The Use of ^{137}Cs in Marine and Lacustrine Sediment Dating*

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Abstract—In this work, the ^{137}Cs dating technique with improved accuracy is shown to be a powerful tool for establishing chronologies in different (lacustrine, marine) young aquatic deposits. The method has special importance when the ^{210}Pb method gives partial or unclear results.

1. INTRODUCTION

In the past, the dating of young sediments by Cs-137 has received much attention, and specially, when used in combination with other radiometric methods (Robbins and Eddington, 1975; Pennington *et al.*, 1976; Jaakkola *et al.*, 1983; Gunten *et al.*, 1987).

However, unsuccessful applications have been reported (Longmore *et al.*, 1983; Davis *et al.*, 1983). There are also situations where Cs-137 records have suffered additional uncertainties from indefinite injections into sea-water from nuclear power stations (Cambray, 1982; Pentreath, 1985).

The relatively simple Cs-137 technique using γ-ray spectrometry, makes this method of measurement accessible to a wide range of laboratories. Nevertheless, it is well known that the assessment of radioactivity at low levels is not a trivial task. Therefore, care should be taken when interpreting the γ-ray spectrometer information for dating purposes.

In this paper Cs-137 measurements in sediments are described, and the derived dates are compared with those obtained by the Pb-210 method. Special attention is paid to the study of a marine sediment for which information on the use of the Cs-137 dating technique is relatively scant in the literature (Chanton *et al.*, 1983; Smith and Walton, 1980).

2. SITE AND SEDIMENT DESCRIPTION

Marine sediments were collected off the coast of Ringhals (Sweden), near Goteborg as shown in Fig. 1, as part of a monitoring programme around the area of the nuclear power station. The Ringhals nuclear facility was started in 1975 and since 1983 four reactors have been in operation (3400 MW).

Frequent monitoring programmes (Notter, 1986) to assess the influence of the Ringhals nuclear facility demonstrated no increased values of Cs-137 in the sediments. The elevated amounts of Cs-137 in these sediments, as compared with similar sediments from the Baltic Sea, at that time showed that, in addition to atmospheric Cs-137, there were major contributions of Cs-137 in the area (40–90% of the total inventory) due to the nuclear facility of Sellafield on the Irish Sea (Notter, 1986).
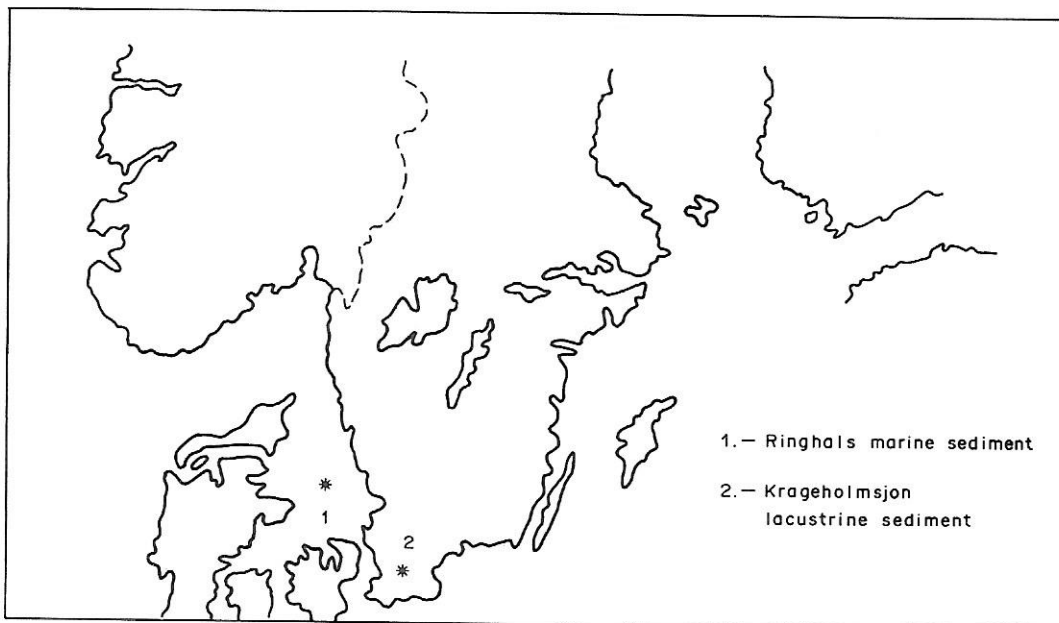
Fig. 1. Outline map of Scandinavia, showing approximately the sites of sediment collection.

The sediment core was collected during June 1984, at 20 km from the coast, from a transportation bottom, and at a depth of 96 m. The length of the core was 20 cm. The sediment was divided into sections, 1 cm thick, and freeze dried. The content of organic matter was determined by measuring the loss on ignition at 550°C. The density of dry matter was evaluated using the organic matter and water content.

Selected lacustrine core was also collected in 1984, from Lake Krageholmsjon which is situated 9 km north west of Ystad, the most southern part of Sweden (Fig. 1). The lake is 43 m above sea level, has an extension of 2.2 km², and is surrounded by a relatively small drainage area. The lake is predominantly surrounded by clayey–sandy moraine with an essentially calcareous bedrock. The major part of the drainage area is cultivated land with zones of forest and peat (Regnell, 1987).

Two important features of the lake need to be considered:

(1) The high content of carbonates in the water and sediment that originate from physically eroded particles from the drainage area. However, carbonates arriving at the lake are generally leached with surface or groundwater and are precipitated either chemically or biologically.

(2) The water-level fell at the end of the nineteenth century (Regnell, 1984).

The sediment core (1.5 m long) was collected at the central part of the lake at a depth of 7 m. It was cut into samples 2.5 cm thick and stored in acid-washed plastic bags. The water-content was determined in each slice and the organic content was estimated by loss on ignition at 550°C, and the carbonate content at 950°C (Regnell, 1984).

The dry bulk-density of the lacustrine sediments increases gradually with depth (Fig. 2), due to compaction effects, as expected. The carbonate values were higher at the surface (15%), decreasing at a constant rate until at 50 cm, where the value was 5% and then constant to 100 cm depth. However, there was a discontinuity in the bulk-density at 50 cm.

3. ACTIVITY MEASUREMENTS

Cs-137 activity was measured with a HPGe coaxial detector, with 1.88 keV resolution, 14% relative efficiency and coupled to a 4 K multichannel analyser. The detector was shielded with lead, 10 cm thick (in a 4 π geometry), and lined with 2 mm of Cu. A background reduction > 3 times
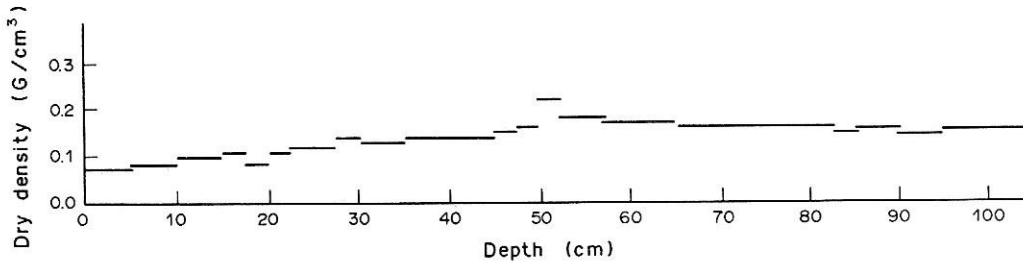
Fig. 2. Dry-density profile in the lacustrine sediment.

Table 1. Efficiency (%) for different γ-ray energies used in 7 different calibration measurements, as well as the mean efficiencies obtained (the uncertainty, in the efficiency of each determination is generally < 4%)

Energy (keV)	Efficiency (%)							Aver. effic. (%)
121.73	6.20	6.40	6.25	6.50	6.30	6.28	6.40	6.33 ± 0.10
244.69	3.65	3.58	3.66	3.58	3.53	3.61	3.56	3.59 ± 0.05
344.27	2.86	2.93	2.91	2.95	2.88	2.94	2.93	2.92 ± 0.03
411.11	2.19	2.19	2.17	2.10	2.17	2.22	2.16	2.17 ± 0.04
661.64	1.45	1.46	1.43	1.43	1.44	1.43	1.40	1.43 ± 0.02
688.63	1.32	1.25	1.28	1.32	1.29	1.36	1.28	1.30 ± 0.04
778.89	1.20	1.23	1.22	1.24	1.23	1.22	1.21	1.22 ± 0.02
964.01	0.96	0.98	0.97	0.98	0.96	0.96	0.97	0.97 ± 0.01
1112.02	0.87	0.87	0.91	0.88	0.83	0.85	0.83	0.86 ± 0.04
1299.04	0.74	0.74	0.73	0.73	0.74	0.74	0.72	0.73 ± 0.01
1407-95	0.68	0.68	0.70	0.68	0.68	0.69	0.68	0.69 ± 0.01

in the 662 keV Cs-137 γ-ray region, was obtained, This system proved to be sufficient for the low-level Cs-137 measurements. A γ-ray spectrum was taken for each sediment layer and the Cs-137 specific activity, A, in mBq/g (dry wt) was obtained using the formula:

$$A = \frac{50}{3} \frac{\Sigma(^{137}Cs)}{I.\,T.\,M.\,\varepsilon_{ph}} \quad (mBq/g) \tag{1}$$

$I = 0.8512$ being the intensity of the 662 keV γ-ray, T the counting time in min, M the mass of the sample in grams and ε_{ph} the photopeak efficiency: Σ (Cs-137) is the net area under the Ba-137m photopeak.

A background of $\simeq 0.0035 \pm 0.0008$ cps from Cs-137 fallout, which is present in the background spectrum, is subtracted from the total number of counts in the studied region, to give Σ(Cs-137). However, a careful analysis of the energy region was carried out in order to avoid interference of the 665.6 keV Bi-214 γ-ray peak when calculating Σ(Cs-137). This is especially important for weak samples and long counting times, for which the presence of this last photopeak becomes significant.

The value of ε_{ph} for Cs-137 was determined by preparing standard samples in which known activities of Eu-152 and Cs-137 were carefully mixed with material from the deepest layers of each core. The deepest did not show Cs-137 activity.

With these standard samples, the dependence of ε_{ph} on γ-ray energy is reproduced, since the counting geometry was fixed for all measurements. Clearly, the mass of the standard samples was similar to the experimental samples and, to minimize coincidence effects in the detector, the activity of Eu-152 was reduced to 1500 Bq. Calibration in the 120–1400 keV energy interval was achieved using Eu-152 (10 γ-ray energies) and Cs-137.

As an example of the calibration procedure the results obtained for the marine sediment are given in Table 1. The standard sample was measured with the HPGe γ-ray spectrometer under the
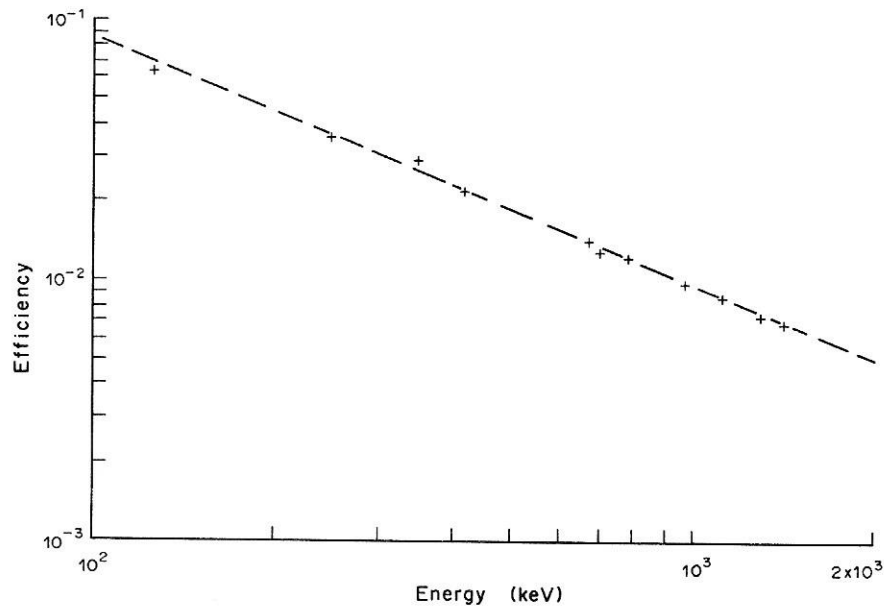
Fig. 3. Plot of the fitting efficiency-energy, using the HPGe detector for the marine calibration sample. Similar results were obtained with the lacustrine calibration sample.

same conditions as the real samples, and ε_{ph} obtained from:

$$\varepsilon_{ph} = \frac{\Sigma_i}{T.I_i.A} \tag{2}$$

where Σ_i is the net area under the ith photopeak, I_i is the intensity of the corresponding γ-ray, A is the activity of the γ-ray emitter and T is the counting time.

To ensure that the Eu-152 and Cs-137 activities were homogeneously distributed in the sediment, the sample was carefully mixed prior to the measurement. This operation was repeated seven times. Values of ε_{ph} which were obtained, are given in columns 2 to 8 of Table 1. In column 9 of the same Table a mean % value for ε_{ph} for each γ-ray energy is presented. The results clearly validate the calibration method and show that the standard activities were well mixed in the sediment.

In Fig. 3 the mean ε_{ph} is plotted against γ-ray energy (E_ϕ), for the marine sediment. To obtain a working expression, these data were fitted to a straight line as

$$\text{Ln}(\varepsilon_{ph}) = a\,\text{Ln}(E_\phi) + b \tag{3}$$

with $a = -0.941 \pm 0.027$, $b = 1.852 \pm 0.169$ and $r = -0.9986$. The fit is also shown in Fig. 3. A $\varepsilon_{ph} = (1.431 \pm 0.028)\%$ for Cs-137 was obtained from equation (3).

It may have been worthwhile to consider more complex ε_{ph} vs E_ϕ functions for fitting: [e.g. Sanchez-Reyes *et al.* (1987)]. However, a negligible improvement would have been obtained in the precision of A for the real samples [equation (1)], since it is essentially governed by the error in Σ (Cs-137) which is a consequence of the low activities of the sediment samples.

A final comment can be made concerning the use of Eu-152 as a standard source in addition to Cs-137. By using both radionuclides an additional test of homogeneity was obtained, since the single ε_{ph} for the Ba-137m γ-ray agrees quite well with prediction based on the Eu-152 data.

4. RESULTS AND DISCUSSION

The marine sediment

The determinations of Cs-137, Pb-210 and Ra-226 concentrations were carried out on aliquot samples of the same core material. In Fig. 4, the results of Pb-210 and Ra-226 are given. They were
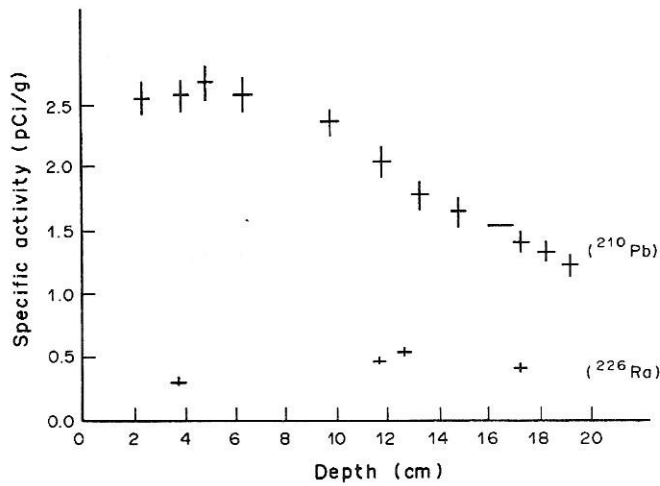
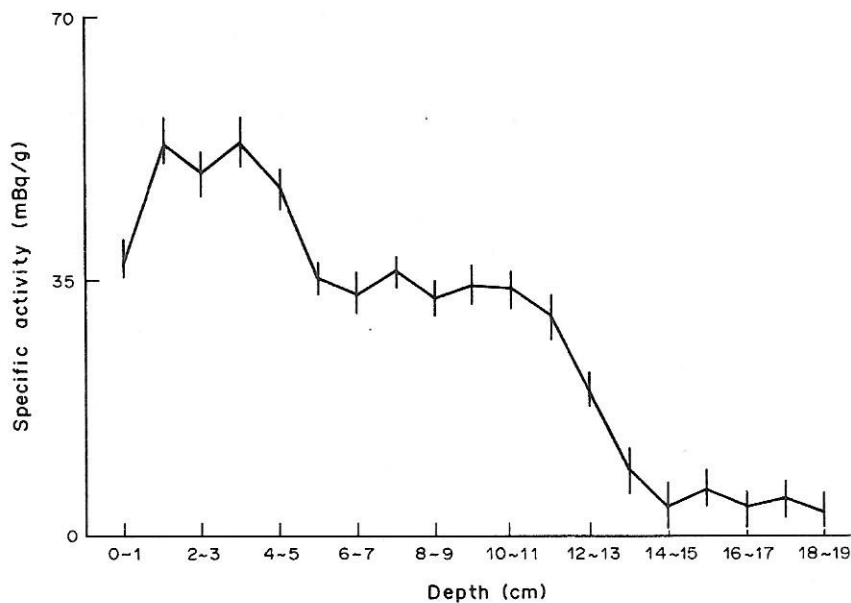Fig. 4. Pb-210 and Ra-226 specific activity profiles in the marine sediment.

Fig. 5. Cs-137 (mBq/g) profile of the marine sediment.

obtained by using well known techniques which can be found, for example in Eakins and Morrison (1976), El-Daoushy (1981) and El-Daoushy and Garcia-Tenorio (1988).

Little information can be obtained from the specific activity profile of Pb-210 alone since the sediment core is short and serious limitations exist for the application of the Pb-210 dating models. Furthermore, the first few cms of the profile show a constant Pb-210 specific activity. This is frequently found in other systems (Smith and Walton, 1980; Chanton *et al.*, 1983) and can be related to physical or biological effects, for example, that may produce mixing and affect data treatment and interpretation.

The Cs-137 activity profile for the same core is more informative (see Fig. 5). Three well-defined zones are apparent: Zone A from 0–1 cm to 5–6 cm; zone B, from 6 cm to 14–15 cm, and the rest, zone C. Some initial remarks are justified.

Since the residence time of Cs-137 in marine water is long, the activity profile in the core cannot directly reflect the Cs-137 atmospheric flux pattern. Indeed, only a fraction k $(0 \leqslant k \leqslant 1)$ of the Cs-137 content in the water column will be incorporated in the sediment each year. Thus, it is possible to find Cs-137 activity in layers corresponding to years where no appreciable amounts of

Cs-137 existed in the atmosphere. However, it seems clear that an increase of Cs-137 activity in the profile should be related to the year 1954, where the Cs-137 in the atmosphere notably increased due to weapon-tests. Also, the Cs-137 maximum should be assigned to the year 1963–64 (Jaakola *et al.*, 1983). Indeed, in the limiting case of $k \to 0$, the pattern of Cs-137 concentration in seawater continues to exhibit a maximum in 1963, since it behaves as the integrated atmospheric flux function.

From this point of view, the layer 14–15 cm can be assigned to the year 1954 (the Cs-137 activity in the interval 14–19 cm should be related to diffusion along the sediment, or to partial mixing.) On the other hand, since the core was sampled in 1984, the layer at 0–1 cm must be associated to this year. Hence, the sedimentation rate, S, can be estimated from the following formula:

$$S = \frac{\Sigma \, \rho_i x_i}{t} \qquad (\text{g} \cdot \text{cm}^{-2} \cdot \text{y}^{-1})$$

where the summation is extended over n layers which cover the time interval t: ρ_i and x_i being the dry-density and thickness respectively of the ith layer. Thus, using the dry densities data previously determined from the water content and the loss on ignition in each layer, a sedimentation rate for the zone from 1984 to 1954 of $S = 0.32$ g $\cdot$ cm^{-2} $\cdot$ y^{-1} is found.

This sedimentation rate should be interpreted as an average sedimentation rate, and hence, it does not exclude the possibility of sporadic changes in the sedimentation rate during the formation of the sediment.

However, this average sedimentation rate can be confirmed by information obtained from the same Cs-137 profile. By using the time frame obtained, the year 1963 can be placed around the layer 9–10 cm. In consequence, as expected, the plateau of zone B should be related to important radioactivity input which occurred during the 1960s. And the zone A, can be explained on the basis of the influence of the reprocessing plant at Sellafield as recommended by Aarkrog *et al.* (1980) who measured Cs-137 in sediments sampled from the neighbouring areas.

In zone A, the increase in Cs-137 from the layer 5–6 cm to the 0–1 cm cannot be explained on the basis of new atmospheric inputs.

Indeed, by using the calculated sedimentation rate (0.32 g $\cdot$ cm^{-2} $\cdot$ y^{-1}), the layer 5–6 cm would correspond to the year 1972–1973, indicating an increasing activity towards recent years. This agrees with available information on Cs-137 released from Sellafield which started in 1971, reaching its maximum value in 1975, and that the Cs-137 transit time from the Irish Sea to the area of Kattegat is *ca* 3 years (Aarkrog *et al.*, 1980; Notter, 1986).

An additional argument can be used to confirm the influence of Sellafield releases on the sediments. As Aarkrog *et al.* (1980) calculated, the Cs-137 activity of fallout origin per unit area in sediments from the zone studied amounts to 40 mCi/km². For the sediment studied a total deposited activity of 88 mCi/km² can be found. Additional non-atmospheric Cs-137 releases are affecting this sediment, as expected if the total deposited activity in zone A (45 mCi/km²) is taken into account.

It is also interesting to note that the Pb-210 and Cs-137 profiles for this sediment, can be explained by using a model, based on an assumption of partial mixing in the core (Abril *et al.*, 1992). It is interesting to indicate that the average sedimentation rate obtained by this model is in agreement with the sedimentation obtained in this section using the Cs-137 data. It is concluded that the Cs-137 profile is a powerful tool for sediment dating.

The lacustrine sediment

The contribution of Cs-137 to core dating differs from the previous case, but it is important. In Fig. 6 the Pb-210 and Ra-226 activity profiles for the core are presented. An unusual activity pattern can be observed which makes dating the core difficult. Bioturbation and/or physical mixing (Benninger *et al.*, 1979; Fisher *et al.*, 1980) or changes in the sedimentation rate (Oldfield and Appleby, 1984) could explain part of, or the whole, activity profile.
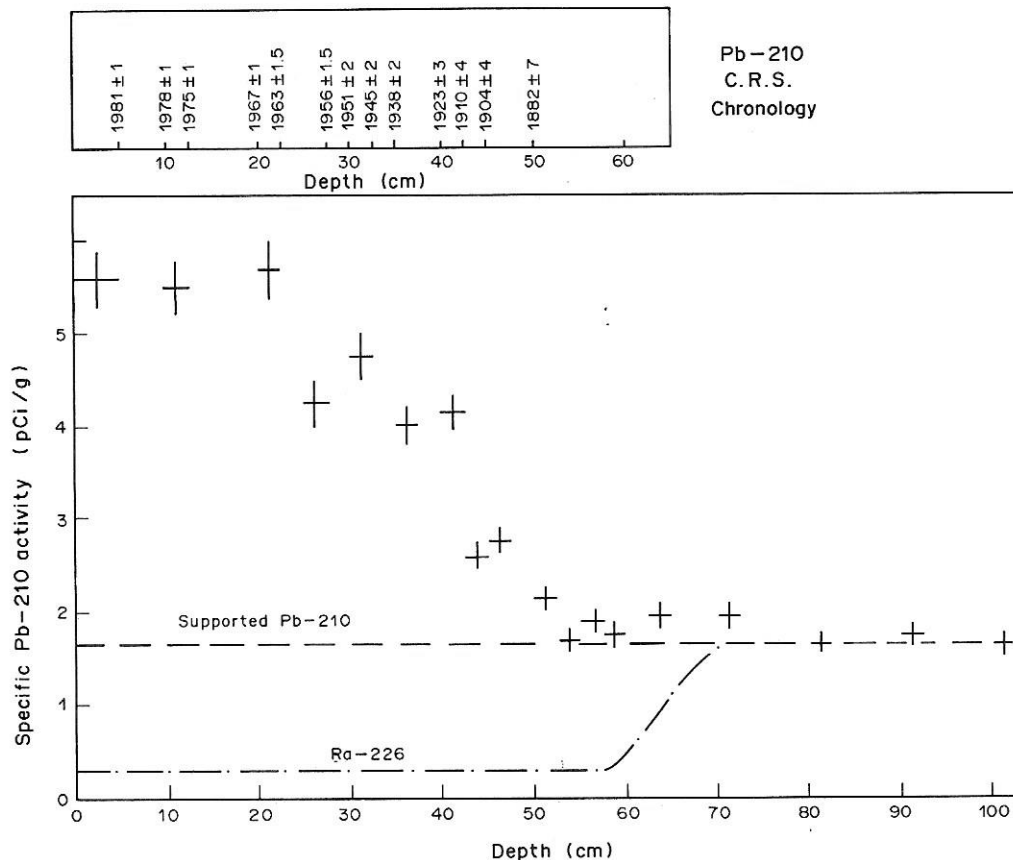
Fig. 6. Pb-210 (pCi/g) profile of the lacustrine sediment along with the chronology established by using the C.R.S. model.

Cs-137 activities for the same core are presented in Fig. 7. It is clear that bioturbation and/or physical mixing seem not to be the reasons for the unusual Pb-210 profile, since they would have produced a flat distribution of the Cs-137 activity (Robbins, 1978).

This suggests that changes in the sedimentation rate could have occurred along the time. The Constant Rate of Supply (C.R.S.) (Appleby and Oldfield, 1983; El-Daoushy, 1987; Oldfield and Appleby, 1984) model was applied to the Pb-210 data, and the chronology obtained is displayed in Fig. 6. Application of the C.R.S. model is only a way of explaining the Pb-210 profile. However, the time-frame deduced from the Cs-137 activity pattern (see Fig. 7) clearly agrees with that obtained by the C.R.S. model for Pb-210. This suggests that the hypothesis used, of changes in the sedimentation rate, could be reflecting the reality. In fact, the C.R.S. model successfully explains the lowering of the lake water level which occurred during the end of the nineteenth century (Section 2). Indeed, the chronology of Fig. 7 places this event roughly in the interval 50–60 cm, where anomalous behaviours of bulk density (Fig. 2) and Pb-210 data (Fig. 7) seem to have occurred.

An additional argument, which may support the hypothesis of changes in the sedimentation rate, is provided by calculating the Pb-210 flux in the sediment core. The normal Pb-210 atmospheric fluxes in Scandinavia range from 3.7 to 7.4 mBq cm^{-2} y^{-1} (El-Daoushy et al., 1982; El-Daoushy, 1986). However, the calculated Pb-210 flux in the core studied amounts to 19.6 $\pm$ 0.19 mBq cm^{-2} y^{-1}, revealing that additional sources of unsupported Pb-210 (from the drainage area) have been regularly deposited in the lake. This fact is confirmed when the integrated amount of Cs-137 in the sediment is calculated: it is found to be twice the expected value if the only source of Cs-137 was the atmosphere.

The Ra-226 data demonstrates a different sediment source to the lake during the past two centuries or so. This has also been documented for other lakes in the region.

These comments would not have been possible without the support of the Cs-137 activity profile,
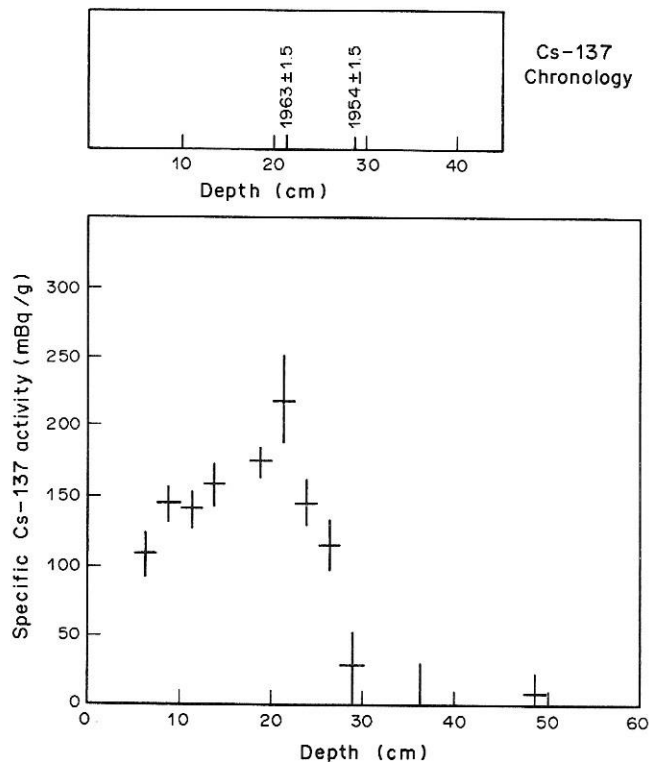
Fig. 7. Cs-137 (mBq/g) profile in the lacustrine sediment, together with the chronology established using this radionuclide.

which provides the hint, in this case, that application of one model is not only a mathematical tool to explain the experimental data. Alternative techniques with improved accuracies are of special importance in cases where Pb-210 data give partial or ambiguous results.

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