Sedmentation in the Central Baltic Sea as Viewed by Non-Destructive Pb-210-dating

Helmar Kunzendorf, Kay-Christian Emeis & Christian Christiansen

Abstract

A non-destructive gamma-spectrometric method was used to assay the sedimentation in the central Baltic Sea. Three cores from the Gotland Deep with water depths exceeding 230 m showed relatively high Pb-210 and Cs-137 but relatively little variation between cores in the curves for unsupported Pb-210. A constant rate of supply (CRS) modelling of the data revealed that there is a tendency of increasing linear sedimentation rates, from 0.5 to 2 mm a^{-1}, since 1950. With the measured bulk density profiles this is accompanied by increasing sediment accumulation rates (from about 200 to 600 g m^{-2} a^{-1}) There are generally moderately increased Cs-137 values found in the deep and these may mainly be ascribed to the Chernobyl accident.

A core from the central Gdansk Basin taken at a water depth of 117 m shows relatively lower Pb-210 values and a more structured unsupported Pb-210 distribution with depth. A very high activity (about 4000 Bq kg^{-1}) sediment slice (8-9 cm depth) can according to the CRS modelling be ascribed to year 1986. The high activity, however, was found to be confined to a small part of the sediment slice material and therefore this has some bearing on the depositional mechanism of Cs-137 in the sediments.

Keywords

Gamma-spectrometry; dating; sedimentation; Baltic Sea.

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At present, there is a renewed focus on the recent sedimentation history of the deep basins of the Baltic Sea. This is mainly because of the increasing concern as regards the state of pollution in the Baltic, viewed by its historical evolution and the probable actions to be taken in the future. The Baltic Sea is also one of the most well-suited areas for multinational and multidisciplinary research because of the easy access even to the more remote areas and also because of the existence of well-equipped research institutes in the countries surrounding the Baltic Sea. However, for obvious political reasons such a type of research was not possible for many years after World War II and therefore, at present several research projects are being or have already been carried out. For instance, the Gotland Basin Experiment (GOBEX) was conducted during the years 1994-95 (Hagen, 1996) and at present, a large EU project, the Baltic Sea System Study (BASYS) is conducted involving all the countries surrounding the Baltic Sea, with project termination in mid 1999.

An evaluation of the sedimentary history of selected depositional centers in the Baltic Sea, both on a century-millenium time scale (C-14 dating) and on a more detailed scale covering the past 200 years, is required for such research. A major tool for the recent sediments is Pb-210 dating. Most dating efforts of recent Baltic Sea sediments in the past were conducted using alpha-spectrometry of the Pb-210 daughter Po-210, but during the past decade, direct non-destructive gamma-spectrometry of Pb-210 has become available and is increasingly being used in dating of marine sediment cores (e.g., Kunzendorf et al., 1996).

In the present paper we report on the dating of some cores from the Gotland Deep, part of the Gotland Basin, and the Gdansk Basin. The focus is on the interpretation of Pb-210 data from the gamma-spectrometric method where the dating is verified by the simultaneous determination of anthropogenic Cs-137. There is only limited discussion as how to explain the unsupported Pb-210 and Cs-137 activities within a geomorphological frame because this can be found elsewhere (e.g. Christiansen et al. 1997).

Geological setting and methods

Setting

The Baltic Sea is basically an estuarine system fed by rivers of several industrialised countries with connection to the open sea (the North Sea – the North Atlantic) via the Danish Straits, the Kattegat and Skagerrak. The Baltic Sea covers an area of some 400 thousand square kilometers and, apart from some deep basins, has an average water depth of about 50 m. The focus in the present paper is on two of the deep sediment basins of the Baltic Sea: the Gotland Basin with water depths exceeding 200 m, particularly in its northeastern part (Gotland Deep) and the shallower Gdansk Basin (water depths of more than 100 m).

Sediment handling

The cores were recovered during a GOBEX cruise of R/V Poseidon (Cruise 215B) in early March 1996 (Emeis, 1996). The cruise was carried out to mainly gather improved shallow seismic profiles in the basin but it also consisted of a sediment sampling program using multi coring devices and long box corers. The cores of the present investigation were taken with multicoring devices allowing the recovery of up to 8 cores in one coring operation, thereby covering a sediment surface area of about 1.5 m^2. The diameter of the core liners used was 10 cm. Penetration depths of the corers were between 30 to 50 cm. The cores usually contained both bottom water and an intact water/sediment interface. A map displaying the position of the two sampling areas in the Baltic Sea is given in Fig. 1 and core station data are compiled in Table 1.

After transport to the laboratory, the cores were cut into 1 cm thick slices. The cutting of the uppermost very liquid sample material was accomplished by careful removing of 1 cm thick material sections while at deeper levels material was really cut into 1-cm slices from the cores. The sediment sections were then freeze dried and the bulk density of the slices was estimated from the simple formula: bulk density (g/cm^3) = dry weight of the slice (g) / wet volume of the slice (cm^3).

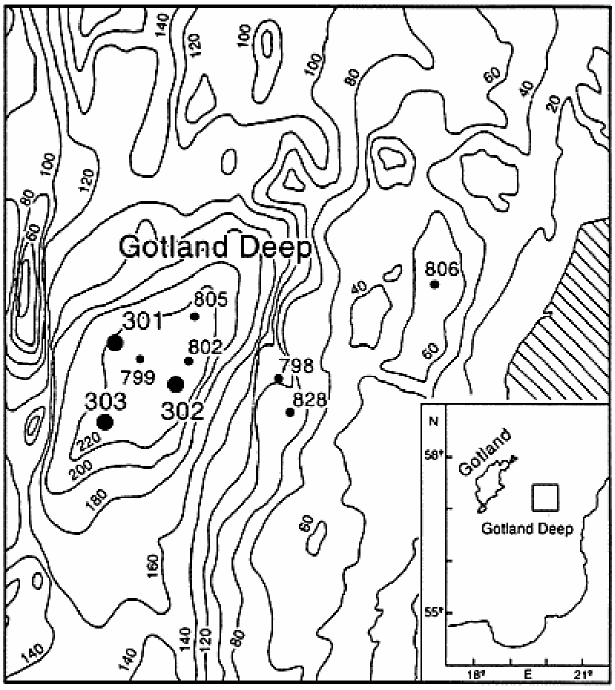

Figure 1: *Coring stations (301, 302 and 303) in the Gotland Deep. Shown are also earlier coring stations. Isolines are in m. Insert position of the two sampling areas.*

Dating technique

Dating of the core material was carried out by non-destructive counting of the Pb-210 gamma-ray activity at 46.5 KeV. The ultra-low background gamma spectrometry systems used consisted of remotely constructed (a long cold finger separates detector and preamplifier) coaxial Ge (Li) detectors with guaranteed rel. efficiencies of 20% The

Table 1: *Coring station characteristics (Emeis, 1996).*

Core	*North*	*East*	*Water depth*
201301-4 (station 301)	57° 20.100'	19° 57.500'	237 m
201302-6 (station 302)	57° 15.280	20° 11.830,	249 m
201303-1 (station303)	57° 11.090'	19° 55.680'	237 m
201308-2 (station 308)	54° 49.870'	19° 18.940'	117 m

detectors were equipped with carbon epoxy windows to reduced absorption of incoming gamma-radiation. Their energy resolutions were guaranteed to less than 850 eV and 1.8 KeV at 122 KeV and 1.3 MeV, respectively. Commercial lead shieldings were used for the detectors to reduce the natural background radiation appreciably. They were constructed of normal and Boliden (old) lead of a total thickness of 10 cm. PC-based multichannel analyser systems were coupled to the spectrometry systems. Count rate calculations were also performed by the PC system.

Five to 20 g of core material was used for gamma-spectrometry each counting lasting about one day. To reduce possible self-absorption effects (Kunzendorf, 1996), in a first approximation, the same amount of sample material was counted throughout a core section. Sample containers were stored for about 3 weeks before counting to allow for re-establishment of secular uranium decay series equilibrium in the sample material having been disturbed during the sample preparation steps.

Calibration of the systems was achieved by using known activities from already analysed Baltic Sea cores. There may be a small relative error in the Pb-210 and Cs-137 data due to the calibration procedure used, but this is of minor importance in the dating procedure because only the depth distribution of activities but not the absolutely correct values are needed for the CRS data modelling.

In the most simple form of the CRS model (e.g., Joshi et al., 1988) the slice age T (in years) at depth z below sediment surface is

$$T = \lambda^{-1} * \ln (A_{total} / A_T)$$

where A_{total} is the total unsupported Pb-210 activity (Bq cm^{-2}) in the sediment core and A_T is the unsupported Pb-210 activity beneath sediments of age T. The decay constant λ for Pb-210 is 0.03114 a^{-1}. The dating therefore depends heavily on the correct estimation of the total unsupported Pb-210 and this in turn requires the systematic measurement of a sufficient number of sediment slices down the core until the unsupported Pb-210 reaches zero values. In practice, for economic reasons, a limited number of measurements can only be carried out for a given sediment core. The total activity along the entire core is then calculated by interpolation between measured slices. The total supported Pb-210 activity (Bq cm^{-2}) calculations are usually done numerically and also require detailed knowledge on the bulk density (g cm^{-3}) or porosity distribution with depth. The numerically integrated or total unsupported Pb-210 activity is then

$$A_{total}\ (\mathrm{Bq\ cm^{-2}}) = \sum_{I=1}^{I=N} m_i(z)\ \Delta z_i\ C_i(z)$$

where $m_i(z)$ is the mass (g cm^{-3}), Δz_i is the thickness (cm) of the sediment slice i and $C_i(z)$ is the unsupported Pb-210 activity (Bq g^{-1}) of the ith sediment slice.

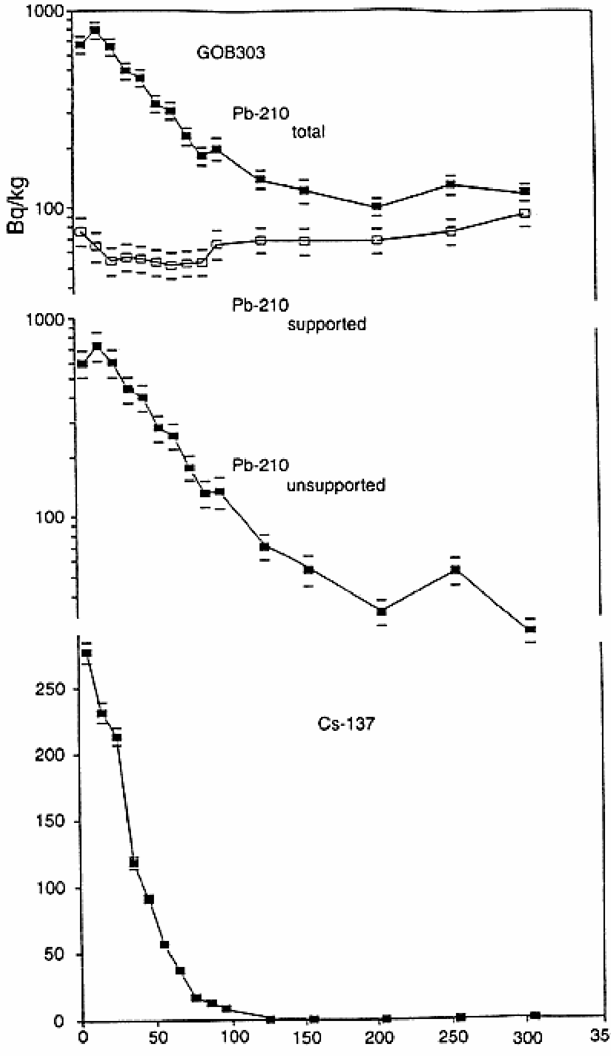

Figure 2: Gamma-spectrometric results for core GOB303, showing total, supported and unsupported Pb-210 activities , and Cs-137 activities with depth in the core. Unsupported activities are obtained by subtracting the supported from the total activity.

Results and discussions

The Gotland Basin

The positions of cores 301, 302 and 303 within the Gotland Deep are shown in Fig. 1. These sites are complementary to sites sampled during an earlier GOBEX cruise conducted in 1994. Data from this study have recently been published (e.g., Neumann et al., 1997; Kunzendorf and Christiansen, 1997).

The sediments of core 301 are grey-olive to olive-grey muds (Emeis, 1996) down to a core depth of about 16 cm showing a distinct darkening towards the sediment surface. The upper 4 cm of the core contained large amounts of water. There is also a distinct lamination of the sediment between about 14 to 16 cm and the sediments then grade into light grey to bluish grey clay below 16 cm. A core depth of 40 cm manganese carbonate layers were observed. Core 302 is a grey-olive to olive grey mud with appearances of slightly visible laminations down to about 68 cm. Gas bubbles were observed especially at a lower depth in a long box core from the same station. Core 302 was also covered with brownish debris on top of a flocculated sediment. The sediments were found to be anoxic.

The evaluation strategy for the measurements is exemplified by the data for core 303, where the total and supported Pb-210 activities, the unsupported Pb-210 activities (supported Pb-210 subtracted from total Pb-210) and the Cs-137 activities with depth in the core are plotted (Fig. 2). Supported Pb-210 activities were estimated directly from the Ra-226 gamma-ray activity or other equivalent radioisotopes, i.e. directly in the same measurement. This is in contrast to the datings by alpha-spectrometry where supported Pb-210 activities have to be estimated by a second analytical step and these values are therefore often not reported. The unsupported Pb-210 and Cs- 137 activities for the other two Gotland Deep cores are shown in Fig. 3. Measured activities are relatively high, which is probably best explained by the fact that very fine-grained sediments (sediment flocs) by their large surface area are able to capture large amounts of very fine Pb-210 and Cs-137 containing material.

The average linear sedimentation rate (in cm a^{-1}), i.e. the rate assuming that there is no compaction throughout the measured core section (constant dry bulk density in g cm^{-3} is then assumed), is calculated according to a Constant Initial Concentration (CIC) model (e.g. Robbins, 1978). The regression line of the unsupported Pb-210 activities (Fig. 2) may be used for this purpose. For core GOB303, a value of 1.6 ± 0.1 mm a^{-1} is calculated. Converting the depth into mass depths (in g cm^{-2}) and using the same calculation strategy yields a mass accumulation rate of 259 $\pm$ 26 g m^{-2} a^{-1}.

These rates are quite often the only cited values in Pb-210 dating applications. However, dating requires the estimation of dates along the sediment. The major tool for Pb-210 dating is use of a Constant Rate of Supply (CRS)

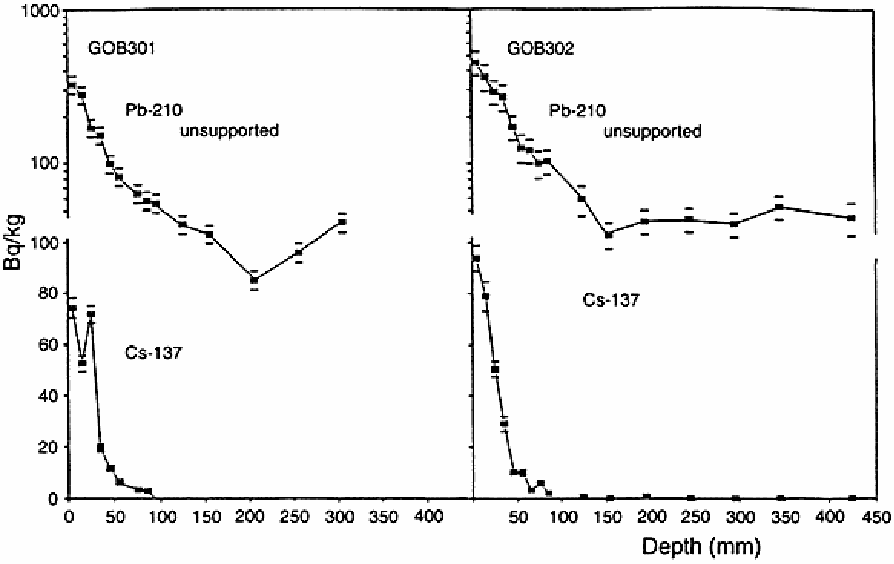

Figure 3: *Cs-137 and unsupported Pb-210 in cores GOB301 and GOB 302.*

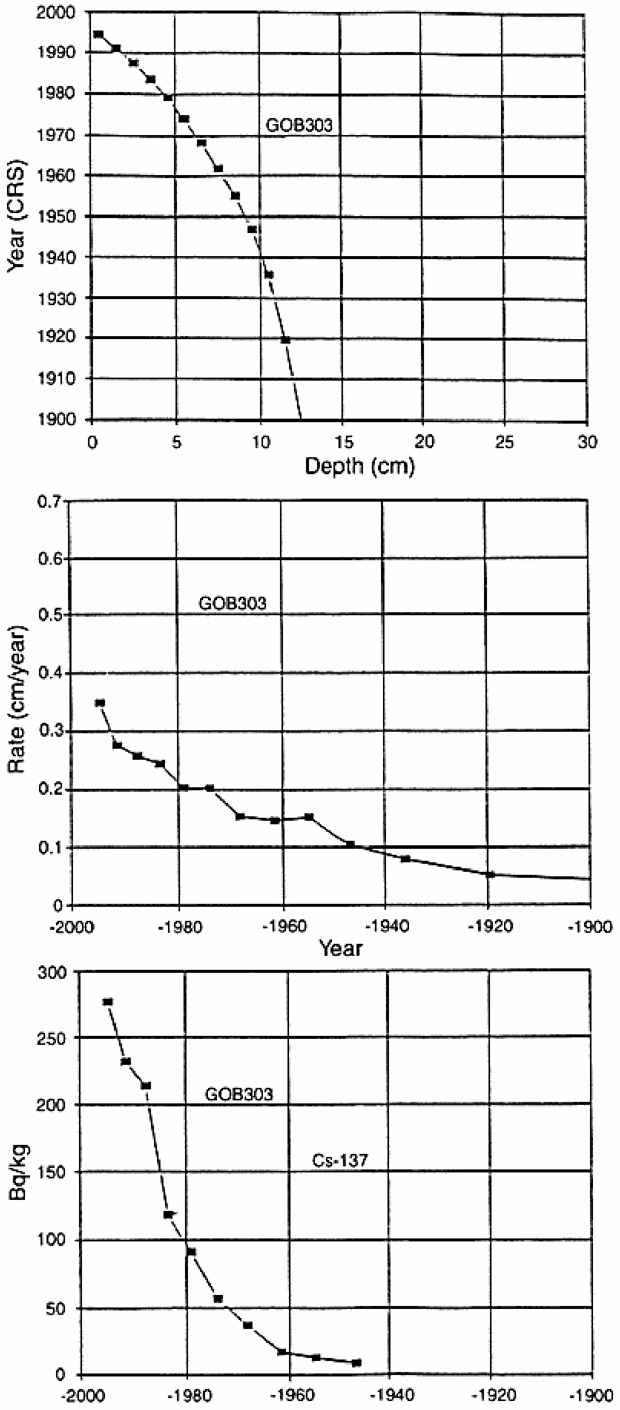

Figure 4: Results of CRS modelling for core GOB303. Sediment accumulation rates not plotted. These may be calculated by multiplying the linear sedimentation rate with the bulk density of the sediment slices.

model (e.g., Robbins, 1978; Joshi, 1986; Appleby and Oldfield, 1992) to the measured data. The CRS model simply assumes that there are varying sedimentation rates along the core. This requires knowledge of the density distribution downcore and the results from this method are therefore, in theory, independent of possible density variations (e.g. compaction) downcore. Bulk densities for the Gotland Deep surface slices are rather low, usually below 0.1 g cm^{-3}, increasing with core depth. Selected results of the CRS modelling for core GOB303 are given in the form of plots in Fig. 4.

As can be seen from the figure, the model is used to estimate the sedimentation rates for each 1-cm sediment slice and it is therefore possible to estimate changes in sedimentation rates during past years. Finally, by plotting dates vs. Cs-137, the dating can be verified because Cs-137 supply to the marine environment in northern Europe has been dominated by 3 significant supply periods and events: atmospheric nuclear bomb testing peaking in 1963, the Chernobyl reactor accident in 1986 and discharge by nuclear fuel reprocessing plants (e.g. Sellafield outlets peaking in the mid-seventies). The data in Fig. 4 show an especially large increase of Cs-137 in the mid-eighties, i.e. material from the Chernobyl accident is deposited in the uppermost core slices. However, from 1960 onwards, an increasing amount of Cs-137 in the core is found and this may be explained by the two other markers mentioned above. Interestingly, the CRS calculations suggest that the sedimentation rates vary and that there are some steps in the sedimentation: relatively low sedimentation with rates below 1 mm a^{-1} before the forties, increase to about 1.5 mm a^{-1} until the seventies, and then a further increase in sedimentation to between 2–3 mm a^{-1} from the seventies onwards. The recent sedimentation rates are 1.5 – 2 times higher than observed nearly three decades ago (Ignatiius et al., 1971). The present findings, however, corroborate the work of Jönsson et al. (1990) using varv counting in recently laminated sediments. Higher or increased sedimentation rates may be explained by a number of factors that work together:

a) The recent eutrophication and higher primary production has caused increased organic matter deposition and this resulted in a generally 1.7-fold increase in sediment organic matter content since the late 1920s (Jönsson and Carmann, 1994).
b) A 16 year long period of bottom water anoxia in recent years (end of seventies to early nineties) may have

resulted in a better preservation of organic matter and authigenic mineral production (Neumann et al., 1997).

c) Shallow water erosion seems to be the major source for deep basin sediments in the Baltic Sea (Jönsson et al., 1990). Increased storminess in recent years (Baerens and Hupfer, 1995) may therefore have caused an increased supply of reworked sediments to the deep basins.

The data reported here were generated from cores taken 1 year after the major GOBEX cruise and they are therefore resembling those described in earlier publications (Kunzendorf and Christiansen, 1997; Christiansen et al., 1997). Coring station 301 refers to the sedimentary conditions in the northwestern part of the basin, with generally more smooth grading of the topgraphical relief compared with the southeastern part of the deep (core 302) with relatively steeper slopes. Accoding to Christiansen et al. (1997), the steeper easternmost part of the deep is responsible for the upper surface sediments being disturbed due to probably winnowing or contourite deposition caused by counterclockwise deep currents around the rim of the Gotland Deep (e.g. Gelumbauskaite and Grigelis, 1996). Consequently it is not surprising that core 302 shows a slightly different Pb-210 profile with generally higher activities although it has a similar Pb-210 depth distribution than core 301. This core also shows higher activities comparable to those of core 303. Core 303 could therefore represent a similar sediment depositional setting than that observed in the southeastern part of the deep. Both these cores also have higher Cs-137 contents in agreement with the fact that the Chernobyl plume transport was from the southeast to the northwest.

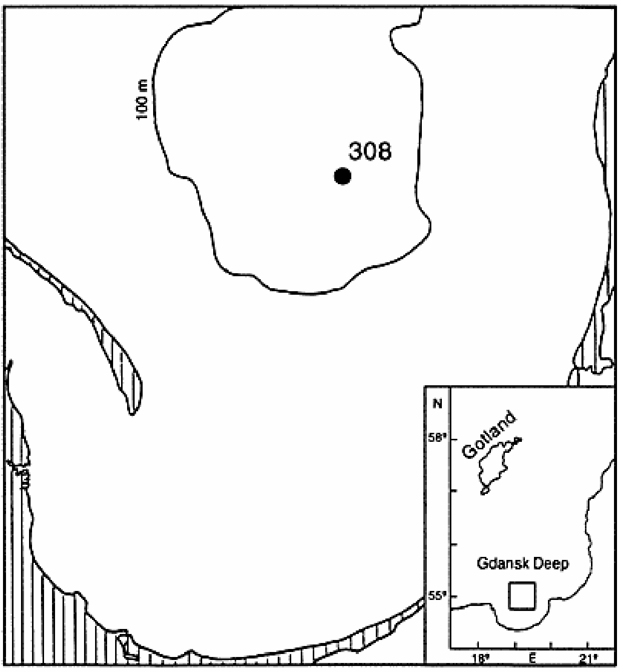

***Figure 5:** Position of coring station GOB308 in the Gdansk Basin.*

The Gdansk Basin

The core taken in the deep part of the Gdansk Basin (Fig. 5) consists mainly of dark grey sandy mud impregnated with sulfides. A thick bacterial mat of up to 2 mm (*beggiatoa)* was observed resting on a black 5 mm thick flocculate. High water contents were measured down to a depth of 60 mm core.

The curves for core GOB308 are shown in Fig. 6. It is clear from the figure that there are significantly higher sedimentation rates in the Gdansk Basin compared with the Gotland Deep. Besides, the curve of unsupported Pb-210 may be divided into several parts, which may express changes in the sedimentation regimes through time, although there is the normal overall exponential decrease of activities with sediment depth. At a sediment depth of 85 mm, a very large Cs-137 peak is observed reaching nearly 4000 Bq/kg. Without applying the CRS modelling we would ascribe this peak to the Chernobyl accident.

Although not as clear as in the Gotland Deep cores, an average linear sedimentation rate may be calculated leading to a value of 4.7 ± 0.7 mm a^{-1}, which is much higher than in most of the Gotland Deep cores. The corresponding value for an average sediment accumulation rate is 966 ± 131 g m^{-2} a^{-1}. The CRS modelling leads to varying very high sedimentation rates (4 to 7 mm a^{-1}) since 1970 while these rates were relatively constant, at 4 mm a^{-1}, from the forties onwards. The plot of Cs-137 vs. date (Fig. 7) simply places the very high peak at 1986, the year of the Chernobyl accident. However, elevated activities are already observed between 1970 and 1980, and an explanation for this could be Sellafield signals either transported from the North Sea during storm events or as aerosols transported directly from the outlet areas.

The plot of unsupported Pb-210 against time (lower part of Fig. 7) suggests that an event-like change of activities is found. These events are confined to the years about 1970 and about 1992. Such minima in the activity distribution may flag erosional events caused by, e.g. enhanced storm activity (e.g. Christiansen et al., 1997). In fact, as already

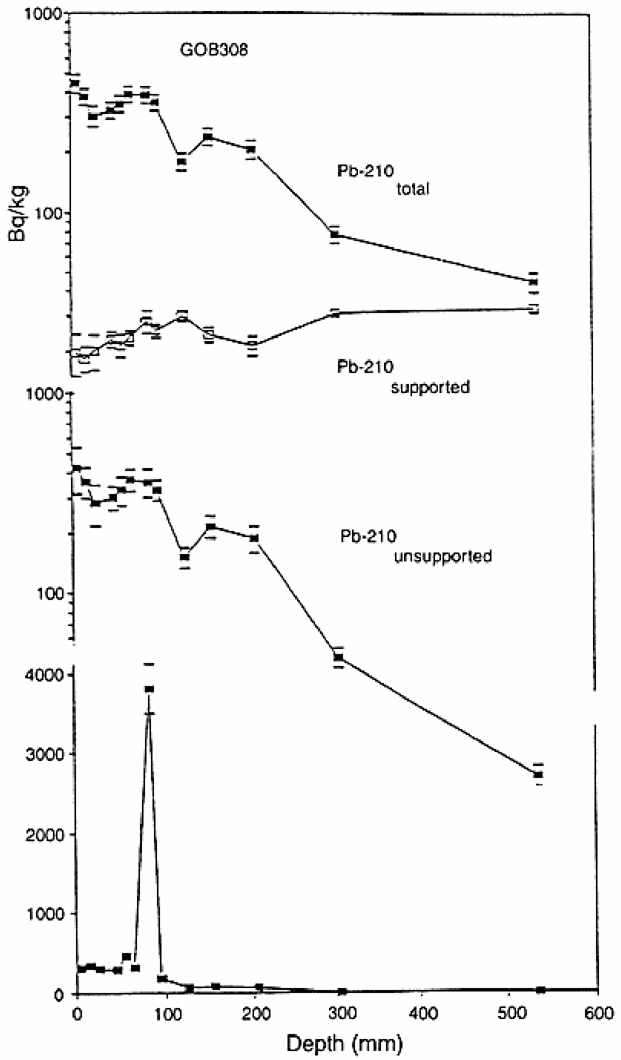

Figure 6: Gamma-spectrometric results for core GOB308.

mentioned, in the years 1971 and 1991 a number of storm surges hit the German Baltic coast (Baerens and Hupfer, 1995).

The Gdansk Basin serves as depositional center for the riverine load of the Vistula River, which drains large Polish land areas that have been opposed to the Chernobyl accident plume. Bojanowski et al. (1995) have compiled Cs-137 inventories into the Polish Exclusive Economic Zone based on cores taken during the years 1986 and 1993. They showed that all subareas in this zone were affected by the Chernobyl accident increasing the depositional figures in Bq m^{-2} by almost an order of magnitude. However, to outline the sedimentation in more detail one needs much more systematic core data that are also based on at least 1 cm sediment slices. Our measurement on core 308 shows Chernobyl derived Cs-137 in the core slice 8–9 cm with strongly decreasing activities at depth.

Re-analysing the high-activity sample of core GOB308 revealed that only a few very high activity grains might be responsible for the peak in the Cs-137 depth distribution because splitting the slice material into 2 parts for check-analysis yielded about 200 Bq kg^{-1} for one part while another subsample showed over 7000 Bq kg^{-1}. The systematic further subdivision of the high-activity sample material into 8 equal parts yielded values of about 200 Bq kg^{-1} for seven of the subsamples. Only one subsample showed over 40000 Bq kg^{-1}.

It is worthwhile mentioning that total or supported Pb-210 values do not change either in the lower activity samples nor in the highly contaminated subsample. This implies that there are two different depositional mechanisms for the two radioisotopes at work are found. Pb-210

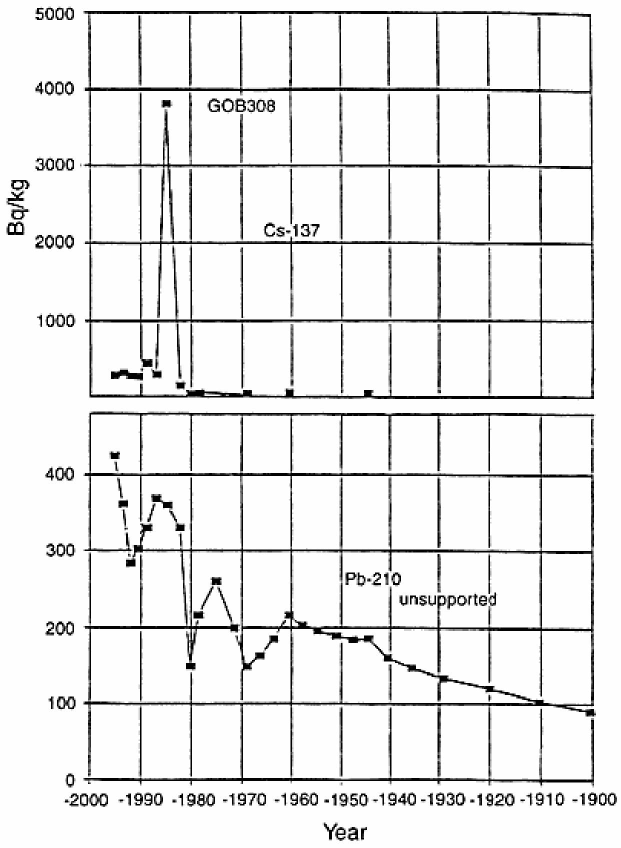

Figure 7: Selected plots of CRS modelling for core GOB308.

is probably attached to very fine particular material and does not fractionate naturally. Cs-137 may be incorporated into the sediment in the same way than Pb-210 but it may also occur as loosely intermingled Cs-137 containing grains that due to their weight may be fractionated in a sediment column by physical forcing. Care has therefore to be taken when evaluating absolute levels of Cs-137 in sediment samples because there may be a fractionation when splitting the sediment slice material into several parts yielding incorrect average activities.

Conclusions

The non-destructive Pb-210 dating of 4 cores from the Baltic Sea suggests that this technique is a natural replacement of the conventional dating technique which uses mainly alpha-spectrometry of Po-210. When properly carried out, unsupported Pb-210 data obtained by gamma-spectrometry may be applied to known modelling techniques in Pb-210 dating. Dried sediments are measured directly and therefore, repeating and checking of analytical procedures are rather straightforward.

In the Gotland Deep, Pb-210 dating of 3 sediment cores suggests that there has been a gradual increase of sedimentation rates since 1950. The most important change seems to be at the beginning of the seventies when sedimentation increased from about 1 to 1.5 mm a^{-1} and increasing to 2–3 mm a^{-1}. For the Gdansk Basin core changes are also outlined by unsupported Pb-210 minima in years ca. 1970 and ca. 1992. These changes may be attributed to hydrographic and meteorological changes and events observed in the past decades.

The Cs-137 data simultaneously obtained are generally useful in that they may be used for the verification of the dating. Plotting the Cs-137 depth distribution on a time base simply (years) expresses whether the CRS modelling assigns the known Cs-137 markers to the correct datum. However, care must be taken because Cs-137 is apparently mixed into the Baltic Sea sediment by other mechanisms than Pb-210.

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