PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY B BIOLOGICAL SCIENCES

Radiometric Dating of the United Kingdom SWAP Sites

P. G. Appleby, N. Richardson, P. J. Nolan and F. Oldfield

Phil. Trans. R. Soc. Lond. B 1990 **327**, 233-238
doi: 10.1098/rstb.1990.0057

Phil. Trans. R. Soc. Lond. B **327**, 233–238 (1990)
Printed in Great Britain

Radiometric dating of the United Kingdom SWAP sites

By P. G. Appleby[1], N. Richardson[2], P. J. Nolan[3] and F. Oldfield[2]

Departments of [1] Applied Mathematics and Theoretical Physics, [2] Geography and [3] Oliver Lodge Laboratory, Department of Physics, University of Liverpool, P.O. Box 147, Liverpool L69 3BX, U.K.

Measurements of ^{210}Pb by direct gamma assay have been used to date sediment cores from Surface Water Acidification Project (SWAP) study sites in the U.K. The results were checked against additional dating evidence from the artificial fallout isotopes ^{137}Cs and ^{241}Am. At one of the sites, Devoke Water in Cumbria, the ^{137}Cs and ^{241}Am data were crucial in identifying a recent sediment hiatus. At sites with recently afforested catchments the sediment record indicated substantial increases in accumulation rates.

Introduction

Accurate dating techniques are crucial to the use of sediment records for palaeolimnological reconstructions of recent environmental change. The precision to which this can be accomplished has been greatly enhanced over the past 20 years by the development of a range of radiometric techniques that use radioisotopes both from natural and artificial sources. The successful utilization of these methods depends on a clear understanding of the mechanisms by which the radioisotopes are incorporated into the sediments. The principal isotope for dating on the timescale of the past 100–150 years, the period of greatest relevance to the Surface Water Acidification Project (SWAP) Palaeolimnology Programme, is ^{210}Pb. By using this isotope, determination of lake sediment dates with a precision of five to ten years is often attainable. Artificial radioisotopes such as ^{137}Cs and ^{241}Am have been present in the environment only for the past 35 years and can be used for dating only the most recent sediments. The use of these isotopes does however provide a valuable check on the accuracy of the ^{210}Pb calculations. By using these techniques, our principal objective within the SWAP programme was to provide a reliable sediment chronology at each site. We report here on the results of this project.

Methods

Sediments from each core were measured non-destructively for ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am by direct gamma assay by using an Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detector (Appleby *et al.* 1986). Background suppression for this detector is achieved by using a 100 mm thick lead castle, a 305 mm diameter × 305 mm long sodium iodide (NaI(Tl)) escape suppression shield and a 3 mm thick copper lining. ^{210}Pb is measured by its gamma emissions at 46.5 keV and ^{226}Ra by the 295 keV and 352 keV γ rays emitted by its daughter isotope, ^{214}Pb; ^{137}Cs and ^{241}Am are measured by their emissions at 662 keV and 59.5 keV, respectively. The absolute efficiency of the detector has been measured by using a series of calibrated sources and sediment samples of known activity. The effect of self absorption of low energy γ rays has been estimated by using sources of different masses. Background counts are done at regular intervals to ensure maintenance of the low background characteristics.

^{210}Pb *dating*

^{210}Pb occurs naturally as one of the radioisotopes in the ^{238}U decay series. Radioactive disequilibrium between ^{210}Pb and its parent isotope, ^{226}Ra (half-life 1600 years), arises through the mobility of the intermediate gaseous isotope ^{222}Rn. A proportion of the ^{222}Rn formed by ^{226}Ra decay in soils diffuses into the atmosphere where it decays to ^{210}Pb. This is precipitated onto the land surface and into lakes where it is adsorbed onto sedimentary particles. Pathways by which ^{210}Pb accumulates in lake sediments are discussed in detail in Oldfield & Appleby (1984). ^{210}Pb activity in sediments in excess of the fraction that derived from decay of the *in situ* ^{226}Ra is called unsupported ^{210}Pb. It declines in accordance with the exponential radioactive-decay law and can be used for age determinations provided there is an appropriate model for estimating the initial activity. The ^{210}Pb half-life of 22.26 years makes it well suited to dating sediments laid down over the past 100–150 years. Unsupported ^{210}Pb is measured by subtraction of ^{210}Pb supported by the parent ^{226}Ra from the total ^{210}Pb activity. In most situations the supported ^{210}Pb can be assumed to be in radioactive equilibrium with the *in situ* ^{226}Ra. Figure 1*a* shows the total and supported ^{210}Pb activity against depth for a sediment core from Loch Chon in the Trossachs, Central Scotland. The unsupported ^{210}Pb activity in this core is shown in figure 1*b*.

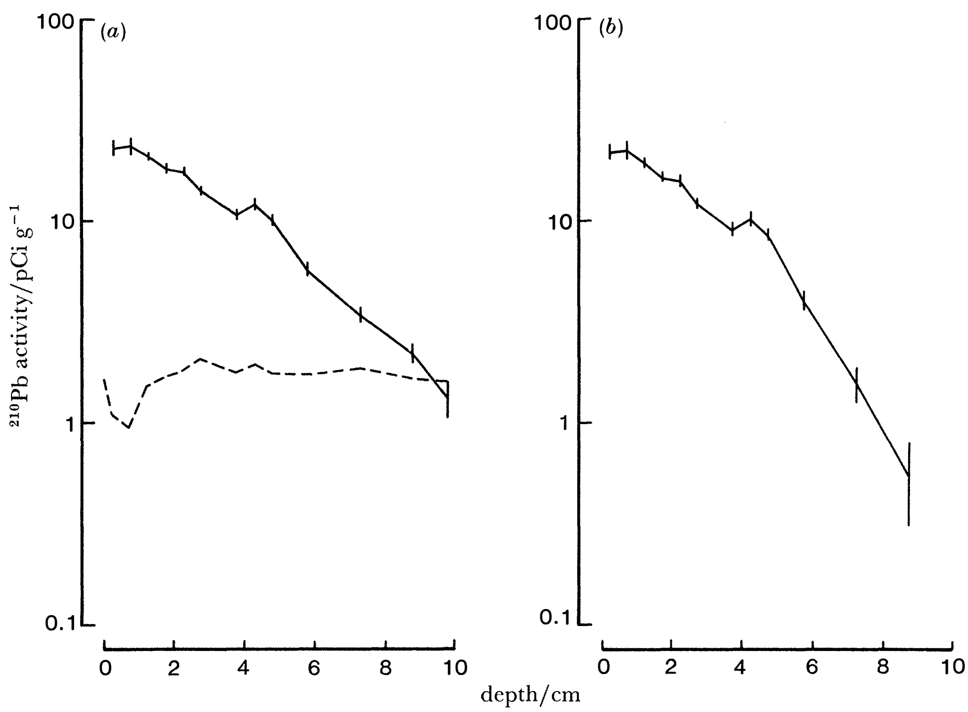

FIGURE 1. ^{210}Pb activity against depth in a core from Loch Chon, Scotland; (*a*) total (——) and supported (– – – –) ^{210}Pb and (*b*) unsupported ^{210}Pb.

There are two principal models for determining the initial activity of a sediment and hence for calculating ^{210}Pb dates, the CRS (constant rate of unsupported ^{210}Pb supply) model and the CIC (constant initial ^{210}Pb concentration) model. Variants of these models have been developed to account for processes such as sediment mixing. The CRS model (Appleby & Oldfield 1978;

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY B BIOLOGICAL SCIENCES

Robbins 1978) is perhaps the most widely accepted. It is based on the hypothesis that the ^{210}Pb supply is dominated by a constant direct atmospheric fallout. There is some evidence (Krishnaswamy & Lal 1978) that the atmospheric ^{210}Pb flux may be subject to short-term fluctuations, but as most sediment samples span several years, short-term variations will generally be smoothed out. This model will not be valid where there are alterations or interruptions to the ^{210}Pb supply due to, for example, sediment focusing or a sediment hiatus. In these cases dates are calculated either by the CIC model (where there is evidence for a constant primary accumulation rate) or by using a composite of both models. Factors governing model choice are set out in Appleby & Oldfield (1983) and Oldfield & Appleby (1984). In this study, the CRS model has generally been used, though dates have routinely been calculated by both models and elements of the CIC model incorporated where this has been thought appropriate.

^{131}Cs *and* ^{241}Am *dating*

Although ^{210}Pb is now routinely used for dating, problems frequently arise over the interpretation of data from sites with disturbed sediment records. In these cases, dates given by the artificial isotope ^{137}Cs may be of considerable value. Until the Chernobyl accident in 1986, the most common source of ^{137}Cs was fallout from the atmospheric testing of nuclear weapons. Where this isotope is strongly adsorbed onto sediments, the variation of the ^{137}Cs activity with depth in a core should reflect the fallout history, with the onset of fallout in 1954 and peak fallout in 1963 providing distinct chronological markers. These may be used to confirm ^{210}Pb dates where they are unambiguous, or to resolve ^{210}Pb dates where they are uncertain. In practice the value of ^{137}Cs dating has often been significantly reduced by the evident mobility of this isotope (Davis *et al.* 1984). The problem has recently been exacerbated by the fallout

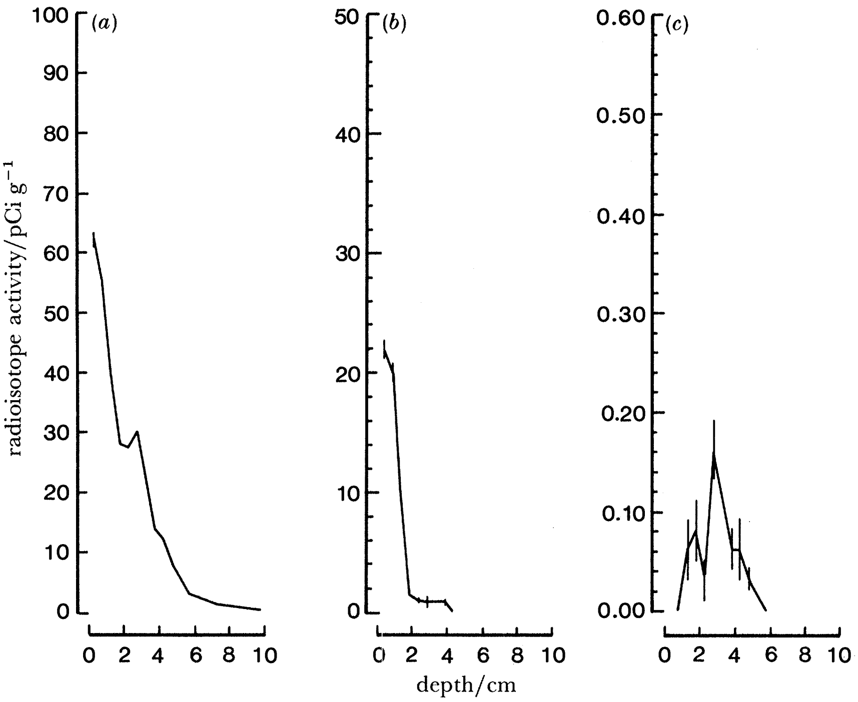

FIGURE 2. Artificial fallout radioisotopes against depth in a core from Loch Chon, Scotland; (*a*) ^{137}Cs, (*b*) ^{134}Cs and (*c*) ^{241}Am.

of ^{137}Cs from the Chernobyl accident. In regions of high fallout, downward diffusion of Chernobyl ^{137}Cs has obliterated the weapons-testing ^{137}Cs profile. In these cases, measurement of ^{241}Am, another fallout product from atmospheric nuclear weapons testing, may provide a useful alternative. The amount of ^{241}Am fallout is very small, only about 0.42% of the ^{137}Cs fallout (Eakins & Cambray 1985). Despite this, ^{241}Am has been detected in cores from many lakes, and evidence from a growing data set suggests that ^{241}Am is significantly less mobile than ^{137}Cs. Graphs of ^{137}Cs, ^{134}Cs and ^{241}Am activity against depth for the Loch Chon sediment core are shown in figure 2.

Results

^{210}Pb chronologies for Loch Chon calculated by using the CRS and CIC ^{210}Pb dating models are illustrated in figure 3. There is good agreement between the two sets of dates for the most recent sediments, both models indicating a reasonably constant accumulation rate since 1954 of 0.013 g cm^{-2} a^{-1}. This is supported by the ^{137}Cs and ^{241}Am data. The ^{137}Cs activity derived from weapons testing fallout (calculated by subtracting Chernobyl ^{137}Cs from the total ^{137}Cs activity) has a well defined peak at 2.75 cm, as does the ^{241}Am activity, and this level is dated to 1966 by the CRS model and 1964 by the CIC model. Both ^{210}Pb models indicate an abrupt transition in the early 1950s from an earlier accumulation rate of 0.0096 g cm^{-2} a^{-1} to the above more recent value. A consequence of this transition (Appleby & Oldfield 1978) is that CIC model dates below 4 cm are about 10 years younger than the CRS model dates. The increase in accumulation rate is presumably associated with the 1950s afforestation programme in the Loch Chon catchment; its onset is indicated by the reduction in unsupported ^{210}Pb activity at a depth of 3.75 cm (dated 1953–1956) shown in figure 1*b*. In view of this direct evidence in support of the CRS model assumptions, the CRS model dates were thought to be the more reliable.

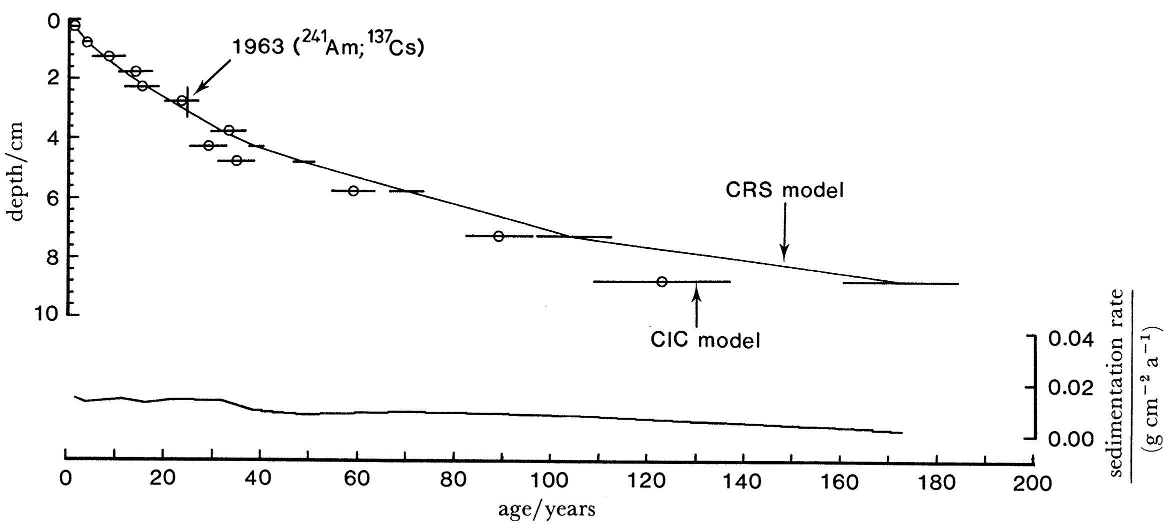

FIGURE 3. Depth against ^{210}Pb age and dry mass accumulation rates in a core from Loch Chon, Scotland, as determined by the CRS and CIC ^{210}Pb dating models. The graph also shows the depth dated to 1963 by the ^{137}Cs and ^{241}Am data.

The very high ^{137}Cs activity in the top 2 cm of the Loch Chon core can be attributed to fallout from the Chernobyl accident in May 1986. This core was obtained in 1987 and the origin of the ^{137}Cs is confirmed by the associated presence of the short-lived isotope ^{134}Cs (figure

2*b*). Chernobyl fallout was detected in most post-1986 cores and provides evidence of the retrieval of an undisturbed sediment–water interface.

Table 1 gives a summary of the radiometric parameters obtained from SWAP sites in the U.K. Mean accumulation rates are given in table 2. Results from individual sites, summarized below, have been presented in separate reports.

Table 1. Radiometric parameters of SWAP lake-sediment cores

site	unsupported ^{210}Pb surf. conc. (pCi g^{-1})	unsupported ^{210}Pb invent. (pCi cm^{-2})	unsupported ^{210}Pb flux (pCi cm^{-2} a^{-1})	^{226}Ra mean conc. (pCi g^{-1})	^{137}Cs invent. (pCi cm^{-2})	^{241}Am invent. (pCi cm^{-2})
Loch Sionascaig	53.5	12.7	0.40	1.03	10.7[a]	0.06
Lochan Dubh	10.3	11.3	0.35	1.65	7.9[a]	0.07
Loch Doilet	6.3	19.3	0.60	1.73	14.4[a]	0.07
Lochan Uaine	78.1	99.7	3.10	23.06	10.1[a]	—
Coire an Lochan	46.6	16.3	0.51	14.10	3.2[a]	—
Loch Chon	21.9	10.4	0.33	1.66	17.8[a]	0.04
Loch Tinker	29.6	20.4	0.64	1.33	23.4	0.10
Round Loch	24.9	10.6	0.33	1.47	6.3	0.06
Devoke Water	24.7	34.6	1.68[b]	0.94	29.3	0.51

[a] Denotes sites cored after May 1986.
[b] ^{210}Pb flux estimated from post-1963 inventory.

Table 2. Comparison of sediment accumulation rates at SWAP sites

site	mean accumulation rates (i) (g cm^{-2} a^{-1}) (*a*) post-1950	(i) (*b*) 1910–1950	(i) (*b*) pre-1910	(ii) (mm a^{-1}) (*a*) post-1950	(ii) (*b*) 1910–1950	(ii) (*b*) pre-1910
Loch Sionascaig	0.0083	0.0086	0.0091	0.75	0.72	0.74
Lochan Dubh	0.0148	0.0104	0.0074	1.56	0.77	0.47
Loch Doilet	0.0540	0.0384	0.0186	4.17	2.30	0.97
Lochan Uaine	0.0223	0.0169	0.0178	0.83	0.52	0.50
Coire an Lochan	0.0096	0.0096	—	0.30	0.30	—
Loch Chon	0.0133	0.0096	0.0096	1.13	0.52	0.44
Loch Tinker	0.0193	0.0223	0.0231	1.61	1.47	1.20
Round Loch	0.0113	0.0120	0.0184	1.51	1.30	1.85
Devoke Water	0.0525	—	—	2.90	—	—

Northwest Scotland

^{210}Pb dates for Loch Sionascaig were unambiguous and gave a constant sediment accumulation rate over the past 150 years of 0.0088 g cm^{-2} a^{-1} or 0.74 mm a^{-1}. The ^{137}Cs results for this core were of no chronological value, even when allowance was made for the contribution from Chernobyl fallout. The ^{137}Cs activity was greatest in the topmost sample, and declined monotonically with depth. A small ^{241}Am peak was detected at a depth consistent with the ^{210}Pb dates.

Loch Doilet and Lochan Dubh both have nonlinear ^{210}Pb profiles, suggesting accelerating sediment accumulation rates. The increase has been very much more pronounced in Loch Doilet, presumably because of the disturbances to its catchment arising from the afforestation programme that began in the 1930s. Both cores have relatively well-defined ^{137}Cs and ^{241}Am peaks at depths reasonably consistent with the ^{210}Pb dates. Traces of Chernobyl fallout were observed at the top of the Loch Doilet core.

The Trossachs

Despite its much smaller catchment, Loch Tinker has experienced significantly higher sediment accumulation rates than Loch Chon. However, whereas the core from Loch Tinker has a linear ^{210}Pb profile suggesting reasonably constant accumulation rates over the past 150 years, the data from Loch Chon indicates accelerated accumulation rates over the past 35 years, presumably due to afforestation in the early 1950s. In both cores the ^{210}Pb dates are supported by ^{137}Cs and ^{241}Am data. The enhanced ^{137}Cs activities at the top of the Loch Chon core derive from Chernobyl fallout and have virtually obliterated the 1963 peak caused by weapons testing fallout.

The Cairngorms

Lochan Uaine and Coire an Lochan are both characterized by very high unsupported ^{210}Pb activities in the topmost sediments. This usually indicates very slow sediment accumulation rates, though at both these sites the very high ^{226}Ra activities may be a contributory factor. In the Lochan Uaine core ^{210}Pb equilibrium (corresponding to *ca.* 150 years of sediment accumulation) is achieved at 8 cm. In Coire an Lochan it appears that equilibrium is achieved at only 2.25 cm. The ^{210}Pb dates for Lochan Uaine suggest a modest increase in sediment accumulation since 1970. This is supported by ^{137}Cs data.

Galloway and Cumbria

The ^{210}Pb data from Round Loch of Glenhead indicates constant sedimentation since *ca.* 1900. There is some suggestion of higher accumulation rates in the 19th century, though this has not been observed in other cores from this lake. The ^{210}Pb dates are consistent with the ^{137}Cs and ^{241}Am data. The data from Devoke Water is contradictory in that ^{210}Pb equilibrium appears to have been achieved at 12.5 cm, though the ^{137}Cs and ^{241}Am data point clearly to an early 1960s date for sediments at a depth of 7 cm. This incompatibility is explained by a sediment hiatus at 9 cm, dated 1954. The high surface concentrations coupled with the high inventories (table 1) suggest that the primary sedimentation rate is quite low but that the core is from a site of intensive sediment focussing. The ^{210}Pb and ^{137}Cs results suggest that the primary sedimentation rate is reasonably constant. On this assumption the ^{210}Pb indicates a loss of *ca.* 50 years from the sediment record.

References

Appleby, P. G. & Oldfield, F. 1978 The calculation of ^{210}Pb dates assuming a constant rate of supply of unsupported ^{210}Pb to the sediment. *Catena* **5**, 1–8.

Appleby, P. G. & Oldfield, F. 1983 The assessment of ^{210}Pb from sites with varying sediment accumulation rates. *Hydrobiologia* **103**, 29–35.

Appleby, P. G., Nolan, P., Gifford, D. W., Godfrey, M. J., Oldfield, F., Anderson, N. J. & Battarbee, R. W. 1986 ^{210}Pb dating by low background gamma counting. *Hydrobiologia* **141**, 21–27.

Davis, R. B., Hess, C. T., Norton, S. A., Hanson, D. W., Hoagland, K. D. & Anderson, D. S. 1984 ^{137}Cs and ^{210}Pb dating of sediments from soft-water lakes in New England (U.S.A.) and Scandinavia, a failure of ^{137}Cs dating. *Chem. Geol.* **44**, 151–185.

Eakins, J. D. & Cambray, R. S. 1985 Studies of environmental radioactivity in Cumbria, part 6. Report AERE-R 11182. Harwell: United Kingdom Atomic Energy Research Authority.

Krishnaswamy, S. & Lal, D. 1978 Radionuclide limnochronology. In *Lakes, chemistry geology & physics* (ed. A. Lerman), pp. 153–177. New York: Springer-Verlag.

Oldfield, F. & Appleby, P. G. 1984 Empirical testing of ^{210}Pb-dating models for lake sediments. In *Lake sediments and environmental history* (ed. E. Y. Haworth & J. W. G. Lund), pp. 93–124. Leicester: University Press.

Robbins, J. A. 1978 Geochemical and geophysical applications of radioactive lead. In *Biogeochemistry of lead in the environment* (ed. J. O. Nriagu), pp. 285–293. Amsterdam: Elsevier Scientific.