# Invited paper

## Metal pollutants and radionuclides in the Baltic Sea – an overview

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**Abstract**

This overview presents in detail the state of knowledge of the abilities of various components of the Baltic Sea environment to accumulate trace elements and radionuclides. Particular components of the Baltic ecosystem (abiotic and biotic) are considered as potential monitors of pollutants. The use of seaweeds, e.g. *Fucus vesiculosus* or *Zostera marina* is recommended, also molluscs, e.g. *Mytilus edulis*, for biomonitoring surveys of metal pollutants and radionuclides in the Baltic Sea. However, several requirements need to be met if results are to be reliable. Since metal levels and radionuclide activities in the growing tips of *F. vesiculosus* reflect exclusively the levels of their dissolved species in the ambient seawater, this alga is very useful for monitoring dissolved species of metal pollutants and radioisotopes in the Baltic ecosystem. In contrast, *M. edulis*, a filter feeder is an appropriate tool for monitoring trace elements occurring in both chemical forms, i.e. dissolved and suspended species. Therefore, full information on the bioavailability and toxicity of heavy metals (depending on their chemical speciation) as pollutants of the Baltic Sea can be obtained if at least two biomonitoring organisms are applied simultaneously, e.g. *F. vesiculosus* and *M. edulis*. Moreover, the data matrix can be interpreted more accurately if not only trace element but also macroelement concentrations (Ca, Mg, Na, K) in these two representatives of Baltic phyto- and zoobenthos are taken into consideration; this point requires special attention.

Two coastal species of fish, i.e. *Zoarces viviparus* and *Perca fluviatilis*, are good biomonitors of metallic contaminants, so their use as sentinels is recommended. The budgets of chemical elements and the ecological status of the Baltic Sea are presented. Several 'black spots', e.g. large estuaries and seaport towns, heavily polluted by trace elements, are identified in the Baltic Sea and other enclosed seas such the Mediterranean and the Black Seas. Of these seas, the Baltic is the most heavily loaded with trace elements and organic pollutants. The overview identifies gaps in our environmental knowledge of the Baltic Sea, and sets out possible priorities, key areas or strategies for future research.

## 1. Introduction

A highly productive basin with intensive fisheries, the Baltic Sea is an interesting object of study. It is a brackish, non-tidal, relatively shallow and semi-enclosed sea. The drainage area is densely populated, heavily industrialised and intensively farmed. The relative ionic concentration of heavy metals is mostly greater in the low-saline Baltic waters than in the high-saline North Sea ones. Therefore, the Baltic Sea is particularly susceptible to pollution, which constitutes a threat to the people, plants and animals of all the states bordering it. This basin represents an integrated system, highly sensitive to chemical pollutants in its contact zones with the adjacent North Sea, the land and the atmosphere. Among the factors influencing the distribution of toxic metals and radionuclides are distance from the transition zone between the North Sea and the Baltic Sea, local hydrological and hydrographic conditions, the catchment area of the adjacent rivers and the extent of conservation measures in the adjacent areas. Since the 1980s, our knowledge of the biogeochemistry of the Baltic Sea has improved remarkably, resulting in much on-going work, the results of which have been published extensively. Unfortunately, the environmental quality of this sea has deteriorated significantly over the last 50 years.

This overview discusses questions concerning the distribution of trace elements and radionuclides in the water, biota and sediments, and the potential abilities of abiotic and biotic components to monitor the chemical pollutants. Trace element budgets for the Baltic ecosystem are also reported.

## 2. Monitors of Baltic Sea pollution

### Trace elements

#### Seawater

Analysis of seawater is the direct way of assessing the pollution status of the Baltic environment. It should be emphasised that levels of dissolved species of trace metals are usually very low, even near the detection limit of the method used, and therefore contamination of a sample during sampling and analysis is always possible (Phillips 1977b, Brügmann 1981, Bryan et al. 1985). The analytical procedures thus require a special approach, because it is necessary to eliminate effectively any analytical contamination. The greatest disadvantage of the spatial analysis of water samples is the large variation in metal levels caused by differences in season, time of day, the extent of freshwater influx, depth of sampling, the intermittent flow of industrial effluent as well as hydrological factors such as currents. In order to avoid these inconveniences, the use of time-integrated biomonitors is recommended (Phillips 1977b, 1980).

Trace elements in seawater are present in dissolved and particulate (organic and inorganic) forms as well as in colloidal or chelated forms. The latter are generally difficult to allot to either soluble or particulate phases. This assignation is therefore arbitrary, because it is based on whether a given element does or does not pass through a filter of certain pore diameter, making the comparison of data difficult or sometimes even impossible (Phillips 1977b). In estuarine and polluted areas trace elements may be removed from the soluble phase to the bottom sediments by precipitation, or to plankton by adsorption. An example of such a dissolved metal lost in this way is the deposition of trace elements with the amorphic Fe- and Mn oxyhydroxide phase at the hydrological front of the Gulf of Gdańsk (southern Baltic), where estuarine mixing of the brackish water with Vistula river water takes place (Szefer et al. 1995). Pempkowiak et al. (1998) reported higher levels of Hg in water from the plume of the Vistula.

Kremling & Streu (2000) recommended analysis of the dissolved species of trace metals (in addition to their determinations in organisms) in order to monitor metal pollution of Baltic waters. These authors observed a significant decrease in the Cd, Cu, Ni, Zn and Pb concentrations in the surface waters of the Baltic Proper during 1982–95. This negative temporal trend pattern is especially clearly marked for Cd and Pb because of their reduced use in industry and agriculture (Cd) and the restrictions on leaded petrol (Pb) in recent decades (Kremling & Streu 2000).

## Suspended matter

According to some authors (Jonsson et al. 1990, Lithner et al. 1996), the settling of suspended particles in the Baltic Sea, in contrast to surficial sediments (0–10 mm), could be used to monitor metal loads, e.g. Pb and As during short periods of time. For instance, since 1975 loads of Pb and As near the Swedish Baltic coast have decreased by 50–70% (Pb) following a reduction atmospheric fallout by more than 90% (As) as a result of remedial action at the Rönnskär smelters (Anon 1991, Rühling et al. 1992, Notter 1994, Lithner et al. 1996). However, the abundances of Pb and As in surficial sediments do not yet significantly reflect the changing loads (Anon 1991, Borg & Jonsson 1996, Lithner et al. 1996).

According to Sokolowski et al. (2001), in the deep zone of the Vistula estuary, Gulf of Gdańsk, desorption from detrital and/or resuspended particles by aerobic decomposition of organic material may be mainly responsible for the enrichment of particle-reactive metals such as Cu, Pb, Zn in the overlying bottom waters. It has been shown that the Vistula river plume is polluted with Cu, Pb and Zn (Sokolowski et al. 2001) and that the data on these particulate metals are in agreement with bottom sediment data, which suggests the anthropogenic origin of these trace elements in the coastal waters of the southern Baltic (Szefer & Skwarzec 1988b, Szefer 1990a, b, 1998, Szefer et al. 1995, 1996, 1998c).

**Bottom sediments**

In general, better agreement has been found between published Baltic data for sediments than for seawater, because in the case of the latter samples the risk of contamination is very much greater (Brügmann 1981). Moreover, the undisturbed deposited material, unlike water, may reflect the developmental history of a sea, including the anthropogenic impact, based on the analysis of dated cores (Clifton & Hamilton 1979, Brügmann 1981, Bryan et al. 1985). The sediments may therefore serve as a better and integrating monitor of long-term and medium-term metal loads (Brügmann 1981, Szefer & Skwarzec 1988b, Szefer 1998, Szefer et al. 1998c). Since in seawater and sediments trace elements occur in various chemical forms, it is important to know which of them are biologically available and capable of having any environmental lability (Phillips 1977b, 1980, Bryan et al. 1985).

In order to evaluate quantitatively metal pollution in the Baltic environment several approaches have been used, e.g. the concentration data obtained were standardised to Al as an element of terrigenic origin (Szefer et al. 1996), or normalised granulometrically to $< 80$ $\mu$m or $< 63$ $\mu$m surficial sediment fractions (Szefer et al. 1995, 1999a), and to $< 2$ $\mu$m (Szefer et al. 1998c). Moreover, analyses of Baltic sediments for concentrations of easily extractable metals were performed in order to discover their bioavailable forms (Szefer et al. 1995). Good agreement between the data from these approaches has been obtained, indicating the anthropogenic origin of Cu, Zn and especially of Pb, Cd and Ag in the coastal, estuarine and lagoonal areas of the southern Baltic. As a result of such approaches, it has been possible to eliminate, according to Phillips' recommendation, the variations in trace levels caused by variations in sediment character (sand, mud, silt) at different locations (Phillips 1977b). The levels of trace elements detected in bottom sediments are associated with the rates of elemental deposition and particle sedimentation, the size and nature of particles, as well as the concentration of organic matter or other major sediment phases, e.g. Fe- and Mn-oxyhydroxides (Phillips 1977b, Szefer et al. 1995, Szefer 1998). In the case of sediments enriched in organic matter, much attention has been focused on eliminating metals naturally bound to organic matter (reflected by an approximately linear increase in increased organic matter concentration) by considering levels of pollutants in sediments (falling above the natural pattern) only in relation to the percentage of total C present (Phillips 1977b). Trace elements of natural and anthropogenic origin (Co, Cu, Ni, Pb, Zn) in sediments from two Baltic basins (Bothnian Sea, Gotland Deep) were estimated by Mälkki (2001) for their affinity towards crustal material and to authigenic elements (Fe, S). The data show that the easily extractable 'labile' fraction is not necessarily an appropriate indicator of metal pollution, since longer stagnation periods result in intensive pyrite formation in the Gotland Deep, which effectively masks pollution trends.

According to Glasby et al. (2002) and Szefer et al. (2002d), sediment samples from the Szczecin Lagoon (Oder Haff) collected in October 1997 (after the exceptional flooding of the River Oder (Odra)) contained significantly higher levels of Mn, Cu, Pb, Co, Sb, Cd, Zn and possibly Fe than those collected in the same area in December 2000. These data suggest that the flood resulted in the enhanced transport of redox-sensitive and anthropogenic elements in the Oder and their subsequent redeposition in the sediments of the Szczecin Lagoon. The sediments of the Szczecin Lagoon also appear to be the most polluted with heavy metals within the Polish Exclusive Economic Zone (EEZ). Several studies have reported heavy-metal pollution in sediments of the Oder Haff (Szczecin Lagoon) (Leipe et al. 1989, 1995, Neumann et al. 1996, 1998, Pohl et al. 1998, Müller & Heininger 1999). However, it has been suggested that, during seasonal periods of salt-water inflow from the Baltic Sea, surficial sediments in the Oder Haff represent a potential source of heavy metals, whereas the sediments in the Achterwasser represent a potential sink for heavy metals (Neumann et al. 1998). However, in spite of this, the heavy metal contents of the Oder Haff displayed the highest average concentration of heavy-metal pollutants in eight boddens and haffs on the Baltic coast of Germany (Müller & Heininger 1999).

The increase in heavy metals (Ag, Cd, Pb, Cu, Zn) in the upper layers of Puck Bay sediments compared to their lower layers reflects the onset of industrialisation, and the resultant increase in heavy-metal pollution, in Poland (Szefer et al. 1998c). According to Pempkowiak et al. (1998) the increase in Hg concentrations in the upper segments of sediments deposited within the last 100–150 years is attributable to the anthropogenic input of this element and its increased scavenging to sediments with organic matter.

Two main factors control the distribution of the rare earth elements (REE) in sediments of the Polish EEZ: the input of Fe-organic colloids from rivers, and the presence of detrital material in the sediments. The highest values of REE are found in individual samples from the Pomeranian Bay, the inner shelf area and the Gulf of Gdańsk. It is suspected that these samples are coarser grained material, probably reworked by erosion. The REE values are so high (La 403 $\mu$g g$^{-1}$, Ce 835 $\mu$g g$^{-1}$) that they cannot be derived from a simple mixing of rock types (e.g. shale, granite, basalt etc.). These samples may be enriched in monazite and apatite, which are high in REE, especially monazite. The lag deposits in the deep parts of the Słupsk Furrow and the southern part of the Gotland Basin within the Polish EEZ are derived from the reworking of granitic material from Scandinavia during the ice ages. They also contain admixtures of Silurian and Tertiary material originating from the ice ages. It is suggested that the local presence of great concentrations of heavy minerals, including monazite, is responsible for the elevated levels of REE in bottom sediments from the Pomeranian Bay, especially from the Odra Bank (Szefer et al. 2002d).

**Ferromanganese nodules**

Ferromanganese nodules have been recommended as monitors of metal pollution in the Baltic Sea. Several authors (Djafari 1976, Suess & Djafari 1977) have reported that the outer layers of ferromanganese nodules from Kiel Bay, Baltic Sea, contain significantly greater levels of Zn, Pb, Cd and Cu, probably anthropogenic in origin. It is considered that the deep-water zone is the ultimate repository for many types of pollution in the Baltic Sea (Hakansson 1990). An interesting study in this respect, confirming the increased Zn contents in the outer layers of the nodules, has also been performed by Hlawatsch (1993) and Hlawatsch et al. (2002a, b) using laser ablation ICP MS, Scanning Electron Microscopy and synchrotron-based micro-X-ray radiation techniques (fluorescence: $\mu$SXRF and diffraction: $\mu$XRD). The usefulness of ferromanganese nodules to monitor heavy metals has been investigated by Ingri & Pontér (1986). It is suggested that natural enrichment processes, governed mainly by the redox conditions, are responsible for the high levels of Zn, Cu and Ni at Fe-Mn concretion surfaces. Furthermore, the presence of ferromanganese micronodules in surface sediments obscures the interpretation of trace element pollution. The use of ferro-manganese for monitoring metallic pollutants in the Baltic Sea is therefore highly questionable, although under controlled circumstances they could be used in this respect (Glasby et al. 1997, Szefer et al. 1998b). Non-anthropogenic elements such as La, Y and Yb are recommended as normalisation elements because their enrichment patterns are similar to those of Cu, Ni and Zn (Ingri & Pontér 1986). Terrigenic elements such as La, Y and Yb are recommended for use as normalising elements. According to Hlawatsch et al. (2002a, b) the enhanced anthropogenic emissions of Zn in the Baltic Sea area are clearly reflected in the ferromanganese nodules. Among the Zn, Cu, Cd, Ni, Co, Mn and Ba studied, trace metals like Cu and Cd were not enriched at all with distinct, early diagenetic Fe/Mn banding. Such a post-accretional transformation of the substrate geochemistry limits the utility of ferromanganese concretions for trace element monitoring, as already pointed out by Ingri & Pontér (1986).

**Phytobenthos**

Marine algae would be expected to be the most suitable indicators of dissolved metal species because, in contrast to animals, the dietary route for trace-element uptake is not involved (Phillips 1979, 1980, 1990, Bryan et al. 1985).

The evidence for the use of bladder wrack *Fucus vesiculosus* as an indicator is based on both laboratory and field observations. According to Bryan (1971) and Bryan et al. (1985) the fact that the lowest levels of metals usually occur in the growing tips of *F. vesiculosus* and that higher, more constant values are recorded in the older tissues can probably be explained by the relatively slow accumulation of trace elements, as well as the synthesis of more binding sites with age. This means that analyses of the younger parts of the alga – the tips – will provide more recent information, while analyses of the older fragments will allow us to know the relevant value integrated over several months (Bryan et al. 1985). Since, especially in estuaries, *F. vesiculosus* can be contaminated by fine particles of sediment adhering to its surface, a standardised analytical procedure should be used. Such a procedure for analysing this brown alga yielded good results for the biomonitoring of Ag, Cd, Cu, Cr, Hg, Ni, Pb, Zn (Bryan & Hummerstone 1973c, Morris & Bale 1975, Phillips 1977b, Melhuus et al. 1978, Phillips 1980, Bryan 1983, Bryan & Langston 1992). According to Phillips (1979) metal concentrations in the growing tips of *F. vesiculosus* from the Sound (Öresund) between Sweden and Denmark agree well with available data on the concentrations of dissolved trace elements in the waters of the Sound. The alga therefore appears to be responding exclusively to metals in the ambient water, as postulated by other authors (Bryan 1983, Bryan et al. 1985). On the basis of concentration data for trace elements in *F. vesiculosus* from the northern Baltic Sea and southern Bothnian Sea, Forsberg et al.

(1988) and Söderlund et al. (1988) recommended the brown seaweed as an excellent biomonitor of metal pollution. Elevated concentrations of metals, e.g. Zn, were found in samples taken close to densely populated and heavily industrialised areas (Söderlund et al. 1988). These bioindicative abilities have also been demonstrated by a significant or tendentious increase in concentrations of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb, V and Zn (but not Cd) in transplanted *F. vesiculosus* near the city of Stockholm, one of the most densely populated areas on the Baltic (Forsberg et al. 1988). The data for Cd were rather surprising, since the lower salinity and the expected higher Cd pollution in this area should be reflected by elevated levels in the *Fucus* biomass. This could be explained by competition from Mn and Zn, which probably suppressed Cd uptake (Bryan 1983, Forsberg et al. 1988). Surprising results have also been obtained for the monitored area of the Stockholm Archipelago using herbarium species collected in 1933 and 1984. The seaweeds from 1933 contained higher levels of Pb, V and Cu, probably a reflection of the mining activities there at that time (Forsberg et al. 1988).

Based on long-term studies, Ostapczuk et al. (1997) have demonstrated that, depending on the objective, *F. vesiculosus* from the North Sea and the Baltic Sea can be a useful tool for trend monitoring. In some cases, however, more information on the chemical form in which the element is present in algal tissue is necessary for proper interpretation of the data. However, consideration of the concentrations of macroelements such as Ca, Fe, K, Mg, Na, P and S in the biomatrices is recommended (Struck et al. 1997) in order to identify and separate independent ecosystem effects, e.g. salinity, temperature. Therefore, the concentrations of trace elements in *F. vesiculosus* do not necessary reflect their total quantities in the ambient water of the Baltic Sea (Kangas & Autio 1986).

Based on data of analyses of *F. vesiculosus* from Swedish and Finnish coasts (Kangas & Autio 1986, Forsberg et al. 1988, Söderlund et al. 1988), the use of *F. vesiculosus* is recommended for biomonitoring metallic pollutants in the Baltic Sea. However, the following precautions should be taken:

- samples should be cut from a fixed part of the *Fucus* thallus and should be free of epiphytes,
- parts of the plants of the same age should be used when comparing spatial distribution,
- samples should be collected at the same or similar time (within a few days) to avoid seasonal variations,
- depth, salinity and water temperature should not fluctuate too much,

- samples should be taken from sites with the same degree of wave-exposure.

Brown seaweed *Pilayella littoralis* from the Gulf of Gdańsk is, as compared to *M. edulis*, less able to regulate Pb uptake from its surroundings (water, sediment); hence it would appear that this Baltic seaweed has a great potential as a biomonitor of Pb in the Baltic environment (Szefer & Szefer 1991).

The green alga *Enteromorpha* sp. has been studied for trace-element contamination in marine ecosystems (Bojanowski 1972, Hägerhäll 1973, Stenner & Nickless 1974, Seeliger & Edwards 1977, Melhuus et al. 1978, Szefer & Skwarzec 1988a). The data indicate clearly that *Enteromorpha* sp. reflects variations in concentrations of As, Cd, Cu, Hg, Pb and Zn in the ambient seawater and can therefore be used as an effective biomonitor of these elements. Bearing in mind that *E. intestinalis* absorbed higher levels of trace elements, e.g. Co, Mn and Zn at lower salinity (Munda 1984), the advantages of this green alga over *F. vesiculosus* are that *E. intestinalis* often penetrates farther upstream, into regions of very low salinity. Moreover it may also reflect changes in ambient element concentrations more rapidly than *F. vesiculosus* (Bryan et al. 1985).

Trace metal concentrations were significantly elevated near the cities of Aalborg (Pb, Cu) and Struer (Cd) in the Limfjord, Denmark. The use of eelgrass as a monitoring organism is highly recommended (Brix et al. 1983). According to Szefer & Szefer (1991), *Zostera marina* may be appropriate for biomonitoring Pb pollution in the Gulf of Gdańsk, Poland.

The results strongly suggest (Brix & Lyngby 1982, 1983, Lyngby & Brix 1982, Brix et al. 1983) that *Z. marina* can be used to monitor trace metal contamination in coastal areas. The properties required of this plant are as follows:

- the concentration of some trace metals in the above- and below-ground parts of *Z. marina* should be used as a measure of the bioavailable fraction of trace metals in the ambient and interstitial water (sediment) in this area,

- in order to obtain information on the dynamics of chemical elements in coastal Baltic waters, data on the distribution of the elements in the individual plants are needed,

- because of significant seasonal variations in trace elements in eelgrass *Z. marina*, parts of the same age should be taken from it at the sampling site at the same or similar time.

## Plankton

According to Phillips (1980) phytoplankton organisms have rarely been used as appropriate biomonitors for the comparison of elemental pollutant abundance at more than one sampling site. The main reasons for this limitation seem to be the difficulty in obtaining a reasonable size of sample free of foreign matter or other organisms, e.g. zooplankton, and the knowledge of the extent to which a particular phytoplankton species is able to accumulate pollutants. The use of these organisms as biomonitors affords little time-integration, although in the case of single species studied, the concentrations of pollutants detected will be a complex composite of the quantities of the available trace elements in the water column as well as the species succession in the phytoplankton community (Phillips 1980). In spite of the questionable abilities of phytoplankton as biomonitors, the uptake of pollutants from the seawater column by these organisms plays an important role in the transferring these trace elements along the successive levels of the trophic chain to its higher organisms (Szefer 1991). Phillips (1977a, 1978) found that the variations in trace element levels in the soft tissue of the blue mussel *M. edulis* from the east and west coasts of Sweden were attributable to the different species composition of the phytoplankton populations inhabiting the two areas. The low-saline waters of the Baltic Sea were dominated by well adapted blue-green algae, while the Danish Straits hosted other phytoplankton species preferring a higher salinity.

The use of zooplankton in the same way as phytoplankton as a biomonitoring tool to detect spatial and temporal trends in the Baltic Sea is not recommended. According to several authors (Martin & Knauer 1973, Boström et al. 1974, Szefer et al. 1985, Diaz & Fernandez-Puelles 1988, Pohl 1992, Weber et al. 1992, Brügmann & Hennings 1994) this is because:

- metal concentrations in different species may vary over a rather broad range,
- some zooplankton species may accumulate the metals in various quantities, depending on their life stage and age,
- some metals seem to be well regulated by zooplankton,
- non-biogenic material adheres strongly to phytoplankton biomass or becomes incorporated into the zooplankton (e.g. rust particles, paint chips, clay particles) and may contaminate zooplankton samples,
- it is not possible to separate phyto- and zooplankton using nets of different mesh sizes, i.e. a higher percentage of phytoplankton in the samples may result in higher metal contents.

Nonetheless, zooplankton has already been used frequently to study metal contamination in the marine environment (Phillips 1980). It may at least be a valuable tool for identifying pollution hot spots (Balogh 1988).

## Zoobenthos

### *Molluscs*

Various species of molluscs are recommended as biomonitors of trace -element pollution in marine ecosystems (Phillips 1980, 1990, Bryan 1985, Bryan et al. 1985, Fowler 1990). It is noteworthy that all requirements are entirely satisfied for the use of these bottom organisms in surveys of metallic pollution. Molluscs are easily identified, widely distributed, common, accessible, sensitive to locally dependent variations in trace elements, available at all times of the year, relatively stationary and sufficiently tolerant of low salinity. The latter property is very important for estuarine and near-estuarine areas, very typical of the Baltic basin. According to Broman et al. (1991) the bioavailability of Cd to the soft tissue of this mussel is dependent on the salinity of the adjacent water. Tissue Cd levels in *Mytilus edulis* inhabiting the southern coastal waters of Sweden were up to one order of magnitude lower as compared to those detected in the northern area, where the salinity is low. The study of metals in *M. edulis* along the Swedish coasts disclosed a tendency towards increasing concentrations of Cd and Zn at some locations in the open coastal archipelagos of Stockholm and Åland as compared to the other coastal parts of the Baltic. This increase in concentration at locations not directly affected by industrial metal discharge was argued to be a result of the influence of low salinity on the forms metals will adopt and on their bioavailability (Phillips 1976a, b, 1977a, 1978, 1980, Struck et al. 1997). According to Möller et al. (1983), *M. edulis* from near the Kiel sewage outlet, southwestern Baltic, accumulated greater amounts of Ag, Au, Cd, Cr, Hg and Ni, reflecting elevated levels of these elements in the ambient water. Therefore, in any biomonitoring survey, especially of areas with a steep salinity gradient, special attention should be paid to the more complex interpretation of the data matrix, considering not only trace elements but also the macroelement concentrations in *M. edulis* and *F. vesiculosus* from the North Sea and the Baltic Sea (Struck et al. 1997). According to Falandysz (1994) the spatial differences in Hg levels in *M. edulis* and other zoobenthic species from the Gulf of Gdańsk are attributable to both pollution from local nonpoint land-based sources and higher accumulation in sedimentation zones.

The best way to conduct a monitoring survey using mussels is undoubtedly to collect a large size-range of mussels at each sampling location, and to analyse the mussels individually. This approach, however, requires a very great number of analyses, and will therefore probably not be realistic in more extensive surveys. Since Cu is regulated in *M. edulis*, *Mytilus* cannot be used as an indicator organism for Cu pollution (Brix & Lyngby 1985). This is in agreement with data obtained by Julshamn (1981), who reported that *M. edulis* from Sorfjorden, Norway, was useless as biomonitor for Cd, Cu and Zn, but was acceptable for Pb and probably Hg. Theede et al. (1979) found in numerous places on the German coast of the Baltic that the Cd content of the edible common mussel *M. edulis* is higher than in individuals from the North Sea coast. The highest Cd content is found in mussels from the innermost part of the Kiel Fjord. Specimens from the outer parts of the Kiel and Flensburg Fjord contain less Cd.

Because of their world-wide distribution and potential as indicators, several species of *Mytilus* as filter feeders have become the subject of various monitoring programmes of the 'Mussel Watch' type (Goldberg et al. 1978, 1983, Koide et al. 1982, Cossa 1988, 1989, Fabris et al. 1994). In *Mytilus*, metals are probably adsorbed both from solution and from ingested phytoplankton and other suspended particles (George 1980). The soft tissue of this mussel appears to be a good bioindicator for Cd, Cr, Hg, Ni, Pb and Zn but not for Cu (Boyden 1975, 1977, Bryan 1980, Bryan et al. 1985). *M. edulis* is unreliable as a biomonitor for Ag and As (Bryan & Hummerstone 1977, Langston 1984). According to Roesijadi et al. (1984) trace elements such as Ag, Cu, Hg and Zn, in contrast to Cd, can be successfully biomonitored using the soft tissue of *M. edulis*.

Szefer et al. (2002c) pointed out that factor analysis (FA) of metal concentrations in the soft tissue and byssus of *M. edulis trossulus* is useful for distinguishing three sub-areas of the southern Baltic inhabited by this mussel. A plot of the samples based on their factor scores showed a clustering of the soft tissue and byssi samples into two main groups, each corresponding to a geographically distinct zone (Szefer 2002, Szefer et al. 2002c). Such differentiation between these two groups could be explained by the differences in environmental parameters in the geographical sectors, e.g. the food supply for *Mytilus* specific to each area, metal runoff, the geochemical composition of the adjacent sediments as substrata for mussels, etc. The Pomeranian Bay, like the Słupsk Bank region, is located in the open part of the study area, in contrast to the Gulf of Gdańsk, which is partly isolated from the open sea by the Hel Peninsula. It is assumed that water mixing occurs in these neighbouring areas; water coming from the Pomeranian Bay mixes with the water mass in the Słupsk Bank region especially during seasonal storms (Szefer 2002).

Szefer et al. (1998a) processed statistically the concentration data for *Mytilus* sp. from the Baltic Sea and other geographical areas. The concentration data for the soft tissue and byssus obtained by examining c. 10 000 specimens of Mytilidae collected in the Baltic Sea and other geographical areas were processed by FA. After removing values corresponding to badly contaminated samples from highly industrialised areas (Saganoseki, Japan and Öxelosund, Sweden) it was possible to distinguish the Baltic population of *M. edulis* from other clusters based on byssus data (Szefer 2002). The grouping of object samples corresponding to the soft tissue overlapped other clusters and was thus inappropriate for identifying the Baltic population. Manganese identified Baltic specimens of *Mytilus*, among others, and can be used as specific determinant in this respect (Szefer 2002).

The common cockle *Cerastoderma edule* usually inhabits the relatively saline waters of estuaries, and, as a filter feeder, is most likely to be able to absorb trace metals from solution and particulate matter (Bryan et al. 1985). Several authors (Boyden 1975, Bryan & Hummerstone 1977, Bryan 1980) have postulated that *C. edule* seems to be an appropriate accumulator of Ag, Cd, Cu and Zn, and a particularly good one for Ni. On the basis of inter-comparison studies of *C. edule* and *F. vesiculosus*, it should be emphasised that Ni levels in this bivalve can underestimate the degree of dissolved Ni pollution at larger concentrations (Bryan et al. 1985). However, changes in Cd levels in this bivalve are approximately proportional to those in *F. vesiculosus*, and levels of Ag increase more rapidly than those in the brown alga in response to pollution of the surrounding area. Szefer et al. (1999b) reported significant spatial and seasonal variations in metal levels in *C. glaucum* from Thau Lagoon, Mediterranean Sea. *Cerastoderma* is therefore not particularly useful as an indicator, although it does reflect environmental pollution with Ag, As, Cd and Ni. It also responds to high levels of Cu and Zn but, probably as a result of regulation, underestimates moderate levels of pollution. Particulate contamination of *Cerastoderma* specimens often causes difficulties in their use as indicators of Cr and Pb (Bryan et al. 1985). Szefer & Wołowicz (1993) statistically processed the tissue concentration data of Cd, Cu, Fe, Mn, Ni, Zn for *Cerastoderma glaucum* from four geographical regions, i.e. the Gulf of Gdańsk (Baltic Sea), Marennes-Oleron Bay, Arcachon Bay (French Atlantic coast) and the Embiez Islands (Mediterranean Sea). It is mainly the Mn and Fe concentrations in these molluscs that are responsible for the differentiation between the populations from Marennes-Oleron Bay and Arcachon Bay. Zn, Cd and partly Ni distinguished the Gulf of Gdańsk cluster from the others (Szefer 2002). Bearing in mind that *Cerastoderma* seems to be an appropriate biomonitor for Cd, Cu, Zn and particularly Ni, such for higher a distribution pattern implies that anthropogenic sources may be responsible levels of Cd and Zn in *C. glaucum* inhabiting the coastal and industrialised zone of the Gulf of Gdańsk (Szefer & Wołowicz 1993).

A deposit-feeding clam, *Macoma balthica*, generally lies within a few cm of the sediment surface and occurs in the majority of estuaries. It appears to act as a biomonitor for Ag, Cd, Cr, Hg, Ni and especially for Zn, although there is some doubt about its use for Cu (Bryan & Hummerstone 1977, Bryan 1980).

Sokolowski et al. (2002) have reported concentration data of Ag, As, Cd, Cu Mn, Pb, Se, V and Zn in soft tissues of the Baltic clam *Macoma balthica* from the Gulf of Gdańsk, southern Baltic Sea. The results obtained were considered in two main phenotypes, i.e. with respect to external shell colour and according to the shell shape of *M. balthica*. Specimens with an elongate posterior end and a notable flexure, referred to as 'irregular' clams, exhibited generally higher concentrations of all elements in their tissues than 'regular' bivalves, i.e. those, whose shells have a rounded posterior end. This paper provides the first information on the potential link between shell deformation and tissue metal levels within a whole population of clams living in the same area.

The concentration levels of trace metals in *M. edulis* from the Limfjord, Denmark, were significantly greater in the soft tissue than in the shells. The results suggest that the shells of this species are of no practical use in the monitoring of the metals investigated (Brix & Lyngby 1985).

According to Szefer (1991) and Szefer & Szefer (1991), *M. edulis* has great potential as a biomonitor of Cd contamination in the southern Baltic ecosystem. Several authors (Rainbow et al. 2000, Szefer et al. 2002c) concluded that *M. trossulus* is suitable biomonitor to employ in programmes designed to trace changes in metal pollution in the Gulf of Gdańsk, Baltic Sea. It has been reported that in comparison to soft tissue, the byssus of *M. trossulus* is a more effective bioaccumulator of trace elements than Cd in the southern Baltic and other regions (Szefer et al. 1998a, 2002c).

The significant relationship between concentrations of Ag, As, Cd and Pb in the perwinkle (*Littorina littorea*) and bladder wrack (*Fucus vesiculosus*) suggests that, directly or indirectly, concentrations in this gastropod reflect those of the ambient water (Bryan et al. 1985). Moreover, a significant correlation exists between these two benthic species for Cu, Fe, Hg and Zn but slope constants were relatively low, perhaps as a result of regulation by the perwinkle. It seems to be a suitable indicator of pollution by dissolved Ag, Cd and Pb and perhaps As and Hg (Bryan et al. 1985). According to Bauer et al. (1997) malformations in male perwinkles are closely related to tributyltin (TBT) contamination: the reduction of male mamilliform penial glands showed strong correlations to TBT concentrations in soft tissues. The intersex index (ISI) – the average value of the intersex stages in *L. littorea* – is recommended as the most sensitive biological parameter for the assessment of TBT contamination in the Baltic Sea and the North Sea, i.e. in those regions where the dogwelk *Nucella lapillus*, the most sensitive species in European surveys, is absent. As results from numerous literature data, *N. lapillus* has been mainly used for TBT biomonitoring in all European programmes (Bryan & Langston 1992, Minchin et al. 1995, 1996, 1997, Evans et al. 1996, 2000, Huet et al. 1996, Skarphédinsdóttir et al. 1996, Morgan et al. 1998, Følsvik et al. 1999, Miller et al. 1999, Santos et al. 2000). This species is well suited to biomonitoring surveys, especially in areas of high contamination like the Kiel/Schilksee sampling site and the Baltic Sea, where alternative monitoring species such as *N. lapillus* are absent (Bauer et al. 1997). TBT levels in the German coastal zone of the Baltic Sea and the North Sea, being much higher than in France, Irish and English waters (Bailey & Davies 1989, Gibbs et al. 1990, 1991, Oehlmann et al. 1993, Minchin et al. 1997), are too high for the survival of more sensitive species, e.g. *N. lapillus* and *Ocenebra erinacea*. The perwinkle therefore represents a monitoring system for many parts of northern Europe and northern America and can be complementary to other recommended TBT-effect monitoring species (Bauer et al. 1997). The field data have been supported by the results of laboratory experiments. Considering the results of the field TBT survey, the reduction of the number of mamilliform penial glands seems to be a good marker for assessing TBT contamination in the Baltic Sea, not only in juveniles but also in adult specimens, because the penis is shed and rebuilt every year (Bauer et al. 1997). According to Albalat et al. (2002) *Mytilus edulis* from the Gulf of Gdańsk exhibited the highest levels of butyltins in conjuction with elevated TBT/DBT ratios, which suggests recent inputs of TBT to this area.

*Polychaete worms*

Several authors considered the ability of the ragworm, *Nereis diversicolor*, to accumulate trace-elements in marine systems (Bryan & Hummerstone 1973a, b, 1977, Bryan 1980, Phillips 1980, Bryan et al. 1985, Bernds et al. 1998). This polychaete is considered an indicator of Ag, Cd, Cu, Cr, Hg, Ni and Pb, and inter-comparison studies between heavy metal concentrations in its body and those of surface sediments have shown that in most cases they are significantly related (Bryan et al. 1985). Such relationships have been reported for Ag, As, Co, Cr, Cu, Hg and Pb (Bryan et al. 1980, 1985, Langston 1980, Luoma & Bryan 1982, Bryan & Langston 1992). The most reliable correlations between concentrations of Ag, As and

Hg in *N. diversicolor* and associated sediments were obtained in the case of sediment levels of these elements normalised to organic matter (Langston 1986, Bryan & Langston 1992). In spite of the close dependence of trace elements in the polychaete on those in the sediments, in some cases tissue concentrations may be additionally influenced by those of the surrounding water. An example of such a situation is Ag and Cd in *N. diversicolor* from the Severn Estuary and Bristol Channel, because their levels in the body were unrelated to those of the associated sediments but were very closely related to those of *F. vesiculosus* which had taken up dissolved species of Ag and Cd from the overlying water (Bryan et al. 1985, Bryan & Langston 1992).

*Crustaceans*

Since many species of crustaceans are highly mobile, they are of no use as indicators in estuaries. The significance of trace metal concentrations in various marine crustacens has been presented by Rainbow (1988, 1995), Rainbow & Phillips (1993) and Rainbow et al. (1990). According to Bryan (1968), decapod crustaceans regulate Zn and Cu against environmental changes, but Cd is accumulated in polluted environments by the shrimp *Crangon crangon* (Dethlefsen 1977). Szefer & Kusak (2002) found significant relationships between concentrations of Ag, Mn and Fe in *Crangon* sp. and those in associated sediments ($< 63$ $\mu$m) from the Gulf of Gdańsk, southern Baltic. Watson et al. (1995) considered the use of barnacle shells as biomonitoring material. The Barnacle (*Balanus improvisus*) from the Gulf of Gdańsk appeared to be a suitable biomonitor of trace elements (Rainbow et al. 2000, Szefer et al. 2002e). This crustacean has shown significant geographical and temporal differences in the local bioavailabilities of trace elements as reflected in the levels of accumulated metallic pollutants (Rainbow et al. 2000). It is concluded (Rainbow et al. 2000, Szefer et al. 2002e) that *B. improvisus* is a promising biomonitor of trace metal pollutants of the Gulf of Gdańsk, Baltic Sea.

Amphipods are appropriate crustaceans for biomonitoring surveys in marine environments (Rainbow & Moore 1990). Comparative studies of decapods, amphipods and barnacles with respect to their abilities to accumulate of Cd, Cu and Zn have been performed by Rainbow & White (1989).

According to Rainbow et al. (1998) the talitrid amphipod *Talitrus saltator* from the strandline of sites around the Gulf of Gdańsk, southern Baltic, indicated spatial trends for trace metal concentrations. This distribution pattern resulted from significant geographical differences in the local bioavailabilities of trace metals, which are variably dependent on outflows from the river Vistula (Cd, Fe, Mn, Zn) or from local sources around the Gulf (Cu, Pb). Therefore, *T. saltator* is an effective biomonitor because it lives on the shore and does not require expensive equipment for its collection (Rainbow et al. 1998).

*Starfish*

The starfish *Asterias rubens* is recommended as a valuable sentinel organism for monitoring metallic pollutants in the sea (Bjerregaard 1988, Phillips 1990, Temara et al. 1996, 1997). This species efficiently bioconcentrates metals such as Cd, Hg, Mn, Pb, Se and Sr (Binyon 1978, Bjerregaard 1988, den Besten et al. 1990, Sorensen & Bjerregaard 1991, Rouleau et al. 1993, Hansen & Bjerregaard 1995, Temara et al. 1996, 1998). Distinct differences and strong relationships in trace element concentrations were found for starfish from the Baltic Sea, and these are attributed to the composition of the sediments being an important substrate for their prey, such as mussels and snails (Brügmann & Lange 1988).

## Fish

The pollution of seafood with heavy metals is still a problem from both the hygienic and ecotoxicological points of view. In several countries, especially those participating in International Council for the Exploration of the Sea (ICES) monitoring programmes, much attention has been focused on determining chemical pollutants such as heavy metals in fish. There are two main approaches: the evaluation of toxic metals in edible tissues (muscle, liver) in relation to human health, and the assaying of metal pollutants in estuarine and coastal areas using fish tissues as biomonitors (Phillips 1980, Cossa et al. 1992, Szefer 2002c).

Fish accumulate trace elements from food and ambient water (Phillips 1977b). Fish muscle is usually monitored in the interests of public health, but such a survey is inconvenient since the metals are either regulated or the tissue concentrations are very low, often being below the detection limit of the method used (Phillips 1977b, 1980, Bryan 1984). However, in contrast to other trace metals, fish muscle contains a high percentage of methyl Hg; hence, muscle is widely used in monitoring this pollutant (Olsson 1976, Schladot et al. 1997). Bryan et al. (1985) recommended the flounder *Platichthys flesus* as potentially the best indicator since it is often distributed throughout the estuary and is able to enter fresh water. According to Moriarty et al. (1984) the miller's thumb *Cottus gobio* from the river Ecclesbourne, Derbyshire, can be used for monitoring heavy metal pollution. The results suggested that it is better to use mass (content) than concentration data for pollutant in a tissue, e.g. Cd in liver and Pb in gills. The use of the northern pike *Esox lucius* to monitor Hg pollution in Swedish waters has been also reported by Johnels et al. (1968) and Olsson (1976). There are significant differences between sexes and ages in respect to Hg concentrations in this species from Lake Marmen, Sweden (Olsson 1976). HELCOM recommends two coastal species of fish as sentinels in the monitoring of contaminants: the marine viviparous blenny (*Zoarces viviparus*) and the limnic perch (*P. fluviatilis*) (Guidelines for Coastal Fish Monitoring and Effects of Harmful Substances and Eutrophication – HELCOM Environment Committee). These two species were selected on certain criteria: stationary behaviour, well-known biology, common and easy to catch, mode of reproduction allowing analytical studies and a size large enough for chemical analyses on individuals. Most Baltic hot spots are situated in areas where the low salinity favours a fish community dominated by limnic species. In such limnic-like areas, perch is the priority species. According to Schladot et al. (1997), eel-pout (*Zoarces viviparus*), a sedentary fish, can be used for monitoring Hg, Me-Hg and As in the shallow waters of the Baltic ecosystem. The muscle and hepatic concentrations of these elements were enhanced relative to the ambient water and were strongly dependent on the sampling site. It is reported by UBA (1996) that the liver is more useful than muscle for monitoring of Pb and Tl where levels of these metals are high.

The temporal changes in hepatic Pb levels in cod liver caught southeast of Gotland and from the Kattegat indicated a negative trend of c. 5% $y^{-1}$ during 1981–94 (Harms 1996). The decline in atmospheric Pb detected over a period of 10 years (from 1979 to 1989) caused by the reduced use of leaded petrol in western European countries should be reflected in decreasing Pb levels in surface water and biota of the Baltic Sea. The temporal negative trend for cod registered under the Swedish monitoring programme seems to support this argumentation.

In the case of hepatic Cd, significant upward temporal trends for herring from the Bothnian Sea, Northern and Southern Baltic Proper were recorded. The continuous decrease in salinity in the Baltic Proper and Bothnian Bay may therefore have created favourable conditions for the gradual enhancement of bioavailable Cd species that are strongly adsorbed by plankton. If trace elements associated with plankton are consumed by fish, their bioaccumulation via uptake into the alimentary tract will be a predominant route of exposure (Harms 1996). In contrast to cod, herring feeds mainly on plankton; hence the respective variations in Cd levels in water and thus plankton could be reflected in herring but not in cod. Therefore, the spatial variations of Cd concentrations at higher levels in the marine trophic web are probably attributable to spatial differences in water salinity. The positive temporal trends for Cd in herring from the Bothnian Sea and the Baltic Proper are associated with the long-term decreasing trends in salinity in these sub-areas. Owing to the lack of major inflows of highly saline North Sea waters during the last two decades until 1993, a continuous decrease of salinity in almost all sub-areas since c. the mid-1970s was noted (Harms 1996). The long-term changes in Cd in the food (plankton) of the herring could be an explanation for the increase in its content in herring over the corresponding time.

The muscle concentrations of Hg in herring from Ängskärsklubb, Bothnian Sea, exhibited a significant decreasing temporal trend of c. $-7.8\%$ $y^{-1}$ during 1981–94 (Harms 1996). Since this sampling location might be influenced by the local input of pollutants from the Gävleån River, the observed trend could not be accepted as representative for the whole of the Bothnian Sea. Although upward trends of c. $4\%$ $y^{-1}$ were identified for muscle Hg levels in herring from the Baltic Proper and the Kattegat, no systematic temporal changes have been detected for those in herring collected in the Northern Baltic Proper (Harms 1996). It appears that temporal changes in muscle Hg in Baltic fish are associated with its emission sources and chemical speciation. The formation and fate of organic Hg species in marine ecosystems are dependent upon several parameters, such as surface water temperature, nutrient supply, and on the abundance of phytoplankton and its species composition (Harms 1996).

The temporal trends for Cu and Zn levels in cod and herring from the Kattegat and in flounder from the Sound as well as for herring from other subareas such as the Baltic Proper and the Bothnian Sea are almost constant over the period (Harms 1996). Cu and Zn as essential trace elements, in contrast to toxic elements Cd, Hg and Pb, are presumably involved in various metabolic processes and play specific physiological roles; hence temporal trends for their concentrations in fish may be associated with natural factors including homeostasis rather than with anthropogenic impact (Harms 1996).

Szefer et al. (2002b) reported that hepatic Pb and Cd are responsible for distinguishing the perch samples caught in winter, while summer samples are identifiable by hepatic Cu, and especially Zn. The observed seasonal variations in selected metals in perch are caused by different metal bioavailability, depending on the ligands present in the biotopes and the chemical speciations between the dissolved and particulate phases (Andres et al. 2000). Moreover, fish metabolism may be dependent on the abiotic conditions, food supply and the stage of the cycle reproduction (Kock et al. 1996, Olsson et al. 1996, Andres et al. 2000).

Perttilä et al. (1982) have reported that Baltic cod liver exhibited spatial differences, which, with the exception of Pb, followed the spatial differences of metal concentrations (Hg, Cd, Cu and Zn) in seawater. These differences suggest that, in spite of the extensive migration of cod, it is a better indicator species for aquatic pollution than herring. Cod feeds mainly on herring and benthic animals, and thus harmful substances are accumulated at higher trophic levels in cod than in herring.

Sures et al. (2001) have demonstrated for the first time that automobile Pd catalyst emitted into the air is bioavailable to European eels (*Anguilla anguilla*). The use of catalytic converters for automobile exhaust purification is responsible for the emission of the platinum-group-metals, i.e. Pt, Pd and Rh.

Björklund (1989) has reported the presence of elevated levels of TBT in flounder, blue mussel and invertebrates from the Swedish coastal waters of the Baltic Sea.

Different species of fish from the Gulf of Gdańsk have been analysed for TBT in muscle, liver and eggs (Kannan & Falandysz 1997, Senthilkumar et al. 1999). According to Albalat et al. (2002), the flounder *Platichthys flesus* can be used as a sentinel species to biomonitor TBT inputs along the Polish coast of the Baltic Sea.

### Sea and shore birds

Located as they are at the top of marine food webs, sea and shore birds have great potential as monitors of metal pollutants, since these are biomagnified along trophic levels. General seabird ecology, the numbers and productivity of many populations, as well as the colonial breeding habits of many of these birds have several advantages suggesting their use for biomonitoring metallic pollutants. According to Furness and Camphuysen (1997) aquatic birds can sometimes be used to monitor fish stocks and fishery activities. The chronic effects of metal pollutants as well as the effects of acidification may have a number of consequences on reproduction, disease, immunosupression and behaviour in these birds (Scheuhammer 1987, 1991). Bearhop et al. (2000a) suggested that both the blood and feathers of the great skua (*Catharacta skua*) reflect Hg intake over the same time period although blood appeared to be a better biomonitor than feathers (Bearhop et al. 2000b). Thompson et al. (1990, 1998) have considered and recommended the use of seabirds to monitor Hg pollution in marine environments.

According to Wenzel et al. (1996), particularly older chicks ($\geq 6$ days old) of kittiwakes *Rissa tridactyla* from the German Bight were suitable biomonitors of Hg and Cd pollution around Helgoland Island because the influence of egg pollution on metal loads in chicks decreased with increasing chick age and dietary metal intake.

This assessment of long-term pollution has been possible owing to the analysis of Hg concentrations in the feathers of museum specimens. Extensive studies of feathers of Swedish birds since 1840 have indicated that the rise in Hg content in several bird species to its present values started in the 1940s. The supply of Hg compounds added to Swedish soils as seed dressings was absorbed in birds' tissues through the digestive system (Berg et al. 1966). In order to elucidate the feasibility of using feathers as a monitoring object, Appelquist et al. (1984) examined the influence of factors such as ultraviolet light, heating, freezing and weathering on the Hg concentration in the feathers of guillemots (*Uria aalge*) and black guillemots (*Cepphus grylle*) from the Northern Baltic as well as from Danish and Greenland waters. Furness & Camphuysen (1997) have reported that pelagic seabirds exhibit a greater increase in Hg pollution, especially seabirds feeding on mesopelagic prey, than most coastal specimens. This finding can be explained by the methylation patterns of Hg in low-oxygen, deeper water. According to Goede & de Bruin (1984) either several parts, or whole feathers of *Calidris canutus* and *Limosa lapponica* reflect the level of Hg pollution and, with time, contamination may occur via the feather oils. The vane is suitable as a monitoring tissue for Zn, sampled just after moult while the shaft reflects the levels of As, Pb and Se deposited in the feather during formation. Elements like Zn should be sampled soon after moult. Although Se is accumulated in significant amounts in the kidney of Scandinavian *Calidris alpina*, after the species' departure from the marine to freshwater environments, levels decline rapidly (Goede et al. 1989).

## Marine mammals

Environmental pollutants can affect all the trophic levels of a food web, including its highest levels, i.e. marine mammals which are doubly injured, directly by pollution and indirectly by decreasing food stocks (Viale 1994). The harbour porpoise, though rarely noted in the Baltic Sea (Skóra et al. 1988, Skóra 1991), constitutes a final link in the Baltic food chain and is an interesting object for pollution studies because of its widespread distribution all over the world. The decline in numbers of live specimens off the Dutch coast since 1960 was related to poisoning, because very high levels of Hg and other organic toxicants were detected in dead animals (De Wolf 1983). Szefer et al. (2002f) have reported for a single harbour porpoise from the southern Baltic elevated levels of renal Hg, most probably anthropogenic in origin. It is well known that the synergic effects of chemical pollutants, including Hg, cause a viral epizootic, which damages some cetaceans and leads to collective pathology (Viale 1994). Therefore, biomonitoring of Hg in sea mammals including Baltic porpoises, is justified bearing in mind that the serious diseases are connected with high levels of toxic metals, especially Hg.

Levels of Hg, Cd, Pb and Se were determined in liver, kidney and muscle of grey seals (*Halichoerus grypus*) from the Baltic Sea and Sable Island (Nyman et al. 2002). According to the data obtained, only Cd levels showed significant spatial differences, being lower in the seals from the Baltic Sea than from Sable Island. No clear reduction in the metal load has been reported for the Baltic grey seal population since the 1970s (Nyman et al. 2002).

Based on FA data (Szefer et al. 2002g), geographical differentiation was considered between harbour porpoise populations from the Baltic Sea, coastal waters of Denmark and Greenland. Specimens inhabiting the southern Baltic were identified by hepatic and renal Fe and Cr while Cd, Mn, Zn and Cu made possible the identification of Greenland specimens. The Danish group confirmed the close association of samples corresponding to the Greenland and Baltic populations. Geographical variations in hepatic and renal metals were caused mainly by the specific feeding habits of the porpoises inhabiting the southern Baltic (principally fish) and Southwest Greenland (fish and squids).

## Radionuclides

### Plankton

Seasonal variations in radionuclide concentrations in the Gulf of Finland have been reported. Phytoplankton inhabiting the Loviisa, Gulf of Finland, was most abundant in $^{103}$Ru, $^{129m}$Te and $^{137}$Cs. Towards autumn, the levels of radionuclides decreased significantly (Ilus et al. 1987). According to Bojanowski et al. (1995), radiocaesium is not preferentially accumulated in either phyto- or zooplankton from the Pomeranian Bay, and hence plankton cannot be used successfully to biomonitor radiocaesium activity in the Baltic environment.

### Phytobenthos

According to several authors (Ilus et al. 1987, 1988, Neumann et al. 1991, Dahlgaard & Boelskifte 1992, Holm 1995) *F. vesiculosus* from the Baltic Sea can be used in monitoring programmes because of its ability to integrate and concentrate low concentrations of radionuclides. According to Dahlgaard & Boelskifte (1992), the dilution effect of radionuclide concentrations caused by growth of *Fucus* is a more significant parameter than the biological loss of radionuclides. Christensen & Strålberg (2000) reported that the contribution of $^{137}Cs$ from Sellafield discharges is now negligible and that the main source of the radionuclide found in *Fucus* along the Norwegian coast is the Chernobyl fallout transported to the sea by runoff from land into rivers entering the Baltic Sea.

According to several reports (Ilus et al. 1981, 1987, 1988, Carlson 1990) *F. vesiculosus* from the Finnish coast is a useful species for monitoring radioactive substances in the vicinity of nuclear power stations and also in surveys of the dispersion pattern and fate of radioactive fallout in marine environments. Dahlgaard & Boelskifte (1992) also reported that *Fucus* can be used successfully as a semi-quantitative indicator for radioactive contaminants. It is concluded that this brown alga is the most sensitive biomonitor of $^{60}Co$ and $^{65}Zn$ and can thus effectively identify the radionuclides originating from the Chernobyl fallout. This finding is in agreement with data reported by Neumann et al. (1991) that *F. vesiculosus* is a sensitive indicator for many radionuclides released into water. For example, it has been reported that as a result of the regular maintenance of the nuclear power plants at Ringhals (Swedish west coast) and Simpevarp (the Baltic Proper), activation products, i.e. $^{60}Co$ and $^{65}Zn$, can be determined in *F. vesiculosus* at long distances along the coast line from the discharge point (Neumann et al. 1991).

The levels of some radionuclides in Baltic algae corresponded closely to their sampling sites, which were affected by the deposition of the Chernobyl fallout (HELCOM 1995). For instance, maximum levels of Chernobyl-derived radiocaesium, $^{110m}Ag$ and $^{106}Ru$ in *F. vesiculosus* from Forsmark and Olkiluoto on the Bothnian Sea were recorded in 1986.

According to Holm (1995), the Chernobyl accident had no significant impact on $^{239,240}Pu$ concentrations in *F. vesiculosus* along the Swedish coast. This finding was strongly supported by the estimate average $^{238}Pu/^{239+240}Pu$ activity ratio for the above mentioned area of 0.032. This ratio is similar to typical worldwide fallout and deviates significantly from the values of 0.47 reported for the Chernobyl fallout over Sweden (Holm et al. 1989). Similar results are reported for seaweeds in the Gulf of Gdańsk by Skwarzec & Bojanowski (1992), suggesting that the contribution of Chernobyl-derived plutonium to Baltic plants was small.

## Zoobenthos

Mussels appear to be appropriate organisms for monitoring radioactive contaminants in the Baltic ecosystem. These organisms collected from the Bothnian Sea in 1986 and 1988 were characterised by higher levels of $^{137}Cs$

and $^{60}$Co than those from the most southerly subareas, e.g. the Bornholm Sea and especially the Kattegat. This means that mussels from the most southerly subareas were influenced by a relatively low Chernobyl-derived fallout (HELCOM 1995). The distribution pattern of $^{60}$Co in *Mytilus edulis* and *Macoma balthica* from Forsmark was similar to the temporal trends recorded in $^{137}$Cs levels in the mussels from the same subarea (HELCOM 1995). Most probably, it is $^{60}$Co emission from the nuclear power plants located at Forsmark that was mainly responsible for the total radioactivity of mussels after 1986–87.

### Fish

Fish muscle appears to be a good biomonitor of radionuclide contamination in the Baltic environment (Ilus et al. 1987, 1992, 1993, HELCOM 1995). Elevated levels of Chernobyl radiocaesium ($^{137}$Cs) in Baltic subareas such as the Bothnian Sea, the Gulf of Finland, the Åland and the Archipelago Seas were attributable to maximum concentrations of this radioisotope in fish in 1986 and 1987, which tended to decrease during the following years. Radiocaesium activity in fillets of fish from the most southerly subareas (Bornholm, Arkona and Belt Seas) exhibited an increasing trend after 1986, although their values were still relatively low. These southernmost subareas are apparently affected by the counter-clockwise movements of Baltic surface waters and their outflow to the North Sea via the Danish straits. In consequence, fish from this latter area, influenced by a relatively low direct Chernobyl fallout, were characterised by much lower levels of radiocaesium than those from other subareas (HELCOM 1995). In 1996–97 Rissanen & Ikäheimonen (2000) detected flesh levels of $^{137}$Cs in salmon (*Salmo salar*) from the River Tornionjoki (Torneälven) two orders of magnitude higher than those from the River Teno. The Tornionjoki salmon migrated to the river from the Gulf of Bothnia and contained from $< 0.2$ to 1.3 Bq $^{134}$Cs kg$^{-1}$ originating from the Chernobyl accident. Similar radiocaesium concentrations have been measured in pike (*Esox lucius*) in the Baltic Sea. It is concluded that salmon flesh reflects the concentrations of some radionuclides in the ambient waters.

Skwarzec et al. (2001) have reported a significant contribution of Chernobyl-derived $^{239,240}$Pu to its total concentrations in fish from the Gulf of Gdańsk collected in 1997.

### Marine mammals

The levels of $^{137}$Cs in the liver, kidney and muscle of harbour porpoises and seals from the Baltic Sea are similar to the hepatic and renal values recorded for these species from British waters (Berrow et al. 1998, Watson et al. 1999). It has been suggested that radiocaesium levels in Baltic mammals originate from the Chernobyl accident (Szefer et al. 2002a). According to Anderson et al. (1990) 66–78% of the $^{137}Cs$ load in grey seals from North Rona and from the Isle of May originated from Sellafield discharges, the remainder from the Chernobyl accident. The radiocaesium contamination in porpoises and seals from coastal British waters decreased with distance from the Sellafield processing plant, indicating that this plant was the major source of this radionuclide (Watson et al. 1999).

## Recommendations and future trends

It should be noted that there is a lack of information concerning heavy metal concentrations in Baltic crabs. An example of such studies are the concentration data reported for these crustaceans from Belgian, French and English coastal waters (Guns et al. 1999, Rainbow et al. 1999).

Because metal levels and radionuclide activities in growing tips of the alga *F. vesiculosus* reflect exclusively the levels of their dissolved species in the ambient seawater, *F. vesiculosus* is very useful for monitoring dissolved species of metallic pollutants and radioisotopes in the Baltic ecosystem. In contrast, *Mytilus edulis* as a filter feeder is an appropriate tool for monitoring trace elements occurring in both chemical forms, i.e. as dissolved and suspended species. Therefore, full information on the bioavailability and toxicity of heavy metals (depending on their chemical speciation) as pollutants of the Baltic Sea may be obtained from the simultaneous use of at least biomonitoring materials e.g. *F. vesiculosus* and *M. edulis*.

According to several authors (Rainbow & Phillips 1993, Rainbow 1995, Rainbow et al. 2000), whole specimens of barnacles can be used to effectively biomonitor metal pollutants in marine ecosystems. Watson et al. (1995) have concluded that barnacle shells cannot be considered an ideal biomonitoring material. These authors proposed potential criteria for the successful use of barnacle shell as biomonitors. The following requirements should be taken into account:

- use an internal standard which is digested with each batch of samples,
- use large numbers of specimens per sample,
- alternatively, sample barnacles of the same size, or sample a wide range of barnacle sizes from each site and compare the regression lines between metal content and shell weight, or use weight-adjusted metal concentrations,
- sample from each different site at the same time.

Seabirds can effectively reflect long-term changes in Hg pollution of epipelagic and mesopelagic marine waters, based on inter-specific dietary preferences. Measured trends in seabirds are in general accordance with model predictions for surface marine waters. Therefore, the use of seabirds as monitors of Hg pollution in the marine environments has been strongly recommended by Thompson et al. (1998).

Aarkrog (2000) in his millennium article wrote that 'Radioecology may briefly be described as the science which studies the interaction between radionuclides and the biogeosphere'. This definition is closely related to the concept of Dahlgaard & Boelskifte (1992), who recommended the study of biological factors such as biomass turnover rates as well as environmental effects on the accumulation of radionuclides and their biological loss in the use of bioindicators in environmental monitoring. This concept can also be applied to trace elements. The SENSI model is helpful in evaluating the ability of *Fucus* to monitor pollutants and contaminants by including ecological factors; this results in an increase in the correlation between expected and measured values. Moreover, the SENSI model can be successfully used to quantify an uncontrolled discharge and to estimate routinely the quality of discharge data (Dahlgaard & Boelskifte 1992).

According to Nielsen (1995, 2000), the most important future radiological changes in the Baltic Sea are expected to be the continuing decrease of $^{137}Cs$ concentrations due to the outflow of water through the Kattegat and to a smaller extent, the increase in $^{99}Tc$ concentrations caused by water inflow from the North Sea. Therefore, future monitoring programmes should follow these changes in order to obtain accurate information on radionuclide exchange between the Baltic Sea and North Sea.

Satellite monitoring of the ionosphere in order to monitor extreme situations caused by natural and man-made accidents is recommended (Boyarchuk 1998). Promising results have been obtained for the box modelling of the radiological consequences of releases of radionuclides into large marine environments such as the Arctic Ocean and the North Atlantic Ocean (Iosjpe & Strand 1998). Mathematical models of environmental radionuclide distribution and transport have been developed to assess the impact on man of potential and actual releases of radioactivity, both planned and accidental, from various nuclear sources (Thiessen et al. 1999).

As far as future radiological studies are concerned, the radiological impact of marine radionuclides is generally lower than that of radionuclides in the terrestrial environment (Aarkrog 1998). Therefore it appears that scientific studies on terrestrial radioactivity are needed. However, radionuclides in the marine environment can be used as effective tracers for biochemical processes (sedimentation processes) and for sea currents, and Aarkrog

(1998) recommends that environmental scientists should concentrate on radiological studies of both the marine and terrestrial environments and consider the global ecosystem in its entirety. Trends in radioecology at the turn of the millennium have been presented in detail by Aarkrog (2000).

Papers dealing with bioconcentration factors of radionuclides for marine fauna and flora as well as the transfer factor for particular trophic levels with special emphasis on man should be continued (Skwarzec & Bojanowski 1992, Holm 1995, Skwarzec 1997).

## 3. Industrial production in the drainage area

Riverine and direct loads of pollutants (heavy metals and nutrients) entering the Baltic Sea are an important environmental problem (Bruneau 1980, Elmgren 1989, Lithner et al. 1990, Backlund et al. 1993, HELCOM 1993, 1998, Rheinheimer 1998, Jansson & Dahlberg 1999). Monitoring surveys of trace elements and radionuclides are therefore required to keep a check on the ecological status of the Baltic Sea (HELCOM 1991, 1993, 1997, 1998). The industries in the Baltic countries are mostly based on locally available raw materials e.g. the deposits of Fe, Cu, Pb and Zn ores which support numerous steel mills and stainless steel works, copper and zinc smelters and aluminium refineries. HELCOM (1998) reported that the riverine heavy metal load is the largest source of the total pollution load (c. 90%). Municipal and industrial wastewater discharges, as well as diffuse discharges constitute the major anthropogenic sources in the riverine load. According to Lithner et al. (1990) the anthropogenically-derived amounts of Cd, Pb and Hg entering the Baltic Proper were from 5 to 7 times higher than their natural load. According to Bruneau (1980) both Finnish and Swedish industries have contributed to the introduction of metal pollutants to both the Bothnian Bay and the Bothnian Sea. Finland (56%), Sweden (44%) and Norway (<1%) belong to the Bothnian Bay catchment area (HELCOM 1998). Both Finland and Sweden have steel mills on the Bothnian Bay. Finnish stainless steel plants may have discharged Ni and Cr from pickling operations. The Finnish fertiliser plant and Swedish forest industries, as well as pulp mills in the catchment area may be responsible for the discharge of pollutants to the Bothnian Sea (Bruneau 1980). These pollutants originate from the catchment area of which 80% belongs to Sweden, 18% to Finland and 2% to Norway (HELCOM 1998). The chemical industry is located mainly in Finland, i.e. refinery, fertiliser and chlorine plants and in Sweden – chlorine and PCV plants. It is important to note that chlorine plants are based on the mercury method, but discharge of this element is very low owing to extensive measures to reduce its environmental impact. Recently non-mercury type plants have come into operation. According to Bruneau

(1980), the Finnish textile industry, located mainly on the Baltic coast or on rivers flowing into the Baltic Sea, could be a major discharger of many chemical pollutants into the Baltic Sea. It is suspected that the major pollutant load in this area is attributed to the Finnish and Swedish pulp and paper industries (Bruneau 1980). Metal pollutants in the Gulf of Finland originate from its catchment area, of which 67% belongs to Russia, 26% to Finland, 7% to Estonia and $< 0.1\%$ to Latvia (HELCOM 1998). Russia has an aluminium works and fertiliser industry (the latter is also located on the Estonian coast), a highly developed chemical industry, petrochemical plants and a pulp and paper industry. Most of the pollutants are transported via the River Neva to the Gulf of Finland. Scandinavian petrochemical installations are located on the Baltic coast; the Finnish steel mills, pulp and paper industry, and manufacturing industry are located in this area and on the lakes and streams, where pollutants are discharged either directly or through Lake Ladoga (Bruneau 1980). The catchment area of the Gulf of Riga belongs to Latvia (39%), Belarus (20%), Russia (18%), Estonia (14%) and Lithuania (9%) (HELCOM 1998). The drainage area of the Gulf of Riga appears to be rather poorly industrialised (Bruneau 1980). However, a very large refinery, petrochemical plant and some paper mills, mostly of small size, are located in this region. The catchment area of the Baltic Proper belongs to all Contracting Parties except Finland, as well as the Non-Contracting Parties of Belarus, the Czech Republic, Ukraine and Slovakia. The catchment area of the Contracting Parties consists of subareas such as Poland (54%), Sweden (15%), Lithuania (9%), Russia (3%), Germany (2.6%), Latvia (2%), Denmark (0.2%) and Estonia (0.2%). Among the Polish rivers, the Vistula and Oder are the largest ones which enter the Baltic Proper, transporting metallic pollutants from industry located even in such distant areas as the borders of the Czech Republic and Slovakia (HELCOM 1998). Poland has smelters, refineries, a petrochemical centre, fertiliser production, mines, textile industry as well as steel, pulp and paper mills. The origin of pollutants entering the Baltic Sea from the Swedish Lake Mälaren is partly from the central industrial district located in the vicinity of many old mines. There are also ammonia plants, fertiliser plants, small steel mills and stainless steel mills on the coast. Mostly pulp and paper mills are located on the Swedish coast and on rivers discharging to the Baltic Proper. In Russia the most important source of pollutants in the Baltic Sea are fertiliser plants located on the southeast side of this basin, while in Germany there are petrochemical plants and steel mills (Bruneau 1980). Lithner et al. (1990) have estimated the collective deposition of trace elements such as Zn, Cu, Cd, Pb, As, Hg, Cr and Ni to the Baltic Sea. The contribution of Chernobyl-derived $^{137}$Cs to all Finnish rivers discharged into the Baltic Sea during 1986–96 was significant (65 TBq), as was that entering five Russian rivers (14 TBq) during only 1986–88 and to the Polish Vistula River (18 TBq) during 1986–96 (Gavrilov et al. 1990, Ilus & Ilus 2000, Saxén & Ilus 2000, Smith et al. 2000). Other rivers from the former USSR such as the Neva, Luga, Narva, Daugava and Neman contributed c. 2.6 TBq of $^{137}$Cs to the Baltic Sea. According to Ilus & Ilus (2000) Swedish rivers provided c. 150 TBq of $^{137}$Cs to the Baltic Sea during 1986–96. The contribution of the Oder River and smaller German rivers in this respect has been estimated at 10 TBq $^{137}$Cs. According to the above data these rivers have transported a total of c. 300 TBq of $^{137}$Cs into the Baltic Sea. Other sources of radionuclides are reprocessing plants in Western Europe, which since the 1970s have introduced c. 150 TBq into the Baltic Sea.

## 4. Global input of chemical elements and pollution status of the Baltic Sea

The Industrial Revolution began in the eighteenth century in England but in the countries around the Baltic Sea it started later, in the 1850–60s. Industrial production in the countries around the Baltic has grown steadily, particularly from the 1950s until the present. In consequence, large quantities of various chemical anthropogenically-derived compounds are introduced to the Baltic Sea every day. These substances come from land and marine point sources such as industrial plants, power plants, waste disposal sites, waste-water treatment plants as well as from diffuse, non-point sources through rivers or land run-off, e.g. agricultural pollution, domestic waste and traffic (Backlund et al. 1993). Riverine and direct point sources of nutrients, i.e. N and P, and of heavy metals, i.e. Cd, Cu, Hg, Pb and Zn entering the Baltic Sea in particular subregions have been estimated by HELCOM (1998). Moreover, both the point-source and diffuse loads of nutrients given for particular Baltic countries have been estimated there. The Baltic drainage basin also receives different pollutants from long-range atmospheric transport from the British Isles, Central and Eastern Europe, and from even more distant regions. There are numerous anthropogenic emitters in the countries bordering the Baltic Sea. The structure of industry is in principle different in particular Baltic countries. The metal, pulp and paper industries are the most important branches in Sweden and Finland, the food industry is dominant in Denmark; by contrast, the industrial structure in Germany is very diversified. Industries in these countries generally use advanced technology, hence direct pollutant emissions have been significantly reduced over last two decades. However, there are still problems involving the diffuse sources of toxic substances and they remain to be solved. On the other hand, in the countries of the former communist block many industrial plants use outdated technology. In those countries there are problems associated with waste handling and excessive quantities of nutrients and industrial pollutants are therefore, transported to the Baltic via rivers (Backlund et al. 1993, HELCOM 1998). The amounts and types of pollutants are thus very different in particular sub-areas of the Baltic. For instance, the load of air-borne pollutants is higher in the southern than in the northern part, because the former part is more densely populated and more heavily industrialised. Moreover, in the south there is a more extended atmospheric transport of pollutants from remote areas (Pacyna 1984, Pacyna et al. 1984). On the other hand, the Bothnian Sea and the Bothnian Bay are polluted mainly by sources in Sweden and Finland, although some minor amounts of contaminants reach this area by means of water currents and winds from the south. It is supposed that various pollutants from industrial wastes discharge into Lake Ladoga and are then taken by the Neva River to the Gulf of Finland, where they finally affect the Baltic (Bruneau 1980). The Baltic Proper is heavily polluted by sources located along the eastern and south-eastern coasts. On the Swedish side, the water entering the Baltic originates partly from the central industrial district with numerous old mines and steel mills, refinery and ammonia plants and others. In Russia (Kaliningrad district), on the southeast side of the Baltic there are fertiliser plants and paper mills. Especially the Polish rivers (Vistula and Oder), the St. Petersburg area and northern Estonia, Latvia and Lithuania contribute considerably to the high total discharges of pollutants to the Baltic Proper (Bruneau 1980, Backlund et al. 1993, Enell 1996, Tammemäe 1998).

## Chemical budget

The first reports on trace element inputs to the Baltic Sea were published in the 1970s (Suess & Erlenkeuser 1975, Åkerblom 1977). However, budgets for chemical elements and nutrients for the Baltic Sea were reported later (Hallberg 1979, Pawlak 1980, Rodhe et al. 1980, Dybern & Fonselius 1981, Boström et al. 1983, Andreae & Froelich 1984, Brügmann 1986, Brügmann & Lange 1990, Lithner et al. 1990, Löfvendahl 1990, Hallberg 1991, HELCOM 1991, 1993, Brügmann et al. 1991/92, Backlund et al. 1993, Kihlström 1992, Pacyna 1992, 1993, Forsberg 1993, Brügmann 1994, Wulff et al. 1994, 1996, Schneider 1995, Enell 1996, Brügmann & Matschullat 1997, Brügmann et al. 1997, Matschullat 1997, Danielsson 1998).

According to Brügmann & Matschullat (1997), 65% Pb, 51% Zn, 48% Cd, 13% Cu and 11% Hg are introduced into the Baltic from the atmosphere with respect to the combined atmospheric and fluvial (riverine, industrial and municipal) inputs of these trace elements. It is also shown that 47% Zn, 34% Cu, 28% Pb, 25% Hg and 20% Cd introduced into the Baltic each year are fixed in the sediments.

Based on the data presented by Matschullat (1997), it can be concluded that the atmospheric input of many anthropogenically-derived trace elements, e.g. Cd, Cu and Pb, exceed their riverine input. This is in agreement with the HELCOM (1991) report emphasising atmospheric input as the predominant entry path of some heavy metals. Inputs of trace elements to the Baltic Sea (t $y^{-1}$) and the respective anthropogenic contribution are presented in Table 1. While the inputs of Cd, Cu, Pb and Zn are almost identical, As, Co, Cr, Hg and Ni are generally transported via rivers. As can be seen from Table 1, the anthropogenic contribution of the total load is very high ($> 70\%$) for As, Cd, Cu, Hg, Pb and Zn; lower values are obtained for Co, Cr and Ni (44–57%). According to Andreae & Froelich (1984) 281 t As, 75 t Sb and 46 t Ge were deposited annually in the Baltic Sea. It is estimated that As is carried to the Baltic Sea mostly by rivers, while for Ge the atmospheric transport predominates. As regards Sb, the atmospheric component is also considered the most important in the transfer of this element to the Baltic Sea. It is shown that the anthropogenic component has contributed significantly to the mass balance for As and Sb, and probably to the greatest extent for Ge (Andreae & Froelich 1984).

**Table 1.** Elemental input to the Baltic Sea [t $y^{-1}$], divided into natural river and atmospheric inputs, total fluvial and airborne inputs, the sum total for the Baltic Sea and the respective anthropogenic percentage. After Matschullat (1997), modified

| | As | Cd | Co | Cr | Cu | Hg | Ni | Pb | Zn |
|---|---|---|---|---|---|---|---|---|---|
| Natural fluv. | 58 | 5.1 | 120 | 270 | 310 | 4.9 | 165 | 140 | 1700 |
| Natural atm. | 10 | 3 | 3 | 20 | 130 | 0.1 | 5 | 20 | 350 |
| $\sum$ Natural | 68 | 8.1 | 123 | 290 | 440 | 5 | 170 | 160 | 2050 |
| Fluv. + diffuse | 200 | 60 | 200 | 440 | 1300 | 50 | 300 | 1500 | 6000 |
| Atmospheric | 50 | 60 | 20 | 100 | 1200 | 20 | 100 | 1300 | 5000 |
| $\sum$ Baltic Sea | 250 | 120 | 220 | 540 | 2500 | 70 | 400 | 1800 | 11000 |
| % Anthropog. | 73 | 93 | 44 | 46 | 82 | 93 | 57 | 91 | 81 |

## Pollution status of the Baltic Sea with respect to other seas

Sekulić & Vertačnik (1997) have estimated the annual load of metals such as Cd, Cr, Cu, Hg, Mn, Ni, Pb and Zn for the Baltic Sea, Adriatic Sea and Black Sea via wastewater and 'natural' waters. The evaluation of data on dissolved species of trace elements concerning the Black Sea and the North Aegean Sea indicate that in this interrelated system, water mass exchanges play an important role in the trace element distribution (Zeri et al. 2000). Although these Seas are almost enclosed basins they do differ significantly with respect to their biological and physical-chemical characteristics.

It should be stressed that a high load of particulate matter is transported annually to all three enclosed seas; this suspended material, enriched in heavy metals, is deposited in the vicinity of its terrestrial source, hence the concentrations of chemical pollutants are elevated exclusively in the narrowest littoral zones, while low levels are detected in deep-waters (Sekulić & Vertačnik 1997). Several 'black spots', e.g. great estuaries and seaport towns, heavily contaminated by chemical elements, are identified in each of the Seas (Sekulić & Vertačnik 1997). Therefore, the present pollution status has ecological implications primarily for the enhanced point-source spots. Among these Seas, the Baltic is the most heavily loaded with trace elements. In contrast, the Adriatic has the lowest load taken absolutely and relatively compared to its volume, while the Black Sea, and especially the Baltic have a significantly higher load. The quantities of 'natural' waters are several orders of magnitude higher than 'anthropological' waters. Owing to the expected 'natural' loading of chemical substances it far exceeds the anthropological one. The relatively low levels found in these three Seas mean that these great natural systems possess very considerable self-purification copabilities and stability.

## General remarks and recommendations

The input of selected chemical elements to the Baltic Sea is not well known. This situation is due mainly to an insufficient data base, uncertainties about how much of the gross input finally arrives in the open sea, and a very limited knowledge of the nature and extent of the exchange at the interface with the sea floor and with the atmosphere (Brügmann 1994). Although the data matrix utilised in mass balance calculations has recently been improved significantly, it is still impossible to present very precise data on trace element inputs to the Baltic Sea. The main reason for this is that reliable data for the water discharge of the rivers alone have not been available so far. This problem is particularly connected with incomplete pollution data given by some Baltic countries, because a lot of the figures were only estimated as totals by sub-regions, and figures from the Kaliningrad region have been lacking completely. Many uncertainties remain with regard to trace elements due to incomplete load data sets, especially from Russia and partly from all Contracting Parties (HELCOM 1998). It is well known that the major part of the pollution load is transported to the Baltic Sea by rivers. It is therefore an important task to start investigations on collecting load data for point and diffuse sources covering the entire Baltic catchment area. National and international research programmes should increase our knowledge of the chemistry, biology, hydrography and meteorology of the Baltic Sea and provide us with indispensable information for effective protective measures (Rheinheimer 1998). The protective measures taken by HELCOM have already contributed to an improvement in the ecological situation in the Baltic Sea. For instance, modern, more effective treatment plants for sewage have been constructed. The concentrations of some pollutants, e.g. Pb in water have decreased significantly. According to Rheinheimer (1998), the harmful substances which accumulated in the sediments, however, will still pose a threat for the near future. The atmospheric inputs are still on a large scale. The increase in traffic, industrial production and tourism, particularly in the eastern countries, will probably contribute to a rise in pollution at least at this early stage before any improvement in the treatment plants for sewage and exhaust gases can be achieved (Rheinheimer 1998).

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