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Short communication

Metal concentration in biota in the North sea: changes and causes

Britta Pedersen

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The article reviews available information on time trends in metal concentration in biota from monitoring studies in the North Sea. Data collected in Danish marine waters are presented as examples of changes in metal concentrations since 1975. Causes of trends and problems associated with the use of measurements in biota when assessing time trends in metals are discussed. Although emissions of many metals have apparently been reduced during the last 15 years due to different regulations, it has only been possible to detect any significant changes in the environment in very few of the more contaminated coastal areas. It is argued that this might well be caused by the lack of understanding of the effects of biological covariables on metal concentrations which may explain part of the variance observed in samples, and also of the physical and biogeochemical processes, which may cause changes in the bioavailability of metals and levels of contamination.

Key words: biota, monitoring, North Sea, trace metals, trends.

Britta Pedersen: National Environmental Research Institute, PO Box 358, DK-4000 Roskilde, Denmark.

Introduction

Trace metals of natural origin are found in the environment and therefore the concentration in, and input to, the North Sea contains a natural and an anthropogenic component, which may both vary over time. In addition, many physical, chemical, and biological processes permanently affect the metal distribution in the marine environment, particularly in the transition zone between fresh and salt water.

The North Sea covers an area of 525 000 km^2 and has a volume of about 47 000 km^3. In general, contaminants entering the North Sea, including metals, disperse according to the currents, but may accumulate locally in sedimentation areas such as the intertidal zones in the Wadden Sea or the deeper parts of the northern North Sea. Therefore, different spatial and temporal patterns are to be expected in the metal concentrations in different areas.

Data on metal concentrations in the marine environment collected before 1975, when the first Århus symposium was held (Hempel, 1978), are generally not very reliable, because the values obtained were close to the then existing analytical detection limit. Since that time, however, the development of more powerful analytical methods and the introduction of improved laboratory conditions, including clean room facilities and systematic quality assurance procedures in the late 1970s, have stimulated the organization of many national and international marine monitoring programmes. An important objective of these has been to assess the effectiveness of measures taken to reduce marine pollution (trend monitoring), the Joint Monitoring Programme (JMP) for the North Sea area of the Oslo and Paris Commissions being one example. The data generated may now supply valuable information about possible trends, provided that the appropriate parameters have been selected.

Trends in trace metals in the marine environment are generally based on measurements of concentrations in the sediment or in biota (e.g. mussels, fish liver), because these represent integrated effects over variable time scales. Less commonly, the water phase is sampled directly because the lower values pose more analytical problems and because the snap-shot nature of such measurements results in higher variability. Even when the primary aim of monitoring is to detect trends which reflect measures taken at the input side, biota are frequently preferred over sediment because a major

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reason for concern regarding metals is their potential effect on the ecosystem. Concentrations in living tissues are expected to reflect these effects better than sediment concentrations.

Laboratory experiments indicate that concentrations in biota are often positively correlated with seawater concentrations, as for instance shown by Pentreath (1977) for the accumulation of ^{115m}Cd in the body tissue of plaice. Field experiments in a restricted harbour area have also confirmed that concentrations of Zn, Cd, Cu, and Pb in sea water and in shrimps followed the same seasonal pattern during a 3-year period (Alliot and Frenet-Piron, 1990).

The recent assessment of the quality status of the North Sea by the North Sea Task Force (NSTF, 1993a) has summarized information for 13 subregions which had been defined in accordance with hydrographic and biological information (ICES, 1983). A limited amount of data on the spatial distribution of metals in the offshore, northern subregions generally indicated very low concentrations, but no trend data were available for these areas. More data existed for the coastal subregions for all matrices, generally indicating elevated levels of metals and particularly high concentrations in many of the larger estuaries (NSTF, 1993a). Studies on temporal trends in metal concentrations in fish and shellfish for several coastal areas were also available for the cooperative work of JMG and ICES (ICES, 1991).

The general conclusion drawn in the Quality Status Report (NSTF, 1993a) was that only a few clear trends could be observed in the concentrations of contaminants in biota over the restricted time periods studied (1978–1988). The few exceptions include a downward trend in Pb in some areas, a downward trend in Hg in fish from the Forth estuary on the English east coast in response to effluent regulation measures (NSTF, 1993b) and in cod and plaice in the western part of the Southern Bight (NSTF, 1993c), and a reduction in Cd concentrations in the horse-mussel from the outer Humber on the English east coast (NSTF, 1993a). Off the Belgian coast, a downward trend has been reported for Hg (NSTF, 1993d), but this has not been supported by observations in neighouring areas, including the Western Scheldt and Dutch coastal area. In all other subregions, including the Skagerrak/Kattegat, the English Channel, and the Wadden Sea, no obvious temporal trends have been found for any metal (NSTF, 1993a).

A new assessment of trends in biota from the JMP data set (Oslo and Paris Commissions, 1994) based on data up to and including 1991 and improved statistical methodology and a more rigorous quality assurance procedure compared to previous assessments revealed 6 significant downward trends among the 16 data sets investigated for Pb. This was also the case in 2 out of 33 data sets for Cd, in 3 out of 22 for Cu and in 6 out of 50 for Hg. Downward trends were mainly observed along the Belgian and Dutch coast and for Hg also in the Wadden Sea and in the Ems–Dollart estuary. For Zn, conflicting trends were found for different organisms, which precluded an overall conclusion.

The variable results from the North Sea trend monitoring studies are in accordance with recent findings for trends of metal concentrations in the American "Mussel Watch Programme". At most individual sites studied, trends were not significant, but among the significant correlations between concentration and year decreases outnumbered increases (O'Connor, 1992).

Overall, the available monitoring data do not allow firm conclusions to be drawn about reduced levels of metal concentrations in the marine environment. This paper addresses some of the problems associated with the use of concentrations in biota as indicators in trend monitoring and the interpretation of the results in relation to the causal factors. Metal concentrations have been measured regularly in the Danish marine and coastal areas since 1979 in different matrices, mainly in the form of the Danish contribution to the monitoring programmes of the Oslo and Paris and Helsinki Commissions. Some of the results obtained from fish tissues are presented here as examples as a background to this discussion.

Methods

In the autumn since 1979, plaice (*Pleuronectes platessa*) have been sampled at three stations along the Jutland west coast and flounder *Platichthys flesus* in the northern part of the Sound (Nivå) and in the Great Belt in the east part of Denmark. The sampling sites are generally located far from any point source of discharge except for the site in the Sound, which is influenced by anthropogenic discharges of metals from the city of Copenhagen and three larger Swedish cities (Jørgensen and Pedersen, 1994). Sampling procedures and analysis followed the guidelines given by the Oslo and Paris Commissions (1990). When possible, 25 fish were sampled at each station and length, weight, sex, age, liver weight, and liver dry weight (DW) were recorded. Concentrations of mercury (Hg) in fish muscle and of cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn) in the liver were measured in each fish separately, as described by Jensen and Cheng (1987).

Results

The average concentrations of Cd and Hg in plaice from Hvide Sande at the Jutland west coast are given in Figure 1 as a function of the sampling year, while data for the same metals in flounder from Nivå in the Sound are given in Figure 2. The standard errors on these values were in the range of 4–14% for Hg and of 5–26%

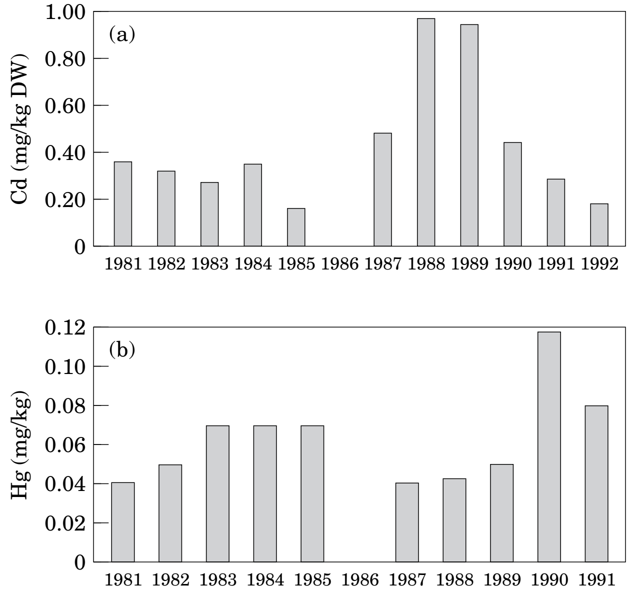

Figure 1. Average annual concentration of Cd in the liver (a) and Hg in the muscle (b) of plaice from the Jutland west coast (Hvide Sande). Cd is given on a dry weight (DW) basis and Hg on a wet weight (WW) basis.

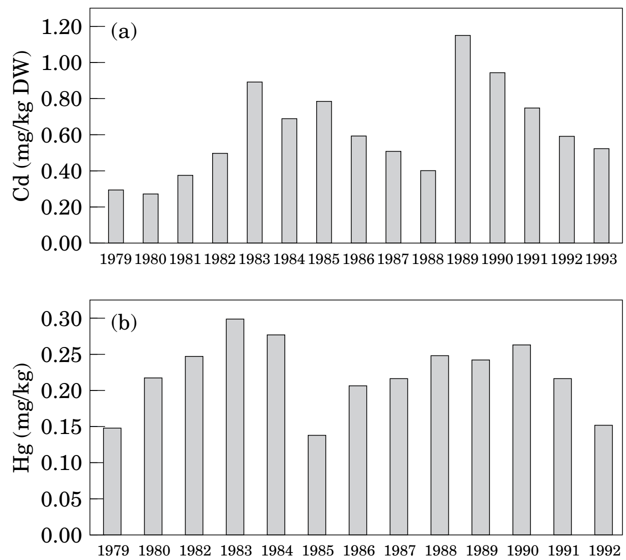

Figure 2. Average annual concentration of Cd in the liver (a) and Hg in the muscle (b) of flounder from the Sound (Nivå). Cd is given on a dry weight (DW) basis and Hg on a wet weight (WW) basis

for Cd. These results may be considered as typical examples of the temporal trends observed in these areas. Although no clear long-term upward or downward trend was observed for any of the metals except for Pb, the patterns indicate large and significant variations, sometimes up to a factor of 5 between years.

The interpretation of the observed variations, however, is not straightforward, because part of the observed differences may actually be caused by biological covariables. As an example, Figure 3 shows the relationship between Hg and size of flounder from the same site in the Sound for two different years. The regressions are significantly different ($p<0.001$; Student's *t*-test), indicating that size may explain a significant part of the variation between individual fish in one year and not in another.

Changes in run-off from year to year often have a great impact on the local marine environment in coastal

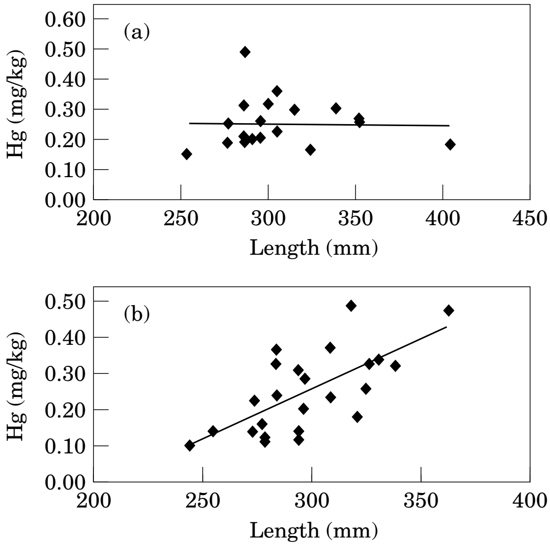

Figure 3. Relationship between the Hg concentration in the muscle and length of flounder from the same site in the Sound in 1981 (a) and 1987 (b). Hg is given on a wet weight basis.

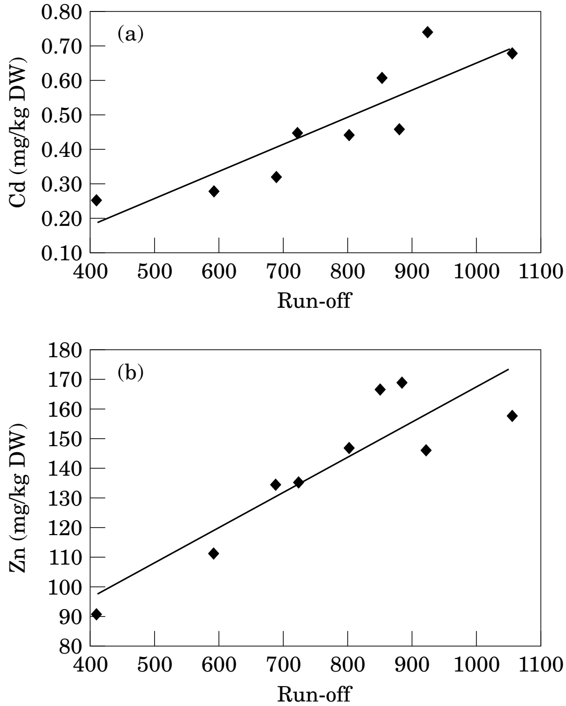

Figure 4. Average annual concentrations of (a) Cd and (b) Zn in the liver of flounder from the inner Danish waters (Sound and Great Belt) as a function of average monthly runoff (km^3) to the eastern part of Denmark with a 3-year time lag, 1979–1988. Cd and Zn are given on a DW basis.

areas, for instance due to changes in the loads of nutrients and contaminants. As an example, the annual average metal concentrations of Cd and Zn at the two stations in the Sound and in the Great Belt were plotted against the run-off to the eastern part of Denmark with a 3-year time-lag (Fig. 4). Cu showed a similar dependence, but there were no correlations between run-off and the Pb and Hg concentrations. Correlations using other time-lags were also investigated, but 3 years gave the best fit.

Discussion

Fluctuating trends are not specific for Danish waters but have also been found in many other coastal areas around the North Sea (NSTF, 1993a). There may be many reasons for the yearly fluctuating patterns often observed. Variability in the metal inputs to an area are certainly important. These inputs to the marine environment may follow different pathways: rivers, disposal of dredged harbour sludge and atmospheric depositions being the most important ones. Direct discharges of industrial and sewage effluent may also add to the inputs.

Total emission has apparently been reduced for a number of metals during the last 10 years owing to a variety of regulations. However, not all types of emission have been monitored sytematically. For instance, Denmark has only regularly measured the discharge of metals in industrial effluent, which makes up only a minor fraction of the total emissions. Because comprehensive collection of input data to the North Sea did not start until 1990, a trend assessment of the input is not yet possible for most areas to support an evaluation of the causes of the patterns found in biota (Oslo and Paris Commissions, 1994).

More data on metal loads are available from some countries bordering the southern part of the North Sea. Although these data are rather variable, total input of some metals appears to have decreased during the period 1985–1989 (NSTF, 1993d). However, the main declines in input from some of the main sources, particularly the rivers Rhine and Meuse, had already occurred before 1985. The decreasing trends in input of, for example, Cd and Hg from rivers are in accordance with the decreasing trends observed in blue mussels (Oslo and Paris Commissions, 1994), while significantly decreased sediment (fraction <63 μm) concentrations of Cd, Cr, Cu, Zn, and Pb between 1981 and 1991 supports the view that the quality of the environment has improved (NSTF, 1993a).

In these studies, sediment concentrations revealed significant trends in more metals compared to concentrations in biota, suggesting that the use of the latter as an indicator is less sensitive in detecting time trends in trace metals. The apparently lower sensitivity of biota compared to sediment may be due to a larger temporal variability of the concentrations as a result of a variety of factors, including:

(1) *Sampling strategy*: Bignert *et al.* (1993) have demonstrated the prerequisite of a sufficiently long time-span and high sampling frequency for detecting any existing trends in the environment, using PCB in fish as an example.
(2) *Effects of biological covariables*: Our knowledge about the effects of for instance fat content, age, sex, length and weight on metal concentrations in the tissues is still fairly limited. Although in many cases metal concentrations have been found to be correlated with the length of an individual, the relationship may vary from year to year, as indicated by the results for flounder in the Sound. Thus, the inclusion of any relationship in statistical models for trend assessment is not straightforward.
(3) *Local conditions*: An understanding of the physical and chemical processes involved in the transport and distribution of metals is essential for selecting the proper sampling sites in relation to the emission source. Although statistical correlations may assist in finding the underlying causes and in understanding the processes, the examples of the observed relationships between metals and run-off cannot be interpreted easily. Although run-off would appear to be of no significance in relation to variability in metal concentrations, it may have an influence on biogeochemical processes in the environment associated with accumulation in biota. So far, there is no clue as to why there would be a 3-year time-lag between these factors, and in fact the correlations may be purely accidental.
(4) *Local biogeochemical processes*: There may be time-lags between a change in input and a change in concentration in biological tissues, because chemical and physical processes in the sediment or the water phase may act as a buffer. Moreover, only some of the metals may be available for uptake by biota and the bioavailability may change with environmental factors such as salinity, temperature, dissolved organic compounds as well as food supply. The effects of (combinations of these) factors are difficult to judge from laboratory experiments, but they may have a decisive influence on the accumulation in biota.
(5) *Accumulation strategy*: Rainbow *et al.* (1990) have highlighted the importance of understanding the processes involved in the accumulation of particular trace metals in evaluating the significance of observed metal concentrations within the context of biomonitoring.

Conclusion

Although the different regulations should have reduced the input load of metals to the North Sea during the last 10–20 years, there is relatively little evidence that concentrations in the system have declined. More systematic quantitative information is needed about inputs, particularly atmospheric input, to support an interpretation of the causes of the trends observed in the marine environment. In addition, more knowledge is required about the physical and biogeochemical processes affecting the distribution of metals in order to be able to link trends in input to trends in the marine environment.

Using metal concentrations in biota as indicators of trends in the marine environment remains a challenge, but the effects of biological covariables must be better understood. When part of the sampling variance can be explained, this will improve the sensitivity of biological indicators for detecting time trends. Attention must also be given to variations in the bioavailability and its effect on accumulation in response to natural changes in the environment. The underlying processes must first be understood before observed correlations may be meaningful. It is only when such problems are resolved that our abilities to relate the monitoring results to the effects of regulations on the input side may improve.

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