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Research paper

A period of calm in Scottish seas: A comprehensive study of ΔR values for the northern British Isles coast and the consequent implications for archaeology and oceanography

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ABSTRACT

The Marine Radiocarbon Reservoir Effect (MRE) is a ^{14}C age offset between contemporaneous marine- and terrestrially-derived carbon. In Northern Hemisphere surface waters it is of the order of 400 years but temporal and spatial deviations, known as ΔR, occur. This study provides a comprehensive dataset of 21 ΔR and MRE values for the east coast of Scotland and 21 recalculated values for the west coast of Scotland and Ireland, for the period c. 3500 BC to 1450 AD. They are presented as mean, site-specific ΔR and MRE values, together with their associated uncertainties, calculated as standard errors for predicted values. The ΔR values range from -320 ± 35 to $+150 \pm 28$ ^{14}C years and show no spatial or temporal trends. The MRE values range from 59 ± 40 to 531 ± 26, show an almost identical distribution pattern to the ΔR values and again show no spatial or temporal trends. Results show that ΔR values calculated for a single site using statistically indistinguishable groups of terrestrial and marine radiocarbon age measurements can produce variability of up to 225 ^{14}C years. ΔR is an important factor in the accurate calibration of samples containing marine-derived carbon for archaeological interpretation but is often also used as an indicator of changes in ^{14}C specific activity of the oceans, and therefore a proxy for changes in ocean circulation and/or climate. Using the methods outlined in this paper, it is apparent that ΔR values for the northern part of the British Isles have been relatively stable, within our ability to quantify non-random variation in the data. The fact that significant climatic shifts have been recorded during this time, yet these are not visible in the ΔR data, presents a cautionary tale regarding the use of ΔR to infer large-scale oceanographic or climatic changes. Upon the exclusion of 5 outliers from the 42 values, the remaining ΔR values are statistically indistinguishable from one another and range from -142 ± 61 to $+40 \pm 47$ ^{14}C years. 34 of these values are from Scottish archaeological sites and can be combined to produce a mean value for Scotland of -47 ± 52 ^{14}C years for the period 3500 BC to 1450 AD, to be used only in the absence of site- and period-specific data.

1. Introduction

The Marine Radiocarbon Reservoir Effect (MRE) manifests itself as a ^{14}C age offset between samples formed in the terrestrial biosphere and contemporaneous samples formed in the marine environment (Stuiver et al., 1986). This occurs due to the difference in mixing rates and residence times of carbon atoms in the two reservoirs, while variations in local conditions and mixing rates prevent there from being a universal ^{14}C offset from the atmosphere for all oceanic environments (Jones et al., 2007a,b; Gomez et al., 2008; Harkness, 1983). Variations in the ocean/atmosphere CO_2 exchange rate, stratification and upwelling of different water masses, etc will all influence the ^{14}C content of water bodies, resulting in a non-uniform ^{14}C concentration (Gordon and Harkness, 1992). On average, the MRE offset between contemporary marine and terrestrial material is of the order of 400 ^{14}C years for the global surface oceans in the Northern Hemisphere (Stuiver and Braziunas, 1993). However, because of the inherently variable nature of this offset, accurate calibration of radiocarbon ages determined from samples containing marine derived carbon can be problematic (Ascough et al., 2004).

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Marine radiocarbon ages are calibrated using a modelled marine curve based on atmospheric data. The current calibration curve (Marine13) (Reimer et al., 2013) uses the ocean-atmosphere box diffusion model (Oeschger et al., 1975; Stuiver and Braziunas, 1993). This modelled marine calibration curve accounts for the global average offset of oceanic ^{14}C with respect to the atmosphere, producing a present-day average surface water reservoir offset of 405 ± 22 ^{14}C yr (Hughen et al., 2004), however, temporal and spatial deviations from this offset, known as ΔR, are evident (Stuiver and Braziunas, 1993; Ascough et al., 2006). Robust ΔR values are calculated using multiple paired samples of terrestrial and marine origin that are of the same calendar age. The ΔR value is calculated by converting the terrestrial/atmospheric ^{14}C age ±1 sigma to a modelled marine age via interpolation between the INTCAL 13 atmospheric curve and the MARINE13 curve (Reimer et al., 2013). ΔR is the difference between this modelled marine ^{14}C age and the measured ^{14}C age of the corresponding marine carbon sample. The 1σ error on the ΔR values is calculated by the propagation of errors on both ages. ΔR is factored into the calibration process by subtracting it from the conventional radiocarbon age (CRA) and then calibrating with the marine curve. A positive ΔR will therefore increase the MRE for the area, relative to the global average, whilst a negative ΔR will decrease it. Globally, ΔR values can show significant variation (Fig. 1) as shown by the data held on the 14 CHRONO Marine Reservoir database at http://intcal.qub.ac.uk/marine/.

1.1. Variations in ΔR as oceanographic indicators

The spread of ΔR values shown in Fig. 1 demonstrates the global variability. This variability in ΔR is often attributed to changes in ocean water ^{14}C activity, related to shifts in circulation patterns. Using this rationale, ΔR is often used as a proxy for identifying past oceanographic changes.

Palaeoclimatic variations that affect the amount of time that water is in contact with the atmosphere can affect the MRE. Colder conditions such as the extension of the Arctic ice sheet would increase sea ice cover, leading to less area available for ocean/atmosphere CO_2 exchange. Deep waters would become increasingly depleted in ^{14}C as they are further removed from contact with the atmosphere. Conditions which induce a higher rate of upwelling of deep, older waters will increase the MRE and vice versa, any conditions which allow the waters to stay near the surface, in contact with the atmosphere, will reduce the MRE. The MRE therefore has the power to reflect large scale shifts in ocean ^{14}C activity, provided that trends and shifts in the MRE (or ΔR values) are accurately identified.

Russell et al. (2011a) suggest a methodological approach to the publication of ΔR values and their errors in an attempt to raise awareness of the inherent variability in ΔR calculations. If this inherent variability is not accounted for in the published ΔR values, using their associated errors, misleading significance of changes in ΔR may well be interpreted as an indicator of oceanographic shifts. Various authors have used ΔR as a climatic/oceanographic proxy using a variety of methods of calculation (e.g. Etayo-Cadavid et al., 2013, Hideshima et al., 2001; Jones et al., 2007a; Kennett et al., 1997; Matos Martins and Monge Soares 2013). In some instances, application of the methodology employed by Russell et al. (2011a) to the data renders the differences in ΔR insignificant and

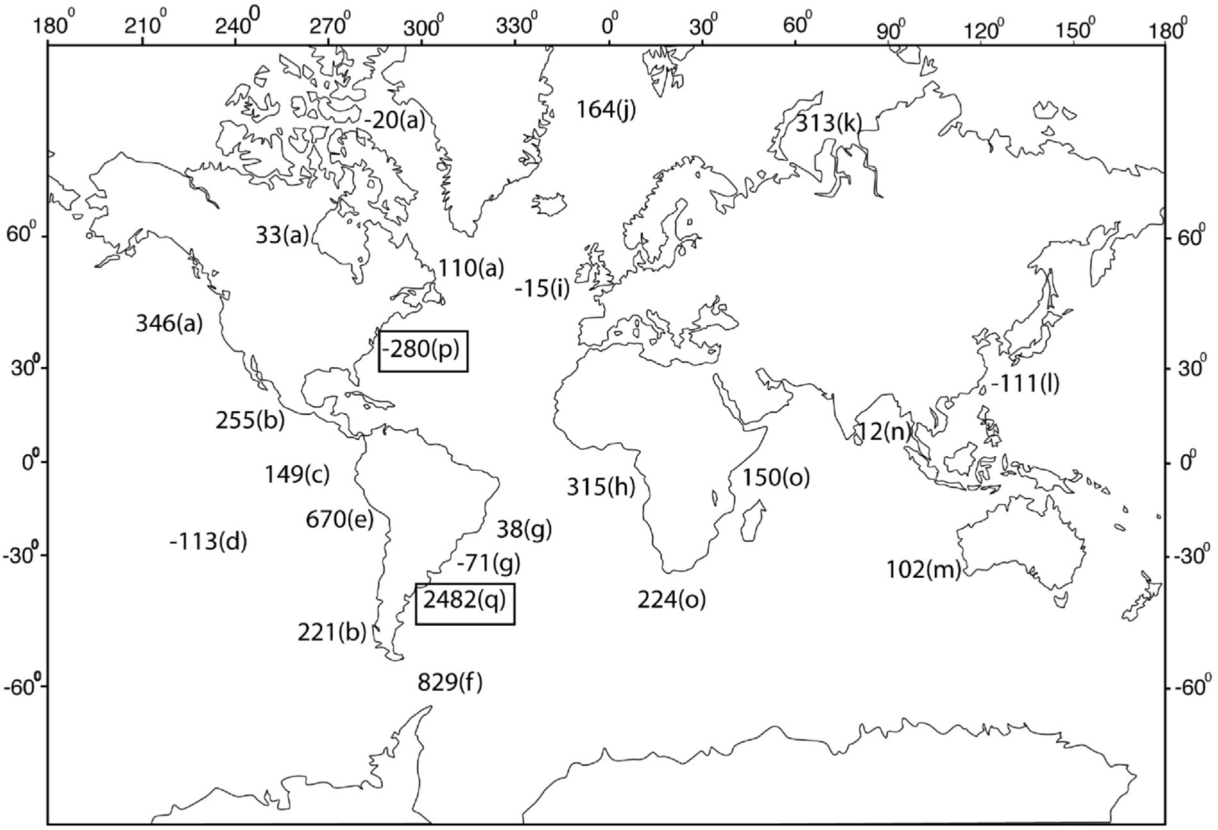

Fig. 1. Global variations in ΔR. Maximum and minimum global values are shown in boxes. All values are taken from the online 14 CHRONO Marine Reservoir database (http://intcal.qub.ac.uk/marine/). References for each value are: a) – McNeeley et al., 2006; b) – Ingram and Southon 1996; c) – Jones et al., 2007a; d) – Beck et al., 2003; e) – Taylor and Berger 1967; f) – Bjork et al., 1991; g) – Nadal de Masi 1999; h) – Lewis et al., 2008; i) – Harkness 1983; j) – Funder 1982; k) – Forman and Polyak 1997; l) – Kong and Lee 2005; m) – Bowman and Harvey 1983; n) – Dutta et al., 2001; o) – Southon et al., 2002; p) – Thomas 2008; q) – Gomez et al., 2008.

therefore the associated oceanographic proxies that have been drawn, invalid.

1.2. ΔR impacts on archaeological calibration

Accurate and precise quantification of the MRE and/or ΔR is of paramount importance for enabling the accurate calibration of ^{14}C age measurements made on samples containing marine-derived carbon that are of archaeological importance. Previous studies at SUERC which refined ΔR values for the west coast of Scotland, northern Iceland and the Faroes (Ascough et al., 2004, 2006; 2007a, 2007b; 2009) have led to significant chronological re-interpretation of Scottish archaeological sites, where conclusions had previously been drawn that were based upon radiocarbon age measurements made on marine derived carbon (e.g. Barber, 2003). The accuracy and precision of ΔR values and their associated errors therefore have the potential to impact significantly on the interpretation and evaluation of archaeological and oceanographic research alike.

ΔR values have been calculated for 21 contexts from 11 archaeological sites on the east coast of Scotland and recalculated for 21 contexts from 13 archaeological sites on the west coast of Scotland and Ireland. The west coast values were previously published using the method of Ascough (2005) but have been recalculated using the method recommended by Russell et al. (2011a) to allow comparability of results. This gives a total of 42 ΔR values for the UK coast which can be used to aid archaeological interpretation and paleo-oceanographic investigation.

1.3. Regional setting

The UK is situated to the North-west of continental Europe, bordered by the North Sea to the east, the English Channel to the south and the Irish Sea/Celtic Sea and Atlantic Ocean to the west (Fig. 2). Warm surface waters from the North Atlantic flow north-west, towards the Norwegian Sea as the North Atlantic Current (NAC), skirting the west coast of the UK as a variety of coastal currents before diverging into the North Sea (Fig. 4). OSPAR (2000), Baxter et al. (2008) and UKMMAS (2010), all provide more detailed discussion on UK coastal circulation. Russell et al. (2010, 2011b), Ascough et al. (2004, 2005a,b, 2006, 2007a,b) and Cage et al. (2006) all relate these specific current patterns and the characteristics of local circulation directly to UK MRE values.

The majority of sites from this study were located in the Northern British Isles, particularly Scotland. The sites range from Quoygrew on Orkney in the North, to Doonloughan in Ireland (Fig. 2) and span a temporal range from the 4th millenium BC to the 15th century AD (Table 1). The sites also occupy a variety of open coastal and estuarine locations. Some of the sites have the potential to be subject to coastal estuarine processes, particularly around the sea lochs of western Scotland and the major estuaries (firths) on the east coast of Scotland and any values calculated from such environments may not represent a true MRE *per se* and instead may demonstrate a local MRE, diluted by freshwater input.

2. Methodology

This study recalculated ΔR values that were previously published by Ascough et al. (2004, 2006, 2007a,b, 2009) and Ascough (2005), as well as those published by Russell et al. (2010, 2011a, 2011b) and Russell (2011), by employing the statistical methodology recommended by Russell et al. (2011a). Most of the radiocarbon measurements were carried out at the SUERC laboratory in East Kilbride, Scotland. All site-specific ΔR values from both studies were determined using the multiple paired sample approach as advised by Ascough et al. (2005, 2009). Secure archaeological contexts were established through close consultations with site excavators and by examination of excavation reports. This identified contexts containing suitable marine (generally mollusc shell or fish bone) and terrestrial entities (roundwood charcoal, charred grains, herbivore bones etc.) which had been relatively unaffected by post-depositional disturbance (e.g. Ascough et al., 2007a, 2009) and which were likely to have been deposited at the same time, suggesting a similar calendar age for both sample types. The methodology advocated the collection of at least 4 suitable marine and 4 suitable terrestrial entities per archaeological context to allow the resulting ages to be tested for contemporaneity. Detection of anomalous age measurements (or outliers) is difficult in very small sets of dates, and we have employed a manual approach (Bronk Ramsay, 2009), informed by a simple chi-squared (χ^2) test of the marine and terrestrial data to demonstrate that the ages are indicative of a single deposition time (within statistical limits). Thus, unrounded radiocarbon ages and their associated errors were χ^2 tested for contemporaneity before calculating ΔR values. The χ^2 test determines whether each sample within a group is statistically indistinguishable at 95% confidence from the remainder and therefore can be considered contemporary. Only samples which pass the χ^2 test are then used to calculate ΔR. The critical value for the χ^2 test differs according to the number of measurements within a group and this value is compared to the *T*-statistic calculated for each group to determine whether the samples are statistically indistinguishable (Ward and Wilson, 1978). The calculation of the *T*-statistic is shown in Eq. (1).

$$T = \sum \frac{(t_i - t)^2}{\sigma_i^2}$$

where: t = the weighted mean of the ^{14}C age group
t_i = the individual ^{14}C measurement
σ_i = the error on the individual measurement

Eq. (1): *T*-statistic calculation.

^{14}C ages that pass the χ^2 test are then used to calculate ΔR. In cases where samples do not pass the χ^2 test, a judgement call has to be made on whether or not the samples from this context are in fact suitable for determining a ΔR value. By using every possible pairing when all samples pass the χ^2 test, 16 estimates of ΔR can be calculated for a context from which the 4 terrestrial and 4 marine entities were selected.

Our approach is closely related to bootstrapping which is a statistical procedure to estimate a parameter associated with a population which may be too difficult or expensive to measure directly. In a similar manner, Jones et al. (2007b) approached the same problem of calculating ΔR in archeological contexts by applying Bayesian analysis, solved using a MCMC approach. Bootstrapping is a nonparametric re-sampling method, not dependent on distributional assumptions, which in this context allows us to estimate the population variance and hence the standard error on delta R, based on a relatively small set of ^{14}C measurements. Our use of a resampling technique is to ensure that we have an appropriate and realistic estimate of the population variance. We sample independently and with replacement from the terrestrial and marine samples.

The spread of ΔR values for each site/context can be fully represented using histograms alongside a weighted mean and the standard error for predicted values. The standard error for predicted values gives the best indication of where future values from

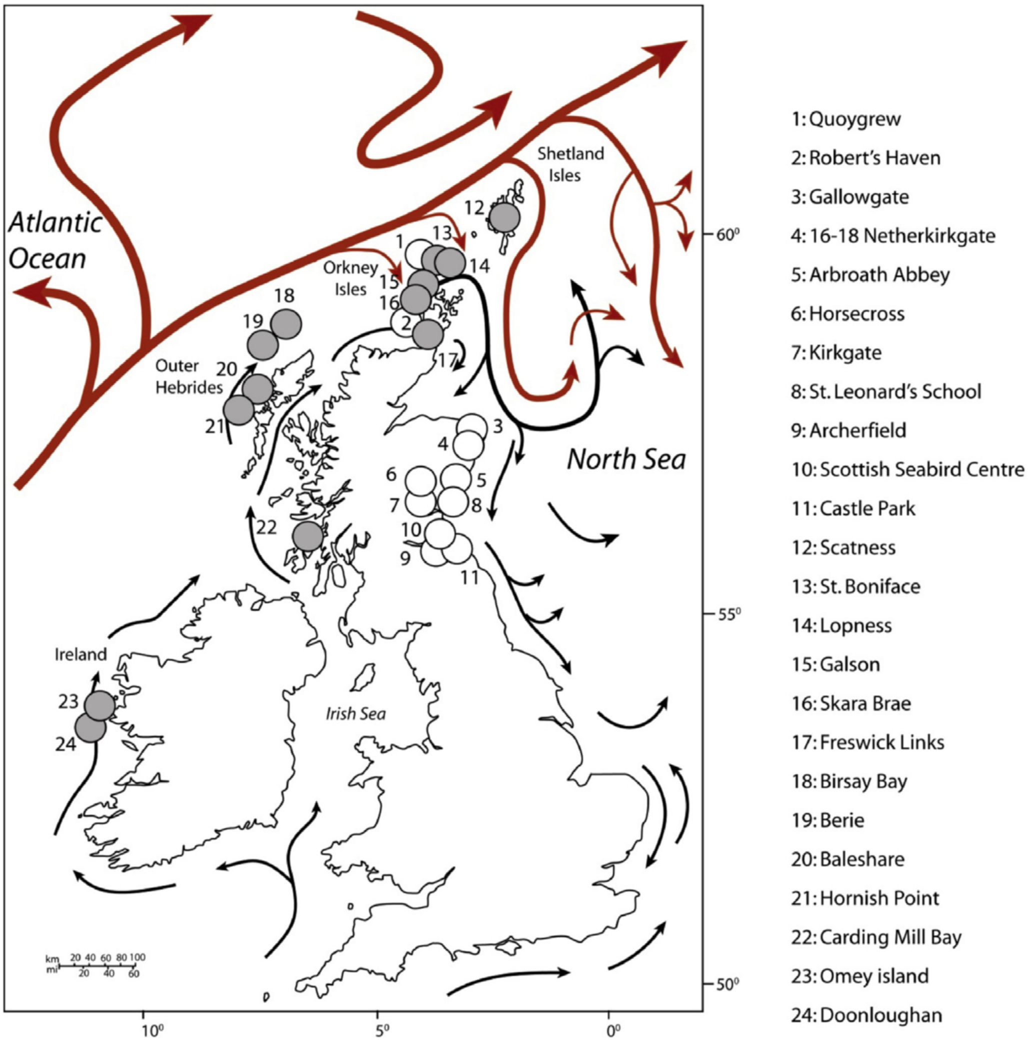

Fig. 2. Location of study sites: open circles (sites 1–11) from Russell et al. (2010, 2011a, 2011b) and Russell 2011; grey circles (sites 12–24) from Ascough et al. (2004, 2006, 2007a, 2009)). Main Atlantic Current is shown in red with coastal currents marked in black.

a similar time and place may lie, using the root sum of squares of the standard deviation and the error on the mean for each group. The benefits of publishing according to this protocol and the potential impact it could have on interpreting ΔR values is discussed in detail by Russell et al. (2011a). This method provides the most robust way of interpreting ΔR values in relation to one another and of statistically addressing the inherent variability within the calculation of ΔR values, and their subsequent use in oceanographic and archaeological interpretation.

3. Results

This paper does not discuss in detail the production of the ^{14}C ages and $\delta^{13}C$ values for each sample used in the study, the references in Table 1 provide all of this supplementary information. In summary, the measured $\delta^{13}C$ values of the terrestrial mammal bones used within this study (−19.4‰ to −23.2‰) fall within the typical range for animals existing on purely terrestrial dietary resources in C3-plant dominated environments (e.g. DeNiro and Epstein, 1978; Chisholm et al. 1982, Post, 2002; Peterson and Fry, 1987, Schoeninger and DeNiro, 1984). A significant marine signal within the mammal's diet would be reflected in a higher $\delta^{13}C$ value, which would have resulted in the sample being rejected on the basis of it being unrepresentative of wholly terrestrial material. The measured $\delta^{13}C$ values of the carbonized cereal grains ranged from −20.2‰ to −27.0‰, representative of a C3 photosynthetic pathway (Craig, 1953; O'Leary, 1981). The $\delta^{13}C$ values for the shells ranged from −2.1 to +2.9, within the accepted range for marine carbonate (Rounick and Winterbourn 1986).

Preliminary studies were undertaken by Ascough et al. (2005b) and Russell et al. (2010) to investigate whether any significant freshwater signals were present at the sites close to estuaries/sea lochs using $\delta^{18}O$ measurements on the mollusc shell samples. This would resolve whether the values for these sites represented a true MRE, or a mixed marine/freshwater offset. The authors concluded that none of the shells were formed in an environment with a significant freshwater input, and therefore the published ΔR values are representative of a true MRE. Also, no correlation could be observed between the variability in the ΔR values and the geographical distribution of the sites.

The radiocarbon ages within each terrestrial/marine group at each site/context were tested for contemporaneity using the χ^2 test. In a few contexts, samples had to be excluded from the χ^2-test as a result of their large contributions to the T value. Where exclusions

Table 1
MRE and ΔR results and corresponding time periods for each site, calibrated using IntCal 13 (Reimer et al., 2013). Contexts in bold are those that failed the X^2 test for comparability of ΔR values.

Site number	Site name	Reference(s)	Grid ref	MRE ± std error for predicted values.	ΔR ± std error for predicted values.	Mean terrestrial age (BP) ± 1σ	2σ calibrated age range
1	Quoygrew A004 Shell	Russell et al., 2011b, Ascough et al., 2009	HY 443 506	276 ± 46	−105 ± 35	941 ± 45	1017–1204 AD
1	Quoygrew A004 Fish	Russell et al., 2011b, Ascough et al., 2009	HY 443 506	286 ± 51	−97 ± 41	941 ± 45	1017–1204 AD
1	Quoygrew A023 Shell	Russell et al., 2011b, Ascough et al., 2009	HY 443 506	327 ± 60	−60 ± 61	902 ± 54	1023–1242 AD
1	Quoygrew A023 Fish	Russell et al., 2011b, Ascough et al., 2009	HY 443 506	373 ± 54	−1 ± 58	902 ± 54	1023–1242 AD
2	Robert's Haven 3019 Shell	Russell et al., 2011b, Ascough et al., 2009	ND 3903 7353	316 ± 39	−57 ± 47	885 ± 36	1039–1220 AD
2	Robert's Haven 3019 Fish	Russell et al., 2011b, Ascough et al., 2009	ND 3903 7353	389 ± 47	18 ± 53	885 ± 36	1039–1220 AD
2	Robert's Haven 3004 Shell	Russell et al., 2011b, Ascough et al., 2009	ND 3903 7353	435 ± 50	32 ± 46	645 ± 24	1284–1393 AD
3	Gallowgate Middle School	Russell et al., 2010	NJ 9421 0659	315 ± 41	−59 ± 48	892 ± 41	1033–1220 AD
4	16 − 18 Netherkirkgate	Russell et al., 2010	NJ 9428 0637	313 ± 44	−96 ± 44	939 ± 41	1020–1185 AD
5	Arbroath Abbey	Russell et al., 2010	NO 642 413	423 ± 58	22 ± 46	641 ± 43	1280–1401 AD
6	Horse Cross	Russell et al., 2010	NO 1187 2388	427 ± 45	12 ± 32	611 ± 45	1288–1410 AD
7	Kirkgate 400	Russell 2011	NO 1196 2360	394 ± 55	−8 ± 56	740 ± 55	1168–1389 AD
7	Kirkgate, 413	Russell et al., 2010	NO 1196 2360	415 ± 54	8 ± 51	641 ± 30	1282–1396 AD
9	Archerfield, 90	Russell et al., 2010	NT 509 841	394 ± 46	−33 ± 43	492 ± 22	1410–1445 AD
9	Archerfield, 142	Russell et al., 2010	NT 509 841	292 ± 50	−130 ± 48	520 ± 43	1310–1450 AD
10	Scottish Seabird Centre 1226 212261226	Russell 2011	NT 55422 85627	442 ± 50	40 ± 47	1322 ± 41	646–771 AD
10	Scottish Seabird Centre 1287	Russell 2011	NT 55422 85627	363 ± 54	−20 ± 40	1469 ± 43	435–656 AD
11	Castle Park, Dunbar 0341	Russell et al., 2010	NT 6776 7917	401 ± 43	4 ± 44	1326 ± 39	646–770 AD
11	Castle Park, Dunbar 3017	Russell et al., 2010	NT 6776 7917	359 ± 42	2 ± 38	1094 ± 40	779–1024 AD
12	Scatness 206	Ascough et al., 2009	HU 3898 1065	409 ± 67	19 ± 58	781 ± 55	1054–1379 AD
12	Scatness 1269	Ascough et al., 2009	HU 3898 1065	276 ± 64	−123 ± 62	1312 ± 44	640–801 AD
13	St Boniface 1063	Ascough et al., 2006	HY 4877 5271	298 ± 32	−102 ± 32	958 ± 29	1021–1155 AD
13	St Boniface 2044	Ascough et al., 2004; Ascough, 2005	HY 4877 5271	340 ± 78	−54 ± 20	2086 ± 16	166–51 BC
13	St Boniface 2136	Ascough et al., 2004; Ascough, 2005	HY 4877 5271	268 ± 57	−56 ± 56	2061 ± 41	190– 24 BC
14	Lopness	Ascough et al., 2007a,b	HY 75840 43960	229 ± 41	−103 ± 39	3700 ± 24	2196–2023 BC
15	Galson	Ascough et al., 2009	NB 4364 5943	285 ± 40	−89 ± 40	1102 ± 30	886–1014 AD
16	Skara Brae 26	Ascough et al., 2007a,b	HY 23125 18745	312 ± 67	−23 ± 72	4103 ± 59	2877–2493 BC
16	Skara Brae 68	Ascough et al., 2007a,b	HY 23125 18745	425 ± 40	24 ± 62	4551 ± 36	3370–3102 BC
18	Birsay Bay	Ascough et al., 2007a,b	HY 2466 2807	316 ± 42	−12 ± 41	3648 ± 26	2133–1941 BC
19	Berie	Ascough, 2005	NB 10348 35171	310 ± 54	−30 ± 69	1662 ± 55	251–536 AD
20	Baleshare 39	Ascough et al., 2004; Ascough, 2005	NF 7763 6157	241 ± 45	−99 ± 46	2013 ± 47	164 BC − 77 AD
20	Baleshare 139	Ascough et al., 2004; Ascough, 2005	NF 7763 6157	271 ± 40	−95 ± 44	2254 ± 29	395–208 BC
20	Baleshare 146	Ascough et al., 2004; Ascough, 2005	NF 7763 6157	260 ± 58	−68 ± 71	2109 ± 58	263 BC − 16 AD
21	Hornish Point	Ascough et al., 2004; Ascough, 2005	NF 758 470	241 ± 26	−101 ± 38	2134 ± 19	357 BC − 16 AD
23	Omey Island	Ascough et al., 2006	L 562 566	241 ± 60	−142 ± 61	991 ± 45	975–1160 AD
24	Doonloughan DL3 F19	Ascough et al., 2009	L 580 459	295 ± 63	−109 ± 69	1265 ± 54	661–882 AD
24	Doonloughan DL11 F2	Ascough et al., 2009	L 580 459	303 ± 52	−80 ± 53	1265 ± 47	663–876 AD
12	**Scatness 543**	Ascough, 2005	**HU 3898 1065**	59 ± 40	**−320 ± 35**	**1705 ± 29**	**252–401 AD**
22	**Carding Mill Bay**	Ascough et al., 2007a,b	**NM 847 294**	531 ± 26	**150 ± 28**	**4783 ± 27**	**3641–3521 BC**
2	**Robert's Haven 3004 Fish**	Russell et al., 2011b, Ascough et al., 2009	**ND 3903 7353**	512 ± 38	**105 ± 34**	**645 ± 24**	**1284–1393 AD**
17	**Freswick Links**	Ascough et al., 2009	**ND 3760 6762**	282 ± 40	**−168 ± 41**	**928 ± 23**	**1033–1159 AD**
8	**St Leonard's School (St Andrews)**	Russell et al., 2010	**NO 51266 16634**	207 ± 36	**−171 ± 47**	**1245 ± 28**	**681–873 AD**

were made, the data were carefully scrutinized to ensure that they were not subject to excessive rejection to allow the remaining samples to pass. If a context contained a large proportion of samples that were not considered contemporary (by failing the χ^2-test), the likelihood of post-depositional disturbance increased, thereby reducing confidence in the security of the context and therefore the validity of any MRE/ΔR that was calculated. It was deemed acceptable to exclude 1 sample from each group of 4 in order for the remainder to pass the χ^2-test and still avoid the risk of calculating an MRE/ΔR based on non-contemporaneous samples. Sites that did not produce suitable results were excluded from the study, owing to archaeological misidentification of the samples as contemporary marine and terrestrial entities. The sites which did pass the χ^2-test were then used to calculate ΔR.

ΔR and MRE values for each context were calculated and the ΔR values displayed as histograms (Supplementary data) showing the spread of values produced by the multiple pairings of marine and terrestrial ^{14}C ages, together with the mean ΔR and standard error for predicted values. This gives a realistic indication of where ΔR values from a similar site and time may lie. The weighted mean ΔR and MRE values for each site/context and their associated errors are provided in Table 1 and illustrated in Fig. 3. These MRE and ΔR values are calculated from sites that range from 492 to 4551 ^{14}C years BP and demonstrate no temporal relationship within the data.

The mean ΔR values for each context range from −320 ± 35 to 150 ± 28^{14}C years and when χ^2 tested as a complete group, fail the test for comparability as T = 193.4245 ($X^2{:}_{005} = 56.942$). When 5 sites are excluded, (Scatness 543, Carding Mill Bay, Robert's Haven 3004 fish, Freswick Links and St Leonard's School), the remaining 37 ΔR values pass (T = 47.271 ($X^2{:}_{005} = 50.998$)). The values which pass the χ^2 test range from −142 ± 61 to 40 ± 47 ^{14}C years and are considered statistically indistinguishable from one another at this level of confidence.

From the results, ΔR values for the northern UK appear to have been relatively stable with little or no temporal or spatial variation over the period represented in the study. On the basis of the δ^{18}O measurements, no freshwater effect is evident and no variability in the MRE or ΔR values can be attributed to estuarine locations. The MRE and ΔR values therefore display no temporal or spatial trends.

A key message from this study reinforces the findings of Russell

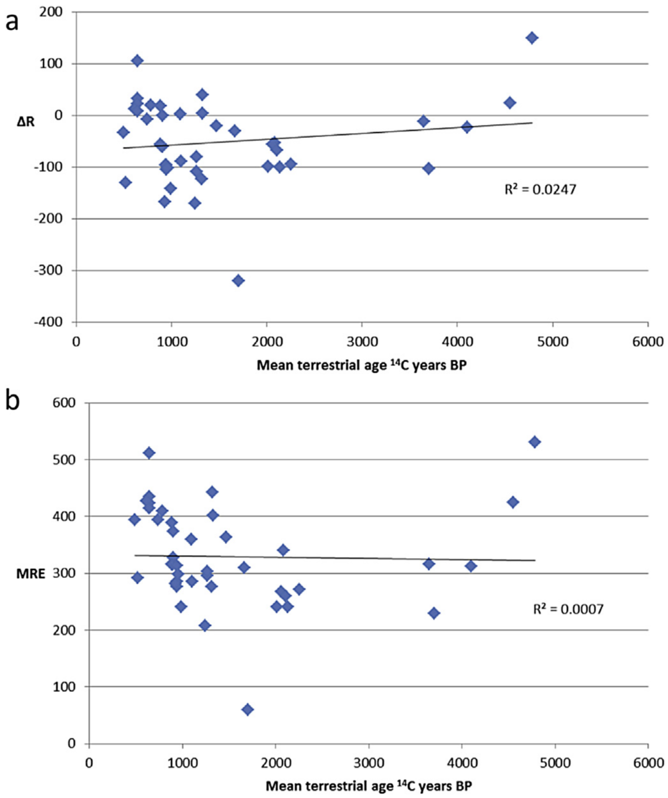

Fig. 3. a. ΔR (^{14}C years) vs mean terrestrial age per context: No linear correlation of changes in ΔR over time. b. MRE (^{14}C years) vs mean terrestrial age per context: No linear correlation of changes in MRE over time.

et al. (2011a); combining multiple pairs of statistically indistinguishable radiocarbon dates in a matrix-style approach can produce variability in the subsequent ΔR values in excess of 200 ^{14}C years. From the sites discussed in this paper, Doonloughan DL3 F319 displays a maximum spread of 225 ^{14}C years, (Fig. 4) and it is this variability which is critical to the justification of whether ΔR can be used as a climatic proxy or not.

Russell et al. (2011a) have already discussed the variability in ΔR values calculated using this method, and concluded that this variability represents uncertainties inherent within the production and calculation of ΔR values and not as a result of oceanographic/climatic changes influencing the ^{14}C activity of the local surface waters. They further concluded that variability in ΔR values of less than 200 ^{14}C years cannot be reliably used as a climatic proxy. This conclusion was reached due to simple reasoning. The samples from which the ΔR values have been calculated were formed at the same time and place and therefore represent concurrent atmospheric and marine signals. If the signals for an individual sample within an atmospheric or marine group were sufficiently enhanced or depleted enough to suggest variability in the source of ^{14}C, the ages would be offset and would consequently fail the X^2 test for compatibility. If ^{14}C ages that are statistically indistinguishable from one another are producing ΔR values with variability of over 200 ^{14}C years, then this variability cannot be as a result of the ages themselves (and therefore source ^{14}C) but must be introduced during the production of the ΔR values in the next step of the calculation. This next step involves a box model which is used to model equivalent marine ages from ages based on terrestrial material (Reimer et al., 2013) and it is uncertainties inherent within this model which have to be attributed as the source of the variable outputs in the ΔR values produced. Investigation of the uncertainties in the model was outside the scope of this study but is highlighted for further research. The source of variation is not necessarily the focus of this research, instead, we wish to raise awareness about how to interpret ΔR data and distinguish whether values are significantly different from one another before making climatic and oceanic inferences.

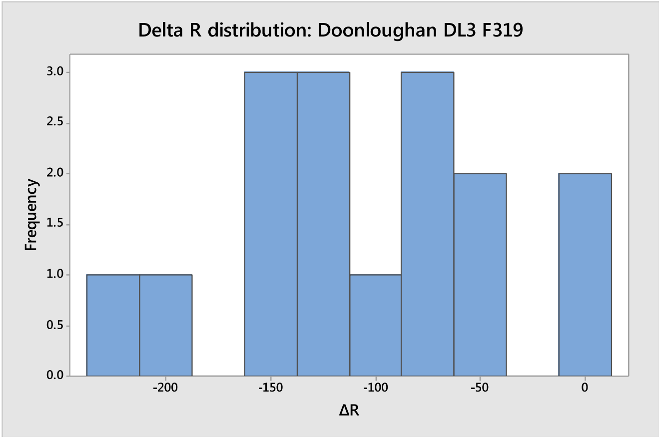

Fig. 4. Spread of ΔR values produced from statistically indistinguishable dates at Doonloughan DL3 F319.

4. Discussion

This study advocates the publication of ΔR values as histograms so that the full range of data for each site is visible and can be interpreted accordingly. This is a large volume of data to make available and in most cases, a mean value is required for the purposes of calibration/ease of publication. Where a mean value is used, we stress that an appropriate error such as the standard error for predicted values should be used. Using a larger error such as the standard error for predicted values alongside the mean ΔR value, may not be desirable (it will increase the calibrated calendar age ranges of marine based samples), but it will offer a more realistic estimate of the range in which future calculations of ΔR values for these sites may lie. This is important when considering that ΔR values are often used as proxy indicators for ocean ^{14}C activity and shifts in oceanic regimes that may force such a change (e.g. Kennett et al., 1997; Kovanen and Easterbrook, 2002). For example, Jones et al. (2007a) document intra-shell variability in ΔR of up to 216 ^{14}C years, which they interpret in relation to El Niño events. This study has shown that the spread of ΔR values from statistically indistinguishable ^{14}C dates can range up to 225 ^{14}C years (Doonloughan DL3 F319). It is therefore entirely possible that variations of 216 ^{14}C years could represent similar variability within the calculation of ΔR values, and not true variability in ^{14}C activity related to oceanographic or climatic changes. Caution should therefore be placed on interpretations drawn from single pairs of ^{14}C ages used to calculate a ΔR value, as any variability in the region of 200 ^{14}C years may be inherent within the calculation method (which is only visible using the multiple paired sample approach) and not representative of oceanographic/climatic changes influencing local ^{14}C activity in surface waters. It could appear, rather unfortunately, that the variability in ΔR may therefore even have the ability to *mask* known climatic changes. The Medieval Warm Period (900–1300AD) followed by the Little Ice Age (1350–1850AD) are two examples of well documented climatic shifts in UK temperatures. No significant differences in the ΔR record appear in the data presented here from these two periods in time (Table 1). A cautionary approach to using ΔR as a climatic tool should therefore be employed with a caveat of suggesting that, on the basis of the present study, ΔR cannot be used as a proxy for oceanographic and or climatic reconstruction unless the shifts are significantly larger than 200 ^{14}C years.

The principle of the proposed approach of resampling is to ensure that we have a realistic estimate of the population variance, which is the key to determining the uncertainty in delta R. The steps taken are:

1. Resample from the individual marine and terrestrial ages, and then compute the differences, a specified number of times.
2. Calculate the mean ΔR and the standard deviation from each resample.
3. Find the standard error of the mean of the group of ΔR values.
4. Calculate the prediction uncertainty by propagating both the standard error of the mean and the population variance.

Despite the inherent variability, this study provides a suite of ΔR values from across the northern UK that will facilitate accurate calibration of radiocarbon ages for samples containing marine derived carbon. A ΔR value chosen for calibration should be as close as possible in time and space to the site which is to be dated in order to achieve a representative estimate of the local MRE at that time. 42 new ΔR values are presented in this paper, which cover a large proportion of coastal Northern British Isles and a vast period of human occupation therein. The data in Table 1 should provide sufficient temporal and spatial information for an appropriate ΔR value to be selected for the majority of calibrations on archaeological, marine derived carbon. This is a critical factor for Scottish archaeology because, owing to our island location, many past communities have typically exploited a large coastal resource base. Consequently, marine-derived material makes a considerable contribution to the national archaeological assemblages and if ^{14}C dating has to rely on marine-derived material from any of these sites, it is of paramount importance to ensure good chronological control.

The range of data, from −142 ± 61 to 40 ± 47 ^{14}C years, presented in this study shows good agreement relative to previously published ΔR values for this region. Reimer et al. (2002) quote a value of −33 ± 93 ^{14}C years for the area encompassing western Ireland, Scotland and the Orkney Islands during the mid to late Holocene (4185–368 BP) whilst Cage et al. (2006) published a value of −26 ± 14 ^{14}C years on samples dating back to 1850 AD from fjordic and coastal waters in north-west Scotland. If mean values were to be presented in this study alongside the site specific values given in Table 1, enough justification would be present to produce values for the east coast, west coast and an overall mean value for Scotland. By removing outliers which do not pass the X^2 tests and removing the sites from Ireland, we propose a weighted mean ΔR value for the west coast of Scotland of −68 ± 90^{14}C years and a weighted mean of −29 ± 53^{14}C years for the east coast. An overall weighted mean value for Scotland from the Neolithic to the Medieval would be −47 ± 52^{14}C years. Calculating a mean value and comparing it with that derived by Reimer et al. (2002), Cage et al. (2006) or Ascough et al. (2004) can only be justified if the presence of definitive temporal or spatial variations in ΔR values are considered to be absent or statistically indistinguishable at the level of confidence at which the ΔR values are reported. This is the case for Reimer et al. (2002) where confidence in a time dependency for ΔR was lacking and thus justified the publication of a mean ΔR value ± the standard deviation on the dataset. A similar case is presented for this study whereby neither spatial nor temporal patterning in the data appears to be present and therefore justifies the publication of a mean value for the dataset ± one standard error for predicted values.

The five sites for which the ΔR values were excluded will be investigated further to determine whether they are genuinely outliers and due to excursions in ^{14}C activity that can be related to climatic/oceanic current changes or whether, again, the values are a product of uncertainties within the modelling that have yet to be understood.

5. Conclusions

Between the Neolithic and Medieval periods, ΔR (and MRE) values for the UK appear to be relatively stable with little or no temporal or spatial variation. However, variability is noted in the spread of ΔR values that can be produced from statistically indistinguishable groups of terrestrial and marine radiocarbon ages. This variability is partly derived from the process of calculating ΔR that uses the box model to produce modelled equivalent marine ages from ages based on terrestrial material. An investigation of the uncertainties in the model was outside the scope of this study but is highlighted for further research. Similar variability is evident in the spread of mean values for the whole region, even for those that pass a χ^2 test for comparability. 42 ΔR values are presented here, which will allow more accurate calibration of ^{14}C age measurements made on archaeological samples from the Northern British Isles containing marine derived carbon.

37 ΔR values from the 42 are statistically indistinguishable from one another. 34 of these values are from Scottish archaeological sites and can be combined to produce a mean value for Scotland of −47 ± 52^{14}C years, applicable from 3500 BC to 1450 AD. This mean value should only be used where site specific data are unavailable for the calibration of marine derived carbon. Many publications on MRE draw interpretations from single pairs of radiocarbon ages used to calculate a ΔR value, and then infer that large apparent shifts in ΔR are as a result of large-scale oceanographic or climatic changes. This study has shown that combining multiple pairs of radiocarbon ages that are statistically indistinguishable in a matrix-style approach can produce variability in the subsequent ΔR values of up to 225 ^{14}C years. This variability represents uncertainties inherent within the production and calculation of ΔR values, not as a result of oceanographic/climatic changes influencing the ^{14}C activity of the local surface waters. A cautionary tale therefore exists regarding the use of ΔR values as a climate proxy.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.quageo.2015.08.001.

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