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FIRST MEASUREMENTS OF NATURAL RADIOACTIVITIES OF ^{210}Pb IN THE INSTITUTE OF PHYSICS, SILESIAN UNIVERSITY OF TECHNOLOGY

TOMASZ GOSLAR, MAŁGORZATA GANOWICZ, JUSTYNA CZERNIK and JAROSŁAW SIKORSKI

Department of Radiosotopes, Institue of Physics, Silesian University of Technology Krzywoustego 2, 44-100 Gliwice, Poland
(e-mail: goslar@zeus.polsl.gliwice.pl, justyna@zeus.polsl.gliwice.pl)

Abstract. In this paper we describe the first measurements of ^{210}Pb concentrations in sediments, performed in the Institute of Physics, Silesian University of Technology, Gliwice. The reliability of our method has been tested by the ^{210}Pb measurements in the sediments of Lake Gościąż. The reasons for the non-monotonic profile of ^{210}Pb in these sediments have been discussed. We also report and discuss the ^{210}Pb measurements in the sediments formed in front of the Skeidarar glacier (Iceland).

1. INTRODUCTION

Since its introduction in the seventies (Krishnaswami *et al.,* 1971; Koide *et al.,* 1973), ^{210}Pb method has been widely used in dating of lake sediments (e.g. Robbins, 1978; Appleby and Oldfield, 1983; Oldfield and Appleby, 1984; Wang and Cornett, 1993). ^{210}Pb in sediments consists of authigenic ^{210}Pb coming from the decay of ^{226}Ra, which is natural component of terrestrial rocks, and of allochthonous ^{210}Pb produced from ^{222}Rn, which liberats from the Earth's surface to the atmosphere. Allochthonous ^{210}Pb is settled at the Earth's surface bound to aerosols or directly to the rain droplets. To the lakes, ^{210}Pb is supplied directly from the atmosphere or from lake surroundings (e.g. due to the soil erosion). Concentration of authigenic ^{210}Pb in sediments is constant, as radioactive decay of ^{210}Pb is counterbalanced by its production from ^{226}Ra. On the other hand, allochthonous ^{210}Pb is not bound to ^{226}Ra, and its concentration decreases after burial in sediments. For that reason, measurements of ^{210}Pb concentration enable dating of sediments, and determination of their deposition rate. Due to the short half-life of ^{210}Pb (22.26 year) the ^{210}Pb method is suitable for dating sediments not older than 100-150 years.

Specific activity of ^{210}Pb may be related to the age of sediments by simple law of radioactive decay if: a) the specific activity of ^{210}Pb in the sediment at the moment of its deposition is known, b) neither vertical mixing of sediments nor vertical migration of ^{210}Pb within sediments occur. To derive ^{210}Pb activity at the moment of deposition, ^{210}Pb activity is measured in modern sediments, and one assumes either that the specific activity of ^{210}Pb (expressed e.g. in Becquerels per gram of sediment) at the moment of sedimentation was constant in the past, or that the flux of ^{210}Pb supplied to the lake (expressed e.g. in Becquerels per year per square meter of sediment area) was constant over time. Both models give the same results if the sedimentation rate was constant.

In the measurement, total activity of ^{210}Pb (allochthonous plus authigenic) is determined. To derive the amount of allochthonous component, one usually assumes uniform concentration of ^{226}Ra and authigenic ^{210}Pb in the whole profile. This can be determined by the measurement of ^{210}Pb in a sufficiently old layer of sediment.

2. THE TECHNIQUE OF ^{210}Pb MEASUREMENT

In 1997, a complete system for determination of ^{210}Pb concentration in sediments was built and tested in the Department of Radioisotopes, Institute of Physics (Ganowicz, 1997). Concentration of ^{210}Pb is derived from the measurement of alpha activity of ^{210}Po, which is in radioactive equilibrium with ^{210}Pb. A sample for alpha measurement has a form of a thin layer of polonium extracted from the sediment and deposited on a silver disc (Flynn, 1968). Initial dry mass of sample is 1-2 g. The sample is treated with a hot hydrochloric acid, the remaining sediment separated from the solution by a few centrifugation steps (4000 rpm during 10 minutes in each step), and then treated with a hot nitric acid (10-20 ml, 95°C, 30 min). In nitric acid, polonium is extracted from sediment. The extraction is continued in a room temperature during the following 24 h, and hence the sediment is separated from the solution by centrifugation (4000 rpm during 10 min). The solution is collected in the teflon vial, while the remaining sediment is washed with the nitric acid and centrifugated again. Both aliquots of the solution are collected in one teflon vial. After adding a few drops of H_2O_2 (to oxygenate organic matter) the solution is evaporated.

The evaporite is dissolved with 5ml portion of HCl and evaporated again. After a few dissolution/evaporation steps, nitric acid is fully replaced with hydrochloric acid.

Polonium is deposited on silver from the 0.5 M solution of HCl, containing 0.4 g of hydroxyloamine and 0.4 g of sodium citrate, at 80°C. Silver disc with a diameter of 20 mm is placed in a special holder, which enables polonium deposition on one side of the disc. To speed up the deposition, a magnetic stirrer is placed below the disc. After ca. 3 h of deposition, the disc surface is covered with a white coating. To control efficiency of deposition and alpha detection, a portion of HCl containing known amount of ^{208}Po (manufactured by AERE Harwell, United Kingdom) is added before evaporation. ^{208}Po does not occur in natural environment. Efficiency of deposition of 30-80% is commonly achieved.

Alpha activity is measured with a spectrometer Canberra model 7401, with a surface-barrier Si semiconductor detector. Sensitive area of the detector is 300 mm^2 and its energy resolution is ca. 20 keV. This enables for a very good separation of peaks from ^{210}Po (E=5.308 MeV) and ^{208}Po (E=5.105 MeV). Absolute activities of both isotopes are rather low and to obtain sufficient precision, one measurement lasts a few thousand minutes. Exemplary spectrum, recorded in a single-day measurement of the sample G43f 15-0(3) is shown in **Fig. 1**.

As the half lifes of both polonium isotopes are quite short (T_{208}=2.898 years, T_{210}=138.4 days), results of calculations are corrected for radioactive decay of ^{208}Po since the moment of its calibration, and decay of ^{210}Pb since the moment of polonium extraction from sediments.

3. ^{210}Pb PROFILE IN LAMINATED SEDIMENTS OF LAKE GOŚCIĄŻ

To verify the applied technique, ^{210}Pb radioactivity has been measured in laminated sediments of Lake Gościąż (**Table 1**). Due to annual lamination, the sediments of Lake Gościąż contain excellent record of environmental changes in the past, and are being extensively investigated. The results of research completed to date have been recently published in a separate monography (Ralska-Jasiewiczowa *et al.*, 1998).

Radioactivities of ^{210}Pb in the Gościąż sediments have been earlier measured in the Department of Physics and Nuclear Techniques, Academy of Mining and Metallurgy in Cracow (Wachniew, 1993). Comparison of results obtained in Gliwice and Cracow is shown in **Fig. 2**. The results roughly agree with each other, but activities measured in Gliwice are generally lower than those measured in Cracow. This is due to radioactive decay of ^{210}Pb in a period between moments of measurement in both institutions. The decay decreases specific activity of allochthonous ^{210}Pb. For better comparison, the activities of allochthonous ^{210}Pb of samples examined in Cracow have been normalised to the spring 1997, when the Gliwice measurements have been made. In calculations of allochthonous ^{210}Pb it was assumed

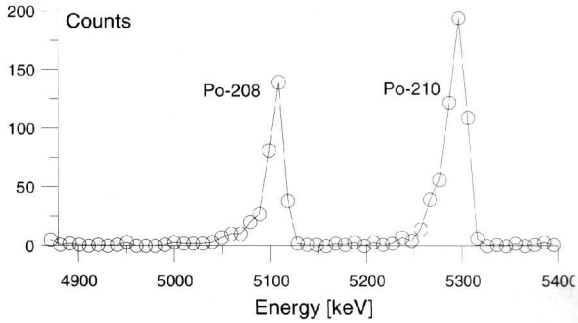

***Fig. 1.** Energy spectrum of alpha radiation for the sample G43f 15-0 (3).*

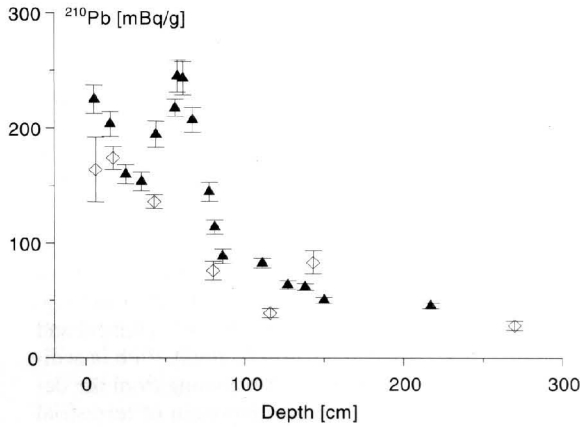

***Fig. 2.** Profile of specific activity of ^{210}Pb in the laminated sediments of Lake Gościąż. The symbols are used to distinguish measuments performed in two different institutions: triangles-Academy of Mining and Metallurgy, Cracow (Wachniew, 1993), diamonds-Silesian Technical University, Gliwice (this work).*

***Table 1.** Results of ^{210}Pb measurements in the sediments of Lake Gościąż.*

No.	Sample	Depth [cm]	Lab. No.	Specific activity of ^{210}Pb [mBq/g]
1	G43f 15-0(3)	4-8	GdPb-1	164±28
2	G43f 22-15(2)	15-18	GdPb-2	174±10
3	G 43f 47-41(2)	41-44	GdPb-3	136±6
4	G 33f 646-650	79-82	GdPb-4	76±8
5	G 33f 621-625	112-120	GdPb-5	39±4
6	G 33f 596-600	142-144	GdPb-6	83±10
7	G 23f 1520-1550	265-275	GdPb-7	28±4

that the activity of authigenic ^{210}Pb ranges between results obtained for samples older than 200 years, i.e. 45.4±2.5 mBq/g for 217 cm (Wachniew, 1993) and 28±4 mBq/g for 270 cm (**Table 2**). The concordance of normalised results is very good (**Fig. 3**), except for the two bottommost samples.

Wachniew (1993) pointed out, that the profile of ^{210}Pb in the sediments younger than 1960 AD is non monotonic, which is fully confirmed by our measurements. Wachniew (1993) suggested also, that the non-monotonic character of the profile was caused by remobilisation of lead in the sediments.

Pb is fallout from the water through the sorption by oxides (hydroxides) of iron and manganese (Carpenter *et al.*, 1981, Benoit and Hemond, 1990). Solubility of iron and manganese oxides in water depends strongly on their oxidation state: they preciptate in oxygenated water and dissolve in reductive conditions. In many lakes, a thin layer of top sediments is rich in oxygen, while decomposition of organic matter causes strong deficit of oxygen in deeper sediments. Due to that Fe and Mn diffuse upwards, and precipitate at the surface of sediments. Then, the maximum of Fe concentration is observed in the top sediments (Engstrom and Wright, 1984). If, however, hypolimnion and top sediments are poor in oxygen, both Fe and Mn diffuse to the water and their maxima occur in hypolimnion. In meromictic lakes, such a maximum occurs permanently (e.g. in Hall Lake, Washington, USA; Balistrieri, 1994). In seasonally stratified lakes, hypolimnic maximum of Fe and Mn disappears during the overturn season. When the overturn goes off, the maximum is rebuilt within a few days or weeks (Dean, 1993; Balistrieri, 1994), and in some lakes (Dean, 1993; Engstrom *et al.*, 1985) both elements circulate repeatedly between oxygenated lake surface and sediments, before being ultimately buried in sediments, or washed out from the lake with the outflowing water.

In some eutrophic lakes, release of Fe and Mn from sediments may be inhibited by formation of carbonates

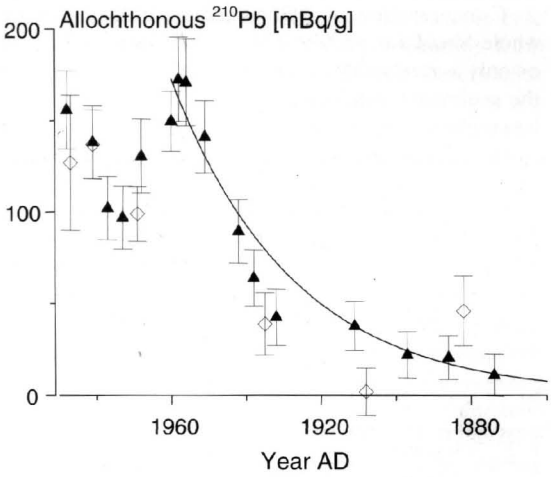

Fig. 3. *Profile of specific activity of allochtonous ^{210}Pb in the laminated sediments of Lake Gościąż. The meaning of symbols is the same as in Fig. 2. The specific activity of ^{210}Pb measured in Cracow (Wachniew, 1993), has been normalised to the spring of 1997 (see the text for details). The scale of calendar years has been elaborated using the varve chronology (Goslar, 1998). The solid line represents radioactive decay of ^{210}Pb, and has been fitted to the data from before 1960.*

(Mayer *et al.*, 1982). Another possibility is a fixation of Fe in extremely insoluble sulfides FeS lub FeS_2, when decomposition of organic matter produces large amounts of H_2S (e.g. Wetzel, 1975). Manganese sulfides are much more soluble than those of iron, and production of H_2S does not influence Mn fixation significantly (Engstrom and Wright, 1984).

For better interpretation of the ^{210}Pb profile in the Lake Gościąż sediments, it is important to know, in which way the shape of profile has been altered by the remobilisation of lead. Basing on few chemical analyses, Wachniew (1993) pointed out, that concentration of iron and manganese was low in the top sediments. One could then guess, that the youngest sediments were the most depleted in oxygen, so Pb dissolved from the topmost sediments diffused downwards, where it was fixed at the boundary of sediment containing more oxygen. High maximum of ^{210}Pb about 1960 AD (**Fig. 3**) could be thus an effect of accumulation of Pb diffused from the overlying sediment. Strong oxygen depletion in the topmost sediments would result from lake eutrophication (incerase of biological productivity of the lake), and intensified decomposition of organic matter.

However, more detailed analysis contradict that interpretation. According to Benoit and Hemond (1991), influence of diffusion on the ^{210}Pb profile is significant only when the sedimentation rate was lower than 1mm/yr, so significant diffussive transport to the depth of 50 cm (^{210}Pb maximum in the Lake Gościąż profile) during <30 years should not be expected. The detailed varve chronology (Goslar, 1998) enables us to reconstruct the changes of initial concentration of ^{210}Pb, i.e. at the moment of deposition. In the section from before 1960, the ^{210}Pb profile can be easily explained by radioactive decay of ^{210}Pb (**Fig. 3**). This is indirectly illustrated in **Fig. 4**, which shows the profile of initial activity of ^{210}Pb, calculated with the assumption that the Pb was perfectly fixed in sediments. **Fig. 4** demonstrates, that between 1860-1960 initial activity of ^{210}Pb was approximately constant, anyway the maximum of ^{210}Pb around 1960 AD appears insignificant. One should rather guess, that concentration of Pb fixed before 1960 was approximately constant, while for some reasons after 1960 fixation of lead was weaker.

Fig. 4 suggests, that the sediment ability of lead fixation is correlated with concentration of iron. The decline of initial ^{210}Pb activity after 1960 occurred at the period when the concentration of Fe was low. Also the minimum of initial ^{210}Pb about 1906 AD coincides with the minimum of Fe. So, at a first approximation, the profile of allochthonous ^{210}Pb in the Lake Gościąż sediments can be explained by changing ability of Pb fixation, proportional to that of Fe. With such an interpretation, the diffusion of Pb within sediments seems of minor importance.

The reasons of Fe variations in the Lake Gościąż sediments were widely discussed by Goslar (1998). At a first glance, lowering of Fe concentration would suggest oxygen depletion in the topmost sediments, e.g.

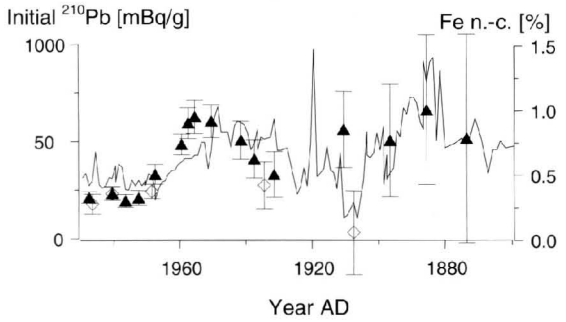

Fig. 4. *Profile of initial activity of ^{210}Pb in the sediments of Lake Gościąż, calculated with the assumption of no lead migration in the sediments. The meaning of symbols is the same as in Fig. 2. One of results included in the table 1, outstanding the plot area (1700±700 mBq/g for 1882AD) has been omitted. Solid line represents the detailed profile of the not bound to carbonates iron content in the sediments (Goslar, 1998).*

due to progressing lake eutrophication. However, the profiles of phosphorus, vivianite, and diatom, algae and zooplankton assemblages (Goslar *et al.*, 1999) strongly demonstrate the decrease of biological productivity in the lake during the last 50 years. This is an effect of abandoning the farms in the lake's vicinity after the Second World War. This is concordant with the abrupt drop of the Cu/Zn ratio in sediments, which demonstrates stronger oxygenation of hypolimnion during the last 40 years. Also gradual deterioration of varve regularity and extinction of annual lamination after 1966 AD is explained by occurrence of short-lasting periods of vertical circulation in the lake (Goslar, 1998). Such a circulation would increase concentration of oxygen in the hypolimnion.

As hypothesised by Goslar (1998), the short-lasting circulation events would cause the decrease of Fe and Mn concentration in sediments. Such short events could increase concentration of oxygen in the hypolimnion only slightly, too weak to inhibit dissolution of Fe and Mn from sediments. In the present lake, hypolimnion is still strongly depleted in oxygen, and due to the diffusion of dissolved Fe and Mn oxides from the sediments, maximum concentrations of these elements occur in the hypolimnion. In the event of circulation, hypolimnion water masse, rich in Fe and Mn, is replaced with the surface water, depleted with these elements. Such a depletion in hypolimnion may stimulate further diffusion from sediments, and in consequence may lead to the decrease of Fe and Mn concentration in the sediments. In such circumstances, atoms of Fe and Mn circulate intensively between hypolimnion and epilimnion, which of course rises probability of their escape with the outflowing water. It must be stressed that this mechanism of modification Fe and Mn profiles does not require long diffusion lengths of these elements in sediments.

As suggested above, the non monotonic profile of ^{210}Pb does not require significant diffusion of lead in sediments, but it migth be caused by a variable rate of Pb fixation, proportional to the concentration of iron. This proportionality reflects identical mechanisms involved; Pb, fixed in sediments on the surface of iron oxides, after their dissolution also diffuses to the hypolimnion, and when the hypolimnion is washed with the surface water, it may easier escape from the lake.

4. PROFILE OF ^{210}Pb CONCENTRATION IN THE SEDIMENTS IN THE FOREGROUND OF THE SKEIDARAR GLACIER (ICELAND)

In the Division of Radioisotopes, a series of ^{210}Pb measurements in the sediments from the foreground of the Skeidarar glacier, Iceland, has been made. The sediments of the Skeidarar area are investigated by the scientists from the Institute of Geography, Nicolaus Copernicus University, Toruń. Three samples (**Table 2**) have been collected from the depth 10-15 cm below the surface of the end moraine (samples 1) and the bottom moraine (samples 2 and 3), both formed during the glacier advance around the middle of this century (E. Wiśniewski, unpublished inf.). Currently, the glacier front is situated ca. 700 m apart of the end moraine, and the maximum distance between places of sample collection is ca. 500 m. The other samples (4-9) come from the 27 m thick profile of glacial and fluvioglacial formation. Measurements of ^{210}Pb have been performed to recognise whether the sedimentation in this profile occurred in the present century, or the sediments were formed during earlier periods of glacier advance.

The results of ^{210}Pb measurements are shown in **Fig. 4** along the lithological profile. This profile does not show the decrease of ^{210}Pb with depth, which should be expected for modern sediments. This may be for a few reasons.

Concentration of ^{210}Pb is almost uniform in the whole Skeidarar profile. This would indicate the lack or only a very small amount of allochthonous ^{210}Pb in the sediments. Additionally, this would imply more or less uniform concentration of ^{226}Ra in the profile, what is quite surprising as the lithology is quite complex (mud intercalated with silts and sands of fluvioglacial and glaciolimnic series). Such a uniformity would favour reasonable dating of sediments, since in such a case the error connected with authigenic ^{210}Pb is small.

Table 2. *Specific activities of ^{210}Pb in the sediments at the foreground of the Skeidarar glacier, Iceland.*

No.	Sample	Lab. No.	Specific activity of ^{210}Pb [mBq/g]
1	Skeidarar 0.15 m - 1	GdPb-15	5.8±0.5
2	Skeidarar 0.15 m - 2	GdPb-16	4.9±0.3
3	Skeidarar 0.15 m - 3	GdPb-17	7.0±0.8
4	Skeidarar 5.5 m	GdPb-8/14	9.2±1.0
5	Skeidarar 11 m	GdPb-9	9.3±1.3
6	Skeidarar 23 m	GdPb-10	12.3±0.8
7	Skeidarar 24 m	GdPb-11	13.5±1.8
8	Skeidarar 25 m	GdPb-12	10.1±1.1
9	Skeidarar 27 m	GdPb-13	8.9±1.4

Therefore, the apparent lack of decline of ^{210}Pb donwards indicates extremely low initial concentration of ^{210}Pb, or the age of sediments being beyond the range of the ^{210}Pb method (>150 years).

Geomorphological situation allows us to guess, that the moraine, where the three topmost samples come from, was formed less than 50 years ago. Despite that, concentration of allochthonous ^{210}Pb in the youngest samples is lower than 5mBq/g. This is little, comparing the top sediments of e.g. Lake Gościąż (ca. 170 mBq/g), a few the biggest Swiss lakes (130-300 mBq/g, Gunten and Moser, 1993) or the Lake Maskinonge, Canada (ca. 600 mBq/g, Wang and Cornett, 1993). This could be due to very low concentration of ^{210}Pb in the air. Both the half-life time of ^{222}Rn and residence time of ^{210}Pb in the air ^{222}Rn are short (a few days), so the concentration of ^{210}Pb in the atmosphere is not uniform. One could speculate, that concentration of ^{210}Pb in the air over Iceland is low, because of a low concentration of ^{226}Ra in the volcanic rocks, covering large part of Iceland. However, significant role may be played by the small area of Iceland, and situation of the island far from continents. Liberation of Rn from oceanic water is negligibly small, and within a few days (half-life of ^{222}Rn and residence time of ^{210}Pb in the air), significant dilution of ^{210}Pb with the ^{210}Pb-free maritime air is possible. Unfortunately, we do not know any results of ^{210}Pb measurements in the air or in modern soils in Iceland.

It is also possible, that the age of sediments is beyond the range of the ^{210}Pb method. This does not need to mean that the material was deposited long ago in the profile. Low concentration of allochthonous ^{210}Pb may indicate, that the clays, sands and silts present in the profile, were rebedded from places, where they had no contact with the surface, and where no allochthonous ^{210}Pb was supplied. Similar effect would occur in case of redeposition of mixed material, descending from different depths. In such a case, if a few meters thick layer is deposited in a very short time (during a few days), the whole profile is almost lacking in allochthonous ^{210}Pb, except of a few top centimeters, which are in contact with the air until they are covered with overlying sediments.

The profile of ^{210}Pb has a maximum at the bottom of the mud layer (ca. 24 m) intermediate values of ^{210}Pb concentration occur in fluvioglacial sediments, and the lowest in the moraine clay. This may reflect the relationship between concentration of allochthonous ^{210}Pb and the length of period, when given layer was exposed to the air. With this interpretation one may understand why the clay material at the depth 15 cm contains least ^{210}Pb; in fact it was never in contact with the surface (first it was buried under the ice, and during the ice retreat it has been immediately covered with the impermeable layer of clay). Also the occurrence of ^{210}Pb maximum in muds seems reasonable, since they probably deposited at the slowest rate. Of course, such an interpretation is reliable only when the whole sediment is relatively young (<150 years). In fact, as ^{210}Pb does not decline with depth, it is still possible, that the observed profile of ^{210}Pb reflects just the non-uniform concentration of ^{226}Ra in the rock.

5. FINAL REMARKS

The system built in the Institute of Physics, Silesian University of Technology enables measurements of specific activities of ^{210}Pb in sediments, with the accuracy of a few mBq/g. ^{210}Pb measurements in the sediments of Lake Gościąż were used as a check of the method, and they confirmed the non-monotonic character of the ^{210}Pb profile. Using calendar chronology of sediments and detailed data on iron concentration we hypothesise, that the non-monotonic character of ^{210}Pb profile reflects changing rate of lead fixation in the sediment. The drop of allochthonous ^{210}Pb fixed after 1960 seems to be connected with the decrease of iron concentration and extinction of lamination, probably as a result of short-lasting events of vertical water circulation in the lake body. The mechanism of lead diffusion, suggested earlier, seems less important. One may thus hypothesise, that the ^{210}Pb method can give reliable ages of sediments only if the profile of iron concentration is uniform.

Glacial, fluvioglacial and glaciolimnic sediments, formed at the foreground of the Skeidarar glacier, contain very little ^{210}Pb. The most probable reason for that, is the depletion of ^{210}Pb in the air over Iceland, due to dilution with the ^{210}Pb-free maritime air, and generally a very short time when the sediment material was exposed to the air. This is in reasonable agreement with the small maximum of ^{210}Pb concentration in the mud, which probably deposited slower than the other facies.

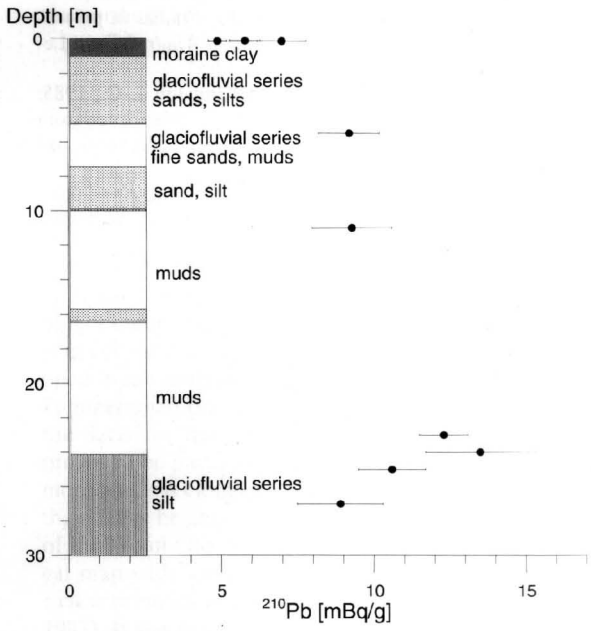

Fig. 5. *Profile of ^{210}Pb activity in the sediments formed at the foreground of the Skeidarar glacier (Iceland). The lithology of sediments is shown to the left.*

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