North Atlantic marine ^{14}C reservoir effects: implications for late-Holocene chronological studies

Authors and affiliations: P. L. Ascough[1*], G. T. Cook[1], A. J. Dugmore[2].

[1] SUERC, Scottish Enterprise Technology Park, Rankine Avenue, East Kilbride G75 0QF, UK. g.cook@suerc.gla.ac.uk

[2]Institute of Geography, School of GeoSciences, University of Edinburgh, Drummond Street, Edinburgh EH9 8XP. ajd@geo.ed.ac.uk

[*]Corresponding author. Tel.: +44 1334463936; fax: +44 1334 463949. E-mail: pla1@st-andrews.ac.uk (P. L. Ascough).

Abstract

We investigated surface ocean-atmosphere ^{14}C offsets for the later Holocene at eight locations in the eastern North Atlantic. This resulted in 11 new ΔR assessments for the west coast of Ireland, the Outer Hebrides, the north coast of the Scottish mainland, the Orkney Isles and the Shetland Isles over the period 1300-500 BP. Assessments were made using a robust Multiple Paired Sample (MPS) approach, which is designed to maximize the accuracy of ΔR determinations. Assessments are placed in context with other available data to enable reconstruction of a realistic picture of surface ocean ^{14}C activity over the Holocene period within the North Atlantic region.

Keywords

Radiocarbon; Marine Reservoir; North Atlantic; Holocene

1 Introduction

Radiocarbon (^{14}C) measurements are an essential chronological tool, providing a rigorous means of absolute dating for carbonaceous samples up to c.50 Ka BP, facilitating extended palaeoenvironmental and archaeological chronologies. Chronological accuracy is dependant however upon equivalence between the ^{14}C activity of measured sample carbon, and the timing of an event of interest in the palaeoenvironmental or archaeological record. The provenance of sample carbon must therefore be carefully considered prior to interpretation. A key complication arises when the sample carbon is derived from the marine environment. This includes both marine organisms e.g. flesh and exoskeletons of marine molluscs, and terrestrial organisms that have consumed marine resources. In such cases, chronological accuracy depends upon post-measurement correction for the Marine Reservoir Effect (MRE). The MRE is a quantitative measure of the offset between the ^{14}C activity of the atmospheric and oceanic carbon reservoirs at any point in time (Stuiver and Braziunas, 1993), and exists due to factors such as gaseous CO_2 transfer times and extended internal circulation of water masses while separated from atmospheric contact (Mangerud, 1972; Kovanen and Easterbrook, 2002). The net effect is a depletion of marine carbon ^{14}C activity relative to that of the contemporaneous atmosphere, which must be corrected for to enable comparison between ^{14}C measurements of marine carbon and material derived from other global carbon reservoirs.

The global average MRE shows time-dependant variations, according to fluctuations in the atmospheric ^{14}C signal. A corresponding time-dependant correction is available via the MARINE04 calibration curve (Hughen *et al.*, 2004), which models the oceanic response to atmospheric ^{14}C variations. However, the atmosphere-ocean ^{14}C difference in a specific geographic area may itself be offset from the average surface ocean MRE (Stuiver *et al.*, 1986). This offset is known as ΔR, and is both spatially and temporally specific. ΔR shows wide spatial variation, from high positive values in regions influenced by old, ^{14}C-depleted water, e.g. $\Delta R = 942 \pm 40$ ^{14}C yr for the Antarctic coast (Stuiver *et al.*, 1981), to negative values in areas where surface waters undergo more extensive exchange with atmospheric ^{14}C, e.g. $\Delta R = -25 \pm 20$ ^{14}C yr for the South China Sea (Southon *et al.*, 2002). As oceanographic and climatic variables are dynamic quantities, fluctuations in these variables will result in temporal variations in MRE and also in ΔR. These are pronounced at periods of extreme climatic change; the MRE value for the Mediterranean during Heinrich event 1 is 810 ± 130 yr (Sini *et al*., 2001), compared to the present average value of 390 ± 80 yr (Sini *et al*., 2000). Holocene ΔR variations are also apparent, e.g. over a c.4500 yr span, the ΔR in Northern and Western coastal waters of the British Isles shows variation between ΔR = 143 ± 20 ^{14}C yr and ΔR = -100 ± 15 ^{14}C yr (Ascough *et al*., 2007a).

Enhanced understanding of ocean-atmosphere ^{14}C offsets is therefore a highly desirable research goal. However, this can only be achieved if ΔR values are quantified with a high degree of both accuracy and precision. Quantification necessitates a comparison between individual terrestrial and marine age assessments that are demonstrably of equivalent absolute age. Typically, the marine age is provided by empirical ^{14}C measurement of a marine sample, while the equivalent terrestrial age is derived by a variety of means. For very young samples, ^{14}C measurement of marine samples where the actual calendar age of the sample is known may be possible (e.g. Yoneda *et al*., 2007). For more ancient samples, measurement of contemporaneous terrestrial carbon (e.g. Ascough *et al*., 2004), or reference to geochronological marker horizons (Waelbroeck *et al*., 2001, Austin *et al*., 1999) may be used. In all approaches it is clear that factors of site and sample selection strongly influence the precision and reproducibility of calculated ΔR values, and therefore consideration of these factors is critical. For example, in the case of marine sediments, uncertainties of association between material representing marine and terrestrial ^{14}C are increased with low sedimentation rates, or significant surface sediment mixing zones (Jones *et al.*, 1989; Paull *et al.*, 1991). Accuracy and precision in ΔR quantification is improved by the application of a rigorous site and sample selection strategy which minimizes sample-dependant uncertainties. One such methodology is a Multiple Paired Sample (MPS) approach, which uses multiple terrestrial and marine ^{14}C measurements (e.g. Ascough *et al*., 2007a). In the MPS approach, sample-dependant uncertainties are reduced by the use of short-lived single entities for measurement (e.g. Ashmore, 1990), as these represent integration of atmospheric or marine ^{14}C over a short time period. However, features such as habitat and potential sources of metabolic carbon may also affect the accuracy of calculated ΔR values. For example, inter-species variation in marine mollusc shell ^{14}C may be the result of specific feeding strategies in areas where different food resources may have non-uniform ^{14}C content, e.g. in regions of carbonate geology, where ^{14}C offsets are observed between grazing and pelagic feeders (Dye, 1994; Forman and Polyak, 1997; Heier-Nielsen *et al*., 1995). In addition, variation in the ^{14}C activity of carbon input to the growing shell may exceed measurement precision in regions where short-term, large-intensity fluctuations in upwelling regimes are documented (Culleton *et al*., 2006). Consideration of these factors is therefore also important in selection of samples from which ^{14}C measurements are to be used in ΔR quantification.

The focus of this paper is therefore: i.) to demonstrate how application of a rigorous methodology can lead to effective reconstruction of a spatial pattern in MRE values for a particular time period, and ii.) to provide a new dataset of ΔR values for a climatically sensitive region during a period of palaeoenvironmental and archaeological significance, when accurate and precise chronologies are crucial. This is the North Atlantic region, which forms a highly suitable location for systematic study of the MRE, as rich coastal resources and a high density of prehistoric settlements have produced extensive deposits of paired marine and terrestrial carbonaceous material. Phases of fluctuation in climatic and oceanic variables over the past 1500 years have coincided with large-scale changes in human societies, during the expansion of the Norse peoples from Scandinavia across the North Atlantic (McGovern *et al*., 2007; Church *et al*., 2005). This period is therefore of considerable importance in the study of human-environment interactions, and is one example of a period when quantification and understanding of the MRE has important benefits for both palaeoenvironmental and archaeological reconstruction. For the work presented here we used a robust multiple paired sample (MPS) approach to assess ΔR for the west and north coast of Scotland and Ireland over the Norse period. This was achieved using extensive ^{14}C measurements of both marine mollusc shells and carbonized cereal grains at eight archaeological sites over a c.900 km transect. The temporal span of the study encompasses the interval immediately prior to and following the expansion of the Norse peoples within the North Atlantic region, and this study utilizes the wealth of material that is currently becoming available from sites across the region.

2 Materials and methods

2.1 Sample selection

Samples were obtained from excavated archaeological deposits at sites on the west coast of Ireland, the Outer Hebrides, the north coast of the Scottish mainland, the Orkney Isles and the Shetland Isles (Figure 1), via consultation with the site excavators. While exposed open coastal locations may be considered well-mixed with regard to coastal ocean surface waters, certain geographic features such as sheltered topography (e.g. fjords) and carbonate geology can modify the ^{14}C content of surface ocean water in a specific location via the introduction of significant amounts of terrestrial organic detritus and carboniferous geological material (e.g. Spiker, 1980; Heier-Neilsen *et al.,* 1995; Ulm, 2002). Specific site selection criteria were therefore applied to ensure the influence of local geographic features was avoided, where material was obtained exclusively from exposed coastal locations away from estuarine settings and significant meteoric water influence. At three sites, (RH, SC and QG), two deposits were selected for sampling, giving a total of 11 deposits. Deposits were selected on the basis of a rigorous protocol (Ascough *et al*., 2004) designed to maximize the likelihood of obtaining contemporaneous marine and terrestrial material. A key methodological feature was the measurement of multiple, individual organisms (single entities), of both marine and terrestrial material, from each deposit. In this way, factors such as the presence of exogenous material and mixing within a deposit could be accounted for, and the (terrestrial and marine) ^{14}C age of the deposit most reliably established. The material obtained from each deposit comprised marine molluscs and carbonized cereal grains. In all cases except SC-206, the mollusc species was limpet (*Patella vulgata*), and the cereal species was barley (*Hordeum sp.*). From SC-206, periwinkle (*Littorina littorea*) and oat (*Avena sp.*) were used. These samples represent a relatively short growth interval, while they inhabited a restricted spatial range during their lifespan. The sampled sites were located in the open coastal zone, away from regions of carbonate geology, estuarine zones and sources of significant freshwater influence.

2.2 ^{14}C measurement

Carbonized plant macrofossils were pre-treated using a standard acid-base-acid procedure, where acid hydrolysis with HCl was followed by sequential removal of organic acids in NaOH solution, followed by a final HCl wash. CO_2 was extracted from the pre-treated plant macrofossils by combustion in pre-cleaned, sealed quartz tubes, using the method of Vandeputte *et al.* (1996). Marine shells were inspected to ensure that only hard, non-porous shells with no surface evidence of carbonate re-precipitation were used (Mangerud, 1972, Mook and Waterbolk, 1985). Surface contaminants were removed by abrasion and ultrasonic cleaning, followed by removal of the outer 20% of the shell by etching in 1M HCl (c.f. Heier-Nielsen *et al.,* 1995). The shell was then homogenized by roughly crushing, and a 0.1g aliquot taken for CO_2 extraction by acid hydrolysis with 1M HCl under vacuum. Immediately prior to the extraction, a further 20% by weight of the sample was removed by 1M HCl etching, ensuring removal of any carbon that might have exchanged after the main pre-treatment.

Evolved gases from either plant or shell samples were purified cryogenically using a solid CO_2/ethanol trap, followed by a liquid N_2 trap. A 2 ml aliquot of the purified CO_2 was converted to graphite by the method of Slota *et al.* (1987), and sample $^{14}C/^{13}C$ ratios were measured at 4.5 MV on a NEC 5 MV terminal voltage AMS instrument with carbon in the 4+ charge state (SUERC). Where possible, samples from a single context were measured in one sample wheel to reduce the effect of random machine error, and laboratory measurement precision at 1 sigma was typically ± 35-40 ^{14}C yr. The error is based on count rates from modern reference standards (Oxalic acid II), background standards (interglacial wood for organic samples and Icelandic doublespar carbonate for shell samples) as well as that on the sample but limited by the standard deviation on between 10 and 13 targets prepared from a dendro dated wood sample and measured in the same batch. $\delta^{13}C$ values were determined using an aliquot of the sample CO_2, measured on a VG SIRA 10 IRMS. Internal standards used were NBS 22 (oil) and 19 (marble), and the isotopic values for the samples are reported as per mille (‰) deviations from the VPDB international standard.

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

The resulting group of terrestrial or marine ^{14}C ages for each context was tested for internal consistency using a chi-squared (χ^2) test (c.f. Ward and Wilson, 1978). This compares the variability within a measurement group with the errors on individual measurements, in order to assess the consistency of the measurements. In several instances, individual samples had been measured more than once, and where these measurements were within 2σ of each other, they were combined as a weighted mean for that sample prior to chi-squared testing, in order to avoid biasing the test. The variability in the measurements is considered to exceed that which could occur by chance if the χ^2 test (*T)* value calculated for the ^{14}C ages exceeds the *T*-statistic for 95% confidence of *N* ^{14}C age measurements ($\chi^2_{:0.05} = T$). If the *T* value of the measurement group was lower than the acceptance limit, the measurements within the group were considered to be contemporaneous. In some instances however, the measurement group *T* value exceeded the acceptance value, indicating either true variability in the group, or variation associated with sample measurement. To determine the source of variation and the measured ages that most accurately represented the terrestrial or marine ^{14}C age of the deposit, further measurements were then made, and reference was also made to separate existing chronological data for the deposit, where available. For deposits where terrestrial ^{14}C measurements showed variability, it was not possible to perform repeat measurements of individual carbonized cereal grains, due to sample size. Instead, additional measurements were made of separate cereal grains from the same deposit. In the case of marine mollusc shell samples, repeat measurements of individual shells were possible, due to the larger size of this sample type. Where repeat measurements were consistent with initial ^{14}C measurement of the same sample, the measurements were combined in a weighted mean ^{14}C age for that sample. Following these measurements, the group of all available (marine or terrestrial) ^{14}C measurements for the deposit was then evaluated as a whole. Measurements that then increased the variability of the measurement group beyond the *T*-value for acceptance were excluded from the group and a new *T*-value calculated for the consistent ^{14}C measurements.

2.4 *Investigation of intra-shell ^{14}C variability*

Oceanic systems that induce rapid ocean ^{14}C variations, such as strong upwelling, are not a feature of the British Isles, however fluctuations in features such as freshwater input to coastal waters may vary over timescales relevant to the lifespan of the species included in this study (i.e. <15 yr). To check this potential source of variation, multiple measurements were made of a single shell which had a significantly different ^{14}C activity from the remainder of the measured marine samples from the deposit (GA-165). This resulted in five separate ^{14}C measurements from an individual entity, the internal variability of which was then assessed using the χ^2 test.

2.5 *Calculation of ΔR values*

A ΔR value was calculated for each possible pairing of terrestrial and marine ^{14}C ages by converting the terrestrial ^{14}C age to a modelled marine ^{14}C age ± 1σ, incorporating the uncertainty in the interpolated calibration curve data. The offset between the midpoint of the modelled marine ^{14}C age bounds and a specific conventional ^{14}C age for a marine sample provides the ΔR for that pairing. The 1σ error for the ΔR determination is obtained by propagation of the errors on the marine age and the modelled marine age.

A group of statistically indistinguishable marine and corresponding terrestrial ^{14}C ages from a specific deposit, determined on the basis of the χ^2 test, was used to calculate ΔR for the period of time represented by the samples. The method considers all possible estimates of ΔR for the group of measured samples by calculating a value of ΔR for each possible pairing of terrestrial and marine ^{14}C ages. The distribution of these values is then summarised by the weighted mean and appropriate standard error for prediction, to account for any additional variability due to the precise pairing of terrestrial and marine samples. Individual ΔR values are calculated using a linear interpolation of the INTCAL04 and MARINE04 calibration data (Reimer *et al.*, 2004, Hughen *et al.*, 2004). The terrestrial calibrated age range represented by the material within the deposit is calculated by calibrating the weighted mean of terrestrial ^{14}C ages for that context. The weighted mean was calculated using measurements that were statistically equivalent on the basis of a χ^2 test, and a calibrated range was provided using the INTCAL04 atmospheric dataset (Reimer *et al.,* 2004) and the OxCal v3.10 calibration program (Bronk Ramsey, 1995; 2001).

3 Results

The $\delta^{13}C$ values of all samples (Table 1) fall within the typical ranges for terrestrial plants, terrestrial mammal bone and marine mollusc shells (Aitken, 1990). Variation in $\delta^{13}C$ values of terrestrial or marine samples within each context was relatively low, with the maximum variation shown in terrestrial plant material. The variation in average $\delta^{13}C$ values between contexts is higher than the within-context figure. In one instance, measurement of a marine shell from deposit DL3 returned an unusually high error (± 125 ^{14}C yr). This sample was re-measured, and two of the other shells were repeated in the same run, to check the measurement consistency between the runs. Repeat measurements confirmed the initial shell ages, and reduced the error on the affected shell to ± 35 ^{14}C yr (Table 1).

3.1 Consistency of ^{14}C measurements within sample groups

For ten deposits, variability in ^{14}C measurements did not exceed the *T*-value for the sample group, and measurements were used in subsequent assessment of ΔR for that context (Table 2). In six instances however, variation within the marine or terrestrial ^{14}C measurements in a sample group exceeded the *T*-value for acceptance, mainly due to a single measurement that outlay the remainder of the group. In all cases, it was possible to identify a measurement group most likely to represent an accurate terrestrial or marine ^{14}C age for the context on the basis of additional and repeat measurements, and reference to existing chronological data. In FL-JM76, SC-206 and GA-165, a high *T*-value was due to an individual measurement, and the remaining larger group of consistent ^{14}C ages (Table 3) was taken as representative of the age of the deposit. For the terrestrial group of ^{14}C ages from BO-64, two measurements were not consistent with the remainder of the group. The larger group of consistent measurements again were taken as representative of the true deposit age. This decision is supported by the fact that the larger measurement group is indistinguishable from 28 previous ^{14}C measurements from the site (Neighbour, 2001; Church, 2002).

Four terrestrial measurements from QG-A004 are equivalent on the basis of a χ^2 test; however, the remaining four measurements are inconsistent with this group, and with each other. The group of four equivalent measurements were used for ΔR calculation from QG-A004, as these are consistent with ^{14}C ages from two other previously dated contexts in the same midden sequence (Barrett and Gerrard, 2002). The equivalent measurements from QG-A004 are also consistent with measurements presented here from QG-A023, which is located in the same midden sequence. QG-A004 was an upper stratigraphic layer, where overlying deposits showed signs of mixing and bioturbation (Barrett and Gerrard, 2002); therefore the variation in ^{14}C ages of terrestrial material in QG-A004 may result from these effects. To establish that this effect was absent in the deeper stratigraphic layer of QG-A023 we performed additional and repeat measurements of the terrestrial and marine samples from QG-A023, which confirmed the lack of variation in ^{14}C measurements in this lower deposit.

3.2 Investigation of intra-shell ^{14}C variability

The results of five individual measurements on a single, homogenized shell from GA-165 (Table 1), were found not to be significantly different on the basis of the χ^2 test for contemporaneity (T= 4.94 ($\chi^2_{:0.05}$ = 9.49)). The empirical standard deviation on the measurement group was 39 ^{14}C y, which is consistent with the error on the individual measurements of ± 35 ^{14}C y. This indicates a lack of variability greatly exceeding measurement precision in ^{14}C concentration within samples of *Patella vulgata* grown within the study area. These results are supported by other repeated measurements on shells made during this study (i.e. from deposits where marine ^{14}C measurements exceeded the T-value for acceptance), where repeat measurements of the same shell returned ages consistent with the initial measurement in nine instances.

3.3 Terrestrial calibrated ranges and calculated ΔR values

The 2σ calibrated ranges (derived from the weighted mean terrestrial ages for each deposit) covered a total of 738 cal yr, over the period 1294-556 BP (see Table 4). The sampled deposits may be grouped, according to the calibrated periods they cover, into the following phases; 1294-1095 BP (DL3; DL11 and SC-1269); 1058-956 BP (BO-

64 and GA-165); 907-732 BP (QG-A004; QG-A023; FL-JM76 and RH-3019) and 735-556 BP (SC-206 and RH-3004). The 728 cal yr calibrated age range is covered almost continuously by the measurements and ΔR values presented in this paper, with a maximum gap of 49 years over the period 956-907 BP (Figure 2). From the deposits, a total of 11 ΔR values were calculated on the basis of contemporaneous terrestrial and marine ^{14}C measurements (Table 4). These ΔR values ranged from +32 ± 14 ^{14}C yr (SC-206) to -121 ± 16 ^{14}C yr (SC-1269), and are statistically different (T = 98.80; (χ^2:0.05 = 19.68).

4 Interpretation

4.1 Intra-shell ^{14}C variation

As noted above, ^{14}C age differences have been identified across marine mollusc shells, attributed to seasonal variations in upwelling and freshwater input on the Californian coast (Culleton *et al.*, 2006). Unaddressed, such variations have real potential to limit the precision to which it is practically possible to define ΔR using affected samples. A previous investigation of inter-species mollusc shell ^{14}C variation at sites within the study area did not show any species-dependant effect on calculated ΔR values (Ascough *et al,* 2005), however in that work, the potential for intra-shell ^{14}C variation was not assessed. In the present study, multiple measurements of individual shells do not indicate the presence of ^{14}C age variability in the marine samples which exceeds measurement precision. Deep water upwelling and geological carbon sources are not a feature of the sample region, and the results suggest that variation in freshwater and terrestrial organic carbon content has not been of sufficient magnitude to influence shell ^{14}C values at the sites selected according to the sampling protocol. Careful use of site selection criteria should therefore be combined wherever possible with repeated analyses of marine carbonates to ensure the accuracy of ΔR values is not compromised by the effect of variation in ^{14}C age across an individual marine sample

4.2 Quantification of ΔR for the North Atlantic

On the west coast of Ireland for the period 1282-1095 BP, values from DL3 and DL11, <2 km apart, are statistically equivalent (T = 1.93; (χ^2:0.05 = 2.84)), where ΔR = -111 ± 18 ^{14}C yr and -75 ± 18 ^{14}C yr, respectively. An earlier assessment is available for the area, at Omey Island (N 52.32, W 10.9) of ΔR = -143 ± 16 ^{14}C yr for 960-790 BP (Ascough *et al*., 2006). Taken together, (T = 7.77; ((χ^2:0.05 = 7.81)) these data indicate steady ocean surface ^{14}C content, higher than the global average, over the late Holocene for the west coast of Ireland. Previous assessments of ΔR at these sites are based on pairings of single terrestrial and marine ages, rather than the MPS approach and produced ΔR = -111 ± 61 ^{14}C yr for 930-760 BP and ΔR = -206 ± 70 ^{14}C yr for 960-870 BP at OI (Omey Island), and ΔR = -113 ± 55 ^{14}C yr for 1290-

1140 BP at DL11 (Reimer *et al*., 2002). These values are consistent with the data presented in this paper.

Further north in the Scottish Outer Hebrides, ΔR values calculated for BO-64 and GA-165 are -56 ± 14 ^{14}C yr and -85 ± 16 ^{14}C yr, (T = 1.77; (χ^2:0.05 = 2.84)), covering the period 956-1063 BP, again suggesting a consistent surface ocean ^{14}C activity for this region over the sampled period. Similar ΔR values have previously been calculated with the MPS approach in this area for 2350-1890 BP (Ascough *et al.*, 2004), ranging between ΔR = -65 ± 21 ^{14}C yr and ΔR = -117 ± 22 ^{14}C yr. This data suggests correspondence between ocean surface ^{14}C activity in the region at ~1000 and ~2000 BP. However, for the periods 8430-8108 BP and 5600-5470, ΔR values in the Outer Hebrides are much higher, ranging between ΔR = +143 ± 20 ^{14}C yr and ΔR = +64 ± 19 ^{14}C yr (Ascough *et al*., 2007a).

Higher ^{14}C activity than the global average in west Irish coastal waters for the periods 1290-1095 BP and 960-760 BP is consistent with a constant influence of the North Atlantic drift current in this area, in which ocean surface waters contain higher amounts of ^{14}C due to extended periods of exchange with atmospheric CO_2 (Campin *et al.,* 1999). Present surface waters around the Outer Hebrides are derived from the Scottish Coastal Current (SCC), originating from Atlantic and Irish Sea waters (Knight and Howarth, 1999), and reinforced by lower-salinity water from the major west coast rivers (McKay *et al.*, 1996). In common with the Irish values, ^{14}C activity for the Outer Hebrides around 1000 BP clearly reflects a strong North Atlantic Current influence, which is also in evidence at ~2000 BP.

In contrast to the consistency in ΔR for single time periods on the west Irish coast and the Outer Hebrides, ΔR values calculated for the Orkney Isles at 920-738 BP, despite rigorous use of the MPS approach, show greater variability. In the same stratigraphic sequence, for one deposit (QG-A004), ΔR = -102 ± 18 ^{14}C, whereas for QG-A023, ΔR = -49 ± 13 ^{14}C yr. It is likely that this greater variation results from fluctuations in environmental variables, responsible for determining ΔR. These fluctuations appear sufficient to result in variation in values of up to 50 ^{14}C yr over the combined temporal period of these 2 deposits (182 cal yr). Similar smaller-scale ΔR variability over periods of c.200 cal yr for localized ocean areas is also observed at deposits on the north Scottish coast. These correspond to the temporal phase covered by the deposits at QG, with values from FL-JM76 (912-781 BP) of ΔR = -90 ± 16 ^{14}C yr, and from RH-3019 (905-732 BP) of ΔR = -56 ± 16 ^{14}C yr. Overall these values at ~900-700 BP continue the trend of dominance by the ^{14}C signature of the North Atlantic-derived waters, with ΔR between 50 and 100 ^{14}C yr below the global average for this period. However, there is also evidence that localized climate and ocean variables modify ΔR to a greater extent in this region, resulting in increased variability over time periods of ~200 cal. yr. A similar effect is observed in determinations for the earlier Holocene for the Orkney Isles at 4138-3898 BP, when values varied from ΔR = -13 ± 18 ^{14}C yr to ΔR = -100 ± 15 ^{14}C yr between the west and east of the Orkney Island system (Ascough *et al.*, 2007a). An important point is therefore that in certain areas, greater variability in ΔR may exist, which is most likely derived from more rapid and/or more extreme fluctuations in local climatic and oceanographic variables that drive ΔR. These are poorly defined at present, but may include localized oceanic circulation influences and bathymetry.

Variation in ΔR at a single site is also observed at the most northerly sample location, SC, however in this case different ΔR values relate to two different time periods. For 1180-1294 BP (SC-1269), ΔR = -128 ± 16 ^{14}C yr. During this period, surface ocean ^{14}C in west Irish waters is also higher than the global average at DL-3 and DL-11. However, for a period up to 600 years later (735-672 BP), ΔR at SC is ΔR = +32 ± 14 ^{14}C yr (SC-206). This period overlaps with a phase (668-556 BP) when similar ΔR is observed on the north Scottish coast at RH, when ΔR = +28 ± 21 ^{14}C yr. A coeval increase in ΔR within northern Scottish waters at these sites suggests that in this instance, the source of variation may not be localized modulation of otherwise constant climatic and oceanographic variables, but rather wider scale fluctuations in these variables, resulting in ΔR variation over a wider area. Regression analysis shows that a significant proportion of the variation in ΔR values presented in this paper is time-dependant, $r^2 = 0.54$ ($P < 0.01$), with an overall increase in values from the earlier to later period (Figure 3). It is likely therefore, that this is an observable response in ocean ^{14}C to environmental forcing. For example, the apparent decrease in surface-ocean ^{14}C observed here at c.730-550 BP (Figure 3), corresponds to a period of climatic cooling in North Atlantic palaeoenvironmental proxy records, known as the Little Ice Age (Grove, 1988; Lamb, 1995; Broecker, 2000) when increasing influence of polar waters may have affected the study region (e. g. Jiang *et al*., 2005).

When data from this study are considered with other ΔR values for the study region obtained using the MPS approach, it appears that over the extended Holocene period, surface-ocean ^{14}C in the study region shows long-term fluctuations. The result is ΔR changes on the order of 200 ^{14}C yr, which fit a third-order polynomial function (Figure 4), where ΔR values decrease from the early to later Holocene until the period at c.730-550 BP, as described above. These changes are likely to reflect a response to larger regional climatic and oceanographic variations, for example, fluctuating strength in the North Atlantic Deep Water production.

Although Harkness (1990) suggested latitude dependence in surface ocean ^{14}C around the British Isles on the basis of modern mollusc shell measurements, analysis of Holocene data by Reimer *et al*., (2002), did not identify such a trend for the British Isles and Ireland. Regression analysis of the data presented in this paper also shows no clear linear dependence of values upon latitude and longitude within the study region (r^2 = <0.1; P = > 0.1), although it is true that modulation of surface ^{14}C by local conditions may induce some non-linear spatial variations. Stronger spatial patterns in the North Atlantic ΔR are apparent when values from sites from Ireland and the British Isles are contrasted with data from higher latitudes, also obtained using the MPS approach (Figure 5). At 940-790 BP, the ΔR for waters around the Faroe Isles is +64 ± 13 ^{14}C yr (Ascough *et al*., 2005), compared to values of between -102 ± 18 ^{14}C yr and -49 ± 13 ^{14}C yr for Scottish waters. For Icelandic waters, ΔR values at 965-1180 BP are +111 ± 14 ^{14}C yr and +96 ± 15 ^{14}C yr (Ascough *et al*., 2007b), in contrast to values of between -85 ± 16 ^{14}C yr and -54 ± 14 ^{14}C yr for Scottish waters. This pattern is continued for the period 670-550 BP, when Icelandic values are ΔR = +132 ± 21 ^{14}C yr (Ascough *et al*., 2007b), in contrast to values from the Shetland Isles and northern Scotland of ΔR = +32 ± 14^{14}C yr and +28 ± 21 ^{14}C yr. When values for the extended time period 1180-550 BP are considered, there is a significant positive correlation between latitude and ΔR values (r^2 = 0.74; P = <0.01). This appears to reflect continuous oceanographic differences between the waters of Ireland, the UK, Faroes and Iceland, with increasing dominance of low-^{14}C Arctic-derived waters, such as the East Iceland Current, at higher latitudes (e.g. Eiríksson *et al*., 2000; 2004).

4.3 Implications for palaeoenvironmental investigation

In order to accurately and precisely quantify ΔR for a specific ocean area, methodological considerations are central. Clearly, even when rigorous sample selection protocols are applied, depositional mixing effects have great potential to decrease the accuracy of calculated values. It is clearly crucial for the accuracy of ΔR values that sources of variation that affect the contemporaneity of marine and terrestrial ^{14}C ages used to calculate ΔR are fully considered, identified, and where possible, empirically assessed. The MPS approach offers a valuable means of achieving this, and the chance to maximize the accuracy of calculated ΔR values. The results of this study demonstrate the utility of a rigorous sample selection process in investigation of surface-ocean ^{14}C over extended time periods, by enabling production of coherent and reproducible ΔR assessments.

The need for accuracy in ΔR quantification is particularly important, given the variations in ΔR that may apparently be in effect at some sites in close proximity. This is of direct relevance in the North Atlantic region, where reconstructions of paleoenvironmental conditions and human-environment interactions have been the subject of especially detailed study, with globally important implications (e.g. Dugmore *et al.*, 2005). Accurate ΔR quantification therefore offers the opportunity of enhanced chronological resolution, particularly if a focus is placed upon periods and locations of particular palaeoenvironmental and archaeological research interest. In addition, a rigorous approach to ΔR quantification could enable integration of surface ocean ^{14}C measurements with climatic and oceanographic forcing mechanisms for MRE variations that at present remain poorly understood.

5 Conclusions

- Assessment of ΔR for the Norse period using the MPS methodology shows a pattern of surface ocean waters in the study region which are relatively homogeneous with respect to ^{14}C.
- Surface ocean waters in the study region are enriched in ^{14}C relative to the MARINE04 global average for most of the studied time period, indicating a dominant influence of North Atlantic current waters.
- Although there is evidence for local modulation of the wider ΔR signal in some locations, it appears that overall surface-ocean ^{14}C in the study region responds to large-scale regional climatic fluctuations, and local variations are subordinate to that of the major oceanic forcing.
- The data presented here fit an emerging pattern of North Atlantic ΔR, which shows significant spatial variation between the islands of Ireland, Britain, the Faroes and Iceland.
- Accuracy and coherence of ΔR determinations is greatly enhanced by the application of a rigorous sample selection methodology, such as the MPS approach.

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Figure legends

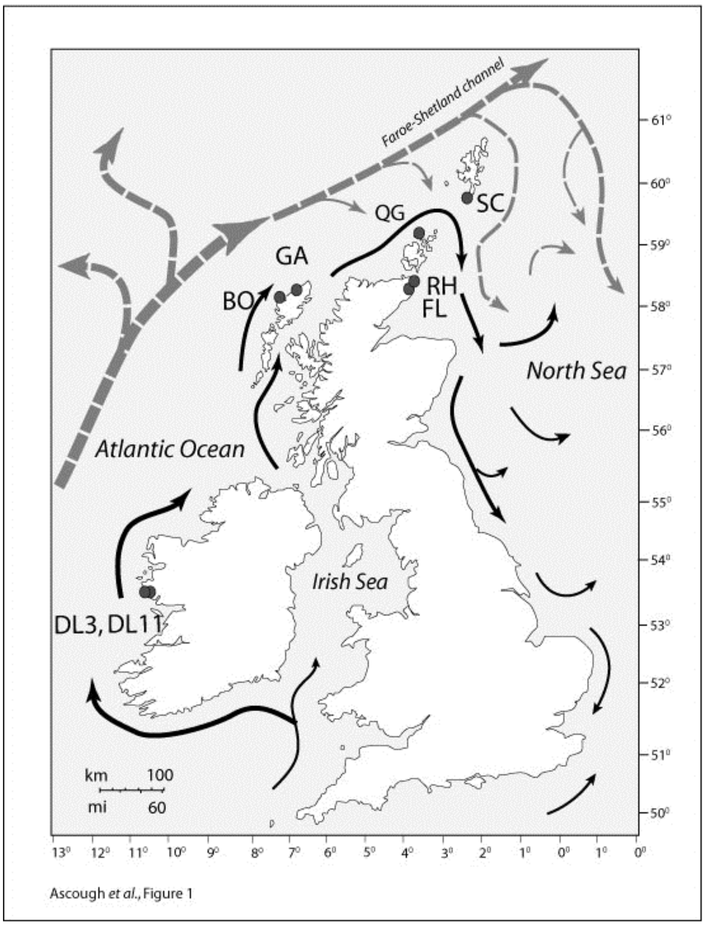

Figure 1: Locations of sample sites included in the study and modern surface circulation in the overall study area showing Atlantic water (grey dashed arrows) and coastal currents including the Scottish Coastal Current (black solid arrows) (after OSPAR, 2000).

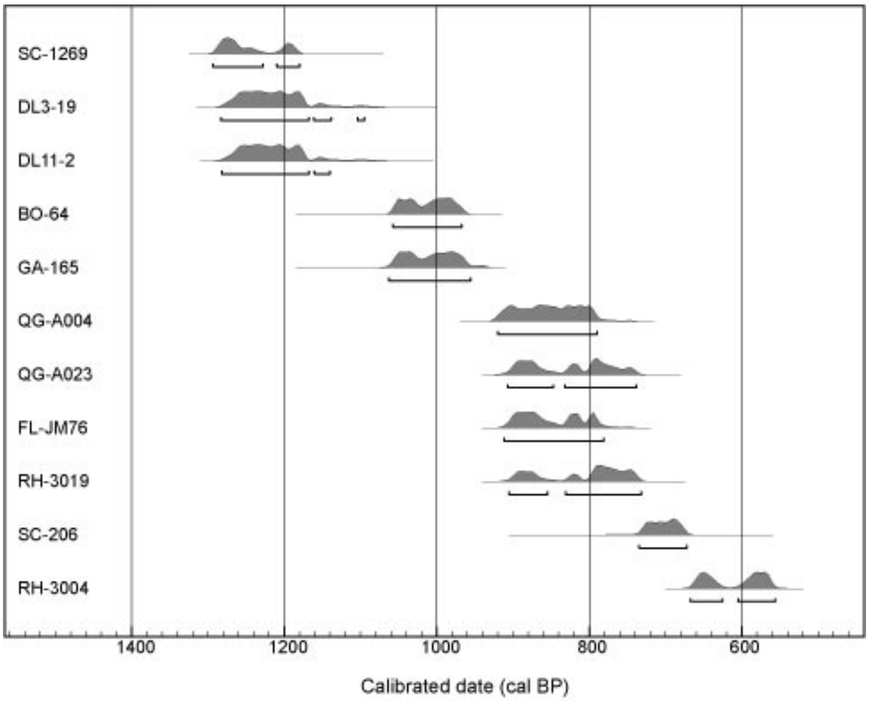

Figure 2: Oxcal plot of calibrated BP age ranges for the weighted mean terrestrial ages for each deposit.

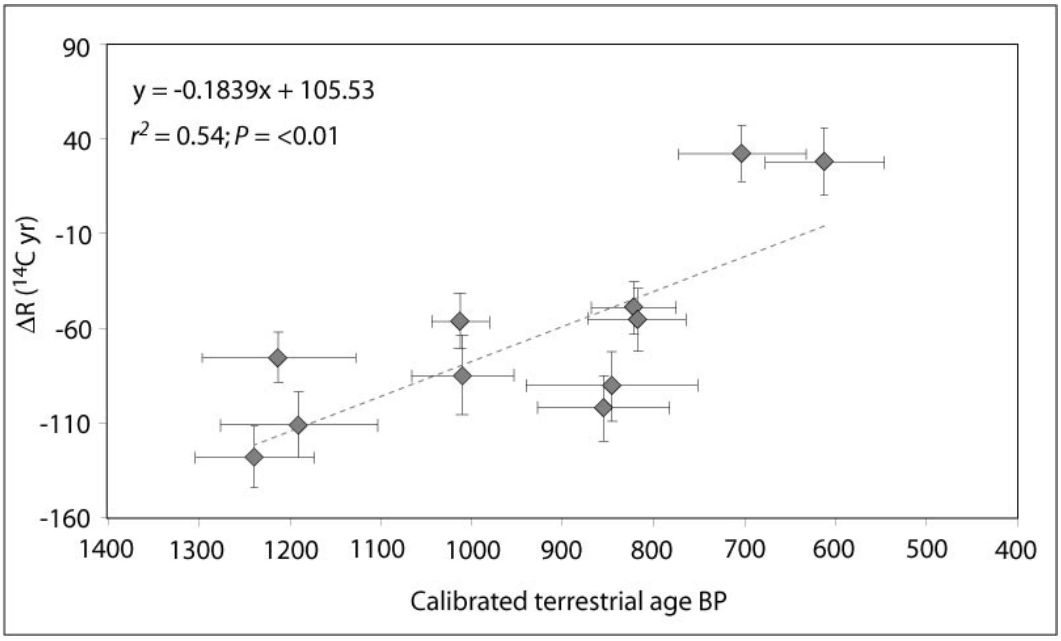

Figure 3: ΔR values calculated from data presented in this paper versus calibrated terrestrial age BP. Calibrated age BP was obtained from the weighted mean of statistically equivalent terrestrial sample ^{14}C measurements. Details of the linear correlation between ΔR and calibrated age BP are given on the graph.

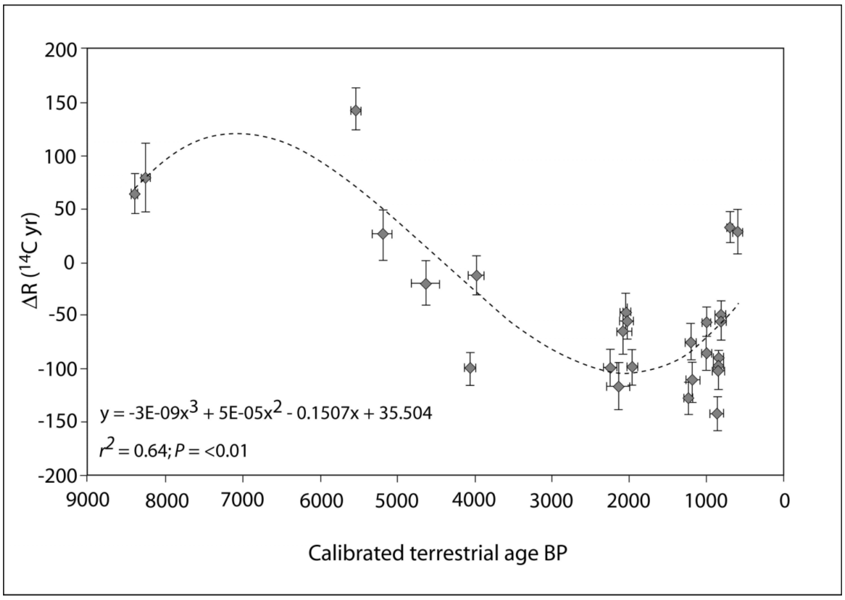

Figure 4: ΔR values for Ireland and the British Isles using the multiple paired sample (MPS) approach for deposits over the extended Holocene period (from Ascough *et al*., 2004; 2006, 2007a and present paper), versus calibrated terrestrial age BP. Details of the fit to a third-order polynomial are given on the graph.

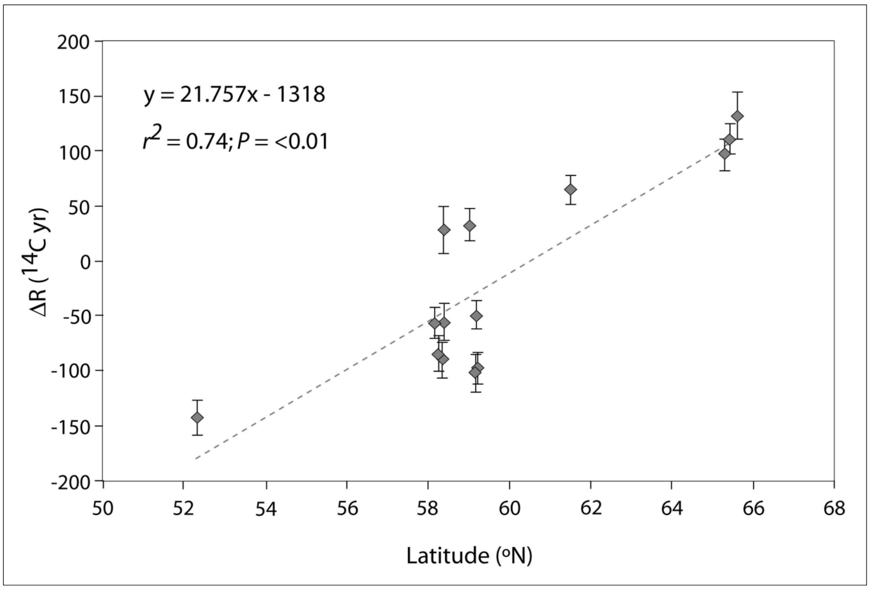

Figure 5: ΔR values versus latitude (°N) in the North Atlantic calculated using the multiple paired sample (MPS) approach (from Ascough *et al*., 2006; 2007b and present paper) for the period 1180-550 BP. Details of the linear correlation between ΔR and latitude are given on the graph, showing how data presented in this paper fits with an overall emerging pattern of spatial variability in the North Atlantic for the period centred on 1000 BP.

Table legends

Table 1: Terrestrial and marine ^{14}C ages and stable isotopic ($\delta^{13}C$) measurement results for each deposit included in the study.

Table 2: Results from each context of χ^2 tests for contemporaneity of ^{14}C measurement values from a group of terrestrial or marine sample materials.

Deposit	**x2 test result (terrestrial samples)**	**x2 test result (marine samples)**
SC-1269	4.65; $(\chi^2{:}_{0.05} = 7.81)$	6.25; $(\chi^2{:}_{0.05} = 7.81)$
DL11-2	5.16; $(\chi^2{:}_{0.05} = 7.81)$	2.06; $(\chi^2{:}_{0.05} = 7.81)$
RH-3019	1.06; $(\chi^2{:}_{0.05} = 7.81)$	0.52; $(\chi^2{:}_{0.05} = 7.81)$
RG-3004	0.70; $(\chi^2{:}_{0.05} = 7.81)$	2.28; $(\chi^2{:}_{0.05} = 7.81)$
DL3-19	7.14; $(\chi^2{:}_{0.05} = 7.81)$	4.05; $(\chi^2{:}_{0.05} = 7.81)$
QG-A023	8.82; $(\chi^2{:}_{0.05} = 11.10)$	4.04; $(\chi^2{:}_{0.05} = 7.81)$
SC-206	4.78; $(\chi^2{:}_{0.05} = 7.81)$	20.04; $(\chi^2{:}_{0.05} = 7.81)$
BO-64	23.62; $(\chi^2{:}_{0.05} = 11.10)$	6.83; $(\chi^2{:}_{0.05} = 7.81)$
QG-A004	114.93; $(\chi^2{:}_{0.05} = 14.10)$	0.65; $(\chi^2{:}_{0.05} = 7.81)$
GA-165	1.07; $(\chi^2{:}_{0.05} = 7.81)$	18.62; $(\chi^2{:}_{0.05} = 7.81)$
FL-JM76	18.86; $(\chi^2{:}_{0.05} = 11.1)$	12.81; $(\chi^2{:}_{0.05} = 7.81)$

Table 3: Data for contexts that contained measurements that were inconsistent on the basis of a χ^2 test, showing consistent measurements and *T*-statistics for measurement groups used to calculate values of ΔR.

Context	**Lab Code**	**Age BP ± 1σ**	**2nd χ^2 test result**
BO-64 (Terrestrial)	SUERC-3169 SUERC-1038 SUERC-1039 SUERC-1040	1095 ± 35 1150 ± 35 1120 ± 35 1065 ± 35	3.20$(\chi^2{:}_{0.05} = 17.81)$
QG-A004 (Terrestrial)	SUERC-3149 SUERC-3542 SUERC-3150 SUERC-3151	980 ± 40 875 ± 35 960 ± 40 925 ± 40	4.61$(\chi^2{:}_{0.05} = 17.81)$
FL-JM76 (Terrestrial)	SUERC-1061 SUERC-1063 SUERC-1064 SUERC-3181 SUERC-3182	950 ± 50 910 ± 35 940 ± 45 870 ± 35 920 ± 35	2.46$(\chi^2{:}_{0.05} = 9.49)$
FL-JM76	SUERC-1065/3186/4941	1232 ± 31	2.04 $(\chi^2{:}_{0.05} = 5.99)$

(Marine)	SUERC-1066/4942 SUERC-1067/3187	1173 ± 28 1186 ± 60	
GA-165 (Marine)	AA-53257/SUERC-4051/4052/4053/4054 AA-53258 AA-53259	1385 ± 17 1360 ± 40 1415 ± 35	1.09 ($\chi^2_{:0.05}$ = 5.99)
SC-206 (Marine)	AA-51173/SUERC-3142 AA-51174 SAA-51175	1202 ± 26 1135 ± 35 1230 ± 35	3.98 ($\chi^2_{:0.05}$ = 5.99)

Table 4: ΔR values for contexts calculated on the basis of contemporaneous ^{14}C measurements

Deposit	**Terrestrial ^{14}C wt mean age**	**Cal time period (AD)**	**Cal time period (BP)**	**ΔR**
SC-1269	1312 ± 23	657-771	1294-1180	-128 ± 16
DL3-19	1265 ± 27	667-856	1283-1095	-111 ± 18
DL11-2	1264 ± 25	668-810	1282-1140	-75 ± 18
BO-64	1108 ± 18	893-984	1058-967	-56 ± 14
GA-165	1102 ± 26	887-995	1063-956	-85 ± 17
QG-A004	931 ± 24	1030-1161	920-790	-102 ± 18
QG-A023	896 ± 22	1043-1213	907-738	-49 ± 13
FL-JM76	912 ± 17	1039-1170	912-781	-91 ± 16
RH-3019	885 ± 24	1045-1219	905-732	-56 ± 18
SC-206	780 ± 27	1216-1279	735-672	32 ± 14
RH-3004	645 ± 25	1283-1395	668-556	28 ± 21