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Effects of morphological changes on metal accumulation in a salt marsh sediment of the Skallingen peninsula, Denmark

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Abstract

In 1931 a red-colored, sandy marker horizon was placed on the emerging Skallingen salt marsh. Sedimentation on top of the marker horizon since then shows two opposing tendencies. Coincident with salt marsh development the sediments display up to 1964 a fining upward sequence with an increasing content of organic matter. Since 1964 a nearby creek has meandered towards the sampling plot. Consequently, the sediments become coarser with a decreasing organic matter content. The morphological induced changes in sedimentary conditions strongly influence metal content in the sediments and thereby hide anthropogenic induced concentration variations. Thus, an apparently diminishing Zn content (per kg dry weight) since 1964 could indicate lesser load to the area. However, corrected for grain size effects there is an increasing content of Zn. Other metal concentrations (e.g. Cu) show a diminishing trend when corrected for grain size effects and therefore indicate a reduced anthropogenic induced load of these metals to the salt marsh.

Introduction

It is suggested that acceleration of present sea-level rise due to global warming could cause substantial losses of coastal salt marshes (Orson et al., 1985; Stevenson et al., 1986). Largely in response to such suggestions, salt marsh accretion processes have been increasingly studied during the last 10 years (e.g. French and Spencer, 1993; Roman et al., 1997). Another line of salt marsh studies has, on a wide basis, dealt with paleoecological and modern pollution trends (see Valette-Silver (1993) and Boorman (1999) for reviews). This type of study can be instrumental in assessing whether recent pollution control programs have been successful (Bricker, 1993; Daoust et al., 1996). Salt marsh sediments from lagoonal settings are specially suited for this purpose because they are usually protected from cyclical erosion and significant bioturbation (Stumpf, 1983) which otherwise complicates historical interpretation (McCaffrey and Thomson, 1980). It has been speculated and shown in laboratory tests that contamination with certain metals should have adverse effects on seedling germination of salt marsh vegetation and thereby potentially contribute to loss of coastal wetlands (Waddell and Krauss, 1990). However, no such effects have been shown in situ around the North Sea (Huiskes and Rozema, 1988; Fletcher et al., 1994).

Varekamp et al. (1992) found that the geochemistry of salt marsh sediments is governed by the supply of inorganic clastic sediments, the in situ accretion rate of organic material, diffusion to porewater and diagenetic processes and in many cases also by atmospheric inputs. Many metals exhibit strong colloidal tendencies towards the organic rich clay and silt fractions due to these fractions large particulate surface areas, surface charge and cation exchange capacities (Horowitz, 1985). Fine grained sediments, thus, serve as the major source of metals to salt marshes (Valette-Silver, 1993). Therefore, changes in grain-size with time

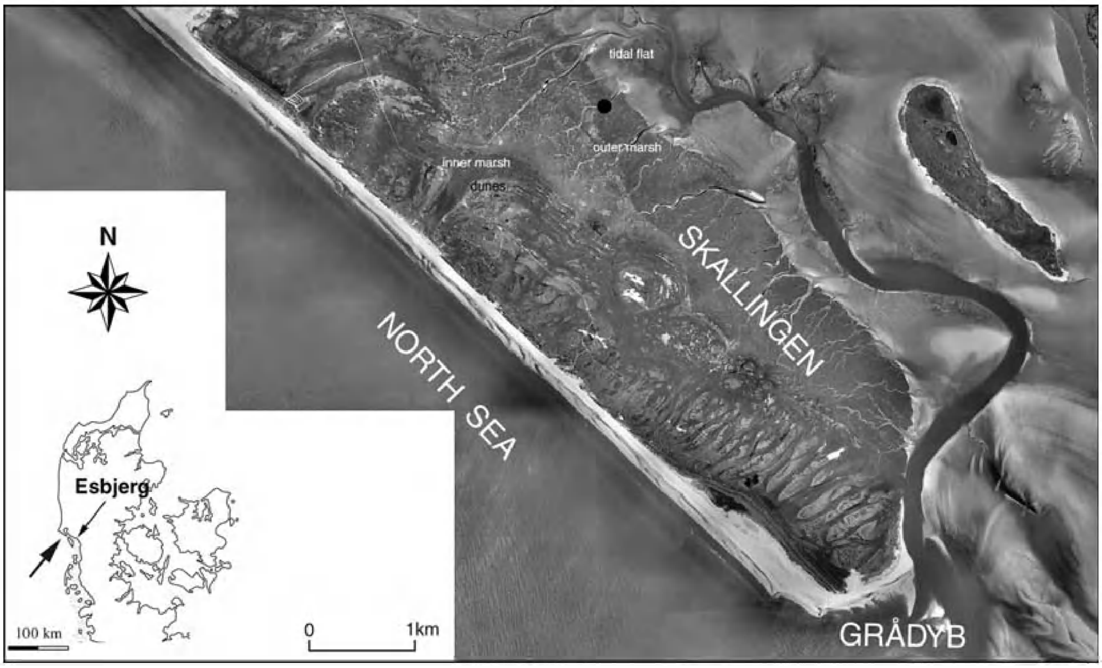

Figure 1. Locational map showing position of sampling station (●) on the Skallingen barrier spit. Inserted map show the location (arrow) of the Skallingen barrier spit in the SW Denmark.

may strongly affect the analytical data. Consequently, several procedures are suggested in the literature for correcting this grain size effect (Ackermann, 1980; Zwolsman et al., 1993).

The aim of the present study is to reveal natural and anthropogenic induced variations in time in metal accumulation rates on the outer lagoonal part of the back barrier salt marsh of the Skallingen peninsula, Denmark. Temporal and spatial variations in accretion rates since salt marsh vegetation invaded the back barrier part of the peninsula in the beginning of the 20th century were also studied. However, these results are not included here but will be published elsewhere (Bartholdy et al., personal communications).

Study area

The Skallingen salt marsh is one of the biggest undiked salt marsh areas in Europe. It covers an area of 31 km^2 and is located on the east (lagoonal) side of the Skallingen barrier spit (Figure 1). The Skallingen salt marsh is young. It started to develop in the beginning of the 20th century when dikes were built between dunes along the shorelines to the west in order to prevent overwash activity (Aagaard et al., 1995). The sampling locality is situated in the outer lagoonal part of the salt marsh at a present height of 1 m above DNN (Danish Ordnance Datum). According to Nielsen and Nielsen (personal communication) this means that the locality is flooded 9% of the year. The sampling plot was covered by a 2 mm thick layer of Sudan-red colored sand in 1931 (Nielsen, 1935). This layer is still visible to the naked eye in a vertical profile through the salt marsh deposits and serves as a marker horizon (Figure 2). Vegetation in the sampling area is made up by *Halimione portulacoides*, *Limonium vulgare*, *Puccinellia maritima*, *Aster tripolium*, *Plantago maritima* and *Artemisia maritima*.

The area is micro tidal with a tidal range of 1.5 m ranging from 1.3 m at neap tide to 1.7 m at spring tide. Meteorological induced sea-level variations are much stronger and sea-levels may reach 4.4 m above DNN during storm surges. The number of storm surges has increased during the last 30 years due to increased storminess (Aagaard et al., 1995). Mean sea-level in the area has been rising with a rate of 1.3 mm a^{-1} since the first tide gauge in Esbjerg was established in 1888. Sedimentation on the Skallingen marsh is variable both in time and in space. But, generally the long-term (1931–1999) rate is 2–3 mm a^{-1} (Christiansen et al., 2001). Thus, yearly accretion rates exceed mean sea-level rise.

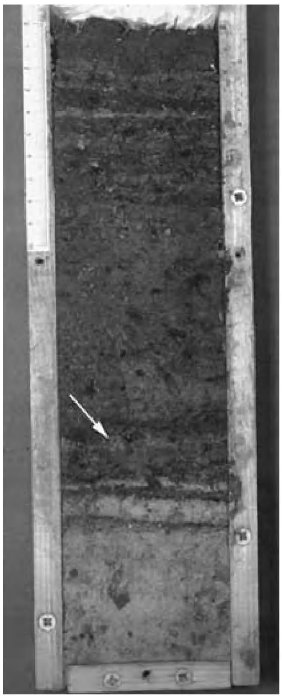

Figure 2. Block cut out of the salt marsh showing the (white) sandy sediment and the red-coloured marker horizon (arrow) underlaying the (dark) salt marsh sediments.

About 85% of the fine-grained sediments deposited in the back-barrier area has been estimated to be imported from the North Sea (Bartholdy and Madsen, 1985). Accumulation rates of Hg and Pb on the Skallingen salt marsh were studied by Madsen (1981). The rates correlated well with consumption in the countries surrounding the North Sea and were estimated to be only marginally influenced by local sources and atmospheric deposition. Import of suspended matter from the North Sea to the Wadden Sea generally takes place during storm surges (Jakobsen, 1961). New evidences, however, suggest that strong import is restricted to strong windy periods following long periods of low energy conditions in the North Sea. During such low energy periods fine-grained material may settle to the bottom and thereby act as a reservoir for import to the Wadden Sea area when resuspended during high energy episodes (Bartholdy and Anthony, 1998).

Methods

Following the method described by Stoodley (1998) a 25 cm high monolithic profile of the saltmarsh sediment was collected using a 10 cm × 6.8 cm casing. This sampling procedure was chosen in order to avoid compaction during sampling. Milan et al. (1995) observed that even if cores are short and of large diameter, sampling of young marsh sediments with cores may result in compaction of up to 25%.

The sediment in the casing was cut in 1 cm slices and subjected to a number of analyses. Bulk density was calculated by dividing the dry weight of the slices by the wet volume of the slices. Grain-size distributions were found using traditional sieving and pipette technique. Content of organic matter was determined as loss on ignition at 500 °C. The temperature used for combustion is sufficient to oxidize all organic matter, but also to drive off hygroscopic water and convert pyrite to ferric oxide. According to Ball (1964) these two latter losses can be assumed to be minimal. Total organic carbon was determined on a Dohrmann DC-190 analyzer. Uncertainty on duplicates of chemical analyzes were $<5\%$. Further, each core slice was screened for 17 selected elements, listed in Table 1, using EDX (Energy Dispersive X-ray fluorescence analysis) (Kunzendorf, 1979). Principal component analysis (SAS© System) was carried out in order to facilitate interpretation and identify key groupings of the elements.

Sediment accumulation rates were determined from ^{210}Pb and ^{137}Cs dating of the profile using low-level gamma-spectrometry with a method described elsewhere (Kunzendorf et al., 1998). The historical profiles were constructed using physical core data (density or porosity) and the constant rate of supply (CRS) model for unsupported ^{210}Pb (Robbins, 1978). Net deposition since 1931 was further measured as sediment accumulation on top of the Sudan-red colored marker horizon of sand placed on the marsh in 1931 (Nielsen, 1935). The two methods gave similar results indicating that compaction of the thin and young salt marsh layers is insignificant (Bartholdy et al., personal communication).

Results and discussion

Sediment accumulation

Bartholdy et al. (personal communication) observed two ^{137}Cs peaks in the present core. A 1987 peak (Chernobyl disaster) was located at 2.5 cm depth and a 1963 peak (atomic weapon tests in the atmosphere) at 7.5 cm depth. The levels of these marker horizons

Table 1. Correlation matrix of elements, organic matter (ORG) and size fractions. Highly significant correlations (p <0.0001) are shown with bold font.

	K	CA	TI	MN	FE	V	CR	CO	NI	CU	ZN	BR	RB	SR	Y	ZR	NB	ORG	MIN63	MIN20
K	**1.00**																			
Ca	0.37	**1.00**																		
Ti	**0.92**	0.36	**1.00**																	
Mn	**0.74**	0.09	**0.81**	**1.00**																
Fe	**0.90**	0.20	**0.92**	**0.88**	**1.00**															
V	**0.64**	0.23	**0.79**	**0.71**	**0.80**	**1.00**														
Cr	**0.72**	0.13	0.70	**0.74**	**0.75**	0.59	**1.00**													
Co	**0.89**	0.22	**0.90**	**0.87**	**0.96**	**0.77**	**0.78**	**1.00**												
Ni	**0.81**	0.19	**0.83**	**0.86**	**0.87**	0.70	0.64	**0.87**	**1.00**											
Cu	**0.77**	0.10	**0.83**	**0.93**	**0.90**	**0.74**	0.63	**0.86**	**0.88**	**1.00**										
Zn	**0.91**	0.34	**0.92**	**0.87**	**0.97**	**0.79**	**0.72**	**0.94**	**0.86**	**0.89**	**1.00**									
Br	**0.74**	0.22	**0.80**	**0.88**	**0.84**	0.69	0.69	**0.84**	**0.85**	**0.89**	**0.86**	**1.00**								
Rb	**0.80**	0.18	**0.88**	**0.78**	**0.92**	**0.79**	0.65	**0.87**	**0.77**	**0.83**	**0.92**	**0.81**	**1.00**							
Sr	**0.75**	0.40	**0.87**	**0.82**	**0.82**	**0.74**	**0.74**	**0.84**	**0.74**	**0.82**	**0.84**	**0.90**	**0.83**	**1.00**						
Y	0.59	0.22	**0.72**	0.70	**0.77**	**0.79**	0.66	**0.73**	0.64	**0.72**	**0.77**	**0.78**	**0.82**	**0.83**	**1.00**					
Zr	0.68	0.39	**0.83**	0.58	0.68	**0.74**	0.57	**0.72**	0.59	0.60	0.66	0.62	0.69	**0.81**	0.64	**1.00**				
Nb	0.38	–0.01	0.43	0.49	0.41	0.40	0.56	0.45	0.36	0.32	0.36	0.46	0.41	0.50	0.45	0.54	**1.00**			
ORG	**0.79**	0.30	**0.86**	**0.91**	0.88	**0.75**	**0.77**	**0.90**	**0.83**	**0.88**	**0.90**	**0.97**	**0.85**	**0.94**	**0.82**	**0.73**	0.51	**1.00**		
<63 μm	**0.86**	0.15	**0.93**	**0.87**	0.96	**0.82**	**0.72**	**0.95**	**0.86**	**0.87**	**0.94**	**0.83**	**0.94**	**0.83**	**0.79**	**0.77**	0.49	**0.88**	**1.00**	
<20 μm	**0.84**	0.09	**0.88**	**0.91**	0.97	**0.79**	**0.73**	**0.94**	**0.89**	**0.91**	**0.96**	**0.88**	**0.94**	**0.81**	**0.79**	0.63	0.44	**0.90**	**0.97**	**1.00**

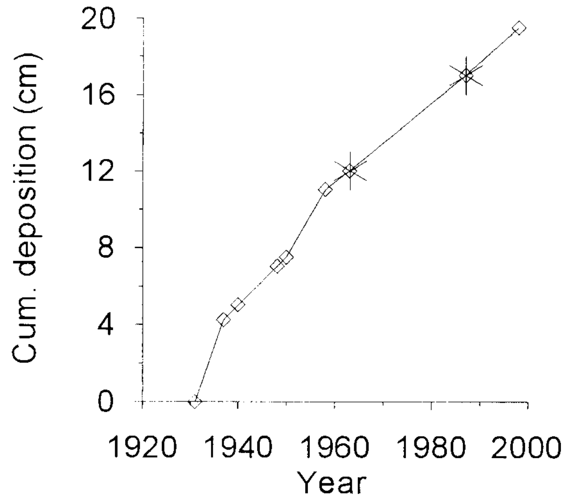

Figure 3. Age vs. depth. From measurements of depth to marker horizon (◇) and from positions of (^{210}Pb) dated ^{137}Cs peaks in the core (*).

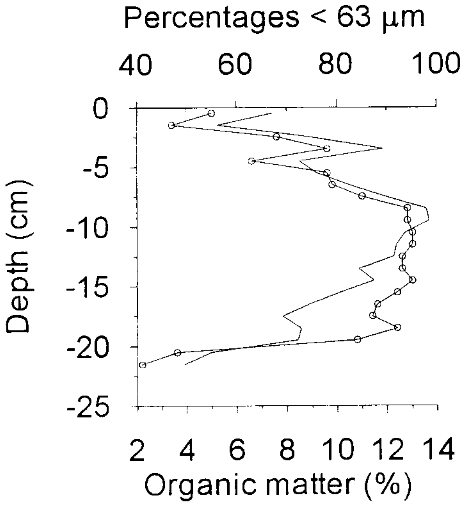

Figure 4. Downcore changes in percentages of sediment < 63 μm (○) and organic matter (loss on ignition) content.

are plotted in Figure 3 together with measurements of depth from the marsh surface to the colored sand layer placed on the marsh surface in 1931. Such measurements were carried out regularly up to 1958 (Nielsen, 1935; B. Jakobsen, pers. com.). Figure 3 indicates a sligth decreasing trend in accretion rates (relatively high at 7–8 mm a^{-1} during the first years and 4 mm a^{-1} on average up to 1958). Since 1958 the rate has been 2 mm a^{-1} on average. These findings are very much in line with more areal wide results (2–3 mm a^{-1}) from repeated levellings, spaced 25 years in time, across the Skallingen salt marsh (Nielsen and Nielsen, personal communication). Figure 3 is used to relate individual layers in the core to a certain year.

Figure 3 shows that the average accumulation rate on the sampling plot are higher than the long-term 1.3 mm y^{-1} raise in mean sea-level and indicates in agreement with the general model set up by Allen (1990) that accumulation on the Skallingen marsh diminishes with increasing level on the marsh and thereby potentially lesser hours of inundation per time unit. However, because of changing wind conditions with more storm surges per year (Christiansen et al., 2001) the number of hours per year with inundation of the sampling plot has gone up from 900 to 1200 during the last 25 years (Nielsen and Nielsen, personal communication). An alternative reason for lower accumulations rates in recent years, therefore, may be found in increasing distance from the sampling plot to the intertidal flats. Christiansen et al. (2001) showed 1) that long term sedimentation rates on the Skallingen salt marsh generally were independent on distance to the nearest creek, but diminished with increasing distance from the intertidal flats and 2) that vegetation in recent years has spread out on the intertidal flat with a rate of about 5 m per year. This has made the distance from the present sample area to the intertidal flat about 150 m longer since 1964.

Morphological changes

Evaluated from existing aerial photos the distance from the sampling plot to the nearest creek was 14 m in 1945. Due to beginning creek meandering towards the sampling plot this distance started to diminish and was 13 m in 1955 (the distance was in the field measured to be 12.7 m in 1958 (Jakobsen, 1965)) and 11 m in 1964. From 1964 the meandering tendency accelerated and the distance was 7 m in 1980 and in 1998 the creek had started to erode parts of the sampling plot. In recent years the sampling plot sediments, therefore, accumulated in the form of deposits similar to levee deposits although no clear levee can be seen in the topography. Most probably, creek meandering takes place too rapidly to allow time for levee deposition to show up in the morphology.

The effect of the creek meandering tendency with time is clearly reflected in the sediment on the sampling plot. Figure 4 shows that the sediment may be divided into three lithostratigrahic units. Sand beneath the 1931 marker horizon has a mean grain-size ranging from 137μm to 90μm, an organic content of

2–4%, and a dry bulk density of 1.2–1.3 g cm^{-3}. Invasion of salt marsh vegetation caused a much finer (mean grain-size = 2μm – 12μm) sediment with a high (11–14%) organic matter content and a low (0.5–0.7 g cm^{-3}) dry bulk density. From 1964 onwards, when the creek meandered towards the sampling plot, the deposits again turned coarser (mean size = 26μm – 90μm) with lower (8–5%) organic matter content and higher (0.8–1.1 g cm^{-3}) dry bulk density. An exception is observed at the 3–4 cm depth level (the mid 1980'ies) which has a finer size distribution. This short period of time was characterized by a relatively low-energy wind regime (Aagaard et al., 1995) and consequently caused no major floodings of the salt marsh with supply of coarse inorganic matter. Generally, storm surges have increased in recent years both in number and in heights. This may also help to explain the general coarsening upward tendency since 1964.

The particle size distribution of the sediment accumulating on the salt marsh surface to-day is very similar to the size distribution in the sand beneath the salt marsh deposits (Figure 5) suggesting that erosion from the creek bottom during storm surge situations and inundation of the salt marsh make up a major part of the source material for near creek deposition on the salt marsh. Creek meandering in the Skallingen salt marsh thus has effects on sediment characteristics but apparently not on sedimentation rates.

Element associations

Associated elements that influence the variance in the total element population through time may be derived from principal component analysis (Siegel et al., 1995). Figure 6 shows the factor-loadings for the elements, organic matter and grain size fractions in the first 4 principal components. These 4 components together account for 90% of the downcore variance.

Factor 1: Explains 76% of the variance. Strong positive loadings can be seen with elements such as Ti, Mn, Fe, Co, Ni, Cu, Zn, Rb and Sr. There is also strong positive loadings from organic matter and the clay fraction. Not unexpectedly this shows that there is a tendency for heavy metal enrichment in very fine grained deposits with a high organic matter content. Elements belonging to this factor should therefore be corrected for grain size effects before their long term development in anthropogenic load to the area can be evaluated form concentration profiles with depth in the salt marsh deposits.

Factor 2: Explains 6% of the variance. In factor 2 the highest positive loadings are observed with Ca, K, Sr and organic matter. This factor is interpreted to represent the biogenic contribution to the variance and to have the strongest independency of grain-size variations.

Factor 3 and 4: Explain 8% of the variance (5% and 3% respectively). In factor 3 the highest positive loadings are observed with Y, Nb, Cr and Zr together with the fine silt fraction. In factor 4 K, Cr, Ti and Nb dominate together with the coarse silt fraction. These two factors together are interpreted to represent the contribution of heavy minerals to the total variance.

In agreement with the principal component analysis, table 1 shows that some of the elements (Ca, Cr, Sr, Y and Nb) are best correlated to the organic matter content whereas others (K, Ti, V, Co and Cr) are best correlated to the < 63 μm fractions and still others (Mn, Fe, Ni, Cu, Zn and Rb) are best correlated to the < 2 μm fractions. The findings in Table 1 of a high correlation between the fine-grained fraction of the samples and Rb corroborate observations (Ackermann, 1980) that in lack of grain-size information, Rb may be used as a proxy for fine-grained sediments. Lowest correlations between Rb concentrations and other metal concentrations were found with Ca, V, Y, Nb and Zr.

A detailed examination (not shown) of correlations between grain-size, organic matter content and element concentrations showed that with the present material generally the best correlations were obtained between element concentrations on one side and the sum of percentages organic matter content plus percentages finer than 20 μm on the other side. This sum will be used to eliminate grain-size effects on metal concentrations.

Changes in element concentrations with time

There are two pre-requisites which should be met in order to use sediment profiles of heavy minerals as evidences for the historical development in anthropogenic heavy metal load. Firstly, chronology should be intact. There were peaks in ^{137}Cs activity in layers which were dated by the ^{210}Pb method to 1963 and late 1980s. These layers coincide with peak deposition of airborne ^{137}Cs from atom weapon tests in the atmosphere and from the Chernobyl disaster, respectively. Their correct location in the stratigraphic column seems to indicate that reworking is insignificant on this locality. This observation corroborates

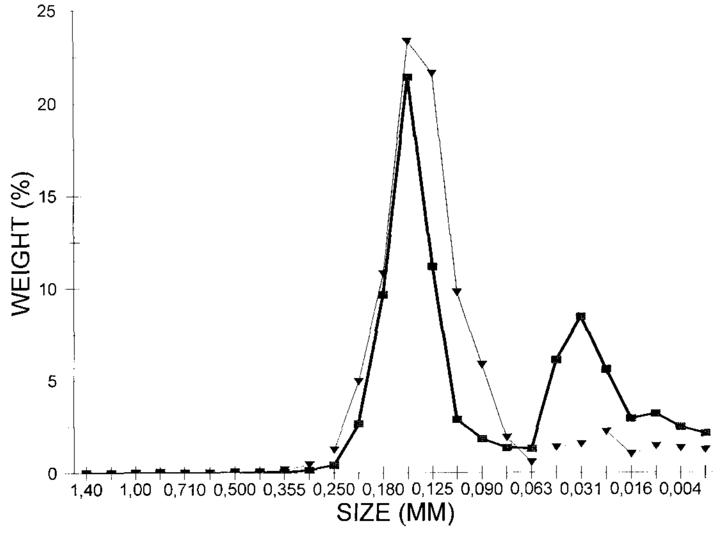

Figure 5. Grain-size distributions of sediments from below the marsh deposits (▼) and from recent surface sediments (■).

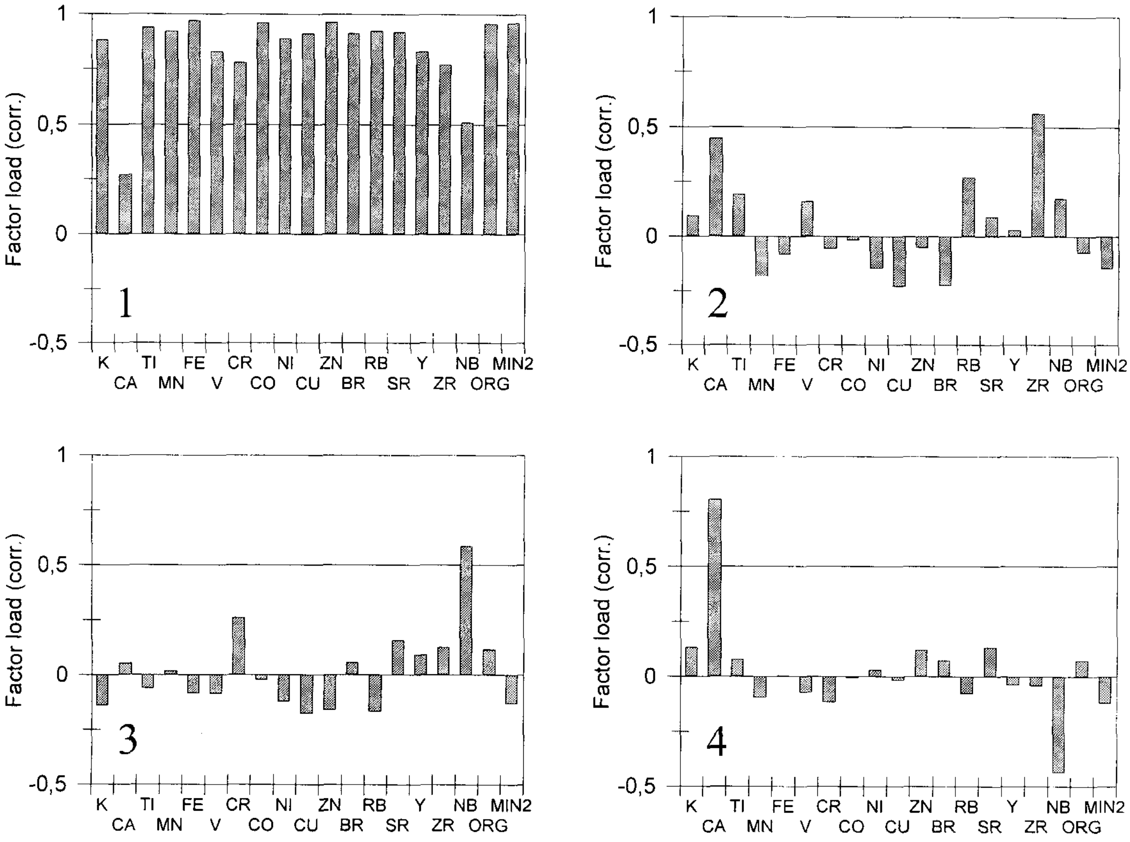

Figure 6. Factor load of the first 4 principal components (On the x-axis ‘org’ means organic matter and ‘min2’ is percentages of grain-size distribution < 2μm).

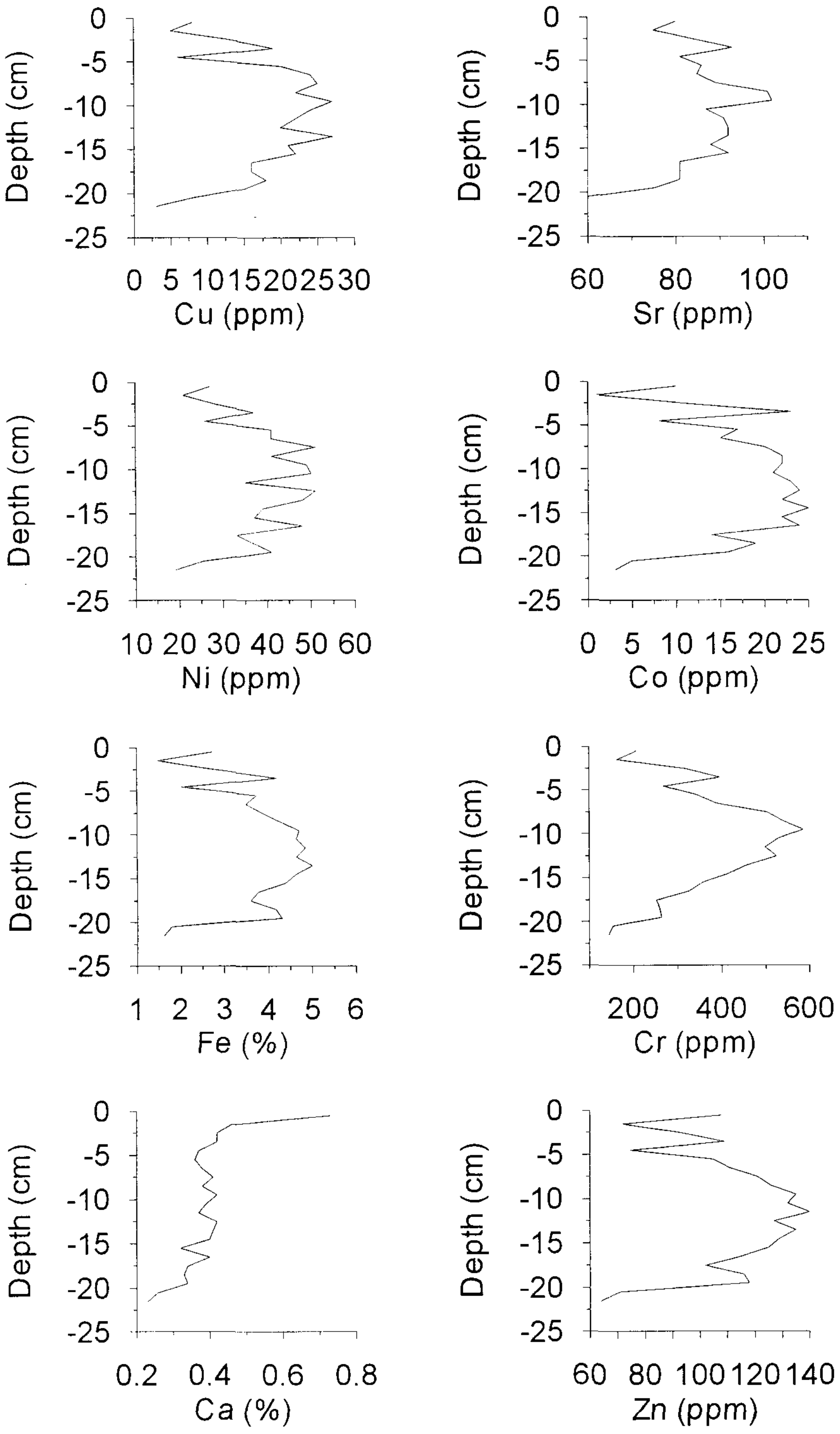

Figure 7. Changes with depth in element concentrations. For comparison is also shown changes with depth in the sum (percentages organic matter + percentages smaller than 20 μm). Note the different scales on the x-axis.

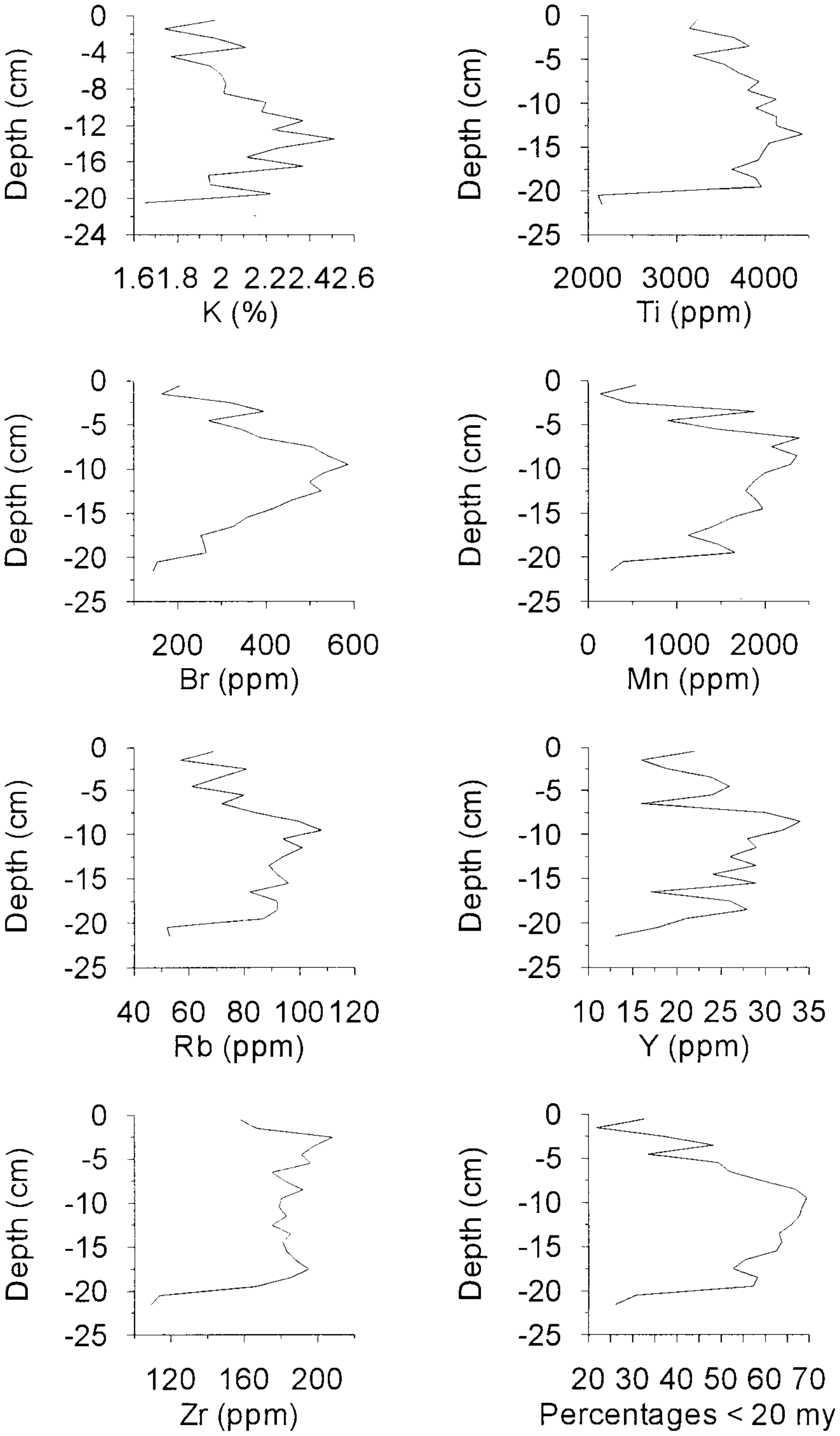

Figure 7. Continued.

findings that salt marsh sediments seldom are strongly affected by bioturbation (Stumpf, 1983). Dating results also indicate that no erosional hiatus exists and that metal accumulation may be considered to be continuous since the red sand was placed on the 1931 surface. Secondly, the metals should stay stabilized in the sediment. This may not be the case in salt marshes with a distinct oxic sediment layer preventing precipitation of metal sulfides from the pore water (Hines et al., 1989). However, heavy metals are strongly adsorbed by organic, clay and ferromanganese oxyhydroxide colloids and many of them form insoluble crystalline compounds with low mobility (Cacador et al., 1996). In spite of that diagenetic processes may modify original metal distributions, a number of authors eg. Zwolsman et al. (1993) have been able to identify anthropogenic maxima for a number of metals in salt marsh sediment. Similarly, Madsen (1981) showed that Pb and Hg content in a dated core from another part of the Skallingen salt marsh very closely reflected consumption of these metals in NW Europe. This may indicate that these two metals are not strongly mobilized in the Skallingen sediment after deposition.

Net sediment accretion has been 2–3 mm a^{-1}. Using sediment slices of 1 cm thickness, means that changes in element concentration with time can not be resolved with a better resolution than about 3–5 years. Figure 7 shows that changes on most element concentrations with time are very similar to changes in the fine (< 20 μm) fraction. Synchronous with the fining upward period starting shortly before the red-sand layer was spread out in 1931 (19.5 cm depth) a sharp increase in element concentrations can be observed and the concentrations are generally doubled after 20–30 years when they reach their maximum in the 1950' in the 10–15 cm depth interval. There are significant drops in concentrations from about 1963 (7.5 cm depth) coincident with the coarsening upward tendency because of the morphological changes in the area. Only Ca does not follow such general trends. The concentration of this element appears to be independent of the change with time in size distribution and increases slightly with time and strongly in the uppermost 2.5 cm.

When adjusted for grain-size effects, the changes with time generally appear very different (Figure 8). Only few elements (e.g Cu) still show a decline in concentrations since 1963. Concentrations of some elements (e.g. Ni, Fe, and Cr) in the fine grained fractions and organic matter only show weak trends or have stayed near to constant throughout the years. There is now a clear increase in the normalized Zn content. The two opposite trends for Cu and Zn between 1970 and 1980 were also noted in the area in cores covering the time up to 1980 (Ribe Amtskommune, 1986). The concentrations of K, Ti, Rb, Zr and Sr in the fine fractions also show strong increases since about 1963. These concentrations are now much higher than in the fine fractions of the sand below the 1931 red-sand layer (19.5 cm depth). The reason for the increase in these mainly 'terrestrial/ feldspar' elements is unknown. As the material depositing on the Skallingen salt marsh for far the greater part is imported from the North Sea (Bartholdy and Madsen, 1985) it is possible that the increased storminess since 1970 from southwesterly directions has supplied the area with material from other sources.

Conclusion

The initial increase up to about 1960 in metal concentrations and the following significant decrease for most of the them since then could, without further proof, easily be misinterpreted to be results of better waste water treatment in the area. When corrected for grain-size effects many metal concentrations in the fine fractions have, however, stayed nearly constant while some (e.g. Zn) concentrations opposite to this still increase.

The observed effects of morphological changes (tidal creek meandering) on metal accumulation thus corroborates findings from the Severn Estuary, UK (French, 1996), that knowledge of both socio-economic activities and system hydrodynamics is important in the successful interpretation of metal/depth trends.

Similar needs for corrections of grain-size effects on metal enhancement with time may be expected near eroding salt marsh edges. Kastler and Wiberg (1996) observed upward coarsening in a core from an eroding edge and explained this by increased exposure to wave and current activity.

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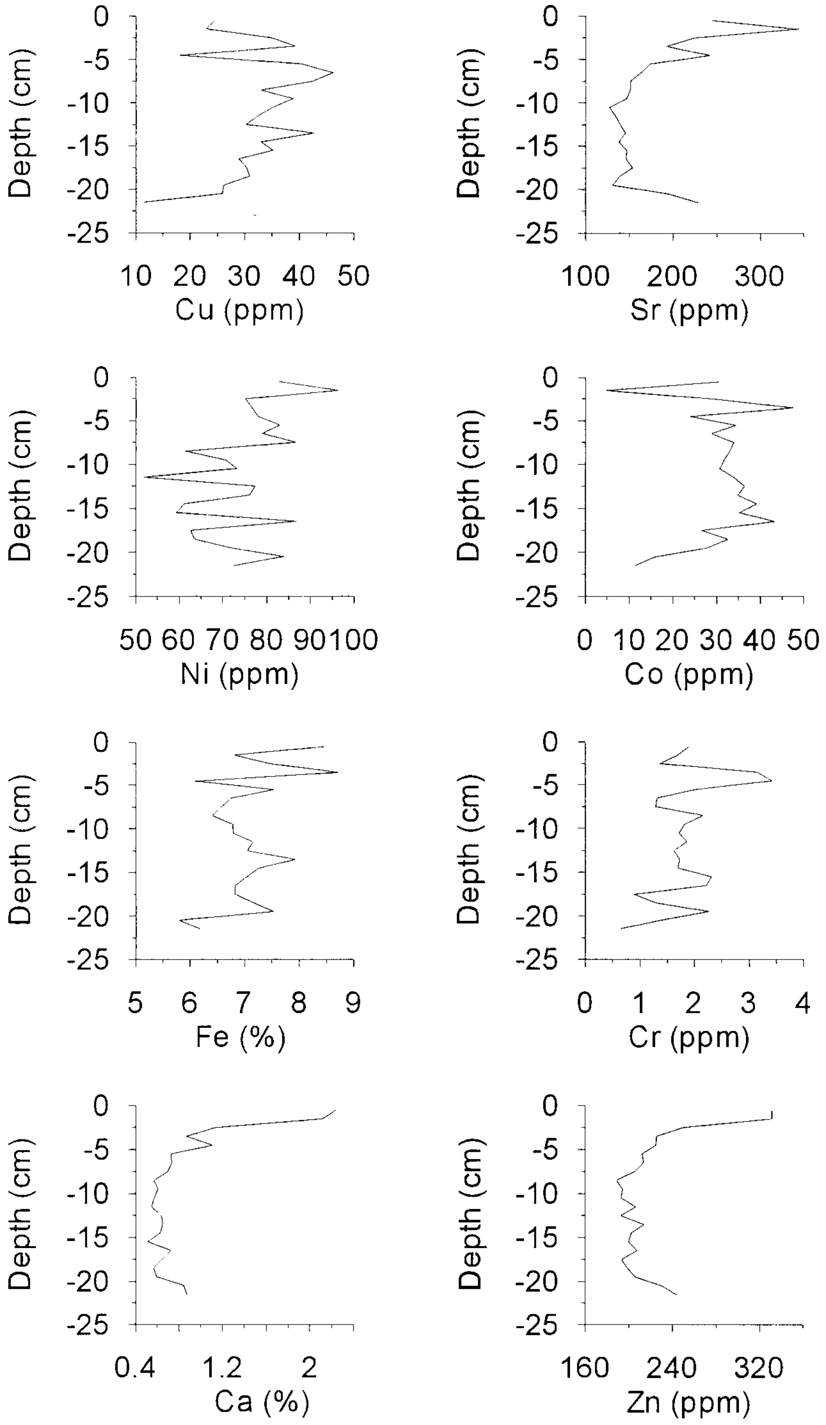

Figure 8. Changes with depth in element concentrations normalized with the sum (percentages organic matter + percentages smaller than 20 μm). Note the different scales on the x-axis.

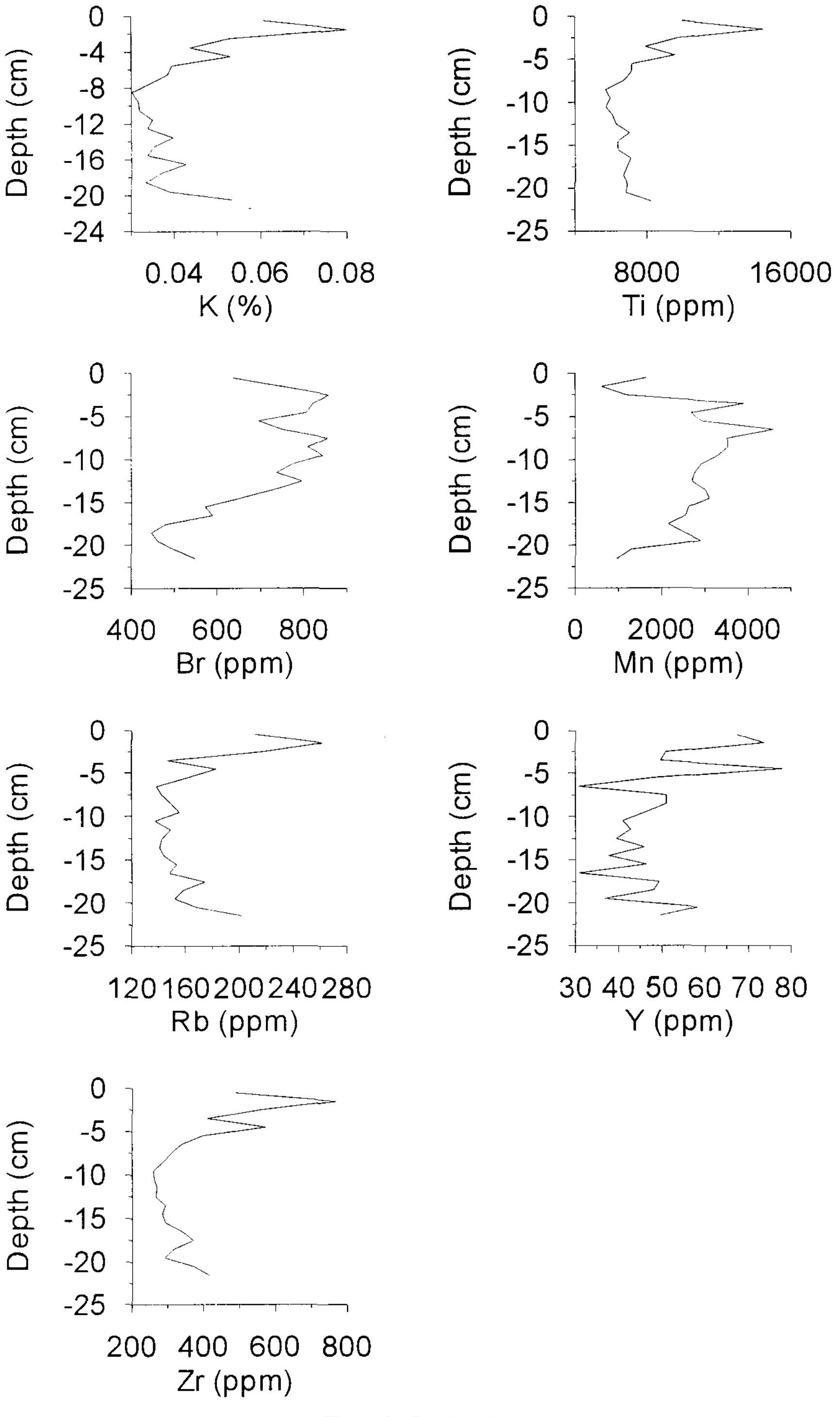

Figure 8. Continued.

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