

PRE-CIVILIZATIONAL AND CIVILIZATIONAL LAYERS IN TWO SEDIMENT CORES FROM THE WESTERN BALTIC SEA

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Summary. Two sediment cores were taken in the Eckernförder and Geltinger Bights (approx. 20 m water depth) using a vibrohammer corer device on board the R/V "Senckenberg". A total of 115 samples obtained from the two cores were analyzed for grain size, organic carbon, carbonate, heavy metal and aluminium contents. Organic-rich (2.5-8.7% Org. C.) and carbonate-poor (less than 1 to 9.4% CaCO_3) mud sediments from the upper core layers (post-1880 sediments) showed metal enrichment in Cu, Pb, Zn, Cd, and Hg by factors of between 1.4 and 15, compared to those in pre-1880 layers (pre-civilizational background). The presence of coal particles, strong and positive correlations, and high enrichment factors, all suggest cultural metal enrichment for Cu, Pb, Zn, Cd and Hg in the upper, post-1880 core layers, for which the increased fossil-fuel (mainly coal) combustion contemporaneous with industrialization in the western Baltic Sea region appeared to be generally responsible. Mn and Co (to some extent), and Ni, Cr, and Fe concentrations seemed to be largely controlled by lithogenic and post-depositional influences.

Riassunto. Nelle Baie di Eckernförder e Geltinger, Mar Baltico occidentale, sono state prelevate due carote alla profondità approssimativa di 20 metri, tramite un carotiere a percussione in dotazione della N/O "Senckenberg". Su un totale di 115 campioni prelevati dalle due carote sono state eseguite analisi granulometriche ed analisi chimiche per la definizione del contenuto in carbonio organico, carbonati, metalli pesanti ed alluminio. I sedimenti fangosi caratterizzati da alti valori in carbonio organico (2.5 ÷ 8.7%) e minori contenuti di carbonati (da < 1 a 9.4% di CaCO_3) hanno evidenziato un arricchimento di metalli pesanti (Cu, Pb, Zn, Cd e Hg) nei livelli più recenti delle carote, con un fattore di incremento variabile da 1.4 a 15 rispetto ai livelli precedenti al 1880 (livello preindustriale). La presenza di particelle carboniose, le evidenti correlazioni positive e l'alto fattore di arricchimento suggeriscono che l'incremento dei metalli pesanti nei livelli post-1880 sia dovuto alle attività industriali, fra le quali l'aumento dell'utilizzo di combustibile fossile (carbone) in relazione allo sviluppo industriale del Mar Baltico occidentale sembra essere la causa principale. La distribuzione di Mn, Co, Ni, Cr e Fe appare invece largamente controllata da fattori litogenici e postdeposizionali.

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1. Introduction

It has been shown by numerous investigations that heavy metals in recent marine sediments can generally be divided into two categories in accordance with their predominant source - "lithogenic" or "anthropogenic". These categories are often simply referred to as "geochemical" (= pre-civilizational) and "man-made" (= civilizational), respectively. The latter has increasingly become of interest in the highly urbanized and industrialized coastal areas of the western Baltic Sea. For example, Erlenkeusser et al. (1974) and Müller et al. (1980) found elevated heavy metal concentrations for Zn, Pb, Cd, Cu, Ni, and Co due to increased human activity in this region. However, Hartman (1964) and Djafari (1977) showed indications of heavy metal enrichment in the surficial sediments as ferromanganese concretions as result of redox changes and syndiagenetic processes in the depositional environment, respectively.

This paper was presented at the National Symposium on the Environment '88 held in June 1988 at Izmir, Turkey (Ergin, 1988a). In the present work, a large number of heavy metal data together with several other chemical and physical parameters are discussed in order to understand the lithogenesis, diagenesis and anthropogenesis in the two temporary anoxic basins of the Western Baltic Sea.

2. Materials and methods

During an ocean cruise of the R/V Senckenberg in 1978 in the Baltic Sea, two vibroham-

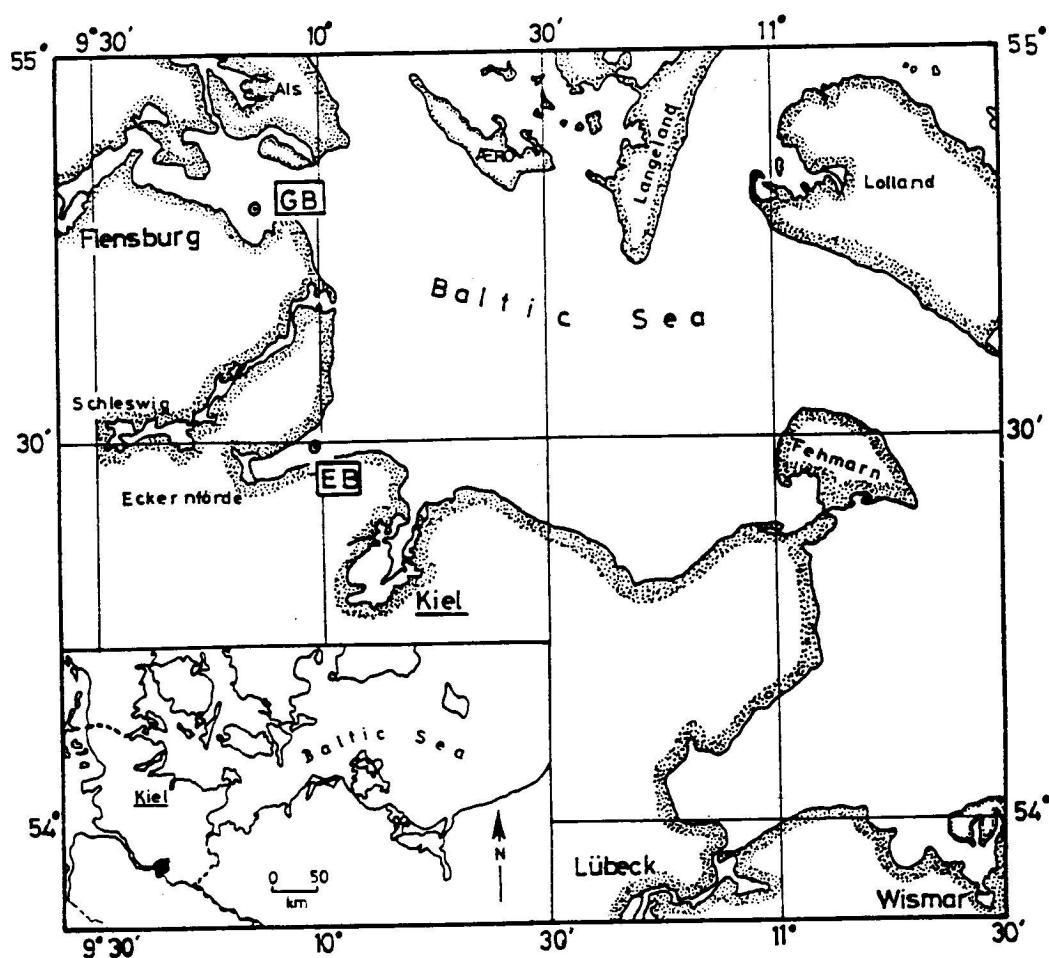


Fig. 1 - Locations of the studied cores EB and GB.

mer cores (of 202 and 258 cm sediment thicknesses) were taken from the relatively deeper parts (20 m water depth) of the Eckernförder and Geltinger Bights in the western Baltic Sea. Sampling locations in the study area are shown in Fig. 1.

Onboard the ship, the sediment cores were split into 2 cm intervals and stored in plastic bags and frozen. A total of 115 sediment samples was air-dried and used for heavy metal and organic carbon determinations.

The upper 40 cm sections of the cores were dated by the Lead-210 technique. For determination of the heavy metals, Fe, Zn, Cr, Ni, Cu, Pb, Co, and Cd, a 0.5 gr dried ground bulk sample was digested with conc. HNO_3 , and the measurements done by standard Atomic Absorption Spectrometry (AAS) procedures, Hg was analyzed separately according to the procedures described by Hatch and Otto (1968).

Total organic carbon was determined gaschromatically with a microcombustion CHN + O/S Elemental Analyzer. Control for all chemical analyses was carried out with known reference sediments ("Rhine" and "Estuary").

3. Results and discussion

Data from the analytical results for the two cores (Core EB = Eckernförder Bight; Core GB = Geltinger Bight) is presented in Figs. 2, 3, 4, and 5 and Tables 1, 2, 3, and 4. Detailed information on the mineralogy, grain size, carbonates, and major elemental geochemistry of

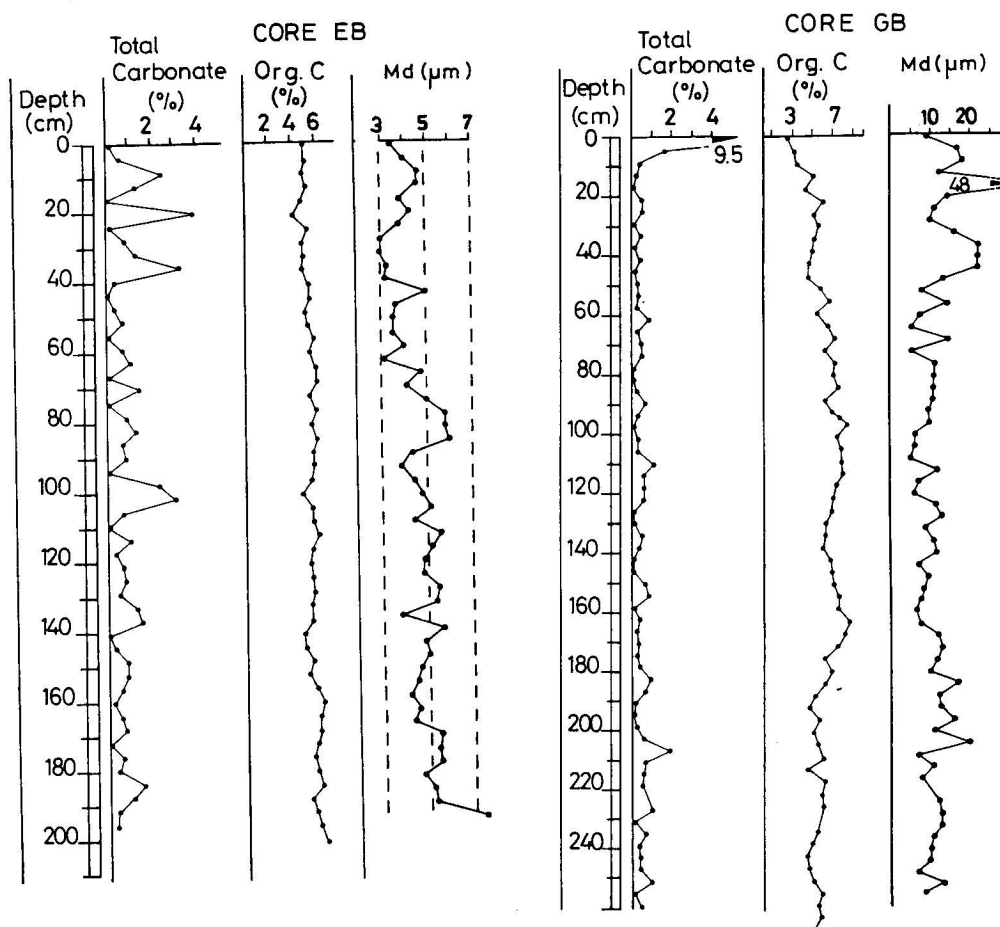


Fig. 2 - Distributions of carbonate and organic carbon contents as well as median grain size in the cores EB and GB. Data for carbonate and median values are taken from Ergin (1988b).

sediments in this study can be found elsewhere (Ergin, 1988b).

3.1. Sediment composition

Compared to the clayey-silt sediments of core EB, the sediments of core GB are usually clayey-sandy silt. Mean grain-size profiles of the two cores are given in Fig. 2 (after Ergin, 1988b). On the average, sediments are composed of 50 and 57% quartz, 32 and 21% clay minerals, 17% feldspars, plus others in core EB and GB, respectively (Ergin, 1988b).

Total carbonate contents of the sediments ranged from less than 1 to 3,8% (mean = 0,8%) in core EB, and from less than 1 to 9,4% (mean = 0,5%) in core GB (Fig. 2; Ergin, 1988b). High carbonate contents were generally confined to upper core layers containing mostly biogenic materials (mollusc shells, planktonic remnants etc.).

Total organic carbon contents of the sediments are remarkably high, ranging from 4,2 to 6,6% in core EB and 2,5 to 8,7% in core GB (Fig. 2). Such high amounts of organic carbon are common in Baltic Sea sediments (Manheim, 1961; Suess and Erlenkussner, 1975; Brüggemann et al., 1980), and can satisfactorily be attributed to the primary productivity (Kühlmorgen-Hille, 1965) and sedimentation rate (3,2 mm/year in core EB and 2,3 mm/year in core GB; Dominik, pers. commun., 1980) which are both relatively high in the study area. In addition to naturally occurring organic materials of marine and terrestrial origin, domestic and indu-

Table 1 - Heavy metal concentrations of the sediments in cores EB and GB compared to average shale. Average shale values are from Turekian and Wedepohl (1961). Enrichment factors (EF) are calculated by comparing the metal data from surface (0-2 cm) and subsurface (= background, or pre-1880 sediments, respectively) layers.

	Range	Mean	S. Dev.	Surface	Background	EF
Fe (%)						
Core EB	2.70-3.66	2.85	0.13	3.66	2.90+0.24	1.26
Core GB	1.50-2.85	2.10	0.24	2.70	2.30+0.50	1.17
Shales					4.7	
Mn (ppm)						
Core EB	468-1562	790	198	544	780+195	0.7
Core GB	100-375	200	65	350	210+122	1.66
Shales					850	
Cr (ppm)						
Core EB	30-49	32	3	42	33+3	1.3
Core GB	14-30	23	2	30	23+5	1.3
Shales					90	
Co (ppm)						
Core EB	7.5-16.2	11.8	2	8.5	11.8+4.3	0.7
Core GB	4.7-10.5	7.2	1.4	8.2	7.6+2.9	1.1
Shales					19	
Ni (ppm)						
Core EB	23-43	32	4	42	29+6	1.4
Core GB	26-44	31	3	44	28+10	1.6
Shales					68	
Cu (ppm)						
Core EB	20-44	26	4	44	25+5	1.8
Core GB	17-34	22	3	30	22+4	1.4
Shales					45	
Zn (ppm)						
Core EB	87-285	110	41	285	100+12	2.9
Core GB	38-172	66	25	160	63+24	2.5
Shales					95	
Pb (ppm)						
Core EB	35-107	49	16	107	44+9	2.4
Core GB	18-62	25	9	62	24+6	2.6
Shales					20	
Cd (ppm)						
Core EB	0.16-1.72	0.52	0.32	1.72	0.41+0.10	4.2
Core GB	0.39-2.98	0.84	0.41	2.21	0.79+0.29	7.6
Shales					0.30	
Hg (ppm)						
Core EB	0.05-0.76	0.30	-	0.76	0.05+0.02	15
Core GB	0.05-0.24	0.14	-	0.18	0.05+0.02	3.5
Shales					0.4	
Al ₂ O ₃ (%)						
Core EB	9.91-12.37	10.80	0.65	11.08	10.85+0.9	1.0
Core GB	7.70-10.86	8.65	0.49	10.86	8.55+0.7	1.3
Shales					16.7 (8.0 Al)	

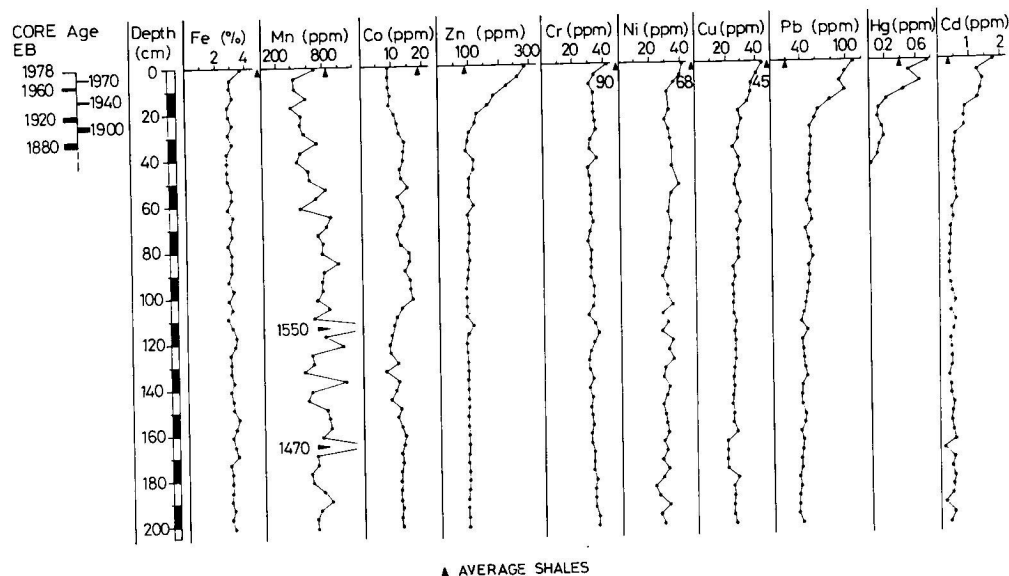


Fig 3 - HNO_3 - soluble heavy metal concentrations in the core EB. Average shale values are from Turekian and Wedepohl (1961). Upper core intervals were dated by the Lead-210 method after Dominik (pers. comm., see also Müller et al., 1980).

strial waste waters must also be considered as further sources delivering high amounts of organic substances to the sediments (Kändler, 1963; Horstman, 1972).

3.2. ^{210}Pb Age of the cores

In order to obtain a time scale for the younger deposits, the upper 40 cm sections were dated by the Pb-210 method (Dominik, pers. commun., 1980), and the corresponding ages are illustrated in Figs. 3 and 4. The Pb-210 determination were also made on surficial sediments (see Müller et al., 1980) taken separately from the coring stations of this study.

The Pb-210 dating revealed that the upper 25 cm section of core GB, and the upper 35 cm section of core EB must have been deposited during the last 100 years approximately, after about 1880 (i.e. post-industrial revolution times). Hence, the pre-1880 sediment layers and related heavy metal concentrations presented here are referred to as "pre-civilizational" and those above the 1880-layers of the cores are defined as "civilizational" (Figs. 3 and 4).

3.3. Data from pre-civilizational layers

As shown in Fig. 3 and 4, heavy metal concentrations in pre-1880 sediments generally

Table 2 - Metal to aluminium ratios in the surface (0-2 cm) and subsurface (= background) layers of the studied cores along with those found in average shale for comparison. Values are $\times 10^{-5}$

	Core EB Surface	Core EB Background	Core GB Surface	Core GB Background	Average Shale
Fe/Al	70000	67000	54000	58000	59000
Mn/Al	920	1300	770	660	1000
Co/Al	14	20	14	16	23
Cr/Al	71	57	52	52	110
Ni/Al	71	50	76	60	85
Cu/Al	75	42	52	47	56
Zn/Al	486	174	278	138	118
Pb/Al	182	76	108	53	25
Cd/Al	2.9	0.7	3.8	1.7	0.4
Hg/Al	1.3	0.1	0.3	0.1	3.1

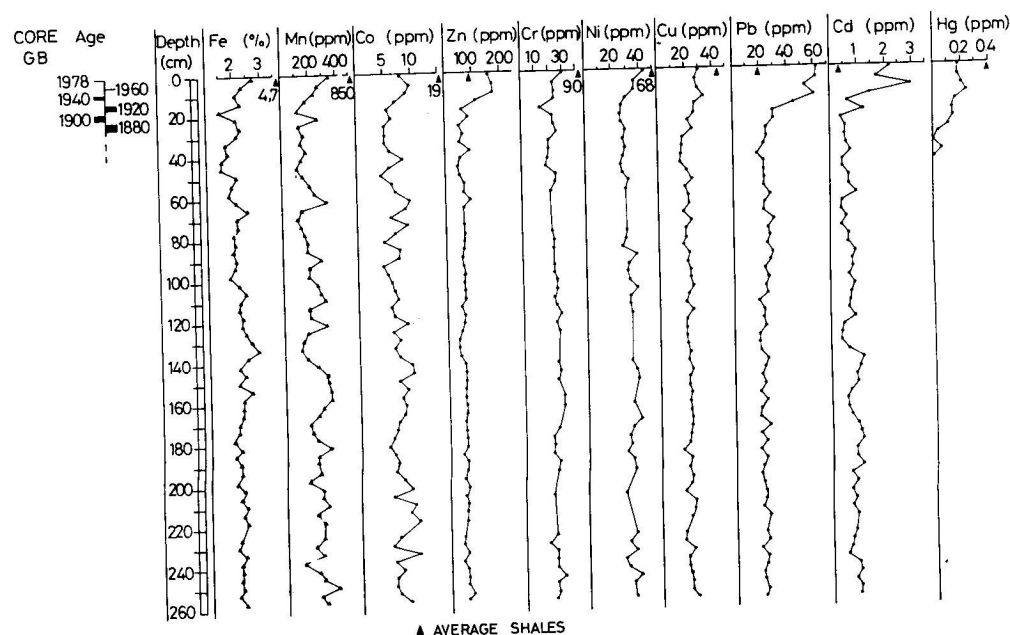


Fig. 4 - As in Fig. 3, but for the core GB.

have uniform values, which are simply referred to as "geochemical background". However, these background values, except for Cd and Pb, were lower in core GB than in core EB. This can be understood best in terms of compositional variations in the two cores; for example, overall high metal contents were confined to finer-grained sediments of core EB, where the presence of relatively high abundances of clay must be considered; a fact that is often referred to as the "grain-size effect".

A comparison of the background values in the two cores with the average shale composition as a global standard value indicated differences in the heavy metal contents (Table 1; Figs. 3 and 4), probably due to considerable variations in the relative abundances of lithogenic admixtures dominated by the aluminosilicates, oxides/hydroxides, such as clay minerals, feldspars, micas, quartz etc. That is why the metal to aluminium ratios between the average shale and the pre-1880 sediments (= background) of the studied cores were somewhat different (Table 2).

3.4. Enrichments of Hg, Cd, Zn, Pb and Cu in civilizational layers

As shown in Figs. 3 and 4, in surface/near-surface layers (post-1880 sediments) most of

Table 3 - Enrichment factors for heavy metals in the studied core. Values are calculated from a comparison of the metal/aluminium ratios between the surface (0-2 cm) and subsurface (= background) core layers. Numbers in parenthesis are taken from Table 1 for comparison.

	Surface/Background (Core EB)	Surface/Background (Core GB)
Fe	1.0 (1.2)	0.9 (1.2)
Mn	0.7 (0.7)	1.1 (1.6)
Co	0.7 (0.7)	0.8 (1.9)
Cr	1.2 (1.3)	1.0 (1.3)
Ni	1.4 (1.4)	1.2 (1.6)
Cu	1.7 (1.8)	1.1 (1.4)
Zn	2.8 (2.8)	2.0 (2.5)
Pb	2.4 (2.4)	2.0 (2.5)
Cd	4.1 (4.2)	2.2 (7.6)
Hg	16 (15)	3 (3.5)

Table 4 - Correlation coefficient matrices for the contents of measured parameters obtained from cores EB (above) and GB (below). Numbers in italics show negative correlations.

	Fe	Mn	Co	Zn	Cr	Ni	Cu	Pb	Hg	Cd	Clay	Al ₂ O ₃	Org.C	CaCO ₃
Fe	1.00	0.26	<i>0.11</i>	0.21	0.06	0.01	0.18	0.21	0.20	0.20	0.07	<i>0.02</i>	0.04	<i>0.20</i>
Mn		1.00	0.14	<i>0.37</i>	<i>0.25</i>	<i>0.35</i>	<i>0.43</i>	<i>0.41</i>	<i>0.42</i>	<i>0.41</i>	<i>0.26</i>	<i>0.28</i>	<i>0.52</i>	<i>0.10</i>
Co			1.00	<i>0.43</i>	<i>0.29</i>	<i>0.09</i>	<i>0.41</i>	<i>0.34</i>	<i>0.44</i>	<i>0.49</i>	0.11	<i>0.08</i>	0.21	0.10
Zn				1.00	0.44	0.45	0.86	0.90	0.92	0.88	0.26	0.41	<i>0.37</i>	0.01
Cr					1.00	0.23	0.47	0.45	0.41	0.57	0.18	0.23	<i>0.21</i>	<i>0.17</i>
Ni						1.00	0.57	0.52	0.41	0.42	0.43	0.25	<i>0.43</i>	<i>0.01</i>
Cu							1.00	0.91	0.87	0.88	0.30	0.61	<i>0.42</i>	0.01
Pb								1.00	0.92	0.91	0.36	0.61	<i>0.51</i>	0.12
Hg									1.00	0.93	0.37	0.49	<i>0.46</i>	0.14
Cd										1.00	0.36	0.52	<i>0.51</i>	0.10
Clay											1.00	0.27	<i>0.49</i>	0.23
Al ₂ O ₃												1.00	0.31	0.13
Org. C													1.00	<i>0.50</i>
CaCO ₃														1.00

Core EB: n = 51

r = 0.27; significance level = 5%

r = 0.36; significance level = 1%

r = 0.44; significance level = 0.1%

	Fe	Mn	Co	Zn	Cr	Ni	Cu	Pb	Hg	Cd	Clay	Al ₂ O ₃	Org. C	CaCO ₃
Fe	1.00	0.20	0.38	0.25	0.065	0.53	0.48	0.25	0.18	0.22	0.35	0.69	0.05	0.37
Mn		1.00	0.30	0.40	0.39	0.60	0.21	0.06	0.03	0.46	0.35	0.26	0.06	0.38
Co			1.00	0.36	0.26	0.51	0.34	0.16	0.04	0.33	0.37	0.24	0.02	0.14
Zn				1.00	0.24	0.49	0.67	0.83	0.70	0.87	0.02	0.52	0.44	0.52
Cr					1.00	0.63	0.34	0.05	0.03	0.22	0.40	0.49	0.36	0.31
Ni						1.00	0.43	0.27	0.06	0.53	0.39	0.53	0.02	0.49
Cu							1.00	0.68	0.66	0.66	0.14	0.56	0.16	0.31
Pb								1.00	0.87	0.68	0.12	0.48	0.46	0.50
Hg									1.00	0.60	0.28	0.38	0.53	0.34
Cd										1.00	0.05	0.43	0.42	0.45
Clay											1.00	0.32	0.43	0.05
Al ₂ O ₃												1.00	0.29	0.62
Org. C													1.00	0.40
CaCO ₃														1.00

Core GB: n = 61

r = 0.24; significance level = 5%

r = 0.32; significance level = 1%

r = 0.40; significance level = 0.1%

the heavy metals increased sharply in concentration, and maximum metal levels were also found there. From a comparison of the metal data from pre-and post-1880 sediment layers lying below and above the 25-35 cm depths, it has been found that the concentrations of Hg, Cd, Zn, Pb and Cu are enriched in the post-1880 layers by factors of between 15 and 1.4. In core EB, the highest metal concentration occurred in the 0-2 cm layer deposited between 1978 and 1975; cadmium (x4.2) next to mercury (x15) showed the greatest enrichment factor, followed by zinc (x2.9), lead (2.4), and copper (x1.8). By contrast, metal enrichments in core GB were considerably lower and the maximum metal levels for Hg, Cd, Zn, and Cu were confined to the 8-18 cm layers deposited during 1940-1920, while Pb seems to be strongly accumulated in the 0-6 cm layers, corresponding to 1975-1940. In these post-1880 layers of core GB, Cd, Hg, Pb, Zn and Cu reached their maximum concentrations, being 7.6; 3.5; 2.6; 2.5 and 1.4 times the metal background values respectively.

These metal enrichment factors are also strongly supported by the data from a comparison of metal to aluminium ratios between the pre-1880 (= background) and surface layers (Table 3).

As shown in Table 4, the strongest positive correlations occurred among the contents of Hg, Zn, Cd, Pb and Cu at the highest significance levels (0.1%; = 99.9); the correlation coef-

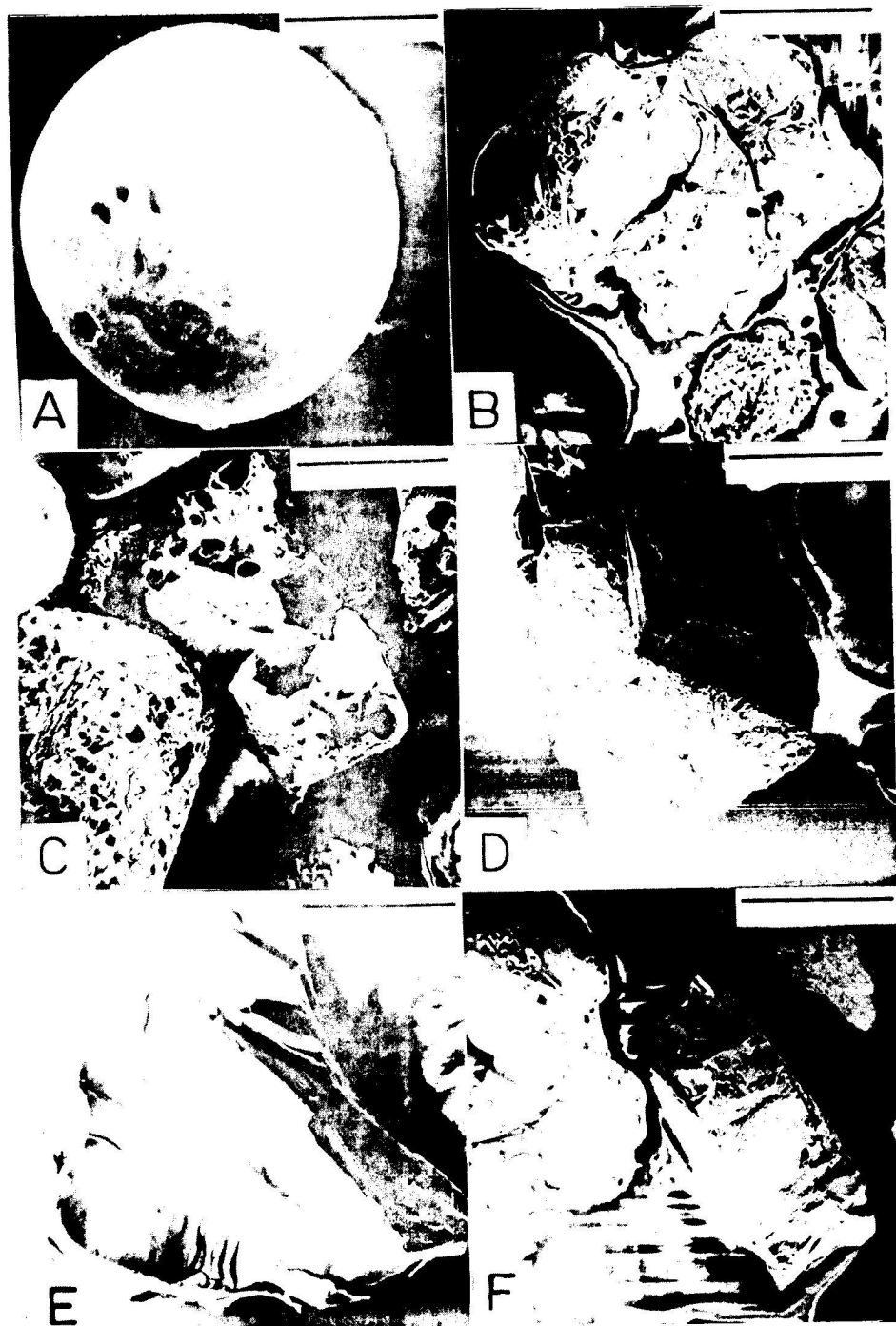


Fig. 5 - SEM-photographs showing anthropogenic coal and slag particles in sand-fractions (larger than 63 micron) of sediments of the cores EB and GB. (A) A slag sphere rich in iron, Core GB, Sample 16-18 cm, Scale bar = 0.2 mm. (B) Aggregated slag particles rich in iron, Core GB, Sample 16-18 cm, Scale bar = 2mm. (C) Slag particles displaying irregular and porous appearance, Core GB, Sample 16-18 cm, Scale bar = 1 mm. (D) A coal particle, Core GB, Sample 18-20 cm, Scale bar = 1 mm. (E) A lignite particle, Core EB, Sample 18-20 cm, Scale bar = 0.4 mm. (F) A charcoal particle (at right) and aggregated metallic slag (at left). Core GB, Sample 16-18 cm, Scale bar = 2 mm.

ficients, r , for the above metals were greater than 0.866 in core EB, and 0.603 in core GB respectively. Such a close relationship between the metals, Zn, Pb, Cu, and Hg was also reported by Suess and Erlenkeusser (1975) in the Baltic Sea, and by Förstner and Reineck (1974) in the North Sea sediments. According to Suess and Erlenkeusser (1975), enrichment of Cd, Pb, Zn, and Cu probably originated from the increased burning of fossil-fuel (e.g. coal) due to rapid industrialization in the Baltic Sea region. As previously reported by Bertine and Goldberg (1971), the burning of fossil fuels can lead to the enrichment of Cu, Zn, Pb, and Cd by factors of 30 to 217 in the coal ash particles. These ideas are also in good agreement with petrographical studies of the sediments, where coal particles occur in appreciable amounts in the upper 20 cm layers of both cores (Fig. 5).

It is believed therefore that the high positive correlations, the presence of coal particles, the high enrichment factors greater than 2, all suggest that the elevated concentrations of Zn, Cd, Pb, Hg and Cu in the post-1880 layers of the cores must have been derived largely from fossil-fuel combustion (mostly coal) in the vicinity of the study area.

3.5. Ni, Cr, Mn, Fe and Co in post-1880 sediments

Compared to the aforementioned heavy metals (Cd, Hg, Zn, Pb and Cu), the concentrations of Ni, Cr, Mn, Fe and Co in the upper core layers, or post-1880 sediments appeared to be less indicative of anthropogenic metal enrichment. Here, these metals revealed 1.1 - to 1.6 - fold increases in their concentrations (Tables 1 and 3), which can be best explained by lithogenic variations in the admixtures of sediments, as well as by diagenetic processes in the depositional environment, rather than an anthropogenic input.

In the case of manganese and cobalt in core EB, the post-1880 layers were marked by upward decreasing metal levels resulting in enrichment factors of less than 1, namely 0.7. This may suggest metal remobilization from the surface/near-surface layers, most probably due to changes in the redox conditions. This point of view is also supported by Djafari (1977) who found unusually high manganese concentrations in the deep water of the Kiel and Eckernförder Bays as a result of anoxic conditions that rise above the sediment-water interface, especially during the summer.

4. Conclusions

On the basis of data obtained from two sediment cores from the Eckernförder and Geltinger Bights in the western Baltic Sea, the following conclusions can be drawn:

- 1) Organic-rich and carbonate-poor mud sediments displayed metal concentrations which was due, more or less, to cultural activity in addition to the natural influxes in this region.
- 2) By a comparison of the metal concentration data from the upper (post-1880 sediments) and lower (pre-1880 sediments = background) core layers, it has been found that Hg, Cd, Zn, Pb and Cu are enriched in the upper core layers by factors of between 15 to 1.4. The presence of coal particles, highest and positive correlations, and high enrichment factors, all suggest that the elevated concentrations of Hg, Cd, Zn, Pb and Cu can be satisfactorily explained by the increased combustion of fossil-fuel (coal) contemporaneous with industrialization in the western Baltic Sea region.
- 3) Ni, Cr, Fe, Mn, and Co concentrations in the upper core layers indicated lithogenic and diagenetic influences to be the main factors controlling the distribution of these metals.
- 4) It has also been inferred that the average shale composition cannot normally be taken as a comparative basis for the metal background concentrations in the study area.

Acknowledgements. This present work is part of a comprehensive investigation of the mineralogy, petrology and geochemistry of sediments from two cores carried out by the author at the Sediment Research Institute of the University of Heidelberg, F.R. Germany, under the supervision of Prof. Dr. German Müller and Prof. Dr. Peter Stoffers to whom I owe great thanks for their helpful and critical discussion. The Konrad Adenauer Stiftung in Bonn, and the Center of International Migration and Development in Frankfurt are acknowledged for financial support. I also extend my thanks to the Master and Crew of the R/V Senckenberg for their help during the sampling.

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