DATING RECENT SEDIMENTS BY ^{210}Pb: PROBLEMS AND SOLUTIONS

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1 Introduction

Accurate dating by ^{210}Pb is of crucial importance to a wide range of programs studying environmental records stored in natural archives such as lake sediments or peat bog accumulations. In a large number of cases the method has proved to be very reliable, particularly in stable environments with uniform sediment accumulation rates where the dating calculations are unambiguous.

The method has also been found to give good result at many sites with non-uniform accumulation, though here the problem is more difficult in view of the need to determine an appropriate dating model. There are two simple models, commonly referred to as the CRS and CIC models (Appleby & Oldfield 1978, Robbins 1978). Of these, the CRS (constant rate of ^{210}Pb supply) model is perhaps the most widely used. The main principles of this model are exemplified in Appleby *et al.* (1979) by cores from three Finnish lakes with annually laminated sediments, all of which contained layers recording dilution of the atmospheric ^{210}Pb flux by increased sedimentation. ^{210}Pb dates calculated using the CRS model were in good agreements with those determined by laminae counting. There are however circumstances where the CIC (constant initial concentration) model is appropriate, e.g. in a core from Devoke Water (Appleby & Oldfield 1992) where the CRS model was invalidated by an abrupt discontinuity in the sediment record.

In a very real sense these models should in the first instance be regarded as tools whose purpose is to determine, as far as practicable, the processes that have generated the data contained in the sediment record. In each case the ^{210}Pb data must be individually assessed in light of any independent chronostratigraphic evidence such as that provided by ^{137}Cs or ^{241}Am. The object of this paper is to highlight the conceptual framework that forms the basis of this

assessment and to show how it can be used to solve problems that have arisen in a number of practical cases. These include the use of hybrid models where there has been a variable ^{210}Pb supply, corrections for inaccuracies in the calculation of radiometric inventories, and the impact of large variations in ^{226}Ra activity.

2 Atmospheric Fluxes

Since dating by ^{210}Pb is based on sediment records of ^{210}Pb fallout, a proper assessment of the record requires a good knowledge of the atmospheric flux. Fallout ^{210}Pb derives from the decay of gaseous ^{222}Rn introduced into the atmosphere by diffusion from soils, and is normally assumed to have a constant flux at any given site (when averaged over a period of year or more). The amount of fallout may however vary from place to place by an order of magnitude, depending on factors such as rainfall and geographical location. This is illustrated by the measurements of ^{210}Pb fallout in Great Britain and Central Europe summarised in Fig. 1. These show that at a regional level there is a strong correlation with rainfall. Overall levels of fallout in Central Europe are however significantly higher than in Great Britain, presumably due to a build of ^{222}Rn concentrations in the atmosphere as the prevailing winds transport air masses over the intervening land surface. The global summary given in Fig. 2 suggests that there is a consistent west to east increase in ^{210}Pb fallout within the major continents, superimposed on a baseline ^{210}Pb flux of c. 30 - 40 Bq m^{-2} y^{-1} per metre of rain at sites remote from major land masses.

3 Long-term Sediment Records of ^{210}Pb fallout

Some of the earliest studies of fallout radionuclides in lake sediments were carried out by Pennington *et al.* (1973 & 1976) on cores from lakes in the Windermere catchment in Cumbria (UK), including Blelham Tarn, Esthwaite Water, Elterwater and Windermere itself. Further analyses carried out during the 1980s and 1990s at the Liverpool University Environmental Radioactivity Research Centre have resulted in a record of measurements from this region that now spans a period of 25 years. These lakes all have similar rainfall and so may be expected to have similar atmospheric fluxes of ^{210}Pb.

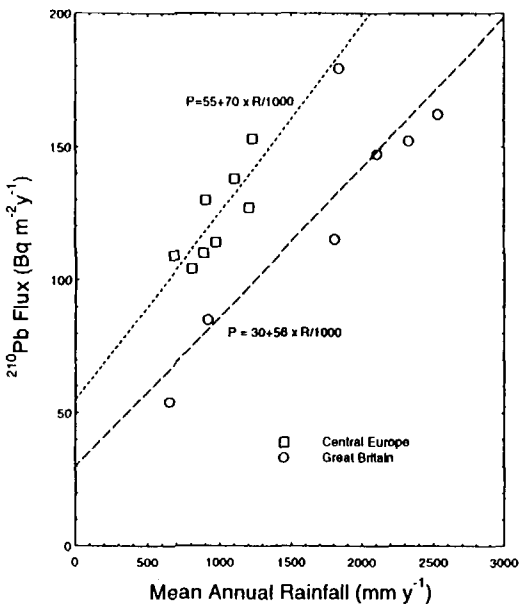

Figure 1. *Values of the atmospheric ^{210}Pb flux versus rainfall for Great Britain (o) and Central Europe (□), summarised from a variety of sources. The data are based on measurement from both soil cores and direct precipitation.*

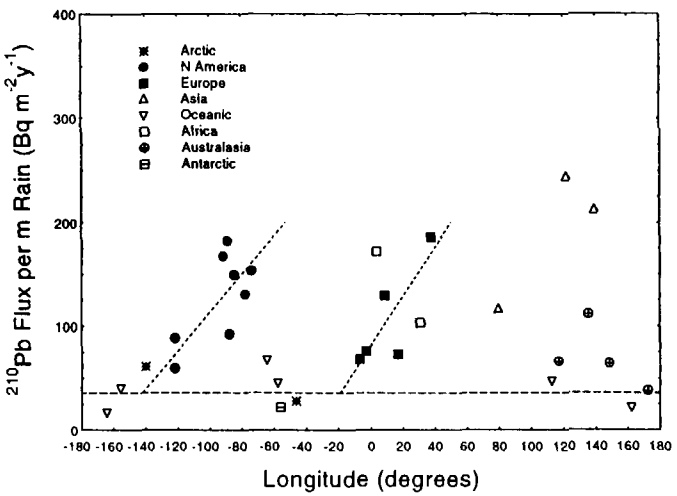

Figure 2. *Values of atmospheric ^{210}Pb flux per m of annual rainfall, plotted against longitude. The plot distinguishes values for each of the major land masses, the poles, and oceanic sites.*

At a given core site, mean ^{210}Pb fluxes to the sediment record can be estimated from the relation

$$P = \lambda A(0),$$

where λ is the ^{210}Pb radioactive decay constant and $A(0)$ is the unsupported ^{210}Pb inventory of the core. Fig. 3a shows the result of these calculations for the Windermere lakes, plotted against the date of coring. This shows that although the ^{210}Pb flux appears to be relatively uniform through time, as assumed by the CRS model, values are about twice as high as the direct atmospheric flux (indicated by the dashed line).

Factors influencing differences between the atmospheric flux and sediment record may include inputs from the catchment, losses from the water column via the outflow, and sediment focussing. The influence of water column processes is shown by comparing ^{210}Pb inventories with those of fallout ^{137}Cs from the atmospheric testing of nuclear weapons. Values of the ^{137}Cs/^{210}Pb inventory ratio (corrected for ^{137}Cs decay) in sediment cores from the Windermere lakes (Fig. 3b) are about half those expected from direct fallout, presumably due to relatively greater losses of ^{137}Cs from the system (via the outflow) arising from its higher solubility (Appleby 1991).

4 Transport Models

These results highlight the fact that transport processes through catchment/lake systems have a strong influence on sediment records of the atmospheric ^{210}Pb fluxes, and thus on ^{210}Pb dating methodologies. Fig. 4 gives a simplistic framework for modelling these processes. Using simple box models, and assuming a constant ^{210}Pb atmospheric flux P, the rate of supply of ^{210}Pb to the bottom sediments is given by the expression

$$P_S = F_{Pb}(1 + \alpha\eta_{Pb})P,$$

(c.f. Appleby & Oldfield 1992, Appleby & Smith 1993) where α is the catchment/lake area ratio, η_{Pb} is a ^{210}Pb catchment/lake transport parameter and F_{Pb} is a water column to sediment record transfer fraction, determined by the water residence time, water depth, particle settling velocity and solubility. Although this equation suggests that essential conditions for validity of the

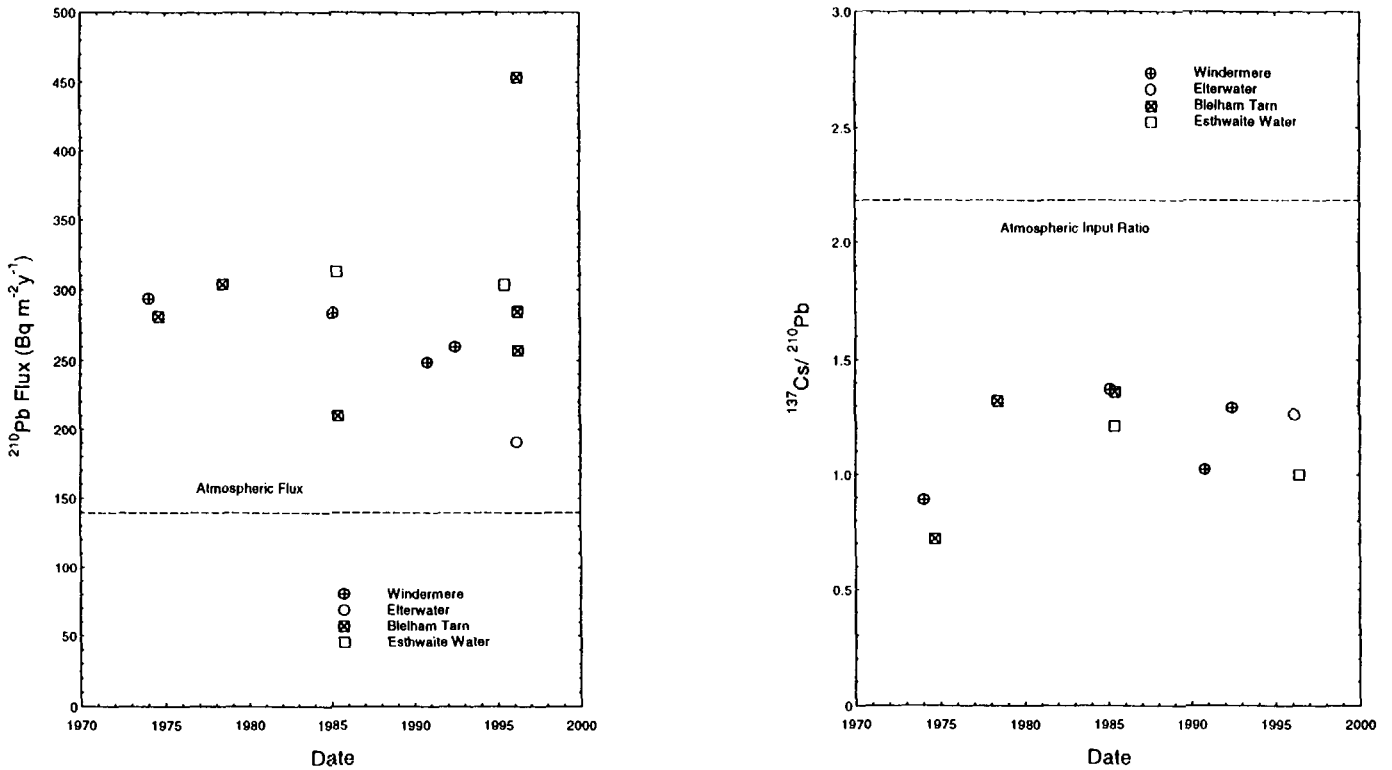

Figure 3. *Values of the mean ^{210}Pb flux (a) and the weapons ^{137}Cs/ ^{210}Pb inventory ratio (b) for sediment cores from lakes in the Windermere catchment, plotted against the date of coring. In both cases the dashed line shows direct atmospheric fallout values.*

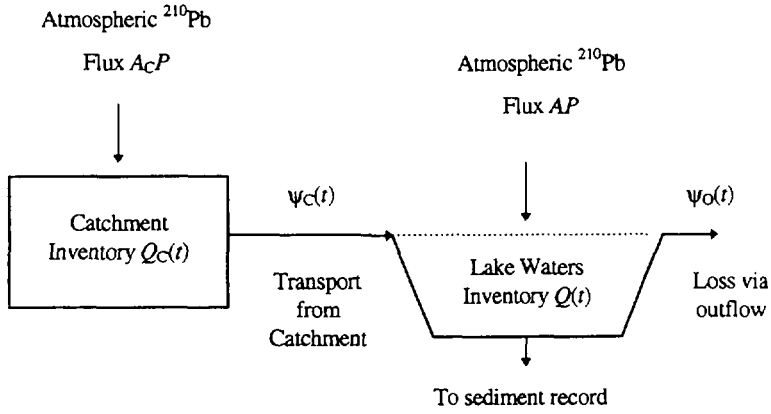

Figure 4. *A simple schematic model of transport processes controlling the supply of fallout ^{210}Pb to lake sediments.*

CRS model include steady inputs from the catchment and constant residence time in the water column, these may not be of major significance in practice. Studies of catchment/lake transport (Lewis 1977; Scott *et al.* 1985; Dominik *et al.* 1987) suggest that just 1 - 2% of the annual ^{210}Pb fallout on the catchment is removed to the lake. Further, since ^{210}Pb is largely associated with particulates only a small fraction of the total amount entering the lake is lost via the outflow (Appleby & Smith 1993). In most cases the validity of either of the simple models will depend on the extent and nature of the processes controlling sediment focussing.

An important exception to these conditions has been observed by Appleby *et al.* (1995) in a number of lakes from Signy Island (maritime Antarctic) where greatly elevated inventories of unsupported ^{210}Pb in the sediments (relative to the atmospheric flux) were explained by the bulk of fallout onto the catchment being delivered to the lakes during the annual thaw. In terms of the above equation this corresponds to the case were $\eta_{Pb} \approx 1$.

5 Validation of ^{210}Pb dates

Although parameters such as the ^{210}Pb inventory of core and the surficial concentration, together with features in the unsupported ^{210}Pb activity versus depth profile (changes in slope, non-monotonic "kinks") all play a role in the

assessment of ^{210}Pb data from sites with varying sediment accumulation (Appleby & Oldfield 1983), independent validation of the chronology is essential to a high level of confidence in the results. The most important means for validating dates for the last 30 - 40 years is via fallout records of artificial radionuclides. This is well illustrated by results from two Windermere cores (Fig. 5), collected in 1985 and 1992. The ^{137}Cs activity versus depth profile in the 1985 core has a single well resolved peak recording the maximum fallout of this radionuclide in 1963 from the atmospheric testing of nuclear weapons. The origin of this feature is corroborated by the presence of a similar (but much smaller) ^{241}Am peak (Appleby *et al.* 1991). The weapons fallout record (^{137}Cs and ^{241}Am) is again clearly present in the 1992 core. Now however there is a second more recent ^{137}Cs peak recording fallout from the 1986 Chernobyl accident. The identification in this case is corroborated by ^{134}Cs. Discharges from the Chernobyl reactor fire included both caesium radionuclides in a characteristic ratio Cambray *et al.* (1987). In many cases the presence of these peaks gives a very good control on the recent chronology. Fig. 6 shows that the dated levels determined from the ^{137}Cs record in the 1992 Windermere core are in excellent agreement with the CRS model ^{210}Pb dates.

Validation of older ^{210}Pb dates near the base of a core remains a major problem, particularly where there have been significant late 19th or early 20th century changes in accumulation rates. The most usual method is by independent chronostratigraphic features in the pollen, diatom or trace metal records.

6 Mixing or Acceleration?

Radionuclides delivered to the bed of the lake on settling particles may be redistributed within the sediment column by two main processes,

- physical or biological mixing at or near the sediment-water interface,
- chemical diffusion or advection within the porewaters.

Sediment mixing typically results in a flattening of the ^{210}Pb activity versus depth profile in the surficial sediment layers, and degradation of features such as ^{137}Cs or ^{241}Am peaks. The effects of these processes have been documented in many studies and a number of models have been developed to take account of them (Robbins *et al.* 1977; Oldfield & Appleby 1984). Since other processes can have similar effects on sediment records, an essential problem in using

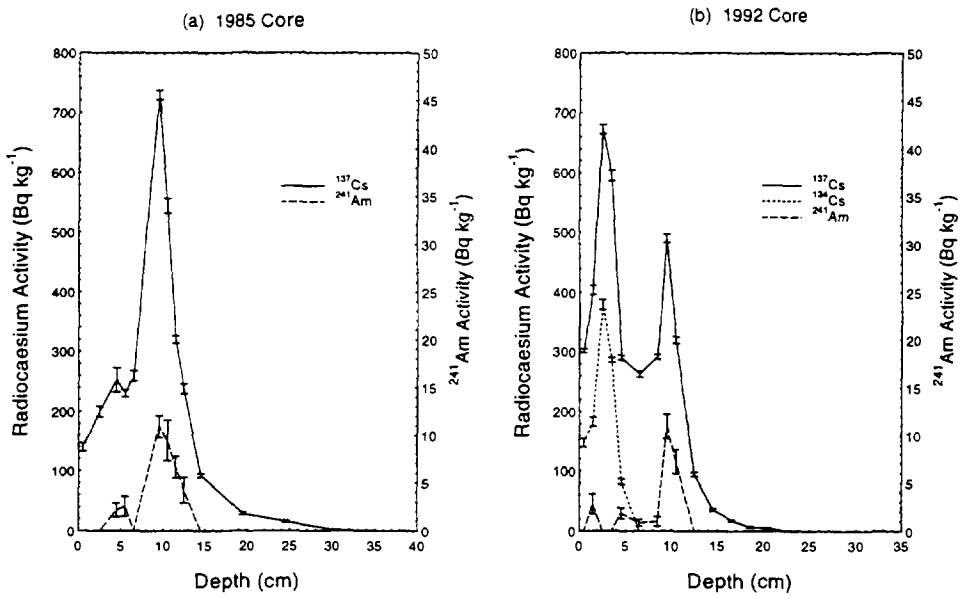

Figure 5. *^{137}Cs (——), ^{134}Cs (.....) and ^{241}Am (-----) activity versus depth profiles in a 1985 (a) and 1992 (b) sediment core from Lake Windermere, Cumbria, UK.*

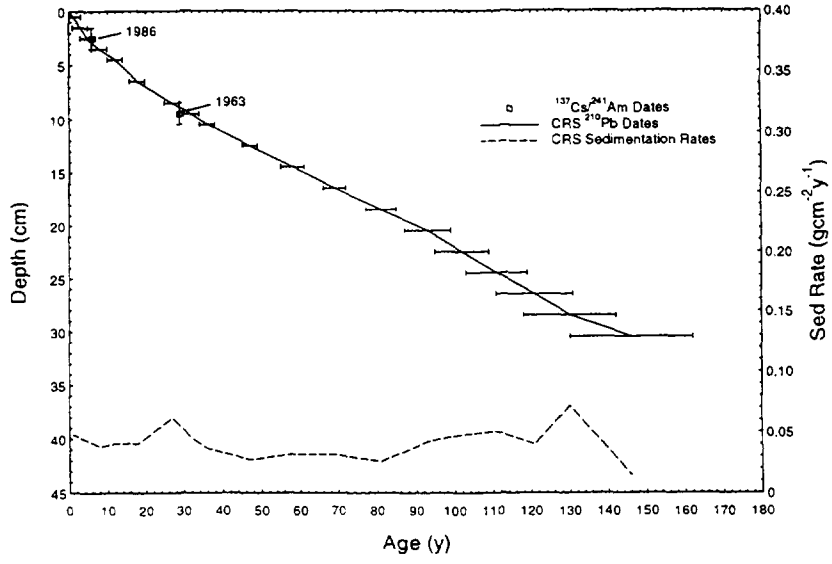

Figure 6. *CRS model ^{210}Pb depth (——) and sedimentation rate(-----) versus age relations for the 1992 Windermere core. Also shown are the 1963 and 1986 depths determined from ^{137}Cs, ^{134}Cs and ^{241}Am records.*

mixing models is the difficulty in making objective determination of the mixing parameters. Estimates of such parameters should be based on at least two independent records. The importance of this is illustrated by the results from Zgornje Krisko Jezero (Slovenia) shown in Fig. 7 in which ^{210}Pb activity in a sediment core was almost uniform throughout the top 20 cm. The initial presumption that this might be due to rapid intensive mixing was shown to be incorrect by the presence of a well resolved $^{137}Cs/^{134}Cs$ peak at a depth of just 4 cm. This feature, recording fallout from the 1986 Chernobyl accident, together with elevated ^{241}Am concentrations below 16 cm, demonstrated that mixing in these sediments was negligible and that the ^{210}Pb profile was almost certainly due to very rapid sedimentation.

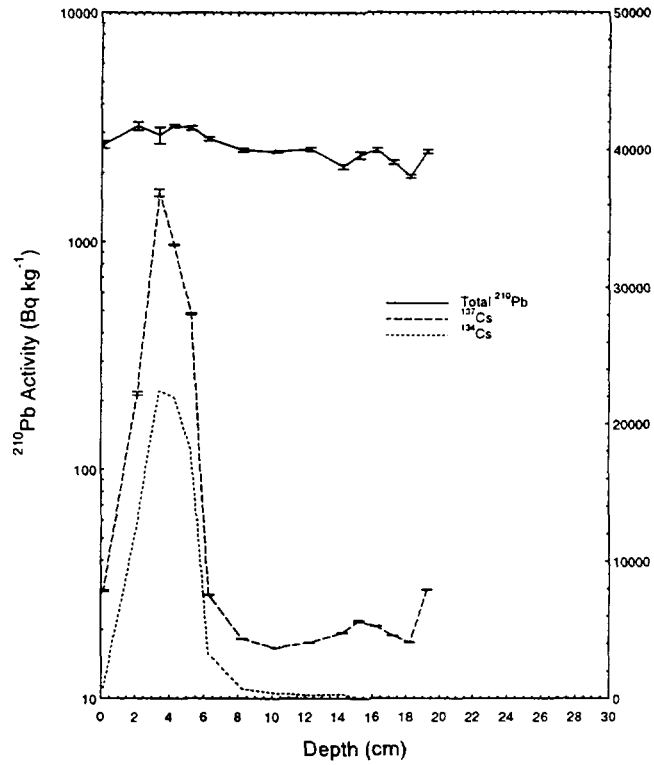

Figure 7. *^{210}Pb (——), ^{137}Cs (-----) and ^{134}Cs (.....) activity versus depth profiles in a sediment core from Zgornje Krisko Jezero, Slovenia.*

In those cases where mixing has occurred, Appleby & Oldfield (1992) have pointed out that corrections will often only be important in those cases where the CIC model is being used. Where the CRS model is applicable, calculations show that the use of uncorrected ^{210}Pb dates in a sediment core with a mixing zone spanning 10 years accumulation gives a maximum error of less than 2 years.

Where physical mixing is demonstrably negligible, the possibility that flattened ^{210}Pb profiles might be due to chemical remobilisation can be tested by comparing ^{210}Pb dates with those determined from the ^{137}Cs stratigraphy. The excellent agreement between ^{210}Pb and ^{137}Cs (or ^{241}Am) in a large number of cases suggests that in most circumstances this is not a significant problem.

7 Corrections to the CRS Model for Incomplete Inventories

CRS model ^{210}Pb dates are calculated using the formula

$$t = \frac{1}{\lambda} \ln \frac{A(0)}{A},$$

(Appleby & Oldfield 1978) where

- λ is the ^{210}Pb decay constant,
- $A(0)$ is the unsupported ^{210}Pb inventory of the entire core,
- A is the unsupported ^{210}Pb inventory below the sample being dated.

A and $A(0)$ are both determined by numerical integration of the ^{210}Pb profile. Accuracy in the dating, particularly in the first few samples above the equilibrium depth, is crucially dependent on reliable estimates of the ^{210}Pb inventories. Under-estimation of A in these samples will cause the ^{210}Pb dates of these levels to be too old.

There are two common sources of error, uncertainties in the numerical procedures for estimating A, and gaps in the sediment record. In both cases the error can be reduced if there is a means for correcting the values given by simple numerical integration. This can be achieved in two ways: by estimating the accumulation rate at a reference point near the base of the core, or the calendar date of that point.

Reference accumulation rates

At sites where ^{210}Pb activity in the older sections is evidently declining more or less exponentially with depth, indicating uniform accumulation, the mean sedimentation rate for that period, r_i, can be estimated from the gradient of the relevant section of the ^{210}Pb profile (plotted logarithmically), using the formula

$$\frac{d}{dm}\ln C = -\frac{\lambda}{r_i},$$

where m is the depth measured as cumulative dry mass. The ^{210}Pb inventory below the first data point above the ^{210}Pb dating horizon (c.100 - 150 years), A_i, can be estimated using the formula

$$A_i = \frac{1}{\lambda} r_i C_i,$$

(c.f. Appleby & Oldfield 1978) where C_i is the unsupported activity at that level. Dates for subsequent levels can then be calculated using the formula

$$t = \frac{1}{\lambda} \ln \frac{A(0)}{\delta A + A_i},$$

where δA is the unsupported ^{210}Pb inventory between the basal sample and the sample being dated.

Reference dates

The above method cannot be applied at sites where there have been non-uniform sediment accumulation rates in the 19th or early 20th century, or a hiatus in the sediment record. Accuracy in these cases can be improved by using an independently dated reference level. If ΔA denotes the entire unsupported ^{210}Pb inventory *above* the reference level, determined by numerical integration of the activity versus depth profile, the inventory below the reference level be estimated using the formula

$$A_i = \frac{\Delta A}{e^{\lambda t_i} - 1},$$

(Oldfield & Appleby 1984), where t_i is the age of the reference level. Revised ^{210}Pb dates are then calculated as above.

The dated reference level must of course be at or above the ^{210}Pb dating horizon. When it is below the ^{210}Pb dating horizon (and there appear to have been significant increases in sedimentation rates in the late 19th century), it may instead be used with the older ^{210}Pb to make estimates of the basal sedimentation rate. The value of A_i can then be estimated by the first method. The process can be iterated to give improved accuracy.

8 Varying ^{210}Pb Supply Rates

Sediment cores frequently record episodes of rapid sediment accumulation, due for example to flood events, turbidity currents of major land-use changes. Even where the profile contains non-monotonic variations indicating significant dilution of the atmospheric ^{210}Pb flux, the event recorded by these features may import significant quantities of additional fallout ^{210}Pb. For a brief period the ^{210}Pb supply rates may be significantly higher than at times of normal accumulation, and where this occurs the simple CRS and CIC models will both give erroneous dates.

Where there are suitably placed independently dated levels, determined by chronostratigraphic markers such as ^{137}Cs, ^{241}Am or pollen, these can be used to calculate the mean ^{210}Pb flux for each zone. Assuming the flux to be uniform within the zone, corrected ^{210}Pb dates and sedimentation rates for intermediate depths can calculated by applying the principles of the CRS model with the relevant ^{210}Pb flux.

The method governing the application of this piecewise CRS model can be summarised as follows:

I Given two core depths x_1 and x_2 with known dates t_1 and t_2, the mean ^{210}Pb flux during the period spanned by this section is

$$P = \frac{\lambda \Delta A}{e^{-\lambda t_1} - e^{-\lambda t_2}},$$

where ΔA is the ^{210}Pb inventory between x_1 and x_2.

II Given the ^{210}Pb flux P for a core section containing a dated level x_1, the date of sediments at depth x in this section is determined by solving the equation

$$\frac{P}{\lambda} e^{-\lambda t} = \frac{P}{\lambda} e^{-\lambda t_1} + \Delta A(x_1, x),$$

where $\Delta A(x_1, x)$ is the ^{210}Pb inventory between x_1 and x.

Fig. 8 shows results obtained by applying this method to a sediment core from Norrviken (Sweden). A highly irregular ^{210}Pb activity versus depth profile with very low unsupported activities suggested rapid sediment accumulation, particularly during the past 40 - 50 years. Well defined ^{137}Cs and ^{241}Am peaks from weapons test and Chernobyl fallout identified the 1986 and 1963 depths and showed that the core did in fact contain a good record of inputs to the lake. Discrepancies between the ^{137}Cs dates and simple CRS model ^{210}Pb dates (dashed line) show that the disturbances must have been associated with major variations in the ^{210}Pb supply. Calculations based on the ^{137}Cs dates indicated an 8-fold increase in sedimentation rates during 1944 - 62, with a 5-fold increase in the ^{210}Pb flux. The solid line shows ^{210}Pb dates calculated using the above equations. The dotted line shows the calculated sedimentation rates.

9 Variations in ^{226}Ra Activity

In many of the early studies, in which ^{210}Pb was determined by alpha spectrometry, it was often assumed that ^{226}Ra activity was uniform throughout the core and that supported ^{210}Pb could be estimated from total ^{210}Pb activity at depths where unsupported activity could safely be assumed negligible. Direct ^{226}Ra measurements, where carried out, were usually limited in number. One of the major advantages of ^{210}Pb assay by gamma spectrometry has been the determination of ^{226}Ra in each sample, usually via the 295 keV and 352 keV emissions from the daughter radionuclide ^{214}Pb following 3 weeks equilibration in a container sealed against ^{222}Rn escape.

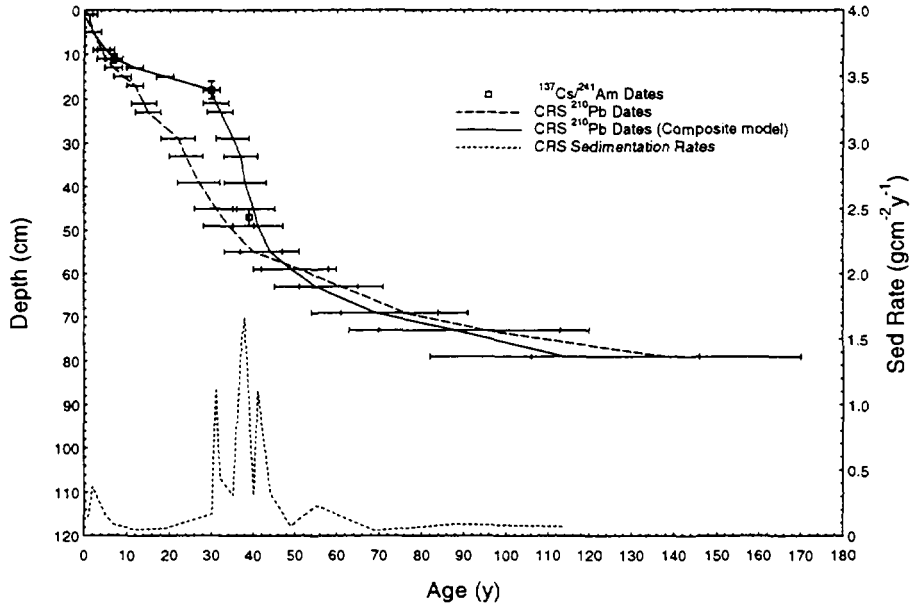

Figure 8. *Depth and sedimentation rate versus age relations for a sediment core from Norrviken, Sweden, showing both the raw CRS model dates (-----) and the corrected dates calculated using the piecewise CRS model. The elevated ^{210}Pb flux during the period of rapid sedimentation was calculated using the 1954, 1963 and 1986 ^{137}Cs dates.*

Although variations in ^{226}Ra activity with depth are in most cases quite small, this is not always the case and there are situations where they can result in significant errors in the estimation of unsupported ^{210}Pb activity. Fig. 9 illustrates an extreme example from Dalvatn, northern Norway (Norton *et al.* 1992), where the sediment record contained a discrete layer (at 3 - 6 cm depth) in which ^{226}Ra activity was an order of magnitude higher than the normal value of 30 - 50 Bq kg^{-1}. In such cases, ^{222}Rn diffusion, sustained over a number of decades, may result in significant differences between the supported (*in situ* generated) ^{210}Pb activity and the ^{226}Ra activity. Calculations using a simple diffusion model show that supported ^{210}Pb activity is reduced in the ^{226}Ra rich sediments and increased in the adjacent layers. Unsupported ^{210}Pb was estimated by subtracting the calculated values from the total ^{210}Pb. CRS model calculations based on the results indicated that the ^{226}Ra rich sediments were associated with an episode of rapid sedimentation around 1910.

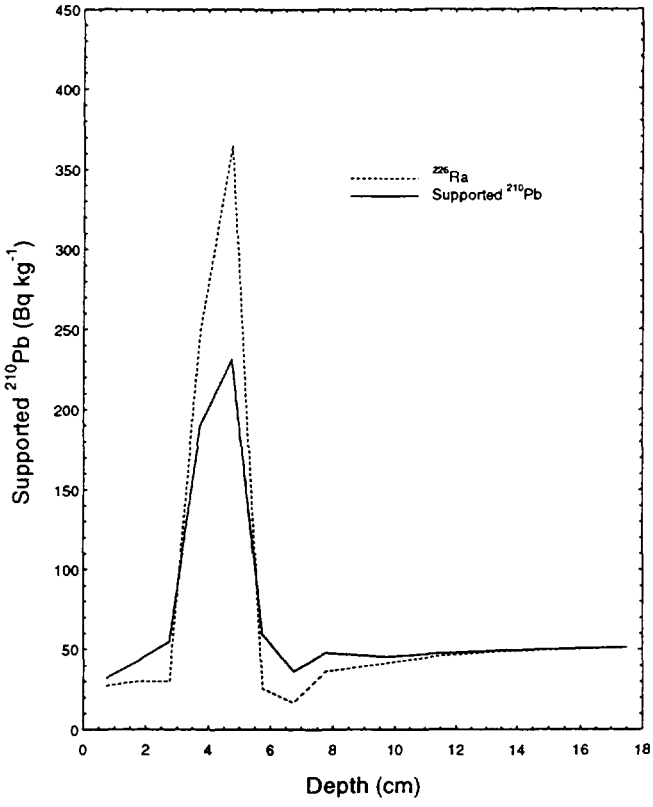

Figure 9. *Supported ^{210}Pb and ^{226}Ra in a sediment core from Dalvatn, Norway (Norton et al. 1992). The ^{226}Ra activities (-----) were determined by direct gamma assay. The supported ^{210}Pb activities (——) were calculated using a simple diffusion model.*

10 Conclusion

It is unlikely that dating by ^{210}Pb will ever be a totally routine procedure. In the relatively closed system of a lake and its catchment where the CRS model has an underlying theoretical basis, potential complexities in transport processes are such that neither of the simple models (CRS and CIC) can be presumed without independent validation. The situation is considerably more difficult in marine environments where there is no *a priori* reason to suppose

that ^{210}Pb supply rates are governed by any simple relationship. In these circumstances the CRS model might just give the best results because of the robustness of the dating parameter. Apart from the fact that integration is a relatively reliable numerical procedure, it also has the effect of smoothing out minor irregularities.

Since validation can at best be achieved at a small number of points, good quality results depend on the adoption of good numerical procedures that take account of the strengths and weaknesses of the particular dating model being used. The CIC model will be prone to large errors if there is significant mixing of the surficial sediments. The CRS model may give nonsensical results if there are gaps in the sediment record. Potential dating problems may be highlighted by routine calculation of ^{210}Pb dates using both models. Comparisons between the two sets of dates will help determine appropriate procedures for correcting or improving the initial calculations. These procedures, some of which are outlined above, are best carried out in the light of a good understanding of the potential transport processes controlling the supply of fallout ^{210}Pb to the sediment record.

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