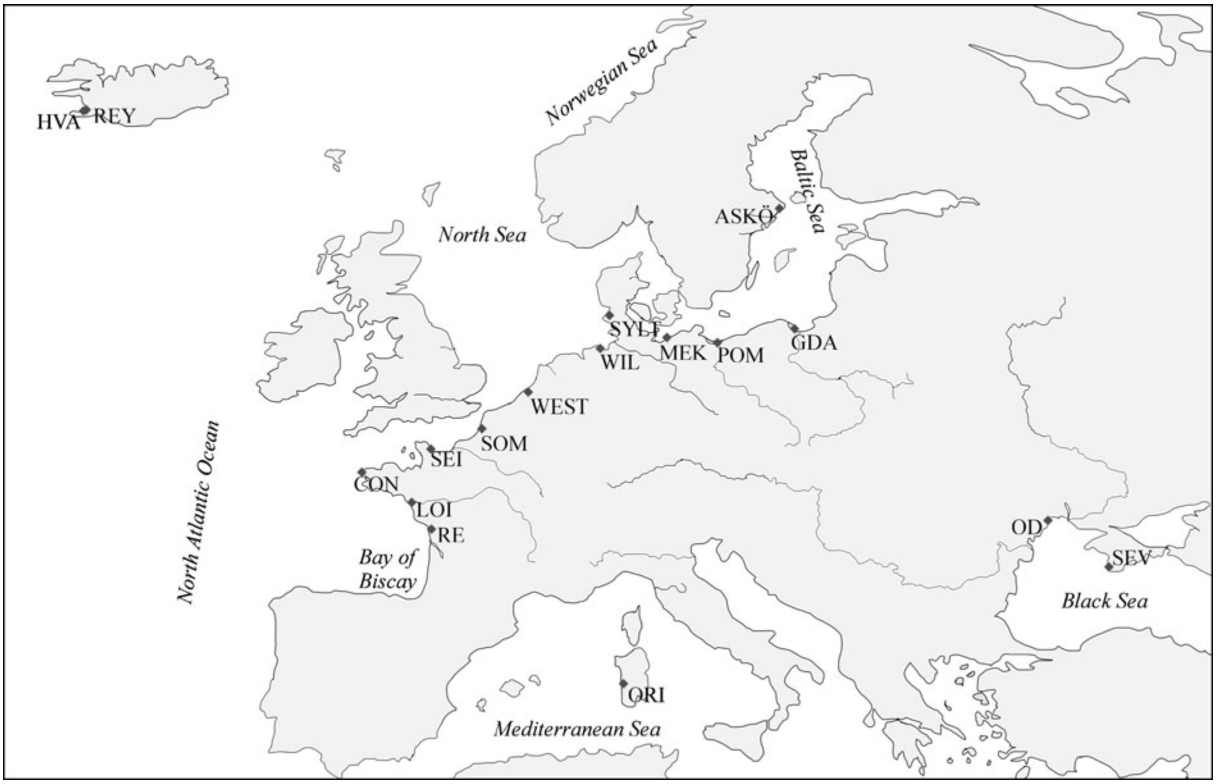

Fig. 1 Location of sampling sites in European coastal waters (⧫). For site codes, see Table 1

as most samples were taken between September and November (Table 2). Metal accumulation patterns in mussels can also vary across different biogeographical zones (Blackmore and Wang 2003; Szefer et al. 2006). Three SVCs were, therefore, calculated for different European coastal water basins, according to their climatic characteristics: (1) the Mediterranean Sea (Angulo 1996); (2) the North Sea (Mubiana et al. 2005), and (3) the Baltic Sea (Rainbow et al. 2004). Accumulated metal concentrations in mussels from the Black Sea were adjusted using the same SVC as for the Mediterranean sites, while those in mussels from Iceland were adjusted using the SVC for the North Sea sites.

Differences in mean concentrations of trace metals in mussel soft tissues among sampling sites were determined with one-way ANOVA tests followed by analyses of normality (Kolmogorov–Smirnov and a test of goodness-of-fit) and homogeneity of variances as prerequisites to the parametric approach. The sampling sites were ranked in ascending order of accumulated metal concentrations, and *R* Spearman correlation analysis was performed to describe relationships between ranks. Correlation analysis, factor, and PCA analyses were conducted using the PC professional software STATISTICA® for Windows (StatSoft 1997). The level of significance was $p < 0.05$ (Sokal and Rohlf 1995).

All specimens analyzed for soft tissue metal concentrations belong to the genus *Mytilus*, which is distributed widely in the northern hemisphere, including on both sides of the Atlantic and Pacific oceans. To date, more than ten species of mussel have been identified, and the current systematic status of the bivalves along the European coast refers to three genetic groups of *Mytilus edulis*, namely the Atlantic mussel *Mytilus edulis edulis*, *Mytilus edulis trossulus*, and the Mediterranean mussel *Mytilus edulis galloprovincialis* (Hummel et al. 2001). Although a number of studies have

Table 1 Details of sampling campaigns (region, site, site code, date, depth, and location in the littoral zone, ambient temperature, and salinity) and collected mussels (taxonomic identification, mean size ± standard deviation, number of pools, and number of individuals in each pool (in brackets))

Region	Sampling						Mussels		
	Site	Site code	Date	Depth (m)	Temperature (°C)	Salinity (PSU)	Species	Size (mm)	No pools
Bay of Biscay	Il de Ré	RE	09.2003	Intertidal	19.5	23.5	Hybridization *Mytilus e. edulis/ galloprovincialis*	27.29 ± 1.17	5 (10)
	Loire	LOI	10.2004	Intertidal	16.0	27.0	*Mytilus e. edulis*	37.26 ± 5.75	5 (10)
	Le Conquet	CON	10.2004	Intertidal	14.6	35.1	*Mytilus e. edulis*	25.59 ± 3.93	5 (8–10)
English Channel	Seine	SEI	10.2004	Intertidal	19.5	13.2	*Mytilus e. edulis*	26.86 ± 1.79	5 (10)
	Somme	SOM	10.2004	Intertidal	17.3	30.0	*Mytilus e. edulis*	33.17 ± 2.99	4 (7–9)
North Sea	Westerschelde	WEST	04.2003	Intertidal	9.0	24.5	*Mytilus e. edulis*	27.53 ± 2.35	6 (5–10)
Bay of Faxaflói	Hvassahraun	HVA	08.2004	Intertidal	9.0	35.0	*Mytilus e. edulis*	24.67 ± 3.54	5 (10)
	Reykjavik	REY	08.2004	Intertidal	10.0	32.0	*Mytilus e. edulis*	24.88 ± 2.64	5 (10)
Southeastern North Sea	Wilhelmshaven	WIL	12.2004	Intertidal			*Mytilus e. edulis*	16.68 ± 3.35	5 (9–13)
	Sylt	SYLT	12.2004	Intertidal	30.0	5.3	*Mytilus e. edulis*	26.80 ± 2.38	5 (10)
Southwestern Baltic Sea	Mecklenburg Bight	MEK	10.2003	1	16.7	14.0	hybridization *Mytilus e. edulis/ trossulus*	26.68 ± 1.49	5 (10)
Southern Baltic Sea	Pomeranian Bay	POM	06.2003	5			*Mytilus e. trossulus*	24.18 ± 1.41	5 (10)
	Bay of Gdansk	GDA	01.2002	38	2.3	6.9	*Mytilus e. trossulus*	28.93 ± 2.14	3 (10)
Northern Baltic Sea	Askö	ASKÖ	05.2003	1	3.8	6.0	*Mytilus e. trossulus*	19.36 ± 1.28	5 (10)
Northwestern Black Sea	Odessa	OD	07.2004	1			*Mytilus e. galloprovincialis*	25.38 ± 2.55	5 (10)
	Sevastopol	SEV	07.2004	1			*Mytilus e. galloprovincialis*	26.54 ± 2.31	5 (10)
Mediterranean Sea	Oristano	ORI	04.2003	6	15.3	37.0	*Mytilus e. galloprovincialis*	44.74 ± 4.97	5 (10)

Table 2 Soft-tissue accumulated concentrations of Fe, Zn, Mn, Cu, and Pb ($\mu g \cdot g^{-1}$ DW) in mussels in different seasons at three locations in European coastal waters and corresponding seasonal variation coefficients (SVC)

Sampling site	Date	Season	Fe		Zn		Mn		Cu		Pb		Reference
			Concentration	SVC	Concentration	SVC	Concentration	SVC	Concentration	SVC	Concentration	SVC	
Westerschelde North Sea	2000	Spring	572	1.40	235	0.54	63.0	1.31	27.4	1.81	16.8	2.05	Mubiana et al. (2005)
	2000	Summer	490	1.20	539	1.24	56.0	1.17	21.3	1.41	12.5	1.52	
	1999	Autumn	409	1.00	436	1.00	48.0	1.00	15.1	1.00	8.2	1.00	
	1999	Winter	273	0.67	566	1.30	45.1	0.94	23.7	1.57	10.3	1.26	
The Gulf of Gdansk, southern Baltic	2001	Spring	2558	1.33	100	1.16	44.4	0.39	9.9	0.80	9.5	0.59	Rainbow et al. (2004)
	2000	Summer	1010	0.52	170	1.98	70.2	0.61	12.6	1.01	13.1	0.81	
	2000	Autumn	1925	1.00	86	1.00	115.1	1.00	12.4	1.00	16.3	1.00	
	2001	Winter	1878	0.98	121	1.41	48.3	0.42	10.8	0.87	10.2	062	
Geneoa, Mediterranean Sea	1974	Spring	1210	1.86	260	1.21	69.8	1.55	12.9	1.12	13.3	2.33	Angulo (1996)
	1974	Summer	373	0.57	127	0.59	30.2	0.67	7.7	0.67	7.4	1.30	
	1974	Autumn	650	1.00	214	1.00	45.1	1.00	11.5	1.00	5.7	1.00	
	1974	Winter	777	1.20	252	1.18	52.9	1.17	13.2	1.15	15.7	2.75	

been undertaken in recent decades to clarify their distribution range, species identification of the genus *Mytilus* in some water basins in Europe still remains debatable. This is primarily because of the broad ecological plasticity of mussels, which are able to adapt to different environmental conditions (Presa et al. 2002). Additionally, intentional human activities such as commercial trade for aquaculture and consumption and unintentional activities such as shipping and the pollution of coastal areas have recently been shown to cause a northeastward shift in the distribution of mussels along the European coast (Hummel et al. 2001). The location of interspecific hybridization zones in overlapping regions has likely shifted too. Since the outer appearance of shells is of low diagnostic value, genetic characters are used to discriminate between the three groups. In this present study, the mussel species were identified with the latest genetic results (based on the concurrent use of allozymes and mitochondrial and nuclear DNA) that define the current distribution of the genus *Mytilus* in European waters (BIOCOMBE 2006). Specimens from Askö, the Gulf of Gdansk, and the Pomeranian Bay were identified as *Mytilus e. trossulus*. Mecklenburg Bight lies in the hybridization zone of *Mytilus e. trossulus* and *Mytilus e. edulis*. Mussels from Iceland, the North Sea, the English Channel, Le Conquet, and Loire belong to the *Mytilus e. edulis* group. The Il de Ré site is a region of hybridization between *Mytilus e. edulis* and *Mytilus e. galloprovincialis* groups, while animals from Oristano and the Black Sea were identified as *Mytilus e. galloprovincialis* (Table 1).

Results

Because of the presence of a seasonal effect in the dataset (one-way ANOVA, $p < 0.001$ for all tests, Table 3) the relevant concentration that permits making geographical comparisons is the season-adjusted concentration, which is the metal concentration of mussel soft tissue adjusted to the autumn period (see Table 2).

Accumulated soft tissue metal concentrations in *Mytilus* spp. showed significant spatial variations among sampling sites along the European coast (Table 4).

Table 3 Results of one-way ANOVA for testing the significance of seasonal differences in metal concentrations on raw data

Metal	Degree of freedom	*F* ratio	Remarks
Fe	16	102.87	$P < 0.001$
Zn	16	67.36	$P < 0.001$
Mn	16	318.70	$P < 0.001$
Cu	16	49.57	$P < 0.001$
Pb	16	37.52	$P < 0.001$

Rank order produced a single ranking of sites for each metal (1, 2, 3, etc.). The 17 sites are ranked in ascending order of accumulated concentrations (Table 5). *R* Spearman correlation coefficients of ranks demonstrated significant relationships between soft tissue mussel concentrations of Fe, Mn, and Pb in European coastal waters (Table 6).

The high correlation in ranking of Fe, Mn, and Pb demonstrates a similar degree of spatial variation in the pollution and bioavailability of these metals throughout Europe, whereas the low correlation with and between Cu and Zn highlights the strong inter-elemental differences in bioavailability and/or degree of pollution of these two trace metals.

The highest concentrations of Fe, Mn, and Pb were noted in mussels from the Southern Baltic, Wilhelmshaven, and some French sites with levels ranging from 1,141 to 3,433 $\mu g \cdot g^{-1}$ DW for Fe, 31.2 to 63.7 $\mu g \cdot g^{-1}$ DW for Mn, and 3.22 to 5.55 $\mu g \cdot g^{-1}$DW for Pb (Figs. 2, 3). Ten- to 20-fold lower concentrations occurred in bivalves from the Northern Baltic, Iceland, and the Mediterranean (95 to 189 μg Fe$\cdot g^{-1}$ DW, 1.8 to 2.49 μg Mn$\cdot g^{-1}$ DW, and 0.27 to 1.20 μg Pb$\cdot g^{-1}$ DW). The mussels from the North and Black Seas generally demonstrated intermediate levels of Fe, Mn, and Pb.

Table 4 Results of one-way ANOVA for testing the significance of geographical differences in metal concentrations

Metal	Degree of freedom	*F* ratio	Remarks
Fe	16	114.40	$P < 0.001$
Zn	16	177.21	$P < 0.001$
Mn	16	457.67	$P < 0.001$
Cu	16	60.61	$P < 0.001$
Pb	16	97.83	$P < 0.001$

Different patterns were noted for Cu and Zn than for Fe, Mn, and Pb; the most striking of which was that high concentrations of these two elements were noted in the Black Sea and in Icelandic waters in addition to those recorded at the Southern Baltic Sea and French sites (Fig. 4).

Thus, higher Cu concentrations were observed in mussels from the Southern Baltic, some French sites, Icelandic waters, and the Black Sea (all ranging within 12.5 to 15.8 μg Cu$\cdot g^{-1}$ DW; Fig. 4). The lowest concentrations were still found in the Mediterranean Sea and also at some North Sea sites at concentrations of 5.2 to 6.3 μg Cu$\cdot g^{-1}$ DW. The differences between the highest and lowest values for Cu in mussels from different regions were an order of magnitude less than those for Fe, Mn, and Pb.

The pattern for Zn was different yet, with the highest concentrations in mussels from the Black Sea and the Dutch Westerschelde (at concentrations ranging from 318 to 407 μg Zn$\cdot g^{-1}$ DW; Fig. 4). Intermediate and the lowest Zn concentrations (range of 46 to 55 μg Zn$\cdot g^{-1}$ DW) were noted in mussels from sites along the Atlantic coast of France).

In summary, the Gulf of Gdansk and Mecklenburg Bight in the Southern Baltic Sea demonstrated the lowest ranking for all metals (Table 5). The other Southern Baltic areas, the Black Sea and most French sites also exhibited lower levels of most metals. The Icelandic and the North Sea coastal waters were ranked higher for the majority of the metals analyzed, with the exception of Wilhelmshaven, where high metal concentrations were mostly recorded. Oristano, in the Mediterranean Sea, appeared to be the site with the lowest metal bioavailabilities of the coastal environments.

Discussion

The SVC was used in this study to resolve issues related to temporal changes in soft tissue metal concentrations in bivalves in a large-scale spatial

Table 5 Rank order of sites in ascending order of accumulated metal concentrations in *Mytilus* spp.

Site	Fe	Mn	Pb	Cu	Zn
Il de Ré	12	14	17	6	12
Loire	10	8	5	8	1
Le Conquet	8	6	11	7	14
Seine	13	10	15	14.5	5
Somme	6	5	12	4	2
Westerschelde	4	4	10	2	15
Hvassahraun	5	3	4	11	8
Reykjavik	2	2	2	13	10
Sylt	11	9	7	3	4
Wilhelmshaven	17	15	6	5	9
Mecklenburg Bight	15	13	14	16	13
Gulf of Gdansk	14	16	16	17	11
Pomeranian Bay	16	17	13	10	3
Askö	3	11	9	9	7
Sevastopol	7	7	8	12	16
Odessa	9	12	1	14.5	17
Oristano	1	1	3	1	6

comparative study of metal bioavailability in coastal waters. Seasonal patterns and the extent of annual variations of accumulated metals can, however, depend partially on biogeographical characteristics (hydrogeological and climatological constraints) and anthropogenic pressure in the area of interest (Riget et al. 1996). Concentration data from three European water basins (i.e., the Baltic Sea, the North Sea, the Mediterranean Sea) demonstrated apparent temporal discrepancies (Table 2). For the same element, inter-site differences were usually most pronounced in summer and autumn, which can result from differences in the physiological handling of metals taken up as was also suggested by Blackmore and Wang (2003) and Mubiana et al. (2005). In most European coastal waters, summer–autumn is the post-spawning period when mussels recover after energy-demanding spawning and gather energy reserves; thus, they are highly dependent on external environmental conditions. Biological (food availability) and hydrological conditions (such as salinity and temperature) of the ambient waters present an additional factor which can affect metal accumulation rates in mytilids in different European areas. This may provide an explanation for the different seasonal patterns of Cu, Pb, and Mn concentrations in the bivalves from the Baltic Sea (Rainbow et al. 2004), which is distinguished from the other seas by low salinity, elevated eutrophication, restricted water exchange with the oceanic system, and a relatively high metal load (Szefer 2002).

The rank order analysis showed that the Gulf of Gdansk and Mecklenburg Bight are sites of low ranking for all metals, which indicates the elevated bioavailabilities of metals in the Southern Baltic Sea. Oristano had the highest ranking for Fe, Mn, and Cu, and this demonstrates that in the Central Northern Mediterranean Sea, the bioavailability of the elements to mussels is relatively low. Based on correlation coefficients from the *R* Spearman tests on rank order ($p < 0.05$) and the ratios of minimal to maximal recorded metal

Table 6 Spearman (nonparametric) rank order correlation for accumulated metal concentrations in *Mytilus* spp.

	Fe		Zn		Mn		Cu	
	Correlation	*p* Value	Correlation	*p* Value	Correlation	*p* Value	Correlation	*p* Value
Zn	−0.07	0.79						
Mn	0.86	< 0.001	0.03	0.90				
Cu	0.32	0.21	0.29	0.26	0.38	0.13		
Pb	0.52	0.03	−0.02	0.94	0.53	0.03	0.17	0.52

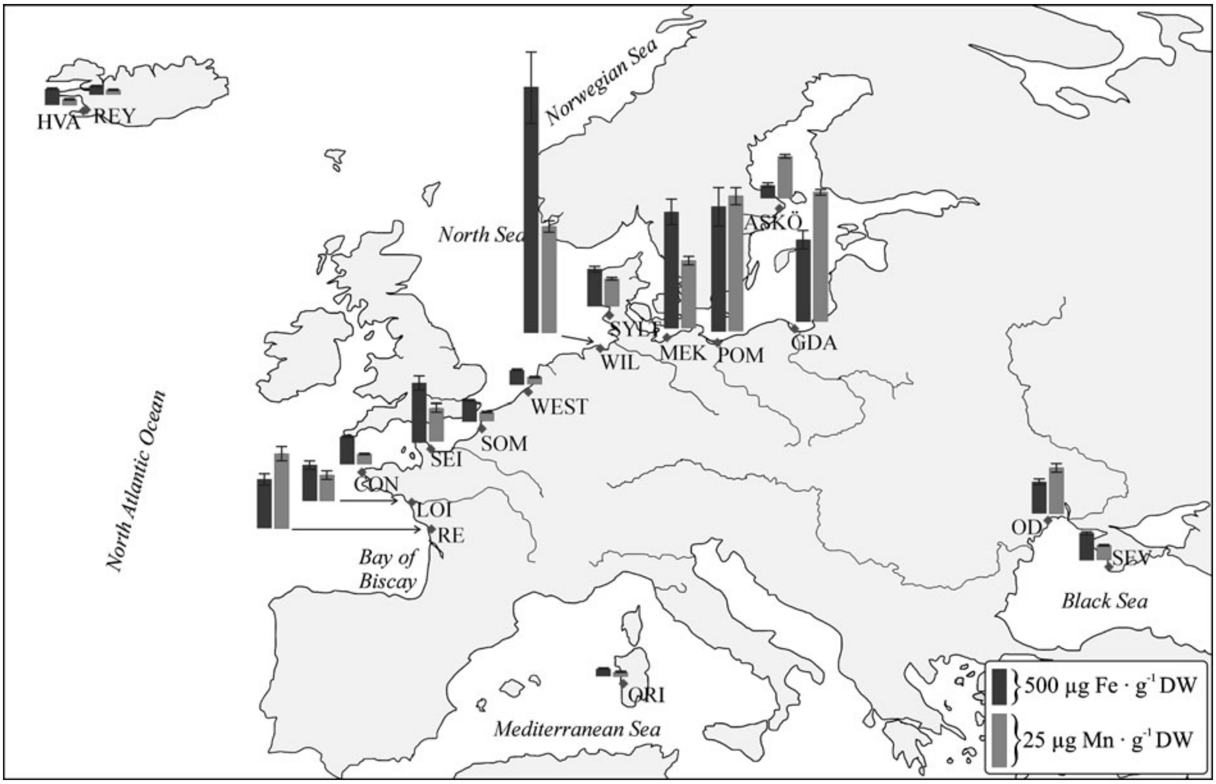

Fig. 2 Season-adjusted concentrations of Fe and Mn in the soft tissues of mussels in European coastal waters. Data are presented as mean and standard deviation ($n = 3 \div 6$)

concentrations, two groups of elements are identified: (1) Fe, Mn, and Pb with high geographic variation (the ratio ranged from 32.6 to 36.3) and (2) Cu and Zn with a relatively low max/min ratio (3.0 and 8.8, respectively; Table 6). The differentiation in the two groups of metals might be related to the partial internal regulation of Cu and Zn body concentrations by mussels and the high affinity of Cu and Zn for organic matter, whereby extremes are buffered, and, on the other hand, the direct reflection by Fe, Mn, and Pb of elevated environmental metal concentrations related to human activities (industry, agriculture, atmospheric fallout).

The highest concentration of Fe was observed in soft tissues of *Mytilus e. edulis* in Wilhelmshaven in the North Sea, which can be attributed to metal loads from the mining, iron, and steel industries. Germany is a world leader in the production of steel, which also involves processing manganese ore. Iron- and manganese-rich industrial effluents from mining sites and ironworks located in the Ruhre field, Salzgitter, and Hamburg enter the coastal waters of the Helgoland Bight mainly with the waters from the Ems, Wesser, and Elba rivers. Despite an open connection with the ocean, local eddy currents reduce the diluting effect and increase metal availability near the shore. Millward et al. (1998) reported remarkably increased concentrations of particulate-bound Fe (10,000 $\mu g \cdot g^{-1}$ DW) and Mn (700 $\mu g \cdot g^{-1}$ DW) in the coastal zone of Helgoland Bight. Elevated concentrations of Fe and Mn were also recorded in Mecklenburg Bight and the two Polish sites in the Southern Baltic Sea, namely the Pomeranian Bay and the Gulf of Gdansk. Szefer et al. (1995) also reported high concentrations of Fe and Mn in surficial sediments from the Southern Baltic Sea. The Gulf of Gdansk and the Pomeranian Bay are supplied by the Vistula and Oder rivers, respectively, the largest and second largest rivers in Poland, which drain

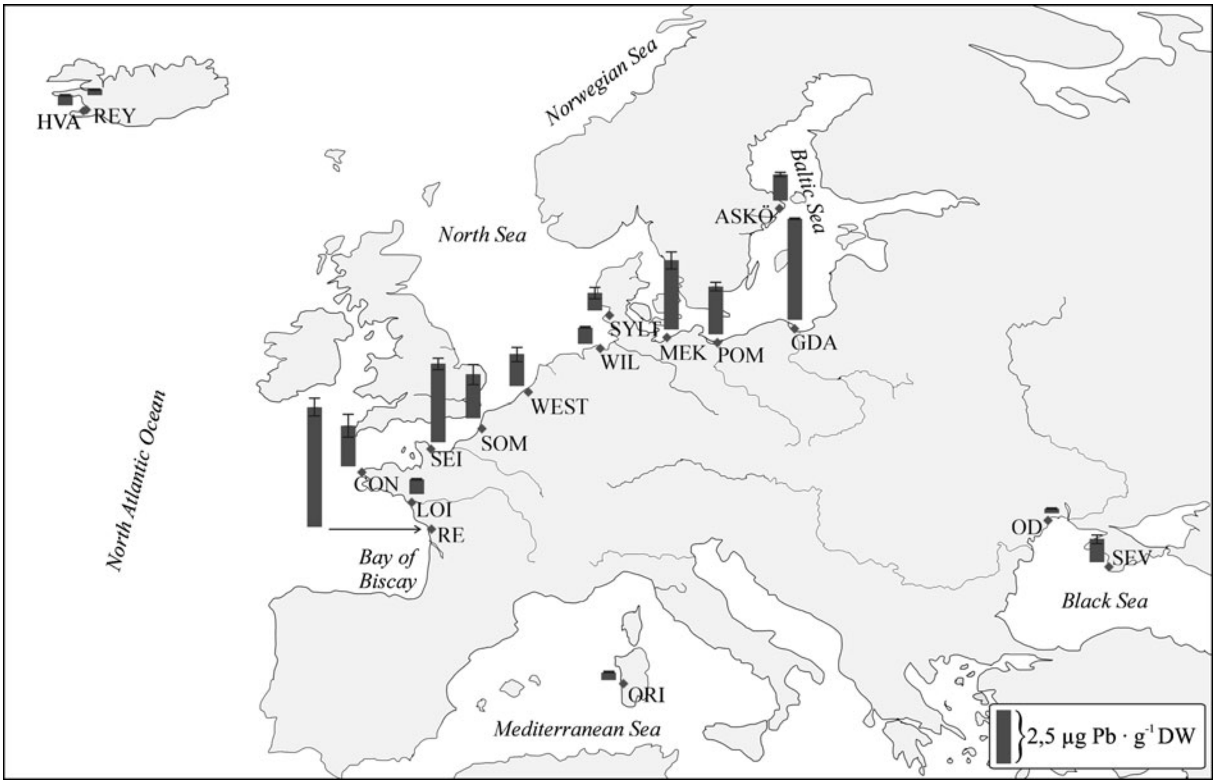

Fig. 3 Season-adjusted concentrations of Pb in the soft tissues of mussels in European coastal waters. Data are presented as mean and standard deviation ($n = 3 \div 6$)

the heavily industrialized heartland of Silesia (Szefer et al. 2002). The rivers and their tributaries flow through highly industrialized areas with manufacturing (iron and steel and electrochemical and chemical petroleum industries) and mining (coal, building stone, and petroleum) industries. Agricultural also contributes nutrients and eroded soil material to the riverine waters. The Gdansk region has been categorized an ecologically endangered area in Poland and a pollution "hotspot" in the Baltic. Both bays are highly dynamic and the sediments are enriched in trace metals with average concentrations that are 5-fold higher than background levels (Szefer et al. 2002). Low tissue concentrations of Fe and Mn were noted in Askö on the eastern coast of Sweden, which underlines its exceptional ecological quality due to low contamination levels and relatively pristine habitats. Askö was added to the list of areas designated as Baltic Sea Protected Areas by HELCOM in 1996 and included in the Ramsar (The Ramsar Convention on Wetlands) list of wetlands of international importance in 2004.

The lowest accumulated concentrations of all analyzed metals were recorded in *Mytilus e. galloprovincialis* from the Gulf of Oristano. This striking observation contrasts with the potential input of metallic contaminants from local food and engineering plants and several art workshops. What is more, Fe, Zn, Pb, Cu, Sb, and Ag ores are actively exploited in the coastal zone in Sardinia. One explanation of the low metal bioavailability in Oristano coastal waters is the northern currents which flow around the eastern coast of Sardinia and further to the Tyrrhenian Sea transporting metal pollutants away from their sources. Indeed, Regoli (1998) found extremely high concentrations of all metals in the gills and digestive glands of mussels collected in the Tyrrhenian Sea, which provide additional support for this hypothesis.

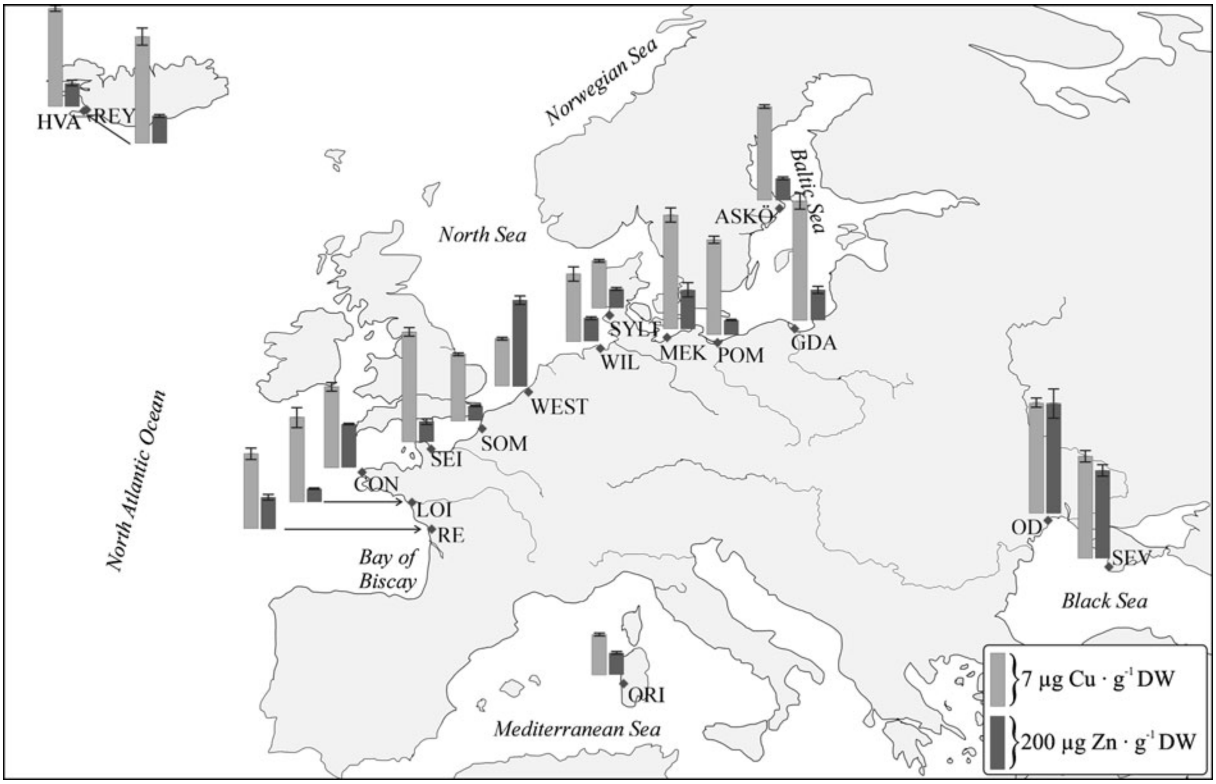

Fig. 4 Season-adjusted concentration of Cu and Zn in the soft tissues of mussels in European coastal waters. Data are presented as mean and standard deviation ($n = 3 \div 6$)

The oligotrophic character of the Mediterranean Sea, as is demonstrated by low nutrient levels and limited phytoplankton blooms, can also contribute to decreased metal bioavailability in this area by reducing suspended organic matter.

The highest concentration of the nonessential trace metal Pb in mussels from the coastal waters of Il de Ré, on the Atlantic coast of France, might be related to the motor-car and aircraft industries in La Rochelle. Radenac et al. (1997) linked elevated Pb levels in the mussels to the natural background metal concentrations in the surrounding area. Increased levels of accumulated Pb were also observed in the Gulf of Gdansk which receives considerable metal inputs from the Vistula River. Falandysz et al. (2000) estimated the annual load of Pb transported by the Vistula River as 196 tonnes, which is as much as 44% of the total annual metal discharge of Polish rivers (Helios Rybicka 1996). According to HELCOM (2006), the highest contribution of anthropogenic Pb emission (17%) to the Baltic is from Poland. High concentrations of Pb in sediments from the southern Baltic Sea were noted by Szefer et al. (1995). Relatively low concentrations of Pb in mussel soft tissues were recorded in Oristano and Odessa, which was presumably due to light automobile traffic on local roads as this is considered an important source of this metal in the coastal waters (Szefer et al. 2006). A similar range of accumulated Pb in *Mytilus e. galloprovincialis* from Sardinia coastal waters (Olbia) was reported by Storelli et al. (2000).

The soft-tissue concentration of Zn was substantially higher in mussels collected at two sites in the Black Sea as compared to other European regions. Two Ukrainian harbors, Odessa and Sevastopol, were designated as “hotspots” (Birkun 2002) since they are exposed to the influence of metal loading from the Danube River.

This is the second longest river in Europe and serves as an important shipping route for eight countries from Germany to Romania. Its annual input of Zn averages 6,000 tonnes while the Dnieper, Don, Cuban, and Belay rivers together introduce 2,600 tonnes of Zn per year to the Black Sea. Highly developed local metallurgic industries, including shipbuilding and engineering plants, are also likely contributors to enhanced Zn emissions. Comparable high accumulations of Zn were noted in *Mytilus e. edulis* from Westerschelde, which reflects the metal input of the Scheldt River and industrial effluents from Antwerp Harbor upstream (Mubiana et al. 2005). Painting and dye industries are an additional source of metal loading in the estuary, which increases metal bioavailability to the adjacent biota. According to Baeyens (1998), the Belgium coast and the Scheldt estuary in particular, can be considered contaminated with elevated concentrations of dissolved and particulate-bound metal forms. The concentration of Zn in the soft tissue of mussels from Westerschelde was within the range measured by Mubiana et al. (2005). The lowest level of this metal was observed in plumes from the Loire and Somme rivers. Amiard et al. (1986) reported similarly low Zn concentrations in bivalves collected adjacent to the mouth of the Loire River (Bay of Bourgneuf), while Miramand et al. (2001) also detected decreased concentrations of this element in the cockle *Cerastoderma edule* in the Somme plume.

High Mn bioavailability in the Gulf of Gdansk was also highlighted by Rainbow et al. (2004), using barnacles and mussels as biomonitors. The lowest levels of Mn were observed in *Mytilus e. edulis* from the Bay of Faxaflói and *Mytilus e. galloprovincialis* from the Gulf of Oristano. Iceland is

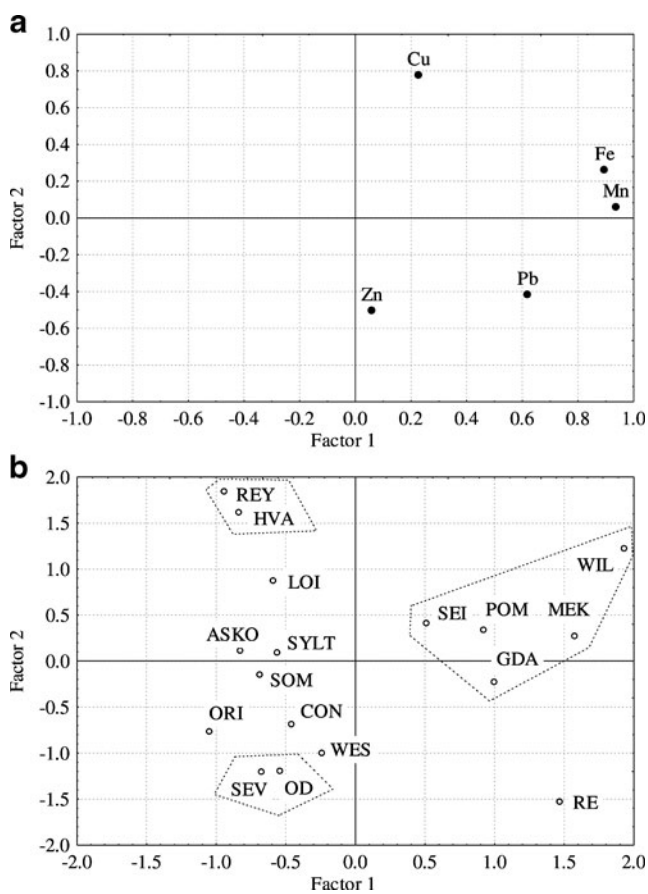

Fig. 5 Two-dimensional scatterplots of factor loading (metals) (**a**) and factor scores of individual data (sites) (**b**) in the space spanned by axes Factors 1 and 2

a volcanic island, and elevated levels of some trace metals, particularly Cd, can occur naturally due to geochemical weathering (Yngvadóttir et al. 2002), but limited heavy industry, the principal source of Fe, Mn, and Pb, does not cause additional pressure on near-shore waters.

Cu was the only element noted to have high accumulated concentrations in mussels from the Bay of Faxaflói. Presumably, this was from industrial inputs from light engineering factories such as telecommunication, electronics, computer, aircraft, and aerospace works as well as the production of medical equipment. Since more than half of the population of Iceland lives in Reykjavik, urban wastewater discharges likely contribute to the higher loading of Cu to the bay. It also cannot be excluded that considerable amounts of Cu enter the Icelandic coastal waters with the Hvita River. The relatively low geographical variation of Cu concentration in mussels (as shown by the ratio of maximal to minimal concentrations) suggests that the bivalves have the ability to partially regulate the accumulated body concentration of this metal as postulated by Phillips and Rainbow (1988) and Rainbow (1995). The low concentration of Cu in the soft tissue of *Mytilus e. edulis* collected from the Westerschelde was remarkable, and this is probably attributable to the high concentration of suspended organic matter with low carrying capacity for Cu in the water column (Gerringa et al. 1996).

Multivariate analysis indicated that 64.4% of total variance is explained by the first two factors denoted F1 and F2 with eigenvalues of 2.15 and 1.07, respectively. The first factor on the scatterplot of F1 against F2 is marked only by positive loading on Fe and Mn, while the second factor is marked by positive loading on Cu and negative loading on Zn and partly Pb (Fig. 5a). In PCA analysis, mussels collected in the southern Baltic Sea (GDA, POM, MEK) and Wilhelmshaven form a separate group (Fig. 5b) that confirms the high metal bioavailability in these regions. The grouping of the site in the vicinity of La Rochelle is substantiated by high values of F1 and low values of F2 (Fig. 5a), indicating contamination with Fe, Mn, Zn, and particularly with Pb. Copper accounts primarily for the identification of the Icelandic sites (REY, HVA), which supports the previous finding on elevated Cu levels in the Bay of Faxaflói presumably due to local industrial inputs. The group of Black Sea sites (SEV, OD), which is characterized by negative loading of F2, reflects high Zn bioavailability in the Ukrainian coastal waters adjacent to two large harbors and the Danube River mouth. The other sites are mid-distance between the four groups mentioned above.

Conclusions

Large-scale geographical variations in accumulated body concentrations of metals in mussels show apparent spatial differences in metal bioavailability along European coasts. The study highlighted coastal areas with elevated metal bioavailability that can have ecotoxicological effects on a local scale. Increased metal concentrations were observed in the North Sea (Fe and Mn in Wilhelmshaven) and in the Southern Baltic Sea (Fe, Mn, Pb, and Cu in Mecklenburg Bight, Pomeranian Bay, and Gulf of Gdansk). High accumulated tissue levels of Pb were also recorded in the Bay of Biscay (in the vicinity of La Rochelle) and of Zn in the Black Sea (near the Ukrainian harbors of Odessa and Sevastopol). The geography of metal bioavailability indicates that riverine discharge and industry are the main sources of metals to the coastal waters in Europe: Fe and Mn originate principally from metalliferous mining sites and ironworks, Pb is emitted by automobile and aircraft factories, while metallurgy and electronic factories contribute most to the loading of Zn and Cu, respectively. In contrast, low environmental metal levels were measured in the Mediterranean Sea and the northern part of Baltic (i.e., in regions of restricted anthropogenic activity and limited riverine input). Relatively low geographic variations of Cu and Zn suggest the ability of mussels to partially regulate accumulated body concentrations, making Cu and Zn somewhat independent of environmental concentrations.

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