

Available online at www.sciencedirect.com



QUATERNARY RESEARCH

Quaternary Research 68 (2007) 325-337

www.elsevier.com/locate/yqres

The use of 'bomb spike' calibration and high-precision AMS ¹⁴C analyses to date salt-marsh sediments deposited during the past three centuries

William A. Marshall^{a,*}, W. Roland Gehrels^a, Mark H. Garnett^b, Stewart P.H.T. Freeman^c, Colin Maden^c, Sheng Xu^c

^a School of Geography, University of Plymouth, Drakes Circus, Plymouth, PL4 8AA, UK

^b National Environment Research Council Radiocarbon Laboratory, Scottish Enterprise Technology Park, East Kilbride, G75 0QF, UK

^c Scottish Universities Environmental Research Centre, Scottish Enterprise Technology Park, East Kilbride, G75 0QF, UK

Received 17 May 2006 Available online 1 October 2007

Abstract

A combination of 'bomb spike' calibration and conventional calibration of AMS ¹⁴C dating has been used to determine a detailed age-depth model for a 1-m sediment section collected from a salt marsh in Poole Harbour, southern England. These data were compared with the chronology obtained from ²¹⁰Pb analysis and ¹³⁷Cs age markers. We report post bomb values of over 1.46 F¹⁴C (>146% modern ¹⁴C), and both the rising and falling limbs of the atmospheric 'bomb spike' are identified. Five pre-bomb samples were analysed using multi-target high-precision 2‰ AMS analysis, and after the replicates were combined the one-sigma uncertainty was as low as ± 9 ¹⁴C yr on some ages. These data, and an additional three normal-precision pre-bomb ¹⁴C samples, were calibrated using CALIB 5.0 and the chronology constrained using the 'prior knowledge' of independent age markers obtained from the analysis of pollen and spheroidal carbonaceous particle (SCPs). No agreement was found between the ¹⁴C 'bomb spike' dates and the CRS ²¹⁰Pb chronology modelled for this sequence. In addition, poor agreement was found between the signal of the 1960s weapons test fallout indicated by the ¹⁴C 'bomb spike' dates and the timing suggested by the ¹³⁷Cs data. This disagreement is attributed to the influence of the local discharge of ¹³⁷Cs from the former UKAEA site at Winfrith. We use our new chronology to confirm the existence of an acceleration in sedimentation rates in Poole Harbour during the last 100 yr previously reported for this site by Long et al. (Long, A.J., Scaife, R. G., Edwards, R.J. 1999. Pine Pollen in intertidal sediments from Poole Harbour, UK; implications for late-Holocene sediment accretion rates and sea-level rise. Quaternary International, 55, 3–16.), and conclude that 'bomb spike' ¹⁴C calibration dating may offer a more robust alternative to the use of ²¹⁰Pb chronologies for dating sediment deposition in salt-marsh environments. In addition, we demonstrate how the use of high-precision AMS analysis

© 2007 University of Washington. All rights reserved.

Keywords: Radiocarbon dating; sea-level change; Spheroidal carbonaceous particles; Poole Harbour; ²¹⁰Pb

Introduction

Salt-marsh sediments can provide an excellent archive of past sea-level changes (Zong and Horton, 1999; Plater et al., 2000; Gehrels, 2000; Haslett et al., 2001; Gehrels et al., 2005).

* Corresponding author. Fax: +44 1752 233054.

Traditionally, the chronologies used in this type of study have depended heavily on 'conventional' ¹⁴C dating for historic and older age-models, with ²¹⁰Pb chronologies, sometimes supported by ¹³⁷Cs data, used to date events in the 19th and 20th centuries. Unfortunately, both ¹⁴C and ²¹⁰Pb dating suffer from a number of limitations and problems that restrict the applicability of these methods to high-resolution palaeo-environmental studies. The conventional approach to the calibration of ¹⁴C data has some well-documented fundamental complications (Stuiver and Braziunas, 1998; Stuiver et al., 1998; Telford et al., 2004). These issues can produce disproportionately high age

E-mail addresses: wmarshall@plymouth.ac.uk (W.A. Marshall), W.R.Gehrels@plymouth.ac.uk (W.R. Gehrels), M.Garnett@nercrcl.gla.ac.uk (M.H. Garnett), s.freeman@suerc.gla.ac.uk (S.P.H.T. Freeman), c.maden@suerc.gla.ac.uk (C. Maden), s.xu@suerc.gla.ac.uk (S. Xu).

^{0033-5894/\$ -} see front matter @ 2007 University of Washington. All rights reserved. doi:10.1016/j.yqres.2007.07.005

uncertainties when ¹⁴C dating is used to determine the age of material from the historic and modern periods. Lead-210 dating is somewhat restricted in both its application and the time span it can cover. For best results it requires a continuous and steady-state deposition of sediment and, even if a suitable sedimentary environment is extant, ²¹⁰Pb chronologies can only be reliably constructed for the last 100 yr (Smith, 2001). Ceasium-137 is sometimes used to constrain ²¹⁰Pb chronologies (Appleby, 2001), but the dependable use of 137 Cs as a dating 'spike' to constrain ²¹⁰Pb chronologies is increasingly viewed as uncertain due to the post depositional migration of caesium under certain conditions (Morris et al., 2000; Abril, 2004; Harvey et al., 2007). Furthermore the discharge of radionuclides from nuclear facilities, notably Sellafield in the UK and the French plant at Cap de La Hague, can overprint the atmospheric fallout signal found in nearby salt marshes (Morris et al., 2000; Harvey et al., 2007). Of special interest to this study is the former UKAEA research and development site at Winfrith, near Wool in Dorset. This site is within 20 km of our study site, and when it was active, between 1958 and 1995, this facility discharged small amounts of fission products and transuranic radionuclides via an offshore outfall into the English Channel (Thomson et al., 2002).

Carbon-14 bomb spike calibration is an alternative method of calibrating ¹⁴C dates which may offer some potential for constructing robust high-resolution chronologies in sediment sequences that span the historic to present period (Turetsky et al., 2004). The rapid post-1950 increase in atmospheric ¹⁴C, produced by the above-ground testing of nuclear weapons, can be used to date contemporaneous organic sediment deposition (Goodsite et al., 2001). This 'bomb spike' or 'bomb carbon pulse' approach has been applied successfully in recent peat sections in Denmark and Greenland (Shotyk et al., 2003), Ireland (McGee et al., 2004) and northern England (Garnett and Stevenson, 2004). These studies have all been in organic-rich terrestrial sites. As yet there have been no reports of the use of this method in a salt marsh.

We report here the use of the atmospheric bomb carbon record to calibrate post-bomb ¹⁴C dates to establish the deposition timing of sediments in a UK salt-marsh. These dates are combined with pre-bomb conventionally calibrated AMS data to construct an age model covering the last 300 yr for a recent salt-marsh sediment sequence from Poole Harbour in Dorset. This part of southern England was not overridden by ice during the Devension, and mean sea-level rise on this section of the English south coast during the late Holocene is proposed to be around 0.5 mm yr^{-1} (Shennan, 1989; Shennan and Horton, 2002). Salt-marsh sediment accumulation is normally closely coupled to relative sea-level rise (Allen, 2003), but previous sea-level research in Poole Harbour (Long et al., 1999; Edwards, 2001) suggests a more rapid local accumulation rate in the last 100 yr than that which can be attributable to a 0.5 mm yr^{-1} rate of sea-level rise. It is suggested that the introduction and expansion of the hybrid salt-marsh grass Spartina anglica (Cord Grass) since 1890 has increased sedimentation rates in Poole Harbour from that time and disrupted the direct coupling of marsh accretion with relative sea-level rise (Long et al.,

1999). However, these previous studies did not have a highresolution chronology for the 19th and 20th centuries so the exact timing of this change in accumulation rates is not well constrained. Therefore, we establish a robust ¹⁴C deposition chronology for the last 300 yr to confirm this timing, and then use this information to test the reliability of the ²¹⁰Pb and ¹³⁷Cs methods of dating recent sediments at this site.

Our ¹⁴C data were produced by a mixture of normal (3‰) and high (2‰) precision analysis and results obtained using these two approaches are compared. We used the constraint of the prior knowledge of stratigraphy, and two independent chronological markers, pollen and spheroidal carbonaceous particles (SCPs), to refine the calibration of our ¹⁴C dates. and reduce the final uncertainty in our chronology. Both pollen and SCPs are useful as independent chronological markers in this part of southern England. The appearance of ragweed (Ambrosia) pollen has been successfully used in western North Atlantic sea-level studies (Gehrels et al., 2002; Gehrels et al., 2005) as an age marker, and Long et al. (1999) used Pinus and Spartina anglica pollen to constrain the history of sea-level change in Poole Harbour during the 18th and 19th centuries. Long et al. (1999) attributed a local increase in Pinus pollen to the regional expansion of pine plantations between the mid 1700s and the mid 1800s, and used the signal of the documented expansion of Spartina anglica around 1895 to identify sediments deposited contemporaneous to this event. SCPs are formed from the incomplete combustion of fossil fuels at high-temperature, and can be used to signal the timing of sediment deposition during specific periods of the 19th and 20th centuries (Jones et al., 1997; Rose et al., 1999; Rose, 2001; Yang et al., 2001). Rose and Appleby (2005) have identified, and dated, a number of changes in the UK regional records of SCP deposition that can be used as age markers.

Site and methods

The salt-marsh site used in this study is on the Arne Peninsula, which is part of the western shore of the enclosed embayment of Poole Harbour located on the English Channel coast (Fig. 1). Poole Harbour has a mean tidal range of 1.5 m at spring tide and 0.5 m at neap tide. The mean high water spring tide height is 0.6 m on the Arne Peninsula. The local geology is carbonate-rich, poorly consolidated, Tertiary sediment of the Bracklesham Group (Bristow et al., 1991), with a scattered cover of Pleistocene fluvial gravels and sands. Spartina anglica is thought to have emerged around the middle of the 18th century in Southampton Water, but the first confirmed formal identification of this species was in 1892 near Lymington in Hampshire (Thompson et al., 1991). Since then this invasive plant had spread south along the English Channel coast, and its first significant expansion was recorded in Poole Harbour between 1890 and 1900 (Long et al., 1999).

An undisturbed 1-m sediment monolith was cut from the side of a pit dug in the highest area of a salt-marsh on the Arne Peninsula (Fig. 1). The surface elevation of the pit was +0.67 m



Figure 1. The Arne Peninsula salt marsh in Poole Harbour showing the location of the sample site.

OD (UK Ordnance Datum) and the site is some 50 m southwest (i.e. more terrestrial) from the location that provided the sediment core analysed by Long et al. (1999). Only the top 76 cm was sampled for ¹⁴C dating. The sediments in this upper section are mainly organic-rich clay, with decreasing organic content from top to bottom (Fig. 2). Below 76 cm the sediments comprised of silts and clay, with coarse flint gravels at the base, and this lower section did not contain any organic material suitable for ¹⁴C dating.

When sampling for the ¹⁴C analysis we targeted only the detrital remains of above-ground parts of salt-marsh grasses. Although the sediments above 55 cm contained many plant fragments, we were very selective and only those fragments, lying horizontal in the clayey matrix were picked out. This minimised the chance of younger root material being selected, and the fragility of these grass fragments ensured that the possibility of transportation and re-deposition was minimal. In addition, only samples that were totally contained in the central section of the monolith and did not extend towards the edge of the monolith were used. The selected fragments of grass stem were isolated from the sediments, carefully washed in deionised water and checked for visible contamination of fine roots under a binocular microscope. A suitable grass stem sample could not be found between 16 cm and 25 cm and a small fragment of a fibrous woody plant stem, Arne-03-20 (SUERC-5234), was used instead. At the NERC Radiocarbon Laboratory the samples were subjected to an acid wash (2 M HCl), combusted

in sealed quartz tubes and the sample CO₂ cryogenically recovered and converted to graphite. The ¹⁴C content was measured at the Scottish Universities Environmental Research Centre (SUERC) AMS Facility. Five samples from the deeper levels, expected to be >100 yr old, were prepared as multiple graphite targets (Table 1) and ran at 2‰ precision instead of the 3‰ normally used for ¹⁴C samples at the SUERC AMS facility. The upper 50 cm of the monolith was sub-sampled at 1-cm intervals for radionuclide analysis. This material was sent to the Liverpool University Environmental Radioactivity Laboratory and the activity levels for ²¹⁰Pb, ²²⁶Ra, ¹³⁷Cs and ²⁴¹Am determined using coaxial low background intrinsic germanium detectors (Appleby et al., 1986).

The upper sediments were sampled for pollen and spheroidal carbonaceous particles (SCPs). Pollen preparation and analysis followed the standard procedure of Moore et al. (1991). SCPs were counted concurrently with the pollen analysis, and the exotic marker (*Lycopodium clavatum*) method was used to standardise the results to sediment volume (Moore et al., 1991).

Results

Radionuclides

Contiguous samples were taken at 0.5 cm intervals. The total ²¹⁰Pb activity in the Arne sediments reaches equilibrium with that of the supporting ²²⁶Ra at a depth of about 35 cm. The



Figure 2. The Arne sediment lithology with the ²¹⁰Pb (a), ¹³⁷Cs (a), ²⁴¹Am (c), SCP (d and e) and *Spartina* pollen (f) results. SCPs shown per cm³ and as an absolute ratio of TLP (Total Land Pollen). *Spartina* shown as % of the TLP sum.

unsupported ²¹⁰Pb activity curve (Fig. 2a), calculated by subtracting ²²⁶Ra activity from total ²¹⁰Pb activity, has an erratic profile. The concentration rapidly increases down profile, and the maximum values occur in the sediments around 4–5 cm deep. Below this depth, the overall ²¹⁰Pb activity trend is of exponential decline, apart from a small peak in the sediment at 24–27 cm. The Arne¹³⁷Cs profile (Fig. 2b) has a well resolved major peak of 78.7 Bq kg⁻¹ at 20.5 cm with a lesser peak of 38.3 Bq kg⁻¹ at 4.5 cm. Between 12 cm and 23 cm there is some ²⁴¹Am activity (Fig. 2c) detected which has its maximum value of 1.9 Bq kg⁻¹ at 22.5 cm.

The unevenness of the ²¹⁰Pb record prevents the use of the Constant Initial Concentration (CIC) model, and therefore a chronology (Fig. 5) was constructed using the Constant Rate of Supply (CRS) model (Appleby and Oldfield, 1978). This model was devised to try and compensate for changes in the dry sediment supply and it is also known as the Constant Flux (CF) model (Robbins, 1978). In this model it is assumed that the ²¹⁰Pb flux has remained constant through time, but the sediment supply is allowed to change. When the unsupported ²¹⁰Pb record is integrated to a specific depth, or dry mass, it will equal the ²¹⁰Pb flux integrated over the time, in years, elapsed since the deposition of those sediments (See Appleby, 2001).

The three 137 Cs age markers commonly used to date sediments are (1) the first significant environmental presence of this radionuclide around 1950–1955, (2) the 1963 weapons peak and (3) the fallout from the 1986 Chernobyl reactor incident. In the first instance we interpret these three signals in the Arne 137 Cs record as being present in the sediments from 28 cm (1), 20.5 cm (2) and 4.5 cm (3). Americium-241 activity

is known to be associated with the signal of the maximum radionuclides fallout in the early 1960s from atmospheric nuclear weapon detonations (Appleby, 2001). It was not present in significant amounts in either the initial fallout from the nuclear weapons tests of the 1950s and 1960s, or the 1986 Chernobyl accident. However, plutonium isotopes did form a significant part of these fallouts (Krey et al., 1976; Appleby et al., 1991; UNSCEAR, 2000), and the majority of the ²⁴¹Am activity in present-day sediment archives is the result of the gamma decay of ²⁴¹Pu, which has a half-life of 14.4 yr (Thomson et al., 2002; Pourchet et al., 2003).

However, in Poole Harbour we know that there have been nearby discharges of ¹³⁷Cs and transuranic radionuclides between 1958 and 1995. The known ¹³⁷Cs emissions from Winfrith peaked at around 2000 GBq in 1980, but the alphaemitting radionuclides in the effluent were not differentiated so it is not known if ²⁴¹Am or ²⁴¹Pu was present. It is only known that discharge of alpha-emitting radionuclides peaked at around 0.005 TBq in the period between 1972 and 1976 (Thomson et al., 2002). Therefore, it is possible that between 1958 and 1995 the discharge from Winfrith may have contributed to the 137 Cs and 241 Am inventory in the Arne sediments. Thomson et al. (2002) found low level ²⁴¹Am activity in salt-marsh sediments from Beaulieu Marsh, located approximately 65 km east of Winfrith. The ²⁴¹Am signal here appeared to agree broadly with the known history of alpha radiation in discharges from the former UKAEA site, but the deposition chronology suggested by the ¹³⁷Cs data from the Beaulieu Marsh section was not in agreement with the known discharge of this radionuclide from Winfrith (Thomson et al., 2002).

Table 1 The results for the high precision AMS Arne samples

| Publication no. | Sample identifier | Index no. | Depth (cm) | Sample dry wt. (g) | $F^{14}C\!\pm\!1\sigma$ | $ \delta^{13}C_{V\text{-PDB}} \\ \%_0$ | ¹⁴ C yr BP±1σ | Combined ${}^{14}C$ yr BP±1 σ | 2σ age ranges AD | CPRAV |
|-----------------|-------------------|--------------|---------------|-----------------------|-------------------------|--|-----------------------------|--------------------------------------|---------------------|-------|
| SUERC-6364 | ARN-03-34 X | | | | $0.9809 \!\pm\! 0.0018$ | -29.5 | 155 ± 15 | | 1677 to 1694 | 0.17 |
| | | | | | | | | | 1726 to 1767 | 0.37 |
| | | | | | | | | | 1772 to 1777 | 0.01 |
| | | | | | | | | | 1799 to 1813 | 0.15 |
| SUERC-6365 | ARN-03-34 Y | 2 | 34 | 0.0450 | $0.9819 \!\pm\! 0.0018$ | -29.5 | 146 ± 15 | 146 ± 9 | 1838 to 1842 | 0.01 |
| | | | | | | | | | 1853 to 1858 | 0.01 |
| | | | | | | | | | 1862 to 1867 | 0.01 |
| SUERC-6366 | ARN-03-34 Z | | | | $0.9831 \!\pm\! 0.0018$ | -29.5 | 137 ± 15 | | 1918 to 1940 | 0.26 |
| | | | | | | | | | 1950 to 1952 | 0.01 |
| SUERC-6369 | ARN-03-42 X | | | | $0.9851 \!\pm\! 0.0018$ | -28.2 | 120 ± 15 | | | |
| | | | | | | | | | 1682 to 1699 | 0.18 |
| | | | | | | | | | 1721 to 1737 | 0.12 |
| SUERC-6370 | ARN-03-42 Y | 3 | 42 | 0.0239 | $0.9843 \!\pm\! 0.0018$ | -28.2 | $127\!\pm\!15$ | 131 ± 9 | 1758 to 1761 | 0.00 |
| | | | | | | | | | 1803 to 1818 | 0.11 |
| SUERC-6371 | ARN-03-42 Z | | | | $0.9890 \!\pm\! 0.0018$ | -28.2 | $147\!\pm\!15$ | | 1833 to 1880 | 0.39 |
| SUERC-6374 | ARN-03-47.5 X | | | | $0.9785 \!\pm\! 0.0018$ | -29.0 | 174±15 | | 1667 to 1683 | 0.20 |
| | | | | | | | | | 1735 to 1782 | 0.54 |
| | | 4 | 47.5 | 0.0980 | | | | 175 ± 11 | | |
| SUERC-6375 | ARN-03-47.5 Y | | | | $0.9785 \!\pm\! 0.0018$ | -29.0 | 175 ± 15 | | 1797 to 1805 | 0.09 |
| | | | | | | | | | 1932 to 1952 | 0.17 |
| SUERC-6376 | ARN-03-55 X | | | | 0.9898 ± 0.0018 | -28.7 | 83 ± 15 | | | |
| | | | | | | | | | 1697 to 1726 | 0.33 |
| | | | | | | | | | 1814 to 1836 | 0.23 |
| SUERC-6377 | ARN-03-55 Y | 5 | 55 | 0.0235 | $0.9864 \!\pm\! 0.0018$ | -28.7 | 110 ± 15 | 92±9 | | |
| | | | | | | | | | 1844 to 1851 | 0.24 |
| SUERC-6380 | ARN-03-55 Z | | | | $0.9898 \!\pm\! 0.0018$ | -28.7 | 82 ± 15 | | 1877 to 1917 | 0.42 |
| SUERC-6381 | ARN-03-62 X | | | | 0.9768 ± 0.0018 | -28.8 | 188 ± 15 | | 1662 to 1677 | 0.25 |
| | | | | | | | | | 1766 to 1772 | 0.03 |
| | | 7 | 62 | 0.0101 | | | | 199 ± 11 | | |
| | | | | | | | | | 1777 to 1800 | 0.48 |
| SUERC-6382 | ARN-03-62 Y | | | | $0.9742 \!\pm\! 0.0018$ | -28.8 | 210 ± 15 | | 1940 to 1951 | 0.24 |

Replicated graphite target results differentiated as X, Y and Z. Radiocarbon content is reported as $F^{14}C$ after Reimer et al. (2004a). CALIB 5.0 age ranges selected are underlined. CPAV is the Calibration Peak Area Value produced by CALIB 5.0.

Pollen and SCPs

Pollen and SCP samples were analysed at 1 cm intervals. No significant numbers of recognisable pollen grains were found below 65 cm. We attribute this to the poor preservation of the pollen grains in the clay-rich sediments found in these lower levels. The Total Land Pollen (TLP) grain count at 64 cm was 21×10^3 per cm³ rising rapidly up profile to 37×10^3 per cm³ at 62 cm, with an average of 38×10^3 per cm³ throughout the sediment section. No significant change was found in the Pinus percentages in the samples that contained significant pollen grains, suggesting that the sediments above 64 cm were most likely deposited after the 18th century Pinus rise described by Long et al. (1999). However, there was a sudden appearance and establishment of Spartina in the pollen diagram (Fig. 2d). Some isolated Spartina grains were found between 46 cm and 41 cm, but this presence rapidly increased to over 1% of the TLP sum at 40-41 cm. We interpret this signal as the wider establishment and rapid expansion of Spartina in the 1890s as described by Long et al. (1999), and therefore use their date of AD 1895 ± 5 yr for this horizon.

The SCP record started at 57 cm, but it was not until 48–50 cm that the count increased to over 1.5×10^3 particles per cm³ (Figs. 2e

and f). Above this level the particle numbers peaked at 41 cm and then declined, before rising abruptly to the major peak of this record of 12.7×10^3 particles per cm³ at 5 cm. However, when the SCP counts were standardised using the TLP deposition, thus reducing the direct influence of local sediment fluxes, a subtly different history was revealed (Fig. 2f). In this curve the post-WW2 rise is more gradual with a later abrupt rise, and the maximum SCP deposition having occurred at 13 cm. This is the depth interpreted as the stratigraphic location of the post-WW2 peak of SCP deposition in England during the 1970s and early 1980s reported by Rose and Appleby (2005). Two of the sedimentary records listed in the Rose and Appleby (2005) study are from sites at Abbotsbury in Dorset and Harbour Farm on the Isle of Wight, and these data confirm that the regional maximum SCP deposition occurred during the late 1970s and early 1980s in this part of southern England (Rose, N. personal communication 2005).

Using the chronology of regional SCP deposition given in Rose and Appleby (2005) three SCP age-marker horizons are identified in the Arne sequence:

(1) 50 cm= 1850 ± 25 yr. The first appearance of SCPs in the sedimentary record.

- (2) 28 cm=1950±15 yr. The late 20th century or post WW2 rise.
- (3) 13 cm=1980±7 yr. The late 20th century maximum deposition peak.

Radiocarbon analyses

In reporting these data we follow the conventions proposed by Reimer et al. (2004a) and present the ¹⁴C data as 'F¹⁴C' and, where appropriate, conventional radiocarbon age (Stuiver and Polach, 1977). The five high-precision samples all had a F¹⁴C one-sigma analytical uncertainty of 0.0018 (Table 1) and the replicates were statistically indistinguishable. CALIB 5.0 (Stuiver et al., 2005) was used to combine the ¹⁴C ages from the multi target results

from the AMS, and they are treated here as if the ${}^{14}C$ data came from one cathode. The resulting calibrated age ranges are shown in Table 1.

The standard precision AMS results are shown in Table 2. On inspecting all the AMS results it was decided that the 1950s transition from pre- to post- bomb $F^{14}C$ values was likely to lie between 26.5 cm and 34 cm. The analysis of the seven samples above 31 cm all produced values over 1.0 $F^{14}C$ suggesting a post-bomb age, with the maximum $F^{14}C$ values being found at 25 cm (1.4609) and 26.5 cm (1.4637). The seven upper $F^{14}C$ results, plus the data from ARN-03-31, were selected for calibration to the bomb carbon record using CALIBomb (Reimer et al., 2004a). The ¹⁴C age produced from analysis of the sample from ARN-03-31 was also calibrated using CALIB 5.0. (Table 2).

Table 2

The standard precision AMS results for the Arne samples

| Publication no. | Sample identifier | Index no. | Depth (cm) | $F^{14}C\!\pm\!1\sigma$ | $\delta^{13}C_{V\text{-PDB}}\ \text{\%}$ | ^{14}C yr BP $\pm 1\sigma$ | 2σ age ranges AD | CPAV |
|-----------------|-------------------|-----------|------------|-------------------------|--|------------------------------|--|--|
| SUERC-6330 | ARN-03-04 | NA | 4 | 1.1262 ± 0.0035 | -29.6 | N/A | 1957.5 to 1958.0 1993.1 to 1995.6 | 0.04 0.96 |
| SUERC-5232 | ARN-03-08 | NA | 8 | 1.1825 ± 0.0038 | -28.8 | N/A | 1958.5 to 1958.8 1986.3 to 1988.8 | 0.08 0.97 |
| SUERC-6331 | ARN-03-11 | NA | 11 | 1.2390±0.0039 | -27.5 | N/A | 1959.2 to 1960.1 1961.4 to1961.8 1981.9 to 1984.0 | 0.09 0.10 0.82 |
| SUERC-5233 | ARN-03-16 | NA | 16 | 1.3363 ± 0.0043 | -27.5 | N/A | 1962.3 to 1962.5 1976.5 to 1978.6 | 0.08 0.92 |
| SUERC-5234 | ARN-03-20 | NA | 20 | $1.0418 {\pm} 0.0035$ | -27.9 | N/A | 1955.5 to 1956.8 | 1.00 |
| SUERC-5235 | ARN-03-25 | NA | 25 | 1.4609 ± 0.0049 | -26.0 | N/A | 1962.9 to 1963.0 1972.4 to 1973.7 | 0.06 0.93 |
| SUERC-5237 | ARN-03-26.5 | NA | 26.5 | 1.4637±0.0049 | -24.7 | N/A | 1962.9 to 1963.0 1971.9 to 1973.5 | $\frac{0.06}{0.93}$ |
| SUERC-5238 | ARN-03-31 | 1 | 31 | 0.9798±0.0031 | -27.0 | 164±25 | CALIBomb 1664.0 to 1667.0 1724.0 to 1788.0 1790.0 to 1815.0 1834.0 to 1878.0 1916.0 to 1951.1 | 0.18 0.43 0.12 0.08 0.20 |
| | | | | | | | CALIB 1665 to 1697 1725 to 1786 1792 to 1815 1835 to 1878 1917 to 1952 | 0.18 0.43 0.11 0.08 0.20 |
| SUERC-5239 | ARN-03-56 | 6 | 56 | 0.9817±0.0031 | -28.8 | 148±26 | 1668 to 1707 1719 to 1782 <u>1797 to 1826</u> 1832 to 1886 1912 to 1953 | $0.17 \\ 0.33 \\ 0.12 \\ 0.19 \\ 0.19$ |
| SUERC-6333 | ARN-03-76 | 8 | 76 | 0.9712±0.0024 | -27.7 | 234±20 | 1643 to 1669 1780 to 1798 1945 to 1951 | |

Radiocarbon content is reported as $F^{14}C$ after Reimer et al. (2004a). CALIBomb (Reimer et al., 2004a) was used to calibrate the upper samples, from 4 cm to 31 cm. CALIB 5.0 was used for the sample from 31 cm. Age ranges selected are underlined. CPAV is Calibration Peak Area Value produced by CALIB 5.0.

Calibration of radiocarbon results

Bomb spike

The eight $F^{14}C$ data were fitted to the post-bomb Northern Hemisphere Zone 1 atmospheric ^{14}C calibration curve (Hua and Barbetti, 2004) using CALIBomb (Reimer et al., 2004a) set to a smoothing of 1 yr with a resolution of 0.2 yr (Fig. 3). The calibration of most of the samples produced two or more possible fits (Table 2), but by considering the stratigraphic order of the samples and the $F^{14}C$ values of the adjacent samples it is possible confidently resolve most of the data to a single solution. For example, the older solutions for the samples from 4–16 cm and 25 cm can all be dismissed because their $F^{14}C$ values and stratigraphic order indicate they all fit on the falling limb of the bomb spike, and therefore all have to be younger than 1963. The resulting fit is shown in Figure 3. The exceptions to this are the samples from 20 cm and 26.5 cm.

The AMS result for the sample from 20 cm is 1.0418 $F^{14}C$. This value has a single calibration solution of 1955.5– 1956.8 (2 sigma), but this date is out of chronological sequence when compared to the adjacent samples, and the result was rejected as unreliable. In contrast, the F14C value of the sample from 26.5 cm has two possible calibrations that can be legitimately fitted to either the rising or falling limbs of the ¹⁴C calibration curve. However, when the next sample above it in the sequence is considered, the sample from 26.5 cm has a higher $F^{14}C$ value than the sample from 25 cm. Therefore, ARN-03-26.5 had to plot isotopically above ARN-03-25, but ARN-03-26.5 can be fitted on the rising or falling limbs of the ¹⁴C calibration curve (Fig. 3). To resolve this the SCP date of 1950±15 yr for 28 cm is used to confirm the older calibration of 1962.9-1963.0 for the sample from 26.5 cm.

Pre bomb

The remaining Arne ¹⁴C results and the data from 31 cm were calibrated in the conventional way using CALIB 5.0 and the Intcal04 calibration curve (Reimer et al., 2004b). Then, by maintaining the inter-sample stratigraphic relationship, and using the concept of chronological 'prior knowledge' of independent age markers, the most likely solutions from the results of the initial calibrations were selected. In the case of the upper three samples, the youngest age solutions are considered valid because the other possible calibrations are older than the AD 1895 ± 5 yr pollen marker at 40 cm. This leaves two options for ARN-03-34, but the solution for ARN-03-31, 1917–1951. constrains the calibration to 1918–1940, not 1950–1952. In the same manner, all the possible age calibrations for ARN-03-47.5 are rejected because they are older than the underlying SCP marker at 50 cm of AD 1850±25 yr, or younger than the overlying sediments.

Below 50 cm all the calibrations younger than the SCP marker at 50 cm can be dismissed, but this still leaves multiple age options for the remaining four samples. After considering the lack the signal of the 18th century expansion of pine plantations reported by Long et al. (1999) in our Pinus pollen data, all the samples above 64 cm are considered to have been deposited after this time. Consequently, all the calibrations for ARN-03-62, ARN-03-56 and ARN-03-55 older than AD 1750 are to be eliminated, but to resolve this chronology further more information is needed. Therefore, the mean long-term accumulation rates indicated by the different calibration options for the remaining samples are examined. These indicate that in the case of the oldest sample, ARN-03-76, if we accepted the younger solution of AD 1780-1798, a mean accumulation rate up to the pollen marker at 40 cm of 3.0-3.9 mm yr⁻¹ is suggested. This is somewhat higher than the $0.5-2.5 \text{ mm yr}^{-1}$ expected (e.g., Stevenson et al., 1986; Wood et al., 1989; Allen and Duffy,



Figure 3. The Arne 14 C post bomb data plotted against sediment depth (a) and fitted to the Northern Hemisphere, Zone 1, atmospheric 14 C curve of Hua and Barbetti (2004) (b). The black triangle shows where the displaced 'woody' sample, ARN-03-20, plots. The open triangle shows the age indicated for this sample using the bomb spike calibrated F¹⁴C value (See Table 2). Analytical two-sigma uncertainty is smaller than the symbols. The location of ARN-03-31 is shown in panel a but because the CALIB 5.0 age range is used in the final chronology it is not shown in panel b.



Figure 4. The Arne pre-bomb 14 C ages plotted against depth (a) and the manual fit of the 2 sigma CALIB 5.0 (Stuiver et al., 2005) calibration of these data to the Intcal04 calibration curve (Reimer et al., 2004b) (b). The 1 sigma uncertainties attached to the 14 C (*y* error) and the 2 sigma CALIB 5.0 calibration age ranges (*x* error) are shown in panel (b). The position of the discarded 14 C age from 47.5 cm is shown by the open circle symbol in panel (a). See text for further explanation.

1998; Bartholdy et al., 2004; Price et al., 2005) in an upper saltmarsh environment, i.e. without any exceptional influences, like invasive sediment trapping alien plant species or anthropogenic catchment disturbance. In contrast, if the older calibration for ARN-03-76 of AD 1643–1669 is used this gives an accumulation rate of $1.4-1.6 \text{ mm yr}^{-1}$ for this section, which is within the range expected for this environment. In addition, it is also in some agreement with the rate of $1.1-1.2 \text{ mm yr}^{-1}$ determined by Long et al. (1999) for the period of AD 1750 to AD 1890, and so the AD 1643–1669 solution is selected.

The three remaining samples each have two possible CALIB 5.0 calibrations that satisfy the constraints discussed above. An argument can be made for simply 'joining the dots' between ARN-03-76 and the pollen marker at 40 cm, but to resolve better the remaining samples the data are manually fitted to the IntCal04 ¹⁴C calibration curve (Reimer et al., 2004b). A non-linear deposition rate is assumed which allows for some vertical adjustment between the samples, and the confirmed age solutions are plotted on to the curve (Fig. 4b). The year-range from the selected CALIB 5.0 age solution is shown in Figure 4b as the Cal AD uncertainty. Using the constraint of the independent age markers and the confirmed CALIB 5.0 solutions it is evident that if the stratigraphy were maintained there is only one solution for both ARN-03-55 and ARN-03-56 (Fig. 5). However, there is no way to decide reliably between the two possibilities remaining for ARN-02-62, AD 1766-1772 or AD 1777-1800. If the simple 'joint-the-dots' approach is used between ARN-03-76 and ARN-03-56 (Fig. 5) then AD 1766-1772 is selected for ARN-03-62, and, after consideration of the other information available here, this is felt to be the most likely solution.

Discussion

Calibration issues

A number of issues were attached to the initial calibration of the 14 C dates. Firstly, the sample from 20 cm, ARN-03-20,

calibrated off the bomb curve in Figure 3, and it was disregarded in the construction of the chronology. This was the only sample in the set not to have been a grass stem fragment. We conclude that the ¹⁴C of this 'woody stem' was assimilation by the plant some years before it was finally deposited on the marsh surface and entombed around 1966. Although the ¹⁴C analysis does not provide us with the date of deposition we are confident that the date of AD 1955.5 to 1956.8, obtained from the 1.0418 F¹⁴C value, accurately dates the assimilation of atmospheric carbon in this material.

Secondly, although the individual calibrations produced by CALIB 5.0 and CAILBomb for ARN-03-31 are different it is only by a matter of one calendar year (Table 2). Therefore, for simplicity only the CAILB 5.0 solution (AD 1917 to AD 1952) is shown as part of the sample array in Figure 4 and subsequently used in Figure 5.

The third issue with the ¹⁴C calibration was the chronological displacement of sample ARN-03-47.5 in the sequence. The CALIB 5.0 calibration for this sample produced four possible solutions which spanned the time period of AD 1667 to AD 1952 (Table 1), but given the location of this sample in the stratigraphy, i.e. below the AD 1855 ± 5 pollen marker at 40 cm, we could immediately reject the youngest solution from CALIB 5.0 (i.e. AD 1932 to AD 1952). However, as shown in Figure 5, to accept any of the remaining solutions (AD 1667 to AD 1805) would necessitate moving this sample physically down sequence to a level below the AD 1850±25 SCP marker at 50 cm to maintain chronological order. A simple explanation of this situation is that this sample was originally deposited lower in the marsh stratigraphy and it has subsequently been eroded, transported inshore and then re-deposited in the high marsh zone sometime between AD 1825 and AD 1880. The alternative explanation is contamination with older carbon during sampling or preparation, but in view of the precautions taken against this kind of occurrence, this explanation is viewed as unlikely.

The only pre-bomb sample not resolved to a single option after the removal of all the unfeasible solutions is ARN-03-62.



Figure 5. Age-depth model for the Arne sediments. The CALIB 5.0 age ranges selected are shown in black and those discarded are shown in grey. The broken line shows the mid-points of the age ranges but is not necessarily the preferred chronology. The index numbers correspond with Tables 1 and 2. Uncertainties not shown are smaller than symbols.

Two solutions are left for this sample and these age ranges are separated chronologically by 7 yr, but, because of this 'dead zone', in finite terms the solutions cannot be legitimately combined. However, even if the younger solution (AD 1777 to AD 1800) is used instead of the one proposed here (AD 1766 to AD 1772) this sample still constrains the fit of the samples from 55 cm and 56 cm to the descending section of the wiggle in the calibration curve between AD 1770 and AD 1830 (Fig. 4). In addition, the selection of the alternative younger solution would not greatly change the mean accumulation rate for this section.

Chronology and accumulation rates

The prior knowledge of the independent age markers is used to establish limiting 'oldest possible' and 'youngest possible' ages for these sections of the stratigraphy (Fig. 5). The *Spartina anglica* pollen marker dates an event that was both short in duration and unique in its occurrence in the record and can, therefore, be considered chronologically precise and used as a reliable signal of that date in the sequence. In contrast, the lower SCP marker has a 50-yr uncertainty attached and so it does not have high chronological precision, but its presence also marks a unique event and so it can be reliably used to constrain the ¹⁴C chronology during the early 18th century. The same reliability and uniqueness is true of the two upper SCP markers, thus allowing them to be used to confirm the CALIBomb calibration of the post-bomb ¹⁴C data, and specifically to select the age-range for ARN-03-26.5.

The new Arne chronology indicates that between the base of our sequence, dated to between AD 1643 and AD 1669, and the AD 1855 ± 5 pollen marker at 40 cm the mean accumulation rate at the site was 1.5 mm yr⁻¹. This broadly agrees with the rate of

 $1.1-1.2 \text{ mm yr}^{-1}$ suggested by Long et al. (1999) for the salt marshes on the Arne Peninsula before the arrival and expansion of *Spartina anglica* during the 1890s. Furthermore, in line with the situation proposed by Long et al. (1999), our data confirm that after the 1890s the mean accumulation rate for the 20th century was much higher than that for the 19th century, i.e. over 5 mm yr⁻¹. Long et al. (1999) calculated a rate of 7.1 mm yr⁻¹ for the sediments deposited after 1900, but our new dates indicate that in the 20th century the accumulation rate in the highest marsh zone remained relatively stable at 1.9 mm yr⁻¹ until the late 1960s and early 1970s, and then it accelerated to 8.3 mm yr⁻¹.

Long et al. (1999) propose that the increase in sedimentation rates in the Arne salt marshes during the 20th century was the result of the very efficient trapping of sediment by Spartina anglica following its introduction in the late 19th century. The ability of this plant to colonise the lower marsh and act as a sediment trap is now well known (e.g., Raybould, 2000; Boorman, 2003; Lacambra et al., 2004). It is also now known that after an initial phase of rapid expansion Spartina anglica populations have experienced periods of catastrophic 'die-back' in Poole Harbour, and other UK locations (See Lacambra et al., 2004 and references therein). In Poole Harbour the first of these die-back episodes began in the 1920s (Long et al., 1999). Long et al. (1999) discuses these episodic collapses of the Spartina anglica populations with reference to changes in tidal flows and water levels in Pool Harbour, and they suggest that there was evidence of increased siltation in nearby tidal channels following these phases. However, sub-populations of Spartina anglica respond with different timings (Thompson et al., 1991; Edwards, 2001; Boorman, 2003) and so it is possible for expansion, die-back and recovery to be occurring simultaneously in different marshes within a relative small area. During these die-back phases rapid marsh erosion takes place on the marsh

front, and the accumulated sediments held by the *Spartina* anglica sward are released back into the environment (Lacambra et al., 2004). Once in suspension fine material from the lower marsh is transported by the rising tide, and eventually redeposited in the high marsh zone (Widdows et al., 2000). This is the mechanism that we propose would explain the timing of the rapid acceleration indicated by our chronology. The arrival of *Spartina anglica* did not increase the accumulation rate in the Arne high marsh significantly during the early 20th century. It appears to have remained at 1.9 mm yr⁻¹ until around 1972 when it accelerated to 8.3 mm yr⁻¹, most likely following the release of sediments during an episode of the localised die-back of *Spartina anglica* in a nearby part of Poole Harbour.

Radionuclides

There is agreement between the post WW2 SCP age marker, the appearance of ¹³⁷Cs in the sediments and the two lower bomb-calibrated ¹⁴C points at 31 cm and 26.5 cm, but there is disagreement between ¹³⁷Cs and the ¹⁴C data as to the location of the 1963-1964 weapons fallout peak. The misalignment of these ¹³⁷Cs and ¹⁴C peaks may indicate that there has been some vertical displacement of ¹³⁷Cs in the Arne sediments. In some environmental situations ¹³⁷Cs and other radionuclides are known to exhibit post-depositional mobility in saturated sediments (Abril, 2004). Caesium-137 is soluble in sea water and behaves conservatively (Beks, 2000) in marine environments, so in some instances it can be remobilised in salt-marsh sediments and migrate vertically. This can result in ¹³⁷Cs being found in sediment horizons that are older than AD 1945 and predate the first dispersal of this artificial radionuclide (Thomson et al., 2002). It is suggested that this tendency for mobility can preclude its use as an independent dating tool (Kim et al., 1997; Abril, 2004; Donders et al., 2004). However we know that the peak of the ¹³⁷Cs discharged from the nearby UKAEA site at Winfrith was in 1980, and this activity may have contributed to the Arne radionuclide inventory during the 1960s. Therefore, the dependability of the sedimentary ¹³⁷Cs in the Arne sequence to signal an atmospheric fallout event during the 1960s must be viewed with some caution. Likewise, the presence of ²⁴¹Am in the Arne sediments between 12.5 cm and 22.5 cm cannot be reliably interpreted. Normally this activity would be viewed as significant because ²⁴¹Am is particle-reactive, and is irreversibly bound into particulate material after deposition (Clifton et al., 1999). This potentially makes it a more reliable marker than ¹³⁷Cs in a sediment sequence, but we do not know if ²⁴¹Am, or its parent ²⁴¹Pu, was part of the cocktail of alpha-emitting radionuclides discharged from Winfrith between 1958 and 1995.

Lead-210, like ²⁴¹Am, is non-conservative (Beks, 2000) in a marine environment and is scavenged by particulate material. We find there is some notable difference between the ²¹⁰Pb chronology on the one hand and the sediment depositional history indicated by the ¹³⁷Cs age-markers (Fig. 5) and the presence of ²⁴¹Am on the other. We note that this is not unique, and point out that other salt-marsh studies have experienced difficulties when attempting to align ²¹⁰Pb chronologies and ¹³⁷Cs age-markers. A study in the USA by Kim et al. (1997)

found a poor fit between ¹³⁷Cs age-markers and a ²¹⁰Pb_{excess} chronology in a Delaware salt marsh, and in the UK Thompson et al. (2002) found significant variability in the alignment of the results of their ²¹⁰Pb_{excess} analysis and the age-markers derived from ¹³⁷Cs at their Beaulieu Marsh and Wry Marsh sites. Thomson et al. (2002) suggest that the assumed steady-state that is required for ²¹⁰Pb_{excess} dating to work reliably does not exist in some salt-marsh environments, and propose that this may be because of fluctuations in accumulation rates. However, simple fluctuations in sediment accumulation rates can, to some extent, be incorporated into the age-modelling by the use of cumulative dry mass/unit area data and the CRS model (Appleby and Oldfield, 1978). Therefore, we must assume that other variables, that are not being incorporating into the modelling, are capable of distorting the result. These variables could include changes in grain size (Maringer, 1996; Dellapenna et al., 2003) or subtle changes in sediment lithology and magnetic properties (McCubbin et al., 2000) that may not change the bulk density significantly, but may preferentially change the affinity of the sediment for ²¹⁰Pb.

Justification of the high precision dates

In addition to our use of two ¹⁴C calibration methods we have used a combination of normal precision 3‰ and high precision 2‰ AMS analysis. To justify the extra commitment of laboratory resources there must be an apparent benefit from the use of higher precision analysis. This advanced type of ${}^{14}C$ AMS analysis demands an organic carbon sample of sufficient size for the production of multiple graphite targets, and requires proportionally more preparation time to produce these replicates. Then we must consider the extended analytical time on the AMS and the cost of running the instrument at 2‰. An inspection of our AMS results shows that the use of high precision analysis reduces the analytical one sigma uncertainty from approximately 0.0030 $F^{14}C$ to 0.0018 $F^{14}C$ (Table 1 and Table 2). This translates to a one sigma ¹⁴C yr uncertainty of ± 15 yr for the 2‰ data (Table 1) instead of the ± 20 to 26 yr (Table 2) associated with the 3‰ data. By combining the replicate data for each ¹⁴C sample in CALIB 5.0 this uncertainty is further reduced to ± 9 ¹⁴C yr (Table 1). It is assumed this reduction in analytical and ¹⁴C yr uncertainty results in a corresponding reduction in the spread of the Cal yr age ranges of each of the calibration solutions. This seems to be the case, but to evaluate this more formally we compare the results obtained from the 2 ‰ analysis of the Arne samples with the same ¹⁴C yr data using an arbitrary ± 25 ¹⁴C yr uncertainty to simulate a 3‰ analysis (Table 3).

The mean and standard deviation is calculated for each of the age-ranges produced by CALIB 5.0 for both the real and simulated ¹⁴C yr data (Table 3). Then the *X*, *Y* and *Z* replicates are combined in CALIB 5.0 and those data evaluated in the same way. The data show that using a ¹⁴C yr uncertainty consistent with a 2 ‰ analytical precision there is a reduction of >45% in most cases of the spread of the age ranges for individual or single-point data. A further reduction in the means and standard deviations of the age ranges is apparent when the

Table 3

The mean ranges and one standard deviations of the age-solutions derived by CALIB 5.0 from the high-precision 2‰ Arne AMS data and simulated 3‰ data

| ¹⁴ C data | Publication no. | ^{14}C yr BP uncertainty $\pm 1\sigma$ | Age-ranges of the Calib 5.0 calibration solutions (Cal yr) | Mean and $[1\sigma]$ of each of the CALIB 5.0 age-ranges |
|--|--------------------------|--|--|--|
| ARN-03-34 2‰ AMS data: | | | | |
| X | SUERC-6364 | 155 ± 15 | 8, 34, 3, 8, 13, 0 | 11.0 [12.13] |
| Y Z | SUERC-6365 SUERC-6366 | 146 ± 15 137 ± 15 | 12, 35, 10, 19, 11 14, 11, 3, 11, 7, 18, 4, 19, 1 | 15.4 [12.70] 9.8 [6.45] |
| Combined ARN-03-34 2‰ data | na | 146 ± 9 | 10, 6, 7, 8, 14, 0 | 8 [4.60] |
| Simulated 3‰ data using±25 yr uncertainty: | | | | |
| Xs | na | 155 ± 25 | 16, 38, 3, 13, 2, 4, 4, 22, 1 | 20.6 [18.31] |
| Ys | na | 146 ± 25 | 18, 41, 6, 13, 4, 14, 1, 22 | 14.9 [12.74] |
| Zs | na | 137±25 | 17, 15, 19, 13, 44, 22, 1 | 18.7 [13.00] |
| Combined simulated 3‰: X_s , Y_s and Z_s | na | 146 ± 14 | 12, 35, 10, 18, 1 | 15.2 [12.64] |
| ARN-03-42 2‰ AMS data: | | 100 - 15 | 10 0 10 10 17 11 0 | 10.1.515.651 |
| X V | SUERC-6369 | 120 ± 15 127+15 | 12, 2, 10, 10, 47, 11, 0 | 13.1 [15.65] |
| Z | SUERC-6371 | 127 ± 13 147 ± 15 | 12, 35, 10, 19, 1 | 15.4 [12.70] |
| Combined ARN-03-42 2‰ data | na | 131±9 | 11 7 6 5 16 3 10 | 8 3 [4 39] |
| Simulated 3% data using +25 yr uncertainty: | iiu | 151-7 | 11, 7, 0, 2, 10, 2, 10 | 0.0 [1.0 7] |
| $X_{\rm s}$ | na | 120 ± 25 | 21, 12, 18, 54, 15, 0 | 20.0 [18.17] |
| Y _s | na | 127 ± 25 | 58, 19, 95, 36, 2 | 42.0 [36.16] |
| Zs | na | 147 ± 25 | 19, 50, 14, 3, 14, 0, 23, 1 | 15.2 [16.34] |
| Combined simulated 3‰: X_s , Y_s and Z_s | na | $131\!\pm\!14$ | 12, 9, 8, 12, 27, 12, 0 | 11.5 [8.08] |
| ARN-03-47.5 2‰ AMS data: | | | | |
| X | SUERC-6374 | 174 ± 15 | 12, 17, 18, 4, 8 | 11.8 [5.93] |
| Y | SUERC-6375 | 175 ± 15 | 12, 16, 18, 4, 8 | 11.6 [5.73] |
| Combined ARN-03-47.5 2‰ data | na | 175 ± 11 | 12, 12, 18, 4, 7 | 10.6 [5.24] |
| Simulated 3‰ data using±25 yr uncertainty: | | | | |
| X _s | na | 174±25 | 15, 45, 8, 13, 0 | 16.2 [17.11] |
| Y _s | na | 175 ± 25 | 14, 46, 7, 16 | 20.8 [17.27] |
| Combined simulated $3\%: X_s$ and Y_s | na | 174 ± 18 | 12, 18, 19, 5, 8 | 12.4 [6.11] |
| ARN-03-55 2‰ AMS data: | | | | |
| X | SUERC-6376 | 83 ± 15 | 13, 12, 31, 2 | 14.5 [12.07] |
| 1 7 | SUERC-6380 | 82 ± 15 | 14, 9, 14, 22, 4, 22, 8 | 13.3 [0.90] |
| Combined ARN-03-55 2% data | n9 | 02+0 | 1 14 13 12 10 | 10.0 [5.24] |
| Simulated 20/ data using +25 yr uncertainty. | lia |)2-1) | 1, 17, 13, 12, 10 | 10.0 [5.24] |
| Simulated 5‰ data using ± 25 yr uncertainty: X ₋ | na | 83+25 | 25 18 38 2 | 20.8 [15:00] |
| Y _s | na | 110 ± 25 | 17, 10, 78, 10, 0 | 23.0 [31.34] |
| Zs | na | 82±25 | 25, 18, 38, 2 | 20.8 [15.00] |
| Combined simulated 3% $X_{\rm s}$, $Y_{\rm s}$ and $Z_{\rm s}$ | na | 92±14 | 22, 15, 16, 12 | 16.3 [4.20] |
| ARN-03-62 2‰ AMS data: | | | | |
| X | SUERC-6381 | 188 ± 15 | 14, 20, 4, 12 | 12.5 [6.61] |
| Y | SUERC-6382 | 210 ± 15 | 6, 16, 3 | 8.3 [6.81] |
| Combined ARN-03-2-62 2‰ data | na | 199±9 | 5, 6, 6, 8 | 6.3 [1.29] |
| Simulated 3 ‰ data using±25 yr uncertainty: | | | | |
| X _s V | na | 188 ± 25 210+25 | 16, 16, 23, 8, 14 22, 21, 10 | 15.4 [5.37] |
| | па | 210±25 | 22, 21, 10 | 11.5 [0.00] |
| Combined simulated 3‰: X_s and Y_s | na | 199±18 | 13, 1, 22, 10 | 11.5 [8.67] |

X, *Y*, and *Z* replicates are combined. However, this exercise also shows that if two or more normal 3‰ precision replicates are analysed, and then combined using CALIB 5.0, a real improvement in the age-range spread of the calibration results can be achieved over individual or single-point data.

Conclusions

We have demonstrated the successful use of the 'bomb spike' calibration method in a salt-marsh sediment sequence, and the use of two types of independent age markers to constrain prebomb ¹⁴C age ranges. The results presented in Figure 5 show the agreement between the ¹⁴C 'bomb spike' dates, the earliest ¹³⁷Cs marker, and the chronological information from lower in the sequence. There is some disagreement between the ¹⁴C and the ²¹⁰Pb_{excess} chronologies, but our results from the Arne site agree with the conclusions of other studies in salt-marsh sites where ²¹⁰Pb dating has been compared with other methods. Therefore, our results suggest that the concerns that have been expressed regarding the use of isolated ²¹⁰Pb chronologies, particularly in dynamic sedimentary environments like salt marshes, are well founded.

We use our new chronology for the Arne sediments to confirm the acceleration of sediment accumulation in the Arne salt marshes since the late 1800s reported by Long et al. (1999). Long et al. (1999) attribute this to the increased sediment trapping at the marsh surface by the newly invading *Spartina anglica* population. However, our data indicate that in the Arne high marsh zone there was no significant change in accumulation during the initial expansion phase of *Spartina anglica*. At our site the highest rates of sedimentation occurred after 1972, possibly as a response to the die-back of a local *Spartina anglica anglica* population.

The benefit of using the higher precision 2‰ AMS analysis is confirmed by the small analytical uncertainties produced. The use of high-precision AMS analysis translates directly into a reduction of possible calibrated age solutions. By using a combination of high-precision AMS data and the constraint of independent age markers this study has produced a robust high-resolution chronology spanning the last three centuries for sediment deposition in the Arne salt marsh. We conclude that these methods of calibrating ¹⁴C data have great potential for the dating of recent salt-marsh sediments, and are useful chronological tools for the study of recent sea-level change.

Acknowledgments

This work has been supported by NERC radiocarbon allocation 1083.0404. It forms part of a PhD project (WM) funded by the School of Geography, University of Plymouth, and is a contribution to IGCP Project 495 ("Late Quaternary Land-Ocean Interactions: Driving Mechanisms and Coastal Responses"). Maria Gehrels and Mel Bugler assisted with fieldwork, and Peter Appleby (Liverpool) performed ²¹⁰Pb and ¹³⁷Cs analyses. Neil Rose (University College London) is thanked for the SCP data from his Harbour Farm and Abbotsbury Embayment sites. We gratefully acknowledge the refinements made to this paper resulting from the comments of the two anonymous reviewers.

References

- Abril, J.M., 2004. Constraints on the use of ¹³⁷Cs as a time-marker to support CRS and SIT chronologies. Environmental Pollution 129, 31–37.
- Allen, J.R.L., 2003. An eclectic morphostratigraphic model for the sedimentary response to Holocene sea-level rise in northwest Europe. Sedimentary Geology 161, 31–54.

- Allen, J.R.L., Duffy, M.J., 1998. Medium-term sedimentation on high intertidal mudflats and salt marshes in the Severn Estuary, SW Britain: the role of wind and tide. Marine Geology 150, 1–27.
- Appleby, P.G., 2001. Chronostratigraphic techniques in recent sediments. In: Last, W.M., Smol, J.P. (Eds.), Tracking Environmental Change Using Lake Sediments Volume 1: Basin Analysis, Coring, and Chronological Techniques, Kluwer Academic, pp. 171–203.
- Appleby, P.G., Oldfield, F., 1978. The calculation of 210Pb dates assuming a constant rate of supply of unsupported 210Pb to the sediment. Catena 5, 1–8.
- Appleby, P.G., Nolan, P.J., Gifford, D.W., Godfrey, M.J., Oldfield, F., Anderson, N.J., Battarbee, R.W., 1986. ²¹⁰Pb dating by low background gamma counting. Hydrobiologia 141, 21–27.
- Appleby, P.G., Richardson, N., Nolan, P.J., 1991. ²⁴¹Am dating of lake sediments. Hydrobiologia 214, 35–42.
- Bartholdy, J., Christiansen, C., Kunzendorf, H., 2004. Long term variations in backbarrier salt marsh deposition on the Skallingen peninsula, the Danish Wadden Sea. Marine Geology 203, 1–21.
- Beks, J.P., 2000. Storage and distribution of plutonium, ²⁴¹Am, ¹³⁷Cs and ²¹⁰Pb_{xs} in North Sea sediments. Continental Shelf Research 20, 1941–1964.
- Boorman, L.A., 2003. Saltmarsh Review. An overview of coastal saltmarshes, their dynamic and sensitivity characteristics for conservation and management. JNCC Report, No. 334. Joint Nature Conservation Committee, Peterborough.
- Bristow, C.R., Freshney, E.C., Penn, I.E. 1991. Geology of the Country around Bournemouth. Memoir for 1:50,000 Geological Sheet 329 (England and Wales). HMSO, London
- Clifton, J., McDonald, P., Plater, A., Oldfield, F., 1999. Derivation of a grainsize proxy to aid the modelling and prediction of radionuclide activity in salt marshes and mud flats of the Eastern Irish Sea. Estuarine, Coastal, and Shelf Science 48, 511–518.
- Dellapenna, T.M., Kuehl, S.A., Schaffner, L.C., 2003. Ephemeral deposition, seabed mixing and fine-scale strata formation in the York River estuary, Chesapeake Bay. Estuarine, Coastal and Shelf Science 58, 621–643.
- Donders, T.H., Wagner, F., van der Borg, K., de Jong, A.F.M., Visscher, H., 2004. A novel approach for developing high-resolution sub-fossil peat chronologies with ¹⁴C dating. Radiocarbon 46 (1), 455–463.
- Edwards, R.J., 2001. Mid- to late Holocene relative sea-level change in Poole Harbour, southern England. Journal of Quaternary Science 16, 221–235.
- Garnett, M.H., Stevenson, A.C., 2004. Testing the use of bomb radiocarbon to date the surface layers of blanket peat. Radiocarbon 46 (2), 841–851.
- Gehrels, W.R., 2000. Using foraminiferal transfer functions to produce highresolution sea-level records from salt-marsh deposits, Main, USA. The Holocene 10, 367–376.
- Gehrels, W.R., Belknap, D.F., Black, S., Newnham, R.M., 2002. Rapid sealevel rise in the Gulf of Maine, USA, since AD 1800. The Holocene 12, 383–393.
- Gehrels, W.R., Kirby, J.R., Prokoph, A., Newnham, R.M., Achterberg, E.P., Evans, H., Black, S., Scott, D.B., 2005. Onset of recent rapid sea-level rise in the western Atlantic Ocean. Quaternary Science Reviews 24 (18–19), 2083–2100.
- Goodsite, M.E., Rom, W., Heinemeier, J., Lange, T., Ooi, S., Appleby, P.G., Shotyk, W., van der Knaap, W.O., Lohse, C., Hansen, T.S., 2001. Highresolution AMS ¹⁴C dating of post-bomb peat archives of atmospheric pollutants. Radiocarbon 43 (2B), 495–515.
- Harvey, M.M., Hansom, J.D., MacKenzie, A.B., 2007. Constraints on the use of anthropogenic radionuclide-derived chronologies for saltmarsh sediments. Journal of Environmental Radioactivity 95, 126–148.
- Haslett, S.K., Strawbridge, F., Martin, N.A., Davies, C.F.C., 2001. Vertical saltmarsh accretion and its relationship to sea-level in the Severn Estuary, UK: an investigation using foraminifera as tidal indicators. Estuarine Coastal and Shelf Science 52 (1), 143–153.
- Hua, Q., Barbetti, M., 2004. Review of tropospheric Bomb ¹⁴C data for carbon cycle modeling and age calibration purposes. Radiocarbon 46, 1273–1298.
- Jones, V.J., Battarbee, R.W., Rose, N.L., Curtis, C., Appleby, P.G., Harriman, R., Shine, A.J., 1997. Evidence for the pollution of Loch Ness from the

analysis of its recent sediments. The Science of the Total Environment 203, 37-49.

- Kim, G., Hussain, N., Church, T.M., Careyb, W.L., 1997. The fallout isotope ²⁰⁷Bi in a Delaware salt marsh: a comparison with ²¹⁰Pb and ¹³⁷Cs as a geochronological tool. The Science of the Total Environment 196, 31–41.
- Krey, P.W., Hardy, E.P., Toonkey, L.E., 1976. The distribution of plutonium and americium with depth in soil at Rocky Flats. USERDA Environmental Quarterly Report. HASL-318.U.S. Atomic Energy Commission. Health and Safety Lab. New York.
- Lacambra, C., Cutts, N., Allen, J., Burd, F., Elliott, M., 2004. Spartina anglica: a review of its status, dynamics and management. English Nature Research Reports, Number 527. English Nature, Peterborough.
- Long, A.J., Scaife, R.G., Edwards, R.J., 1999. Pine pollen in intertidal sediments from Poole Harbour, UK; implications for late-Holocene sediment accretion rates and sea-level rise. Quaternary International 55, 3–16.
- Maringer, F.J., 1996. The partitioning of natural radionuclides in a large Alpine river. Environment International 25 (1), 323–331.
- McCubbin, D., Leonard, K.S., Maher, B.A., Hamilton, E.I., 2000. Association of ²¹⁰Po(²¹⁰Pb), ²³⁹⁺²⁴⁰Pu and ²⁴¹Am with different mineral fractions of a beach sand at Seascale. Cumbria, UK. The Science of the Total Environment 254, 1–15.
- McGee, E.J., Gallagher, D., Mitchell, P.I., Baillie, M., Brown, D., Keogh, M., 2004. Recent chronologies for tree rings and terrestrial archives using ¹⁴C bomb fallout history. Geochimica et Cosmochimica Acta 68 (11), 2509–2516.
- Moore, P.D., Webb, J.A., Collinson, M.E., 1991. Pollen Analysis. Blackwell Scientific Publications, London.
- Morris, K., Butterworth, J.C., Livens, F.R., 2000. Evidence for the remobilization of Sellafield Waste Radionuclides in an Intertidal Salt Marsh, West Cumbria, U.K. Estuarine. Coastal and Shelf Science 51, 613–625.
- Plater, A.J., Horton, B.P., Haworth, E.Y., Appleby, P.G., Zong, Y., Wright, M.R., Rutherford, M.M., 2000. Holocene tidal levels and sedimentation rates using a diatom-based palaeoenvironmental reconstruction: the Tees estuary, northeastern England. The Holocene 10 (4), 441–452.
- Pourchet, M., Magand, O., Frezzotti, M., Ekaykin, A., Winther, J.-G., 2003. Radionuclides deposition over Antarctica. Journal of Environmental Radioactivity 68, 137–158.
- Price, G., Winkle, K., Gehrels, W.R., 2005. A geochemical record of the mining history of the Erme Estuary, south Devon. UK Gregory D. Marine Pollution Bulletin 50, 1706–1712.
- Raybould, A.F., 2000. Hydrographical, ecological and evolutionary change associated with Spartina anglica in Poole harbour. In: Sherwood, E.B.R., Gardiner, B.G., Harris, T. (Eds.), British Saltmarshes, Forrest Text for the Linnean, pp. 129–142.
- Reimer, P.J., Reimer, R.W., 2004. Discussion: reporting and calibration of postbomb ¹⁴C data. Radiocarbon 46 (3), 1299–1304.
- Reimer, P.J., Baillie, M.G.L., Bard, E., Bayliss, A., Beck, J.W., Bertrand, C., Blackwell, P.G., Buck, C.E., Burr, G., Cutler, K.B., Damon, P.E., Edwards, R.L., Fairbanks, R.G., Friedrich, M., Guilderson, T.P., Hughen, K.A., Kromer, B., McCormac, F.G., Manning, S., BronkRamsey, C., Reimer, R.W., Remmele, S., Southon, J.R., Stuiver, M., Talamo, S., Taylor, F.W., van der Plicht, J., Weyhenmeyer, C.E., 2004. INTCAL04 Terrestrial radiocarbon age calibration, 26–0 ka BP. Radiocarbon 46, 1029–1058.
- Robbins, J.A., 1978. Geochemical and geophysical applications of radioactive lead. In: Nriagu, J.O. (Ed.), Biogeochemistry of Lead in the Environment. Elsevier Scientific, Amsterdam, pp. 285–393.

- Rose, N.L., 2001. Fly ash particles. In: Last, W.M., Smol, J.P. (Eds.), Tracking Environmental Change Using Lake Sediments. Physical and Chemical Techniques, vol. 2. Kluwer Academic Publishers, Dordrecht, The Netherlands, pp. 319–349.
- Rose, N., Appleby, P.G., 2005. Regional applications of lake sediment dating by spheroidal carbonaceous particle analysis I: United Kingdom. Journal of Paleolimnology 34 (3), 349–361.
- Rose, N.L., Juggins, S., Watt, J., 1999. The characterisation of carbonaceous flyash particles from major European fossil-fuel types and applications to environmental samples. Atmospheric Environment 33, 2699–2713.
- Shennan, I., 1989. Holocene crustal movements and sea-level change in Great Britain. Journal of Quaternary Science 4, 77–89.
- Shennan, I., Horton, B., 2002. Holocene land- and sea-level changes in Great Britain. Journal of Quaternary Science 17, 511–526.
- Shotyk, W., Goodsite, M.E., Roos-Barraclough, F., Frei, R., Heinemeier, J., Asmund, G., Lohse, C., Hansen, T.S., 2003. Anthropogenic contributions to atmospheric Hg, Pb and As accumulation recorded by peat cores from southern Greenland and Denmark dated using the ¹⁴C "bomb pulse curve". Geochimica et Cosmochimica Acta 67 (21), 3991–4011.
- Smith, J.N., 2001. Why should we believe ²¹⁰Pb sediment geochronologies? Journal of Environmental Radioactivity 55, 121–123.
- Stevenson, C.J., Ward, L.G., Kearney, M.S., 1986. Vertical accretion in marshes with varying rates of sea-level rise. In: Wolfe, D. (Ed.), Estuarine Variability. Academic Press Inc., San Diego, pp. 241–259.
- Stuiver, M., Polach, H.A., 1977. Discussion: reporting of ¹⁴C data. Radiocarbon 19 (3), 355–363.
- Stuiver, M., Braziunas, T.F., 1998. Anthropogenic and solar components of hemispheric C-14. Geophysical Research Letters 25 (3), 329–332.
- Stuiver, M., Reimer, P.J., Bard, E., Beck, J.W., Burr, G.S., Hughen, K.A., Kromer, B., McCormac, G., VanderPlicht, J., Spurk, M., 1998. INTCAL98 radiocarbon age calibration, 24,000-0 cal BP. Radiocarbon 40 (3), 1041–1083.
- Stuiver, M., Reimer, P.J., Reimer, R.W. 2005. CALIB 5.0. [WWW_program_ and_documentation]. Online at : http://radiocarbon.pa.qub.ac.uk/ (Accessed 02/12/05).
- Telford, R.J., Heegaard, E., Birks, H.J.B., 2004. The intercept is a poor estimate of a calibrated radiocarbon age. The Holocene 14 (2), 296–298.
- Thompson, J.D., McNeilly, T., Gray, A.J., 1991. Population variation in *Spartina anglica* C.E. Hubbard. New Phytologist 117, 115–128.
- Thomson, J., Dyer, F.M., Croudace, I.W., 2002. Records of radionuclide deposition in two salt marshes in the United Kingdom with contrasting redox and accumulation conditions. Geochimica et Cosmochimica Acta 66 (6), 1011–1023.
- Turetsky, M.R., Manning, S.W., Wieder, R.K., 2004. Dating Recent Peat Deposits. Wetlands 24 (2), 324–356.
- UNSCEAR, 2000. Sources and Effects of Ionizing Radiation. UN, New York.
- Widdows, J., Brown, S., Brinsley, M.D., Salkeld, P.N., Elliott, M., 2000. Temporal changes in intertidal sediment erodability: influence of biological and climatic factors. Continental Shelf Research 20, 1275–1289.
- Wood, M., Kelley, J.T., Belknap, D.F., 1989. Paterns of sediment accumulation in the Tidal Marshes of Maine. Estuaries 4, 237–245.
- Yang, H., Rose, N.L., Battarbee, R.W., 2001. Dating of recent catchment peats using spheroidal carbonaceous particle (SCP) concentration profiles with particular reference to Lochnagar, Scotland. The Holocene 11, 593–597.
- Zong, Y.Q., Horton, B.P., 1999. Diatom-based tidal-level transfer functions as an aid in reconstructing Quaternary history of sea-level movements in the UK. Journal of Quaternary Science 14 (2), 153–167.