Dating of sediments from Lake Zurich (Switzerland) with ^{210}Pb and ^{137}Cs

By H. N. Erten*, H. R. von Gunten**, E. Rössler* and M. Sturm***

  * Eidg. Institut für Reaktorforschung, CH–5303 Würenlingen, Switzerland

 ** Eidg. Institut für Reaktorforschung and Anorg.-chemisches Institut, Universität Bern, CH–3000 Bern 9, Switzerland

*** Swiss Federal Institute for Water Resources and Water Pollution Control (EAWAG/ETH), CH–8600 Düben-dorf, Switzerland

ABSTRACT

^{210}Pb- and ^{137}Cs-measurements, and varve counting have been used to date sediment cores from Lake Zurich (Switzerland). Two cores from different water depths were dated with ^{210}Pb/^{210}Po and revealed sediment accumu-lation rates of (0.055 ± 0.015) g·cm^{-2}·y^{-1} and (0.09 ± 0.03) g·cm^{-2}·y^{-1}, respectively. A comparable rate of (0.07 ± 0.01) g·cm^{-2}·y^{-1} has been obtained from ^{137}Cs measurements. These rates were confirmed by annual layer (varve)-counts which lead to rates of 0.07 g·cm^{-2}·y^{-1}. Constant ^{210}Pb activities were observed in the top 6 cm of the sediment cores. This constancy is generally explained in the literature by mixing processes caused by bioturba-tion and by distortion during coring operations of the uppermost water-rich fluffy sediments. However, the distinct ^{137}Cs-maxima and the regular and undisturbed varve lamination of the top sediment observed in the cores of Lake Zurich contradict this assumption. In addition, measurements of ^{7}Be at the water/sediment interfaces proved complete sediment core recovery and mechanically undisturbed sediments. Remobilization processes are assumed to cause the observed constant ^{210}Pb activities. Remobilization may also be the reason for an incomplete ^{210}Pb inventory in the sediments which contain only about 50% of the fallout from atmosphere. The results of the ^{210}Pb dating should therefore be considered with some care. If existent, varve counting represents the easiest and most reliable means for dating lake sediments.

1. Introduction

The sediments of a lake contain the information on its history. Dating of sediments combined with other measurements allow to study the impact of nature and man on a lake and its environment. Goldberg [1] was the first to use ^{210}Pb to date the sedimentary record of the last 100 years. Since then this method has been refined and has been used in many geochronological studies (cf. references in [2]). ^{222}Rn, a noble gas in the decay chain of ^{238}U, is emanating from the earth's surface and from airborne dust into the atmosphere. Decay products of ^{222}Rn, including ^{210}Pb, are removed by dry fallout and wet precipita-tion, thus providing a continuous flux of ^{210}Pb onto land and water. In lakes or in the marine environment ^{210}Pb is scavenged by particles and is accumulated in the sediments. Here it decays with a half-life of 22.3 years and can be used for dating. Besides this

"unsupported" [210]Pb there is always a fraction of ^{210}Pb in the sediments which is in radioactive secular equilibrium with [226]Ra. This "supported" [210]Pb causes a background activity which limits the sensitivity of the method, and causes additional uncertainties. Extensive testing of nuclear weapons in the atmosphere between 1954–1963 caused a significant enhancement of the radioactive fall-out. The maximum fall-out in 1963 is usually well preserved in sediments of lakes and other depositional basins and can thus be used as a time marker. Among other nuclear fission products ^{137}Cs can easily be detected in the sediments by γ-ray spectroscopy [3]. This method requires complete recovery of the topmost sediment layers in order to obtain correct rates for the sediment accumulation since 1963. A prove for a complete core is the presence of [7]Be in the uppermost layer. [7]Be (half-life 53 days) is produced in the atmosphere by cosmic ray interactions and is carried to the sediments by particles.

The observation of well conserved annual layers (varves) proves the recovery of an undisturbed sediment profile. Likewise, varve counting represents an excellent and simple additional method to determine the age of a sediment [4].

2. Experimental

Sediment cores from Lake Zurich were recovered using a gravity corer with transparent PVC-tubes (inner diameter 6.3 cm). The cores were continuously sampled in 0.5 cm and 1 cm intervals immediatly after recovery. Varve counting was performed on photographs of cores which were sliced lengthwise with an electro-osmotic blade [5]. Two cores (no. I and II) were taken in August 1979 in the northern part of Lake Zurich in 61 m and 28 m
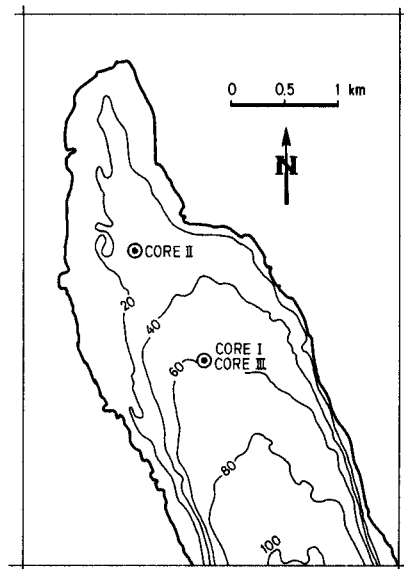

Figure 1: Map of the northern part of Lake Zurich showing the locations of the studied sediment cores (water depths are given in meters). Cores I and II were taken on August 21, 1979 at 61 m and 28 m water depths, respectively, core III was taken on December 21, 1981 at same location as core I.

water depth, respectively (fig. 1) and were used for ^{210}Pb and ^{137}Cs dating. An additional core (no. III) was sampled in December 1981 at the same location as core no. I to perform ^{7}Be-measurements and recheck porosity data. The measured water content of the three cores and an average sediment density of 2.65 g·cm^{-3} was used for the correction of compaction with depth. Parallel cores from each location were analyzed for their sedimentary structures and were used for varve counting. For the determination of ^{137}Cs and ^{7}Be the samples were freeze-dried and homogenized, and were measured simultaneously in a calibrated well-type γ-ray Ge(Li) detector.

Lead-210 was determined through its daughter ^{210}Po (half-life 138 days) which was at the moment of measurement in radioactive equilibrium with its parent. About one gram of dry sample was used. A known amount of ^{208}Po was added to the sample as yield tracer following the procedure of [6] and [7]. Coating of one side of a silver disc (diameter 1.5 cm) with RUTEX liquid rubber provided deposition of the activity on one side only. An almost quantitative plating was achieved in approximately 7 hours. The overall chemical yield was (90 ± 10)% based on the ^{208}Po tracer. The samples were positioned in a vacuum chamber at a distance of 1 mm from a Si surface-barrier detector (ORTEC 300 mm^2) with an α-energy resolution of about 20 keV full-width at half maximum (FWHM). The
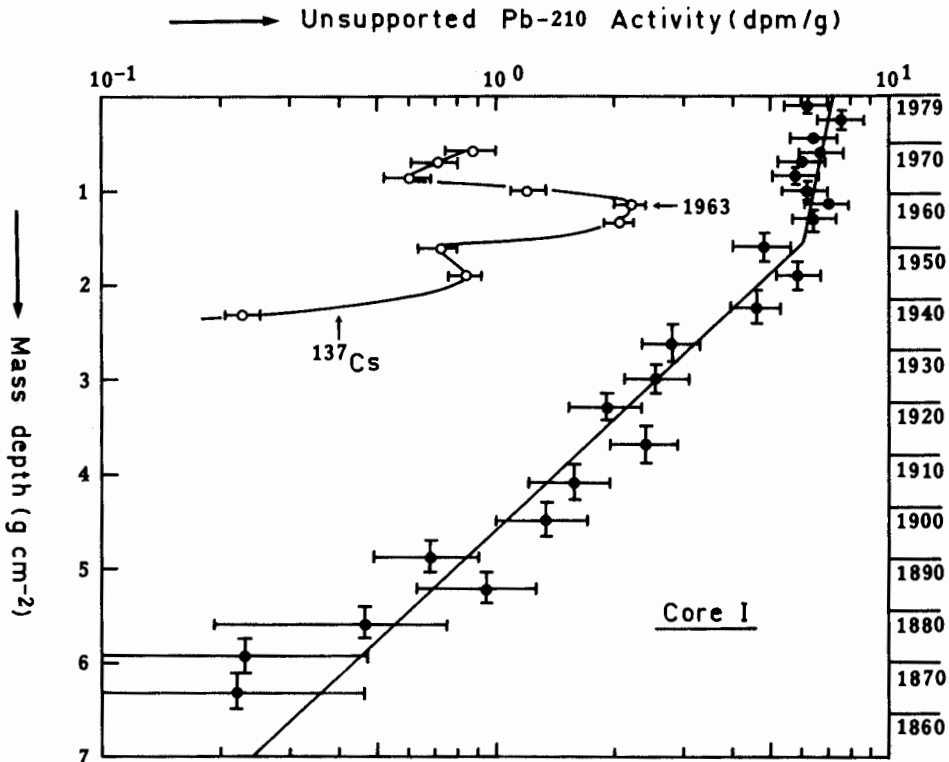

Figure 2: Activities of "unsupported" ^{210}Pb (see text) and ^{137}Cs from core no. I. Note constant ^{210}Pb activity in the uppermost 6 cm (1.5 g·cm^{-2}). These values have not been used for the least squares fit of the ^{210}Pb curve. The significant peak of ^{137}Cs (units in dpm·g^{-1}) rules out mixing of the top of the sediments.

efficiency for the 5.3 MeV alpha-line of ^{210}Po was determined to be $(18 \pm 2)\%$. The ^{210}Po activities were converted into ^{210}Pb activities applying corrections for the decay of ^{210}Pb during the time between sampling and separation of ^{210}Po, and for the decay of ^{210}Po since its separation.

3. Results and discussion

The results of the ^{210}Pb measurements from core I and II are shown in tables I and II. Total activities consist of a nearly constant "supported" fraction of ^{210}Pb, which is assumed to be in radioactive equilibrium with ^{226}Ra, and an "unsupported" (excess-) activity which decreases with depth. Mean "supported" activities of (2.11 ± 0.28) dpm $\cdot$ g^{-1} and (1.86 ± 0.25) dpm $\cdot$ g^{-1} for cores I and II, respectively, were obtained from the deepest sediment samples. These contain no or only negligible amounts of "unsupported" ^{210}Pb. The mean "supported" activities were substracted from the measured total activities of the respective core to yield the "unsupported" ^{210}Pb. The "supported" activities agreed to $\pm 20\%$ with activities of ^{226}Ra determined in some of the samples by γ-ray counting ^{214}Bi on a Ge (Li) detector. Errors (1σ) in the activities include counting statistics, uncertainties in the detector efficiency calibration and in the determination of the chemical yields. Errors in the "unsupported" ^{210}Pb activities, in addition, include the uncertainty in the value of "supported" ^{210}Pb. Z is the actual sediment sample depth below the sediment/water interface which is converted into mass-depth using the measured water content and a sediment density of 2.65 g $\cdot$ cm^{-3}.

Figure 2 shows a graphical presentation of the results from core I. One observes a nearly constant ^{210}Pb activity within about 6 cm $(1.5$ g $\cdot$ cm$^{-2})$ below the sediment/water interface, a range which contains the sediment accumulation of about 25 years. Equally constant ^{210}Pb activities near the water/sediment interface have been reported in sediments from Lake Michigan [8], from Lake Huron [9] and from other deposits [6, 10]. In order to explain this constant activity Robbins et al. developed a steady state rapid mixing model [9]. Its basic assumption is that sediments within this region are mixed at a rate much larger than the rate of deposition of sediments.

The results from Lake Zurich contradict a fast mixing of the topmost layers of sediment. First, the distribution of ^{137}Cs in the cores shows a distinct maximum in activity at about 6 cm sediment depth which is assumed to correspond to the year 1963. The ^{137}Cs distribution in core I is shown in figur 2. Its pronounced peak and the observed fine structure of the curve prove that physical mixing could not have been occured within this part of the sedimentary record. Second, ^{7}Be was measured $[(0.5 \pm 0.1)$ dps $\cdot$ g$^{-1}]$ in the uppermost sediment sample of core III, thus demonstrating the completeness of our core recovery. Since deeper samples contained no measurable ^{7}Be activity (lack of γ-ray peak at 478 keV), the assumption of mixing of the topmost centimeters cannot be valid for these sediments. Third, texture analysis of the sediment cores showed well developed annual layers, as described by [11] and [12], exhibiting an undisturbed succession of distinct light (summer) and black (winter) laminations since about 100 years. Therefore mixing or bioturbation did not occur during the last decades at least not for the solid parts of the sediment. A possibility to explain the constant values of ^{210}Pb in our cores as well as in the sediments described in the literature could be pore water remobilization of ^{210}Pb and/or ^{210}Po from the top layers of the sediment. Preliminary results of ^{210}Pb

Table 1. *Total activities of ^{210}Pb* (sum of "supported" and "unsupported" activities, see text) of sediment core I. Z is the actual sample depth below the water/sediment interface, while *mass-depth* describes sediment depth below the water/sediment interface corrected for compaction using the measured water content of the cores *(porosity)* and an average sediment density of 2.65 g·cm^{-3}.

z (cm)	porosity $\rho = 2.65$	mass-depth (g·cm^{-2})	total ^{210}Pb activity (dpm·g^{-1})	z (cm)	porosity $\rho = 2.65$	mass-depth (g·cm^{-2})	total ^{210}Pb activity (dpm·g^{-1})
0–1	0.97	0.08	8.37 ± 0.84	18–19	0.87	5.91	2.34 ± 0.23
1–2	0.94	0.24	9.69 ± 0.97	19–20	0.85	6.31	2.33 ± 0.23
2–3	0.92	0.45	8.63 ± 0.86	20–21	0.85	6.70	2.27 ± 0.23
3–3.5	0.91	0.57	8.89 ± 0.89	21–22	0.84	7.13	2.18 ± 0.22
3.5–4	0.90	0.70	8.15 ± 0.82	22–23	0.84	7.55	2.22 ± 0.22
4–4.5	0.89	0.85	7.95 ± 0.80	23–24	0.84	7.98	2.04 ± 0.20
4.5–5	0.89	0.99	8.26 ± 0.83	24–25	0.84	8.40	2.66 ± 0.27
5–5.5	0.89	1.14	9.12 ± 0.91	25–26	0.84	8.82	2.30 ± 0.23
5.5–6	0.88	1.30	8.58 ± 0.86	26–28	0.84	9.67	2.29 ± 0.23
6–7	0.88	1.62	6.79 ± 0.68	28–30	0.84	10.52	1.90 ± 0.19
7–8	0.88	1.93	8.04 ± 0.80	30–32	0.83	11.42	1.66 ± 0.17
8–9	0.87	2.28	6.73 ± 0.67	32–34	0.83	12.32	1.59 ± 0.16
9–10	0.87	2.62	4.94 ± 0.49	34–36	0.83	13.22	1.68 ± 0.17
10–11	0.87	2.97	4.70 ± 0.47	36–38	0.83	14.12	1.89 ± 0.19
11–12	0.87	3.31	4.03 ± 0.40	38–40	0.83	15.02	1.88 ± 0.19
12–13	0.86	3.68	4.51 ± 0.45	40–42	0.83	15.93	1.92 ± 0.19
13–14	0.85	4.08	3.69 ± 0.37	42–44	0.82	16.88	2.48 ± 0.25
14–15	0.85	4.48	3.46 ± 0.35	44–46	0.82	17.83	2.11 ± 0.21
15–16	0.85	4.88	2.80 ± 0.28	46–48	0.82	18.79	2.15 ± 0.22
16–17	0.87	5.22	3.05 ± 0.31	48–50	0.82	19.74	2.40 ± 0.24
17–18	0.87	5.56	2.58 ± 0.26				

Table 2. *Total activites of ^{210}Pb* (sum of "supported" and "unsupported" activites) of sediment core II (cf. table I).

z (cm)	mass-depth (g·cm^{-2})	total ^{210}Pb activity (dpm·g^{-1})
1–2	0.24	6.61 ± 0.66
2–3	0.45	6.05 ± 0.61
3–3.5	0.57	7.86 ± 0.79
3.5–4	0.70	7.05 ± 0.71
4–4.5	0.85	7.33 ± 0.73
6.50	1.62	6.96 ± 0.70
11.50	3.31	4.03 ± 0.40
16.50	5.22	3.04 ± 0.30
21.50	7.13	2.18 ± 0.22
27.00	9.67	2.36 ± 0.24
37.00		1.91 ± 0.19
43.00		1.81 ± 0.18
47.00		1.79 ± 0.18

measurements in the interstitial water [13] seem, indeed, to support the remobilization hypothesis. This remobilization process could also explain the significant deficit in the ^{210}Pb inventory of the sediments of Lake Zurich. The expected inventory in the sediments was calculated for an atmospheric input of about 0.75 dpm m$^{-2} \cdot$ y^{-1}, which is obtained using a transfer velocity for ^{210}Pb of 0.9 cm $\cdot$ s^{-1} [14] and a ^{210}Pb concentration in the air of Switzerland of 12 fCi $\cdot$ m^{-3} [15]. The measured inventory of the two sediment cores shows a deficit which amounts to almost 50% of the calculated ^{210}Pb fall-out.

Using ^{210}Pb for the determination of sedimentation rates a constant flux of ^{210}Pb into the sediments was assumed. This assumption was justified by measurements of ^{210}Pb in monthly air samples at Fribourg (Switzerland) during the last 13 years, which proved constant average yearly concentrations of ^{210}Pb of (11.9 ± 0.4) fCi $\cdot$ m^{-3} [16]. It was further assumed that the sedimentation rate within the studied sediment range remained constant and that no postdepositional mobilisation and migration of ^{210}Pb had occured. The latter assumption is in contradiction with the observed constant activities in the top centimeters of the cores and with the ^{210}Pb inventory, but holds probably more or less for the deeper parts of the sediment. Finally it was assumed that the "supported" ^{210}Pb values do not vary within the sediment. These assumptions correspond to the Constant Initial Concentration (C.I.C.) model which leads to sedimentation rates of $(0.055 \pm 0.014$ g $\cdot$ cm$^{-2} \cdot$ y^{-1} for core I and (0.09 ± 0.03) g $\cdot$ cm$^{-2} \cdot$ y^{-1} for core II. Samples from the uppermost sediment, which showed constant ^{210}Pb activities were not used in this calculation. Due to a limited number of measured samples in core II this result is less accurate. The results are in agreement with rates for sediment accumulation derived from ^{137}Cs measurements in core I, which show a rate of (0.071 ± 0.01) g $\cdot$ cm$^{-2} \cdot$ y^{-1} for the last 16 years. Counting of annual layers in cores I and III showed values of 0.07 g $\cdot$ cm$^{-2} \cdot$ y^{-1} for the last 30 years. It should be noted that the errors in the ^{210}Pb dating method are rather large, if they are taken realistically. But even so, they do not include the additional uncertainties which may be caused by the possible remobilization of ^{210}Pb. Nevertheless, the results prove the applicability of these methods for dating recent lake sediments and support dating results of Lake Zurich sediments from other authors [11, 17]. One should, however, strongly point to the fact that varve counting in sediments is, if applicable, much simpler and more accurate than nuclear dating methods.

ZUSAMMENFASSUNG

Sedimente aus dem Zürichsee wurden mit Hilfe von drei unabhängigen Methoden datiert. Für ^{210}Pb ergaben sich Sedimentationsraten von 0.055 g $\cdot$ cm^{-2}y^{-1} bzw. 0.09 g $\cdot$ cm^{-2}y^{-1}. Die ^{137}Cs-Datierung und die Auszählung von Jahreslagen (Varven) ergab 0.07 g $\cdot$ cm^{-2}y^{-1}. Ähnlich wie in anderen Seen wurden auch im Zürichsee in den obersten 6 cm aller untersuchten Kerne konstante ^{210}Pb-Aktivitäten festgestellt. Dies wird in der Literatur in der Regel auf Bioturbation oder auf sekundäre Durchmischung der stark wassergesättigten Sedimente bei der Proben-

nahme zurückgeführt. Im Zürichsee widersprechen die Erhaltung von millimetermächtigen Jahreslagen, das Auftreten eines ausgeprägten ^{137}Cs-Maximums und ganzjährig anoxische Bedingungen an der Sediment/Wassergrenzfläche solchen Deutungsversuchen. Auch das Vorhandensein des kurzlebigen Isotopes [7]Be (HWZ 53 Tage) in den obersten Sedimentschichten beweist eine komplette und ungestörte Probennahme. Für die beobachtete konstante ^{210}Pb-Aktivität werden daher Remobilisationsprozesse nach der Ablagerung der Sedimente verantwortlich gemacht. Nachträgliche Remobilisierung könnte auch die Ursache für das beobachtete unvollständige ^{210}Pb-Inventar in den Sedimenten sein, das nur ca. 50% des atmosphärischen Ausfalls beträgt. Die Untersuchungen zeigen, dass die Ergebnisse der ^{210}Pb-Datierung in diesem eutrophen See mit Vorsicht interpretiert werden müssen und dass die Auszählung von Jahreslagen – falls vorhanden – die am wenigsten aufwendige und genaueste Methode zur Datierung von Seesedimenten darstellt.

REFERENCES

1   Goldberg E. D., Radioactive Dating, IAEA STI/PUB/68, 121 (1963)
2   Krishnaswami S. and Lal D., In: Lakes-Chemistry, Geology, Physics (Ed. A. Lerman), Springer (1978)
3   Krishnaswami S., Lal D., Martin J. M. and Meybeck M., Earth Planet, Sci. Lett. *11*, 407 (1971)
4   Ludlam S. D., Chem. Geol. *44*, 85 (1984)
5   Sturm M. and Matter A., J. Sed. Petrol. *42*, 987 (1972)
6   Eakins J. D. and Morrison R. T., Int. J. of Appl. Rad. Isotopes *29*/531 (1979)
7   Figgins P. E., The Radiochemistry of Polonium, U. S. Atomic Energy Commission, NAS-NS-3037, p. 29–30 (1961)
8   Robbins J. A. and Edgington D. N., Geochim. Cosmochim. Acta *39*, 285 (1975)
9   Robbins J. A., Krezoski J. R. and Mozley S. C., Earth Planet. Sci. Lett. *36*, 325 (1977)
10 Smith J. N. and Walton A., Geochim. Cosmochim. Acta *44*, 225 (1980)
11 Züllig H., Schweiz. Z. Hydrol. *44*/1 (1982)
12 Sturm M., EAWAG-News *16*, 6 (1983)
13 Santschi P., personal communication
14 Turekian K. K., Benninger L. K. and Dion E. P., Geophys. Res. *88* (C9), 5411 (1983)
15 KUeR Report, 25 Jahre Radioaktivitätsüberwachung in der Schweiz, Eidg. Kommission zur Überwachung der Radioaktivität, Fribourg, Switzerland (1982)
16 von Gunten H. R. and Wegmüller F. (in prep.)
17 Kelts K. and Hsü K. J., Freshwater Carbonate Sedimentation, In: Lakes-Chemistry, Geology, Physics (Ed. A. Lerman), Springer (1978)

Addresses of the authors: 1) H. N. Erten, Dept. of Chemistry, Middle East Technical University, Ankara, Turkey
2) H. R. von Gunten, EIR, CH–5303 Würenlingen and Anorg.-chemisches Institut, Universität Bern, CH–3000 Bern 9, Switzerland
3) E. Rössler, EIR; CH–5303 Würenlingen, Switzerland
4) M. Sturm, EAWAG, CH-8600 Dübendorf, Switzerland*
*) EAWAG-Contribution Nr. 1099