

Constructing recent peat accumulation chronologies using atmospheric fall-out radionuclides

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SUMMARY

Radionuclide dating is a robust technique for establishing the recent accumulation rate for most peatlands. In this article we review the production of ‘fall-out’ radionuclides and the concepts underlying the dating method. Some inherent problems such as the issues surrounding reduced natural radionuclide deposition, e.g. ‘continentality’, are highlighted. We describe the common radionuclide analysis methods, their application to peat studies, and the production of ²¹⁰Pb activity inventories. To illustrate the radionuclide dating method we use case studies to demonstrate how these data can be used to derive peat accumulation rates during the last 100–150 years at contrasting sites using ²¹⁰Pb supported by anthropogenic radionuclides.

KEY WORDS: ²⁴¹Am, cross-validation, CRS model, ¹³⁷Cs, mire, ²¹⁰Pb dating.

1. INTRODUCTION

Radionuclide dating methods can be used to produce age markers and accumulation chronologies for peat sequences spanning many thousands of years. However, this paper will describe the use of ‘fall-out’ radionuclides to construct chronologies for young peat sequences, typically deposited during the last 150 years. Detailed introductions to radioactivity and the development of radiometric dating can be found in texts like Smart & Frances (1991) and Lowe & Walker (1997), and the reader is directed to these texts for background information. Technical descriptions covering the use of different radionuclides and methods are presented in Rutter & Catto (1996), Dickin (1997) and Carroll & Lerche (2003). We shall focus here on the practical application of this dating method for peat studies, specifically peat bog studies, but also draw attention to the critical underlying concepts.

2. RADIONUCLIDES

A radionuclide is an atom with an unstable nucleus that undergoes α or β radioactive decay, or transmutation, by emitting or receiving sub-atomic particles. The decay is exponential in trend, and the time taken for half of the original number of atoms to decay is termed one half-life. The basic principal of radiometric dating is to derive the number of

half-lives that have passed since decay began by comparing the number of parent nuclide atoms remaining in a sample with the number of daughter atoms present.

The two natural radionuclides that are commonly used to date recent peat sequences are radiogenic ²¹⁰Pb and cosmogenic ¹⁴C (Oldfield *et al.* 1995, Appleby *et al.* 1997, Piotrowska *et al.* 2011). Lead-210 is a natural radionuclide belonging to the ²³⁸U radioactive chain, while ¹⁴C is produced naturally in the upper atmosphere from collisions of free neutrons (produced by cosmic rays) with ¹⁴N₂ molecules. Beryllium-7 is a less-used cosmogenic radionuclide which is relevant to some peat studies. This radionuclide has a half-life of only 53.3 days, and so is of limited use as a dating tool. On the other hand, because its activity very quickly drops below detection limits (Walling & Quine 1995, Mabit *et al.* 2008), it can be used to confirm the presence of an active accretion surface at the top of a sequence, as well as the growth of new vegetation. Because of its short half-life, it must be measured as soon as possible after sample collection.

2.1 Lead-210

Lead-210 is commonly used to date sediments and peats accumulated during the last 150 years in both terrestrial and marine environments. The original proposal of ²¹⁰Pb as a dating tool is attributed to Goldberg (1963), but it was not widely used until after the first application to lake sediments by

Krishnaswami *et al.* (1971). The total ^{210}Pb ($^{210}\text{Pb}_{\text{total}}$) activity initially supplied to the sediment or peat is a combination of the supported ($^{210}\text{Pb}_{\text{sup}}$) and unsupported or excess ($^{210}\text{Pb}_{\text{ex}}$) fractions. In order to date a peat sample using ^{210}Pb , we need to determine both fractions. To understand some of the issues that can arise with ^{210}Pb dating and why it does not work at some sites, it is desirable to understand the distinction between these two components of total ^{210}Pb activity and the processes involved in their emplacement (Appleby 2001).

Lead-210 is produced by the eventual decay of primeval ^{238}U , via intermediate daughters, to ^{222}Rn gas. Some of this diffuses through rocks and soils and escapes to the atmosphere, but some decays within the sediment matrix to ^{210}Pb (Figure 1). Radon-222 production is dependent upon the local geology and the uranium concentrations in rocks and soils. In western Europe, high ^{226}Ra , ^{222}Rn and ^{210}Pb activity in sediments is closely associated with the uranium-rich granites emplaced during the Variscan Orogen in the south-west of Ireland and western England, central France and the Iberian Peninsula (Plant *et al.* 2003). Other geologies, including volcanic terrains (e.g. D'Alessandro & Vita 2003), can also produce ^{222}Rn and ^{210}Pb activity; but at reduced levels when compared to the activity found in soils and sediments formed over crystalline granites (Plant *et al.* 2003). The amount of ^{210}Pb activity in a sample is presumed to be produced by the *in situ* decay of ^{226}Ra and then

^{222}Rn . This is termed the supported fraction ($^{210}\text{Pb}_{\text{sup}}$), and under monotonic conditions it can be expected to remain relatively constant through time because it is being constantly replenished as it decays (Figure 1).

The unsupported or excess ^{210}Pb fraction ($^{210}\text{Pb}_{\text{ex}}$) is produced by the decay of gaseous ^{222}Rn ($T_{1/2} = 3.8$ days) in the atmosphere, and is not supported by the local ^{226}Ra activity. Once formed, ^{210}Pb is rapidly bound to atmospheric particles ($\sim 0.5\text{--}1\ \mu\text{m}$) and deposited on the earth's surface (Figure 1). Precipitation plays an important role here because it scavenges lead from the air, along with other microscopic dust particles and aerosols, and carries it down to the ground. In ombrotrophic peatlands (bogs), we assume that atmospheric fallout supplies all inorganic material so that the main component of ^{210}Pb in the peat is unsupported (Appleby *et al.* 1997).

2.2 Nuclear fall-out materials

In addition to natural radionuclides, nuclear activities and accidents associated with both military weapons development and civil energy production have released radionuclides into the environment (e.g. ^{90}Sr , ^{134}Cs and ^{137}Cs , ^{239}Pu and ^{240}Pu , ^{241}Np , ^{241}Pu and ^{241}Am , and $^{14}\text{C}_{\text{bomb}}$). If the signals of these events were detectable in a peat sequence, they could be used as chronological markers. Figure 2 shows the ^{137}Cs deposition histories for the Northern and Southern Hemispheres and the timings of the

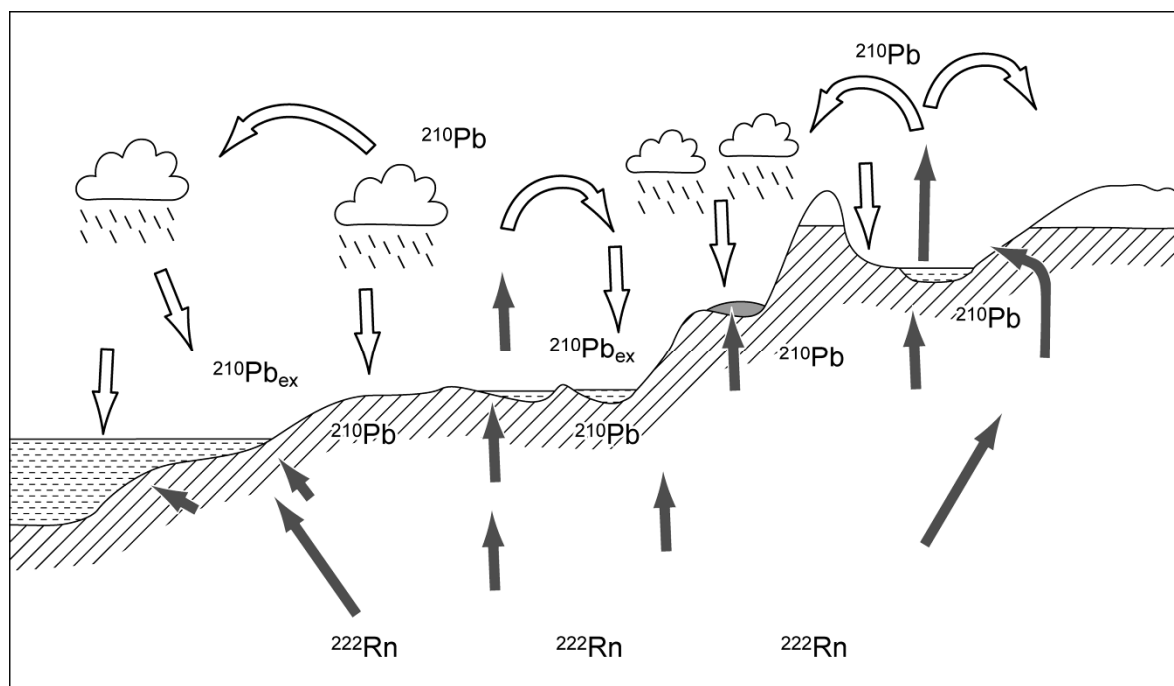


Figure 1. Lead-210 production and relative atmospheric deposition levels (after Preiss *et al.* 1996).

activities recorded at three specific European sites. The Northern Hemisphere curves have two distinct peaks. The older is the signal of the atmospheric nuclear weapons tests, and the younger arises from the catastrophic nuclear reactor explosion and fire at Chernobyl in the Ukraine on 26 April 1986. This incident initially released approximately $2 \cdot 10^{18}$ Bq of radioactive materials into the lower atmosphere (Smol 2002, Kashparova *et al.* 2003), but unlike the weapons tests did not propel radioactive particles and dust sufficiently high into the atmosphere for

global distribution to occur (Appleby 2001), so that we do not see its signal in distal records.

Not all European locations received fall-out from Chernobyl because the dose deposited on the ground was dependent upon regional air flows and local precipitation patterns in the days immediately following the event (Table 1). At some locations the Chernobyl contribution overprints the earlier weapons signature and the two peaks become blurred, but at other sites two distinct signals can be identified.

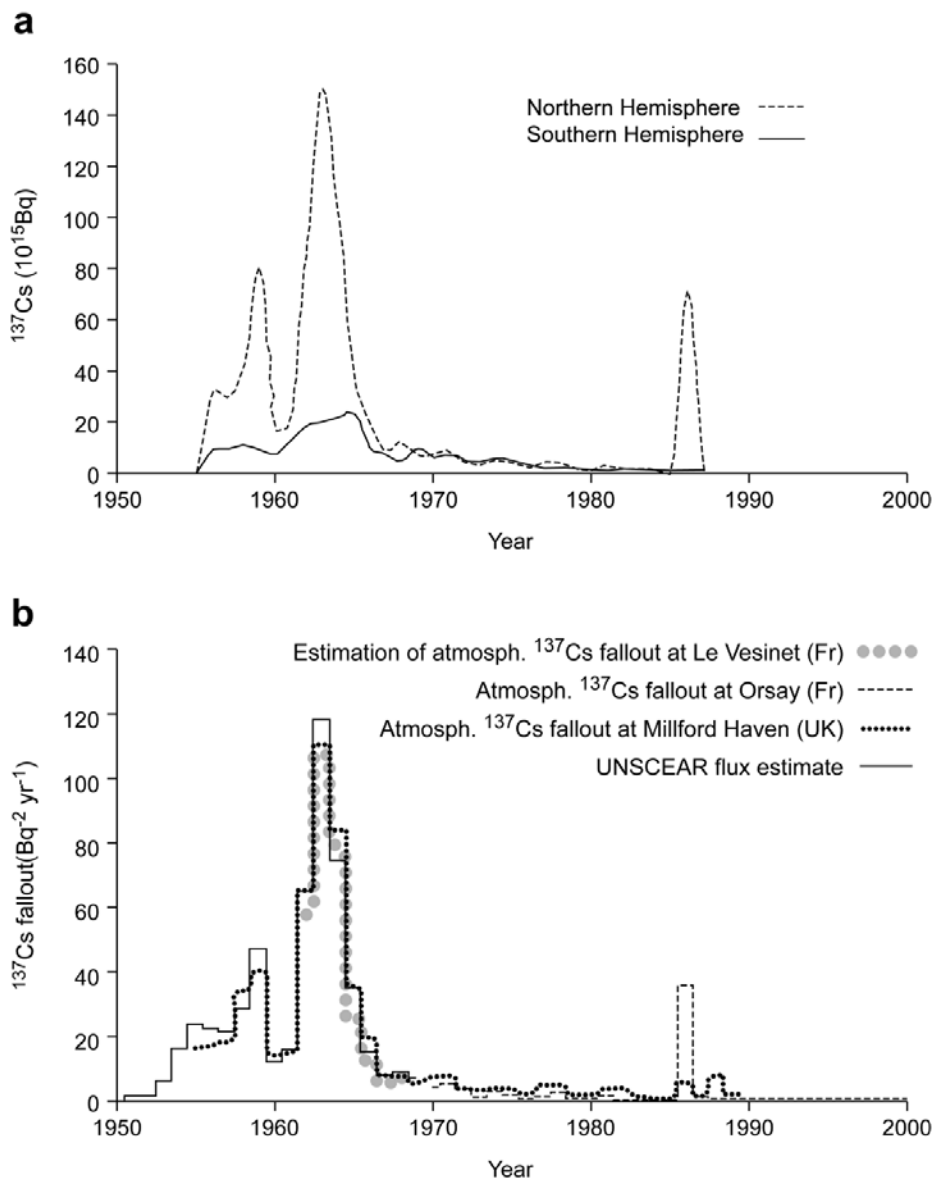


Figure 2. (a) The deposition of ^{137}Cs fall-out per year for the Northern and Southern hemispheres up to 1987. From Cambray *et al.* (1989). (b) Annual atmospheric ^{137}Cs fallout at Orsay, expressed in $\text{Bq m}^{-2} \text{ yr}^{-1}$ (Barker & Bouisset 2001), and at Millford Haven from 1957 to 1991 (EML 2000), estimate of ^{137}Cs deposition for Paris (UNSCEAR 1982) and Le Vésinet (Renaud & Louvat 2004), modified from Monna *et al.* (2009).

Table 1. Spatial variation of ^{137}Cs radioactivity in soils in selected parts of the Northern Hemisphere. Measured in the top 5cm, and decay corrected to 03/05/86. Data from Cambray *et al.* (1989). See also Figure 1.10 in Smith & Beresford (2005).

Location	Total ^{137}Cs activity Bq m^{-2}	^{137}Cs activity attributed to Chernobyl Bq m^{-2}
UK		
Holmrook, Cumbria	13000	10000
Lerwick, Shetland	6100	4600
Antrim, Northern Ireland	4000	2200
Trawsfynydd, Gwynedd	3100	1850
Dounreay, Caithness	3000	1250
Eskdalemuir, Dumfries and Galloway	1300	820
South Brent, Devon	700	<40
Carmarthen, Carmarthenshire	610	43
Culham, Oxfordshire	370	<70
Ashbury, Wiltshire	330	25
Northern Europe and Scandinavia		
Budapest, Hungary	13000	8800
Belgrade, Serbia	8900	7300
Bucharest, Romania	5600	4300
Warsaw, Poland	3600	2860
Bonn, Germany	1800	1550
Vienna, Austria	1600	1150
Helsinki, Finland	1400	990
Stockholm, Sweden	600	430
Remy, France	430	290
Moscow, Russia	370	210
Reykjavik, Iceland	66	7

3. ANALYSIS AND MEASUREMENT

For a detailed description of the analytical methods involved in ^{210}Pb dating, the reader is directed to Appleby (2001) and Ebaid & Khater (2006) and references therein. Three techniques are available for measurement of ^{210}Pb :

- directly by measurement of ^{210}Pb gamma emission;
- indirectly by measurement of its alpha-emitting grand-daughter ^{210}Po ; and
- indirectly by measurement of its beta-emitting daughter ^{210}Bi .

We focus on the first two techniques because the third, despite its high sensitivity, has to our knowledge never been used for peat and is rarely used for peat-age dating. To make these measurements we can use two different analytical methods, gamma spectrometry or alpha spectrometry.

Gamma spectrometry has the advantage of being a non-destructive method which directly measures the gamma emissions from a sample (Appleby 2001). Some preparation, e.g. drying and pulverising, is required; but because the method is non-destructive the measurements can be repeated

and the sample can be used subsequently for other analyses, e.g. geochemistry. The total ^{210}Pb activity in peat samples can be determined directly by gamma spectrometry. However, because of the low energy of ^{210}Pb gamma emission (46.5 keV), a spectrometer with a high-energy spectrum is necessary. Also because of the low emission energy and the problem of self-adsorption, difficulties can be experienced with large-volume samples, but these can be minimised by using well-type detectors designed for low-volume samples (Appleby 2008). This said, gamma spectrometry has the advantage of being able to provide simultaneous measurements of ^{210}Pb and other radionuclides, including the artificial 'nuclear fall-out' species (^{241}Am : 59.54 keV; ^{137}Cs : 662 keV); as well as ^{214}Pb activity (295keV and 352keV), which can be used to estimate the $^{210}\text{Pb}_{\text{sup}}$ - ^{226}Ra activity in a sample (Appleby 2001). Normally, samples are placed in sealed containers and left for three weeks before counting to ensure $^{222}\text{Rn}/^{226}\text{Ra}/^{214}\text{Pb}$ equilibration. Therefore, ^{214}Pb activity will be in equilibrium with its progeny and we can use the ^{214}Pb activity in a sample as an estimation of $^{210}\text{Pb}_{\text{sup}}$.

If we do not need to measure ^{210}Pb , ^{137}Cs and ^{241}Am simultaneously, alpha spectrometry offers an alternative to gamma spectrometry. This method can produce better precision with low-activity ^{210}Pb samples. However, the preparation of peat samples for alpha spectrometry is more complex and time-consuming than for gamma spectrometry, and because it requires digestion of the sample it is destructive. In addition, ^{210}Pb can be determined only indirectly in peat samples, e.g. by measurement of its decay product ^{210}Po or by assuming ingrowth of the ^{210}Pb in equilibrium with ^{226}Ra . To use the activity of ^{210}Po as a proxy for ^{210}Pb , the Po must be chemically extracted from the material. After digestion and chemical treatment the ^{210}Po in solution is deposited onto a silver disk for analysis. To determine the efficiency of the extraction and ensure no loss of ^{210}Po during sample preparation, each sample is spiked with a known amount of ^{208}Po or ^{209}Po , which have the same chemical properties as ^{210}Po but emits alpha particles at a different energy level.

There is no standard peat digestion for ^{210}Pb determination (*via* ^{210}Po) using alpha spectrometry. In geochemical studies, peat samples are often digested using H_2O_2 (to break down organic matter) followed by a combination of concentrated acids, e.g. HNO_3 and HCl . When total digestion is required, and when assaying samples containing siliciclastic material, more aggressive reagents like hydrofluoric acid (HF) are used in an additional stage. It is possible to use heat to break down the

organic fraction before digestion, but this requires careful control of the burn temperature to minimise the loss of volatiles. It is also possible to use a microwave digestion system. However, because unsupported ^{210}Pb is mainly bound, like stable Pb, to organic matter and iron hydroxides and is not present in the peat matrix, a total digestion using HF is not usually needed; a simple digestion using $\text{HNO}_3 + \text{H}_2\text{O}_2$ with filtration is sufficient to ensure good recovery of ^{210}Pb .

In ombrotrophic peatlands, especially after alpha measurements, some authors use measurements taken in deep peat sections to estimate $^{210}\text{Pb}_{\text{sup}}$, which should be low because ombrotrophic peatlands are fed exclusively by atmospheric inputs.

4. INITIAL ACTIVITY LEVELS AND INVENTORIES

Lead-210 has a relatively short half-life of 22.3 years, so the amount of $^{210}\text{Pb}_{\text{ex}}$ activity remaining in most samples is statistically undetectable after about seven half-lives. For ^{210}Pb dating to work, the surface activity of $^{210}\text{Pb}_{\text{ex}}$ must sufficiently high that activity levels in sub-surface peat remain measurable, allowing for their exponential decay, when the time period of interest has elapsed. If the surface activity is low then the activity in sub-surface peat will rapidly drop below the minimum detection level. This means that if the peat on the surface of a bog retains 100 Bq kg^{-1} of ^{210}Pb , exponential decay of the lead will reduce the activity to $\sim 25 \text{ Bq kg}^{-1}$ after the passage of two half-lives (44.6 years). Given that some laboratories typically report an analytical uncertainty of $\pm 15\text{--}25 \text{ Bq kg}^{-1}$ for gamma-counted ^{210}Pb data, we can see that low activity levels can curtail the precision of the age model. Low activity problems may be caused by poor local retention of atmospheric ^{210}Pb at some sites due to local environmental conditions leading to, for example, removal by wind during droughts or water during floods. Alternatively, the site may be in a zone of low regional atmospheric ^{210}Pb deposition. Radon-222 distribution in surface air, and hence atmospheric ^{210}Pb production, is not spatially uniform across the surface of the Earth (Table 2).

As a general rule, atmospheric ^{210}Pb levels tend to be depleted in rain-shadow zones on the 'wrong sides' of mountain ranges, and trends of decreasing ^{210}Pb deposition are observed when moving westwards across continents (Appleby 2001). If produced under a lake or ocean, a significant fraction of the gaseous ^{222}Rn will diffuse into the overlying water body and be contained within it,

Table 2. Variations of annual mean ^{210}Pb concentration in surface air at different geographical locations. Selected from Preiss *et al.* (1996).

Location	^{210}Pb concentrat ⁿ . (mBq m ⁻²)	Location	^{210}Pb concentrat ⁿ . (mBq m ⁻²)	Location	^{210}Pb concentrat ⁿ . (mBq m ⁻²)
Anchorage	310	Dye 3 (Greenland)	180	Calcutta	945
Salt Lake City	707	Tromso	167	Chiba (Japan)	370
Los Angeles	540	Moscow	310	Pretoria	750
Washington (W.D.C.)	465	Chilton (UK)	230	Reunion Island	110
New York City	580	Dublin	135	Honolulu	178
Winchester (USA)	635	Galway	130	Brisbane	260
New Orleans	770	Rosslare	205	Melbourne	190
Miami	283	Brunswick	370	Lower Hutt (S. NZ)	75
Panama City	116	Warsaw	320	Falkland Islands	50
Lima (Peru)	247	Paris	480	Antarctica Peninsula	14
Santiago	332	Bordeaux	590	South Pole research station	32

and permanent ice cover will significantly reduce ground surface emissions of ^{222}Rn . Therefore, surface air concentrations of ^{222}Rn tend to be higher at sites close to the centres of continental land masses (Figure 3a), and lower near ice sheets and in oceanic locations (Chevallard *et al.* 2002, Conen & Robertson 2002). However, in some mountainous areas, alpine bogs can receive an enhanced ^{210}Pb deposition flux (and therefore $^{210}\text{Pb}_{\text{ex}}$ inventory) due to specific meteorological processes like the 'feeder-seeder effect' and aerosol canopy interception (Le Roux *et al.* 2008). To summarise, we find that sites with high ^{222}Rn emissions and high annual rainfall produce the highest ^{210}Pb fluxes and, therefore, greater total activity inventories for soils (see Figure 3b).

5. LEAD-210 AGE MODELS

Sequential $^{210}\text{Pb}_{\text{ex}}$ data are processed to obtain sediment ages using an appropriate model of radioactive decay through time. A number of subtle models with varying degrees of sophistication and complexity have been developed (Carroll & Lerche 2003), but the original model of ^{210}Pb dating was the Constant Flux, Constant Sedimentation (CF-CS) model developed by Krishnaswami *et al.* (1971) and Robbins (1978). This model extends the basic radiometric dating principal and makes the following assumptions:

1. there has been a constant rate of ^{210}Pb deposition from the atmosphere;
2. the $^{210}\text{Pb}_{\text{ex}}$ activity in the peat is due to atmospheric deposition only;
3. there has been no post-accumulation disturbance or redistribution of the accumulated peat; and
4. there has been steady-state dry-mass peat accumulation.

Under these conditions, a logarithmic plot of $^{210}\text{Pb}_{\text{ex}}$ per unit mass of peat vs. depth should indicate a linear relationship, and the peat accumulation rate can be determined from its slope. However, the number of environments that satisfy all prerequisites for the CF-CS model to perform satisfactorily is limited (Carroll & Lerche 2003). Short-term fluctuations superimposed on longer-term changes in accretion rate are common features of many sedimentary environments.

To facilitate ^{210}Pb dating at sites where the sedimentation or peat accumulation rate has not been constant, a more sophisticated model that could compensate for changes in the dry sediment supply was devised. This became known as the Constant Flux (CF) model (Robbins 1978) or the Constant Rate of Supply (CRS) model (Appleby & Oldfield 1978, Appleby *et al.* 1979). In this model it is assumed that the ^{210}Pb flux has remained constant through time, but the peat accumulation rate may have changed. The CRS model is most commonly used in ^{210}Pb dating today. It is known to produce reasonable results for most

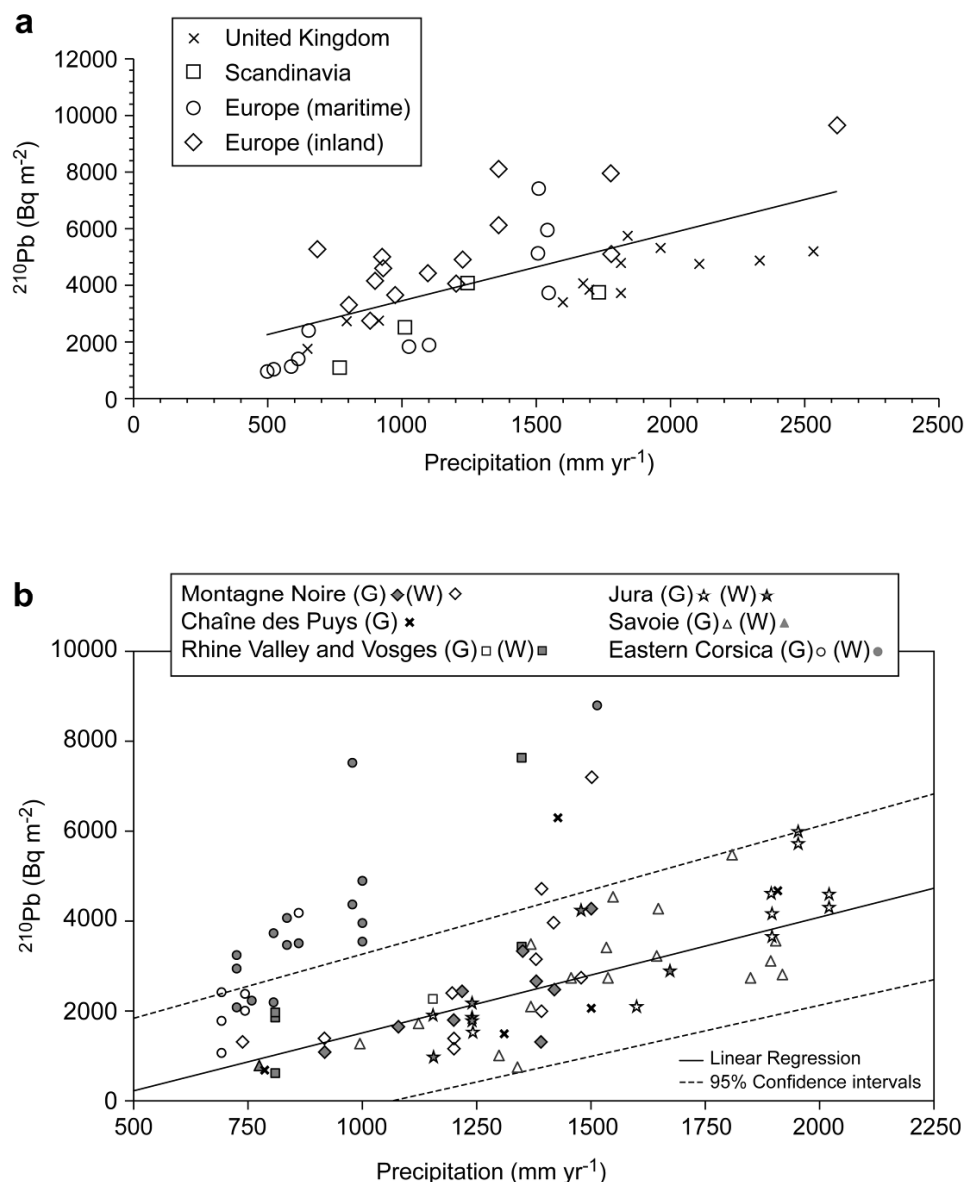


Figure 3. (a) ^{210}Pb inventory vs. precipitation rate observed in Europe and Scandinavia, from Appleby (2001) and personal communication. (b) ^{210}Pb inventory vs. precipitation rate for French soils from different mountain massifs, from Le Roux *et al.* (2008).

environments that have received a steady flux of unsupported ^{210}Pb directly from atmosphere (Appleby 2001), even where the organic matter content changes with depth. Therefore, this model can compensate for inter-annual variations in the mass accumulation rate of *Sphagnum* peat driven by short-term variability in local weather.

An alternative to the CRS model is the Constant Initial Concentration (CIC) model (Appleby 2001). Sometimes termed the Constant Initial Activity (CIA) (Appleby & Odfield 1978) or the Constant Specific Activity (CSA) model (Robbins 1978), this

model was developed for environments where the unsupported ^{210}Pb flux is not supplied directly by local atmospheric fall-out. It is designed to work when the local ^{210}Pb flux is responding to remote changes in larger sedimentary systems (e.g. abyssal environments in deep ocean basins or large lakes), and so is not an obvious choice for peatland work. This model allows the sedimentation or peat accumulation rate to change through time, but unlike the CRS model, the CIC model assumes that the initial unsupported ^{210}Pb activity has remained in proportion to the sediment supply (Appleby 2001).

6. PEAT SAMPLE PREPARATION

We begin with the assumption that we want to use radionuclide analysis to create a chronology for a typical high-latitude peat core. We first need to prepare sequential samples, including the top living vegetation layers (Olid *et al.* 2008), usually at 1 cm intervals cut as slices across our peat core to a depth that we estimate to span the last 150 years. This estimate might be based on a similar core from the same area or a simple peat growth model, but 50–60 cm is usually deep enough. Normal good laboratory practices and care should be exercised when taking the samples (Givelet *et al.* 2004, De Vleeschouwer *et al.* 2010). For alpha spectrometry we need 0.5–1.0 g of dry peat for the acid digestion described in Section 2, but if we are using gamma spectrometry we shall need significantly larger samples. The actual sample size depends on the geometry of the counter used, and more than 100 g of peat is required for some planar geometries, but a typical germanium well-detector designed for low-volume samples will take 1–3 g of peat powder. Low-density material can be troublesome due to self-absorption of ^{210}Pb gamma emissions in large-volume samples. Typically, the sample powder is firmly packed into a screw-top plastic cylinder approximately 10 mm in diameter and 40 mm high

using a glass bar as a ram-rod so as to achieve maximum activity for the geometry. Once packed, the cylinders are sealed using adhesive tape and left undisturbed for 21 days or more to allow radiometric equilibration between ^{226}Ra and ^{222}Rn .

If we intend to use the CRS model we shall need to obtain bulk density data for the peat samples intended for radionuclide analysis. Ideally, we want bulk density for each slice of peat and not an average for the core. A simple method consists of measuring or calculating the cross-section of the core (cm^2) and dividing this into the dry weight (g) of the (1 cm thick) slice to give a result in g cm^{-3} . If we know that peat growth is likely to have been slow, we may need to use 0.5 cm slices to capture data at the required resolution, e.g. to fully resolve the ^{137}Cs record.

7. VALIDATION AND PITFALLS

An in-depth explanation of the use of ^{210}Pb age models is beyond the scope of this guide and the reader is directed to Appleby (2001) and Carroll & Lerche (2003). However, there are a number of common inherent problems that the user needs to be aware of, and certain constraints that may require consideration (see Table 3). For example, before

Table 3. Summary of the common problems and issues that may be encountered when using ^{210}Pb CRS modelling to date peat.

During field coring	➤ site with very low, spasmodic or highly variable peat accumulation rates;
	➤ the presence of too many vascular plants (e.g. <i>Calluna vulgaris</i> , <i>Molinia caerulea</i> , <i>Pinus</i>) resulting in root disturbance of the peat stratigraphy;
	➤ minerotrophic site (e.g. valley mire) with excessive temporal variability of detrital clastic input;
	➤ disrupted stratigraphy or peat compression due to using an inappropriate coring device.
In the laboratory	➤ inaccurate measurements of peat bulk density;
	➤ incomplete digestion of peat samples for alpha spectrometry;
	➤ using an inappropriate geometry for ^{210}Pb gamma spectrometry measurements, e.g. sample is too small for the container selected;
	➤ unreliable estimation of ^{210}Pb supported activity due to incorrect instrument calibration;
	➤ low precision due using inappropriate counting times.
Modelling	➤ hiatus in the profile;
	➤ lack of validation and cross checking with other independent dating methods (e.g. $^{14}\text{C}_{\text{bomb-pulse}}$, pollen, SCP);
	➤ the ^{210}Pb activity is too low for reliability because of low initial deposition at surface, or poor retention in some samples;
	➤ wrong equilibrium point selected between supported and unsupported ^{210}Pb fractions.

modelling the ^{210}Pb data, we can check whether the ^{210}Pb activity measured at the surface is realistic and within the range expected for the region. We also need to check that the unsupported inventory is essentially complete and that the reduction in activity in the lower peat horizons broadly fits the exponential decay model. Any significant abrupt departures from this model indicate a non-monotonic record which may prevent successful modelling of the chronology using the standard approach. In such situations, more data may be needed to understand the record.

It is always desirable to validate ^{210}Pb chronologies by comparing them with results obtained using independent dating methods on the same peat sequence (Turetsky *et al.* 2004). A common approach is to use the ^{137}Cs signals from nuclear weapons testing during the 1950s and 1960s, and in some areas of the Northern Hemisphere the Chernobyl incident of 1986. In some cases, other independent chronological markers can be derived, e.g. from the timing of establishment of pine plantations or *Cannabis* planting (pollen) (Appleby *et al.* 1997), the introduction and withdrawal of fuel additives (stable lead isotopes) and the rise of industrial coal burning (SCPs) (Swindles 2010).

8. WORKED EXAMPLES

We present here examples of radionuclide dating from two bog sites, one in northern Europe and the other in North America. Both sites were cored using a Wardenaar corer, thus avoiding peat compression, and the samples were processed using the advanced cutting and preparation techniques described by Givélet *et al.* (2004). Lead-210, lead-214, ^{137}Cs and ^{241}Am activities (Figure 4) were determined using a Canberra gamma spectrometer at the University of Heidelberg. Instrumental details are described by Givélet *et al.* (2004) and Rausch *et al.* (2005).

The European site is located in the Viurusuo mire complex some 8 km south-west of the town of Outokumpu in Finland. A number of peat cores were obtained and used to investigate past atmospheric deposition of heavy metals (Rausch *et al.* 2005). One of these cores was particularly suitable for ^{210}Pb age dating, with an unsupported near-surface ^{210}Pb activity in excess of 170 Bq kg^{-1} . The ^{210}Pb activity decayed with depth to below 40 cm, where it dropped beneath the detection limit of the gamma counter. The decay rate conformed to an approximate exponential model (Figure 4a), suggesting that this was a monotonic site which had been receiving a relatively steady-state supply of

^{210}Pb , and that more than five half-lives had elapsed since deposition of the lead on the palaeo-surface at 40 cm depth. In addition, ^{137}Cs and ^{241}Am activities were detected in the peat (Figure 4a).

Caesium-137 and ^{241}Am were both produced by thermonuclear weapons tests in the 1950s and 1960s, but the majority of the ^{241}Am activity detected in peat is the in-growth product of the decay of ^{241}Pu released during the explosions. Therefore, when we find low levels of ^{241}Am activity coexisting with a maximum in ^{137}Cs activity in an ombrotrophic peat profile, this normally confirms that the ^{137}Cs in question was deposited during the height of the weapons-test fallout in the 1960s (Appleby 2008, Le Roux *et al.* 2010). Nonetheless, it is important to note that this interpretation does not always hold true if the nature of the site is such that its sediments contain radionuclide contributions that are independent of the simple atmospheric pathway, e.g. tidal salt-marsh (Marshall *et al.* 2007) or minerotrophic peatland.

When we examine the Viurusuo ^{137}Cs record, we find that it does not fit the model described above. There is a single large peak in ^{137}Cs activity which is significantly closer to the surface than the peat showing ^{241}Am activity. To resolve this we must consider the regional deposition of radionuclides in the Northern Hemisphere. We know that, in addition to atmospheric fall-out from thermonuclear tests during the 1950s and 1960s, some locations (Table 1) received fall-out containing ^{137}Cs from Chernobyl in 1986, and significant amounts of this material fell in Finland. Indeed, Table 2 indicates that some two-thirds of the total ^{137}Cs activity detectable in soils in this part of Scandinavia is attributable to the 1986 reactor fire at Chernobyl; and thus that this signal is likely to be a prominent feature of any Finnish ^{137}Cs record.

To check of the robustness of the Viurusuo ^{210}Pb record we can compile the ^{210}Pb peat inventory and compare it with the annual precipitation rate. The precipitation rate at Outokumpu is 600 mm yr^{-1} , whereas the complete $^{210}\text{Pb}_{\text{ex}}$ inventory is 6000 Bq m^{-2} . A comparison with the information in Figure 3 indicates that presence of the complete ^{210}Pb inventory can be assumed because the expected $^{210}\text{Pb}_{\text{ex}}$ activity values in Scandinavia for a precipitation rate of 600 mm yr^{-1} are around 4000 Bq m^{-2} . A chronology for Viurusuo was produced from the $^{210}\text{Pb}_{\text{ex}}$ data using the CRS model (Figure 5a), and to validate this we first use the ^{241}Am activity to mark the location of peat horizons deposited in the early 1960s and find reasonable agreement with the CRS model. We next express the Viurusuo ^{137}Cs activity as an area flux

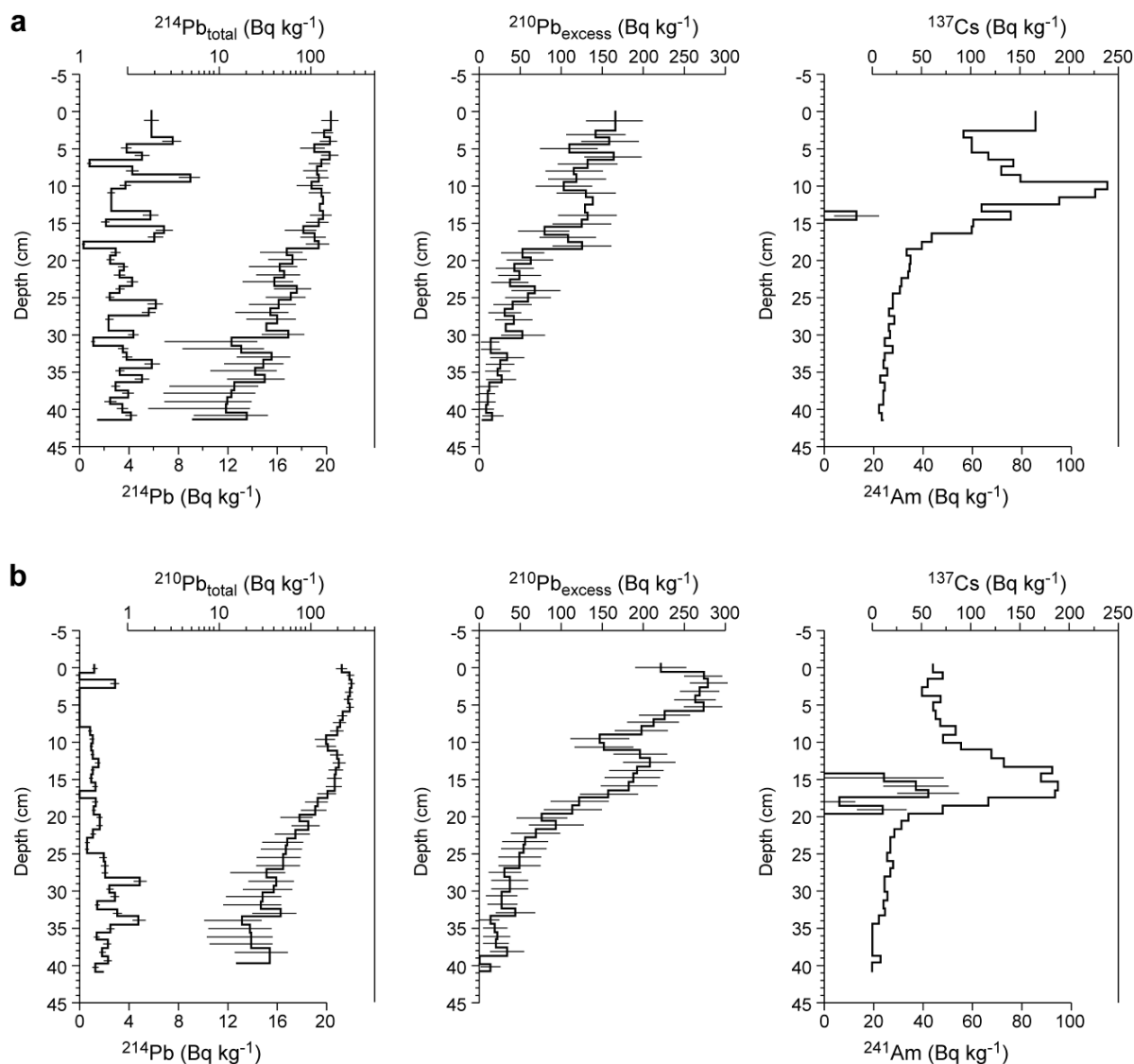


Figure 4. (a) Radionuclide analysis, Viirusuo, Outokompu, Finland (Rausch *et al.* 2005); (b) radionuclide analysis, Sifton Bog, Canada (Givelet *et al.* 2003).

(Figure 5a) and find that the peak of maximum ^{137}Cs activity is in agreement with the CRS model age of 1987 ± 2 years. Dates produced for the Viirusuo core using ^{14}C data calibrated against the bomb-spike curve (see Goodsite *et al.* 2001, Piotrowska *et al.* 2010) also support the ^{210}Pb CRS chronology in the post-bomb period (Figure 5a).

The second example is from Canada. Givelet *et al.* (2003) used peat cores to investigate past atmospheric deposition of mercury at Sifton Bog near the city of London, Ontario. One of the cores was processed in a similar manner to the Finnish core discussed above to produce a suitable dataset for radionuclide dating (Figure 4b). As the total ^{210}Pb inventory for Sifton Bog was measured at

4500 Bq m^{-2} and this agreed with the value expected for a precipitation rate of 850 mm yr^{-1} , it was possible to produce a chronology using the CRS model as for Viirusuo (Figure 5b).

To validate the Sifton Bog CRS chronology, we use the nuclear fall-out radionuclides, and find that the ^{241}Am data supports the CRS ages. However, this region of Canada did not receive significant levels of fall-out from the Chernobyl incident (Taylor *et al.* 1988). Therefore, when we examine the ^{241}Am and ^{137}Cs data, we find the maximum activity for both at 14–15 cm, confirming that this is the stratigraphic location of the 1963 peak in fall-out from atmospheric nuclear weapons tests. This date is in agreement with the CRS model. Furthermore,

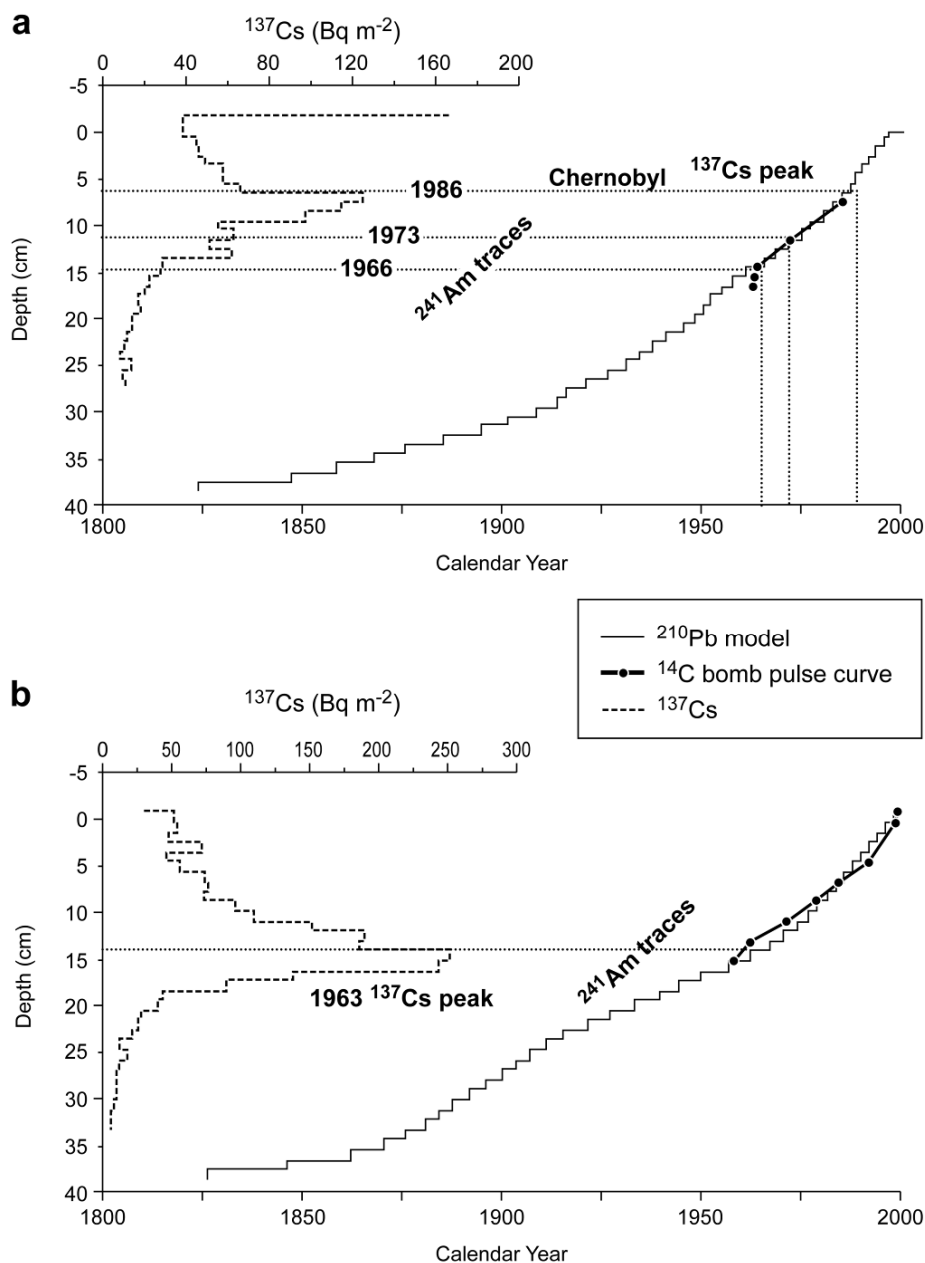


Figure 5. Examples of age-depth models for peat cores derived from radionuclides: (a) Viirusuo, Outokompu, Finland (Rausch *et al.* 2005); (b) Sifton Bog, Canada (Givelet *et al.* 2003).

when we integrate $^{14}\text{C}_{\text{bomb-pulse}}$ dates we find these are also in agreement with the ^{210}Pb chronology produced using the CRS model (Figure 5b).

It is desirable for all ^{210}Pb chronologies to be independently validated (Smith 2001), and we have shown here how the nuclear fall-out radionuclides ^{241}Am , ^{137}Cs and $^{14}\text{C}_{\text{bomb-pulse}}$ can be successfully used for this purpose. As demonstrated in Figure 5, ^{14}C bomb-pulse dating and the ^{210}Pb CRS model are complementary. These two dating models are

independent of one another because the mechanisms of ^{210}Pb and ^{14}C integration in peat are completely different. The lead is deposited onto the bog surface as particulate metallic fall-out from atmosphere which resides on the outside of the plant material and slowly becomes entombed in the peat. The carbon, on the other hand, originates from the atmospheric CO_2 reservoir and is assimilated into the plant tissue during photosynthesis. The use of ^{241}Am and ^{137}Cs adds further support to a

chronology because, although both these radionuclides are delivered to the bog as particulate material, they have different geochemical attributes. For example ^{210}Pb , ^{241}Am and its parent ^{241}Pu are particle reactive nuclides which, like stable lead, are relatively immobile in peat sequences; but ^{137}Cs is mobile and soluble in acidic pore water. Therefore, these nuclides can be expected to respond differently to changes in redox conditions, bog hydrology and other local factors.

The cross-validation approach described above works best in peat bogs with recent high accumulation rates which receive ^{210}Pb exclusively from the atmosphere, but good results can still be obtained in slow accumulation sites and/or minerotrophic sites. Furthermore, although the ^{210}Pb CRS model assumes relatively steady peat accumulation and retention of the full radionuclide inventory, the fall-out radionuclides can be used to produce chronological markers in truncated sequences from peatlands that have been de-watered after deposition. For example, by using ^{241}Am , ^{137}Cs and $^{14}\text{C}_{\text{bomb-pulse}}$, it is possible to locate peat horizons accumulated at the time of the maximum weapons test fall-out during the 1960s in a sequence that contains a hiatus or a thick tephra layer, or at a site that was drained in the 1990s which would be very difficult to date using the standard ^{210}Pb CRS model.

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