Comparaison of ^{210}Pb chronology with $^{238,239\text{-}240}Pu$, ^{241}Am and ^{137}Cs sedimentary record capacity in a lake system

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Introduction

Transuranic elements have been released into the environment on a global scale since the early 1950s. The main source was atmospheric testing of nuclear weapons though releases from nuclear installations have been significant on local and regional scales. Large quantities of radioactive debris from the explosion of high-yield thermo-nuclear weapons were injected into the stratosphere and widely dispersed around the world. Gradual re-entry of this debris to the troposphere was followed by fallout onto the earth's surface. Fallout on lakes was deposited on the bed of lake as part of the sediment record.

The purpose of this paper is to report the results of a study of the transuranic radionuclides $^{238,239\text{-}240}Pu$, ^{241}Am in the sediments of

Blelham Tarn in Cumbria (UK) and forms part of a project concerned with the fate of transuranic elements deposited on lakes and their catchments.

As sediments accumulate they capture a high quality record of the changing levels of contamination. In this study sediment cores taken from the two locations in the lake were sectioned at 1 cm intervals and analysed by alpha spectrometry for $^{238,239-240}Pu$, ^{241}Am to determine the historical fallout record at this site. The sub-samples were also analysed by gamma spectrometry for the fission product ^{137}Cs and the natural radionuclide ^{210}Pb.

^{137}Cs was also part of the debris from nuclear weapons tests though further amounts were also deposited in 1986 as a result of fallout from the Chernobyl reactor accident. Comparisons between ^{137}Cs and the transuranic elements provide information on their relative transport rates through the environment.

The ^{210}Pb flux is proportional to the annual precipitation on the lake and provides a chronology of the sub-samples of the cores.

Site Description

Blelham Tarn is a small lake situated off the North-West shore of the North basin of Windermere. The basic morphometric characteristics are given in Table 1. It has a fairly simple bathymetry, with two main basins (figure 1). The lake is mildly enriched, receiving inputs from some of the area's naturally richer soils on the western side of Windermere. The streams are nutrient enhanced due to local agriculture and a small sewage works for Outgate (a small village 2 miles away). Blelham Tarn has been a site of scientific interest since the 1930's and has been the subject of many palaeolimnological studies including some of the earliest studies of fallout radionuclides in recent sediments (Pennington *et al.*, 1976).

	Blelham Tarn
Location	54° 24' N, 2° 59' W
Altitude	42
Max. Relief	284 m
Rainfall	1814
Catchment Area	4.27 km^2
Lake Area	0.102 km^2
Length	0.65 km
Width	0.17 km
Max. Depth	15 m
Mean Depth	6.8 m
Volume	0.693 x 10^6 m^3
Inflows	5 small streams

Table 1
Morphometry of Blelham Tarn.

Experimental

Sample collection

Two sediment cores were collected on 24 March 1997 from the deepest parts of the two sub-basins of the lake (sites A and B, Figure 1) using a 10.3 cm diameter Mackereth corer. The cores were extruded vertically at the Institute of Freshwater Ecology, Windermere, core A at 1 cm intervals down to the base of the core and core B at 1 cm down to 30 cm and thereafter at 2 cm. The wet sediment samples (excluding the sub-samples taken for algal analysis) were stored in sealed plastic bags and returned to the ERRC Liverpool where they were dried at 50°C to determine the water content and dry bulk density. Each sample was divided into two parts, one being retained at Liverpool for dating by ^{210}Pb and ^{137}Cs and the other sent to the Laboratoire de Radiochimie et Radioécologie, Université de Nice-Sophia Antipolis where they were analysed for Pu and Am analysis by alpha spectrometry and ^{137}Cs by gamma spectrometry.

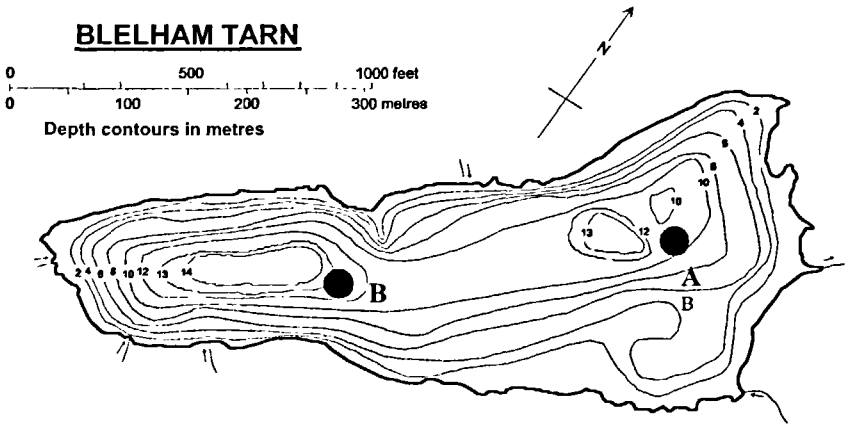

Figure 1a / b
The English Lake District showing (a) location and (b) the bathymetry of the lake. The locations of the sediment cores used in this study are marked.

Alpha spectrometry measurements

The radiochemical separation of Pu and Am in sediments is a complex procedure (Anonymous, 1989; Michel *et al.*, 1999a; 1999b). The first step is to pre-concentrate the radionuclides. The organic matter is removed by ashing and known quantities of the yield tracers ^{242}Pu and ^{243}Am are added to the residue. The transuranic elements are then removed by leaching with 8M HNO_3 and co-precipitation with ferric hydroxide in ammonia medium. This is followed by two extraction procedures: the first to separate the plutonium isotopes, the second to separate the fraction containing americium, before electrodeposition and counting by a-spectrometry. The separation of plutonium is achieved by elution with different acid solutions (8M HNO_3, 10M HCl and 10M HCl + 0.1M NH_4I) on an anionic exchange column mainly to discard thorium. After evaporation and addition of H_2O_2 in order to eliminate iodide, Pu isotopes are electrodeposited in sulfuric acid-ammonia media at pH = 2.4, on a stainless-steel disk, with a current of 1 A applied for 1 hour. The americium fraction is purified by co-precipitation with calcium oxalate. After extraction into HDEHP (diethylhexyl phosphoric acid) and elution through a two-layer exchange column (cationic + anionic), in order to separate remaining traces of Th and Pu, another anion-exchange process is used to eliminate rare earths. The eluate is evaporated and few drops of concentrated HNO_3 are added in order to eliminate thiocyanates. Americium is then electrodeposited two hours by the same procedure as for plutonium.

All reagents used in the separations were of analytical grade and solutions were prepared in unionized water. Anionic columns were: Bio Rad AGMP1 and Dowex 1 x 4 100-200 mesh in chloride form and cationic columns: Aldrich Dowex 50W x 8 100-200 mesh in chloride form.

The α-spectra were determined using EG & G Ortec 576A Dual Alpha Spectrometers equipped with boron implanted silicon detectors equipped with a multichannel buffer analyser (spectrum master ORTEC 919).

Gamma-spectrometry measurements

^{137}Cs measurements at Nice were carried out by standard g-spectrometry using methods described in (Barci-Funel *et al.*, 1988; 1992; Holm *et al.*, 1994). The sediment samples were oven-dried at 100°C, homogenized and packed into plastic containers and counted on a coaxial HPGe detector (EG&G ORTEC) of 17% relative efficiency with an energy resolution FWHM (Full Width at Half Maximum) of 1.9 keV at 1.33 MeV. The efficiency of detector was determined using standard sources with the same geometrical configuration as the samples being measured. Background radiation was reduced by placing the detector inside a 5 cm thick lead castle with a 2 mm thick copper lining. The spectra were collected using a multi-channel buffer analyzer (Spectrum Master ORTEC 919).

Results and discussion

Figure 2 plots concentrations of the transuranic radionuclides versus depth in core A. The corresponding results for core B are given in Figure 3. Figure 4 shows the ^{137}Cs results for both cores. The transuranic radionuclides all have a single well-resolved peak in their activity. In core A these occur at a depth of 14-16 cm. In core B they occur at 15-17 cm. The peaks in transuranic activity coincide with similar well-resolved peaks in the ^{137}Cs activity and almost certainly record the fallout maximum in 1963 from the atmospheric testing of nuclear weapons. Concern about fallout levels following the early tests resulted in a moratorium in atmospheric testing in 1958. Its breakdown in 1961 was followed by a period of intensive testing resulting in very high fallout during the years 1962-63. Implementation of the 1963 test ban treaty led to a rapid decline in fallout during the next few years and values in 1966 were just 10% of those in 1963. Nearly 50% of all fallout occurred during 1962-64, a space of just 3 years, giving rise to the very high concentrations recorded in the sediments of Blelham Tarn from this period.

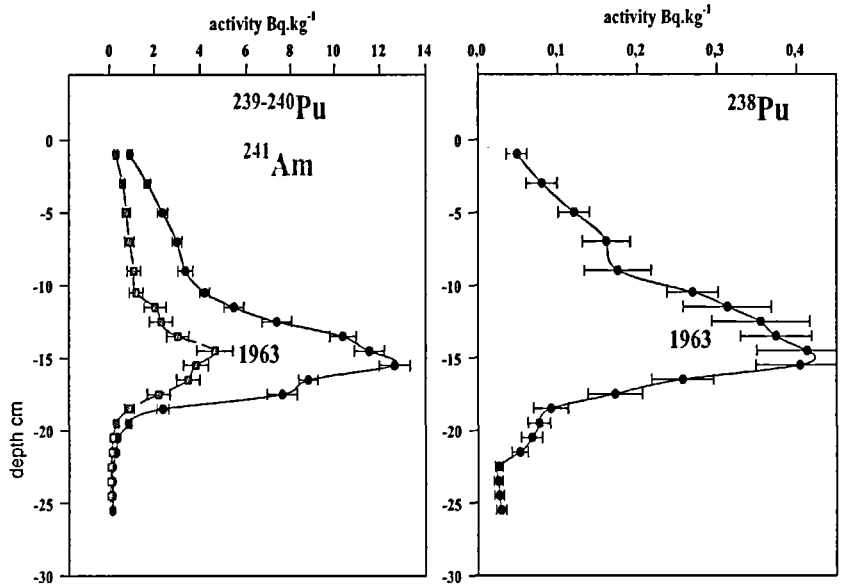

Figure 2
$^{238,239-240}$Pu and ^{241}Am profile in the dating core A.

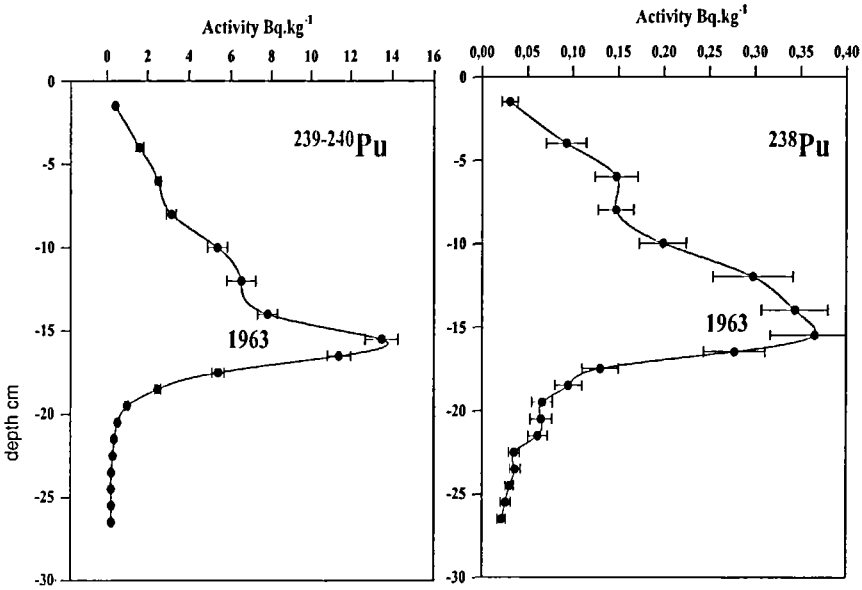

Figure 3
$^{238,239-240}$Pu profile in the dating core B.

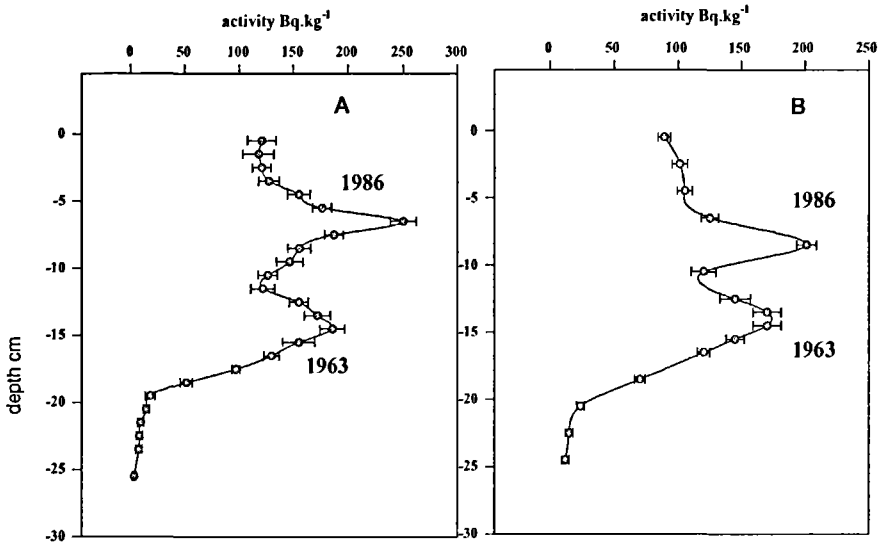

Figure 4
^{137}Cs profile in the dating core A and B.

In contrast to the transuranic elements, the ^{137}Cs profiles have a second more recent peak, at depths 6-7 cm in core A and 8-9 cm in core B. These features are almost certainly a record of fallout from the Chernobyl accident in 1986. Although the short-lived radionuclide ^{134}Cs associated with Chernobyl fallout was below limits of detection in the present cores, it was observed clearly in an earlier (1990) core from Blelham Tarn (van der Post, 1997). The ^{134}Cs peak in the 1990 core occurred at the same depth as the ^{137}Cs peak associated with Chernobyl fallout, and in the correct activity ratio. The absence of transuranic radionuclides in Chernobyl fallout is due to the fact that these radionuclides were mainly on heavier refractory particles and were deposited relatively near to the accident site.

Taken together the above results illustrate the extent to which sediments accurately recorded contamination of surface waters, in this case by radioactive fallout. They also show how dated events can be used to help provide accurate chronologies of recent sediments on timescales of just a few years. Sediments at 6-7 cm depth in core A

and 8-9 cm in core B can confidently be dated 1986. Sediments at 14-16 cm in core A and 14-17 cm in core B can be dated 1963. From these results the mean sedimentation rates at each core site during 1963-86 and 1986-97 are calculated to be:

Core A	Core B	
1963-86	0.079 ± 0.006 $g.cm^{-2}.y^{-1}$	0.100 ± 0.006 $g.cm^{-2}.y^{-1}$
1986-97	0.061 ± 0.008 $g.cm^{-2}.y^{-1}$	0.053 ± 0.002 $g.cm^{-2}.y^{-1}$

The date are compared to the ^{210}Pb chronology in the Figure 5. Using the ^{210}Pb chronology, the depths 15 cm and 7 cm respectively correspond to 1958±4 and 1983±4. The chronology is in good agreement with the transuranics and cesium dating profiles.

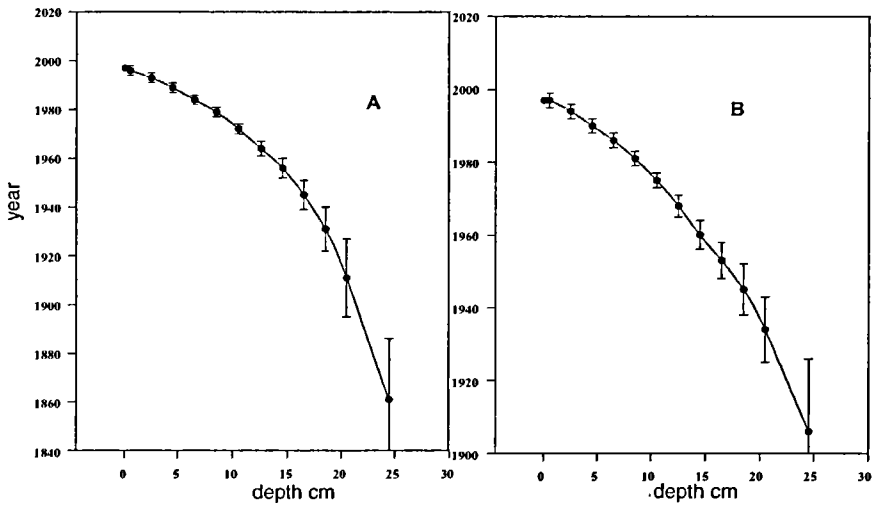

Figure 5
^{210}Pb chronology in the dating core A and B.

In conjunction with ^{210}Pb and other techniques such as diatom stratigraphy (Haworth, 1980) they can further be used to construct detailed sediment chronologies spanning the past 150 years.

Aknowledgements
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