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Sediment Accumulation Rates in the Gotland Deep, Baltic Proper Obtained by ^{210}Pb and ^{137}Cs Methods[1]

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1. Introduction

Geochronology of sediments has important contribution to limnology and geochemistry including studies of accelerated eutrophication or regional erosion rates. It is also widely used to determine recent history of anthropogenic pollution and material balance [2]. Several different methods may be used to determine the ages of sediment layers. Historically, chronology has been based on qualitative methods such as stratigraphy, tectonics and morphology. However, the best results are obtained by means of radiometric dating, based on radioactive element activity e.g. radiocarbon, thorium, radiolead [11, 13].

The last few decades have seen dramatic growth in the use of ^{210}Pb geochronology. The radiolead method can provide valuable information on sediment mixing and sediment accumulation rates within the last 100-150 years [5]. Determination of sediment accumulation rates is important for understanding of geochemical process in surface sediments, especially origin of particulate matter and estimating flux of organic carbon to the bottom sediments [10, 14].

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The radiolead method seems to be most useful when applied to sedimentation rates in coastal marine, estuarine and lacustrine environments, especially when supported by the ^{137}Cs validation [6, 7].

The method is based on measurements of ^{210}Pb (half life – 22,3 years) activity – the end product of the ^{238}U decay series. It's presence in the atmosphere is due to the diffusion of ^{222}Rn from the rocks into the atmosphere where it decays to ^{210}Pb [9, 12]. This ^{210}Pb is then removed from the atmosphere by dry and wet deposition. After falling into lakes or oceans it is deposited on the bottom of the lake. ^{210}Pb is also produced by the disintegration of ^{222}Rn derived from ^{226}Ra dissolved in water and trapped in mineral rocks. The ^{210}Pb derived from atmosphere is called excess – $^{210}Pb_{ex}$, the ^{210}Pb from the mineral matrix is called supported – $^{210}Pb_{supp}$. The amount of the $^{210}Pb_{ex}$ changes over time, it depends on the atmospheric fallout while the amount of the $^{210}Pb_{supp}$ is always constant [4].

The dependence between activity concentration and the depth below sediment-water interface, the so called radiolead profile, serves as a basis for sediment dating. Two methods are often used in this respect: CRS (usually) and CIC (sparsly).

This papers compares the results obtained from applying the constant rate of supply (CRS) model to the ^{210}Pb activity data with the results obtained by means of ^{137}Cs data. The study was carried out on the sediment cores collected from two stations in the Baltic Sea (Gotland Basin).

2. Materials and methods

Sediment cores from the Gotland Deep were collected in 1989 (II) and 2007 (I) (Table 1 and Figure 1). Samples were analysed in the Institute of Oceanology, Polish Academy of Sciences (IOPAS). The dried samples were ground with a mortar and pestle. Sediment samples of 0,1g were spiked with ^{209}Po and dissolved in perchloric acid followed by hydrofluoric acid. After removing acids, the dry residue was dissolved in 0.5 mol/dm^3, and ^{209}Po and ^{210}Pb were deposited from acidic solution onto silver disks. The activities of radionuclides on disks were counted for 1 day in a multi-channel analyzer (Canberra).

Table 1. Location of sediment cores
Table 1. Współrzędne geograficzne stacji pobierania próbek

Station	Latitude	Longitude	Depth [m]	Porosity
I	56°37' N	19°20' E	135	0,84-0,98
II	56°19' N	18°36' E	85	0,71-0,94

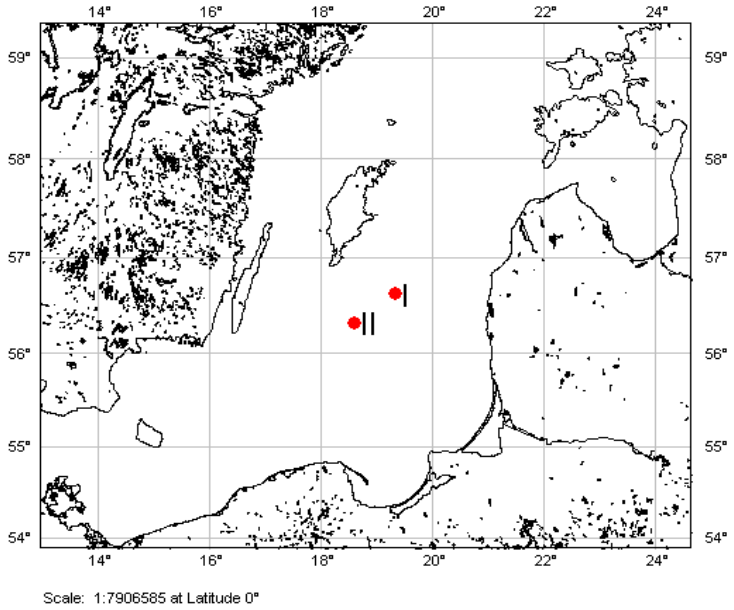

Fig. 1. Map of the Baltic Sea (southern and central) showing the location of sampling sites
Fig. 1. Mapka Morza Bałtyckiego z zaznaczonymi stacjami pobierania próbek

Sediment accumulation rates were calculated using CRS model [3]. The model assume the variable initial concentration of $^{210}Pb_{ex}$ and sediment accumulation rate but the constant flux of $^{210}Pb_{ex}$ that reaches the sediment-water interface.

Sediment accumulation rates were calculated according to the formula:

$$A_t = A_0 e^{-\lambda t}$$

where: A_t is the activity at time t, A0 is the activity at time 0, λ is the radionuclide decay constant.

When t is replaced by t=x/ν (x – depth of a given sediment layer, ν – sedimentation rate) the above formula can be rewritten:

$$A_t = A_0 e^{-\lambda\ x/\nu}$$

$$\ln A^{210}Pb_{ex}(x) = \ln A^{210}Pb_{ex}(0) - (\lambda/\nu)x$$

where: $A^{210}Pb_{ex}(x)$ is activity at layer x, $A^{210}Pb_{ex}(0)$ is activity at layer 0, λ is decay constant, ν is sedimentation rate.

In the CRS model:

$$A_x = A_0 e^{-\lambda t}$$
$$x/\nu = 1/\lambda \ln \Sigma A_{cx} / \ln \Sigma A_{c0}$$

and ΣA_{cx} is the cumulative residual $^{210}Pb_{ex}$ activity beneath sediments of depth x, ΣA_{c0} is the total $^{210}Pb_{ex}$ activity in the sediment column, x is the cumulative depth (cm), ν is the sediment accumulation rate (cm/year), t is the age of sediment (years).

The ^{137}Cs activity was determined using gamma spectrometry. Samples of known mass were placed in a gamma counter equipped with an HP Ge detector with an energetic resolution of 1.8 kV for the ^{60}Co line. Calibration of the counter was performed based on the reference IAEA materials. The mean counting time was 24 h.

3. Results

The sediments were black silty clay with shell fragments. Humidity and porosity were necessary for dating sediments. The sediment cores collected from two stations exhibited different porosity and humidity.

Total ^{210}Pb activities are higher at station II (Table 3). They ranged from 590 Bq/kg at the surface layer to 44 Bq/kg at lower sediment section. Station II is a deeper station so there is more time for sorption of ^{210}Pb than seafloor deposition. The activity of ^{210}Pb at station I range from 378 Bq/kg to 27 Bq/kg (Table 2).

The supported $^{210}Pb_{supp}$ activity concentration was calculated as the mean of activities in layers with constant activity below the decay zone.

$^{210}Pb_{ex\ activity}$ concentrations were calculated as the difference between total ^{210}Pb and $^{210}Pb_{supp}$ activities:

$$^{210}Pb_{ex} = {}^{210}Pb_{tot} - {}^{210}Pb_{supp}$$

The ^{210}Pb activities decrease exponentially until $^{210}Pb_{supp}$ levels are reached. It occures at depth about 10cm at station I (Figure 2a) and 7cm at station II (Figure 2b). Sediment accumulation rates were calculated using CRS model. At station I sediment accumulation rate equals 0,7 mm/year, while sediment accumulation rate at station II equals 0,5 mm/year.

The sediment accumulation rate determined by ^{137}Cs is equal to the depth of ^{137}Cs occurrence derived by 54 (time of analyses time of radio cesium introduction to the environment; 2008-1954). At station II it is higher than that determined from the ^{210}Pb profiles. It is equal to 1,7 mm/year. At station I sediment accumulation rate determined by ^{137}Cs is equal to 1,5 mm/year.

^{137}Cs has been introduced in the environmental as a consequence of nuclear weapons tests from 1954-1976. The significant quantities of ^{137}Cs introduced also as a result of Chernobyl accident in 1986. ^{137}Cs method is used commonly for validation ^{210}Pb results [1, 8]. However, cesium may be introduced into deeper sediment layers due to surface sediments mixing. The mixing can be executed by boths activity of benthic organisms, and physical factors.

Table 2. Activities of $^{210}Pb_{tot,}$, $^{210}Pb_{supp,}$ $^{210}Pb_{ex}$ and ^{137}Cs (Bq/kg) at station I
Table 2. Stężenie aktywności $^{210}Pb_{tot,}$, $^{210}Pb_{supp,}$ $^{210}Pb_{ex}$ i ^{137}Cs (Bq/kg) w osadach dennych pobranych na stacji I

Layer (cm)	^{210}Pb activities (Bq/kg)			^{137}Cs activity (Bq/kg)
	Total	Supported	Excess	
0-1	378	30	348	341
1-2	298	30	268	-
2-3	258	30	228	-
3-4	185	30	155	133
4-5	128	30	98	
5-6	68	30	38	55
6-7	54	30	24	-
7-8	68	30	38	33
8-9	36	30	6	-
9-10	24	30	-	3
10-11	31	30	-	-
11-12	25	30	-	0
13-14	32	30	-	-
15-16	30	30	-	-
17-18	32	30	-	-
19-20	33	30	-	-
21-22	31	30	-	-
23-24	30	30	-	-
25-26	29	30	-	-
27-28	30	30	-	-

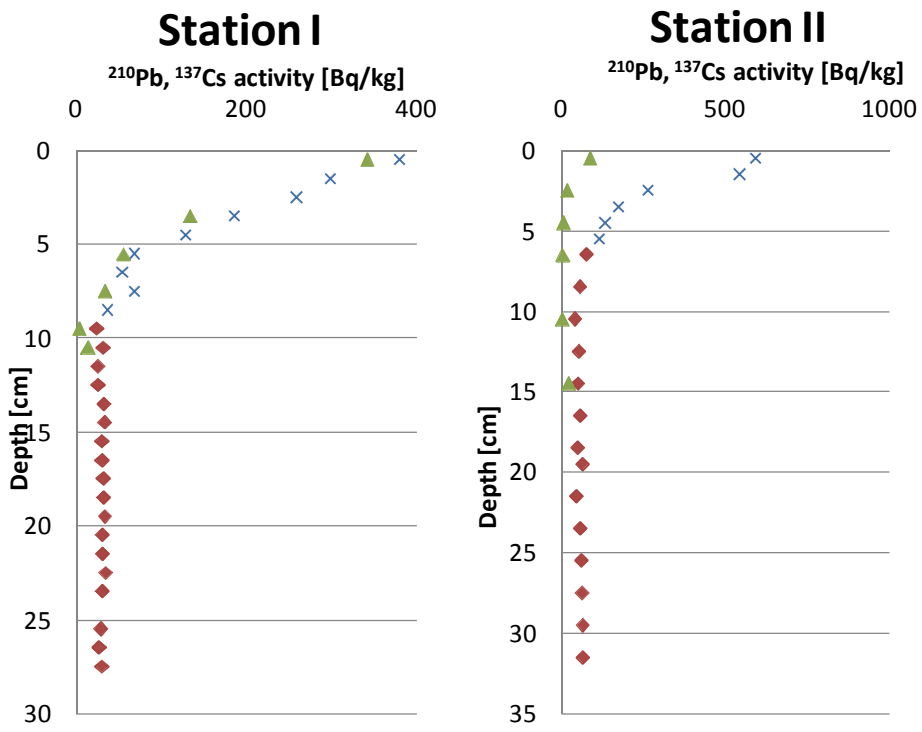

Fig. 2. ^{210}Pb activity against sediment depth profiles. Rhombs mark $^{210}Pb_{supp}$, x mark $^{210}Pb_{ex}$, triangles mark ^{137}Cs

Fig. 2. Profile stężenia aktywności mierzonych radionuklidów; romby $^{210}Pb_{supp}$, x $^{210}Pb_{ex}$, trójkąty ^{137}Cs

Table 3. Activities of $^{210}Pb_{tot,}$, $^{210}Pb_{supp}$, $^{210}Pb_{ex}$ and ^{137}Cs (Bq/kg) at station II
Table 3. Stężenie aktywności $^{210}Pb_{tot}$, $^{210}Pb_{supp}$, $^{210}Pb_{ex}$, i ^{137}Cs (Bq/kg) w osadach dennych pobranych na stacji II

Layer (cm)	^{210}Pb activities (Bq/kg)			^{137}Cs activity (Bq/kg)
	Total	Supported	Excess	
0-1	590	56	534	86
1-2	541	56	485	-
2-3	261	56	205	18
3-4	172	56	116	-
4-5	131	56	75	7
5-6	114	56	58	-
6-7	74	56	-	-
8-9	55	56	-	3
10-11	40	56	-	-
12-13	52	56	-	-
14-15	49	56	-	-
16-17	56	56	-	-
18-19	48	56	-	-
19-20	62	56	-	-
21-22	44	56	-	-
23-24	56	56	-	-
25-26	59	56	-	-
27-28	61	56	-	-
29-30	64	56	-	-
31-32	64	56	-	-

4. Discussion

Using ^{210}Pb and ^{137}Cs method can help to understand better geochemical process in surface sediments and provide information on pollution chronologies. ^{210}Pb have been widely applied to estimate the age of the sediment and the rate of sedimentation. The ^{210}Pb method of dating sediments is not only used in studies of early diagenesis but also to interpret the geochemical changes caused by man's activities during the last decade. We can use different models (e.g. CRS) which allow us to precisely determine the sediment accumulation rate.

^{137}Cs method usually confirm results obtained from ^{210}Pb geochronology. It's used to validate ^{210}Pb method. The rate of sediment accumulation derived from both techniques, ^{137}Cs and ^{210}Pb are usually similar.

Results obtained by ^{137}Cs method were higher than those determined by lead model. The differences could be caused by sediment mixing, especially physical mixing process. But not only physical processes caused mixing. It also could be caused by bioturbation, which can be defined as sediment displaced by the activity of bottom-living animals. Chemical mobility and resuspension also caused different results between sediment accumulation rates obtained by radio-lead and ^{137}Cs methods.

References

1. **Abril J.M.:** *Constraints on the use of ^{137}Cs as a time-marker to support CRS and SIT chronologies.* Environmental Pollution 129, 31-37, 2003.
2. **Alvarez-Iglesias P., Quintana B., Rubio B., Perez-Arlucea M.:** *Sedimentation rates and trace metal input history in intertidal sediments from San Simon Bay (Ria de Vigo, NW Spain) derived from ^{210}Pb and ^{137}Cs chronology.* Journal of Environmental Radioactivity 98, 229-250, 2007.
3. **Appleby P.G., Oldfield F.:** *The concentration of lead-210 dates assuming a constant rate of supply and unsupported ^{210}Pb to the sediment.* Cantena 5, 1-8, 1978.
4. **Appleby P.G., Oldfield F.:** *The assessment of ^{210}Pb data from sites with varying sediment accumulation rates.* Hydrobiology 103, 29-35, 1983.
5. **Baskaran M., Naidu A.S.:** *^{210}Pb derived and the fluxes of ^{210}Pb and ^{137}Cs isotopes into continental shelf sediments, East Chukchi Sea, Alaskan Arctic.* Geochimica and Cosmochimica Act, Vol. 59, No. 21, 1995.
6. **Benoit G., Rozan T.F.:** *^{210}Pb and ^{137}Cs dating methods in lakes: a retrospective study.* Journal of Palelimnology 25, 455-465, 2000.
7. **Lima A.L., Hubeny J.B., Reddy Ch., King J.W., Hughen K.A., Eglinton T.:** *High-resolution historical records from Pettaquamscutt River basin sediments: 1. ^{210}Pb and varve chronologies record of ^{137}Cs released by the Czernobyl accident.* Geochimica and Cosmochimica Acta, Vol. 69, No. 7, 2004.
8. **Lu X.:** *Application of the Weidbull extrapolation to ^{137}Cs geochronology in Tokyo Bay and Ise Bay, Japan.* Journal of Environmental Radioactivity 73, 169-181, 2004.
9. **McDonald C.P., Urban N.R.:** *Sediment radioisotope dating across a stratigraphic discontinuity in a mining-impacted lake.* Journal of Environmental Radioactivity 92, 80-95, 2007.
10. **Pempkowiak J., Knapińska-Skiba D.:** *The determination of accumulation rates in the recent Baltic Sea sediments on the basis of Pb-210 and Cs-137 profiles.* Proc. 16th CBO, Kiel 1988, Vol, 2, 824-832, 1988.
11. **Pempkowiak J., Widrowski H.:** *Zanieczyszczenie metalami ciężkimi wierzchniej warstwy osadów dennych południowych rejonów Morza Bałtyckiego.* Arch. Ochrony Środowiska, 1-2, 55-66, 1988.

12. **Saxena D.P., Joos P., Grieken R.V., Subramanian V.:** *Sedimentation rate of the floodplain sediments of the Yamuna river basin (tributary of the river Ganges, India) by using ^{210}Pb and ^{137}Cs techniques.* Journal of Radioanalitycal and Nuclear Chemistry, Vol. 251, No. 3, 2002.
13. **Turner L.J., Delorme L.D.:** *Assessment of ^{210}Pb data from Canadian lakes using the CIC and CRS models.* Environmental Geology 28(2), 1996.
14. **Zaborska A., Carroll J., Papucci C., Pempkowiak J.:** *Intercomparison of alpha and gamma spectrometry techniques used in ^{210}Pb geochronology.* Journal of Environmental Radioactivity 93, 38-50, 2007.

Szybkość sedymentacji powierzchniowych osadów bałtyckich zmierzona przy użyciu ^{210}Pb i ^{137}Cs

Streszczenie

Metoda ołowiowa jest powszechnie używana do oznaczania wieku osadów. Dodatkowo pozwala ona lepiej zrozumieć procesy geochemiczne zachodzące w osadach. W celu poprawy jakości wyników metodę ołowiową weryfikuję się używając ^{137}Cs. Poniższy artykuł dotyczy porównania szybkości sedymentacji osadów uzyskanych przy pomocy metody ołowiowej (model CRS- Constant Rate of Supply) z wynikami uzyskanymi za pomocą ^{137}Cs. Obliczenia wykonane zostały dla dwóch osadów pobranych z Głębi Gotlandzkiej. Wyznaczone metodą ołowiową wartości współczynników sedymentacji (0,7 mm/rok dla I stacji i 0,5 mm/rok dla II stacji) były niższe od współczynników sedymentacji obliczonych za pomocą ^{137}Cs (1,5 mm/rok dla I stacji i 1,7 mm/rok dla II stacji). Rozbieżność była spowodowana mieszaniem powierzchniowym osadów.