

2 **^{210}Pb dating: thirty-five years on**

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6 It is now 35 years since I first became involved in
8 ^{210}Pb dating. Until then I had been quietly working
9 away on the mathematics of classical space–time.
10 Little did I anticipate that I was about to continue with
11 this general theme, though in a rather more down to
12 earth (or should I say limnological) setting. Following
13 a chance conversation with Frank Oldfield I soon
14 found myself on quite a different path, investigating
15 the mathematics of paleolimnological space–time
16 where the time dimension was now measured as depth
17 in a column of lake sediment!

18 I first met Frank in the mid-1970s as a member of that
19 pioneering Interdisciplinary Research Centre of the
20 University of Liverpool, the West Kirby Car Pool
21 (Oldfield 2010). Including at various times mathemati-
22 cians, geographers, engineers, veterinary scientists, the
23 University Archivist, and the Director of the Computer
24 Laboratory, it really did cover a wide range of academic
25 interests. Frank had recently arrived back in Liverpool
26 from New Guinea, and one day when we were travelling
27 to work he mentioned that he had been using the recently
28 developed ^{210}Pb method (Krishnaswamy et al. 1971) for

dating lake sediment cores, though there were problems. 29
Sediments from a lake in the New Guinea Highlands 30
known to date from the mid-eighteenth century or earlier 31
were yielding dates that were far too young. The rest, as 32
they say, is history. Using Frank’s diagrams and a little 33
bit of calculus we came up with the constant rate of 34
supply (CRS) model, which managed to produce much 35
more credible results (Oldfield et al. 1978). However, 36
when we tried to publish a paper describing the model, 37
we ran into difficulties. The referees, acknowledged 38
experts in the field at that time, described the model as 39
naïve and not particularly original. In light of what we 40
know now their criticisms were well justified. The only 41
thing wrong about their advice was that the approach we 42
developed, when tested empirically using data from a 43
number of different sites, in fact worked very success- 44
fully. The basic methodology, published originally in 45
Appleby and Oldfield (1978), has since been widely used 46
in numerous studies all round the world. We learnt two 47
lessons from this. The first was that there are times when 48
it pays to be a little naïve. The second was that one 49
shouldn’t always follow the advice of experts. 50

51 A highlight of those early years following my
52 introduction to Paleolimnology was a trip to Joensuu,
53 Finland at the beginning of September 1981 to attend
54 the 3rd International Symposium (Fig. 1). Getting to
55 Joensuu was itself quite an adventure involving a
56 journey by ferry and minibus across Sweden and
57 Finland, in the company of many of the other UK
58 participants. Following the opening session in Joen-
59 suu, the Symposium moved 45 miles north to the

A1 The author was the recipient of a “Lifetime Achievement
A2 Award” presented by the International Paleolimnology
A3 Association (IPA) in Glasgow, Scotland on 22 August 2012.

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60 beautiful hilltop hotel at Koli where the lecture
61 sessions took place. The meeting was memorable
62 both scientifically and socially, with many discussions
63 taking place around the sauna and pool. At the
64 conference dinner each national group was expected
65 to make a contribution to the evening's entertainment.
66 The UK's well-lubricated rendition of 'Strip the
67 Willow,' ably lead by Elizabeth Haworth, was an
68 unforgettable introduction to the paleolimnological
69 community. The journey home gave me the opportu-
70 nity to visit two sites, Laukunlampi and Pääjärvi, that
71 had figured in one of our early papers on ^{210}Pb records
72 in lakes with laminated sediments (Appleby et al.
73 1979), and see for the first time paleolimnologists in
74 action. Paddy O'Sullivan's love of Sibelius also
75 resulted in another highlight, a visit to Ainola.

76 After a couple of years of model testing Frank had a
77 second bright idea, why don't we develop a ^{210}Pb
78 radiometric laboratory at Liverpool? Now I knew the
79 meaning of the word radioactivity, but that was about as
80 far as it went—my first-year Physics was mostly long
81 forgotten. The idea seemed fairly adventurous to say the
82 least. Nonetheless, a visit from a supplier was arranged,
83 a demonstration organised, buttons pressed, and it all
84 looked fairly impressive, though the equipment did
85 seem very expensive. In the course of the demonstration,
86 the salesman said—"by the way, they have one of these
87 over in the Physics department, you might like to go and
88 have a look at it." So I went over to Physics, found Paul
89 Nolan, and a bright and shiny well detector sitting on a
90 bench in the old Van de Graaf Laboratory waiting for
91 someone to come along and use it. We quickly loaded it
92 with a lake sediment sample and came back next day to
93 find a beautiful 46.5 keV ^{210}Pb photo-peak in the

spectrum. We were in business. So our third lesson
was—perhaps fairy godmothers do exist.

The development of radiometric assay by gamma
spectrometry using hyper-pure germanium well-type
detectors proved to be a major step forward. Advantages
of this technique included non-destructive measure-
ments, minimal sample preparation, and significantly
higher detection efficiencies (particularly when using
well detectors) that allowed simultaneous determination
of a range of radionuclides, including ^{210}Pb , ^{226}Ra , ^{137}Cs ,
 ^{241}Am , etc. (Appleby et al. 1986) in relatively small
samples. Direct measurement of ^{226}Ra (supported ^{210}Pb)
concentrations in all samples analysed for ^{210}Pb removed
much of the uncertainty in determining the unsupported
component of the total ^{210}Pb activity needed for the
dating calculations. Further, the determination of ^{137}Cs
(and ^{241}Am) records alongside those of ^{210}Pb has proved
to be of crucial importance in the assessment of ^{210}Pb data
and the validation or correction of ^{210}Pb dates.

At Frank's instigation we formed the "Radiometric,
Mineral Magnetic, and Palaeoenvironmental Research
Centre," though after a few years we took pity on
people and shortened it to the "Environmental
Radioactivity Research Centre," ERRC. Any suc-
cesses that this Centre may have had were very much
due to its collaborative nature. Key people have of
course included Professor Frank Oldfield (Geography)
whose initiatives and leadership were central to this
whole development, and Professor Paul Nolan (Phys-
ics) who has very generously provided the technical
support so essential to the establishment and mainte-
nance of our Environmental Radiometric Laboratory
(Fig. 2).



Fig. 1 In conversation with Rick Battarbee at the 3rd interna-
tional symposium on Paleolimnology, Joensuu, 1981

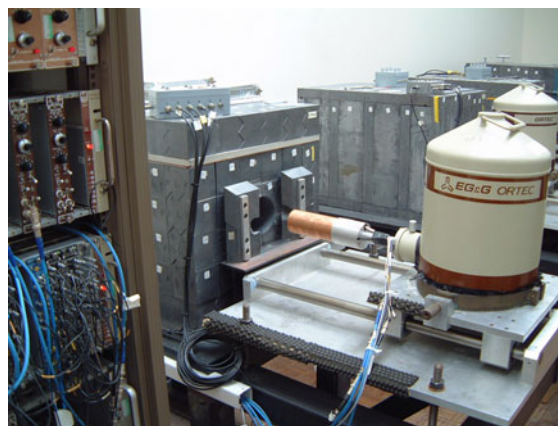


Fig. 2 Well-type hyper-pure germanium gamma detectors in
the ERRC Environmental Radiometric Laboratory

127 A further interesting challenge was the hosting of
 128 the 5th International Symposium on Paleolimnology
 129 held at Ambleside in the English Lake District in
 130 September 1989. This took place just about at the
 131 dawn of the internet age when communication systems
 132 and software packages were very rudimentary com-
 133 pared to those on display this year in Glasgow, at a
 134 time when FAX was dominant. Highlights (apart from
 135 the Scottish dancing) included the spectacle of a 6-m
 136 Mackereth corer in action, and a display of fieldwork
 137 by helicopter.

138 The main emphasis of my work during the next
 139 decade was the development of more precise models of
 140 the pathways by which ^{210}Pb and other fallout radio-
 141 nuclides, following deposition on the lake and in the
 142 catchment, are transported to the sediment record. A
 143 first step was the creation of mass balances for
 144 catchment/lake systems that involved determining
 145 inputs from the atmosphere, rates of transport from
 146 the catchment to the lake, and losses via the outflow.
 147 Since many of the key parameters were not well
 148 determined, this gave me a wonderful excuse to become
 149 involved in some fieldwork myself, a particular treat
 150 being trips to a number of high mountain lakes in
 151 spectacular settings in the Pyrenees, Austrian Alps, and
 152 Tatra mountains. Figure 3 shows colleagues from the
 153 University of Barcelona measuring soluble and partic-
 154 ulate concentrations of ^{210}Pb and ^{137}Cs in the water
 155 column of Redo Lake in the Spanish Pyrenees. Work at
 156 these sites, and also at Blelham Tarn in Cumbria
 157 (Appleby et al. 2003), revealed that although the bulk of
 158 fallout ^{210}Pb remained locked up in the catchment for
 159 very long periods of time, losses from the catchment to
 160 the lake could make a significant contribution to the
 161 amount reaching the sediment record. Although this
 162 might sound like bad news for the CRS model, in
 163 practice the effects were fairly marginal. It did however
 164 emphasise the need to constantly assess ^{210}Pb data and
 165 only accept dates as reliable if they have been
 166 independently validated. The most important means
 167 for achieving this was by comparing them with
 168 chronostratigraphic dates, most usually from sediment
 169 records of the 1963 ^{137}Cs (or ^{241}Am) fallout maximum
 170 from the atmospheric testing of thermonuclear weap-
 171 ons, or more recently, the 1986 Chernobyl reactor
 172 accident. In the early days of ^{210}Pb dating the 1963
 173 ^{137}Cs peak was too recent to be of any great value apart
 174 from indicating the quality of records in near-surface
 175 sediments. As time passes, it is now two ^{210}Pb half-lives

176 old, the 1963 ^{137}Cs peak is becoming of increasing
 177 importance in calibrating ^{210}Pb calculations.

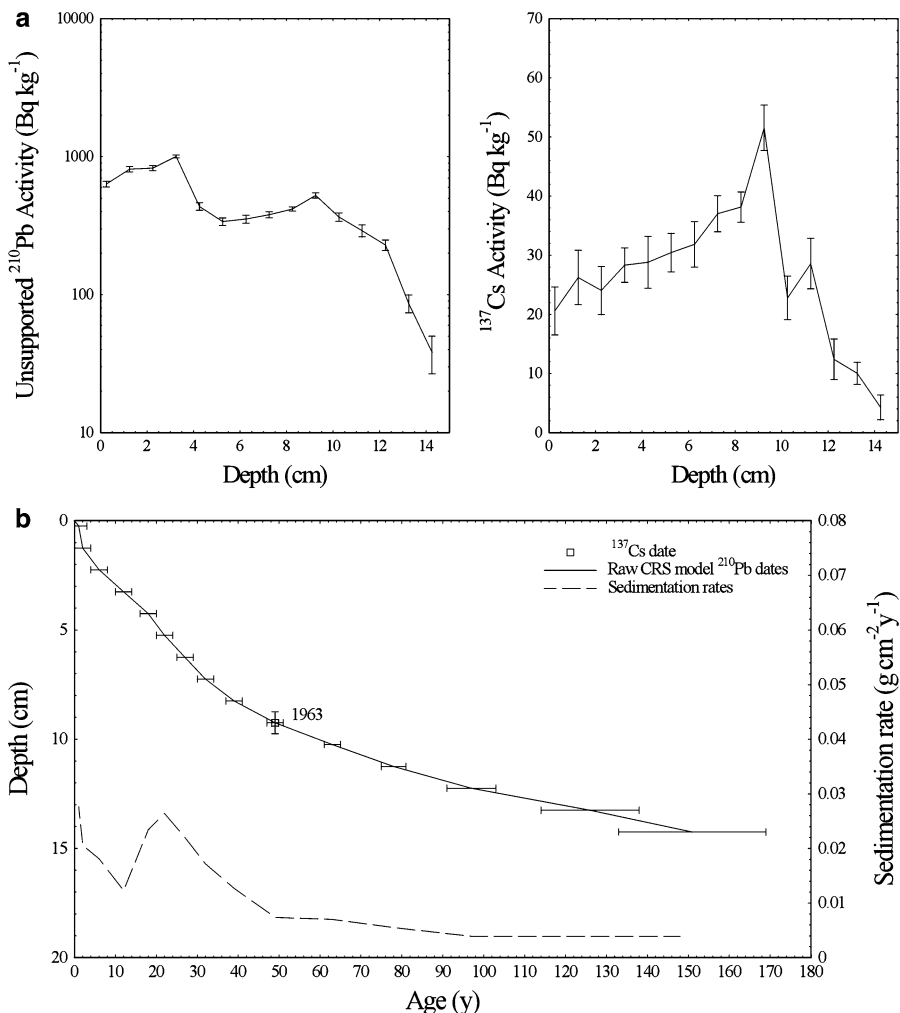
178 Calculating the ^{210}Pb date of a sample in essence
 179 requires an estimation to be made of the original ^{210}Pb
 180 concentration of the sample when laid down on the bed
 181 of the lake. The two simple models for making this
 182 estimation are the constant initial concentration (CIC)
 183 model, which assumes a steady state system in which
 184 sediments laid down at different times all had the same
 185 initial concentration, and the CRS model, which
 186 assumes that the initial concentrations were inversely
 187 proportional to the sedimentation rate, the coefficient
 188 of this proportionality being determined from the
 189 ^{210}Pb inventory of the core. Dates given by these two
 190 models are of course significantly different only at
 191 sites where there have been substantial changes in the
 192 sedimentation rate during the ^{210}Pb time-span
 193 (~ 130 years). In light of the complexities of the
 194 transport processes controlling the supply of fallout
 195 ^{210}Pb to the bottom sediments, it is surprising that the
 196 simple models work as well as they do. Figure 4 shows
 197 results from a site in Finland where in spite of the
 198 highly irregular nature of the ^{210}Pb record, the raw
 199 CRS model dates are in excellent agreement with the
 200 1963 ^{137}Cs date determined from the well-defined
 201 peak in the ^{137}Cs activity versus depth record.

202 Although the CRS model has proved to be generally
 203 the more reliable, there have nonetheless been many
 204 cases where there have been significant discrepancies
 205 between ^{210}Pb and ^{137}Cs dates. These would typically
 206 be due to systematic changes in the rate of supply of
 207 ^{210}Pb to the core site, or singular events such as



Fig. 3 Measuring ^{210}Pb concentrations in the water column of Redo Lake, Spanish Pyrenees, with colleagues from the University of Barcelona

Fig. 4 a Radionuclide records in a core from Finland. Although the ^{210}Pb record is highly irregular, the ^{137}Cs record does have a well-defined peak identifying the 1963 depth. **b** The raw CRS model ^{210}Pb dates are in excellent agreement with the 1963 ^{137}Cs date. The irregular ^{210}Pb record is attributed to a large increase in the sedimentation rate in recent decades



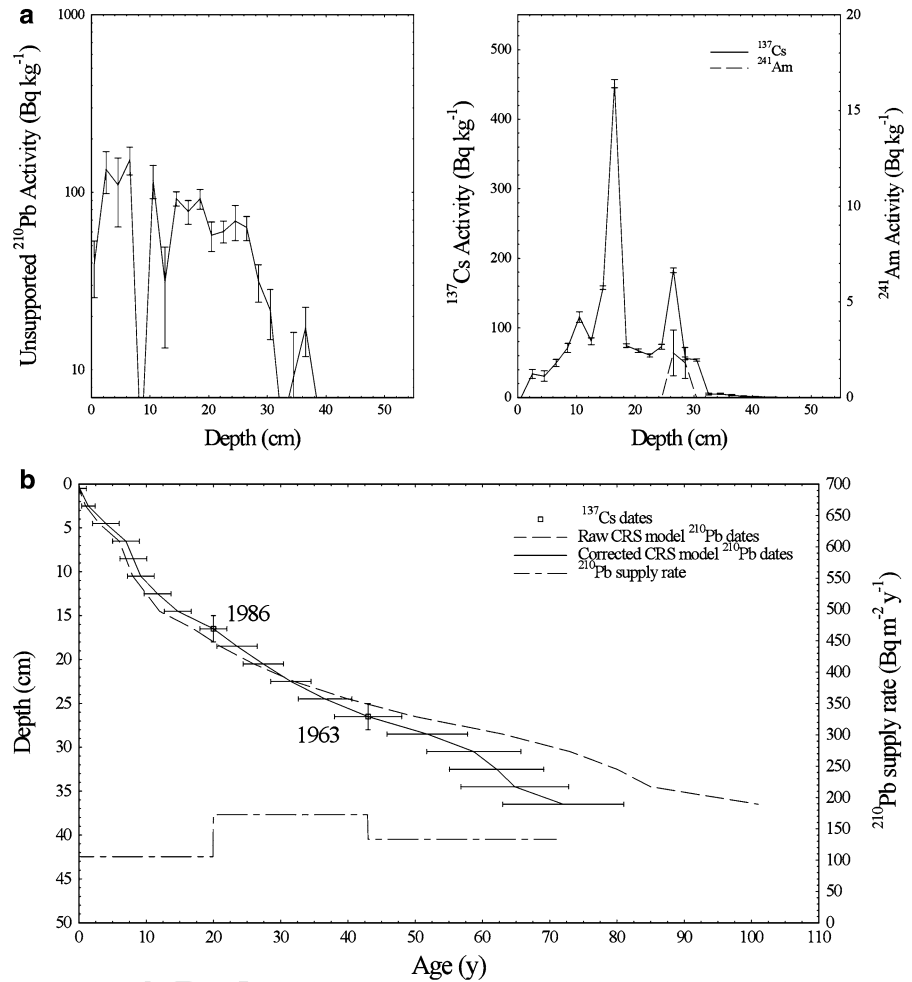
208 sediment slumps, or hiatuses in the sediment record
 209 that have affected the ^{210}Pb inventory. Where dating
 210 discrepancies are significant it may be necessary to
 211 apply corrections to the ^{210}Pb calculations. Any
 212 correction procedure must however be *simple, prac-*
 213 *ticable, and evidence-based.* Although such discrep-
 214 ancies imply a departure from the assumptions of one
 215 or the other of the simple models, the complicated
 216 nature of the transport processes governing the supply
 217 of ^{210}Pb makes it unlikely that any more general and
 218 widely applicable process-based model can be found.
 219 The approach we have taken is to apply the CRS model
 220 in a piecewise way to different sections of the core,
 221 using ^{137}Cs or other chronostratigraphic dates as
 222 reference points (Appleby 2001). Figure 5 shows
 223 results from a lake in northern Germany in which the
 224 raw CRS model dates, calculated by assuming a single

^{210}Pb supply rate, differed significantly from those
 determined from the ^{137}Cs record which had two
 distinct peaks identifying the 1986 and 1963 depths.
 Figure 5b shows mean ^{210}Pb supply rates for each of
 the three different time periods, post-1986, 1963–1986
 and pre-1963, calculated from the ^{210}Pb inventories
 contained within those sections of the core, and also
 the corrected ^{210}Pb dates calculated by applying the
 CRS model in a piecewise way using these values.

Quite apart from their role in dating lake sediments,
 in view of their well-defined origin, radionuclide
 records also play an important role as indicators of
 the quality of sediment records. If a core has good-
 quality ^{210}Pb and ^{137}Cs records it is reasonable to
 suppose that records of other environmental indicators
 should also be well preserved. Although judgement of
 quality and reliability from the record in a single core at

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Fig. 5 a Radionuclide records in a core from northern Germany. Well-defined peaks in the ^{137}Cs record identify the 1986 and 1963 depths, and also demonstrate that the irregular ^{210}Pb record is not caused by sediment mixing. **b** Errors in the raw CRS model ^{210}Pb dates are attributed to changes in the ^{210}Pb supply rate calculated using the 1986 and 1963 ^{137}Cs dates as reference points. The corrected ^{210}Pb dates were calculated by applying the CRS model in a piecewise way using different ^{210}Pb supply rates for each time period



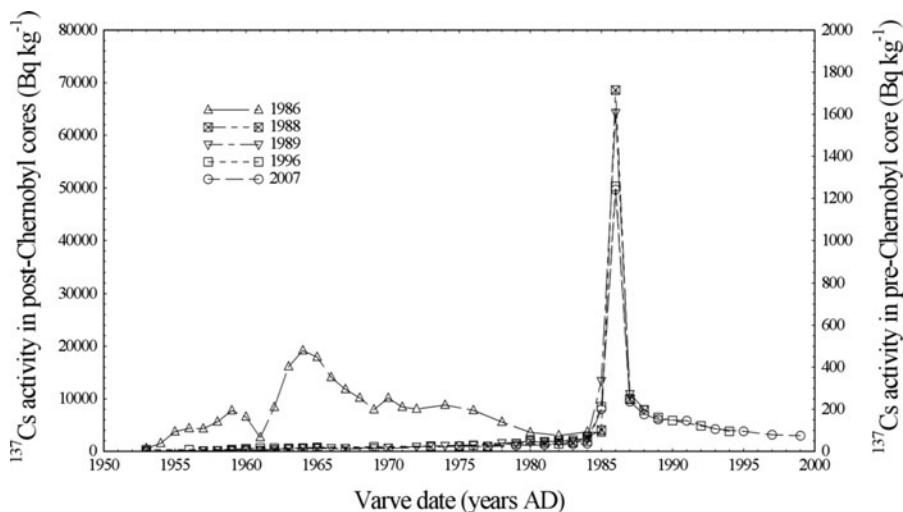
242 a particular point in time is somewhat circumstantial,
 243 the evident mobility of ^{137}Cs having led some authors to
 244 question the reliability of ^{137}Cs dates, we are now in the
 245 fortunate position of being able to investigate directly
 246 the stability of sediment records by revisiting sites that
 247 were cored decades earlier. This has been demonstrated
 248 in a recent study by Klaminder et al. (2012) using
 249 varved sediment cores from Nylandssjon (Sweden)
 250 collected over a 21-year period, 1986–2007. The ^{137}Cs
 251 records from these cores (Fig. 6) clearly demonstrate
 252 the persistence and accuracy of the key 1963 and 1986
 253 ^{137}Cs chronostratigraphic features over a period of
 254 several decades. Annual laminations allowed very
 255 precise dating of sediments from this lake. The pre-
 256 1986 core collected just before the Chernobyl accident
 257 preserved a well-defined peak in the 1964 layer
 258 recording the year of maximum fallout from the
 259 atmospheric testing of nuclear weapons. The post-

1986 cores, collected in 1988, 1989, 1996 and 2007, all
 had well-defined peaks in the 1986 layer recording
 fallout from the Chernobyl nuclear reactor fire. The
 evident mobility of a soluble fraction, presumably by
 pore water diffusion, in no way diminished the
 reliability of these ^{137}Cs dates. Because of the very
 high levels of Chernobyl fallout at this site, downward
 migration of ^{137}Cs from this source did however mask
 the weapons fallout peaks in the post-1986 cores.

Working with the many friends and colleagues I
 have made and met in the paleolimnology community
 has in itself been an immense source of satisfaction
 quite apart from that gained from working in a field
 with direct relevance to many contemporary issues. In
 particular I owe a great debt of gratitude to Frank
 Oldfield whose guidance, advice and encouragement
 were instrumental in setting me off on this particular
 journey, and also to Rick Battarbee and his colleagues

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Fig. 6 Records of fallout ^{137}Cs in annually laminated sediment cores collected during the period 1986–2007 from Nylandssjön, Sweden (Klaminder et al. 2012). The 1986 core was collected just before the Chernobyl reactor fire



278 at the UCL Environmental Change Research Centre
 279 with whom we at Liverpool have enjoyed an
 280 extremely valuable and fruitful collaboration over
 281 many years. I am also greatly indebted to Elizabeth
 282 Haworth and her colleagues at the IFE/FBA Ferry
 283 House Laboratory on Windermere for their invaluable
 284 help and advice, and also for providing me with an
 285 excellent excuse for visiting the many lakes and tarns
 286 of the English Lake District.
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