

Regional distribution of manganese, phosphorus, heavy metals, barium and carbon in sea-bed sediments (0-2 cm) from the northern part of the Norwegian Skagerrak

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The regional distribution of manganese, phosphorus, heavy metals, barium and organic carbon in sea-bed sediments (0-2 cm) from the northern part of the Norwegian Skagerrak has been mapped and the results discussed in the light of dispersion processes from natural and human sources. Similar distribution patterns are found for manganese (Mn), phosphorus (P) and the heavy metals Cr, Cu, Ni, Pb, V and Zn which all intercorrelate well. The highest levels of these elements occur in the deepest parts of the Skagerrak Basin. Barium (Ba) is concentrated at stations located in the shallower waters in the southern part of the investigated area. Mercury (Hg), on the contrary, has a bimodal distribution, being enriched in the northernmost area and along the Norwegian south coast. The highest concentrations of total carbon and organic carbon are found at intermediate water depths around the perimeter of the deep Skagerrak Basin. According to the State Pollution Authority's (Statens Forurensningstilsyn 1993) classification of the condition of sediments based on concentrations of heavy metals (Category I = Good, II = Fair, III = Poor, IV = Bad, V = Very bad), the sea-bed sediments in the northern part of the Norwegian Skagerrak fall in Category II with respect to Ni and Pb and in Category I with respect to Cr, Cu and Zn.

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Introduction

The Geological Survey of Norway, in cooperation with the University of Bergen and several other scientific institutions, has had responsibility for coordinating geochemical, geological and geophysical mapping of the sea-bed and its immediate subsurface in the Norwegian part of the Skagerrak. The Skagerrak is one of three principal areas in the North Sea (the other two are the Wadden Sea and the German Bight) where deposition of natural and anthropogenic suspended matter occurs today. In this contribution we present the results of the geochemical mapping of sea-bed samples collected during the 1992 and 1993 cruises with emphasis on the lateral distribution of manganese, phosphorus, heavy metals, barium, and total and organic carbon in the top 0-2 cm. These parameters have been chosen because some of them are perturbed by anthropogenic activity and are indicators of pollution by man. In addition, some of them act as active adsorbates or sinks for the heavy metals investigated and thus affect their distribution. These data reflect the effect of transport and depositional processes on the distribution of contaminants, and give important information about the current extent of sediment pollution within this key area of the North Sea.

The bathymetry of the Skagerrak is characterised by a 600-700 m deep, elongated trough, the Norwegian Trench, with its long axis oriented northeast-southwest (Holte Dahl 1986) (Fig.1). Several currents merge in the Skagerrak. The Baltic Current drains brackish water north-

wards through the Kattegat and into the eastern part of the Skagerrak (Stigebrandt 1983) where it converges with the northeasterly longshore Jutland Current, which sweeps the western side of the Jutland Peninsula and is fed by some of the largest rivers in northwestern Europe (e.g. the Elbe, Ems, Meuse, Rhine, Thames and Weser). Water masses from the northeast Atlantic Ocean, which enter the Skagerrak basin at depth from the west (Danielsen 1991), are high in nutrient elements and total dissolved solids. The influx of water masses is balanced by water leaving the Skagerrak in a southwesterly direction as the Norwegian Coastal Current (Larsson & Rohde 1979, van Weering 1981). The coalescence of these major currents causes an increase in sediment transport capacity and subsequent sedimentation in the deeper, central parts of the Skagerrak. However, the sedimentation processes are complex because of stratification of the water masses, resuspension of bottom sediments during extreme weather conditions, and a mixture of anthropogenic and natural sources. The provenance of the sediments is thus by no means straightforward.

The highest sedimentation rates within the studied area are measured in the southern and northeastern parts (Eisma & Kalf 1981, van Weering 1981). The sediments within the Skagerrak are transported by suspension and consist mainly of clay and silt, but samples from the southernmost stations (64, 65 and 66, see Fig. 1) and single stations close to the Norwegian mainland (Eisma 1981, van Weering 1981, van Weering et al. 1987, 1993, Eisma & Kalf 1987, Bøe et al. this volume) consist of a sig-

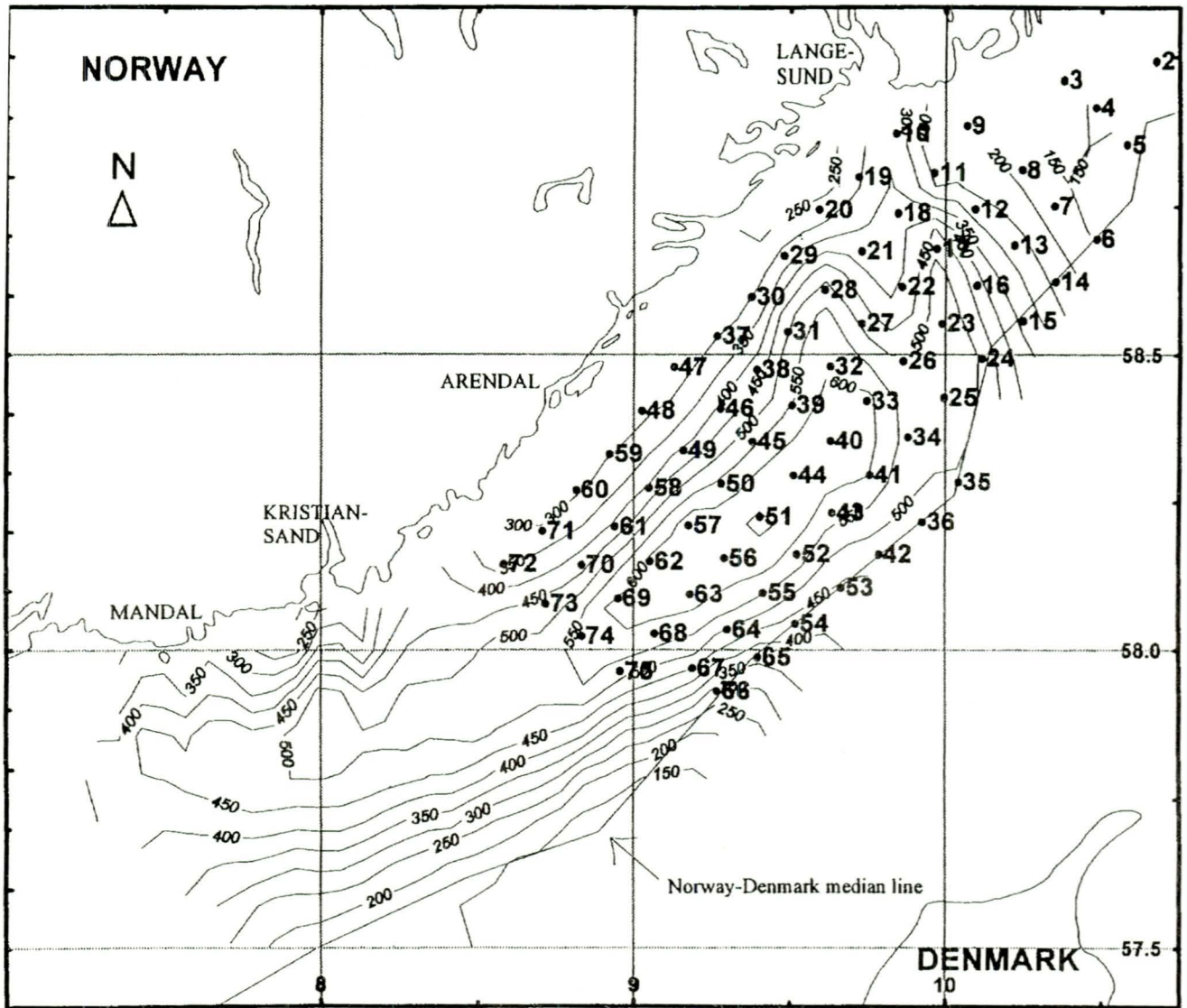


Fig.1. Sampling stations (numbered 2-75) and bathymetry (contour interval 100 m) of the northern part of the Norwegian Skagerrak.

nificant portion of fine and very fine sand.

Cores of accumulated bottom sediments from the Skagerrak should give the best indication of the general trends in North Sea water quality over time and particularly of the contaminant content of the suspended particulate matter in the water masses (Turrell 1992). Earlier geochemical work on sediments from the Skagerrak has focused on a wide range of dispersion processes and sources which might have caused the present vertical and lateral distribution of heavy metals (Anton et al. 1993, Förstner & Reineck 1974, Dominik et al. 1978, Kuijpers et al. 1993, Kunzendorf et al. this volume). Attempts to estimate the enrichment factors of heavy metals in Skagerrak sediments have been made by Cato (1977, 1986), Erlenkeuser & Pederstad (1984), Müller & Irion (1984), Holtedahl (1986) and Pederstad et al. (1993).

Sampling and analytical methods

Sediment cores were collected down to a sediment depth of about 50 cm using a Niemistö® corer (Niemistö 1974) at each of the 74 sampling stations (numbered 2-55 and 56-75 for stations sampled during the 1992 and 1993 cruises, respectively) (Fig.1). Sub-samples for geochemical analyses were collected onboard by cutting the sediment core into ten 2 cm-thick slices down to 20 cm depth and then at 25-27 cm, 35-37 cm, 45-47 cm and 55-57 cm depth below the sediment/water interface. The sliced geochemical samples were immediately frozen at -18°C and later freeze-dried. The inorganic components were extracted with 7N HNO_3 in an autoclave at 120°C for one hour in the laboratory (Norsk Standard 1980). The element contents were determined by using inductively coupled plasma (ICP-AES) for barium (Ba), chromium (Cr),

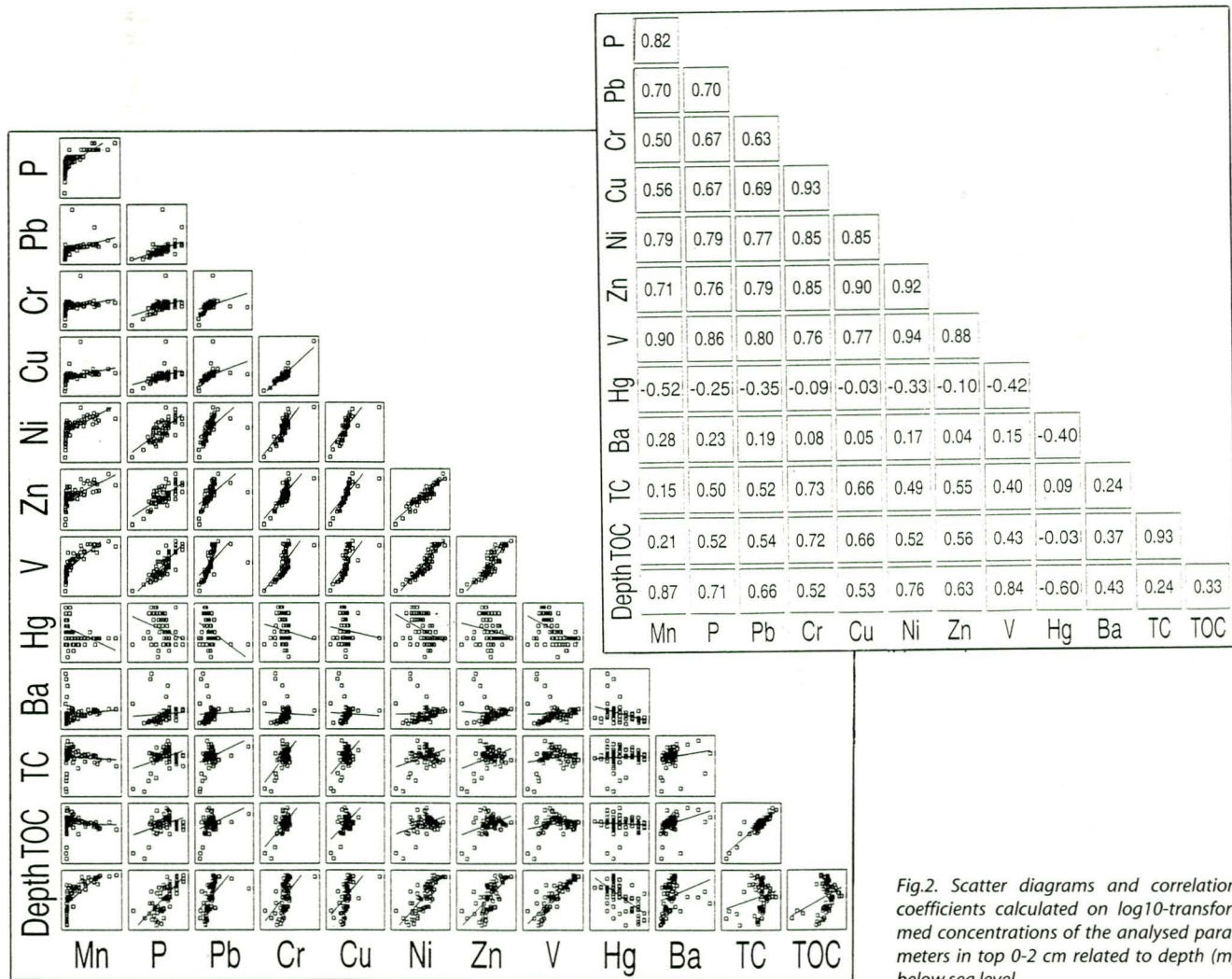


Fig.2. Scatter diagrams and correlation coefficients calculated on log10-transformed concentrations of the analysed parameters in top 0-2 cm related to depth (m) below sea level.

copper (Cu), manganese (Mn), nickel (Ni), phosphorus (P), lead (Pb), vanadium (V) and zinc (Zn) (Ødegård 1987), and by cold vapour atomic absorption spectroscopy (AAS) for mercury (Hg) (Hatch & Ott 1968). In addition, total carbon (TC) and total organic carbon (TOC) were determined by ignition using a LECO® stove.

Results and discussion

Co-variations between parameters

A correlation matrix for the log₁₀-transformed data and a scatter-diagram with the linear data, all plotted versus water depth, are shown in Fig. 2. The positive correlation (r>0.50) between the heavy metals Pb, Cr, Cu, Ni, V, Zn, and partially TC and TOC, with Mn and P, suggests some similarities with regard to source and fate in this environment. Between this group of elements and Hg or Ba there is no significant correlation. Nor is there a significant correlation between Hg and Ba. This indicates that these elements have separate sources and modes of occurrence.

Manganese correlates very well with water depth (r=0.87), V (r=0.90) and P (r=0.82) and fairly well with Zn (r=0.71), Cu (r=0.56), Cr (r=0.50) and Pb (r=0.70). Vanadium correlates very well with Mn (r=0.90) and P (r=0.86), with the heavy metals Pb (r=0.80), Cr (r=0.76), Cu (r=0.77), Ni (r=0.94) and Zn (r=0.88), and with water depth (r=0.84). Vanadium co-precipitates with Mn during diagenesis, and shows a more linear increase in concentration as a function of water depth than manganese (see Fig. 2). This probably reflects the property of V to appear in several oxidation states (+II, +III, +IV and +V), e.g. it is the key transition metal in homogeneous catalytical respiration of some lower marine biota (Hägg 1966); and the oxidation state of vanadium is also sensitive to the periodically reduced oxidising conditions in the water column and the reducing conditions interstitially in the sediments just below the sediment/water interface. This could possibly be associated with a gradually diminishing transport capacity of the water currents with respect to particulate matter, including vanadium-bearing particles, as a result of increased water depth or an increased flocculation with depth of Mn-particles scavenging vanadium and

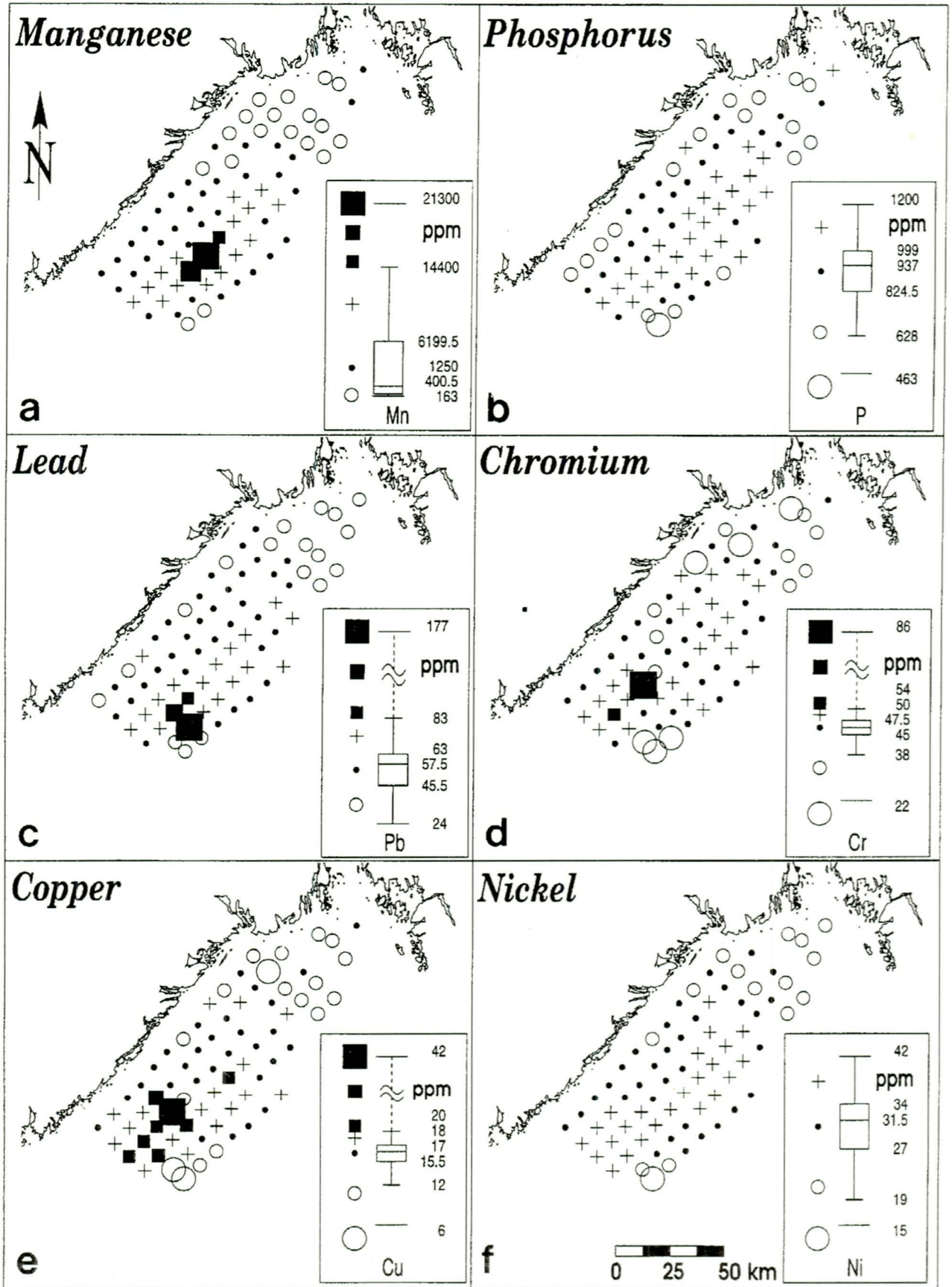
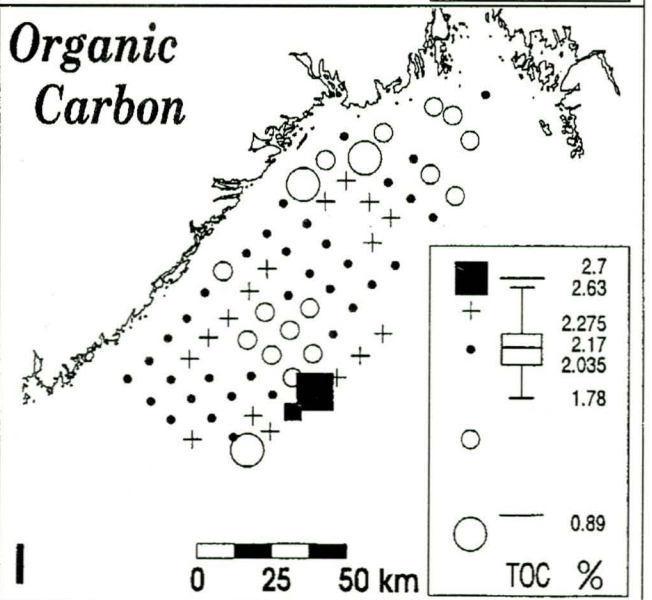
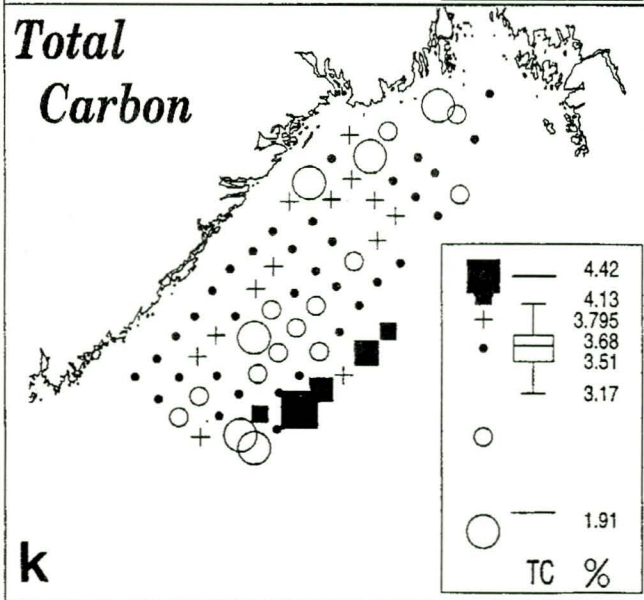
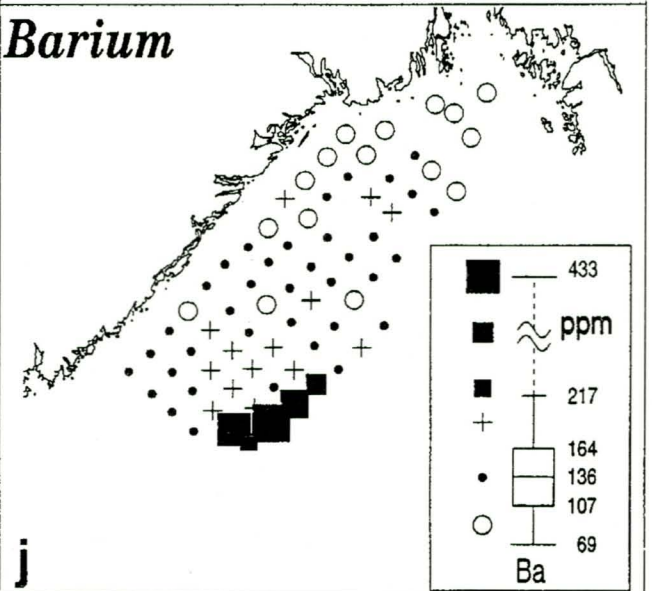
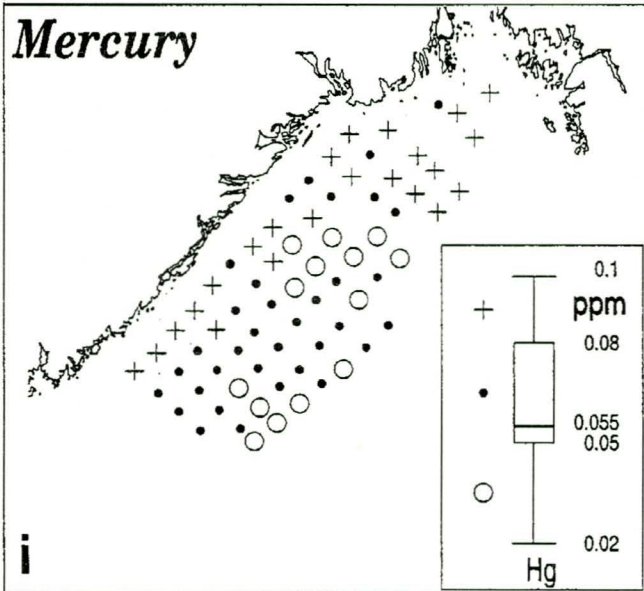
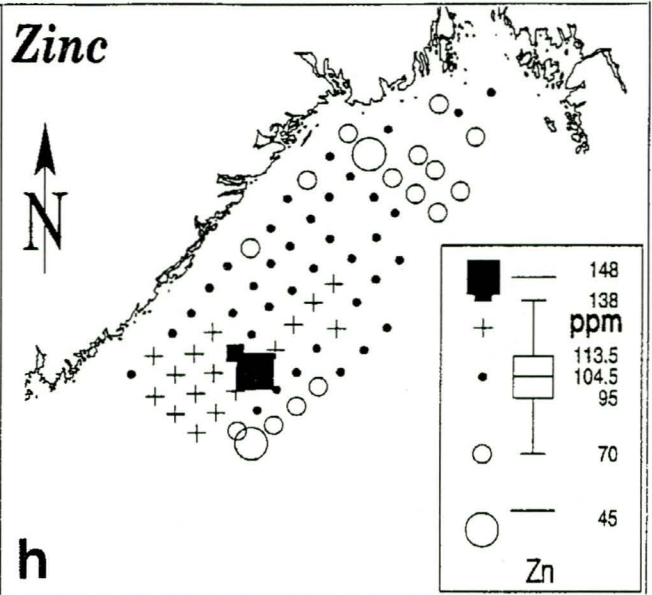
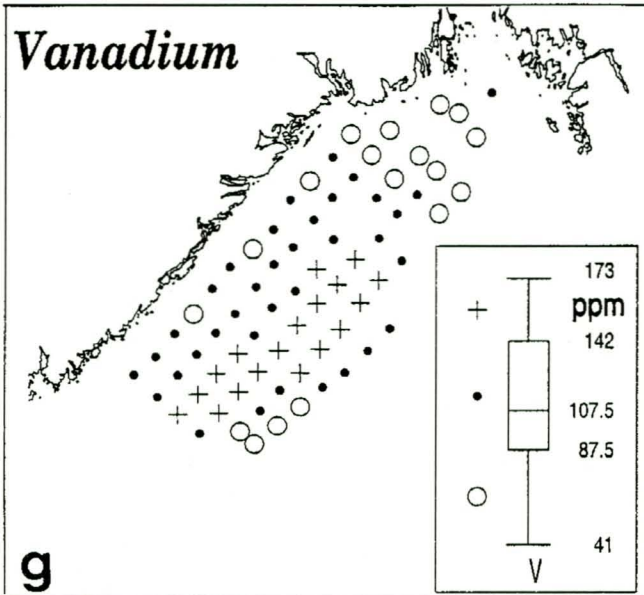


Fig.3. The concentration in surface sediments (0-2 cm) of a) Mn, b) P, c) Pb, d) Cr, e) Cu, f) Ni, g) V, h) Zn, i) Hg, j) Ba, k) TC and l) TOC. The symbols on the maps are chosen according to procedures developed for Exploratory Data Analysis (EDA) as shown by Tukey (1977) and Hoaglin et al. (1983). The symbols are linked to a boxplot showing the concentration level for the <25 (o), 25-75 (-) and >75 (+) percentiles. The bar-line in the box-plot indicates the 50%-value. Larger open circles (O) and solid squares (■) are chosen as symbols for samples with extremely low and extremely high concentrations as indicated in the legend in the lower right corner.



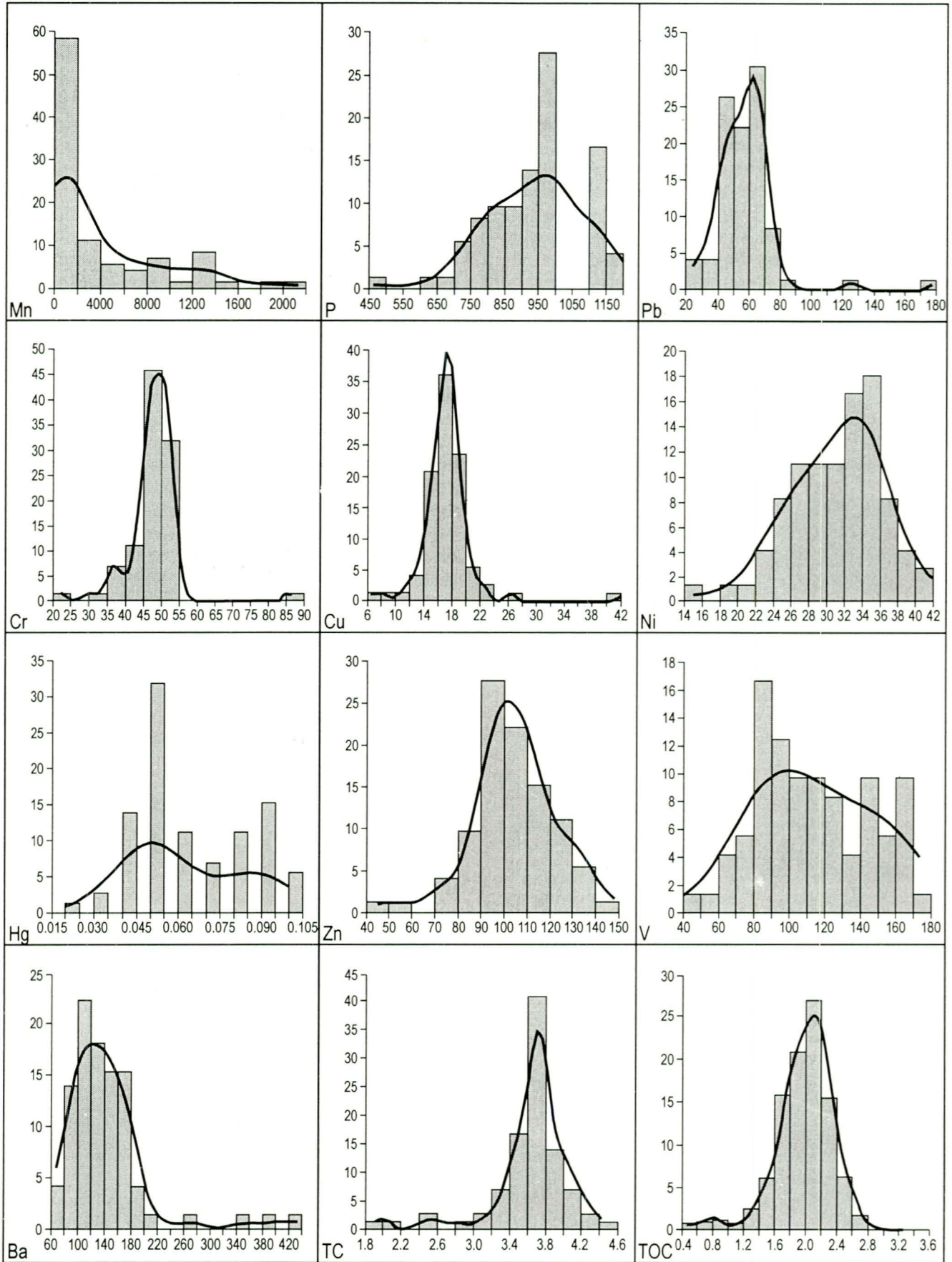


Fig. 4. Histogram and frequency distribution of elements mapped in Fig. 3.

other heavy metals. The heavy metals Pb, Cr, Cu, Ni and Zn show, except for a few outlying values, a nearly log-linear positive dependency with water depth. The concentration of Hg decreases as a function of water depth, whereas Ba, TC and TOC are enriched at intermediate water depths (Fig. 2).

Regional distribution of element contents

The regional distributions of Mn, P, Pb, Cr, Cu, Ni, V, Zn, Hg, Ba, TC and TOC in the sediments 0-2 cm below the sediment/water interface are presented in the geochemical maps shown in Fig. 3. This sequence of presentation is chosen because of the link between Mn and P with Cr, Cu, Ni, V and Zn revealed by the correlation analysis. The distributions of Hg, Ba and TC/TOC are each distinctly different.

Manganese (Mn)

Sediments with the highest concentrations of manganese are found in the deeper parts of the investigated area at more than 500 meters of water depth. At these depths the concentration of manganese in the sediments is above 5,000 mg/kg with a maximum value of 21,300 mg/kg. The median level of manganese is 1,250 mg/kg for the whole dataset, reflecting the positive skewness of its frequency distribution (Fig.4). Manganese is of considerable interest in this study, because it is widely used in industry and also affected by diagenetic processes. The latter take place when manganese in the sediments is mobilised by reducing processes and transported by diffusion to the sediment/water interface where it is oxidised and precipitates with iron as Fe-Mn-oxy-hydroxides. Moreover, these processes affect the concentration of other heavy metals like Cu, Ni, V and Zn, which are scavenged by the oxy-hydroxides (Jenne 1976).

The high concentrations of manganese at stations with water depths greater than 500 m partly reflect the reducing conditions in the sediments immediately below the water/sediment interface. This situation has probably been stable for many centuries. However, the general redox state of the local environment should be monitored in the future, especially with respect to indicators of chemical processes leading to the decomposition of organic matter, such as: 1) oxygen consumption, 2) nitrate reduction, 3) sulphate reduction and 4) methane formation (Berner 1971). Stagnant conditions leading to decomposition of organic matter and mobilisation of trace metals have only been reported from uncontaminated fjords in which circulation is hindered by sills at the outlet (Piper 1971, Skei 1983), and from fjords with an unusually heavy anthropogenic load.

Phosphorus (P)

Phosphorus shows a similar distribution to that of manganese with concentrations ranging from about 500

mg/kg in the northeast and southern parts of the investigated area and along the south coast of Norway to above 1,000 mg/kg in the deeper (more than 400 m depth) parts of the basin (compare with Fig.1). However, although the areal distributions of these two elements coincide and as it is known that P is incorporated in Mn-oxyhydroxides (Cronan 1976), the factors influencing the distribution of phosphorus are thought to be more of a biological and sedimentological nature than diagenetic. Since a major source of phosphorus is sewage effluent, it is most likely associated with fine particulate matter, e.g. dead algae, which settles in the deeper parts of the basin where the currents are weak. Another possible anthropogenic source of unknown magnitude is effluent from the producers of fertilizers for agricultural purposes.

Lead (Pb)

Lead is enriched in the deeper parts of the investigated area and ranges from 24 to 177 mg/kg; with a median value of 58 mg/kg; which is well above the average of 23 mg/kg cited for siltstone (Lovering 1976) and the 25-30 mg/kg shown on maps of the concentration of Pb in overbank deposits in southeastern Norway (Bogen et al. 1992). However, the measured values are comparable to the median 60 and maximum 240 mg/kg reported for 100 argillaceous bottom sediments from the Atlantic Ocean and the Caribbean Sea (Ericson et al. 1961).

Lead has been used by man over the last three millennia, but a significant increase in lead emissions to the atmosphere has occurred in this century, and in Europe especially since the 1950's when lead alkyl compounds were added to petrol as an anti-knock agent. During this process it is injected into the air by car exhaust as minute elemental or oxide particles and dispersed in the lower atmosphere until it precipitates with rain or snow. Worldwide consumption of leaded petrol peaked at c. 253,000 tonnes in 1970-71 (Nriagu 1990). Other uses of lead, e.g. in car batteries, as a component in anti-corrosion boat coating, and in ammunition both for hunting and for military purposes (World War I and II), might directly or indirectly have been the sources for some of the lead measured in these sediments. Air pollution is probably the dominant source of lead not bonded in the crystal lattice of silicate minerals in these sediments (Pacyna, pers. comm. 1995).

Chromium (Cr), copper (Cu), nickel (Ni), vanadium (V) and zinc (Zn)

The regional distributions of the elements Cr, Cu, Ni, V and Zn are very similar to those of manganese, phosphorus and lead, as is to be expected from the correlation analysis. The frequency distribution (Fig. 4) of these elements is unimodal with a fairly narrow range around the mean, except for V which seems to have two modes. This could reflect a clastic and diagenetic or biological mode of occurrence, or a separate anthropogenic source in

addition to the natural ones. Chromium and copper has, as is the case for lead, distinct outlying values, possibly reflecting the natural and anthropogenic sources.

In general, the lowest concentrations are measured at the northernmost and southernmost stations where the water depths are small. At more than about 400 m depth, the concentrations of the metals in the sediments are slightly higher than the median value with the highest concentrations present in the deepest part of the Skagerrak.

Mercury (Hg)

Mercury is present at rather low concentrations throughout the investigated area and ranges from 0.02 to 0.10 mg/kg with a median value of 0.06 mg/kg. The highest values are found in the northernmost part of the study area (stations 2-10), where the sedimentation rates are the highest (Bøe et al. this volume), and along the south coast of Norway at the sampling stations 19, 30, 37, 48, 59, 60, 71 and 72 which are located at relatively shallow depths (see Fig. 1). Mercury shows a distinct bimodal distribution suggesting that there might be at least two different modes of origin (Fig. 4). One of these is probably the chlor-alkaline industry located along the Swedish west coast (Cato 1977, 1986) and along the Glomma River at Sarpsborg and in the Langesundfjord area in southeastern Norway. Mercury has also been used in the paper industry to protect timber from infestation and in agriculture to protect cereal seeds from decay before sprouting. A third general source of mercury is public sewage containing contributions from dental offices. This amounted to a release of about one tonne Hg/yr in the mid-1980's (Statens Forurensningstilsyn (SFT) 1990). Emissions of Hg to the atmosphere from point sources in southeastern Norway have been estimated to be at 55 tonnes Hg in the period 1940-1980 (Anderson et al. 1987).

Barium (Ba)

Although Ba is not a heavy metal with proven toxic effects, it is of interest in this study because barite (BaSO_4 with a density of 4.48 g/cm^3) is abundantly used as an additive to drilling mud in the offshore industry. Barium

as barite may thus act as a tracer of drilling for petroleum offshore. The concentrations of barium in the sediments range from about 69 mg/kg to a maximum of 433 mg/kg. In the southernmost part of the sampled area it occurs in concentrations of more than 200 mg/kg and is clearly associated with the coarser sediments (Bøe et al. 1995). Using a scanning electron microscope (JEOL) with a backscatter imaging capability, we have proven that, at Stations 53, 54, 65, 66 and 67, barium occurs as discrete grains of barite. The barite grains in these samples could be either diagenetic or detrital. Barite is virtually insoluble in water. To what extent Ba is affected by diffusion processes and diagenetic enrichment in the top of these seabed sediments is not known. A diagenetic formation of barite crystals would presumably lead to authigenic, well crystallised mineral grains. Since the barite grains observed in these samples seem to be pseudospherical with about $10 \mu\text{m}$ diameter, i.e. smaller than the median diameter of the sediment grains in these samples, we assume they are detrital, probably derived from drilling muds used by the offshore industry in the North Sea. In the shallower waters along the coast and in the northernmost areas where the sedimentation rates are high, the concentration of Ba is below 100 mg/kg.

Total carbon (TC) and total organic carbon (TOC)

Total carbon and total organic carbon correlate very well ($r=0.91$) and exhibit similar distribution patterns, which are distinctly different from those of manganese and phosphorus. The highest values are found at intermediate water depths in the southern part of the investigated area and around the perimeter of the basin. The C/N ratio of the organic matter in the sediments from the Skagerrak is between 2 and 14 (Olausson 1975) indicating mainly a marine origin. According to van Weering et al. (1987) 400-600,000 tonnes of organic carbon accumulate each year in the Skagerrak.

Comparison with pollution standards

The statistics of our results are listed in Table 1 together with the limits used for classification into five categories

Table 1. Minimum, median and maximum concentrations of manganese, phosphorus, heavy metals, barium and carbon in sea-bed sediments (0-2 cm) from the northern part of the Norwegian Skagerrak compared to classification limits (Class I-V) given by the Statens Forurensningstilsyn (1993).

	Min	Median	Max	I	II	III	IV	V
Mn	163	1250	21300	-	-	-	-	-
P	463	937	1200					
Pb	24	58	177	<30	30-120	120-600	600-1500	>1500
Cr	22	48	86	<70	70-300	300-1500	1500-5000	>5000
Cu	6	17	42	<35	35-150	150-700	700-1500	>1500
Ni	15	32	42	<30	30-130	130-600	600-1500	>1500
Zn	45	105	148	<150	150-700	700-3000	3000-10000	>10000
V	41	108	173	-	-	-	-	-
Hg	0.02	0.055	010	<0.15	0.15-0.6	0.6-3	3-5	>5
Ba	69	136	433	-	-	-	-	-
TC	1.91	3.68	4.42	-	-	-	-	-
TOC	0.89	2.17	2.70	-	-	-	-	-

(Category I = Good, II = Fair, III = Poor, IV = Bad, V = Very bad) of the general condition of sediments with respect to their contamination of heavy metals (Statens Forurensningstilsyn 1993). Because of the unknown effect of different mineralogical compositions and applied analytical methods, a direct comparison should be carried out only with caution. However, if one compares the median value of our samples with that of each class, it seems as if these sediments are at least contaminated according to the levels in Category II with respect to Ni and Pb (except for at Stations 63 and 64, which have higher concentrations of lead and belong to Category III), and Category I with respect to Cr, Cu and Zn. The levels of Hg are all below the upper limit of Category I.

Conclusions

The distribution of the heavy metals Cr, Cu, Ni, Pb, V and Zn in the top 0-2 cm of sea-bed sediments from the northern part of the Norwegian Skagerrak seems to be strongly dependent on water depth, and on the distribution of manganese and phosphorus. A regional dispersion pattern with deposition in the deeper parts of the basin seems to be dominant for most of these heavy metals. There is a wide range of both natural and anthropogenic sources for the heavy metals in these sediments and an assessment of the contributions from different natural and anthropogenic sources is part of a separate study. The distribution of Hg is different from the other heavy metals and probably reflects the contribution from several anthropogenic sources in addition to the natural ones. One of these is the industrial activity in the northern and eastern areas outside the investigated area, and a second source is public sewage effluent from towns along the south coast of Norway (Statens Forurensningstilsyn 1990). Barium is not a heavy metal, but shows an enrichment as detrital grains of barite in sediments in the southern part of the studied area, where the coarsest sediments occur. The source of this barite is thought to be drilling mud used in the offshore industry.

The general level of contamination in the investigated area ought to be monitored in the future with reference to the data presented here. Compared to the State Pollution Authority's (Statens Forurensningstilsyn 1993) classification (I-V) of the general condition of the contamination level of sediments based on their concentration of heavy metals, the sea-bed sediments (0-2 cm) in the northern part of the Norwegian Skagerrak fall into category II with respect to Ni and Pb and in category I with respect to Cr, Cu and Zn. No classification is available for the other parameters discussed in this paper.

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