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Reassessing Neolithic Diets in Western Scotland

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Abstract: Although marine resources are known to have been exploited by both foragers and early farmers in Scotland, the importance of seafood to the diets of Neolithic groups has been widely debated. Here we present paired stable isotope ($\delta^{13}C$ and $\delta^{15}N$) and radiocarbon measurements on Early Neolithic human remains from Raschoille Cave in Oban. These are compared with published data for other sites in western Scotland and used to re-evaluate the use of marine resources by the first farmers. The diets of Late Mesolithic foragers and Early Neolithic farmers were modelled from stable isotope data using both Linear and Bayesian (FRUITS) mixing models. Our FRUITS dietary models indicate that Mesolithic foragers obtained much of their dietary protein and calories from marine resources, consistent with the predominance of shellfish, fish and sea mammal remains in their shell middens. Of note is the large proportion of dietary calories obtained from plant foods, which is like that of the early farming groups. The diets of Early Neolithic farmers appear relatively homogeneous across Scotland. Plant foods were the primary source of calories. Meat and/or dairy from terrestrial mammals were the most important source of dietary protein. Marine resources were, at most, a minor component of the 'lifetime' diet.

Keywords: Raschoille Cave; Scotland; Neolithic; Mesolithic; diet; stable isotopes; FRUITS

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1. Introduction

Changes in human bone collagen carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope values have been cited as evidence of a rapid and comprehensive shift from marine to terrestrial resource dependence at the Mesolithic–Neolithic transition in Scotland [1–4], an interpretation that has gained support from the lipid residue analysis of pottery sherds that found virtually no evidence for processing of marine products in the pots [5]. However, the simple proposition of a large-scale shift from marine to terrestrial resource exploitation, which is based largely on biomolecular evidence, fails to accommodate the archaeological evidence for the use of marine resources in Neolithic Scotland [6]. The remains of shellfish, fish and/or sea mammals are by no means absent from Neolithic sites in coastal areas of northern and western Scotland (Figure 1).

Although the use of marine resources in the Scottish Neolithic is beyond dispute, the importance of those resources to the diet is uncertain [7]. The timing and duration of the Mesolithic–Neolithic transition in Scotland and, by extension, the rapidity of the change from foraging to farming are complex issues that are not easily resolved for want of more precise information on chronology, subsistence, technology and genetic ancestry. Nevertheless, the suggestion that the change was gradual and variable geographically has started to gain traction. Archaeological evidence points to multiple 'pulses' of farmer immigration into Scotland over several centuries, beginning perhaps as early as 4200 cal BC [8], while paired stable isotope and radiocarbon analyses of human remains from Cnoc Coig (on the Inner Hebridean island of Oronsay) suggest continued dependence on marine resources into the fourth millennium BC, leading the researchers to conclude that

"both hunter-gatherer-fisher and farming lifestyles potentially co-existed on the West Coast of Scotland for several hundred years" [9].

Figure 1. Scottish Neolithic sites with evidence for exploitation of marine resources. Small dots = individual sites. Large dots = multiple sites.

The human remains recovered from Raschoille Cave provide an opportunity to further explore Early Neolithic diets in western Scotland. Genetic evidence points to a group with a mixed hunter-gatherer/immigrant farmer ancestry [10], the admixture likely occurring in Scotland in the preceding ten generations [11]. Given the coastal setting and the presence of shellfish and fish remains among the Neolithic deposits, both terrestrial and marine resources may have been exploited by the Raschoille population. Scatterplots and Bayesian models of the stable isotope values of the human remains were generated to gain information on diet and, by inference, the subsistence practices.

1.1. Archaeological and Environmental Context

Raschoille Cave (56°24′12″ N, 5°28′41″ W) is a small cave on the west coast of Scotland, in the town of Oban (Figure 2). The cave lies about 13 meters above sea level and close to the altitudinal limit of the Holocene marine transgression. During the Neolithic, when the relative sea level was still several meters higher than today, the cave occupied a position at the edge of a shallow marine embayment in the lower part of a sheltered coastal

valley known locally as Glenshellach ('Valley of the Willows'), which has an archaeological record of human settlement extending back to the Late Mesolithic c. 6500 cal BC [12].

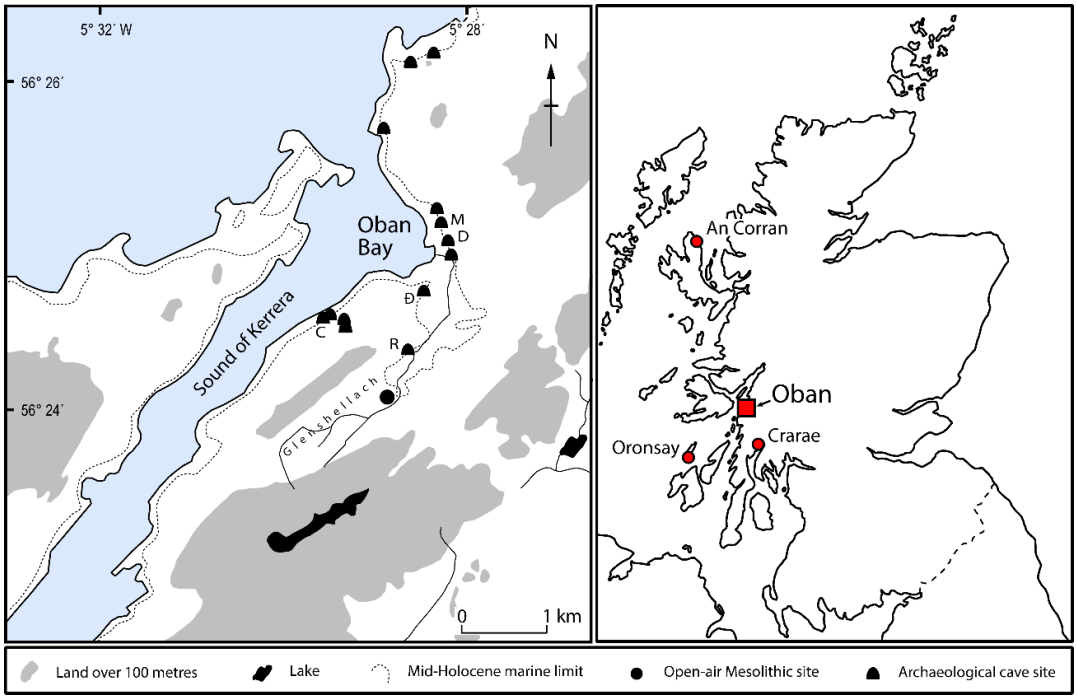

Figure 2. Location of Raschoille Cave and other key sites in the Oban area and central-west Scotland: R—Raschoille Cave; C—Carding Mill Bay I; D—Distillery Cave; Đ—Druimvargie Rockshelter; M—MacArthur Cave.

The cave entrance was revealed during 'groundworks' in 1984. A rescue excavation by local archaeologists recovered c. 2000 disarticulated and commingled human bones from unconsolidated sediments in the cave. These were reported to have occurred in distinct clusters, which were assigned unique context numbers and were interpreted as the result of secondary disposal of excarnated remains [13].

There is no firm evidence of the domestic use of Raschoille Cave during the Neolithic. Most likely it was used as a burial chamber by people living elsewhere in the Glenshellach Valley, with access to farmland as well as natural terrestrial, freshwater and marine resources. A variety of habitats (rocky shores, salt marsh, tidal flats) likely existed within the embayment, as reflected in the diversity of shellfish remains from the cave. Glacially eroded basins within the Glenshellach Valley, which today contain mires, were likely occupied by small freshwater lakes or ponds in the Neolithic.

Post-excavation work on the human remains from Raschoille Cave was beyond the expertise and resources of the local archaeologists and, eventually, a part of the human bone collection was transferred to the Archaeology Department at Edinburgh University for scientific study, supervised by one of us (CB). The remainder of the collection ended up in the Hunterian Museum in Glasgow and was subsequently acquired by the National Museum of Scotland (NMS) in Edinburgh. Ultimately, the two sub-assemblages will be reunited and curated at the NMS.

1.2. Stable Isotope Analysis for Dietary Reconstruction

Carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope analysis of bone collagen is a long-established and reliable tool for reconstructing the diets of archaeological populations [14,15]. The $\delta^{13}C$ and $\delta^{15}N$ values of human tissues, including bone collagen, reflect those of foods consumed. Variation in the $\delta^{13}C$ and $\delta^{15}N$ values of different foods facilitates the reconstruction of the paleodiet. Carbon is assimilated by plants during photosynthesis.

Plant $\delta^{13}C$ values depend on the environmental source of carbon and the photosynthetic fixation pathway for carbon: most plants use either a three-carbon or a four-carbon photosynthetic pathway and are known as C_3 or C_4 plants, respectively. In terrestrial plants, carbon is fixed from atmospheric CO_2. C_3 plants incorporate a greater proportion of the lighter isotope ^{12}C into tissues than C_4 plants. The mean $\delta^{13}C$ value of C_3 plants is −26.5‰, while that of C_4 plants is −12.5‰. There is an offset of c. +5‰ between C_3 plants and consumers' bone collagen; human consumers of C_3 resources typically have bone collagen $\delta^{13}C$ values of c. −21.5‰, while human consumers of C_4 resources typically have bone collagen $\delta^{13}C$ values of c. −7.5‰ [16]. Most temperate region grasses and cereal crops, such as wheat and barley, as well as most fruits and vegetables, are C_3 plants. Many tropical grasses, including cultivars such as maize, millet and sorghum, are C_4 plants. The British Isles with their cool climate have very few native C_4 plants, mostly found growing in salt marsh and coastal dune habitats. Moreover, there is no archaeobotanical evidence that introduced C_4 crops, such as millet, were grown in any part of the British Isles in prehistory [17].

More pertinent to the present study is the utility of carbon and nitrogen stable isotope analyses to determine the proportion of marine vs. terrestrial resources in diet. Most marine plants fix carbon through the C_3 photosynthetic pathway—i.e., using the same photosynthetic pathway as most temperate-region terrestrial plants. However, marine plants generally have higher mean $\delta^{13}C$ values than terrestrial C_3 plants. This is because marine plants fix carbon from oceanic carbonate, which has a relatively enriched ^{13}C content (+7‰) in comparison to atmospheric CO_2. This enrichment in marine plant ^{13}C is passed on through the food web into the tissues of consumers. Humans relying entirely on marine resources typically have bone collagen $\delta^{13}C$ values of c. −13‰ [18]. Analysis of the $\delta^{13}C$ value of human bone collagen may therefore indicate the proportion of the diet drawn from terrestrial C_3 vs. marine food webs.

Co-analysis of a second dietary discriminant, $\delta^{15}N$, adds resolution to paleodietary reconstruction. The $\delta^{15}N$ values of plants also vary, again reflecting differences in the environmental source [19]. The nitrogen incorporated into plants may be drawn from atmospheric N_2 as well as soil nitrogen. Soil $\delta^{15}N$ values vary spatially and temporally. Further variation in $\delta^{15}N$ values is introduced with each step (or trophic level) of the food chain. This trophic level effect is the result of isotopic fractionation during metabolic processes. Within a single biome, plants have lower $\delta^{15}N$ values than herbivores, which in turn have lower values than carnivores [20,21]. The $\delta^{15}N$ values increase by c. 3–6‰ with each trophic stage [22–24]. Bacterial activity in aquatic systems may enrich ^{15}N in aquatic food webs. Additionally, aquatic food chains generally have a larger number of trophic levels than those of terrestrial environments [15,25]. Consequently, human consumers of aquatic resources generally have elevated $\delta^{15}N$ values in comparison with those relying entirely on terrestrial resources. Evaluating bone collagen $\delta^{13}C$ *and* $\delta^{15}N$ values, therefore, distinguishes the relative proportions of terrestrial and aquatic (i.e., marine and freshwater) resources in the diet and may also indicate the trophic level (e.g., plant vs. animal; shellfish vs. fish vs. sea mammal) of resources consumed [14,26,27].

2. Materials and Methods

2.1. Sampling

In total, 28 human bones from Raschoille Cave were sampled for ^{14}C, $\delta^{13}C$ and $\delta^{15}N$ analyses. They comprised three series selected on different occasions for individual research projects. Sample details are provided in Table 1.

Series 1 and 2 were selected from the sub-assemblage studied at Edinburgh University. Series 1 comprised fragments cut from 14 postcranial bones specifically for the dietary reconstruction of the Raschoille population [28,29]. Every effort was made to select each bone from a different individual, based on archaeological context and osteometric criteria. Series 2 consists of five petrous bones that were selected for an archaeogenomic study of prehistoric populations in Scotland [10,11]; these were from different contexts and (according to genetic data) different individuals.

The nine Series 3 samples came from the sub-assemblage in the National Museum of Scotland; the information in Table 1 relating to these samples is reproduced from [30,31]. Five samples (RC.20–25) were petrous bones from separate individuals, but no information is available for the other four samples.

2.2. AMS Dating and Stable Isotope Analyses of Human Bone

The AMS ^{14}C dates and stable isotope ratios of the Series 1 samples were measured at the Oxford Radiocarbon Accelerator Unit (ORAU) in 1999. The samples were submitted as bone fragments, each weighing at least 0.5 g. The Series 2 (petrous) samples were subsampled for a DNA analysis at the David Reich Lab at Harvard University in 2015 before being transferred to the Penn State University AMS Radiocarbon Facility (PSUAMS) for ^{14}C dating and stable isotope analysis.

The two labs used similar procedures for extraction and purification of bone collagen for ^{14}C and stable isotope analyses, based on the modified Longin method [32], in which bone samples were first cleaned manually, decalcified and gelatinized, and the resulting gelatin lyophilized and weighed to determine per cent yield as a measure of collagen preservation. The procedure used at ORAU is essentially that outlined in [33] but excluded the ultrafiltration step (ultrafiltration was not used routinely at ORAU until 2000). Details of the collagen preparation procedure used at PSUAMS, which did include either ultrafiltration or XAD pretreatment, can be found in [34]. As is standard practice in both labs, the collagen quality and chemical integrity were assessed using the atomic ratio of carbon to nitrogen (C:N atomic ratio), the percentage of collagen extracted compared with the starting weight of bone (wt% collagen) and the carbon yield of the collagen on combustion. Bone is considered acceptable if measured C:N ratios of collagen fall between 2.9 and 3.6.

AMS ^{14}C dating and stable isotope analysis of the Series 3 samples were undertaken at the Scottish Universities Environmental Research Centre (SUERC) in East Kilbride. The collagen extraction protocol (which included ultrafiltration) and quality control procedures are comparable to those used at ORAU and PSUAMS and are described in [29].

Table 1. $\delta^{13}C$ and $\delta^{15}N$ values and radiocarbon determinations of human bone collagen samples from Raschoille Cave. Abbreviations: cv—cervical vertebra; L—left; R—right; M—Male; m—probably male; F—female; *—based on DNA evidence; n.d.—no data.

Sample No.	Lab ID	"Layer"	Context	Skeletal Element	Sex	Age	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	%C	%N	C/N	^{14}C Age BP (±1σ)	Cal BC Age Range (2σ)
Series 1 [28,29]**:**													
RC.01	OxA-8537		14	L humerus		1–2 yr.	−21.8	11.9	41.6	14.9	3.3	4535 ± 50	3486–3035
RC.02	OxA-8434	IVa	77.1	R femur		c. 3 yr.	−21.1	8.7	45.0	15.4	3.4	4720 ± 50	3633–3372
RC.03	OxA-8431	III	10.106	L femur		?3–5 yr.	−20.6	8.6	38.7	13.8	3.3	4930 ± 50	3909–3634
RC.04	OxA-8399		17/19	cv		3–7 yr.	−21.4	10.2	39.4	13.7	3.4	4630 ± 65	3628–3104
RC.05	OxA-8432	III	18.17 (+18.21)	R humerus		8–10 yr.	−20.4	7.6	36.4	13.0	3.3	4980 ± 50	3945–3646
RC.06	OxA-8401	II	78.1	L femur		?10 yr.	−21.1	9.1	32.4	11.5	3.3	4565 ± 65	3515–3031
RC.07	OxA-8400		14	rib		Adult	−20.3	9.7	40.3	14.6	3.2	4640 ± 65	3630–3108
RC.08	OxA-8444	V	39.8	R humerus	F	Adult	−21.1	9.6	38.6	13.0	3.5	4715 ± 45	3631–3372
RC.09	OxA-8443	V	39.5	R humerus		Adult	−20.4	9.4	36.7	13.4	3.2	4825 ± 55	3710–3381
RC.10	OxA-8433	IV	79.1	L humerus	m	Adult	−20.2	9.4	38.1	13.7	3.2	4920 ± 50	3903–3543
RC.11	OxA-8404	III	10.119	R humerus		Adult	−21.6	7.7	23.4	8.6	3.2	4850 ± 70	3791–3381
RC.12	OxA-8441	IVa	87.1	R humerus		?Adult	−21.2	9.1	39.0	13.7	3.3	4900 ± 45	3786–3540
RC.13	OxA-8435	V	39.26	Humerus			−22.5	10.3	43.5	15.1	3.4	4680 ± 50	3626–3363
RC.14	OxA-8442	IVa	89.5	R humerus			−21.0	8.7	38.1	13.5	3.3	4890 ± 45	3786–3534
Series 2 [10]**:**													
RC.15	PSUAMS-2068	III	19.1	petrous	M*		−21.5	9.4	43.4	15.7	3.2	4770 ± 30	3638–3386
RC.16	PSUAMS-2069	III	21.3	petrous	M*		−21.0	9.0	46.6	16.3	3.3	4665 ± 30	3516–3370
RC.17	PSUAMS-2154	III	10.4	petrous	M*		−21.8	9.6	21.9	8.6	3.0	4725 ± 20	3627–3377
RC.18	PSUAMS-2155	III	17.21	petrous	M*		−22.2	11.0	25.5	9.4	3.2	4730 ± 25	3630–3377
RC.19	PSUAMS-2156	IVa	87.6	petrous	F*		−22.1	9.4	27.1	10.0	3.2	4415 ± 25	3312–2920
Series 3 [30,31]**:**													
RC.20	GU-40818	III	13.18	petrous	M*		−21.9	8.4	20.0	6.7	3.5	4550 ± 29	3371–3102
RC.21	GU-40819	III	15.13	petrous, L	F*		−21.7	7.7	37.8	13.4	3.3	4738 ± 31	3633–3378
RC.22	GU-40820	IV	34.80	petrous, R	F*		−21.6	7.7	36.3	12.9	3.3	4817 ± 31	3646–3528
RC.23	GU-40821	II	95.20	petrous, R	F*		−22.1	8.8	40.5	14.4	3.3	4490 ± 29	3347–3040
RC.24	GU-40822	III	12.10	petrous, L	F*		−21.5	9.2	28.5	10.1	3.3	4499 ± 29	3351–3096
RC.25	GU-40823	I	2	n.d.			−21.9	9.5	37.8	13.4	3.3	4668 ± 29	3517–3371
RC.26	GU-40824	I	2	n.d.			−22.3	10.2	34.6	12.4	3.2	4432 ± 31	3329–2926
RC.27	GU-40825	I	2	n.d.			−22.4	10.4	41.1	14.6	3.3	4731 ± 29	3631–3377
RC.28	GU-40826	III	31.32	n.d.			−22.2	9.3	39.9	14.0	3.3	4638 ± 31	3516–3359

2.3. Statistical Modelling

Bayesian chronological models of burial activity in Raschoille Cave were created in *OxCal 4.4.4* [35] based on the three ^{14}C date series.

Individual diets were modelled using *Food Reconstruction Using Isotopic Transferred Signals (FRUITS) v. 3.0 beta* [36], assuming five food sources—terrestrial plants, terrestrial herbivores and omnivores, shellfish, fish and seals.

Graphs were prepared using Excel, PAST [37] and Adobe Illustrator.

3. Results

3.1. Archaeological Samples

All the human bones analyzed produced sufficient collagen of acceptable quality (judging by the C:N ratios) for dating and stable isotope analysis. As noted above, the data were produced by three different laboratories, employing similar collagen extraction protocols and quality control procedures, with the exception that the protocol used at Oxford at that time did not include an ultrafiltration step to remove potential contaminants. While this may have influenced the ^{14}C measurements (see Discussion), it appears to have had little or no effect on the stable isotope values which seem consistent between sample series 1–3 (cf. Sealy et al., 2014) [38].

Tables 2 and 3 present our baseline stable isotope data for terrestrial and marine food sources, which were used to inform the FRUITS models. The terrestrial data are drawn from two sites in Oban, Raschoille Cave and Carding Mill Bay I (Figure 2), to provide a 'local' baseline, while those for marine resources are (of necessity) derived from a wider area of central-west Scotland.

Table 2. $\delta^{13}C$ and $\delta^{15}N$ data for 'local' terrestrial fauna used for the FRUITS models.

Lab ID	Site	Context	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	Reference
GUsi3497	Carding Mill Bay I	medium mammal (*Ovis aries* or *C. capreolus*)	−21.6	3.5	[39]
GUsi3507	Carding Mill Bay I	medium mammal (*Ovis aries* or *C. capreolus*)	−22.9	3.7	[39]
GUsi3498	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−23.3	3.4	[39]
GUsi3500	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.8	2.8	[39]
GUsi3501	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−23.1	3.1	[39]
GUsi3502	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.8	2.8	[39]
GUsi3503	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.5	2.7	[39]
GUsi3504	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.5	3.1	[39]
GUsi3505	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−23.2	3.7	[39]
GUsi3506	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.5	2.4	[39]
GUsi3508	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−23.2	2.3	[39]
GUsi3511	Carding Mill Bay I	large mammal (*C. elaphus* or *Bos taurus*)	−22.8	3.9	[39]
GU39625*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.3	3.5	[39]
GU39626*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.2	3.3	[39]
GU39627*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.3	3.5	[39]
GU39628*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.4	3.4	[39]
GU39629*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−22.6	3.8	[39]
GU39630*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.3	4.2	[39]
GU39631*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−23.4	3.4	[39]
GU39632*	Carding Mill Bay I	probable cattle (*Bos taurus*)	−22.0	3.0	[39]
C XXIV:2	Carding Mill Bay I	pig (*Sus* sp.)	−21.9	3.2	[2]
GUsi3509	Carding Mill Bay I	red deer (*C. elaphus*)	−23.2	3.0	[39]
CVII:123	Carding Mill Bay I	red deer (*C. elaphus*)	−21.9	2.0	[2]
C XVII:4	Carding Mill Bay I	red deer (*C. elaphus*)	−22.9	2.5	[2]
OxA-8396*	Raschoille Cave	red deer (*C. elaphus*)	−21.8	2.9	[28,29]
OxA-8397*	Raschoille Cave	red deer (*C. elaphus*)	−21.5	2.8	[28,29]
OxA-8398*	Raschoille Cave	red deer (*C. elaphus*)	−21.6	2.6	[28,29]

Table 3. $\delta^{13}C$ and $\delta^{15}N$ data for marine resources from central-west Scotland used for the FRUITS models.

Lab ID	Site	Species	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	Reference
0055-r	An Corran	cod	−13.6	15.3	[40]
703	Bornish 12th-13th C	cod	−12.9	14.5	[41]
706	Bornish 13th C	cod	−11.3	15.4	[41]
708	Bornish 13th C	cod	−13.1	13.8	[41]
713	Bornish 12th-13th C	cod	−13.2	13.8	[41]
C VI:21	Carding Mill Bay I	otter	−12.0	16.0	[2]
n/a	Oronsay	grey seal	−11.9	19.1	[42]
10502	Cnoc Coig	seal	−11.6	18.8	[9]
10420	Cnoc Coig	seal	−11.8	19.5	[9]
GUsi3201/3208	Airds Bay, Loch Etive	limpet flesh	−14.1	6.3	[30]
GUsi3202/3209	Airds Bay, Loch Etive	limpet flesh	−15.0	6.7	[30]
GUsi3204/3211	Airds Bay, Loch Etive	limpet flesh	−15.1	6.3	[30]
GUsi3205/3212	Airds Bay, Loch Etive	limpet flesh	−15.0	7.1	[30]
GUsi3206/3213	Airds Bay, Loch Etive	limpet flesh	−14.0	7.0	[30]
GUsi3207/3214	Airds Bay, Loch Etive	limpet flesh	−15.2	6.7	[30]
GUsi3215/3221	Oban (SAMS)	limpet flesh	−13.6	7.8	[30]
GUsi3216/3222	Oban (SAMS)	limpet flesh	−15.5	6.9	[30]
GUsi3217/3223	Oban (SAMS)	limpet flesh	−14.7	6.2	[30]
GUsi3451/3603	Oban (SAMS)	periwinkle flesh	−13.3	8.5	[30]

All animal and human bones yielded enough collagen for reliable measurement of $\delta^{13}C$ and $\delta^{15}N$, with atomic C:N ratios in the accepted range of 2.9–3.6 [42–48].

3.2. Diet Reconstruction

The diets of the individuals recovered from Neolithic deposits in Raschoille Cave are reconstructed here from the $\delta^{13}C$ and $\delta^{15}N$ values of 28 directly-dated human bone collagen samples, but with emphasis on those individuals for which we have osteometric age-sex determinations (Table 1, RC.01–12).

Four samples analyzed by [30] (Table 1, RC.25–28) are problematic since there is no associated information on age-at-death or sex for the skeletal elements sampled. The petrous data in Series B and C are also problematic in that $\delta^{13}C$ and $\delta^{15}N$ values may vary according to which part of the petrous bone is analyzed. The inner periosteal layer is formed in utero and does not remodel after c. 2 years of age, such that bone collagen samples combining material from the inner and outer periosteal layers (which may be the case here) can "reflect a mixed dietary signal of foetal diet, breast feeding and diet consumed later in life" [49] (p. 203).

The $\delta^{13}C$ and $\delta^{15}N$ ranges of adult individuals from Raschoille Cave are plotted in Figure 3 relative to the values recorded for terrestrial and marine food sources from the region. FRUITS individual diet reconstructions for the Raschoille Neolithic population are presented graphically in Figure 4, alongside the corresponding data for Mesolithic and Neolithic populations elsewhere in western Scotland. The detailed model outputs are provided in Tables 4–6.

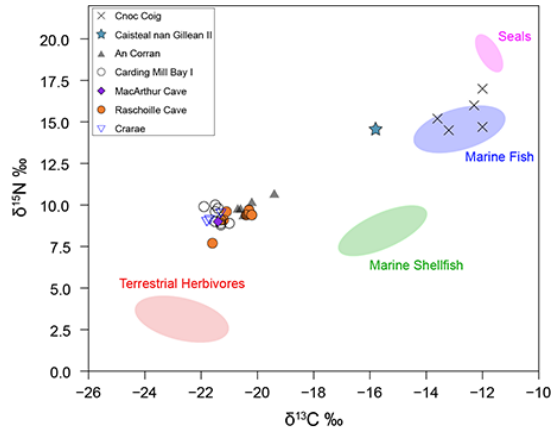

Figure 3. $\delta^{13}C$ vs. $\delta^{15}N$ in bone collagen from Late Mesolithic and Early Neolithic adults in western Scotland relative to the ranges recorded for seals, marine fish, marine shellfish and terrestrial herbivores (plotted as ellipses). Shellfish values are for modern flesh and, for comparability with the other food sources, have been adjusted for the Suess effect and the fish flesh to bone collagen offset.

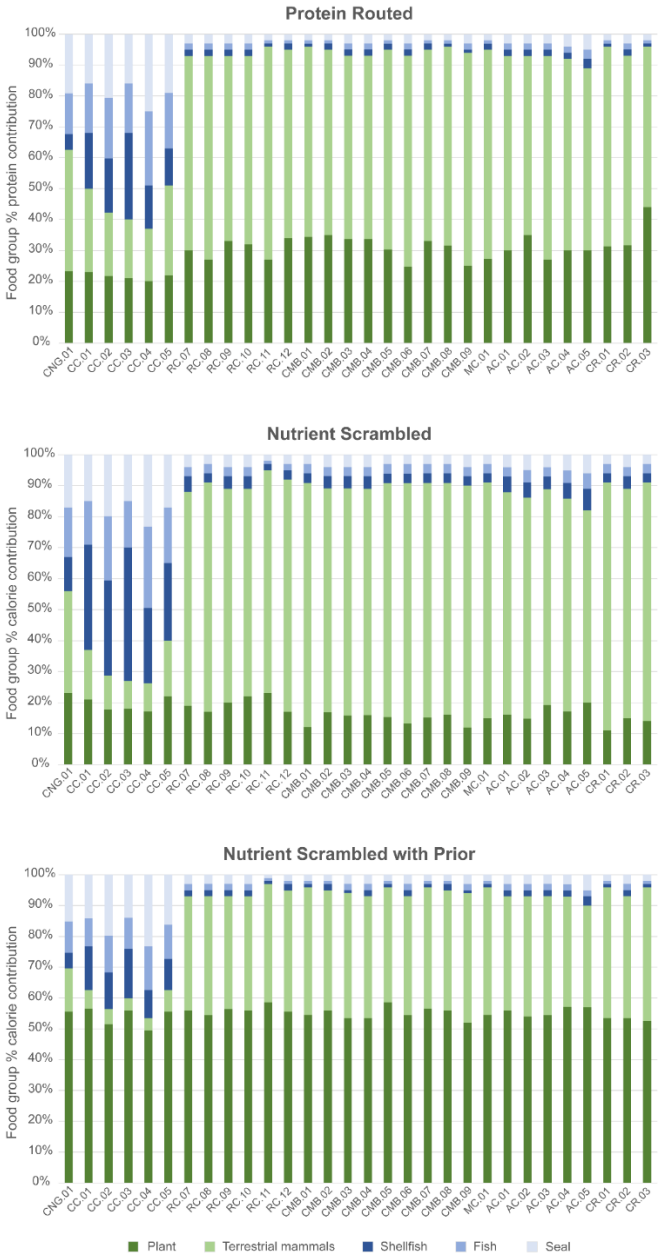

Figure 4. Comparison of FRUITS diet reconstructions for Mesolithic and Neolithic adult populations in central-west Scotland: CG—Caisteal nan Gillean II (Oronsay); CC—Cnoc Coig (Oronsay); RC—Raschoille Cave; CMB—Carding Mill Bay I; MC—MacArthur Cave (Oban); AC—An Corran (Skye); CR—Crarae (Argyll).

Table 4. Protein-routed Model (Model 1). Proportion of protein sources (% ± 1σ) in the diets of Mesolithic and Neolithic adult humans from central-west Scotland modelled in FRUITS (no priors were assumed; Mesolithic diet modelled using carbon and nitrogen isotope values from wild omnivore and herbivore species from west Scotland; Neolithic diet modelled using carbon and nitrogen isotope values from both wild and domestic terrestrial omnivore and herbivore species from across Scotland).

ID	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	Terrestrial Plants	Terrestrial Mammals	Shellfish	Fish	Seal
Caisteal nan Gillean II							
CNG.01	−15.8	14.6	23 ± 18	39 ± 13	5 ± 5	13 ± 9	19 ± 11
Cnoc Coig							
CC.01	−13.2	14.5	23 ± 18	27 ± 12	18 ± 12	16 ± 10	16 ± 11
CC.02	−12.3	16.0	21 ± 16	20 ± 10	17 ± 11	19 ± 11	20 ± 13
CC.03	−12.0	14.7	21 ± 17	19 ± 11	28 ± 14	16 ± 10	16 ± 11
CC.04	−12.0	17.0	20 ± 15	17 ± 9	14 ± 10	24 ± 15	25 ± 15
CC.05	−13.6	15.2	22 ± 17	29 ± 11	12 ± 9	18 ± 11	19 ± 12
Raschoille Cave							
RC.07	−20.3	9.7	30 ± 23	63 ± 21	2 ± 2	2 ± 2	3 ± 2
RC.08	−21.1	9.6	27 ± 23	66 ± 21	2 ± 2	2 ± 2	3 ± 3
RC.09	−20.4	9.4	33 ± 26	60 ± 24	2 ± 2	2 ± 2	3 ± 3
RC.10	−20.2	9.4	32 ± 23	61 ± 22	2 ± 2	2 ± 2	3 ± 3
RC.11	−21.6	7.7	27 ± 20	69 ± 19	1 ± 1	1 ± 1	2 ± 2
RC.12	−21.2	9.1	34 ± 25	61 ± 24	2 ± 2	1 ± 1	2 ± 2
Carding Mill Bay I							
CMB.01	−21.5	9.0	34 ± 27	61 ± 25	1 ± 1	1 ± 1	2 ± 2
CMB.02	−21.0	8.9	35 ± 28	60 ± 26	2 ± 2	1 ± 1	2 ± 2
CMB.03	−21.5	9.6	34 ± 28	60 ± 27	2 ± 2	2 ± 2	3 ± 2
CMB.04	−21.4	9.8	34 ± 27	60 ± 26	2 ± 2	2 ± 2	3 ± 2
CMB.05	−21.3	8.8	30 ± 24	64 ± 23	2 ± 1	1 ± 1	2 ± 2
CMB.06	−21.5	10.0	25 ± 20	69 ± 19	2 ± 1	2 ± 2	3 ± 3
CMB.07	−21.3	8.9	33 ± 27	62 ± 26	2 ± 2	1 ± 1	2 ± 2
CMB.08	−21.3	9.1	31 ± 25	63 ± 24	1 ± 1	1 ± 1	2 ± 2
CMB.09	−21.9	9.9	25 ± 20	69 ± 20	1 ± 1	2 ± 2	3 ± 2
MacArthur's Cave							
MC.01	−21.4	9.0	27 ± 25	67 ± 24	2 ± 2	1 ± 1	2 ± 2
An Corran							
AC.01	−20.7	9.8	30 ± 23	63 ± 22	2 ± 2	2 ± 2	3 ± 3
AC.02	−20.6	9.8	35 ± 26	58 ± 25	2 ± 2	2 ± 2	3 ± 3
AC.03	−20.5	9.4	27 ± 21	66 ± 20	2 ± 2	2 ± 2	3 ± 3
AC.04	−20.2	10.2	30 ± 24	62 ± 22	2 ± 2	2 ± 2	4 ± 3
AC.05	−19.4	10.7	30 ± 23	59 ± 21	3 ± 3	3 ± 3	5 ± 4
Crarae							
CR.01	−21.8	9.0	31 ± 24	64 ± 23	1 ± 1	1 ± 1	2 ± 2
CR.02	−21.3	9.5	32 ± 25	62 ± 24	2 ± 1	2 ± 1	3 ± 2
CR.03	−21.7	9.1	44 ± 31	52 ± 30	1 ± 1	1 ± 1	2 ± 2

Table 5. Nutrient Scrambled Model (Model 2). Proportion of foods (%cal ± 1σ) in the diets of Mesolithic and Neolithic adult humans from central-west Scotland modelled in FRUITS (no prior assumed; Mesolithic diet modelled using carbon and nitrogen isotope values from wild omnivore and herbivore species from west Scotland; Neolithic diet modelled using carbon and nitrogen isotope values from both wild and domestic terrestrial omnivore and herbivore species from across Scotland).

ID	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	Terrestrial Plants	Terrestrial Herbivores	Shellfish	Fish	Seal
Caisteal nan Gillean II							
CNG.01	−15.8	14.6	23 ± 17	33 ± 12	11 ± 9	16 ± 10	17 ± 11
Cnoc Coig							
CC.01	−13.2	14.5	21 ± 16	16 ± 9	34 ± 15	14 ± 10	15 ± 11
CC.02	−12.3	16.0	18 ± 14	11 ± 7	31 ± 13	21 ± 13	20 ± 13
CC.03	−12.0	14.7	18 ± 14	9 ± 7	43 ± 15	15 ± 10	15 ± 11
CC.04	−12.0	17.0	17 ± 13	9 ± 6	24 ± 12	26 ± 15	23 ± 15
CC.05	−13.6	15.2	22 ± 16	18 ± 10	25 ± 13	18 ± 11	17 ± 12
Raschoille Cave							
RC.07	−20.3	9.7	19 ± 18	69 ± 16	5 ± 4	3 ± 2	4 ± 3
RC.08	−21.1	9.6	17 ± 15	74 ± 14	3 ± 3	3 ± 2	3 ± 3
RC.09	−20.4	9.4	20 ± 17	69 ± 15	4 ± 4	3 ± 2	4 ± 3
RC.10	−20.2	9.4	22 ± 20	67 ± 18	4 ± 4	3 ± 2	4 ± 3
RC.11	−21.6	7.7	23 ± 23	72 ± 22	2 ± 2	1 ± 1	2 ± 2
RC.12	−21.2	9.1	17 ± 16	75 ± 14	3 ± 3	2 ± 2	3 ± 3
Carding Mill Bay I							
CMB.01	−21.5	9.0	12 ± 11	78 ± 11	3 ± 3	3 ± 2	3 ± 3
CMB.02	−21.0	8.9	17 ± 17	73 ± 16	4 ± 3	3 ± 2	4 ± 3
CMB.03	−21.5	9.6	16 ± 18	74 ± 16	4 ± 3	3 ± 3	4 ± 3
CMB.04	−21.4	9.8	16 ± 18	73 ± 16	4 ± 4	3 ± 3	4 ± 3
CMB.05	−21.3	8.8	15 ± 15	74 ± 16	3 ± 3	3 ± 2	3 ± 3
CMB.06	−21.5	10.0	13 ± 14	76 ± 13	3 ± 3	3 ± 2	3 ± 3
CMB.07	−21.3	8.9	15 ± 15	75 ± 13	3 ± 3	3 ± 2	3 ± 3
CMB.08	−21.3	9.1	16 ± 16	74 ± 14	3 ± 3	3 ± 3	3 ± 3
CMB.09	−21.9	9.9	12 ± 13	78 ± 12	3 ± 3	3 ± 3	4 ± 3
MacArthur Cave							
MC.01	−21.4	9.0	15 ± 14	76 ± 13	3 ± 3	3 ± 2	3 ± 3
An Corran							
AC.01	−20.7	9.8	16 ± 16	71 ± 15	5 ± 4	3 ± 3	4 ± 4
AC.02	−20.6	9.8	15 ± 14	72 ± 13	5 ± 4	4 ± 3	5 ± 4
AC.03	−20.5	9.4	19 ± 17	69 ± 15	4 ± 4	3 ± 3	4 ± 4
AC.04	−20.2	10.2	17 ± 17	68 ± 15	5 ± 5	4 ± 4	5 ± 4
AC.05	−19.4	10.7	20 ± 18	62 ± 15	7 ± 5	5 ± 4	6 ± 5
Crarae							
CR.01	−21.8	9.0	11 ± 10	80 ± 10	3 ± 3	3 ± 2	3 ± 3
CR.02	−21.3	9.5	15 ± 14	74 ± 13	4 ± 3	3 ± 3	4 ± 3
CR.03	−21.7	9.1	14 ± 15	77 ± 14	3 ± 3	3 ± 2	3 ± 3

Table 6. Nutrient Scrambled Model with prior (Model 3). Proportion of foods (%cal ± 1σ) in the diets of Mesolithic and Neolithic adult humans from central-west Scotland modelled in FRUITS (prior assumed, protein intake of 5 to 40% [36]; Mesolithic diet modelled using carbon and nitrogen isotope values from wild omnivore and herbivore species from west Scotland; Neolithic diet modelled using carbon and nitrogen isotope values from both wild and domestic terrestrial omnivore and herbivore species from across Scotland).

ID	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	Terrestrial plants	Terrestrial herbivores	Shellfish	Fish	Seal
Caisteal nan Gillean II							
CNG.01	−15.8	14.6	55 ± 11	14 ± 7	5 ± 4	10 ± 7	15 ± 9
Cnoc Coig							
CC.01	−13.2	14.5	56 ± 8	6 ± 5	14 ± 6	9 ± 7	14 ± 9
CC.02	−12.3	16.0	52 ± 8	5 ± 4	12 ± 5	12 ± 8	20 ± 12
CC.03	−12.0	14.7	56 ± 7	4 ± 4	16 ± 6	10 ± 7	14 ± 9
CC.04	−12.0	17.0	49 ± 9	4 ± 3	9 ± 5	14 ± 9	23 ± 13
CC.05	−13.6	15.2	55 ± 9	7 ± 5	10 ± 6	11 ± 7	16 ± 10
Raschoille Cave							
RC.07	−20.3	9.7	56 ± 14	37 ± 13	2 ± 2	2 ± 2	3 ± 3
RC.08	−21.1	9.6	55 ± 14	39 ± 13	2 ± 2	2 ± 2	3 ± 2
RC.09	−20.4	9.4	57 ± 14	37 ± 13	2 ± 2	2 ± 1	3 ± 2
RC.10	−20.2	9.4	56 ± 13	37 ± 12	2 ± 2	2 ± 2	3 ± 3
RC.11	−21.6	7.7	58 ± 16	38 ± 15	1 ± 1	1 ± 1	1 ± 1
RC.12	−21.2	9.1	55 ± 12	39 ± 12	2 ± 1	1 ± 1	2 ± 2
Carding Mill Bay I							
CMB.01	−21.5	9.0	54 ± 13	41 ± 12	1 ± 1	1 ± 1	2 ± 2
CMB.02	−21.0	8.9	56 ± 13	39 ± 13	2 ± 2	1 ± 1	2 ± 2
CMB.03	−21.5	9.6	54 ± 13	41 ± 13	1 ± 1	2 ± 1	3 ± 2
CMB.04	−21.4	9.8	54 ± 13	40 ± 12	2 ± 1	2 ± 1	3 ± 2
CMB.05	−21.3	8.8	58 ± 16	37 ± 15	1 ± 1	1 ± 1	2 ± 2
CMB.06	−21.5	10.0	55 ± 14	39 ± 13	2 ± 1	2 ± 1	3 ± 2
CMB.07	−21.3	8.9	56 ± 13	39 ± 12	1 ± 1	1 ± 1	2 ± 2
CMB.08	−21.3	9.1	56 ± 14	39 ± 13	2 ± 1	1 ± 1	2 ± 2
CMB.09	−21.9	9.9	52 ± 11	42 ± 10	1 ± 1	2 ± 1	3 ± 2
MacArthur Cave							
MC.01	−21.4	9.0	54 ± 12	41 ± 12	1 ± 1	1 ± 1	2 ± 2
An Corran							
AC.01	−20.7	9.8	56 ± 13	37 ± 12	2 ± 2	2 ± 2	3 ± 3
AC.02	−20.6	9.8	54 ± 13	39 ± 12	2 ± 2	2 ± 2	3 ± 3
AC.03	−20.5	9.4	55 ± 13	39 ± 12	2 ± 2	2 ± 2	3 ± 2
AC.04	−20.2	10.2	56 ± 13	35 ± 12	2 ± 2	2 ± 2	3 ± 3
AC.05	−19.4	10.7	57 ± 12	33 ± 11	3 ± 3	3 ± 2	5 ± 4
Crarae							
CR.01	−21.8	9.0	53 ± 13	42 ± 13	1 ± 1	1 ± 1	2 ± 2
CR.02	−21.3	9.5	54 ± 12	40 ± 12	2 ± 1	2 ± 1	3 ± 2
CR.03	−21.7	9.1	52 ± 12	43 ± 11	1 ± 1	1 ± 1	2 ± 2

3.3. Radiocarbon Dating

Figure 5 compares the original series of AMS ^{14}C dates on human bone from Raschoille Cave measured at ORAU (Table 1, series 1), which were not pretreated using

ultrafiltration, with the newer series of dates (Table 1, series 2 and 3) based on ultrafiltration. Although the same bones were not dated, the two sets of dates are essentially random samples of the Raschoille population. Ultrafiltration has resulted in younger ^{14}C ages, suggesting the OxA- dates may be too old by c. 100–150 years.

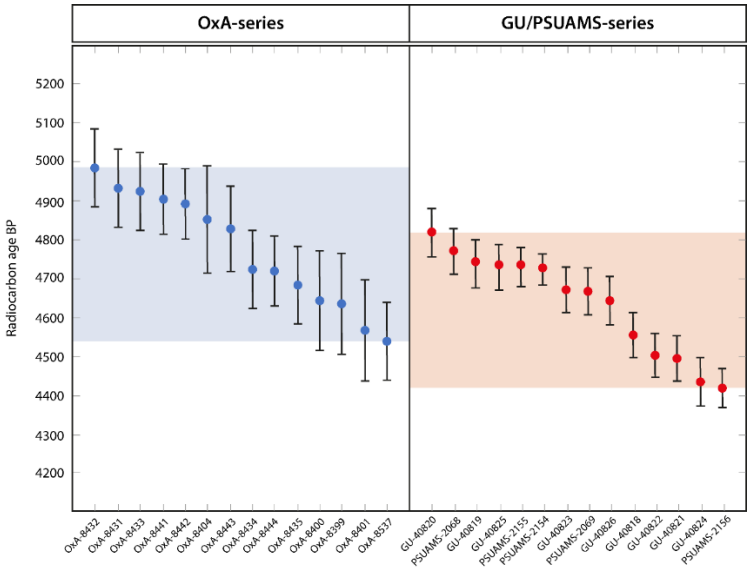

Figure 5. Comparison of non-ultrafiltered (blue) and ultrafiltered (red) AMS dates on human bones from Raschoille Cave.

The chronology reflected in the ultrafiltered series was examined further using OxCal 4.4.4 [35] and the IntCal20 radiocarbon calibration curve [50]. Figure 6 shows the results of a simple *sequence model* applied to the ultrafiltered date series (Table 1, series 2 and 3) in which the dates are assumed to represent a single group of events that occurred one after another over an extended period. The model output suggests the human remains all date to the period 3650–3090 cal BC (95% probability) and probably 3640–3210 cal BC (68% probability).

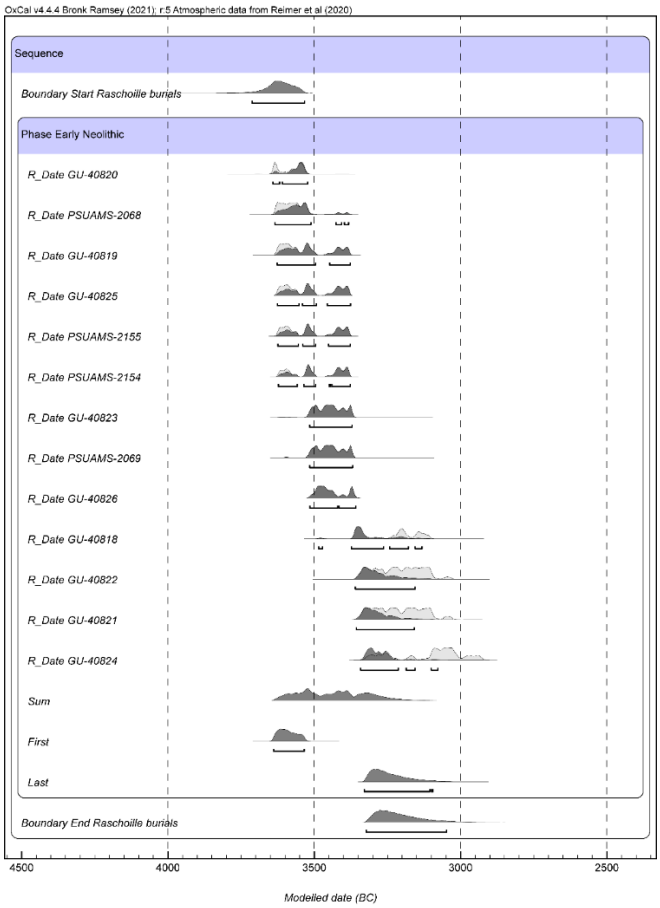

Figure 6. Bayesian statistical model estimating the start and end of Neolithic burial activity in Raschoille Cave, based on the GU and PSUAMS radiocarbon dates on human bone. The model assumes individuals were buried soon after death or excarnation, and estimates this activity began 3640–3530 cal BC (95% probability; Start Raschoille burials) and ended 3330–3100 cal BC (95% probability; End Raschoille burials). The duration of burial activity was between 220 and 510 years (95% probability) and probably 260–400 years (68% probability).

4. Discussion

4.1. Inter-Individual Variation in Stable Isotope Values

The $\delta^{13}C$ and $\delta^{15}N$ values of the Raschoille population as a whole range from −22.5‰ to −20.2‰ and from 7.6‰ to 11.9‰, respectively. Although differences in the diet are largely responsible for variation in individual stable isotope values, variation can also result from non-dietary factors including developmental stage and physiological stress related to disease or pregnancy [51–53].

The dietary timespan reflected in bone collagen stable isotope values largely depends on the age-at-death of the individual and the skeletal element sampled [54,55]. In infants and young children, bone growth and collagen turnover are very rapid, such that bone collagen isotope values in younger children generally reflect the average diet over less than a year before death [55]. During adolescence, there is rapid bone growth, collagen synthesis and remodeling; it is estimated that complete collagen turnover may occur in cortical bone (e.g., the femoral shaft) in as little as two years [55,56]. Moreover, increased protein requirements may result in reduced discrimination, or fractionation, of dietary $^{14}N/^{15}N$ [51,52]. It is widely assumed that in adults collagen turnover is much slower and stable isotope signatures reflect average dietary intake over at least ten and as much as thirty years before death [54,57,58]. However, a study of collagen turnover in cortical bone of femora shafts of present-day large mammals [59] found that the isotopic composition of cortical bone collagen reflects that of the diet *during the period of skeletal growth* with the

implication that collagen recovered from the femoral cortical bone of adults (irrespective of age-at-death) represents isotopic values during adolescence. In comparison, the turnover of collagen in cancellous bone, such as ribs and vertebrae, is relatively rapid and continuous [54,56]. For example, adult rib collagen can be completely replaced in as little as one year and up to five years, and therefore represents the shorter-term dietary intake [56].

It follows that comparing the isotopic values of different skeletal elements in separate individuals, as well as individuals of differing ages (i.e., adult vs. sub-adult), may lead to the assumption of variation in dietary intake where none exists. Detailed evaluation of the significance of the differences in $\delta^{13}C$ and $\delta^{15}N$ between individuals requires the demography of the sampled population, as well as the skeletal element analyzed, to be known. Unfortunately, the specific skeletal element sampled and associated demographic information were not documented for all the analyzed Raschoille Cave remains. Those specimens for which the information was recorded include both adults and sub-adults. The majority (12/14) of the Series A samples were taken from the cortical tissue of a limb bone. However, in one individual a cervical vertebra was sampled, while in another individual a rib was analyzed. The following discussion of individual differences in the diet focuses on those samples in Series A for which both age-at-death and the skeletal element sampled are known (Table 1).

Although there is no statistically significant difference between the adult and sub-adult stable isotope values (Mann–Whitney U-test $\delta^{15}N$, $p = 0.688921$; $\delta^{13}C$, $p = 0.378478$), the sub-adults have a wider spread of values, with $\delta^{15}N$ ranging from 7.6‰ to 11.9‰. This relatively larger range may relate in part to ^{15}N enrichment associated with nursing [60] and to the use of lower trophic level foods, such as cereals, during weaning [55].

No statistical correlation between age and $\delta^{15}N$ value (Spearman Rank Correlation Test $r_s = -0.23424$, $p = 0.61315$) was evident in the Raschoille Cave population (n.b., for the Spearman Rank analysis, the age of the sub-adults was taken as the mid-point of the range quoted in Table 1). However, sample RC.01, which has the highest $\delta^{15}N$ value of 11.9‰ is from the only individual identified as an infant, approximately 1–2 years old at death. The ^{15}N enrichment may therefore reflect a nursing signal. Fuller et al. [60] demonstrated that nursing infants had $\delta^{15}N$ values that were elevated by 2–3‰ over those of their mothers. The $\delta^{13}C$ values also exhibited a slight enrichment of c. 1‰ during breastfeeding but decreased rapidly when solid foods were introduced to the infant diet. The Raschoille Cave infant (RC.01) had a $\delta^{15}N$ value that was 2.7‰ above the mean value of the adults from the site. In contrast, the infant's $\delta^{13}C$ value of −21.8‰ was lower than the adult mean $\delta^{13}C$ value of −20.8‰. The combination of relatively elevated $\delta^{15}N$ and depleted $\delta^{13}C$ suggests the child was in the process of being weaned, with solid foods introduced to complement breastfeeding.

Bayesian dietary modelling is presented *only* for those individuals in Series A identified as adults because of the influence of developmental stage on non-dietary related variations in stable isotope values.

4.2. Diet Reconstruction Using Linear Mixing Models

Linear mixing models (LMMs) have been widely employed in archaeology to estimate past human diets from stable isotope data. The results of a simple, one proxy ($\delta^{13}C$), two food source (terrestrial vs. marine) LMM of west-central Scottish Mesolithic and Neolithic groups are presented in Table 7. Dietary endpoints used for the model were $\delta^{13}C = -20.0$‰ for 100% terrestrial diets and $\delta^{13}C = -12.0$‰ for 100% marine diet [61].

Table 7. Mean $\delta^{13}C$ and $\delta^{15}N$ of adults from Mesolithic and Neolithic sites in central-west Scotland, and percentage proportion of marine resources in diet calculated using a simple linear mixing model with $\delta^{13}C$ endpoint for 100% terrestrial diet of −20.0‰ and for 100‰ marine diet of −12.0‰. Note: uncertainty over dietary endpoints of ±1‰ equates to uncertainty in the proportion of marine resources in diet of c. ±10% [62]. Data from [2,9,10,29,30,40,63,64].

Site	∑	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	% Marine (±10%)	ID	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰	% Marine (±10%)
MESOLITHIC					**Caisteal nan Gillean II**			
Caisteal nan Gillean II	1	−15.8	14.6	52.5	CNG.01	−15.8	14.6	52.5
Cnoc Coig	5	−12.6	15.5	92.5	**Cnoc Coig**			
NEOLITHIC					CC.01	−13.2	14.5	85.0
An Corran	5	−20.3	10.0	0.0	CC.02	−12.3	16.0	96.3
Carding Mill Bay I	9	−21.4	9.3	0.0	CC.03	−12.0	14.7	100.0
Crarae	3	−21.6	9.2	0.0	CC.04	−12.0	17.0	100.0
Raschoille Cave	6	−20.8	9.2	0.0	CC.05	−13.6	15.2	80.0
MacArthur Cave	1	−21.4	9.0	0.0	**Raschoille Cave**			
					RC.07	−20.3	9.7	0.0
					RC.08	−21.1	9.6	0.0
					RC.09	−20.4	9.4	0.0
					RC.10	−20.2	9.4	0.0
					RC.11	−21.6	7.7	0.0
					RC.12	−21.2	9.1	0.0
					Carding Mill Bay I			
					CMB.01	−21.5	9.0	0.0
					CMB.02	−21.0	8.9	0.0
					CMB.03	−21.5	9.6	0.0
					CMB.04	−21.4	9.8	0.0
					CMB.05	−21.3	8.8	0.0
					CMB.06	−21.5	10.0	0.0
					CMB.07	−21.3	8.9	0.0
					CMB.08	−21.3	9.1	0.0
					CMB.09	−21.9	9.9	0.0
					MacArthur's Cave			
					MC.01	−21.4	9.0	0.0
					An Corran			
					AC.01	−20.7	9.8	0.0
					AC.02	−20.6	9.8	0.0
					AC.03	−20.5	9.4	0.0
					AC.04	−20.2	10.2	0.0
					AC.05	−19.4	10.7	0.1
					Crarae			
					CR.01	−21.8	9.0	0.0
					CR.02	−21.3	9.5	0.0
					CR.03	−21.7	9.1	0.0

LMMs have limited applicability. Using LMMs to evaluate dietary constituents from two variables (i.e., $\delta^{13}C$ and $\delta^{15}N$) can distinguish the contributions of, at most, three isotopically distinct food sources [65].

Conventionally, it has been widely accepted that in individuals consuming protein-adequate diets, dietary protein will be directly routed to bone collagen. Consequently, bone collagen $\delta^{13}C$ and $\delta^{15}N$ values reflect dietary protein sources [66,67]. Where protein intake is inadequate, there is de novo synthesis of non-essential amino acids from dietary carbohydrates and/or lipids. In such instances, the carbon isotope ratios of bone collagen reflect 'whole' diet, i.e., protein, carbohydrate and lipid sources [62,67].

The effects of nutrient scrambling on collagen isotope values may not only be problematic in individuals with low protein diets. Fernandes et al. [68] demonstrated that in mammals a proportion of collagen carbon is derived from dietary lipids and/or carbohydrates and that this is uniform, c. 26 ± 4%, irrespective of dietary intake.

Differential routing of nutrients (or nutrient scrambling) undermines the notional linear correlation of $\delta^{13}C$ and $\delta^{15}N$ assumed in LMMs and may mask the contribution of certain food sources to the diet in conventional LMMs. Milner et al. [6] observed that Neolithic groups with a low-protein diet could have derived up to 20% of their dietary protein from marine resources without any evident impact on the $\delta^{13}C$ value. Hedges [62] contrasted models of a low protein diet, in which 20–30% of the protein was derived from marine sources, with a high protein 100% terrestrial diet. The difference in the $\delta^{13}C$ values between the two hypothetical diets was just 0.3‰ [62].

4.3. Diet Reconstruction Using FRUITS

Both protein-routed and nutrient-scrambled Bayesian mixing models were generated, and the results are discussed below. Bayesian mixing models (BMMs) potentially offer higher-resolution reconstructions of the diet compared to simple LMMs with two fixed endpoints. The Bayesian mixing model Food Reconstruction Using Isotopic Transferred Signals (FRUITS) can accurately evaluate the contribution of multiple food sources and allows users to incorporate uncertainty in trophic level offsets and food source isotope values into dietary reconstructions [36,69].

The diets of the Raschoille Cave individuals were reconstructed using FRUITS (version 3.0 Beta). Since the accuracy of dietary reconstructions may be affected by the developmental stage of the individual (as discussed above), only individuals thought to be adults or adolescents (age-at-death >10 years) were included in the analysis (Table 1, RC.07–12). Children plus individuals for whom *reliable* demographic information is lacking (Table 1, RC.1–6, 13–28) were excluded.

The existence of offsets between the diet and consumer collagen stable isotope ratios is well-established [20,70]. The $\delta^{13}C_{diet\text{-}collagen}$ offset is widely acknowledged to be c. +5‰ [16,36]. However, the size of the $\delta^{15}N_{diet\text{-}collagen}$ offset has been the subject of some debate [22–24]. Here we use the diet to collagen offsets set out in [69]—$\delta^{13}C_{diet\text{-}collagen}$= +4.8 ± 0.5‰ and $\delta^{15}N_{diet\text{-}collagen}$= +5.5 ± 0.5‰.

There are also uncertainties in the stable isotope values of consumers (largely reflecting instrument measurement error); these were set at 0.5‰ for both $\delta^{13}C$ and $\delta^{15}N$ [36,69].

Uncertainties in food-source stable isotope values (reflecting differences in preparation methods and seasonal and physiological variations in animal metabolism) should also be incorporated into dietary reconstructions. Following [36,69] this uncertainty was set at a minimum of 1‰ for all food sources.

A further consideration for dietary reconstructions that rely on the isotope values of archaeological animal bones as a proxy for food web values is the offset between bone collagen and tissue (e.g., muscle, fat) consumed. These offsets are:

- terrestrial herbivores $\Delta^{13}C_{protein\text{-}collagen}$ = −2.0‰, and $\Delta^{15}N_{protein\text{-}collagen}$ = 0‰, $\Delta^{13}C_{lipid\text{-}protein}$ = −8.0‰ [71];
- fish $\Delta^{13}C_{protein\text{-}collagen}$ = −1.0‰, and $\Delta^{15}N_{protein\text{-}collagen}$ = +1.5‰, $\Delta^{13}C_{lipid\text{-}protein}$ = −7.0‰ [71].

The offset for seals was taken from Pickard and Bonsall [72].

4.4. Food Sources

There are five categories of food sources that may have contributed to the diets of the Early Neolithic individuals from Raschoille Cave: (i) plant foods, including domesticated cereals and wild plants; (ii) meat or dairy from terrestrial mammals; (iii) shellfish; (iv) fish; and (v) sea mammals.

The $\delta^{13}C$ and $\delta^{15}N$ values for plant foods were drawn from Fernandes et al. [69] and based on data originally published by Bogaard et al. [73]. The $\delta^{13}C$ and $\delta^{15}N$ values of carbonized plant remains have been established to be appropriate proxies for the raw plant stable isotope ratios [74,75]. Non-manured plants had mean energy $\delta^{13}C$ of −23.5‰, protein $\delta^{13}C$ of −26.0 ± 1.0‰ and $\delta^{15}N$ of 2.0 ± 1.0‰, while manured plants had an elevated

mean protein $\delta^{15}N$ of 4.5 ± 1.0‰ [69]. Evidence for manuring in Neolithic Scotland is limited. High levels of soil phosphate in a paleo land surface at North Mains, Strathallan, preserved beneath a Bronze Age mound, indicated the presence of manure. However, it is not certain that this relates to the manuring of arable land; it may reflect animal grazing or penning [76]. Consequently, non-manured plant values were used for the models evaluated below.

A review of published stable isotope data for terrestrial fauna from Scottish Mesolithic and Neolithic sites reveals significant regional variability in the $\delta^{13}C$ and $\delta^{15}N$ of both wild game and domestic livestock. Therefore, for this study, we have attempted to reconstruct the *local* food web by using baseline indicators from the Oban area. Isotopic signatures for terrestrial mammals were determined from archaeological samples from two sites, Raschoille Cave and Carding Mill Bay I (Table 2). Not all samples have been directly dated, but they are all assumed to derive from the time range c. 6500–3000 cal BC, corresponding to the later part of the Mesolithic and the Early Neolithic. We detected no significant temporal shifts in mammalian baseline values over this period nor any indications of herbivores grazing on seaweed or salt marsh plants (cf. Britton et al.; Jones et al.) [77,78].

Mean bone collagen $\delta^{13}C$ and $\delta^{15}N$ values of all terrestrial herbivores and omnivores (n = 28) are −22.7 ± 0.6‰ and 3.1 ± 0.5‰, respectively. There is a small but significant difference in the mean $\delta^{13}C$ and $\delta^{15}N$ values for red deer (n = 6, −22.2‰, 2.6‰) and domestic cattle (n = 8, −23.1‰, +3.5‰) (Mann–Whitney U test $\delta^{13}C$ p = 0.011821, $\delta^{15}N$ p= 0.002415). Incorporating the bone collagen to protein tissue offset, the terrestrial animal protein values used for our FRUITS models for Early Neolithic groups were $\delta^{13}C$ = −24.7 ± 1.0‰ and $\delta^{15}N$ = 3.1 ± 1.0‰. For the Mesolithic groups, wild terrestrial animal protein values were used, $\delta^{13}C$ =—22.2 ± 1.0‰ and $\delta^{15}N$ = 2.6 ± 1.0‰.

The baseline isotope values of marine resources (represented by fish and sea mammal bones) were drawn primarily from archaeological specimens from central-west Scotland (see Table 3). Mean carbon and nitrogen stable isotope bone collagen values of archaeological fish were $\delta^{13}C$ = −12.8 ± 0.9‰ and $\delta^{15}N$ = 14.6 ± 0.8‰ (n = 5), while the mean values for seals were $\delta^{13}C$ = −11.8 ± 0.2‰ and $\delta^{15}N$ = 19.1 ± 0.4‰ (n = 3). Incorporating the bone collagen to protein tissue offset and food source uncertainties, the fish protein values were $\delta^{13}C$ = −13.8 ± 1.0‰ and $\delta^{15}N$ = 16.1 ± 1.0‰ and seal protein values were $\delta^{13}C$ = −14.1 ± 1.0‰ and $\delta^{15}N$ = 18.5 ± 1.0‰.

The archaeological evidence for shellfish consumption by Neolithic communities in the Oban Bay area is equivocal. Abundant shellfish remains (mainly periwinkles and bivalves) were recovered from the Neolithic deposits in Raschoille Cave and appear to have been closely associated with concentrations of human bones [13]. Possible explanations for the co-occurrence of shells with human bones are that the shells are: (1) remains of food offerings to the dead; or (2) food refuse from funeral feasts or external domestic activities (or even pre-existing midden deposits) that were brought into the cave and heaped over human remains as part of the burial ritual. At Carding Mill Bay I, the offset between the ^{14}C ages of human and animal bones from the same contexts [39] (Tables 7 and 8) suggests the human remains were either inserted into pre-existing midden deposits or that 'ancient' midden deposits were transported to the site from elsewhere and heaped over human remains in symbolic acts of burial.

Since shellfish flesh is not recovered from archaeological sites, the isotope signatures of modern shellfish may be used in diet reconstructions [63]. However, using modern samples as proxies for prehistoric resources introduces further uncertainties into dietary reconstructions and therefore modern comparanda should be used cautiously. Carbon isotope ratios of modern specimens do not directly mirror the values of prehistoric resources. The use of fossil fuels in the industrial era has resulted in ^{13}C-depleted atmospheric CO_2—known as the 'Suess effect' [79–81]. Modern atmospheric CO_2 carbon isotope values are lower than pre-industrial values [82]. Therefore, a correction factor needs to be applied to the $\delta^{13}C$ values of modern samples when used as proxies for prehistoric food webs. Keeling et al. [82] described a c. 2‰ difference between modern (i.e., AD 2014) and

pre-industrial $\delta^{13}C$ values of atmospheric CO_2 and noted that this depletion may be larger in terrestrial plants and by inference in terrestrial food webs. The Suess effect has had a smaller impact on oceanic $\delta^{13}C$; in the North Atlantic, ^{13}C-depletion of up to 0.8‰ is evident [83].

The effects of lipid removal pre-treatments on the $\delta^{15}N$ values of modern samples are also problematic. Lipid $\delta^{13}C$ is ^{13}C-depleted in comparison to bone collagen values—by up to 7‰—and it has been recommended that lipids should be removed from samples prior to isotope ratio measurements [84]. However, the solvents used to remove lipids have been demonstrated to alter $\delta^{15}N$ values [85,86]. Sotiropoulos et al. [87] recommended a two-fold approach—measurement of two separate portions of a single sample, $\delta^{13}C$ from a lipid-extracted portion of a sample and $\delta^{15}N$ from an untreated portion of the sample.

Additionally, regional-scale spatial variation in intra-species $\delta^{13}C$ and $\delta^{15}N$ values of marine organisms are evident in British coastal waters, largely reflecting the extent of mixing of waters, as well as differences in phytoplankton communities, microalgae and cyanobacteria [88]. Where possible, the modern comparanda should be sourced locally to the archaeological sites under investigation. The shellfish flesh isotope values used for this study were selected from data published in [30]. The measurements from two genera of shellfish, limpets and periwinkles (the most frequently recovered species from archaeological shell middens and the most common species in the intertidal zone along the west coast of Scotland) were utilized. Only those specimens with C:N ratios in the range 2.9–3.6 after lipid extraction were used as proxies for prehistoric shellfish. Additionally, the $\delta^{13}C$ of lipid-extracted samples were combined with the $\delta^{15}N$ values from the untreated portion of the sample. Mean carbon and nitrogen stable isotope values of shellfish flesh were $\delta^{13}C = -14.6 \pm 0.7$‰ and $\delta^{15}N = 7.4 \pm 1.6$‰ ($n = 11$).

4.5. FRUITS Model Predictions

Three models were evaluated using FRUITS. The different models were designed to test the impact of varying parameters to reconstruct the diet and to assess how this might impact our understanding of the role of marine resources in diet:

- MODEL 1: protein-routed model, which assumes dietary protein was directly routed to bone collagen;
- MODELS 2 and 3: 'nutrient-scrambled' (fraction weighted, concentration-dependent) models, which assume collagen carbon was derived from dietary protein and energy sources, in the proportions 74 ± 4% and 26 ± 4% %, respectively [68]. Model 3 assumed a prior restriction on dietary protein intake of between 5 and 40% [36].

The results of the diet reconstructions using FRUITS are presented in Tables 4–6.

For each of the three models, FRUITS' reconstructions indicate that terrestrial food sources (i.e., plant foods and meat and/or dairy from terrestrial mammals) dominated dietary intake at Raschoille Cave.

The nutrient-scrambled model with the prior protein restriction (Model 3) suggests that similar proportions of calories were derived from terrestrial plant and animal sources. However, because food source data (and local plant food isotope values, in particular) are limited, there is uncertainty in the precise contribution these resources made to individual diets.

Both the protein-routed and the nutrient-scrambled models suggest that marine resources were a minor component of the diet with shellfish comprising 0.0–5.9% (95% CI) of protein intake (Model 1) and whole diet calories (Model 3). In the same way as shellfish, fish likely made a very small or no contribution to the diet (0.0–5.9%, 95% CI). Seals *may* have provided more protein and calories to the diets of the people interred at Raschoille Cave than fish and shellfish, but this contribution would still have been small (protein (Model 1) 0.0–8.9%, 95% CI; and calories (Model 3) 0.0–8.9%, 95% CI).

4.6. Neolithic Diets in Central-West Scotland

Bownes et al. [39] determined, using Bayesian modelling, that the protein sources of four individuals from Carding Mill Bay contained "modest amounts (15 ± 11 to 21 ± 14%)" of marine foods. However, Bownes et al. [39] did not factor wild plant foods or cereals into their FRUITS calculations but assumed that 100% of dietary protein was derived from either terrestrial meats or the flesh of aquatic animals. Bownes [30] (p. 150) acknowledged that plant foods would have been exploited by both Mesolithic and Neolithic groups in Scotland but excluded them from dietary models on the basis that "plants contain very little … protein in relation to animal meat".

This assumption is problematic for two main reasons:

1. The protein content of animal meats varies depending on the species and the cut of meat. Some cereals and wild plants have similar protein content to that of certain animal products (see Table 8). Hazelnuts and barley, which are among the most frequently identified plant foods recovered from Scottish Neolithic sites [89], have relatively high protein yields;
2. Deriving dietary protein exclusively from animal products is atypical [23]. Modern mean protein intake in developed countries is 57% animal protein and 43% plant protein, while in developing countries the relative proportions of animal and plant protein are 30% and 70%, respectively. Animal protein as a proportion of total protein intake may be higher among groups that consume large quantities of dairy products [23].

Table 8. Protein content (in grams) per 100g of wild plants, cereals and animal products. The plants listed have all been identified at Scottish Neolithic sites [89]. All food protein data were drawn from the *USDA Food Composition Database* [90].

Food	Protein Content (g) per 100 g
Plant products	
Barley, hulled	12.48
Flax seed	16.67
Hazelnut	14.95
Oat	12.50
Rye	6.98
Wheat—durum	13.68
Wheat—spelt	14.57
Animal meats	
Beef mince (grass-fed cattle)	18.75
Beef steak (grass-fed cattle)	23.01
Lamb shoulder steak	16.48
Lamb loin	20.88
Pork carcass	13.91
Pork loin	20.71
Venison (grass-fed deer)	22.32
Dairy products	
Sheep's milk	5.98
Sheep's milk cheese	25.00
Whole cow's milk	3.33
Cow's milk cheese, hard	25.00
Cow's milk, cream cheese	6.15

While the use of wild plant foods in Neolithic western Scotland is well documented [89,91], cereal cultivation has been suggested to be of minor importance based on a perceived lack of evidence for large-scale vegetation clearance before the Bronze Age [92]. This argument must be weighed against the sharp increase in cereal (*Hordeum*)-type pollen after c. 3800 cal BC recorded in pollen diagrams across the Oban region [12] (Figure 4) and archaeobotanical and other evidence for cereal cultivation in the Hebrides from c. 3700 cal BC [93–95].

The Raschoille individuals have similar stable isotope signatures, and by inference likely had similar diets, to other west Scottish Early Neolithic populations. Both FRUITS (each model) and LMM reconstructions indicate that terrestrial resources dominated diets at all Neolithic sites (Figure 4; Tables 4–7). At Crarae, there may have been a greater reliance on terrestrial animal food sources than among Early Neolithic populations elsewhere. However, what is perhaps surprising in the FRUITS reconstructions is that at all the sites considered in this study, including both Mesolithic and Neolithic, plant foods made a significant contribution to dietary calories.

The LMM estimations of the proportion of marine resources in Neolithic diet do not exceed uncertainty (i.e., >10% [62]; see Table 7). Our FRUITS models confirm that marine resources were likely a minor component in the diets of Early Neolithic groups in western Scotland. The proportion of calories derived from marine food sources modelled slightly higher in some individuals from An Corran than in the individuals from the other Neolithic sites included in this study. As with plant foods, reliance on marine resources may have varied regionally in Neolithic Scotland [63]. However, these differences are *very* small.

5. Conclusions

The reconstruction of diet from bone collagen carbon and nitrogen stable isotope ratios indicates that the Neolithic individuals interred in Raschoille Cave consumed predominantly terrestrial resources comprising C_3 plants, meat and/or dairy products. Comparisons with other groups suggest diets were relatively homogeneous across Early Neolithic western Scotland. Both BMM and LMM reconstructions of diets indicate that terrestrial resources dominated. Mean proportions of plants are relatively high. This contrasts with the paucity of evidence for cereal cultivation in western Scotland and hints at the continuing importance of wild plant foods in diet.

Our findings are consistent with the Neolithic 'neglect of the sea' hypothesis. Based on the available evidence, and the limitations of the Mesolithic dataset notwithstanding, a shift from a largely marine to a predominantly terrestrial diet at the Mesolithic–Neolithic transition is supported.

One of the most startling findings of the FRUITS modelling of prehistoric diets in coastal western Scotland is the high proportion of terrestrial resources in the diets of the Late Mesolithic groups on Oronsay. This contribution to the diet is largely undetectable using traditional LMMs.

We recognize that our research has limitations. Our model predictions are likely biased by inadequate baseline $\delta^{13}C$ and $\delta^{15}N$ data for wild terrestrial plants and cereals, as well as freshwater and estuarine resources. Moreover, it is not certain that the individuals interred at Raschoille Cave were permanent coastal dwellers, nor that they were responsible for the accumulation of the midden deposits at the site. Evaluation of sulfur-stable isotope ratios may resolve this issue. Research into incremental dentine carbon, nitrogen and sulfur-stable isotope ratios, which facilitates the reconstruction of the short-term diet during the development of the tooth analyzed and has the potential to indicate mobility, would enhance our understanding of marine resource exploitation, consumption behavior (i.e., sporadic vs. continued use) and settlement patterns in the Neolithic.

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