CHRONOLOGIES FOR RECENT PEAT DEPOSITS USING WIGGLE-MATCHED RADIOCARBON AGES: PROBLEMS WITH OLD CARBON CONTAMINATION

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ABSTRACT. Dating sediments which have accumulated over the last few hundred years is critical to the calibration of longer-term paleoclimate records with instrumental climate data. We attempted to use wiggle-matched radiocarbon ages to date 2 peat profiles from northern England which have high-resolution records of paleomoisture variability over the last ~300 yr. A total of 65 ^{14}C accelerator mass spectrometry (AMS) measurements were made on 33 macrofossil samples. A number of the age estimates were older than expected and some of the oldest ages occurred in the upper parts of the sequence, which had been dated to the late 19th and early 20th century using other techniques. We suggest that the older ^{14}C ages are the result of contamination by industrial pollution. Based on counts of spheroidal carbonaceous particles (SCPs), the potential aging effect for SCP carbon was calculated and shown to be appreciable for samples from the early 20th century. Ages corrected for this effect were still too old in some cases, which could be a result of fossil CO_2 fixation, non-SCP particulate carbon, contamination due to imperfect cleaning of samples, or the "reservoir effect" from fixation of fossil carbon emanating from deeper peat layers. Wiggle matches based on the overall shape of the depth-^{14}C relationship and the ^{14}C minima in the calibration curve could still be identified. These were tested against other age estimates (^{210}Pb, pollen, and SCPs) to provide new age-depth models for the profiles. New approaches are needed to measure the impact of industrially derived carbon on recent sediment ages to provide more secure chronologies over the last few hundred years.

INTRODUCTION

There is a growing interest in documenting environmental and climatic changes over recent time periods, especially the last few hundred years. This period is important for several reasons. First, it is the time in which instrumental data overlap paleoenvironmental data, and thus there is an opportunity to test and calibrate paleo-records against instrumentally measured time series. Second, it is a period in which human impacts on climate and environment have been most intense and have accelerated most rapidly. A key issue in establishing paleoenvironmental records which can be matched securely with instrumental data is to develop chronologies that are sufficiently accurate and precise over the last few hundred years. In annually resolved records such as tree rings, the problem does not arise, but in non-annually resolved records such as lake sediments and peat, this is still a major limitation.

In peats, a number of possible techniques for dating are available. Short-lived radioisotopes (^{210}Pb, ^{241}Am, ^{137}Cs) are the most commonly used (Appleby et al. 1997), but this approach has shown variable results in some situations and often needs to be cross-checked with other chronological markers (Oldfield et al. 1995). Stable isotopes of lead have also been used successfully, especially to provide age markers in the 20th century (Martínez-Cortizas et al. 1999). Other useful age markers are spheroidal carbonaceous particles (SCPs; Yang et al. 2001), historical tephras (Schoning et al. 2005), and pollen markers (Barber et al. 1998). Used together, these techniques can provide a high-quality chronology for the last 100–150 yr. However, the quality of chronologies tends to decline for the early 20th and late 19th century, and dating is reliant on only a few markers before AD 1850. In addition, radiocarbon dating of single samples is of limited value for sediments from between AD 1650 and 1950 because of the plateau in the relationship between ^{14}C activity and calendar age over this period. One approach to extend peat chronologies back to around AD 1600, as well as to provide a cross-check on other techniques from AD 1850–1950, is to apply wiggle matching to a

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series of ^{14}C ages. This technique has been used with success in older peats (Kilian et al. 2000; Kilian et al. 1995; Mauquoy et al. 2002; Speranza et al. 2000), but has been attempted rather infrequently for more recent peats (Clymo et al. 1990; Gedye 1998).

In this paper, we report the results from an attempt at using wiggle-matched ^{14}C ages on 2 short peat profiles from northern England. In particular, we explore the problems associated with obtaining accurate assessments of ^{14}C age from such profiles in the light of results which suggest significant contamination from older carbon.

METHODS AND DATING RATIONALE

Two profiles were sampled from Butterburn Flow (55°5′N 2°30′W), a large raised mire in northern England, using a large volume sampler (Wardenaar 1987). The cores (BFA and BFB) were taken from the central dome of the northern section (intersection of the transects in Hendon et al. 2001) from separate *Sphagnum magellanicum* lawns approximately 10 m apart. The profiles were the subject of an investigation into the relationships between high-resolution paleohydrological reconstructions, land use, and climate (Hendon and Charman 2004; Charman et al. 2004). These records probably span the last 300–400 yr of peat accumulation. Although short-lived radioisotopes (^{210}Pb, [241]Am), pollen, and SCP markers have provided estimated chronologies for the period AD 1800 to present, the accuracy of the dating declined in the earlier part of the record and there were no estimated ages from before AD 1800. The aim of the analyses reported here was to establish a wiggle-matched ^{14}C chronology to provide a cross-check with age estimates in the 19th and early 20th century, as well as to extend the chronology back in time.

Samples from 0.5-cm-thick slices of the monoliths were washed through a 125-μm mesh using distilled water. The residue was examined under a dissecting microscope (10× to 50×) in sterile petri dishes, and subsamples of *Sphagnum* leaves and stems were picked and placed in a separate petri dish of distilled water. Where sufficient quantities of *Sphagnum* were not available, leaves of *Erica tetralix* and *Calluna vulgaris* were picked either to supplement *Sphagnum* remains or to provide additional replicate samples. Visible roots and other extraneous organic material were removed using fine forceps, and the subsamples were washed through distilled water again. Samples were sent to the ^{14}C laboratory in distilled water in polypropylene vials. In the ^{14}C laboratory, the samples were subjected to a standard pretreatment (acid wash), dried, and homogenized. The total carbon in the pretreated sample was recovered as CO_2 by heating (900 °C) with CuO in a sealed quartz tube. The gas was converted to graphite by Fe/Zn reduction and analyzed for ^{14}C by the University of Arizona NSF-AMS facility. To achieve higher ^{14}C precision, samples with sufficient CO_2 were split into sub-samples after CO_2 production and up to 3 replicate graphite targets were prepared and analyzed for [14]C.

RESULTS AND DISCUSSION

Radiocarbon Ages

Adequate material for the preparation of 3 replicate graphite targets was available from 12 of the samples and 7 of the samples had adequate material for 2 replicate targets. A single age was obtained for the remaining samples (Table 1). Replicates from each level had overlapping error terms at 2 σ, except 1 replicate from BFB 36–36.5 cm depth (AA-49801) which we rejected from further analysis.

Table 1 Sample characteristics and results of ^{14}C analyses. Key to sample composition:
A = *Sphagnum* leaves and stems, B = *Erica tetralix* and *Calluna vulgaris* leaves.

Publication code	Identifier	Mid-point (cm)	Replicate	Composition	^{14}C age (yr BP)	±1 σ (yr)	$\delta^{13}C_{PDB}$ (%)
BFA							
AA-49754	BFA 17.5–18.0 cm	17.75	1	A	304	58	−25.5
AA-49755			2	A	181	46	
AA-49756			3	A	326	49	
AA-49757	BFA 20.0–20.5 cm	20.25	1	A	146	44	−25.5[a]
AA-49758			2	A	150	46	
AA-49759			3	A	76	47	
AA-49760	BFA 21.5–22.0 cm	21.75	1	A	457	51	−25.6
AA-49761			2	A	487	45	
AA-49762			3	A	470	49	
AA-49763	BFA 24.5–25.0 cm	24.75	1	A	346	66	−26.5
AA-49764			2	A	264	43	
AA-49765			3	A	135	55	
AA-49766	BFA 29.0–29.5 cm	29.25	1	A	414	41	−25.2
AA-49767			2	A	313	39	
AA-49804	BFA 33.0–33.5 cm	33.25	1	A	447	38	−25.5[a]
AA-49768	BFA 34.5–35.0 cm	34.75	1	A, B	262	40	−26.0[a]
AA-49769			2	A, B	138	38	
AA-49770			3	A, B	195	40	
AA-49771	BFA 37.0–37.5 cm	37.25	1	A, B	225	44	−28.8
AA-49772			2	A, B	320	53	
AA-49773			3	A, B	154	42	
AA-49805	BFA 38.0–38.5 cm	38.25	1	A, B	186	39	−26.7
AA-49774	BFA 39.5–40 cm	39.75	1	A, B	253	40	−26.8[a]
AA-49775			2	A, B	315	39	
AA-39729	BFA 41.0–41.5 cm	41.25	1	A	324	42	−26.0[a]
AA-49776	BFA 44.5–45.0 cm	44.75	1	A	379	70	−27.0
AA-49777			2	A	384	43	
AA-49778	BFA 46.0–46.5 cm	46.25	1	A	450	40	−27.4
AA-49779			2	A	334	38	
AA-49780	BFA 47.0–47.5 cm	47.25	1	A	332	39	−26.9
AA-49781			2	A	269	39	
AA-49782			3	A	359	42	
AA-49783	BFA 49.0–49.5 cm	49.25	1	A	428	30	−26.8
AA-49784			2	A	379	41	
AA-49785			3	A	389	30	
AA-39730	BFA 52.0–52.5 cm	52.25	1	A	333	46	−26.2
AA-39731	BFA 66.5–67.0 cm	66.75	1	A	300	140	−26.0[a]
BFB							
AA-49806	BFB 18.0–18.5 cm	18.25	1	A	1272	41	−26.7
AA-49807	BFB 20.5–21.0 cm	20.75	1	A	870	46	−26.6
AA-49786	BFB 21.5–22.0 cm	21.75	1	A	784	31	−26.7
AA-49787			2	A	818	43	
AA-49788	BFB 23.5–24.0 cm	23.75	1	A	487	32	−27.0
AA-49789			2	A	502	44	
AA-49790	BFB 26.5–27.0 cm	26.75	1	A	318	30	−26.8
AA-49791			2	A	301	39	
AA-49792			3	A	517	31	

Table 1 Sample characteristics and results of ^{14}C analyses. Key to sample composition:
A = *Sphagnum* leaves and stems, B = *Erica tetralix* and *Calluna vulgaris* leaves. *(Continued)*

Publication code	Identifier	Mid-point (cm)	Replicate	Composition	^{14}C age (yr BP)	±1 σ (yr)	$\delta^{13}C_{PDB}$ (%)
AA-49793	BFB 28.5–29.0 cm	28.75	1	A	343	30	−27.5
AA-49794			2	A	326	44	
AA-49795			3	A	413	30	
AA-49808	BFB 29.5–30.0 cm	29.75	1	A	559	41	−26.8
AA-49809	BFB 31.0–31.5 cm	31.25	1	A	552	44	−27.1
AA-49796	BFB 31.5–32.0 cm	31.75	1	A	551	31	−26.8
AA-49797			2	A	558	43	
AA-39732	BFB 32.5–33.0 cm	32.75	1	A	213	52	−26.0
AA-49798	BFB 34.0–34.5 cm	34.25	1	A	296	29	−26.7
AA-49799			2	A	527	49	
AA-49800			3	A	308	29	
AA-49801	BFB 36.0–36.5 cm	36.25	1	A, B	891	47	−26.6
AA-49802			2	A, B	239	40	
AA-49803			3	A, B	295	29	
AA-49810	BFB 36.5–37.0 cm	36.75	1	A	541	47	−26.3[a]
AA-49811	BFB 36.5–37.0 cm		2	A, B	420	43	−26.3[a]
AA-39733	BFB 37.5–38.0 cm	37.75	1	A	315	54	−26.5
AA-49812	BFB 39.5–40.0 cm	39.75	1	A	485	66	−26.3[a]
AA-39734	BFB 43.0–43.5 cm	43.25	1	A	377	44	−26.1

[a]Estimated δ^{13}C value due to insufficient sample material for an independent measurement.

The replicate age estimates were combined to produce single values using OxCal 3.5 (Bronk Ramsey 2000). The ages range from 126 ± 26 BP to 473 ± 28 BP for BFA and from 213 ± 52 BP to 1272 ± 41 BP for BFB. These ages compare with an expected range of 79 BP to 365 BP in IntCal98 for the period cal AD 1505 to cal AD 1945 (Stuiver et al. 1998). Many of the ages are clearly much older than expected. In particular, some of the oldest ages occur at the top of the profiles, especially at the top of core BFB where the ages above 24 cm depth are all >490 yr, at least 300 yr older than expected. There are a number of possible reasons for these older ages, including a reservoir effect (Kilian et al. 1995) and contamination during the preparation process (although quality assurance standards processed concurrently with the samples suggested contamination at this stage had not occurred). However, the fact that the most recent peats show a greater deviation from the expected ages points to a factor that has changed over time.

The Influence of Industrial Emissions

One possible explanation for the apparently older ages obtained for the more recent peats is that they have been affected by inputs of older carbon from industrial emissions. These inputs would include the fixation of carbon in CO_2 derived from coal burning as well as the fall-out of particulates. The latter would include SCPs, which we have already established are present in the cores (Hendon and Charman 2004). While these may have been removed by washing during the preparation process, it is well known that these particles have a strong affinity for *Sphagnum* leaves, even being retained within the pores on the cells (Punning and Alliksaar 1997). Therefore, we made estimates of the potential influence of contamination by SCPs on the peat ages (Table 2) using the following mass balance equation applied to the counts of SCP particles made previously (Hendon and Charman 2004):

$$Au = (Mt \times At - Mc \times Ac) / (Mt - Mc)$$

where Au = activity of sample corrected for contamination, Mt = carbon mass in total sample, At = measured activity of total sample, Mc = carbon mass of SCP contaminant, and Ac = activity of SCP contaminant.

Table 2 Estimated sample ages from combining replicates and the possible influence of SCP carbon on ages. The column "SCP effect" is the total aging effect of SCPs in the sample (see text for details).

Material ID code	Mid-point (cm)	^{14}C age (yr BP)	^{14}C age 1 σ (yr)	Sample carbon (mg)	SCP mass (mg)	SCP effect (yr)	Adjusted age (yr BP)
BFA							
BFA 17.5–18.0 cm	17.75	264	29	4.706	0.01562	27	237
BFA 20.0–20.5 cm	20.25	126	26	2.727	0.01562	46	80
BFA 21.5–22.0 cm	21.75	473	28	2.995	0.01562	42	431
BFA 24.5–25.0 cm	24.75	243	30	2.567	0.01562	49	194
BFA 29.0–29.5 cm	29.25	361	28	1.647	0.00731	36	325
BFA 33.0–33.5 cm	33.25	447	38	0.556	0.00121	17	430
BFA 34.5–35.0 cm	34.75	197	23	2.176	0.00121	4	193
BFA 37.0–37.5 cm	37.25	221	26	8.112	0.00000	N/A	221
BFA 38.0–38.5 cm	38.25	186	39	1.797	0.00000	N/A	186
BFA 39.5–40.0 cm	39.75	285	28	2.348	0.00000	N/A	285
BFA 41.0–41.5 cm	41.25	324	42	0.695	0.00000	N/A	324
BFA 44.5–45.0 cm	44.75	383	37	2.225	0.00000	N/A	383
BFA 46.0–46.5 cm	46.25	390	28	3.369	0.00000	N/A	390
BFA 47.0–47.5 cm	47.25	318	23	3.679	0.00000	N/A	318
BFA 49.0–49.5 cm	49.25	402	19	4.316	0.00000	N/A	402
BFA 52.0–52.5 cm	52.25	333	46	1.754	0.00000	N/A	333
BFA 66.5–67.0 cm	66.75	300	140	0.348	0.00000	N/A	300
BFB							
BFB 18.0–18.5 cm	18.25	1272	41	1.481	0.12378	701	571
BFB 20.5–21.0 cm	20.75	870	46	1.658	0.05332	263	607
BFB 21.5–22.0 cm	21.75	796	25	1.995	0.03468	141	655
BFB 23.5–24.0 cm	23.75	492	26	1.834	0.00993	44	448
BFB 26.5–27.0 cm	26.75	390	19	6.845	0.00993	12	378
BFB 28.5–29.0 cm	28.75	368	19	3.834	0.00993	21	347
BFB 29.5–30.0 cm	29.75	559	41	1.112	0.00993	72	487
BFB 31.0–31.5 cm	31.25	552	44	1.572	0.00993	51	502
BFB 31.5–32.0 cm	31.75	552	25	2.096	0.00993	38	513
BFB 32.5–33.0 cm	32.75	213	52	1.428	0.00993	56	157
BFB 34.0–34.5 cm	34.25	337	19	3.957	0.00993	20	317
BFB 36.0–36.5 cm	36.25	276	23	3.118	0.00192	5	271
BFB 36.5–37.0 cm	36.75	476	32	0.497	0.00192	31	445
BFB 37.5–38.0 cm	37.75	315	54	0.738	0.00192	21	294
BFB 39.5–40.0 cm	39.75	485	66	0.749	0.00192	21	464
BFB 43.0–43.5 cm	43.25	377	44	1.257	0.00000	N/A	377

In addition, we have assumed the following:

1. The concentration of SCPs in the samples counted for pollen/SCP is the same as that in the samples prepared for dating;
2. The mean mass of an SCP is 4.2×10^{-9} g (Rose 2001);
3. The *Sphagnum* samples retain all the SCPs present in the original sample after processing;
4. The SCPs are 100% dead carbon.

The potential influence of SCPs on ^{14}C age is relatively small for most of the profiles, except in the top part of BFB where including the effects of SCPs results in an adjustment of up to 700 yr for the topmost sample (Table 2). However, the ages of these upper samples and the ages for samples with relatively low or zero counts of SCPs are still too old by several hundred years when compared with likely age ranges from IntCal98 (Figure 1). This suggests that either the estimate of the level of contamination from industrial sources is too low or that there is an additional source of older carbon.
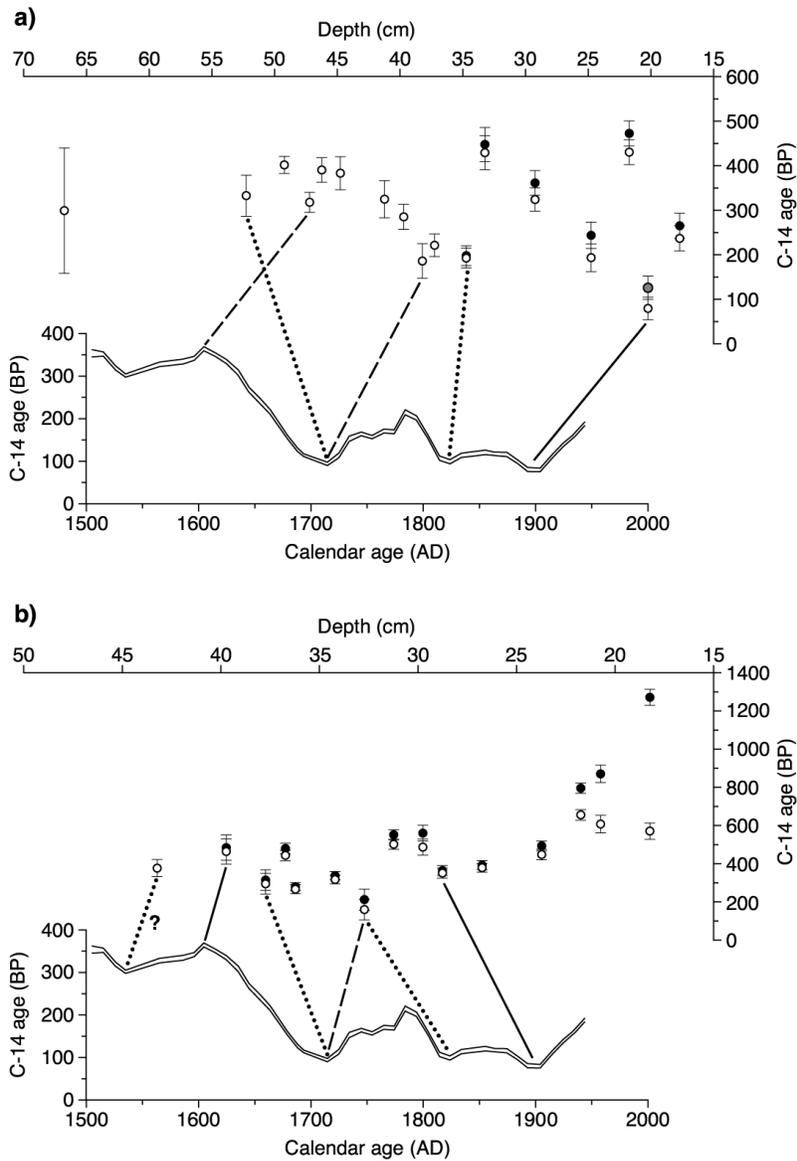
Figure 1 Possible wiggle matches of a) core BFA and b) core BFB with the IntCal98 calibration curve (Stuiver et al. 1998), shown below each peat profile. Closed symbols show original ^{14}C age estimates and open symbols show adjusted ages after taking into account the possible effect of SCP contamination. Dashed lines represent wiggle match 1 and dotted lines represent wiggle match 2. Solid lines represent both wiggle match 1 and 2. Error bars are ±1 σ.

Additional sources of older carbon in the samples could include non-SCP particulate carbon or the fixation of CO_2 containing older carbon. The latter process may be less likely than the former because CO_2 is a well-mixed gas in the atmosphere. However, Levin and Hesshaimer (2000) have shown that fossil fuel-derived CO_2 frequently depletes the atmospheric $^{14}CO_2$ signal at their Heidelberg site by up to 5 pMC, which, had it occurred in early 20th century samples, would age them by about 400 yr. Also, ^{13}C analyses of urban and rural grasses by Lichtfouse et al. (2003) suggested that fossil fuel-derived carbon can contribute even more to urban plant tissues, with up to 29.1% of plant tissue carbon being fossil fuel-derived. Our study site is relatively close to the areas that were some of the largest industrial sources of CO_2 in the late 19th and early 20th century (Manchester/Leeds/Sheffield ~150 km to the south and the Tyne-Tees areas 70–100 km to the east) and, although overall rates of fossil fuel CO_2 emissions were lower then, it is conceivable that there was significant local depletion of atmospheric $^{14}CO_2$ at this time. The $\delta^{13}C$ values do not help in determining the source of carbon (Table 1), as there is no relationship between $\delta^{13}C$ and the calculated amount of SCP contamination. The $\delta^{13}C$ of SCPs is likely to be similar to that of peat, as it is derived from coal and oil and therefore primarily of plant origin.

Other Influences on ^{14}C Ages

There have been few other attempts to use wiggle-matched ages over this time period, but those that have been carried out also show an offset between measured ages and those expected. The data of Clymo et al. (1990) and Gedye (1998) both show offsets of ~125 yr on samples covering this period. Clymo et al. (1990) do not discuss the offset, but Gedye (1998) attributes it to the reservoir effect, which was proposed by Kilian et al. (1995). This suggestion arose from dating older late-Holocene peat, which showed systematic offsets of ~100–150 yr. The exact mechanisms behind this reservoir effect are unclear, but the main suggestion is that fungi attached to the roots of higher plants fix older C in methane derived from deeper peat. Others have suggested that CO_2 derived from decay of older peat is fixed by *Sphagnum* and higher plants, which may derive up to 20% of their carbon from this source (Jungner et al. 1995). However, measurement of ^{14}C in growing *Sphagnum* has shown activity levels apparently unaffected by this process (Nilsson et al. 2001). In addition, Blaauw (2003) and Blaauw et al. (2004) found that this systematic offset was not present in their late-Holocene samples and attributed this to more intensive cleaning and careful selection of aboveground plant remains only. In particular, thorough cleaning is thought to be important to remove fungal contamination. Although we paid careful attention to cleaning, we may have not removed every single fine root, and since our samples were treated only in distilled water during preparation rather than in KOH (cf. Blaauw 2003), they may not have been cleaned of all the adhering fine organic material.

Wiggle Matching Corrected ^{14}C Ages

Despite the fact that ages of recent peats may be affected by several possible sources of contamination from older carbon, it may still be possible to use the shape of the relationship between ^{14}C age and depth to provide wiggle-match age estimates. Figure 1 shows 2 possible wiggle matches for BFA and BFB. Although many ages are significantly older than expected, the youngest ages may be more reliable than the older ages, where peaks in activity are hard to identify. Low points in ^{14}C activity are present at 2 main points in BFA and at 3 main points in BFB. However, the distortion in the curve from the old carbon contamination makes it difficult to know whether the lower minimum value in each profile relates to the ^{14}C minimum at AD 1825 or AD 1715. We can test these alternative interpretations by comparison with other chronological markers (Figure 2).
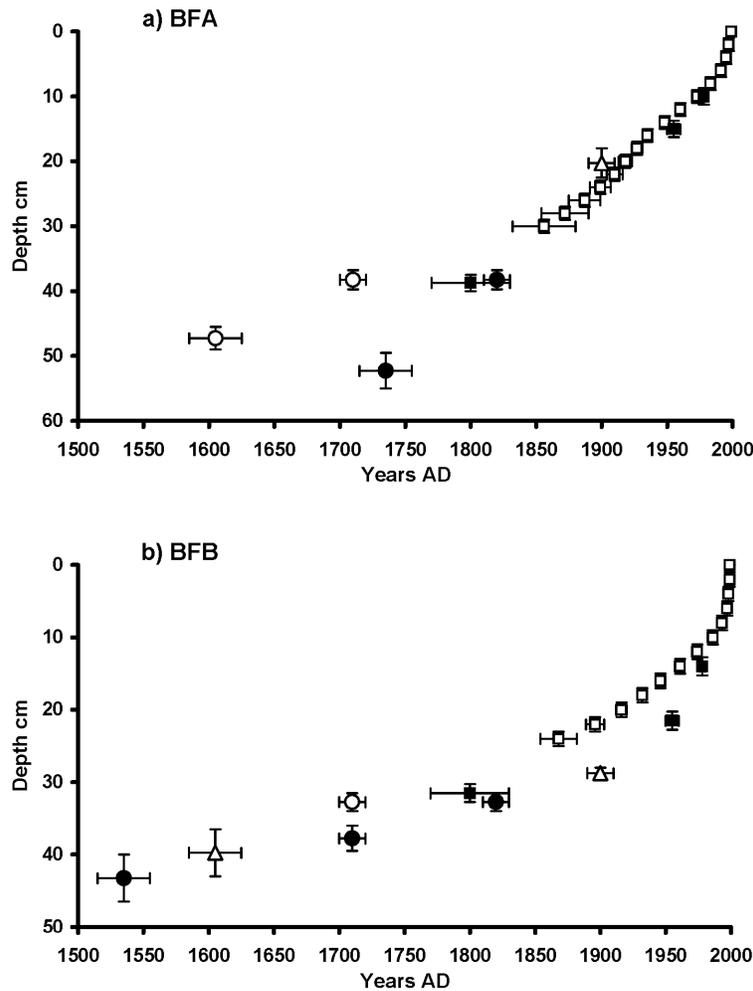
Figure 2 Comparison between age estimates based on ^{14}C wiggle matching, ^{210}Pb, SCP, and pollen markers for a) core BFA and b) core BFB. Open squares = ^{210}Pb ages; closed squares = pollen and SCP ages; open circles = wiggle match 1; closed circles = wiggle match 2; triangles = both wiggle match 1 and 2. Age error bars are 1 σ for ^{14}C age estimates and ^{210}Pb. SCPs and pollen age errors are given according to Rose et al. (1995) and Barber et al. (1998), respectively. Error bars on the depth scale refer to uncertainty arising from sample thickness and distance between sample depths.

For BFA, the age estimates based on the second interpretation of the ^{14}C age-depth relationship are strongly supported by the other age markers. The minimum value at 20.0–20.5 cm overlaps with the 2-σ age ranges of the ^{210}Pb age estimates. The minimum at 38.0–38.5 cm is supported by the pollen marker at AD 1800 (the rise in pine pollen; see Figure 3 and Hendon and Charman 2004). The suggested age at 52.0–52.5 cm—associated with the shoulder of the peak in ^{14}C activity—has no supporting marker but is in line with the extrapolated curve from ^{210}Pb, pollen, and SCP markers. In the case of BFA then, the ages from the wiggle-matched ^{14}C ages agree well with the other age markers, which also have a high degree of internal consistency.
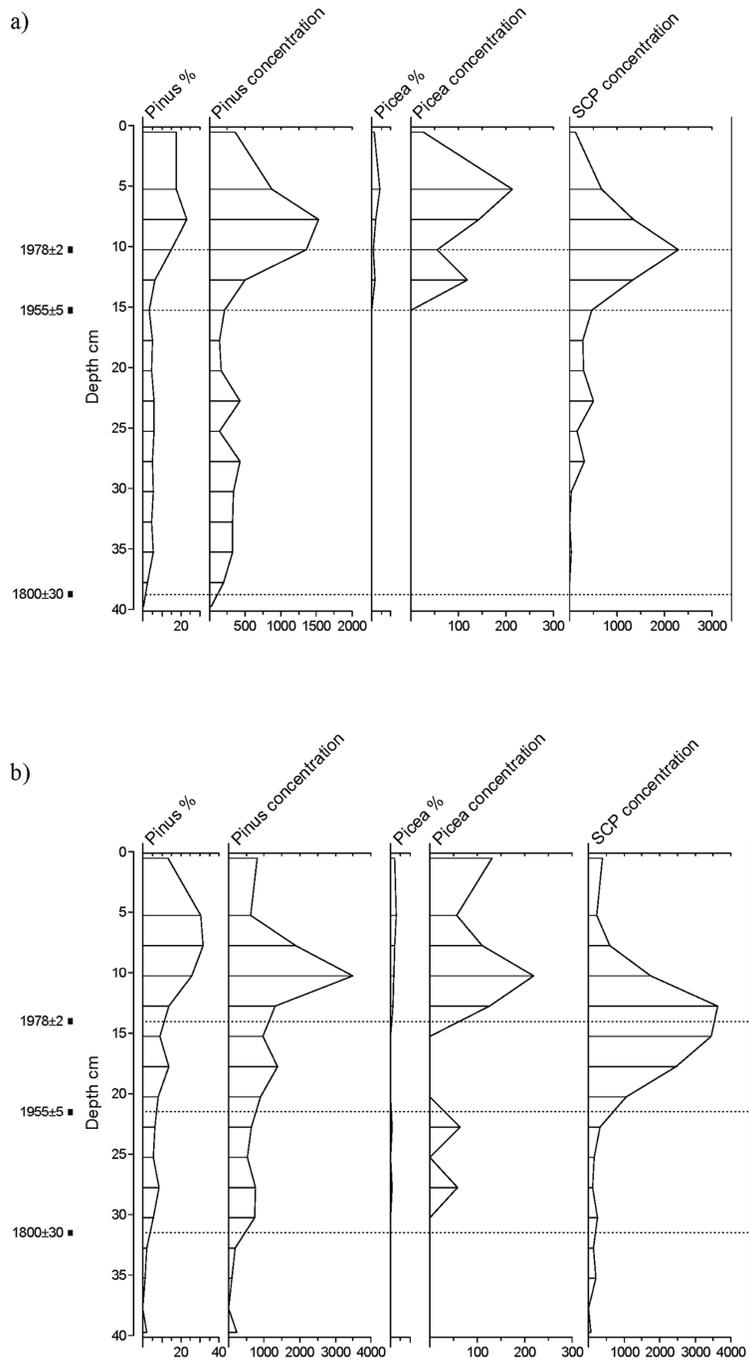
Figure 3 Age estimates based on changes in pine pollen and SCPs for a) core BFA and b) core BFB. The AD 1800 age estimate is based on the initial rise in pine pollen. The 2 SCP markers are based on the rapid increase in SCPs during the 1950s and the peak in SCPs from the late 1970s (Rose et al. 1995). The secondary pine rise reflects the expansion of forest plantations after the 1950s. Pollen % curves expressed as % total land pollen; concentrations expressed as number cm^{-3}.

In BFB, there was a disagreement between the ages derived previously from ^{210}Pb and the age estimates based on SCPs (Figures 2b and 3). The wiggle-matched ^{14}C minimum at 28.5–29.0 cm is much younger than an extrapolation of the ^{210}Pb chronology for an equivalent depth. The minimum at 32.5–33.0 cm is close to the AD 1800 pine pollen rise, which has an estimated error of ±30 yr (Barber et al. 1998). Therefore, it seems unlikely that this minimum represents the AD 1715 minimum in the calibration curve, but is more likely to be AD 1825. The older age estimates based on wiggle matching are not supported by other age estimates. In this case, the new age estimates from wiggle-matched ^{14}C ages shed light on the previously unresolved disagreement between ^{210}Pb and SCP/pollen age estimates. Up until now, we had rejected the age estimates based on SCPs, suspecting some downward movement of SCPs within the profile. However, the wiggle-matched ^{14}C age estimates are more in line with the SCP and pollen ages than they are with the [210]Pb. Given that ^{210}Pb chronologies generally require cross-validation with other techniques (Oldfield et al. 1995), the new results suggest that the ^{210}Pb chronology should be rejected in favor of one based on a combination of the SCP, pollen, and wiggle-matched ^{14}C estimates.

CONCLUSIONS

Wiggle matching ^{14}C ages for the period AD 1600–1950 is an attractive approach to cross-check chronologies based on ^{210}Pb, SCPs, and pollen markers, as well as to extend the chronology over a period that is impossible to date accurately using fewer ^{14}C ages. However, we have shown that the approach is not without problems and that ages older than expected may be obtained. Contamination with SCPs seems to be the most likely cause of the largest aging effects in late 19th- and early 20th-century samples, probably augmented by other industrial carbon contamination. This effect is likely to be particularly acute in regions close to industrial sources, such as northern England. The previously reported "reservoir effect" may also be present in the samples analyzed here, although we have no direct evidence for this. Careful cleaning of samples as recommended by Blaauw (2003) may solve the problem of older carbon contamination to some extent, but other approaches designed to assess the influence of industrial carbon directly are also needed. Differential temperature combustion of samples is one possible approach to separating carbon fixed by plants from industrial particulate carbon.

ACKNOWLEDGMENTS

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