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## RADIOCARBON



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## MARINE RADIOCARBON RESERVOIR EFFECT IN LATE PLEISTOCENE AND EARLY HOLOCENE COASTAL WATERS OFF NORTHERN IBERIA

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### ABSTRACT

Radiocarbon dating of closely associated marine mollusk shells and terrestrial material (mammal bones or charred wood) collected from archaeological contexts in northern Atlantic Iberian coastal areas is used to quantify the marine <sup>14</sup>C reservoir effect ( $\Delta R$ ) for the coastal waters off the Cantabrian coast (northern Iberia). For the first time,  $\Delta R$  values were reliably determined for these coastal waters and, also for the first time, a  $\Delta R$  was calculated for the Late Pleistocene in Atlantic Iberia. Pairs of coeval samples of different carbon reservoirs selected from Upper Paleolithic (Late Pleistocene) and Mesolithic (Early Holocene) contexts yielded  $\Delta R$  weighted mean values of  $-117 \pm 70$  <sup>14</sup>C yr and  $-105 \pm 21$  <sup>14</sup>C yr, respectively. These values show oceanographic conditions characterized by a reduced offset between atmospheric and surface water <sup>14</sup>C contents, suggesting a nonexistent or a very weak upwelling and some stratification of the water column. Similar oceanographic conditions have been recorded in other areas of Atlantic Iberia during the Holocene, such as off Andalusian and northwestern Galician coasts. Results not only provide useful information on environmental conditions but also a framework to obtain more precise and reliable absolute chronologies for the Late Pleistocene and Early Holocene in northern Iberia.

**Keywords:** Marine <sup>14</sup>C reservoir effect; Cantabrian coast; Late Pleistocene; Early

Holocene

## INTRODUCTION

Radiocarbon ( $^{14}\text{C}$ ) dates on marine samples have not been used as extensively as terrestrial biosphere (charcoal or bone) dates to set up absolute chronologies because interpretation is complicated by oceanographic factors. Nevertheless, marine shellfish were widely used by prehistoric populations in various locations, namely in coastal areas, and their shells are abundant and usually well preserved in archaeological deposits. To set up chronologies for a particular coastal area using marine samples, background research concerning the oceanographic conditions and the marine reservoir effect for the area in question is needed in order to obtain accurate and reliable results.

The residence time of carbon in the deep ocean is about 1000 yr (Sigman and Boyle 2000). Thus, a fraction of the  $^{14}\text{C}$  atoms have time to decay (half-life=5730 yr) while the deep water is out of contact with the atmosphere. The deep ocean is therefore depleted in  $^{14}\text{C}$  relative to the atmosphere, and the surface seawater (mixed layer) also has a  $^{14}\text{C}$  specific activity lower than that of the atmosphere but greater than that of the deep ocean. Marine waters are therefore deficient in  $^{14}\text{C}$  compared with the terrestrial biosphere and, consequently, marine organisms have an apparent age, the so-called reservoir age. This reservoir age,  $R(t)$ , is defined as the difference between conventional  $^{14}\text{C}$  dates from a pair of coeval samples that lived in different carbon reservoirs (Stuiver et al. 1986).  $R(t)$  can vary with time, since the atmospheric  $^{14}\text{C}$  content is not constant and also varies from region to region of the ocean since the oceanographic conditions present in each region are different due to the variability in water mass mixtures, wind regime, bathymetry, and upwelling of deep water.

Considering these issues, Stuiver et al. (1986) modeled the response of the world ocean to atmospheric  $^{14}\text{C}$  variations. From this modeling, a calibration curve for marine samples related to the sea surface water (mixed layer) has been derived. In order to take into account the difference in  $^{14}\text{C}$  content between the surface water of a specific region and the average surface water, a parameter, denoted as  $\Delta R$  (regional marine  $^{14}\text{C}$  reservoir effect) and defined as the difference between the reservoir age of the mixed layer of the regional ocean and the reservoir age of the mixed layer of the average world ocean at time  $t$ , is required (Stuiver et al. 1986). Usually,  $\Delta R$  values are determined for a particular geographical region by  $^{14}\text{C}$  dating of marine mollusk shells of historic (known) age, collected alive before 1950, i.e. of "pre-bomb" age or, perhaps more often,

by  $^{14}\text{C}$  dating of paired samples of the same age but of different origin (terrestrial and marine). The  $^{14}\text{C}$  age of the terrestrial biosphere sample is then converted into a marine model age using the calibration curve Marine13, the most recent curve (Reimer et al. 2013); this marine model age is then deducted from the  $^{14}\text{C}$  age of the associated marine sample to yield  $\Delta R$  (Stuiver and Braziunas 1993). Although reservoir ages are time-dependent,  $\Delta R$  is not according with the model mentioned above, unless some change of oceanographic conditions has occurred that is restricted to the regional ocean under consideration. This happens in regions affected by an active upwelling of deep water. Rates of regional upwelling can vary in the course of time and as the intensity of  $^{14}\text{C}$  depletion in the mixed layer depends upon the upwelling activity, it is likely that  $\Delta R$  values can also vary in the course of time (Stuiver and Braziunas 1993, Kennett et al. 1997, Ingram 1998, Ascough et al. 2005). For instance, for the western Portuguese coast, a  $\Delta R$  mean value of  $250 \pm 25$   $^{14}\text{C}$  yr was determined for modern times (AD 1880-1940), while for the period 3000 - 600 BP  $\Delta R$  takes a mean value of  $95 \pm 15$   $^{14}\text{C}$  yr (Soares and Dias 2006). On the other hand,  $R(t)$ , taking into consideration its definition mentioned above, always takes a positive value, but  $\Delta R$  can be either positive or negative (see, for instance, Stuiver and Braziunas 1993: Fig. 16, Stuiver et al. 2016). Large positive  $\Delta R$  values are usually associated with a strong upwelling, while negative values can be associated with a non-existent upwelling and some stratification of the water column.

Previous research concerning the variability of  $\Delta R$  in coastal waters off Atlantic Iberia allowed the quantification of this parameter (Figures 1 and 2), which is of crucial importance for the correct calibration of  $^{14}\text{C}$  ages of marine samples.  $\Delta R$  values were thus determined for the western Portuguese coast, which weighted mean values are mentioned above, also for western and northwestern Galician coasts ( $\Delta R = -8 \pm 12$   $^{14}\text{C}$  yr and  $\Delta R = -192 \pm 24$   $^{14}\text{C}$  yr, respectively; see Soares and Dias 2007), and southern Atlantic Iberian coast (Barlavento  $\Delta R = 69 \pm 17$   $^{14}\text{C}$  yr, Sotavento  $\Delta R = -26 \pm 14$   $^{14}\text{C}$  yr, and Andalusian coast  $\Delta R = -108 \pm 31$   $^{14}\text{C}$  yr; see Martins and Soares 2013). So, concerning the Atlantic coasts of the Iberian Peninsula, only the quantification of  $\Delta R$  for the northern Atlantic Iberian coast (Cantabrian Sea) was missing (see Figure 1).



Figure 1- $\Delta R$  weighted mean values for the Atlantic coasts of the Iberian Peninsula.

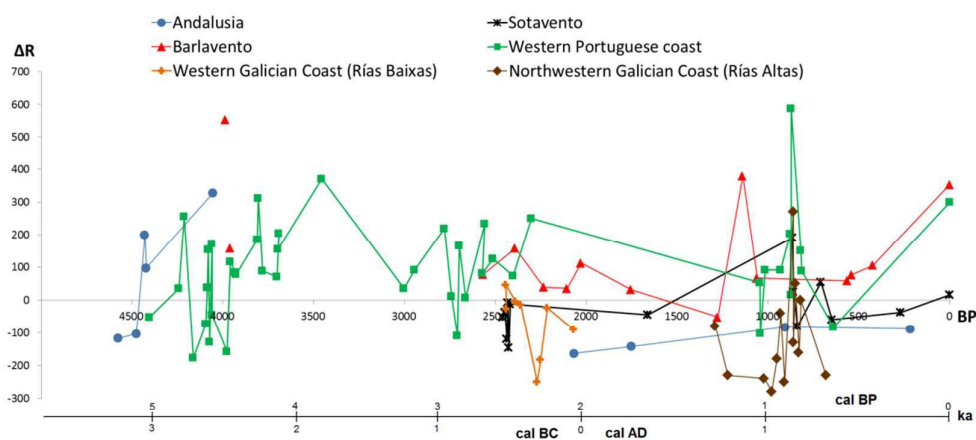


Figure 2 -The variability of marine  $^{14}\text{C}$  reservoir effect off Atlantic coasts of Iberia

Northern Iberia is a key place for the study of long term changes in hunter-fisher-gatherer societies. Numerous Upper Paleolithic and Mesolithic sites have been recorded in the region, providing one of the richest archaeological records in the world for the study of the Pleistocene-Holocene transition. Recently, three different (but related) research projects led by two of us (IGZ and MGM) have addressed questions regarding environmental conditions and human occupations in Late Pleistocene and Early Holocene coastal locations from northern Iberia (see, for example, García-Escárgaza et al. 2015b; Gutiérrez-Zugasti, 2011; Gutiérrez-Zugasti et al. 2014, 2015). Building absolute chronologies using archaeological samples is crucial to make reliable interpretations of past human behavior and environmental changes through time. Given the nature of the investigated sites (huge Mesolithic shell middens and more discrete Upper Paleolithic shell lenses) and the sometimes limited amount of terrestrial datable materials (especially in Mesolithic middens), dating of shells is fundamental to establish accurate chronologies in the region. However, as stated above, shell-based dating is biased by the regional marine  $^{14}\text{C}$  reservoir effect ( $\Delta\text{R}$ ), which must be known in order to calibrate the conventional marine dates. In addition, information on oceanographic conditions derived from the study of the  $\Delta\text{R}$  can be related to data obtained from other environmental proxies (e.g. stable isotopes and elemental analysis). In this paper, we present radiocarbon dates on paired coeval samples of different carbon reservoirs (terrestrial and marine) in order to quantify the  $\Delta\text{R}$  for the Cantabrian Sea during the Late Pleistocene and the Early Holocene.  $\Delta\text{R}$  results will provide an accurate tool for calibration of marine radiocarbon dates and they will also be used for discussion on oceanographic conditions in the region and comparison with other areas of the Atlantic Iberian coast.

## **OCEANOGRAPHIC CONDITIONS OFF WESTERN AND NORTHERN ATLANTIC IBERIAN COASTS**

The western coast of the Iberian Peninsula (37-43°N) has a North-South orientation and is the northern boundary of the NW Africa coastal upwelling system, while the northern Atlantic Iberian coast, with a W-E orientation, develops along ~43°N and is not affected by that upwelling system. At these latitudes, shelf winds follow a seasonal pattern connected with the large-scale climatology of the Northeastern Atlantic Ocean. The

dominant wind pattern on these coasts is a consequence of the location of the Azores high, which causes changes in wind direction and intensity. These changes, consequently, modify the hydrographic structure of the water column. Hence, the atmospheric circulation associated with the Azores high corresponds to westerly winds off the Atlantic Iberian coast in the winter (the Azores high occupies its southern position) and to considerably stronger northerly and northwesterly winds in the summer, since the Azores high moved to its most northern position. These northerly summer winds induce Ekman transport offshore along the western coast, i.e. they are clearly upwelling favorable from June to September. Besides, the western Iberian shelf forms a complex oceanographic system due to its location, size, coastline, and bathymetric features, where a variety of micro-, meso-, and macro-scale physical processes occur, including coastal upwelling and coastal downwelling (Wooster et al. 1976). Northerly winds cause upwelling; downwelling occurs from October to March, when the coast is under the influence of southerly winds due to the reinforcement of the Iceland low, while the Azores high occupies its most southerly position (Fiúza 1982, 1983; Fiúza et al. 1982; Ferreira 1984; Nogueira et al. 2003; Lorenzo et al. 2005; Varela et al. 2005).

Galicia is located at the northern limit of the upwelling area of the NE Atlantic. Cape Finisterre is the most northwesterly point in the Galician region and at this distinctive topographic feature, the western coastline abruptly changes its near South-North orientation to a Southwest-Northeast direction as far as Cape Ortegal and to a West-East direction beyond this Cape (Cantabrian coastline along  $\sim 43^\circ\text{N}$ ).

The wind field in the Cape Finisterre area has high spatial and temporal variability throughout the year. Nevertheless, an outstanding feature of the Galician coast is the persistent upwelling near Cape Finisterre (Torres et al. 2003). This recurrent upwelling center off Cape Finisterre divides the Galician coast into two regions, which differ not only in the occurrence and intensity of winds, but also in the origin of the upwelled water. Eastern North Atlantic Central Water of subpolar origin (ENACW<sub>sp</sub>) has been recorded as upwelled water north of Cape Finisterre when northeastern winds predominate during summer. South of the Cape, ENACW of subtropical origin (ENACW<sub>st</sub>) prevails during the summer upwelling (Castro et al. 2000). Besides the different origin of the upwelled waters, also a lesser intensity of upwelling occurs in the northwestern Galician shelf, and the existence of a thermohaline front near the coastline



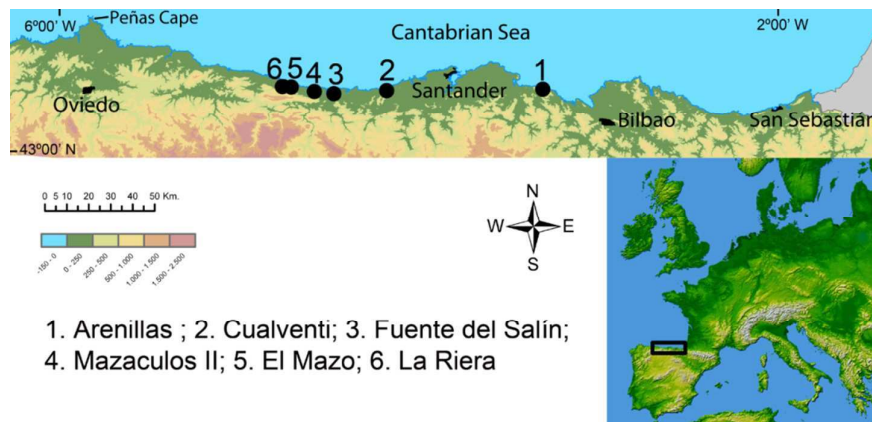
impedes to a great extent the penetration of upwelled water into the Rías Altas (Prego and Bao 1997; Prego et al. 1999).

The Cantabrian Sea is the southernmost part of the Bay of Biscay, in the Eastern North Atlantic. The Cantabrian shelf lies in a West-East orientation and presents narrow and abrupt features, with a mean width of 30-40 km and crossed by a number of canyons and irregularities. In winter there is a prevalence of westerly winds, while easterlies prevail during summer due to the location of the Azores high. Since the Cantabrian coastline has a West-East orientation, easterly winds trigger the occurrence of coastal upwelling. The upwelling season extends from June to August and the upwelling system is characterized by short-lived events concentrated around the favorable season (Botas et al. 1990; Gil et al. 2002; Gil 2003; Llope et al. 2006). This upwelling system differs from that on the western coast, where a nearly constant upwelling is usually present throughout the year (Alvarez et al. 2011). An important topographic feature located in the central Cantabrian coast is the Cape Peñas, which reinforces wind-driven upwellings to the west of the Cape. The sea surface temperature follows the expected seasonal warming and cooling pattern, which determines processes of stratification and mixing of the water column. During summer the stratification of the water column occurs to a depth of 50 m, which can be broken by upwelling events, while during winter the water column remains mixed (Lavin et al. 1998). Comparing the upwelling phenomenon on the western and northern coasts of Iberia, it can be concluded that upwelling events are more frequent and intense along the western coast than along the northern one. On the other hand, the most favorable upwelling conditions correspond to the spring-summer period on the western coast and only to summer in the Cantabrian Sea, when the stratification of the water column is well established (Alvarez et al. 2011).

## **MATERIAL AND METHODS**

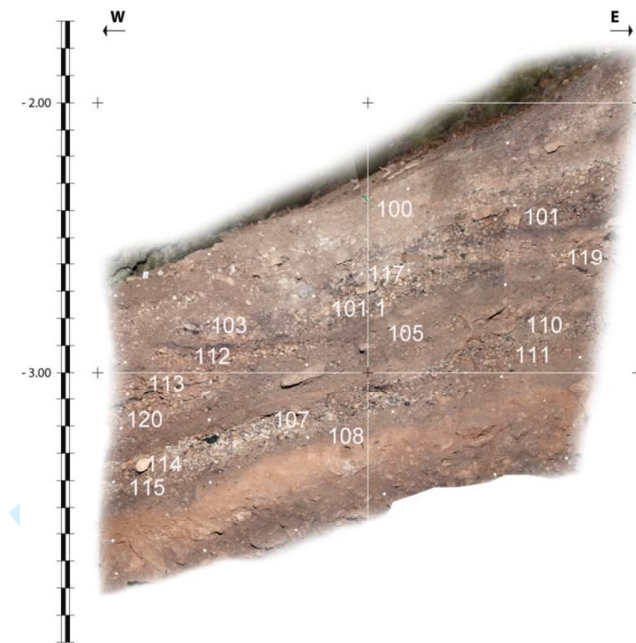
Pairs of closely associated archaeological samples (marine shells and bones/charred wood) from several depositional contexts were collected for radiocarbon dating in order to quantify the marine  $^{14}\text{C}$  reservoir effect of coastal waters in the Cantabrian Sea (northern Iberia) (Table 1). Twenty one samples were selected from the material recovered at six archaeological sites (see Figure 3 for a location of the sites), following two different strategies: 1) paired samples from five sites previously excavated by other

scholars were collected from museums and universities; 2) paired samples were collected from our own excavations at the Mesolithic shell midden site of El Mazo (Figure 4) (Gutiérrez-Zugasti and González-Morales 2014; Gutiérrez-Zugasti et al. 2014).



**Figure 3** - Locations of sampled archaeological sites on the Cantabrian coast.

In the first case, samples from the Mesolithic levels of Arenillas (Bohigas and Muñoz, 2002), the Lower Magdalenian levels of Cualventi (Lasheras et al. 2005-2006) and the Gravettian site of Fuente del Salín (González-Morales and Moure-Romanillo, 2008) were collected from the Museum of Prehistory and Archaeology of Cantabria (Santander, Spain), whilst samples from the Solutrean levels of La Riera (Straus and Clark, 1986) were collected from the Museum of Archaeology of Asturias (Oviedo, Spain). Samples from the Mesolithic site of Mazaculos II (González-Morales and Marquez-Uría, 1978; González-Morales et al. 1980) were collected from the storage of the Institute of Prehistory (IIIPC) at the University of Cantabria (Santander, Spain). Each selected pair comes from the same excavation unit, from the same square and, when possible, from the same subsquare and spit (see Table 1). In the second case, terrestrial and marine samples that were found together or separated apart by just a few centimeters were selected for radiocarbon dating, i.e. samples where very accurate data on the provenance of the samples and on their stratigraphic relationships is known.



**Figure 4** - Stratigraphic profile at the Mesolithic site of El Mazo. Numbers indicate the location of different stratigraphic units. Paired samples for radiocarbon dating were taken from units 101, 113, 120, 105 and 108.

Bone, shell and charred wood samples from the six sites were subjected to species identification prior to dating. In the case of bones, the anatomical part was also identified. Bone samples belong to roe deer *Capreolus capreolus*, red deer *Cervus elaphus* and ibex *Capra pyrenaica*. Sometimes species identification was not possible and bones were simply identified as ungulates. Shell samples collected from museums/universities belonged to the limpet *Patella vulgata*, whilst shell samples recovered at El Mazo were topshells *Phorcus lineatus* (Table 1). Both marine species were certainly collected alive by humans to be used as food. The charred wood samples used in the study were identified as *Corylus avellana*, the common hazel, a shrub that can live for a few decades. The characteristics of the sites, the excavation techniques used and the analysis of sample taphonomy confirm the integrity of the samples included in our study (García-Escárzaga et al. 2015a; Gutiérrez-Zugasti, 2009; Gutiérrez-Zugasti et al. 2013).

All samples were radiocarbon dated by AMS. Ten shells, seven bones and two charred wood samples were run at the Oxford Radiocarbon Accelerator Unit (ORAU) following routine pretreatments and standard dating procedures that included ultrafiltration and

measurement of stable isotopic composition (by IRMS) and carbon and nitrogen content (C:N ratio determination) for bones (see Brock et al. 2010 and references therein for a description of procedures). Two additional bones were dated at the Center for Applied Isotopes Studies (CAIS) of the University of Georgia (USA) following standard procedures described in Cherkinsky et al. (2010) and Vogel et al (1984).  $^{14}\text{C}$  ages were calculated in accordance with the definitions recommended by Stuiver and Polach (1977).

$\Delta R$  values were calculated by converting the terrestrial biosphere sample  $^{14}\text{C}$  age into a marine model age (see Table 2), which was subtracted from the  $^{14}\text{C}$  age of the associated marine shell sample to yield  $\Delta R$  (Stuiver and Braziunas 1993; Reimer et al. 2002). We also followed the recommendations of Ascough et al. (2005, 2007, 2009) and Russell et al. (2011) for calculating the  $\Delta R$  values.

The original focus of the research projects involved in this study was to set up absolute chronologies for Upper Paleolithic and Mesolithic contexts, which led to a reduced number (only one case –Mazaculos II, see Table 1) of multiple paired samples from each archaeological context. This handicap made the fulfillment of the published methodology difficult to follow. Nevertheless, for the calculation of the  $\Delta R$  values we employed the following recommendations: *i*) unrounded  $^{14}\text{C}$  ages; *ii*) interpolation between calibration curves IntCal13 and Marine13 (Reimer et al. 2013) for converting the terrestrial biosphere sample  $^{14}\text{C}$  age into a marine model age; *iii*) chi-squared test ( $\chi^2_{:0.05}=T$ ) as a statistical criterion in the definition of contemporary samples from each group (terrestrial or marine) collected in the same archaeological context (when more than 1 sample was available); and *iv*) establishment of the set of  $\Delta R$  values that can integrate the weighted mean calculation according to the chi-squared test ( $\chi^2_{:0.05}=T$ ) results. The  $1\sigma$  error for the  $\Delta R$  determination is obtained by propagation of the errors on the marine age and the modeled marine age from each pair of samples.

The reservoir age  $R(t)$  was also determined following Stuiver et al. (1986).

Table 1. <sup>14</sup>C age of samples from archaeological contexts of sites located on the Cantabrian coast.

Site	Chronoculture	Unit (A#* - B/#** - #***)	Sample description	δ <sup>13</sup> C (‰)	14C Date (BP)	Lab nr
<i>Arenillas</i>	Mesolithic	Shell midden	<i>Capreolus capreolus</i> (distal humerus)	-21.36	7143±36	OxA-X-2488-43
			<i>Patella vulgata</i>	0.00	7374±38	OxA-27154
<i>Mazaculos II</i>	Mesolithic	1.3 (H11)	Ungulate (phalanx)	-21.40	7700±30 <sup>a, b</sup>	UGAM-9081
			<i>Capreolus capreolus</i> (unfused proximal tibia)	-21.15	7755±38 <sup>a, b</sup>	OxA-26953
			<i>Patella vulgata</i>	+0.31	8133±39	OxA-27155
<i>El Mazo</i>	Mesolithic	101 (X15 - C - 2)	<i>Capreolus capreolus</i> (metatarsus)	-22.14	7105±40	OxA-30780
			<i>Phorcus lineatus</i>	+1.62	7310±40	OxA-30806
		113 (X16 - C)	<i>Corylus avellana</i> (charred wood)	-24.52	7212±35	OxA-28403
			<i>Phorcus lineatus</i>	+1.00	7565±34	OxA-28404
		120 (X16 - C)	Ungulate (carpal)	-20.92	7412±36	OxA-28405
			<i>Phorcus lineatus</i>	+1.43	7625±45	OxA-30976
		105 (X15 - D - 2)	<i>Corylus avellana</i> (charred wood)	-25.61	7380±55	OxA-30535
			<i>Phorcus lineatus</i>	+1.23	7595±40	OxA-30977
108 (X16 - C)	<i>Capreolus capreolus</i> (rib)	-20.85	8022±39	OxA-28411		
	<i>Phorcus lineatus</i>	+0.51	8222±36	OxA-27904		
<i>Cualventi</i>	Lower Magdalenian	E (H6 - 1 - 9)	<i>Cervus elaphus</i> (unfused distal metapod)	-19.77	15635±75	OxA-26954
			<i>Patella vulgata</i>	+0.75	15990±65	OxA-27115
<i>La Riera</i>	Solutrean	4 (E9)	<i>Capra pyrenaica</i> (mandible right M2)	-19.66	19710±120	OxA-X-2491-15
			<i>Patella vulgata</i>	+0.94	19990±90	OxA-27116
<i>Fuente del Salín</i>	Gravettian	2 (K6 - 3)	Ungulate (diaphysis)	-20.30	23040±50	UGAM-9077
			<i>Patella vulgata</i>	+0.78	23160±90	OxA-27195

\* Square \*\* Subsquare \*\*\* Spit

<sup>a</sup> 1.291; ( $\chi^2_{.05}=3.84$ )<sup>b</sup> <sup>14</sup>C date weighted mean value 7721±24 BP

## RESULTS AND DISCUSSION

R(t) and  $\Delta R$  values of the terrestrial/marine pairs collected from the archaeological contexts mentioned above are listed in Table 2. The context age, also presented in Table 2, was determined by using the samples of terrestrial origin. These ages are presented as conventional  $^{14}\text{C}$  dates and also as calendar dates using the IntCal13 calibration curve (Reimer et al. 2013) and the program CALIB (Stuiver and Reimer 1993).

It must be noted that all the dated samples are short-lived samples with the exception, perhaps, of the two charred wood samples of *Corylus avellana*. Nevertheless, it is unlikely that radiocarbon dates from this species are affected by the "old wood" effect, since usually this shrub does not live for long. The "old wood" effect problem can lead to a reduction of the offset between marine and terrestrial  $^{14}\text{C}$  ages of the pair, but this does not happen with the pairs in question from El Mazo. From Mazaculos II, two bone samples from the same context were dated. The results of these two samples are not statistically different (1.291;  $\chi^2_{:0.05}=3.84$ ), see Table 1), which should make the  $\Delta R$  obtained for Mazaculos II very reliable.

Since, as is known, the climatic conditions and, consequently, the oceanographic conditions and the carbon cycle were different during glacial times compared with the Holocene, two sets of  $\Delta R$  values were considered in our case: one for the Upper Paleolithic period (Late Pleistocene) and another for the Mesolithic (Early Holocene). These two sets of  $\Delta R$  values and respective weighted mean values are presented in Table 2.

The  $\Delta R$  obtained for Mesolithic sites (dated to the Early Holocene) show a homogeneous pattern, except for Mazaculos II, which is significantly different from those recorded at Arenillas and El Mazo (24.80;  $\chi^2_{:0.05}=12.51$ ). Values from Mazaculos II should be considered as reliable since two terrestrial samples were used in the calculations. Two hypotheses can be proposed to explain the difference: 1) the shell sample used for radiocarbon dating at Mazaculos II comes actually from an older level; 2) oceanographic conditions suffered changes during the Early Holocene in northern Iberia. Excavations at Mazaculos II were carried out using modern techniques (González-Morales and Marquez-Uría, 1978; González-Morales et al. 1980), differentiating when possible independent depositional events that have occurred.

**Table 2.** Values for the reservoir effect for coastal waters off Cantabria (Northern Iberian Peninsula).

Site	Unit	<sup>14</sup> C Terrestrial Date (BP)	Calibrated date 2 $\sigma$ (cal BC)	Calibrated date 2 $\sigma$ (cal BP)	<sup>14</sup> C Marine Date (BP)	Model Marine Date (BP)	R(t) ( <sup>14</sup> C yr)	$\Delta$ R ( <sup>14</sup> C yr)	$\chi^2$ test results
<b>MESOLITHIC (EARLY HOLOCENE)</b>									
<i>Arenillas</i>	Shell midden	7143±36	6070-5930	7880-8020	7374±38	7521±30	231±52	-147±48	(n=7) 24.80 ; ( $\chi^2_{:0.05}=12.51$ ) (Mazaculos II $\Delta$ R rejected)
<i>El Mazo</i>	101	7105±40	6050-5900	7850-8000	7310±40	7464±52	205±57	-154±66	
	113	7212±35	6210-6010	7960-8160	7565±34	7565±25	353±48	0±42	
	120	7412±36	6380-6220	8170-8330	7625±45	7748±17	213±58	-123±48	
	105	7380±55	6390-6100	8050-8340	7595±40	7747±40	215±68	-152±57	
	108	8022±39	7070-6780	8730-9020	8222±36	8363±60	200±52	-141±70	
<i>Mazaculos II</i>	1.3	7721±24	6600-6480	8430-8550	8133±39	8030±38 or 8093±41	433±49 or 378±54	103±54 <sup>a,b</sup> or 40±57 <sup>a,b</sup>	(n=6) 8.65 ; ( $\chi^2_{:0.05}=11.07$ ) <b><math>\Delta</math>R weighted mean value</b> <b>-105 ± 21<sup>14</sup>C yr</b>
<b>UPPER PALEOLITHIC (LATE PLEISTOCENE)</b>									
<i>Cualventi</i>	E	15635±75	17100-16770	18720-19050	15990±65	16040±75	355±99	-50±99	(n=3) 1.22 ; ( $\chi^2_{:0.05}=5.99$ ) <b><math>\Delta</math>R weighted mean value</b> <b>-117 ± 70<sup>14</sup>C yr</b>
<i>La Riera</i>	4	19710±120	22110-21460	23410-24060	19990±90	20115±120	280±150	-125±150	
<i>Fuente del Salín</i>	2	23040±50	25580-25230	27180-27530	23160±90	23395±100	120±103	-235±135	

<sup>a</sup> 0.624; ( $\chi^2_{:0.05}=3.84$ )<sup>b</sup>  $\Delta$ R weighted mean value 74±39 <sup>14</sup>C yr (Rejected in the calculation of  $\Delta$ R weighted mean value for the Mesolithic)



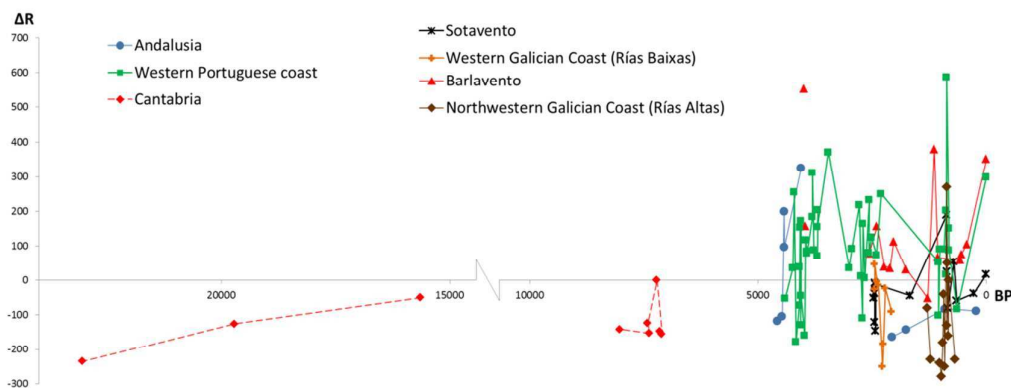
In fact, level 1 was divided into three different sublevels due to variations in the characteristics of the shell deposits identified during excavation. Therefore, shell samples should have been recovered successfully according to their separate stratigraphic provenance. Nevertheless, it is possible that the shell sample used for radiocarbon dating in our study was recovered at the contact between level 1.3 and level 2, where shells from both layers could have been mixed together, in which case the dated shell would be older than other shells recorded in layer 1.3. Unfortunately it is not possible to corroborate this hypothesis since no coordinates were recorded on individual shells. Although the most part of the Holocene was characterized by climatic stability, some important environmental changes have also been recorded (Bond et al. 1997, 2001; von Grafenstein et al. 1998; Rasmussen et al. 2007). Among them, the 8.2 ka cold event stands out in many North Atlantic Early Holocene climate records as the most prominent climate anomaly, being responsible for changes in oceanographic conditions and, consequently, in the  $\Delta R$  value (a high value is expected for  $\Delta R$  due to the huge amount of  $^{14}\text{C}$ -depleted fresh water from ice sheets - "many thousands of years of stored precipitation", following Teller et al. (2002) - that was injected in the North Atlantic at high latitudes). But calibrated radiocarbon dates from Mazaculos II (see Table 2) place the occupations around 8.5–8.4 ka, at least 200 years earlier than the Bond event 5, as recorded in the Greenland Ice Cores (NGRIP members, 2004). However, a comparison between oxygen stable isotope ratios on shells from Mazaculos II and Arenillas (including the shells from both sites used for radiocarbon dating in this study) suggests that winters were slightly colder and summers significantly colder in Mazaculos II than in Arenillas (unpublished data), at least in terms of sea surface temperatures, opening up the possibility that a significant change in the climatic conditions and thereafter in the oceanographic conditions in northern Iberia occurred at 8.5–8.4 ka. In fact, a minor cold climatic feature has been identified in the Greenland Ice Cores around 8.5 ka (Rasmussen et al. 2007), which might be related to colder conditions as reflected in the stable isotope ratios of the Mazaculos II shells. In any case, further research is required to provide a precise explanation for the intriguing results from Mazaculos II.

$\Delta R$  values from Upper Palaeolithic sites (dated to the Late Pleistocene - 19, 23 and 27 ka cal BP) are very similar, statistically not differentiable from those determined for the Mesolithic (Early Holocene - 8-9 cal BP). This finding was unexpected since during glacial times, at these high latitudes ( $\sim 43^\circ\text{N}$ ), an increase of  $R(t)$  and, consequently, of



$\Delta R$  should be expected due to subpolar front movements (southward shift of the polar front) and variations in the mode and rate of North Atlantic Deep Water production, increasing the influence of old polar waters relative to young (tropical) surface waters (Bard 1988, Bard et al. 1994, Stern and Lisiecki 2013). Consequently, further research using more pairs of samples from Upper Paleolithic contexts is needed, in order that the temporal (and spatial) variability of the marine  $^{14}\text{C}$  reservoir effect in this region can be more reliably measured and changes in oceanographic conditions precisely recorded.

Data from our study can be compared to results from other Atlantic coastal areas of Iberia (Figure 5). Given that no other data on the  $\Delta R$  exists for the Late Pleistocene in Iberia and the results put the above mentioned questions, only information corresponding to the Holocene will be discussed next. The Holocene  $\Delta R$  weighted mean value determined for the Cantabrian coast ( $-105 \pm 21$   $^{14}\text{C}$  yr) can be compared with those determined for the northwestern Galician coast and Andalusian Atlantic coast, respectively  $-192 \pm 24$   $^{14}\text{C}$  yr, between 1300 and 900 BP (Soares and Dias 2007), and  $-108 \pm 31$   $^{14}\text{C}$  yr, between 4600 and 200 BP (Martins and Soares 2013). As already mentioned, a weak upwelling occurs in the northwestern Galician shelf, and the existence of a thermohaline front near the coastline does not allow the penetration of upwelled water into the Rías Altas, from where the dated marine shell samples were certainly collected. Besides, the strong stratification of the water column is a common summer phenomenon in the Rías, which leads to a greater equilibrium with the atmosphere and, consequently, to an enrichment in  $^{14}\text{C}$  of the surface waters, which explains the difference in  $\Delta R$  value compared to the other regions. The  $\Delta R$  value obtained for the coastal waters off Cantabrian coast is not statistically different from the one obtained for the Andalusian Atlantic coastal waters, where there is no upwelling and also some summer stratification of the water column occurs, similar to the situation that prevails in the coastal waters of the Cantabrian Sea.



**Figure 5** - The variability of marine  $^{14}\text{C}$  reservoir effect off Cantabrian coast compared with the variability for the other Atlantic Iberian coastal regions.

## CONCLUSIONS

A record of past reservoir ages is preserved in the  $^{14}\text{C}$  content of contemporaneous marine and terrestrial material. Using an archaeological dataset of radiocarbon dates, the marine  $^{14}\text{C}$  reservoir effect ( $\Delta\text{R}$ ) was calculated for coastal waters off northern Iberia (Cantabrian Sea). Results provide a  $\Delta\text{R}$  mean value of  $-117 \pm 70$   $^{14}\text{C}$  yr for the Late Pleistocene and  $-105 \pm 21$   $^{14}\text{C}$  yr for the Early Holocene. These values reflect the oceanographic conditions present in the area at that time, pointing to a reduced offset between atmospheric and surface water  $^{14}\text{C}$  contents, and therefore suggesting a nonexistent or a very weak upwelling and some stratification of the water column. These conditions are similar to other coastal areas of Atlantic Iberia during the Late Holocene, such as the Andalusian and the Galician northwestern coasts. For the first time, a  $\Delta\text{R}$  mean value was calculated for the Late Pleistocene in Iberia, suggesting oceanographic conditions not very different to those of the Holocene. However, further research is needed to accurately establish the temporal and spatial variability of the  $\Delta\text{R}$  in glacial times. Using the  $\Delta\text{R}$  values established in this study and the most recently published marine calibration curve (Marine13), accurate and reliable calendar dates should be obtainable from shell samples collected in archaeological contexts on the Cantabrian coast. Therefore, data on the marine  $^{14}\text{C}$  reservoir effect not only provide environmental information, but also a framework to obtain more accurate and reliable chronologies for the Late Pleistocene and Early Holocene in northern Iberia.

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