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ABSTRACT – This paper presents the results of a study using strontium, oxygen and carbon isotopes, strontium concentrations, infrared analyses and radiocarbon dating to investigate human mobility as seen in individuals from the Neolithic court tomb of Parknabinnia, Co. Clare, Ireland. Taking advantage of the recent demonstration that it is possible to obtain reliable *in vivo* strontium isotope signals from calcined bone, we compare measurements on cremated bone (n = 4) and uncremated tooth enamel (n = 4). The results suggest that two out of four uncremated enamel samples can be considered 'local' while the other two, and all four cremated bone samples, represent 'non-local' individuals. New radiocarbon dates obtained on two of the cremated bone fragments place them in the Chalcolithic/Early Bronze Age, rather than the Neolithic ages for the uncremated remains, demonstrating re-use of the monument. Assuming that our small sample is representative, it seems that the court tomb was used for burial by both 'locals' and 'non-locals' during the Neolithic and predominantly by 'non-locals' in the Chalcolithic/Early Bronze Age. This stands in contrast to the nearby Early Neolithic portal tomb of Poulnabrone where only one individual (of 17 analysed) appears to be an 'outsider'. This study suggests that, even within a small region, mobility and landscape use may have differed significantly within the Neolithic and also between the Neolithic and the Chalcolithic/Early Bronze Age.

INTRODUCTION

Funerary practices in the Irish Neolithic included the placement of both uncremated and cremated human remains in mortuary monuments, including court tombs. These monuments were constructed in the earlier Neolithic (from c. 3750 cal. BC), but use could have continued for centuries during the Neolithic and also sometimes re-used in the Chalcolithic (c. 2450–2150 BC) and the Early Bronze Age (c. 2150–1500 BC). Use and re-use involved both cremation and inhumation (Jones et al. 2015; Schulting et al. 2012). The reasons behind the choice of one funerary rite over the other are poorly understood. One possibility is that the cremated human bone represents only token deposits brought to the site from cremations that took place elsewhere, while non-cremated human remains represent the local dead. Given the long use-histories of some court tombs, might the occurrence of both practices within a single tomb reflect a diachronic shift in funerary rites?

In this paper, carbon, oxygen and strontium isotope ratios, strontium concentrations, radiocarbon dating, and infrared analyses are applied to address these questions of human mobility, funerary practice and tomb re-use at the earlier Neolithic court tomb at Parknabinnia, Co. Clare. We analysed both cremated bone and uncremated tooth enamel to investigate the possibility whether the former were part of token deposits brought to the monument from other locations. If so, we might expect their strontium isotope results to reflect a wider variety of lithologies than seen in the uncremated enamel, which could represent the deceased of the local community. Our results are compared to those previously published on uncremated enamel from the nearby Early Neolithic portal tomb at Poulnabrone, Co. Clare (Ditchfield 2014; Kador 2010; Kador et al. 2014).

The skeletal tissue of choice in C, O and Sr isotope studies is dental enamel, since this has been shown to be far more resistant to diagenesis than either dentine or bone (Tuross et al. 1989; Budd et al 2000; Hoppe et al. 2003; Lee-Thorp & Sponheimer 2003). Recent experimental work, however, has demonstrated that calcined bone, because of its substantially greater crystallinity and thus resistance to external influences, also provides a reliable substrate for strontium isotope analyses (Harvig et al. 2014; Snoeck et al. 2015). Carbon and oxygen isotopes, however, have been heavily altered by the very high temperature reached during cremation but can still be used to look into the cremation ritual. These observations open up new possibilities not only for Irish prehistoric archaeology (Snoeck et al. 2016a), but for that of other periods and places as well (e.g. Snoeck et al. 2018).

Strontium isotope analyses

Two isotopes of strontium, ^{86}Sr and ^{87}Sr, are widely used in mobility studies of humans and fauna. Their ratio (^{87}Sr/^{86}Sr) varies between different types of bedrock because of the radioactive decay of ^{87}Rb, with values ranging from about 0.7 to more than 4.0 due to different geological age, original Rb/Sr values and initial ^{87}Sr/^{86}Sr (Faure 1986). After being incorporated into the body through drinking water and the consumption of plants and animals, ^{87}Sr/^{86}Sr can be measured in bone and teeth to reflect the consumer's location at the time the tissue in question formed. Measurements on tooth enamel relate to the time during which the tooth crown formed, and so present an indication of conditions during infancy through to early adolescence, depending on the tooth measured, from c. 4 months to 2-3 years in the case of the first molar (M1) and from c. 3 to 7 years in the case of the second molar (M2), etc. Bone on the other hand continues to remodel, and so provides information relating to the later years of adult life (Hedges et al. 2007; Robin & New 1997).

To study the strontium isotope variations observed in human remains, it is necessary to construct a baseline for the biologically available strontium (BASr) around the site of interest (Bentley 2006; Evans et al. 2009; 2010). The map of the BASr for Ireland (Snoeck et al. submitted) is based on many samples coming from the Burren and surroundings and can thus be used here to contextualize the strontium isotope results obtained on the human remains. With this map, it is possible to compare the local BASr values to those measured on uncremated enamel and cremated bone and identify individuals who consumed the majority of their food close to or far from the site, following the method described in Snoeck et al. (2016a). In short, individuals having strontium isotope ratios compatible with the BASr values of catchments with a radius up to 5 km will be considered to be 'locals'. Farmers are likely to grow most of their crops and keep their animals within this range most of the time (Chisholm 1968; Jones et al. 1999). Individuals are defined as 'regional' if they exhibit a strontium isotope ratio consistent with catchments with a radius of 5–20 km and as 'outsiders' if their strontium isotope ratio is more than two standard deviations from the average BASr value measured for the 20 km radius catchment.

Carbon and oxygen isotope analyses and infrared measurements

In tooth enamel, carbon isotopes reflect overall sources of dietary carbon (Ambrose and Norr 1993), while oxygen isotopes primarily reflect drinking water. Following the expected rainfall gradient, oxygen isotopes in groundwaters are more depleted in ^{18}O in the east than in the west of Ireland (Darling et al. 2003). However, because of the high pyre temperatures reached during cremation, carbon and oxygen in bone exchange with the surrounding combustion atmosphere and in particular with carbon dioxide from the fuel. The final isotope composition represents a complex mixture of carbon and oxygen from the endogenous bone mineral and collagen fractions and that contributed by the fuel used for the cremation (e.g. Zazzo et al. 2011; 2013; Snoeck et al. 2014a; 2016b). It is therefore not possible to directly compare carbon and oxygen isotopes from cremated bone and tooth enamel. Nevertheless, in the case of uncremated enamel, it is possible to observe variations in diet and place of origin, and in the case of cremated bone, together with FTIR (Fourier Transform Infrared) spectroscopy, to assess pyre characteristics such as temperature and ventilation (Lebon et al. 2010; Snoeck et al. 2014b; 2016b). Infrared analyses allow, among other things, for the detection of cyanamide, which has been suggested to appear in bone burned under reducing conditions (Zazzo et al. 2013; Snoeck et al. 2014b; 2016b)

Radiocarbon dating of calcined bone

Twenty years ago, radiocarbon dating of calcined bone (completely white bone burned at temperatures above 650°C) provided reliable results (Lanting and Brindley 1998; Lanting et al. 2001). This has been put to good use in a wide range of archaeological contexts, including Neolithic Ireland (Schulting et al. 2012). While it is important to keep in mind that the old wood effect might impact on the radiocarbon dates of calcined bone (see Hüls et al. 2010; Zazzo et al. 2011; 2012; Snoeck et al. 2014a), reliable results can still be obtained if one can assume that the wood used to burned the deceased, and the deceased are of contemporary age.

Parknabinnia court tomb and its context

The Parknabinnia court tomb (Cl 153 – Figure 1) is located on Roughan Hill within an upland limestone region known as the Burren in north-west County Clare on the west coast of Ireland (Figure 2), an area that seems to have been a particular focus for Neolithic farmers (Jones 2003). The tomb is atypical in that it has a narrow, straight-sided ‘court’ rather than the more usual open court with curving sides, and a short heel-shaped cairn rather than a long, trapezoidal cairn (Jones & Walsh 1996). These features, however, are shared with other court tombs in the immediate and wider region and they seem to form a morphologically distinct north Munster type (Jones 2019). Radiocarbon dates on uncremated human bone suggest that, like many other court tombs across the country, Parknabinnia was probably initially used c. 3700–3570 BC (the earliest date is 3690–3375 cal. BC at 2σ; GU–10578; 4785±60 BP), but unusually, it continued to be used, if intermittently, up into the first half of the third millennium (GU–10575; 4195±55 BP; 2905–2620 cal. BC at 2σ) (Schulting et al. 2012). Two of the four cremated bone fragments on which isotope analyses were carried out were also dated as part of this project (see below).

The skeletal remains from Parknabinnia were recovered from the tomb's two chambers, both of which were partially filled with a matrix of loose stones interspersed with disarticulated human bone (both inhumed and cremated), animal bone, potsherds, and lithic and bone artefacts. The potsherds probably represent 5–10 vessels including two Carinated Bowls, at least two simple bowls, and a decorated bowl. These vessels all probably date to the second quarter of the 4th millennium BC (Brindley 2010). The lithics recovered include leaf-shaped arrowheads, a plano-convex knife and a bifacial knife, and two flat stone beads. The bone artefacts include what appear to be two halves of the same toggle with expanded 'golf-tee' shaped heads, and a portion of what may be a long, tubular bead with an expanded head. The skeletal remains were highly fragmented, but at least twenty individuals are represented, comprising fifteen adults and five sub-adults. The stratigraphy of the site, the radiocarbon dates, and a taphonomic analysis of the skeletal collection indicate that the Neolithic burial rite at Parknabinnia consisted of successive primary inhumations that were rearranged and disturbed by subsequent inhumations (Beckett 2005a; b; 2011; Beckett & Robb 2006).

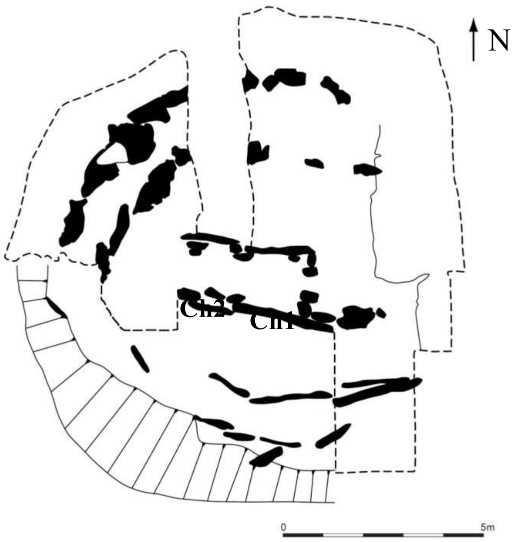

Figure 1 – Plan for Parknabinnia court tomb (Cl 153); re-drawn by Libby Mulqueeny (Schulting et al. 2012)

Geology, palaeoenvironment, and archaeology of the Burren and surrounding region

In order to interpret the strontium isotope results from Parknabinnia, it is important to understand the region's geological, landscape, and human histories. The site is situated on Carboniferous limestone, while Carboniferous shale and sandstone occurs less than 5 km to the west, with older Silurian and Upper-Devonian formations to the east and south-east in the Slieve Aughty Mountains (Geological Survey of Ireland). The soils on Roughan Hill today are thin rendzinas (Finch 1971) with bare bedrock exposed in places, but they are nevertheless more developed than in many parts of the Burren. Micromorphological analysis of the sediments underlying the Parknabinnia court tomb indicated the pre-tomb presence of rendzinas and a clay-rich soil that was probably developed, in part, from glacial drift (Lewis 2003).

There is of course the possibility of soil change since the Neolithic on the Burren, which is now a karstic landscape with limited and fragmentary pockets of soil development (Drew 1982, 1983, 2001; Drew & Treacy 2014). In the past these may have been more extensive, and would have included shale and granite-bearing soils deriving from Co. Galway to the north, via glacial movement (Moles & Moles 2002). These materials would be expected to have higher $^{87}Sr/^{86}Sr$ signatures than the Carboniferous limestone. Analyses of pollen cores from the Burren and its immediate surrounds indicate changes in the vegetation that are probably related to changes in soil cover, beginning with initial clearances of the original pine and hazel forest by Neolithic farmers in the earlier 4th millennium BC (Crabtree 1982; Jelicic & O'Connell 1992; Lamb & Thompson 2005; O'Connell & Molloy 2001; Watts 1984).

The palaeoenvironmental evidence, together with the distribution of known Neolithic monuments, suggests a marked preference for well-drained limestone substrates. A core area of Neolithic settlement is present in the south-east Burren with other, possibly less dense, occupation to the west on the Atlantic coast, to the north around the inner shores of Galway Bay, and to the east across the lower-lying limestone of east Clare and west Tipperary (De Valera & Ó Nualláin 1961; 1972; 1982). In contrast, only limited archaeological evidence of Neolithic settlement exists on the poorly drained sandstone and shale south-west of the Burren, although there is a possible court tomb (Cl 50) located in this area, where the small Moy River meets the Atlantic (De Valera and Ó Nualláin 1961). While the preference for siting Neolithic monuments on limestone is clear, there also appears to be a preference for geological/environmental boundaries. This may reflect economies that utilized ecotonal environments and can be seen in the concentration of Neolithic monuments near the southern edge of the Burren where it borders sandstone and shale-derived soils and also in the siting of the east Clare monuments which are within 3–6 km of the contrasting geological formations of the Slieve Bernagh and Slieve Aughty mountains (De Valera and Ó Nualláin 1961).

Although not particularly abundant, evidence for the spatial distribution of Neolithic settlement other than megalithic tombs does seem to be consistent with the monument distributions, which show a particular focus on the southern Burren and lower levels of activity elsewhere (Hull & Comber 2011; Jones 2003). This impression is reinforced by pre-development work in advance of pipelines and roads passing through the region but avoiding the Burren. Although there are likely sampling biases in these studies, the low number of sites and features pre-dating the Chalcolithic encountered on these projects is striking (Delaney et al. 2012; Grogan et al. 2007; Hull & Taylor 2010).

In the subsequent Chalcolithic and Early Bronze Age periods, areas which had previously witnessed Neolithic activity seem to become particularly important activity foci (Jones et al. 2015), and many formerly more ‘quiet’ parts of the landscape show evidence for settlement and ritual activity as well (Delaney et al. 2012; Hull & Taylor 2010). The Neolithic Parknabinnia court tomb is, in fact, set in the midst of a ‘busy’ Chalcolithic/Early Bronze Age landscape consisting of the densest concentration of Chalcolithic megalithic wedge tombs in the country along with Chalcolithic/Early Bronze habitation enclosures and field walls (Jones 2016).

MATERIALS AND METHODS

Samples

Only a small number of calcined bone fragments (c. 50 identifiable as human, with a larger number of small unidentified fragments; Beckett 2005a: 227) were encountered in the excavations, and are insufficient in weight to represent even a single individual. The assumption is, then, that these are token deposits from an unknown number of cremations carried out elsewhere. Uncremated teeth, all isolated specimens, were also limited in number. Here, four enamel (T) and four cremated bone samples (B) were selected for infrared, strontium, carbon and oxygen isotope analyses (Table 1). Two cremated bone samples (B1 and B2) were selected for radiocarbon dating.

Table 1 – Human remains from Parknabinnia

Find	Chamber	Context	Element	Notes
B1_986.01	1	443	skull frag	/
B2_1018.01		443	skull frag	/
B3_1959.02		582	skull frags	/
T1_1728.04		565	lower M1	minimal wear
T2_1908.11		582	upper M2	minimal wear; root fully formed
B4_1433.01	2	533	skull frag	/
T3_1952.01		584	incisor	lightly worn, dentine exposed
T4_2099.01		586	lower M1	cusps lightly worn, exposing dentine

Three cremated bone and two enamel samples came from Chamber 1, while the other two enamel samples and one cremated bone sample came from Chamber 2 (Figure 1). One of the cremated bone samples was analysed in triplicate (B3a–c). Based on the stratigraphic evidence and their state of attrition, T3 and T4 (slightly worn, exposing dentine) could be from the same individual, as could T1 and T2 (minimally worn, with no dentine exposure). The calcined bone fragments are assumed to belong to different individuals than those represented by the teeth, since cremation would not leave these unaffected. B1 and B2 were found in the same context and could, therefore, derive from the same individual. As discussed below, however, radiocarbon dating of these two samples shows that they represent distinct individuals, both post-dating the Neolithic. The demonstrable problems with stratigraphic integrity make it difficult to determine the number of individuals represented. At minimum, there must be four individuals: two uncremated and two cremated. We return to this below.

Analytical procedures and definition of 'local'

Details on the various analytical procedures and the geographic assignment of each individual can be found in Supplementary Information.

RESULTS

Biologically available strontium (BASr) in the Burren and surroundings

The map of the biologically available strontium for Ireland (Snoeck et al. submitted) shows that only one geological area within 75 km of Parknabinnia presents lower strontium isotope values relative to the local BASr range (0.7086 ± 0.0004 – Table 2): Carboniferous volcanics and minor intrusions to the south-east (Figure 2). However, several geological formations present higher $^{87}Sr/^{86}Sr$ values: Upper Devonian to Lower Carboniferous Old Red Sandstone of the Slieve Bernagh mountains east of Parknabinnia; the Ordovician granite formation in Connemara to the north; and the Carboniferous sandstone and shales of west Clare, etc. To evaluate their possible contribution to the strontium isotope values of the studied individuals, average BASr values for 1, 5, 10 and 20 km catchments were calculated for both Parknabinnia and Poulnabrone (Table 2).

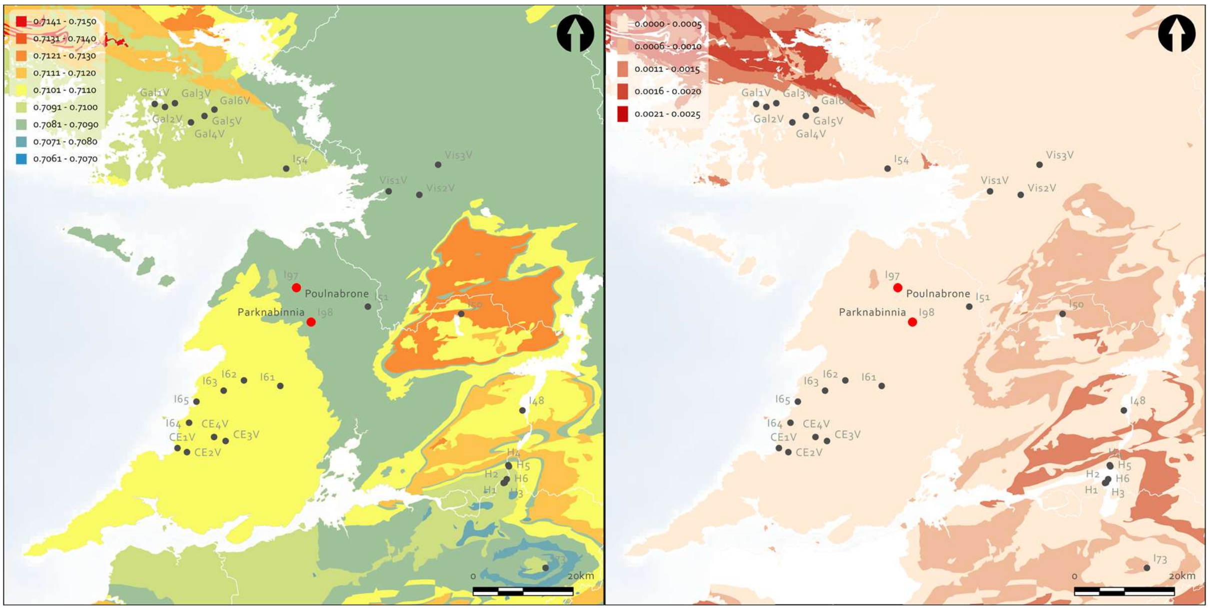

Figure 2 – BASr map of the region around Parknabinnia and Poulnabrone (left = median and right = median absolute deviation) based on Snoeck et al. (submitted). A thin coastal strip (50m) is expected to exhibit values near 0.7092 due to the sea-spray effect.

Table 2 – BASr for the outcrop on which each site lies ('local BASr') and the average BASr values calculated for 1, 5, 10 and 20 km catchments (± 2SD); the values between brackets represent the number of different geological formations included in the calculation of the average BASr

	Site BASr	1km BASr	5km BASr	10km BASr	20km BASr
Poulnabrone	0.7086 ± 0.0004 (1)		0.7089 ± 0.0004 (2)	0.7090 ± 0.0005 (2)	0.7091 ± 0.0006 (2)
Parknabinnia			0.7089 ± 0.0004 (2)	0.7091 ± 0.0006 (2)	0.7093 ± 0.0006 (3)

Radiocarbon dating results

Two cremated bone fragments were radiocarbon dated (B1 and B2). Stratigraphically, these were incorporated into Neolithic deposits, but the results show that they are much later intrusions. B1 returned an early Chalcolithic date (UBA-31468: 2456 – 2415 cal. BC) while B2 is significantly more recent, falling within the Early Bronze Age (UBA-32467: cal. 1971 – 1687 BC) (Table 3). Both dates are several centuries younger than the latest dates on uncremated bone from the court tomb and indicate a previously unknown re-use of the tomb (Figure 3).

Table 3 – Radiocarbon dating results for the cremated bone

Find no.	Lab code	Element	^{14}C yr BP	cal BC (95%)
B1_986.01	UBA-31468	skull frag	3824 ± 34	2456 – 2415 BC
B2_1018.01	UBA-32467	skull frag	3502 ± 55	1971 – 1687 BC

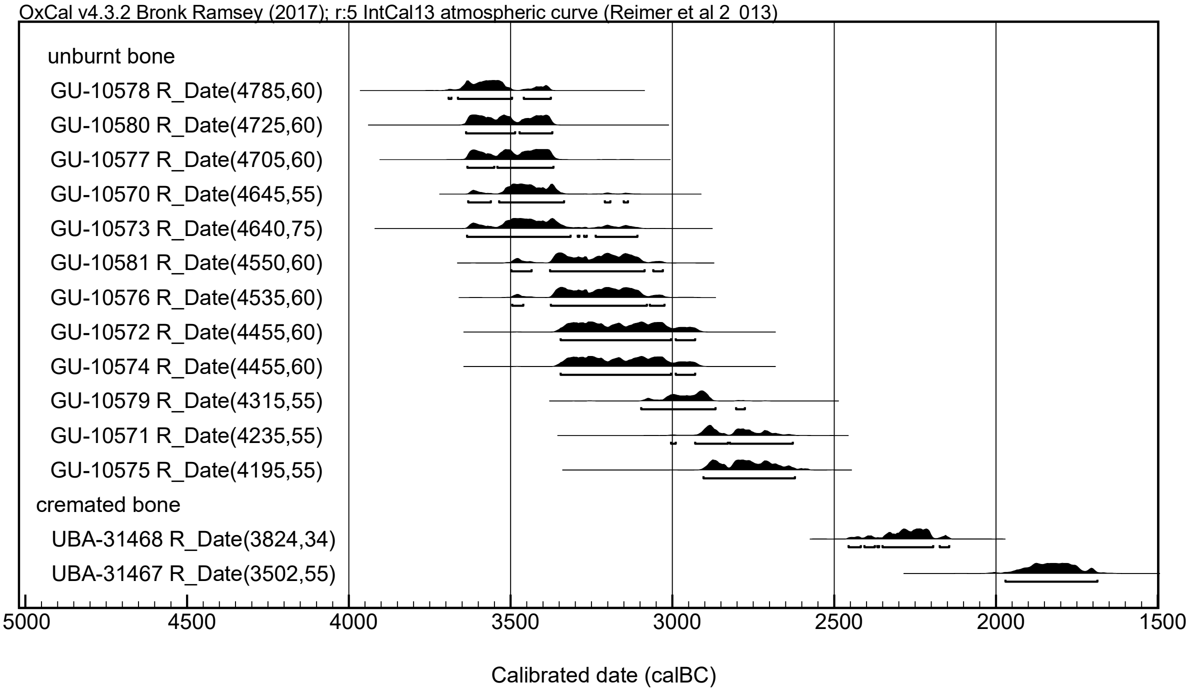

Figure 3 – Radiocarbon dates obtained on uncremated (Schulting et al. 2012) and cremated bone (this study) from Parknabinnia.

Infrared, elemental and isotope results

The infrared and carbon isotope results (Table 4) show that cremated bone samples B1 and B4 differ substantially from B2 and B3, having lower carbonate content (BPI – Type B Carbonate to Phosphate Index) and higher cyanamide content (CN/P). Furthermore, B1 and B4 are more enriched in ^{13}C than B2 and B3. B2 also has higher δ^{18}O values compared to the other samples (Figure 4a). The variability observed in the δ^{13}C and δ^{18}O values of the three B3 samples is not altogether surprising, as this depends mainly on the composition of the combustion atmosphere during cremation, which will be highly variable (Snoeck et al. 2014a; Snoeck et al. 2016b). Even taking into account the large variability observed in B3, the δ^{13}C values of B1 and B4 are still distinguished from B2 and B3.

The tooth enamel samples have similar carbon and oxygen isotope ratios (Figure 4b). The δ^{13}C values are typical of a terrestrial C$_3$-plant based diet (Zazzo et al. 2010). As would be expected, the oxygen isotope values are all enriched in ^{18}O compared to those observed for human and faunal remains from Dublin on Ireland's east coast (Knudson et al. 2012).

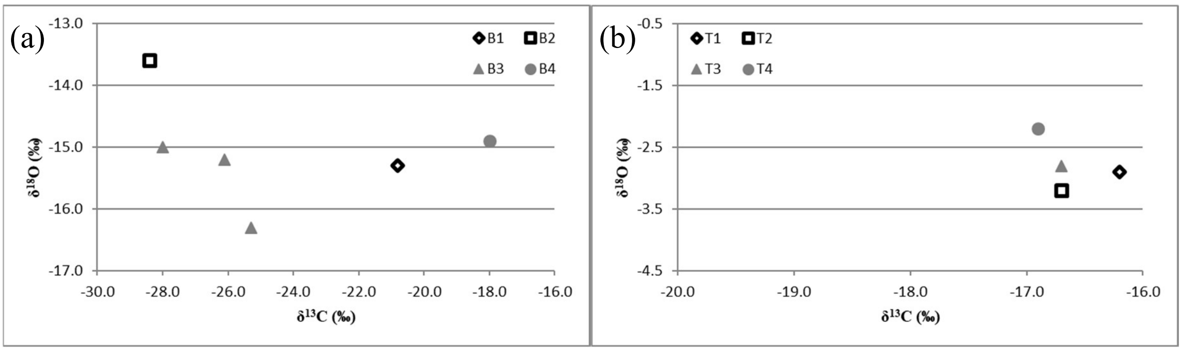

Figure 4 – $\delta^{13}C_{VPDB}$ vs $\delta^{18}O_{VPDB}$ of (a) the cremated bone and (b) the tooth enamel samples; note the difference in the $\delta^{13}C$ scales

Table 4 – Infrared, elemental and isotopic results for tooth enamel (T) and cremated bone (B)

	BPI	IRSF	CN/P	$^{87}Sr/^{86}Sr$ (± 2σ)	[Sr] (ppm)	$\delta^{13}C_{VPDB}$ (‰)	$\delta^{18}O_{VPDB}$ (‰)
B1	0.21	4.17	0.52	0.711166 ± 10	47	-20.8	-15.3
B2	0.49	3.76	0.12	0.709881 ± 08	39	-28.4	-13.6
B3a	0.40	4.62	0.12	0.710134 ± 07	37	-25.3	-16.3
B3b	0.45	4.69	0.13	0.709818 ± 07	*	-26.1	-15.2
B3c	0.48	4.36	0.12	0.710212 ± 10	36	-28.0	-15.0
B4	0.18	4.29	1.34	0.711371 ± 08	44	-18.0	-14.9
T1	0.53	3.52	/	0.709612 ± 11	81	-16.2	-2.9
T2	0.50	3.47	/	0.710142 ± 07	98	-16.7	-3.2
T3	0.60	3.40	/	0.708883 ± 07	50	-16.7	-2.8
T4	0.56	3.37	/	0.708728 ± 08	48	-16.9	-2.2

*Not enough material remaining from B3b for strontium concentration measurement

The $^{87}Sr/^{86}Sr$ results clearly fall into three groups: one (B1 and B4 – Chamber 1 and 2 respectively) with ratios higher than 0.7112; a second (B2, B3, T1 and T2 – all Chamber 1) with intermediate ratios from 0.7096 to 0.7102; and a final group (T3 and T4 – both Chamber 2) with lower ratios of 0.7086/7 (Figure 5). As would be expected, the results obtained for the three sub-samples (B3a, B3b and B3c) of the same calcined bone fragment exhibit limited variation in $^{87}Sr/^{86}Sr$ values (0.0004). Once again, B1 and B4 differ from B2 and B3.

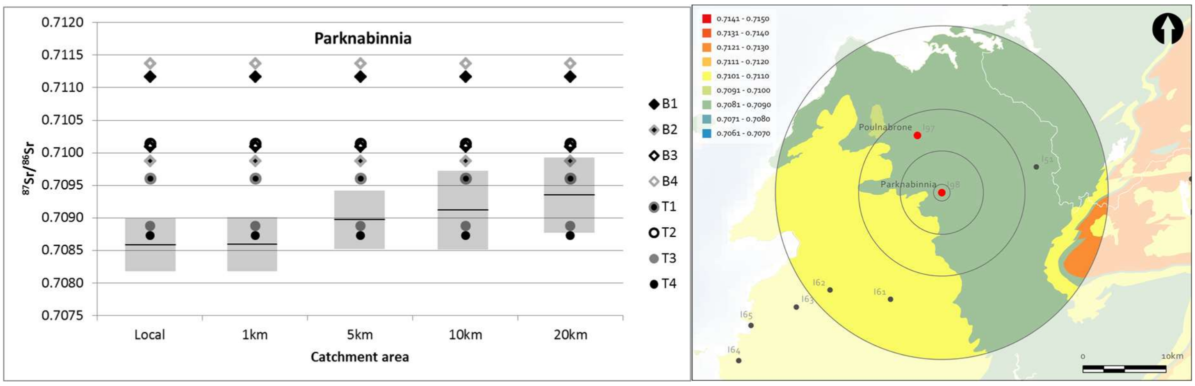

Figure 5 – Strontium isotope results for the uncremated enamel and calcined bone samples from Parknabinnia; the grey shaded areas correspond to the average BASr values calculated for the different catchments (± 2SD) (see Table 2)

Combining the strontium isotope ratios with strontium concentrations further highlights the differences between B1/B4 and B2/B3 as well as between T1/T2 and T3/T4. Two sub-samples of B3 (a and c) returned identical concentrations. The similar isotope ratios and concentrations measured for B1/B4, B2/B3 and T3/T4 suggest that each group originated from a similar place and had a similar dietary plant/meat ratio. T1 and T2, however, present different strontium isotope ratios and concentrations. Strong positive linear correlations are observed for both the tooth enamel ($r^2 = 0.99$, $p = 0.003$) and calcined bone samples ($r^2 = 0.78$, $p = 0.116$). While the correlation for the enamel is statistically significant, both results should be taken as provisional given the small sample sizes, especially as it is difficult to be certain of the number of individuals represented by these samples.

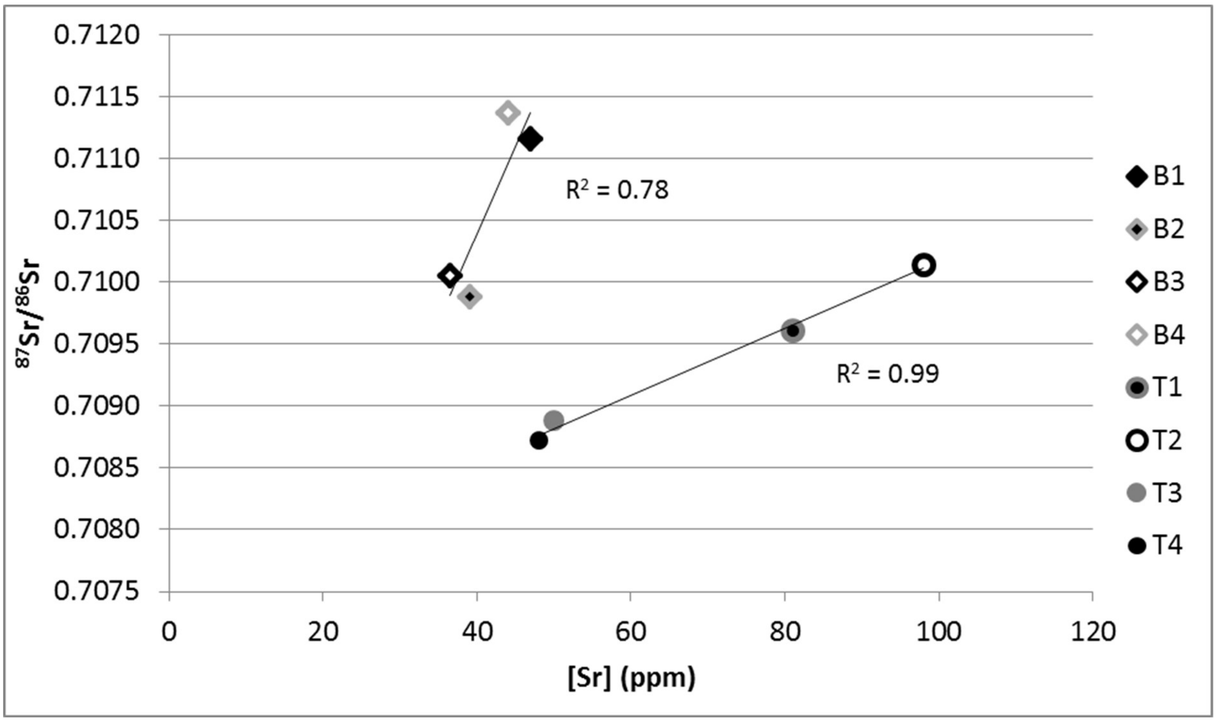

Figure 6 – Strontium isotope ratios versus strontium concentration results for the uncremated enamel and calcined bone samples from Parknabinnia

DISCUSSION

The present project sought to compare $^{87}Sr/^{86}Sr$ values for uncremated dental enamel and cremated bone from Parknabinnia, testing the hypothesis that the former would be more likely to represent individuals from the immediate locality, while the latter might represent the token remains of individuals from further afield, since cremation would facilitate their transportation. The results suggest that the situation is more complex than this, particularly since radiocarbon dates reveal that at least some, and probably all, of the cremated bone substantially post-dates the Neolithic use of the tomb. This highlights the need for caution when material from potentially mixed deposits is being analysed. Without the dates, the interpretation of the ^{87}Sr/^{86}Sr results would have been very different, offering some support for the hypothesised difference between the subsistence catchments represented by uncremated and cremated human remains.

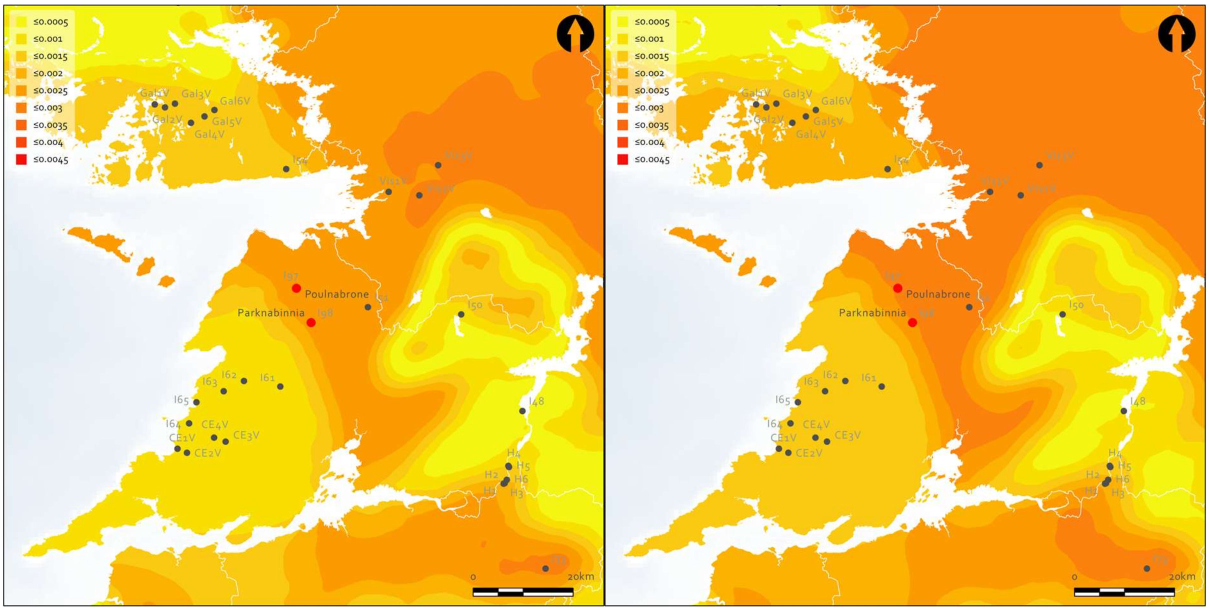

Figure 7 – Geographic assignments of 'non-local' samples B1 (left) and B4 (right), based on the absolute values of the residuals between the sample strontium isotope ratio and the 5km focal mean for the BASr.

Two enamel samples (T3 and T4) from Chamber 2 have ^{87}Sr/^{86}Sr values consistent with the biologically available strontium isotope range for the immediate vicinity of the site, as well as its 1 and 5 km catchments (together defined as 'local'). The two other enamel and two cremated bone samples (defined as 'regional individuals' and all from Chamber 1) have strontium isotope ratios intermediate between the local BASr value and the adjacent Carboniferous sandstone and shale BASr value located 2–3 km to the west. This falls outside of the 'local' range because of the small proportion of the more radiogenic lithological formations contained within the 0-5 km catchment. The last two samples (B1 and B4) can be clearly characterised as 'outsiders' (see SI), having values above 0.7110, consistent with the Slieve Aughty uplands to the east and south-east (Figure 7). The strontium isotope ratios of T3 and T4 (0.7087–0.7088) suggest that the limestone ^{87}Sr/^{86}Sr signal of the Burren was only minimally affected by surficial glacial soils with significantly different values, and hence, at least some of the individuals represented by the calcined bone are likely to be 'outsiders'. While it is relatively straightforward to evaluate the possible place of origin of T3 and T4 ('locals'), and the later lives of B1 and B4 ('outsiders' probably from the Slieve Aughty uplands), the situation with the remaining four samples (T1, T2, B2 and B3) requires further discussion. Indeed, their ^{87}Sr/^{86}Sr values could be the result of consuming a combination of local resources and resources originating from the Slieve Aughty mountains (mudstone/sandstone), and/or those of the adjacent shale and sandstone region of west Clare.

The provisional linear correlation observed in the strontium concentration versus strontium isotope ratios of the tooth enamel fragments (Figure 6) suggests two main sources of strontium, with different strontium-to-calcium ratios. The individual or individuals represented by T3 and T4 clearly consumed foods locally, while T1 and T2 moved between a region with higher Sr/Ca ratios and $^{87}Sr/^{86}Sr$ values reflecting the local limestone. The difference in strontium isotope ratio and concentration for T1 and T2 suggests that these represent two distinct individuals, and that T2 either consumed more food with a higher Sr/Ca ratio than T1, or, those resources were obtained from a geological context with higher Sr/Ca ratio.

The fact that B1 and B4 differ in infrared and in $\delta^{13}C$ and $^{87}Sr/^{86}Sr$ values compared to B2 and B3 suggests that the former two derive from a different region. Incidentally, this, together with non-overlapping radiocarbon dates, also indicates that B1 and B2 are distinct individuals as both are cranial fragments in which we would expect to see similar turnover. Furthermore, the low $\delta^{13}C$ values and the presence of cyanamide in B1 and B4 could indicate that these bones were cremated in different conditions compared to B2 and B3 (Zazzo et al. 2013; Snoeck et al. 2014b), possibly on smaller and/or less well ventilated pyres, where low O_2 facilitates the incorporation of cyanamide (Snoeck et al. 2016b). Considering all of the above, at least five individuals are represented: T1, T2, T3/T4, B1/B4 and B2/B3.

Parknabinnia and Poulnabrone

While there are limited strontium isotope data available to compare with Parknabinnia, the nearby portal tomb of Poulnabrone, Co. Clare, situated on the same geological formations, provides a $^{87}Sr/^{86}Sr$ dataset on uncremated enamel (Ditchfield 2014; Kador 2010; Kador et al. 2014). The onset of burial activity begins earlier here than at Parknabinnia by one to two centuries, but the two sites overlap in use (Schulting 2014). All but one individual at Poulnabrone exhibit values between 0.7082 and 0.7092, consistent with the local BASr value (0.7086 ± 0.0004; n = 17) as well as with those calculated for 1, 5, 10 and 20 km catchments, suggesting that most individuals interred at the site originated in this part of north-west Clare. The single 'non-local' ('regional' in the present framework) at Poulnabrone has a value of 0.7102, similar to those from chamber 1 at Parknabinnia (T1, T2, B2, B3), and could have consumed food from the limestone/shale geological divide to the west or from the Slieve Aughty mountains. This individual has been directly dated to the earlier Neolithic (Ditchfield 2014: Table 4.38). The evidence for only one ‘non-local’ (6%) at Poulnabrone (Figure 8), contrasts with Parknabinnia where only T3/T4 is consistent with the local BASr.

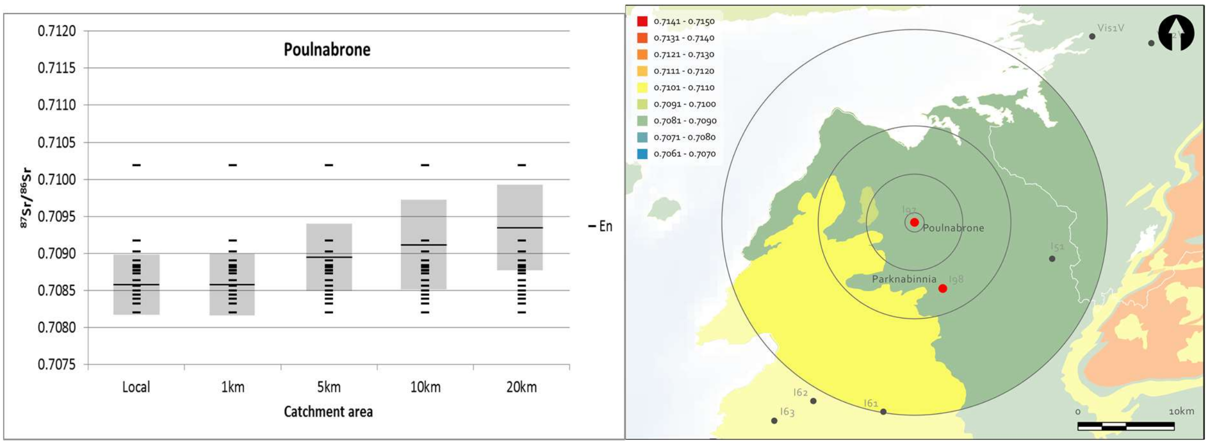

Figure 8 – Strontium isotope results for the uncremated enamel samples from Poulnabrone (Kador 2010; Kador 2014; Ditchfield 2014); the grey shaded areas correspond to the average BASr values calculated for the different catchments (± 2SD)

The calcined remains at Parknabinnia could have been brought to the site as token deposits after cremation at a more distant location during the Chalcolithic and Early Bronze Age re-use of the tomb, which is in line with the observation that the cyanamide content and carbon isotope ratios are highly variable, and also with the small amount of material recovered. In contrast, it appears that inhumation of local and regional individuals characterized the Neolithic deposits. It is also possible that the differing local and regional ^{87}Sr/^{86}Sr values obtained from the tooth samples indicate that different subsets of the local population at Parknabinnia and Poulnabrone used different areas of the landscape as their fields and pastures in the Neolithic (e.g., Bogaard et al. 2011).

Given that the strontium isotope analysis of cremated bone is relatively new, few data are available for comparison; nevertheless, cremated bone from northern Ireland dating to the Neolithic and Bronze Age also showed a majority of ‘non-local’ individuals with nine out of fifteen samples showing a ‘regional’ or ‘outsider’ signal (Snoeck et al. 2016a) which is consistent with the results obtained for Parknabinnia where all four calcined samples can be classified as ‘non-local’ (‘regional’ or ‘outsider’).

CONCLUSION

Taking advantage of the recent demonstration that cremated bone provides a reliable substrate for strontium isotope mobility studies, we investigated human mobility represented by calcined and uncremated human remains at the earlier Neolithic court tomb of Parknabinnia on the Burren of Co. Clare, western Ireland. At both Parknabinnia and Poulnabrone, uncremated tooth samples belong to local or regional individuals, while calcined bone from Parknabinnia belongs to regional individuals or migrants from other parts of Ireland or further afield, suggesting that a selection of their remains were brought to the site as token deposits after cremation elsewhere. Both calcined bone samples that were radiocarbon dated in this study (two out of the four samples subjected to strontium isotope analysis) dated to the Chalcolithic and the Early Bronze Age, and so it is likely that all the cremated material post-dates the Neolithic use of the tomb. However, leaving the calcined bones aside, there is still a contrast with Poulnabrone in that two out of three (or four) individuals represented by the uncremated tooth samples from Parknabinnia belong to regional individuals.

Many excavated court and portal tombs have been found to contain both cremated and uncremated human remains; some, such as Creggandevesky, Co. Tyrone, held only cremated remains, though this may be because of differential survival. Further studies incorporating this material will no doubt provide new data for the investigation of mobility not only in the Irish Neolithic, but also in the Chalcolithic and Early Bronze Age (periods that saw the re-use of both monument types and the continued use of both funerary rites), as well as new insights into the reasons underlying the two funerary rites. In this regard, the finding that the cremated remains at Parknabinnia appear to represent Chalcolithic and Early Bronze Age individuals spending most of their final years away from the site is of particular interest. The extent to which it can be extended to other sites remains to be determined.

The results of this study clearly show that, by combining a wide range of analytical methods, it is possible to better evaluate the minimum number of individuals, ascertain the re-use of a Neolithic monument in later periods, and assess landscape use and possible origins of both uncremated and cremated individuals in prehistory.

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Isotopic evidence for changing mobility and landscape use between the Neolithic and Early Bronze Age in western Ireland

SUPPLEMENTARY INFORMATION

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Infrared analyses

Cremated bone was screened using Fourier Transform Infrared Spectroscopy in Attenuated Total Reflectance mode (FTIR-ATR – Agilent Technologies Cary 640 FTIR with GladiATRTM from Pike Technologies). Various infrared indices were calculated for each sample (Table S1 – see Lebon et al. 2010 & Snoeck et al. 2014b for more details). The cyanamide to phosphate ratio (CN/P) is of particular interest as it characterizes the presence of cyanamide, which has been suggested to be present when bone is cremated in reducing conditions (Zazzo et al. 2013; Snoeck et al. 2014b; 2016b).

Table S1 – Useful infrared indices for the study of calcined bone apatite

Name	Measurement	Significance	Reference
BPI	A_{1415}/A_{605}	Amount of type B carbonates	1,2
IRSF	$(A_{605} + A_{565}) / A_{590}$	Crystallinity	3
CN/P	A_{2010} / A_{605}	Amount of cyanamide	4,5

[1]LeGeros & LeGeros 1983; [2]Sponheimer & Lee-Thorp 1999; [3]Weiner & Bar-Yosef 1990; [4]modified from Zazzo et al. 2013; [5]Snoeck et al. 2014b

Radiocarbon dating of cremated bone

The cremated bones were cleaned with a dremel tool, ground in a mortar and pestle and placed in 100 mL beakers. Sodium hypochlorite solution (20 mL, 1.5%) was added to the samples which were allowed to stand for 48 hours to remove any remaining protein. The samples were then vacuum-filtered on a pre-baked glass-fibre filter, washed with deionised water and returned to the beaker. 20 mL of 1M acetic acid were added and allowed to stand for 24 hours to remove contaminant carbonate. The samples were again vacuum-filtered, washed with deionised water, returned to the beakers and placed in a drying oven overnight at 60º. The samples were stored in a sealed vial prior to hydrolysis with orthophosphoric acid (85%, 15 mL per g of bone). The sample gas obtained by the hydrolysis was combusted in a closed tube using an excess of copper oxide and a strip of silver foil ribbon (to remove sulphur) at 850°C for 8 hours prior to graphitization.

Samples were converted to graphite on an iron catalyst using the zinc reduction method (Slota et al. 1987). The $^{14}C/^{12}C$ and $^{13}C/^{12}C$ ratios were measured by accelerator mass spectrometry (AMS) at the ^{14}CHRONO Centre, Queen's University Belfast. The sample $^{14}C/^{12}C$ ratio was background corrected and normalised to the HOXII standard (SRM 4990C; National Institute of Standards and Technology). The radiocarbon ages were corrected for isotope fractionation using the AMS measured $\delta^{13}C$ which accounts for both natural and machine fractionation. The radiocarbon age and one standard deviation were calculated using the Libby half-life of 5568 years following the methods of Stuiver and Polach (1977).

The resulting AMS determinations were calibrated using the IntCal13 curve (Reimer et al. 2013), using OxCal v4.2.4 (Bronk Ramsey 2013). All calibrated ages are reported at 95.4% confidence.

Carbon and oxygen isotope analyses

For carbon and oxygen isotope analyses, the cremated bone samples were first treated for 1 hour in sodium hypochlorite (NaOCl 1%) followed by 4 hours in acetic acid (1M). The enamel samples were soaked in sodium hypochlorite (NaOCl 1%) for 1 hour but only for 30 minutes in calcium acetate buffered acetic acid (1M). The samples were then analysed on a Kiel IV carbonate device, interfaced with a Delta V Advantage isotope ratio mass spectrometer at the SSMIM (Service de Spectrométrie de Masse Isotopique du Muséum national d'Histoire naturelle), Paris, France. A laboratory carbonate standard (LM marble) normalized with NBS

19 and giving mean $\delta^{13}C$ and $\delta^{18}O$ values of +2.08 ± 0.04 ‰ and +1.70 ± 0.05 ‰ respectively was used to check the accuracy of the data. We used these standard deviations as an indicator of analytical precision over the period of analysis.

Strontium isotope analyses

Before analyses, the tooth enamel and cremated bone samples (c. 50 mg) were rinsed three times with milliQ water. For each rinsing, the samples were placed for 10 minutes in an ultrasonication bath. Cremated bone fragments were treated with 1M acetic acid for 3 minutes in the ultrasonication bath and then rinsed with milliQ water and 10 minutes ultrasonication. Tooth enamel fragments underwent the same pre-treatment but were left for 30 minutes in the ultrasonicating bath during the acetic acid treatment (Snoeck et al. 2015).

Strontium was extracted from the samples and purified following the protocol described in Snoeck et al. (2015) and measured on a Nu Plasma MC-ICP Mass Spectrometer (Nu015 from Nu Instruments, Wrexham, UK) at the Université Libre de Bruxelles (ULB). During the course of this study, repeated measurements of the NBS987 standard yielded $^{87}Sr/^{86}Sr$ = 0.710214±40 (2SD for 15 analyses), which is, for our purposes, sufficiently consistent with the mean value of 0.710252±13 (2SD for 88 analyses) obtained by TIMS (Thermal Ionization Mass Spectrometry) instrumentation (Weis et al. 2006). All the sample measurements were normalised using a standard bracketing method with the recommended value of $^{87}Sr/^{86}Sr$ = 0.710248 (Weis et al. 2006). Procedural blanks were considered negligible (total Sr (V) of max 0.02 versus 7–8V for analyses; i.e. ≈ 0.3%). For each sample the $^{87}Sr/^{86}Sr$ value is reported with a 2σ error (absolute error value of the individual sample analysis – internal error).

Strontium concentration analyses

Small sample fractions (~2 to 5 mg) pre-treated as above were digested in precleaned Teflon beakers (Savillex) using subboiled 7 M HNO_3 at 120˚C for 24h, evaporated to near-dryness and subsequently digested with a drop of concentrated HNO_3. Following dilution with 2% HNO_3, Sr concentrations (ppm) in the sample digests were determined using a Thermo Scientific Element 2 sector field ICP mass spectrometer at the Vrije Universiteit Brussel (VUB, Belgium) in low (^{86}Sr and ^{88}Sr) and medium (^{43}Ca and ^{44}Ca) resolution using Indium (In) as an internal standard and external calibration versus three ISO certified standards (BAS CCB01 cremated bone apatite, BAS CRM512 dolomite and BAS ECRM782-1 dolomite) from the Bureau of Analyzed Samples Ltd. (BAS, Middlesbrough, UK). Based on repeated digestion and measurement of this reference material, the analytical precision (1SD) of the procedure outlined above is estimated to be better than 3%.

Biologically available strontium and definition of non-locals

A map of the biologically available strontium (BASr) for Ireland was generated using modern plant data and Geological Survey Ireland (GSI) Bedrock Geology 500k (1:500,00) polygon data (https://www.gsi.ie/en-ie/data-and-maps/Pages/Bedrock.aspx#500k) (Snoeck et al. in press). Single part features were created from the GSI polygons, with individual polygons for spatially discrete outcrops of each geological formation. Descriptive statistics (count, minimum, maximum, range, mean, standard deviation, lower quartile, median, upper quartile, interquartile range and median absolute deviation) were calculated by aggregating samples that intersect each polygon. Where no samples intersect a polygon, descriptive statistics are calculated based in order of preference on samples from other polygons of the same formation or other polygons of the same age/type. The 83 formations in Ireland are grouped into 7 groups, namely: 1) metamorphic; 2) felsic/ intermediate igneous; 3) clastic sedimentary (Ordovician to Devonian); 4) clastic sedimentary (Carboniferous to Cretaceous); 5) chemical sedimentary; 6) mafic igneous; 7) clay/sand. The method used to generate the BASr map is described in Snoeck et al. (submitted) and takes into account inputs from sea spray by defining a polygon for the coastal zone as a 50m buffer and inputs from rainfall and superficial geology by creating single part polygons.

To calculate the BASr values of the catchments used to classify individuals as ‘local’, ‘regional’ or ‘outsider’, the BASr values of the geological formations were averaged, and weighted by their proportional representation in the catchment. It is assumed that plants growing on the different soil types within a catchment contribute to this average according to their proportional representation (Snoeck et al. 2016a).

Geographic assignments for the ‘outsiders’ were calculated by: 1) converting the map of BASr to a raster dataset with a cell size of 100m; 2) calculating focal statistics for the raster dataset based on a 5km cell neighbourhood; 3) calculating the absolute value of the residuals between the strontium isotope ratio for the individual and the 5km focal mean (Pouncett in press). The method used to calculate the geographic assignment is a logical extension of the method used to determine whether an individual is local or non-local. Instead of calculating an average for the location where the individual was buried, averages are calculated for every cell in the study area. Calculating the absolute value of the residuals, rather than converting the residuals to a probability density function, allows easier evaluation of how sources of error are likely to impact upon the geographic assignments for each of the individuals.

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