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Marine Radiocarbon Reservoir Effects for the Mesolithic and Medieval Periods in the Western Isles of Scotland

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Abstract:

We present new values for the marine radiocarbon reservoir effect (MRE) for the west coast of Scotland during the latest Mesolithic at 4540-4240 BC (6490-6190 BP), and the later medieval period at AD 1460-1630 (490-320 BP). This gives a ΔR of -126 ± 39 ^{14}C years for the latest Mesolithic, and a ΔR of -130 ± 36 ^{14}C years for the later medieval. We recalculate previously published MRE values for the earlier Holocene period in this region, at 6480-6290 BC (8430-8180 BP). Here, MRE values are slightly elevated, with a ΔR of 64 ± 41 ^{14}C years, possibly relating to the 8200 BP cold event. New values for the late Mesolithic and medieval indicate lower MRE values, broadly consistent with an extant dataset of 37 assessments for the mid to late Holocene for Scottish coastal waters, indicating stable ocean conditions. We compared the results of the intercept and Probability Density Function (PDF) methods for assessing ΔR. The ΔR values are indistinguishable, but confidence intervals are slightly larger with the PDF method. We therefore apply this more conservative method to calculate ΔR. The MRE values presented fill important gaps in understanding radiocarbon dynamics in Scottish waters, and are discussed in context with previous data. They will provide confidence when calibrating material from critical periods in Scotland's prehistory, particularly for the Mesolithic, when the use of marine resources by coastal populations was high.

1. **Introduction**

The North Atlantic region has a very rich archaeological and palaeoecological record in which marine resources feature prominently; these sample types are almost ubiquitous in, for example, coastal middens, where they can be essential materials for radiocarbon (^{14}C) dating. Marine artefacts and ecofacts on archaeological sites arise via significant use and consumption of marine resources at particular time periods, by prehistoric and historic communities across the North Atlantic. Specific examples include Mesolithic societies along the Atlantic coast of Europe (Richards and Hedges, 1999, Noe-Nygaard 1988, Lubell *et al.* 1994), at the Mesolithic-Neolithic transition in the UK (Schulting and Richards, 2002; Montgomery et al., 2013), and during the Viking period on the North Atlantic islands (including Scotland, Faroes, Iceland and Greenland; Barrett et al., 2001; Ascough et al., 2006; 2012; Arneborg et al., 1999; 2012). Stable isotopes of carbon and nitrogen (δ^{13}C and δ^{15}N) have been used in these studies to demonstrate the incorporation of significant amounts of marine material in human diets, according well with the archaeology of these time periods, in which fishing vessels, equipment for fish and shellfish collection and processing, and other material remains of these activities are found. One location in which marine resources were used almost continuously during the Holocene is Scotland, with its extensive coastline and island archipelagos to the west and north. Scottish archaeology represents a very detailed record of North Atlantic communities over the past c.10,000 years and is important for its position at the interface between Europe and the North Atlantic region, making it key to our understanding of factors such as cultural adaptation to climatic and environmental changes in marginal environments, human-environment interactions, and trade and exchange over extended distances.

In order to understand the chronology of events in Scottish archaeology, ^{14}C dating is crucial for building absolute chronologies within the archaeological and palaeoenvironmental sciences. However, the use of marine resources introduces the need to correct ^{14}C dates for the marine reservoir effect. A reservoir effect occurs when the carbon within one of Earth's carbon reservoirs (i.e. the terrestrial biosphere, marine or freshwater hydrospheres, or the cryosphere) has a lower ^{14}C activity (and hence an older 'apparent' ^{14}C age) than carbon in the atmosphere. This can occur if ancient carbon, (e.g. carbon from carbonate rocks such as limestone) enters the reservoir, or if carbon undergoes 'aging' within the reservoir as a result of time spent in that reservoir without exchange. As global circulation of $^{14}CO_2$ in the atmosphere is rapid, being on the order of 5-10 years (Levin and Hesshaimer, 2000), and uptake of $^{14}CO_2$ by plants and subsequent transfer through the food chain is equally rapid (Nydal, 1968), terrestrial environments do not typically have a reservoir effect, with the exception of material in close proximity (< 1 km) to volcanic CO_2 sources (Bruns et al., 1980). In contrast, the marine reservoir exhibits a substantial ^{14}C reservoir effect, due to the 'aging' of deep water masses when separated from the atmosphere. When these water masses return to the surface they 'dilute' the ^{14}C content of the surface ocean, and this dilution is passed to organisms inhabiting the marine reservoir (e.g. fish and molluscs). Importantly, the reservoir effect is also transferred to terrestrial organisms, such as humans, that consume marine resources.

Therefore, radiocarbon dated remains of marine material from archaeological sites require correction for the marine reservoir effect (MRE), as do the remains of humans and other omnivores that have demonstrably consumed a significant proportion of marine carbon in their diet. Without correction, samples can appear several hundred years 'too old', leading to incorrect chronologies of events in the archaeological record. For example, uncorrected dates on marine material from wheelhouse sites on the Western Isles of Scotland appear to show that these structures are equivalent in age to the demonstrably earlier architectural form of brochs, yet when corrected for the MRE this discrepancy is removed (Barber 2003; Ascough et al., 2004). Clearly, in order for the resulting ^{14}C dates to be accurate, the MRE correction needs to be appropriate to the individual site, period and samples. The marine calibration curve (currently Marine13 (Reimer et al., 2013)) gives a global average MRE correction that varies with time. However, individual locations around the globe are offset from this average value, where the offset is known as ΔR (Stuiver et al., 1986; Stuiver and Braziunas, 1993). These ΔR values are location-specific and can vary at a single location through time, making their quantification an important issue for ^{14}C dating of marine material. Spatiotemporal variability in MRE and ΔR values can result from several different oceanographic, environmental or climatic factors. These include changes in ocean circulation that bring water masses of varying ^{14}C content to an area, changes in ocean ventilation or stratification that increase or reduce the input of ^{14}C-depleted waters from depth, fluctuations in wind speed, air/water temperature or ice cover affecting ocean uptake of atmospheric ^{14}C, and in estuarine settings, changes in the admixture of fresh (high ^{14}C) and marine (low ^{14}C) waters.

The North Atlantic has been the setting of extensive efforts to quantify regional MRE and ΔR values. Modern ΔR values range from 225 ± 51 ^{14}C yr in Kollafijord, Iceland (Broecker and Olson, 1961), to -119 ± 54 ^{14}C yr at Skelmorlie Bank, Scotland (Harkness, 1983), with a clear geographic gradient from Arctic waters containing proportionally 'older' carbon (i.e. high MRE) in the north, to Atlantic waters with lower MRE values further south, a trend that appears to have been in existence through at least the last 1000 years (Ascough et al., 2006). In waters surrounding Scotland, modern values of ΔR range from -119 ± 54 ^{14}C yr to +94 ± 30 ^{14}C yr (Harkness, 1983), while non-modern values have been measured at -123 ± 62 ^{14}C yr to +143 ± 20 ^{14}C yr (Ascough et al., 2007; Russell et al., 2015) during the Holocene. In the pre-Holocene North Atlantic there are large shifts to higher MRE values on the order of several hundred years during the Younger Dryas (i.e. c. 11,000 yr BP (Austin et al., 1995)), and shifts on the order of 1000 years during the Last Glacial (Skinner and Shackleton, 2004). Recent work by Russell et al. (2015) failed to detect significant shifts in ΔR in Scottish coastal waters over the latter half of the Holocene, although five outliers from this trend were detected. This work was based upon multiple paired sampling and a statistical approach that involves ‘bootstrapping’ to determine the likelihood that repeat measurements would give the same ΔR for a location if different samples were selected. The approach involves taking multiple samples of terrestrial and marine material, and for every possible terrestrial-marine sample pairing, calculating a ΔR value. The weighted mean of these values is taken as the overall ΔR for a context, and the uncertainty on this weighted mean is obtained by combining the standard error of the weighted mean with the standard deviation of all calculated ΔR values (i.e. the standard error for predicted values). In conclusion, Russell et al. (2015) recommended using a ΔR value of -47 ± 52 ^{14}C years for the period 3500 BC to AD 1450 in Scottish coastal environments if no further information for a specific site and time period is available. This value overlaps with a previous determination for the subpolar eastern North Atlantic (including Scotland and Ireland), for the mid- to late-Holocene by Reimer et al. (2002), which was -33 ± 93 ^{14}C years.

The five outliers from the Russell et al. (2015) dataset include material from the Neolithic period (Carding Mill Bay, 3640-3520 BC) and the Medieval period (Roberts Haven, 1280-1390 AD), both of which are critical for understanding the chronology of Scotland’s archaeology. The Scottish Mesolithic/Neolithic transition c.6k cal BP saw the introduction of organized farming practice for the first time, while the Medieval period saw the expansion of trade routes with Europe and further afield, based upon an emergent fishing industry (Barrett et al., 2008). We therefore sought further information on the MRE in Scotland for these time periods by ^{14}C analysis of paired marine and terrestrial samples from four archaeological sites. We also recalculated ΔR values for two further sites that were not contained within the Russell et al. (2015) paper, but which relate to the Mesolithic periods in Scotland. The aim of this work was therefore to clarify MRE values for important periods in Scottish prehistory to improve archaeological chronologies. In addition, we examined the effect of taphonomic bias upon MRE values, and critically assessed the multiple paired sample approach for MRE and ΔR quantification.

2. **Methods**

2.1. Sample selection

Samples were selected for new quantifications of the MRE from individual stratigraphic contexts at four sites; Context 14 at Northton (NO-14); Context 1 at Tràigh na Beirigh 1 (TNB1-1), Context 5 at Tràigh na Beirigh 2 (TNB2-5); and Context 177/83 at Guinnerso (GUN-177/83). Northton (NGR: NF 9753 9123) is located on the Isle of Harris, Scotland, while Tràigh na Beirigh 1 & 2 (NGR: NB 1002 3628 & NB 1003 3633) and Guinnerso (NGR: NB 0350 3631) are located on the Isle of Lewis, Scotland (Figure 1). The archaeological evidence at Northton consists of a series of Mesolithic ground surfaces with mixed anthropogenic material within the soils that is overlain by machair, a calcareous shell-sand soil unique to the Western Isles of Scotland. The site represents the first archaeological evidence for Mesolithic human occupation in the Western Isles (Gregory et al., 2005) and the samples for this project were taken from the latest Mesolithic layer, immediately under the machair (Bishop et al., 2010). The sites of Tràigh na Beirigh 1 & 2 consist of two open-air Mesolithic shell-middens, again overlain by machair (Church et al., 2012; Bishop et al., 2013). The samples for this project (TNB1-1, TNB2-5) were taken from the main body of the shell middens at both sites. The final samples GUN 177/83) come from the Medieval occupation of a sheiling (a stone hut forming a summer dwelling on a seasonal upland pasture) located in the multi-period landscape at Guinnerso in the moorland of the Uig Peninsula in Lewis (Church & Gilmour, 1998).

The selected sites are exposed to the Atlantic Ocean, away from significant sources of freshwater or carbonate geology, either of which could compromise ^{14}C dates used to quantify the MRE. Selection of contexts followed the processes described in Ascough et al. (2005). Briefly, material was only selected from discreet, sealed contexts of limited spatial extent without visible signs of disturbance. These sites were selected after an initial programme of range-finder ^{14}C dating sponsored by Historic Environment Scotland indicated that NO-14 corresponded to the mid-Mesolithic period, TNB1 -1 and TNB2-5 to the latest Mesolithic period, and GUN-177/83 to the Medieval period. At each site, four paired samples of terrestrial (carbonised plant macrofossils) and marine material (marine mollusc shells) were selected from bulk samples taken for environmental archaeological analysis, using an on-site ‘total’ sampling strategy, following Jones (1991). Bulk samples were processed using a flotation tank (Kenward et al., 1980), with the residue held by a

1.0 mm net and the flot caught by 1.0 and 0.3 mm sieves respectively. All the flots and residues were air-dried and sorted using a low-powered stereo/binocular microscope at x15-x80 magnification. Hazel nutshell were chosen as the terrestrial single-entity samples from the Mesolithic sites, as hazelnuts are short-lived, single-season plant remains and are very common on Mesolithic sites in Scotland (Bishop et al., 2014, 2015). Barley grains were chosen from the Medieval phase at Guinnerso as they too are short-lived, single-season plant remains. Common limpet shells (*Patella vulgate)* were selected from all four sites as the marine sample to which the ^{14}C ages of the hazelnut and barley (terrestrial) samples were compared. The lifespan of the common limpet ranges from ca.5 to ca. 20 years (Lewis and Bowman, 1975), introducing the possibility of inbuilt ages of up to 20 years when using limpets to calculate MRE and ΔR. In this study, shells were inspected to estimate age based upon growth bands where possible, and to select shells <10 years old. Shell morphology was also checked to ensure this was consistent with the faster-growing, shorter-lived individuals at the lower shoreline (Lewis and Bowman, 1975). Any inbuilt age associated with marine shells used in this study will therefore be low, compared to the typical uncertainties associated with MRE and ΔR determinations. Although species-specific MRE and ΔR values for marine mollusc shells have been observed at locations world-wide, these are highly unlikely for the study region. Species-specific effects arise where there are differences in ^{14}C age of resources consumed by molluscs, typically in areas of carbonaceous geology where infaunal feeders will ingest ^{14}C-dead carbon during feeding (c.f. Forman and Polyak, 1997). Species-specific effects can also arise where there are significant differences in ^{14}C age of the water column over small geographical areas, such as estuaries (e.g. Holmquist et al., 2015). Neither of these applies in the study area, and previous work has showed no inter-species variability in mollusc MRE and ΔR values for the region (Ascough et al., 2005).

In addition to new MRE quantification, recalculations of the MRE and ΔR were performed for two sites relating to the early Holocene and Mesolithic period in Scotland; Northton on the Isle of Harris (context NO-5) and Sand on the Scottish mainland (context SA-13) (Figure 1) (Ascough et al., 2007), in order to assess these data in light of the findings presented in Russell et al. (2015). ΔR values and terrestrial calibrated age ranges for these sites were therefore recalculated using the Intcal13 and Marine13 datasets (Reimer et al., 2013), and the standard error for predicted values, outlined in Russell et al. (2011a,b; 2015), was calculated for each ΔR value obtained. This is particularly important for these two sites as they previously gave ΔR values of 64 ± 19 ^{14}C yrs (SA-13) and 79 ± 32 ^{14}C yrs (NO-5) (Ascough et al., 2007). These data were taken to indicate that ΔR values were higher in the early Holocene/ Mesolithic as they related to the periods 6480-6420 BC (SA-13) and 6390-6230 BC (NO-5) (Ascough et al., 2007). By recalculating these data using the standard error for predicted values (Russell et al., 2015), we can assess whether this more robust method of estimating the error on ΔR values still gives values for Scottish waters that are significantly different from those later in the Holocene period.

2.2. Radiocarbon measurement of samples for MRE/ΔR quantification

Carbonized plant macrofossils were pre-treated with a HCl wash to remove carbonates (0.1 M at 80°C for 2 hours), followed by removal of organic acids in 0.1 M NaOH (2 hours at 80°C), then a final HCl wash to remove any CO_2 adsorbed in the base step. The pre-treated macrofossils were converted to CO_2 by combustion in pre-cleaned quartz tubes (Vanderputte et al., 1996). Marine shells were inspected to establish that there was no evidence of carbonate re-precipitation (Mangerud, 1972; Mook and Waterbolk, 1985). Shells were cleaned ultrasonically and by abrasion to remove surface contaminants, and then etched in 1 M HCl to remove the outer 20% of the shell. The whole shell was then crushed and a 0.1g aliquot was hydrolysed with 1 M HCl under vacuum. CO_2 from plant or shell samples was purified cryogenically using solid CO_2/ethanol and liquid N_2 traps. 3 ml aliquots of the purified CO_2 were converted to graphite by the method of Slota et al. (1987), and sample $^{14}C/^{13}C$ ratios were measured by accelerator mass spectrometry (AMS). $\delta^{13}C$ values (as per mil (‰) deviations from the VPDB international standard) were measured on CO_2 from all samples using a VG SIRA 10 with NBS 22 (oil) and 19 (marble) as internal standards. The full methodology is given in Dunbar et al. (2016).

2.3. Consistency of ^{14}C measurements within sample groups

The groups of measured terrestrial and marine ^{14}C ages for the individual contexts were tested for internal consistency using the chi-squared (χ^2) test (c.f. Ward and Wilson, 1978). The test establishes whether a group of ^{14}C ages can be considered to be contemporaneous by comparing the variability within a measurement group with the errors on individual measurements. Measurement variability is considered to exceed that occurring by chance (i.e. χ^2 test fail) if the χ^2 test value (T) for a group of ^{14}C ages exceeds the T-statistic for 95% confidence of N ^{14}C age measurements (χ^2: $_{0.05}$ T). If a group of samples failed the χ^2 test, the measurements were scrutinized to establish the source of the variation. Where the χ^2 test fail was due to a single measurement, this measurement was excluded from the sample group, and the remaining consistent ^{14}C measurements used to calculate ΔR. In instances where the χ^2 test fail was due to multiple measurements, the ^{14}C dating of the context was repeated where possible, using additional samples (c.f. Ascough et al., 2007).

2.4. Calculation of ΔR values

For each context, multiple values of ΔR were calculated using samples that passed the χ^2 tests. Two slightly different methods of calculating ΔR exist, therefore we performed a sensitivity test, comparing the results obtained with both methods to check for any significant differences. The first method involves converting individual terrestrial ^{14}C ages to modeled marine ^{14}C ages using an interpolation of the IntCal13 and Marine13 datasets (Reimer et al., 2013). The conversions incorporate the uncertainty in the interpolated calibration curve data. The ΔR for each pairing of terrestrial and marine ^{14}C ages is the difference between the midpoint of the modeled marine ^{14}C age boundaries and the measured marine ^{14}C age. The 1σ error on individual ΔR values was calculated by propagation of the errors on the terrestrial and marine ^{14}C ages.

The second method differs slightly in that it incorporates the probability density function (PDF) of the marine calibration curve when obtaining ΔR (Reimer and Reimer, in prep). The individual terrestrial ^{14}C ages are calibrated using the IntCal13 calibration curve. This produces a PDF, the discreet points of which are reverse-calibrated using the marine calibration curve. The offset between the radiocarbon dated marine sample and the reverse-calibrated terrestrial sample PDF gives ΔR. To determine the confidence interval of ΔR, a convolution integral is used, approximated as a normal distribution (Reimer and Reimer, 2016).

For both methods, ΔR was calculated for each possible pairing of marine and terrestrial ^{14}C measurements for a context, giving multiple ΔR values for that context. The weighted mean of the ΔR values was then calculated to give an overall ΔR value for that context. The standard error on the weighted mean was evaluated based upon the measurement uncertainties (Equation 1).

$$\sigma_1 = \sqrt{\frac{1}{\sum 1/s_i^2}} \qquad \text{Equation 1}$$

The final 1σ error associated with a weighted mean ΔR for a context was then obtained via the standard error for predicted values. This accounts for any additional variability due to the precise pairing of terrestrial and marine samples used to calculate ΔR.

$$\sigma = \sqrt{(x^2 + y^2)} \qquad \text{Equation 2}$$

Where:

x = the error on the weighted mean

y = the standard deviation on all the ΔR values calculated for a context.

2.5. Terrestrial calibrated age ranges

To calculate a calendar age range that is represented by the material in the deposit (and for which the ΔR values are applicable), the weighted mean of the terrestrial ^{14}C ages that passed the χ^2 test for each context was used. Calibrated ranges at 95% confidence (i.e. 2σ) were obtained using the IntCal13 atmospheric dataset (Reimer et al., 2013), and the OxCal v4.2 calibration program (Bronk Ramsey, 1995; 2001).

3. Results

3.1. New ΔR values for the Mesolithic and Medieval Periods

The δ^{13}C values for carbonised plant macrofossils and marine shells fall within the expected ranges for these sample types (i.e. C_3 vegetation in the northern hemisphere and marine carbonates (Aitken, 1990)). The χ^2 test results for the groups of terrestrial and marine samples for each context are given in Table 1, along with the ^{14}C ages and δ^{13}C values for each sample. The reported χ^2 test results are for groups of samples where the variability in ^{14}C measurements did not exceed the T-value, and results were used in assessment of MRE/ ΔR for that context. Samples that caused the ^{14}C measurements in a group to fail the χ^2 test are indicated; these measurements were excluded from ΔR calculation.

The sensitivity test between the two methods of calculating ΔR showed no significant differences, with a maximum of 12 ^{14}C yr between ΔR values calculated using different methods. The confidence interval of the probability density function (PDF) method is, however, slightly larger than that obtained using the intercept method, therefore we use the PDF method to report ΔR values in the following, as the results are the more conservative of the two methods.

For TNB1-1, the weighted mean ^{14}C age of the terrestrial samples gives a calibrated age range of 4330-4240 BC (6280-6190 BP) at 95% confidence, placing this site at the latest phase of the Mesolithic in Scotland (Ashmore, 2004). For this time period, the calculated MRE is 300 ± 51 ^{14}C yr and the ΔR = -109 ± 55 ^{14}C yr. This ΔR value overlaps with that calculated for TNB2-5, which is -143 ± 54 ^{14}C yr, corresponding to a MRE value of 229 ± 41 ^{14}C yr for the period 4540-4470 BC (6490-6410 BP), in the late Scottish Mesolithic (Ashmore, 2004). For GUN-177/83, the weighted mean terrestrial ^{14}C age corresponds to the Late Medieval period, at AD 1460-1630 (490-320 BP). For this time interval the calculated MRE is 305 ± 24 ^{14}C yr and the $\Delta R = -130 \pm 36$ ^{14}C yr. The group of terrestrial ^{14}C ages for NO-14 are statistically consistent, with a χ^2 value for the group of T= 2.13 (χ^2 :$_{0.05}$ = 7.81), giving a weighted mean age of 7450 ± 17 ^{14}C yr BP, and a calibrated age range of 6390-6250 BC (8330-8200 BP). The group of marine ages are also internally consistent, with a χ^2 value for the group of T= 0.53 (χ^2 :$_{0.05}$ = 7.81). The weighted mean of the terrestrial group of samples is 2361 ^{14}C years older than the group of marine samples. When the MRE is responsible for an age offset between terrestrial and marine samples, the marine material is always older than the terrestrial samples. As the reverse is true in this instance, the age offset between terrestrial and marine samples for NO-14 must be that the two sample types are of different actual ages, and entered the context c. 2000 ^{14}C years apart. It is therefore not possible to calculate a MRE for NO-14 using these samples.

3.2. Recalculation of ΔR values using the standard error for predicted values

Recalculated ΔR and MRE values with the standard error for predicted values for SA-13 and NO-5 are given in Table 2. For SA-13 this gives a MRE of 416 ± 35 ^{14}C yr and a ΔR of 63 ± 49 ^{14}C yr for the period 6480-6420 cal. BC (8430-8370 cal. BP). For NO-5 this gives a MRE of 440 ± 69 ^{14}C yr and a ΔR of 67 ± 78 ^{14}C yr for the period 6390-6290 cal. BC (8340-8180 cal. BP)

4. Discussion

4.1. Archaeological significance of the dating programme

The terrestrial dates from the hazel nutshell and barley carbonised macrofossils from the four sites are important in determining the chronology of the sites excavated. The 4 hazel nutshell dates from Northton (NO-14) have demonstrated that the latest palaeosol in the site sequence is of the same date as the main Mesolithic archaeological phase at the site dating to the 7th millennium BC (Gregory et al. 2005), albeit with some later intrusion from the later Neolithic archaeology in the machair overlying the palaeosol sequence. The hazel nutshell dates from the two open-air shell middens at Tràigh an Beirigh (TNB1-1 & TNB2-5) date the activity at these sites to the 5th millennium BC, furnishing the archaeology of the Western Isles of Scotland with Terminal Mesolithic shell-midden sites for the first time. These open-air shell middens are one of the main site types of the Late Mesolithic in

Scotland and the wider European Atlantic seaboard (Hardy, 2015; Milner et al. 2007) and the lack of these sites in the Western Isles until this point has been viewed an enigmatic problem in North Atlantic archaeology (Edwards, 1996; Hardy, 2015). The barley dates from the Medieval sheiling at Guinnerso (GUN-177/83) also demonstrate the antiquity and importance of transhumance practice in the Western Isles.

4.2. New determinations of MRE and ΔR values for the Mesolithic and Medieval Periods in the Western Isles of Scotland

The results of this study fill important gaps in our knowledge of ^{14}C dynamics in ocean systems surrounding Scotland that relate to the ^{14}C dating of historic and prehistoric communities in the region. For the earliest period covered, the recalculated values for SA-13 and NO-5 relate to the periods 6480-6420 cal. BC, and 6390-6290 cal. BC, respectively, corresponding to the Mesolithic period. There is only a 29 year gap between the two calibrated age ranges, meaning that the ΔR values from these sites both correspond to one of the earliest periods represented in Scottish archaeology. The ΔR values for SA-13 and NO-5 are statistically indistinguishable on the basis of a χ^2 test, and can be combined to give a weighted mean of 64 ± 41 ^{14}C yr. The recalculated ΔR values are equivalent (on the basis of a χ^2 test with *df* = 38) to 37 other values for Scottish coastal waters in the Holocene, presented in Russell et al. (2015). However, as the results from SA-13 and NO-5 give slightly higher MRE/ ΔR values for the earliest period covered, possible factors underlying this can be considered. One possibility is that the age ranges for SA-13 and NO-5 follow the 8.2 Kyr event in palaeoenvironmental records for the North Atlantic region (Alley and Ágústsdóttir, 2005). A proposed mechanism for this event is the catastrophic drainage of two large glacial lakes, Agassiz and Ojibway, into the North Atlantic (Barber et al., 1999). This influx of freshwater may have resulted in a slowdown of the North Atlantic Deepwater (NADW) Conveyor, consequently resulting in colder conditions in the region (Ellison et al., 2006). A NADW slowdown period is thought to be followed by phases of 'older' surface ocean ages, as 'aged' deep waters are returned to the surface (Thiagarajan et al., 2014). Regardless of the mechanism for change in MRE/ ΔR, the use of values from NO-5 and SA-13 are recommended for this time period until further data become available.

For the latest Mesolithic period, values from TNB1-1 and TNB2-5 are statistically equivalent, with a χ^2 value of T= 0.19 ($\chi^2 :_{0.05} = 3.84$). This indicates a ΔR of -126 ± 39 ^{14}C years for the western isles of Scotland during the period 4540-4240 BC. It is important to note that there is a 135 calibrated year hiatus between the upper and lower limits of the two calibrated ranges making up this timespan. Both TNB1-1 and TNB2-5 are statistically indistinguishable from (on the basis of a χ^2 test with *df* = 37) the values presented in Russell et al., (2015). The closest ΔR values in time for this geographic region are obtained from Carding Mill Bay (CMB), which has a lower calibrated age limit of 3641 BC, putting a gap of 596 cal. years between this and the upper limit of TNB1-1. The ΔR for CMB was an outlier from other Holocene values in Russell et al. (2015), being significantly higher (ΔR = 150 ± 28 ^{14}C years for the period 3641-3521 BC). Two previous values for CMB in Reimer et al., (2002) give a ΔR of -44 ± 91 ^{14}C years for the period 3965-3714 BC and ΔR = 86 ± 67 ^{14}C years for 3942-3653 BC. The spread in these determinations is large, although the calibrated age range for the positive ΔR obtained in Reimer et al. (2002) is closest in time to the highly positive ΔR presented in Russell et al. (2015). It is possible that in coastal waters surrounding CMB there were significant fluctuations in ΔR over the time period 3965-3521 BC. Potential mechanisms for these fluctuations include varying proportions of high-^{14}C content Atlantic water reaching the site through time due to oceanographic shifts. If this were the case, other sites in the region would also be expected to show concurrent ΔR changes, however we currently lack these measurements. Overall, a reassessment of data from CMB would be useful in light of these data. For the latest Mesolithic period in the Western Isles of Scotland we therefore recommend using the ΔR values calculated for TNB-1-1 and TNB2-5. This correction would be applicable to marine samples that return ^{14}C ages around 5908 ± 21 years BP to 5697 ± 21 years BP (the weighted means of marine ^{14}C ages for TNB-1-1 and TNB2-5, respectively). Prior to these new ΔR calculations there was a gap of 2647 calendar years for which no values were available. Determinations of accurate MRE/ ΔR values for this period are especially important given the evidence for marine consumption during the Mesolithic in Scotland (Schulting and Richards, 2002) and the debate surrounding whether the use of marine resources continued into the Neolithic (Milner et al., 2004; Montgomery et al., 2013).

Previous data for the medieval period suggested a slightly elevated ΔR relative to the preceding Norse period (Ascough et al., 2009). However, when the standard error for predicted values was applied, these values were not found to be significantly different from other values for Scottish waters during the period 3500 BC- AD 1450 that were used to calculate an average ΔR for this time period of -47 ± 52 ^{14}C years (Russell et al., 2015). The exception to this was values of ΔR for Atlantic cod (*Gadus morhua*) at Roberts Haven (1284-1393 AD), which were higher (105 ± 34 ^{14}C years) (Russell et al., 2011b), which may indicate integration of ΔR values over a wider geographic range, including northern waters, where higher ΔR values are found. For the time period of AD 1457-1632, the ΔR value at GUN-177/83 is -130 ± 36 ^{14}C years, which is consistent (on the basis of a χ^2 test with *df* = 37) with the -47 ± 52 ^{14}C years of Russell et al., (2015). It is worth pointing out here that the T-statistic for this grouping is very close to the critical value (T = 52.000 and χ^2 :$_{0.05}$ = 52.192, respectively) The value of GUN-177/83 can be used for the later medieval period in coastal waters of Scotland, corresponding to a later date than the previously available range of ΔR values available for the Holocene period in Scottish waters.

4.3. Issues of taphonomy in calculation of MRE and ΔR values using archaeological samples

While the ^{14}C ages of samples from NO-14 are internally consistent within the groups of terrestrial and marine material on the basis of a χ^2 test, the two groups of sample ages show a large difference of 2361 ^{14}C years, with the younger samples in this instance (with a weighted mean of 5085 ± 18 ^{14}C years) being the marine shell samples. A negative ΔR value on the order of -1000 ^{14}C years would mean substantially higher ^{14}C content in the oceans than in the atmosphere. While this may be a future prospect in the field of ^{14}C measurement due to the high input of fossil fuels to the atmosphere (Graven, 2015), it is highly unlikely to have been a feature of past environmental systems on the timescale of the ^{14}C method. It is therefore most likely that the discrepancy in ^{14}C ages at NO-14 is due to issues of taphonomy, namely post-depositional disturbance of a context into which younger (marine) material was entrained. The context from which ΔR values were to be determined in this study were carefully selected on the basis of no apparent evidence of such post-depositional mixing, therefore the ^{14}C ages from NO-14 serve as an example of the need for multiple measurements from contexts, not only for determination of ΔR values, but for contexts where dating is critical to archaeological interpretation, and where material returns an anomalous ^{14}C age contrary to expectations. The experience of NO-14 provides a possible explanation for another of the ΔR values in Russell et al., (2015) that did not pass the overall χ^2 test; Scatness, context 543. In this instance the calculated MRE was 59 ± 40 ^{14}C years and ΔR = -320 ± 35 ^{14}C years at AD 252-401. Such an extreme negative ΔR may well be explained by intrusion of younger marine material into a context at a later calendar date than when the terrestrial material was deposited. This emphasises the need for a programme of MRE/ ΔR assessments for a region in order to obtain a correction value that is accurate as well as precise. The issues of taphonomic bias will always be present on archaeological sites, although these can be mitigated by techniques such as the multiple paired sample approach to MRE/ ΔR quantification (c.f. Ascough et al., 2009).

5. Conclusions

We present new determinations of the marine radiocarbon reservoir effect (MRE) for key periods in Scottish history and prehistory. We calculate ΔR based on two different methods, the more commonly used intercept method (c.f. Russell et al., 2015), and the Probability Density Function method (c.f. Reimer and Reimer, in press). The findings were that ΔR values were indistinguishable using the two methods, with a maximum difference of 12 ^{14}C yr, however confidence intervals are slightly larger when using the PDF method, making this the more conservative of the two. We present an interpretation of recalculated values for the earliest period of the Holocene for which MRE values are available. The latter data indicate that in the early Holocene, during the Mesolithic period, MRE/ ΔR values were slightly higher than values obtained for the remainder of the Holocene, with a weighted mean ΔR = 64 ± 41 ^{14}C yr. The new values presented also relate to the latest Mesolithic period in western Scotland, for which no data were previously available. These data suggest a MRE that is slightly higher than values obtained for the remainder of the Holocene, where ΔR = -126 ± 39 ^{14}C yr. These values can be used for calibration of samples where the measured marine ages are in the range 5910 - 5700 ^{14}C yr BP, and which are geographically close to the sampled sites. For the later medieval period, values from the Isle of Lewis indicate a ΔR of -130 ± 36 ^{14}C years for AD 1457-1632. This is consistent with previous ΔR determinations for the period 3500 BC- AD 1450 where a weighted mean ΔR of -47 ± 52 ^{14}C years was determined (Russell et al. 2015). Underlying reasons for the early variations in ΔR that are observed in the data presented here remain elusive, although the 8.2 Kyr cold event and associated flux of freshwater to the surface Atlantic Ocean is a possible explanation for the slightly elevated ΔR values. Finally, the findings of this study strongly emphasise the benefit of a programme of ^{14}C dating, rather than individual, isolated dates, when seeking accurate chronological information for archaeological deposits, particularly when quantifying the marine ^{14}C reservoir effect, in any geographic area, for any time period.

Further research to build upon the results of this study has the potential to yield valuable insight into the dynamics of MRE and ΔR values in the North Atlantic. Despite the wide temporal range of the values, data is lacking for ΔR in several time periods through the Holocene (e.g. 6000-5000 BC). Sites where a wide variability in MRE/ ΔR appears over short timescales (e.g. Carding Mill Bay) warrant more investigation to properly understand this variability. Values from the Iron Age to the Medieval period (i.e. 200 BC-AD 1600) show a variability in ΔR values of between +100 to -200 ^{14}C years. Further research could usefully examine whether this variation is replicated in earlier time periods, in order to improve understanding of the range in values that can be expected for a single geographic area. Finally, future determinations of MRE and ΔR values should focus on periods where large-scale changes in oceanographic changes, particularly salinity, are known to have occurred (e.g. the Little Ice Age, in order to examine whether these changes occur concurrently with specific MRE/ΔR values and could be a driving mechanism for the latter.

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Tables

Table 1: Results of δ^{13}C values, ^{14}C measurements ± 1**σ**, and χ^2 test results for samples measured in this study. *Measurements excluded from the sample group on the basis of the χ^2 test.

Sample ID	**Site-context**	**Material type**	**δ^{13}C**	**^{14}C age BP ± 1σ**	**χ^2 test result**
SUERC-33736	NO-14	Hazel nutshell (*Corylus avellana*)	-23.5	7470 ± 30	
SUERC-33737	NO-14	Hazel nutshell (*Corylus avellana*)	-23.3	7440 ± 30	
SUERC-34911	NO-14	Hazel nutshell (*Corylus avellana*)	-25.0	7460 ± 40	
SUERC-34912	NO-14	Hazel nutshell (*Corylus avellana*)	-21.9	7400 ± 40	2.13 (χ^2 :$_{0.05}$ = 7.81)
SUERC-34913	NO-14	Limpet (*Patella vulgate*)	1.5	5070 ± 35	
SUERC-34914	NO-14	Limpet (*Patella vulgate*)	0.5	5080 ± 35	
SUERC-34918	NO-14	Limpet (*Patella vulgate*)	1.4	5105 ± 35	
SUERC-34919	NO-14	Limpet (*Patella vulgate*)	1.2	5085 ± 35	0.53 (χ^2 :$_{0.05}$ = 7.81)
SUERC-44850	TNB2-5	Hazel nutshell (*Corylus avellana*)	-24.5	5687 ± 18	
SUERC-44854	TNB2-5	Hazel nutshell (*Corylus avellana*)	-26.1	5677 ± 23	
SUERC-44855	TNB2-5	Hazel nutshell (*Corylus avellana*)	-24.0	5654 ± 23	
SUERC-44856	TNB2-5	Hazel nutshell (*Corylus avellana*)	-26.3	5692 ± 23	1.71 (χ^2 :$_{0.05}$ = 7.81)
SUERC-44858	TNB2-5	Limpet (*Patella vulgate*)	0.5	5911 ± 23	
SUERC-44860	TNB2-5	Limpet (*Patella vulgate*)	0.3	5853 ± 28	
SUERC-47247	TNB2-5	Limpet (*Patella vulgate*)	0.8	5953 ± 26	
SUERC-47137	TNB2-5	Limpet (*Patella vulgate*)	-1.5	5904 ± 39	6.88 (χ^2 :$_{0.05}$ = 7.81)
SUERC-33731	TNB1-1	Hazel nutshell (*Corylus avellana*)	-27.4	5415 ± 30	
SUERC-33732	TNB1-1	Hazel nutshell (*Corylus avellana*)	-26.9	5415 ± 30	
SUERC-34902	TNB1-1	Hazel nutshell (*Corylus avellana*)	-26.0	5355 ± 35	
SUERC-34903	TNB1-1	Hazel nutshell (*Corylus avellana*)	-27.9	5280 ± 35*	2.15 (χ^2 :$_{0.05}$ = 5.99)
SUERC-34904	TNB1-1	Limpet (*Patella vulgate*)	0.7	5560 ± 35*	
SUERC-34908	TNB1-1	Limpet (*Patella vulgate*)	1.0	5675 ± 40	
SUERC-34909	TNB1-1	Limpet (*Patella vulgate*)	1.1	5690 ± 40	
SUERC-34910	TNB1-1	Limpet (*Patella vulgate*)	1.3	5720 ± 35	0.77 (χ^2 :$_{0.05}$ = 5.99)
OxA-8482	GUN-177/83	Charred barley grain (*Hordeum sp.*)	-24.5	360 ± 35	
OxA-8483	GUN-177/83	Charred barley grain (*Hordeum sp.*)	-24.9	380 ± 35	
SUERC-34924	GUN-177/83	Charred barley grain (*Hordeum sp.*)	-23.0	345 ± 35	
SUERC-34928	GUN-177/83	Charred barley grain (*Hordeum sp.*)	-22.7	355 ± 35	0.53 (χ^2 :$_{0.05}$ = 7.81)
SUERC-34920	GUN-177/83	Limpet (*Patella vulgate*)	-0.3	685 ± 35	
SUERC-34921	GUN-177/83	Limpet (*Patella vulgate*)	0.2	660 ± 35	
SUERC-34922	GUN-177/83	Limpet (*Patella vulgate*)	1.2	670 ± 35	
SUERC-34923	GUN-177/83	Limpet (*Patella vulgate*)	0.6	645 ± 35	0.69 (χ^2 :$_{0.05}$ = 7.81)

Table 2: MRE values, ΔR values, and calibrated terrestrial calendar age ranges (95% confidence interval) for samples analysed in this study. *Values not calculated due to taphonomic disturbance.

Site-Context	**MRE (^{14}C yr) ± 1σ**	**ΔR (^{14}C yr) ± 1σ Intercept method**	**ΔR (^{14}C yr) ± 1σ probability Density Function method**	**^{14}C weighted terrestrial mean age BP ± 1σ**	**Calibrated age range (95% confidence)**
SA-13	416 ± 35	62 ± 34	63 ± 49	7600 ± 26	6480-6420 BC (8430-8370 BP)
NO-5	440 ± 69	77 ± 56	67 ± 78	7424 ± 30	6390-6290 BC (8340-8180 BP)
NO-14	*	*	*	7446 ± 17	6390-6250 BC (8330-8200 BP)
TNB2-5	229 ± 41	-137 ± 41	-143 ± 54	5679 ± 11	4540-4470 BC (6490-6410 BP)
TNB1-1	300 ± 51	-109 ± 56	-109 ± 55	5399 ± 19	4330-4240 BC (6280-6190 BP)
GUN-177/83	305 ± 24	-118 ± 28	-130 ± 36	360 ± 18	1460-1630 AD (490-320 BP)

Figures

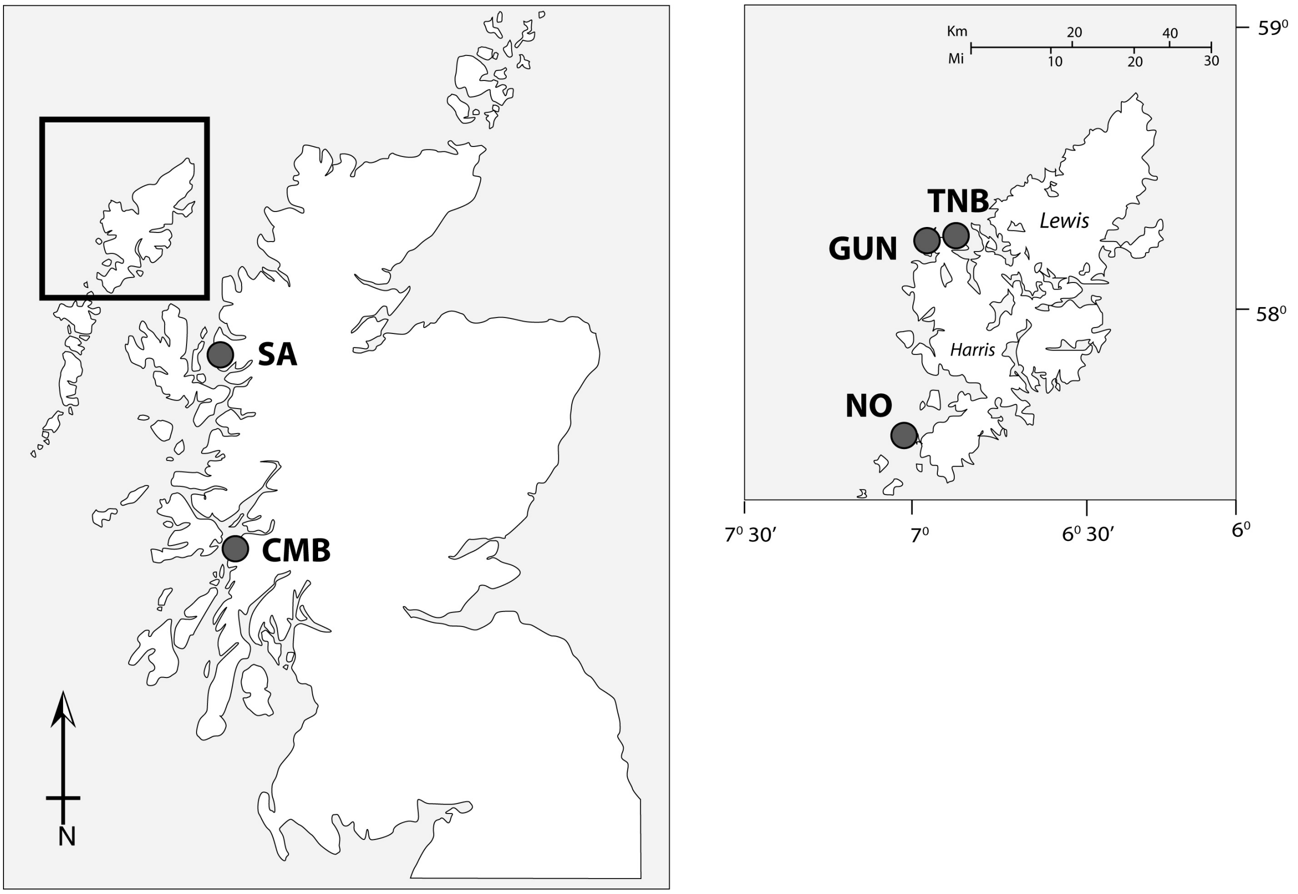

Figure 1: Location of sample sites from which material was obtained for MRE/ΔR, quantification, from which data was recalculated, and locations mentioned in the text. SA = Sand; CMB = Carding Mill Bay; NO = Northton; TNB = Tràigh na Beirigh; GUN = Guinnerso.

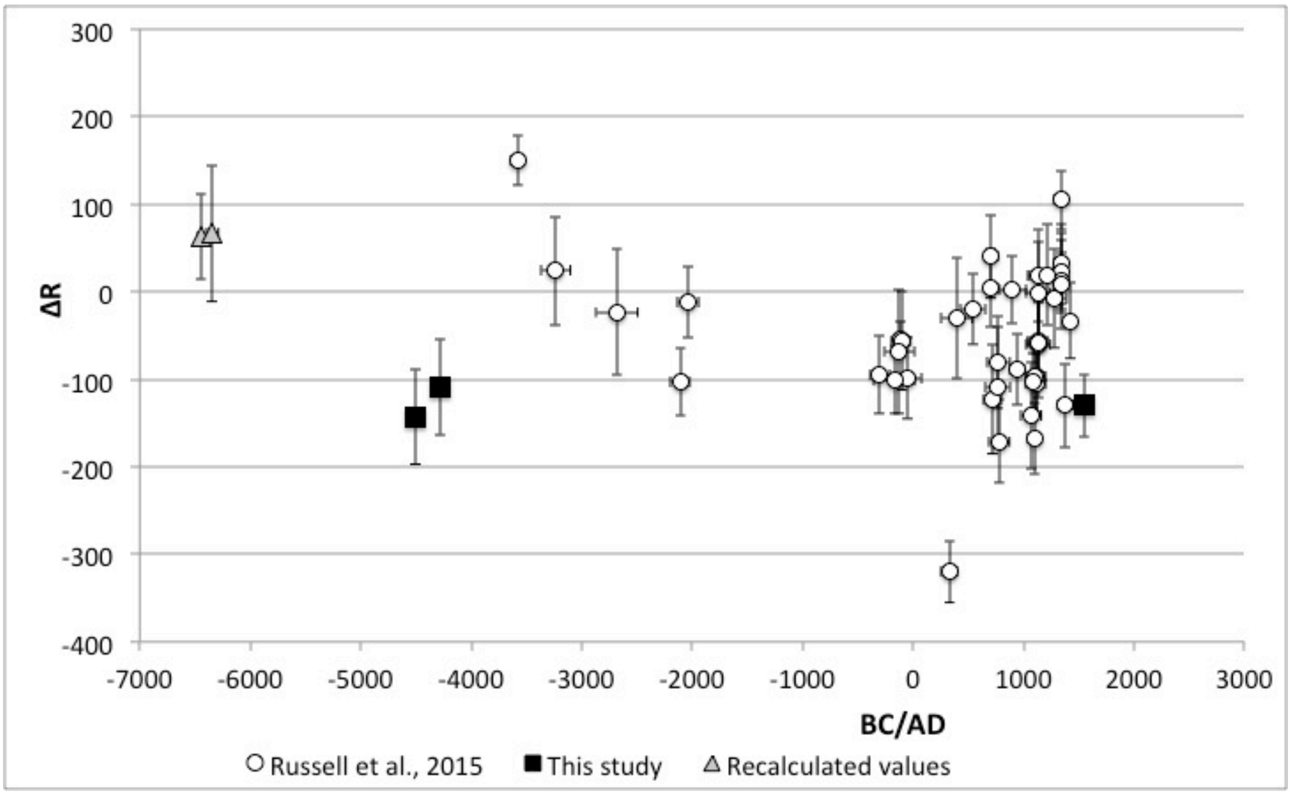

Figure 2: Graph of ΔR values for Scottish coastal waters through the Holocene showing new values (black squares) and recalculated values (grey triangles) alongside previous values for Scottish waters (white circles: Ascough et al., 2004, 2006, 2007, 2009, Russell et al., 2010, 2011b, 2015).