# MARINE RADIOCARBON RESERVOIR CORRECTIONS FOR THE MEDITERRANEAN AND AEGEAN SEAS

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**ABSTRACT.** Radiocarbon measurements of nine known age shells from the Mediterranean and the Aegean Seas combined with previous measurements provide an updated value for  $\Delta R$ , the local variation in the reservoir correction for marine samples. Comparison of pre-1950s samples from the Algerian coast, with one collected in 1954, indicates early incorporation of nuclear weapons testing <sup>14</sup>C into the shallow surface waters of the Mediterranean. Comparisons between different basins indicate the surface waters of the Mediterranean are relatively homogenous. The recommended  $\Delta R$  for calibration of the Mediterranean marine samples with the 1998 marine calibration dataset is  $58 \pm 85$  <sup>14</sup>C yr, but variations in the reservoir age beyond 6000 cal BP should be considered.

# INTRODUCTION

Calibration is essential for interpretation of radiocarbon ages, especially when comparing to historical records or to other data with a different chronological basis. <sup>14</sup>C ages of marine samples also require a correction for the reservoir age of the ocean where they were formed. Because the ocean is a large carbon reservoir, the residence time of <sup>14</sup>C is long compared to the atmosphere. Together with upwelling of older carbon from the deep ocean, this results in an apparent age of marine samples several hundred years older than contemporaneous atmospheric samples (Mangerud 1972). While the pre-industrial global mean reservoir correction (R) is about 400 years, local variations ( $\Delta$ R) can be several hundred years or more (Stuiver et al. 1986).

Until recently, there was only one  $\Delta R$  value from a known age shell for the entire Mediterranean despite the archaeological significance of the region (Broecker and Olson 1959, 1961). This shell collected in 1954 near Algiers yielded a  $\Delta R$  value of  $-135 \pm 85$  <sup>14</sup>C yr (Stuiver et al. 1986). The negative  $\Delta R$  value would imply either a smaller reservoir age for the Mediterranean than the global mean or that the mollusc incorporated some <sup>14</sup>C from nuclear weapons testing. Although most marine records do not show incorporation of nuclear weapons carbon this early, the mixing time varies considerably among localities.

Recent work by Siani et al. (2000) has provided a more robust estimate of  $\Delta R$  for the Mediterranean. Their analysis, based on measurements of 26 modern, pre-nuclear testing mollusc shells and a few previously reported apparent ages, was mainly concentrated in the western Mediterranean, the Tyrrhenian Sea, the Black Sea, and the Adriatic Sea. Here we report  $\Delta R$  measurements on seven known age, pre-nuclear weapons testing mollusc shells from the eastern Mediterranean and the Aegean Sea and two from the Algerian coast in the western Mediterranean. We then incorporate our dataset with previously published data for the region and with three measurements from the Tyrrhenian and the Adriatic Seas supplied by M Taviani and A Correggiari (personal communication) in order to investigate possible regional differences in the  $\Delta R$  for the Mediterranean and adjacent seas.

# METHODS

Nine known age shells from the Mediterranean and the Aegean Sea were selected for dating from the collections of the Museum National D'histoire Naturelle, the Museum fur Naturkunde der Humboldt-Universitat zu Berlin; the Hebrew University, and Zoologische Stattssammlung Muenchen

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(Figure 1). Although records were not always adequate to determine that the molluscs had been collected live, all shells were unweathered and all of the bivalves were either articulated or had both shells collected together, which would indicate collection at or shortly after death. Only the outer edges of the shells were used for dating so that shell deposited nearest to the time of death was sampled. <sup>14</sup>C analyses were performed at the Center for AMS dating at Lawrence Livermore National Labs. <sup>14</sup>C ages presented here (Table 1) are corrected for isotope fractionation using the measured  $\delta^{13}$ C values and normalized to –25 ‰ PDB (Stuiver and Polach 1977).



Figure 1 Map showing the approximate collection location for samples in this study

Data were compiled from our measurements and <sup>14</sup>C ages from the literature (Table 2). Several reported measurements were omitted from the compilation because the authors suspected an older source of carbon from freshwater carbonates (Siani et al. 2000). The reservoir age R was calculated from the difference between the conventional <sup>14</sup>C age and the atmospheric age interpolated to the nearest year from the 1998 calibration dataset (Stuiver et al. 1998a).  $\Delta R$  values were calculated from the difference in the conventional <sup>14</sup>C age and the 1998 decadal marine calibration dataset (Stuiver et al. 1998b), which was interpolated to the year of shell growth.

## **RESULTS AND DISCUSSION**

The <sup>14</sup>C ages and  $\Delta R$  values measured for this study are given in Table 1 and in the on-line marine reservoir correction database at http://www.calib.org (Reimer and Reimer 2001). The two shells from Algeria had a relatively large spread in <sup>14</sup>C ages but were statistically the same at the 95% confidence level using a chi-squared test (Ward and Wilson 1978). Together with three measurements reported by Siani et al. (2000), a mean  $\Delta R$  value of  $83 \pm 33$  yr was calculated for the Algerian coast. This value is statistically different at the 95% level from the earlier  $\Delta R$  value of  $-135 \pm 85^{-14}$ C yr ( $-116 \pm 80^{-14}$ C yr as recalculated with the 1998 marine dataset) from a shell (species unknown) collected in AD 1954 (Broecker and Olson 1959). If the mollusc were an intertidal species it could have incorporated atmospheric <sup>14</sup>C due to wave action (Hogg et al. 1998). However, an unreasonably large amount of atmospheric carbon would be needed to offset  $\Delta R$  to this degree unless the shell incorporated <sup>14</sup>C produced in nuclear weapons testing. Mixing of atmospheric <sup>14</sup>C into shallow water may have occurred more rapidly than was observed in surface water samples from the central basin of the western Mediterranean (Broecker and Gerard 1969). A similar rise in 14C is seen in Red Sea corals from near Hurghada, which exhibit a  $\Delta^{14}$ C increase of 12‰ (~90 <sup>14</sup>C yr) between 1953 and 1955 (Cember 1989). This may be compared to the  $\Delta^{14}$ C of Arctica islandica shells from the North Sea collected at 37 m, which did not increase until after 1955 (Weidman 1995). Because of the uncertainty surrounding the uptake of bomb <sup>14</sup>C into the shallow waters of the Mediterranean, the Broecker et al. measurement was not included in the regional mean  $\Delta R$ .

Location	Mollusc species	Collector, Museum nr	Lab ID	Collection year	<sup>14</sup> C BP	δ <sup>13</sup> C (‰)	$\Delta R$ (yr)	Reservoir age (BP)
Eastern Mediterranean								
Zante, Greece	Chlamys varia	Friedrich #19971367 <sup>a</sup>	CAMS-69547	1942	$620 \pm 40$	0.3	$153 \pm 41$	439
Beirut, Lebanon	Pinctada radiata	?b	CAMS-69545	1929	$490 \pm 40$	2.4	$32 \pm 40$	344
Beirut, Lebanon	Chlamys varia	?2	CAMS-69546	1929	$400 \pm 50$	1.9	$-58\pm50$	254
Netamiya, Israel	Oslimus turbinatus	G Haas <sup>c</sup>	CAMS-69540	1937	$510\pm40$	1.4	$47 \pm 41$	342
<i>Western Mediterranean</i> Castiglione, Algeria Castiglione, Algeria	Tellina planata Ruditapes decussatus	Dieuzede <sup>2</sup> Dieuzede <sup>2</sup>	CAMS-69543 CAMS-69544	1931 1931	$\begin{array}{c} 620 \pm 40 \\ 510 \pm 40 \end{array}$	1.8 1.6	$\begin{array}{c} 161 {\pm}~40 \\ 51 {\pm}~40 \end{array}$	468 358
Aegean Sea Smyrna (Izmir), Turkey	Mytilus edulis	T Loebbecked	CAMS 60530	1893/94	$750 \pm 40$	13	$288 \pm 40$	652
Nauplia, Greece	Diodora italica	Friedrich #200028281	CAMS-69541	1940	$500 \pm 40$	1.3	$35 \pm 40$	324
Piraeus, Greece	Patella caerula	Friedrich #20001478 <sup>1</sup>	CAMS-69542	1943	$610 \pm 40$	-0.8	$143 \pm 41$	427

Table 1 <sup>14</sup>C ages,  $\delta^{13}$ C, and  $\Delta R$  values of known age shells from the Mediterranean and Aegean Seas from this study

<sup>a</sup>Zoologische Stattssammlung Muenchen (E Schwabe, Curator)

<sup>b</sup>Museum National D'histoire Naturelle (P Bouchet, Curator)

<sup>c</sup>Hebrew University, Jerusalem (J Heller, Professor)

<sup>d</sup>Museum fur Naturkunde der Humboldt-Universitat zu Berlin (M Glaubrecht, Curator)

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Table 2a Compilation of <sup>14</sup>C ages and  $\Delta R$  values for known age shells from the western Mediterranean, the Adriatic, and the Tyrrhenian Seas. References as numbered: 1) Siani et al. 2000; 2) Delibrias 1989, as quoted in reference 1; 3) Pelc 1995 as quoted in reference 1; 4) this study; 5) Taviani and Correggiari (personal communication; 6) Langone et al. 1996. A few samples from reference 1 were not included because of the possible influence of estuarine carbonates (Siani et al. 2000).

		Collection	<sup>14</sup> C age	$\Delta R$		
Location	Lab ID	year	(BP)	(yr)	Reference	
Western Mediterranean						
Alger, Algeria	GifA 96710	1881	$620 \pm 35$	$148 \pm 35$	1	
Cherchel, Algeria	Gif 4067	1905	$460 \pm 35$	$7 \pm 35$	2	
Mahdia, Algeria	Ly 6948	1948	$500 \pm 50$	$29 \pm 51$	3	
Castiglione, Algeria	CAMS-69543	1931	$620 \pm 40$	$161 \pm 40$	4	
Castiglione, Algeria	CAMS-69544	1931	$510 \pm 40$	$51 \pm 40$	4	
Antibes, France	GifA 96726	1873	$450 \pm 40$	$-27 \pm 40$	1	
St Raphael, France	GifA 96724	1892	$455 \pm 35$	$-8 \pm 35$	1	
La Seyne, France	GifA 96699	1892	$470 \pm 40$	$7 \pm 40$	1	
Marseille, France	GifA 96711	1873	$510 \pm 35$	$33 \pm 35$	1	
Marseille, France	GifA 96709	1874	$550 \pm 40$	$74 \pm 40$	1	
Banyuls, Spain	GifA 96716	1906	$570 \pm 35$	$118\pm35$	1	
Toulon, France	Gif 4068	1837	$405 \pm 35$	$-88 \pm 35$	2	
Banyuls, Spain	Ly 6900	1900	$565 \pm 55$	$110 \pm 55$	3	
Malaga, Spain	GifA 96715	1929	$430 \pm 35$	$-28\pm35$	1	
Tyrrhenian Sea						
Central Tyrrhenian Sea	CAMS-16300	1920	$430\pm60$	$-23 \pm 60$	5	
Central Tyrrhenian Sea	CAMS-16301	1920	$480 \pm 60$	$28 \pm 60$	5	
Bastia, Corsica	GifA 96704	1921	$495 \pm 40$	$42 \pm 40$	1	
Naples, Italy	GifA 96717	1873	$535 \pm 40$	$58 \pm 40$	1	
Naples, Italy	GifA 96725	1892	$610 \pm 110$	$147 \pm 110$	1	
Sicily	Ly 6863	1900	$525\pm50$	$70 \pm 50$	3	
Adriatic Sea						
Barletta, Italy	CAMS-16299	1906	$570 \pm 60$	$118 \pm 60$	5	
Rimini, Italy	CAMS-12144	1911	$587 \pm 28$	$137 \pm 28$	5,6	
	CAMS-12901					
	CAMS-13120					
	CAMS-13121					
Dalmatia	GifA 96707	1873	$380\pm35$	$-97 \pm 35$	1	
Rovigne, Croatia	GifA 96718	1926	$390 \pm 50$	$-66 \pm 50$	1	
Adriatic Sea	GifA 96722	1867	$540\pm30$	$60 \pm 30$	1	

Regional mean values of  $\Delta R$  were also calculated for the Eastern Mediterranean, Western Mediterranean, Adriatic, Tyrrhenian, and Aegean Seas from the new and compiled measurements (Table 3) and are available on-line at www.calib.org. There is little difference in the mean  $\Delta R$  values for any of the regions with the possible exception of the Aegean Sea. The Aegean  $\Delta R$  is statistically different from that of the rest of the Mediterranean at the 95% confidence level. However, all but one of the Aegean samples fit closely to the other Mediterranean data from the same approximate collection dates (Figure 2). The one higher  $\Delta R$  value at AD 1893 is from a mussel shell (*Mytilus edulis*), a species that often inhabits estuarine environments. The slightly high  $\Delta R$  for this shell could be due to carbonates derived from limestone depleted in <sup>14</sup>C. If this sample is excluded then the Aegean Sea,  $\Delta R$  is similar to the mean Mediterranean value as well as the mean of 75 ± 65 reported for the Black Sea (Siani et al. 2000). This result confirms the need for consideration of species in <sup>14</sup>C dating marine shells as has been previously noted (Hogg et al. 1998). The empirical standard deviation, that is the square root of

the variance or "scatter" in the data, is fairly large except for the Tyrrhenian Sea. This is probably the result of the differing habitats and feeding patterns of the various species as well as local variations in the <sup>14</sup>C content of the seawater. For purposes of <sup>14</sup>C calibration, the empirical standard deviation, which is a measurement of the dispersion of the data from the mean, provides a better estimate of the true uncertainty in  $\Delta R$  than the uncertainty in the mean. The mean  $\Delta R$  for the Mediterranean of 58 ± 85 <sup>14</sup>C yr (empirical s.d.) is comparable to the 35 ± 70 <sup>14</sup>C yr previously reported by Siani et al. (2000) for predominately western Mediterranean measurements.

A time-dependency in the reservoir age of the Mediterranean Sea and the North Atlantic was previously demonstrated for the early twentieth century (Siani et al. 2000). Our eastern Mediterranean and Aegean data add support to a decline in  $\Delta R$  from AD 1900 to 1930 (Figure 2). Larger variations in the Mediterranean  $\Delta R$  have occurred in the past due to changes in reservoir age of the North Atlantic water entering the Mediterranean or fluctuations in continental runoff, which contributes <sup>14</sup>C depleted carbonates and may alter the ventilation of deep and intermediate waters (Mercone et al. 2000; Siani et al. 2001).

Table 2b Compilation of <sup>14</sup>C ages and  $\Delta R$  values for known age shells from the eastern Mediterranean and the Aegean Seas. References as numbered: 1) Siani et al. 2000; 2) Delibrias 1989, as quoted in reference 1; 3) Pelc 1995, as quoted in reference 1; 4) this study; 5) Taviani and Correggiari (personal communication); 6) Langone et al. 1996.

		Collection year	<sup>14</sup> C age	$\Delta R$		
Location	Lab ID	(AD)	(BP)	(yr)	Reference	
Eastern Mediterranean						
Zante, Greece	CAMS-69547	1942	$620\pm40$	$153\pm41$	4	
Beirut, Lebanon	CAMS-69545	1929	$490\pm40$	$32 \pm 40$	4	
Beirut, Lebanon	CAMS-69546	1929	$400\pm50$	$-58\pm50$	4	
Netamiya, Israel	CAMS-69540	1937	$510\pm40$	$47 \pm 41$	4	
Aegean Sea						
Izmir, Turkey	CAMS-69539	1893	$750\pm40$	$288\pm40$	4	
Nauplia, Greece	CAMS-69541	1940	$500 \pm 40$	$35\pm50$	4	
Piraeus, Greece	CAMS-69542	1943	$610\pm40$	$143\pm41$	4	
Exact location unknown						
Mediterranean Sea	GifA 96700	1887	$585\pm35$	$118\pm35$	1	



Figure 2 ΔR values for the western Mediterranean including the Adriatic and the Tyrrhenian Seas (diamonds), the eastern Mediterranean (triangles), the Aegean Sea (open circles), and an unknown location in the Mediterranean (square) plotted versus collection year of the shell sample. The error bars represent one standard deviation based on counting statistics
1950 and the uncertainty in marine calibration dataset.

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Table 3 Regional means of  $\Delta R$  values and reservoir ages. The uncertainty in the mean is the larger of the standard deviation based on counting statistics and the "scatter sigma," which is the square root of the variance divided by the number of samples. The empirical standard deviation (s.d.) is the square root of the variance. The regional mean for the western Mediterranean includes samples from the Algerian coast, and the Tyrrhenian and the Adriatic Seas. The eastern Mediterranean regional mean does not include the Aegean Sea.

	Mean $\Delta R$	Nr of		Reservoir age
Region	( <sup>14</sup> C yr)	samples	Empirical s.d. (yr)	( <sup>14</sup> C yr)
Western Mediterranean	$40 \pm 15$	25	75	$400 \pm 22$
Eastern Mediterranean	$53 \pm 43$	4	86	$353 \pm 47$
Algerian coast	$83 \pm 33$	5	75	$413 \pm 51$
Tyrrhenian Sea	$45 \pm 21$	6	30	$390 \pm 21$
Adriatic Sea	$43 \pm 48$	5	108	$396 \pm 61$
Aegean Sea	$154 \pm 52$	4	105	$480 \pm 72$
Aegean Sea w/o sample of <i>Mytilus edulis</i>	$109 \pm 37$	3	65	$420\pm55$
All Mediterranean except the Aegean Sea	45 ± 14	30	75	$390 \pm 15$
All Mediterranean	$58\pm15$	34	85	$400\pm16$

Currently, no marine reservoir age data for the Mediterranean is available between the nineteenth century and ~4500 cal BP. However, at present the Atlantic Ocean surface waters flow into the Mediterranean through the Strait of Gibraltar and overturn in the eastern Mediterranean with a residence time of ~100 years (Broecker and Gerard 1969; Stuiver and Ostlund 1983). This circulation pattern appears to have been unchanged for the past 18,000 years except during the S1 sapropel formation between about 6000–9000 <sup>14</sup>C yr BP (~ 6800–10,200 cal BP) (Kallel et al. 1997).  $\Delta R$  in the subpolar North Atlantic has been shown to be constant within ± 95 <sup>14</sup>C yr over the past 6000 years based on measurements of contemporaneous marine and terrestrial samples (Reimer et al. 2002).

Reservoir ages based on <sup>14</sup>C dates of planktonic foraminifera associated with dated tephra layers and on charcoal/shell pairs from archaeological cave excavations support a relatively constant reservoir age for the eastern Mediterranean from about 4000–6000 <sup>14</sup>C yr BP (~4400–6800 cal BP) (Siani et al. 2001). Between about 7400–8500 <sup>14</sup>C yr BP (~8200–9500 cal BP), reservoir ages appear to have increased slightly. Facorellis et al. (1998) derived a reservoir age of 515 ± 22 ( $\Delta R = 149 \pm 30$ ) from paired mollusc shells (*Patella ulyssiponensis*) and charcoal samples from Cyclope cave on the island of Youra in the Aegean Sea. This increased reservoir age corresponds to the time of the S1 sapropel formation which may be related to changes in ventilation (Mercone et al. 2000; Siani et al. 2001). After this event reservoir ages return to near modern pre-bomb values.

Siani et al. (2001) measured a reservoir age of  $380 \pm 100$  near the beginning of the Younger Dryas (about 10,500 <sup>14</sup>C yr BP) which is comparable to the reservoir age of 320-345 yr about 10,700 <sup>14</sup>C yr BP calculated from paired marine and terrestrial samples from Cyprus (Simmons and Wigand 1994). Reservoir ages rose through the Bølling/Allerød reaching a maximum of  $820 \pm 120$  yr in the Older Dryas interval before declining to around the modern pre-bomb value during the Last Glacial Maximum (Siani et al. 2001). We discuss reservoir ages rather than  $\Delta R$  values beyond the tree-ring record (about 11,900 cal BP), because there the atmospheric <sup>14</sup>C calibration dataset is based on the marine record with an estimated reservoir correction (Stuiver et al. 1998a).

### CONCLUSIONS

 $\Delta R$  values are indistinguishable for different Mediterranean basins including the Aegean Sea. The recommended  $\Delta R$  for <sup>14</sup>C calibration of marine samples with the 1998 marine calibration dataset is  $58 \pm 85$  <sup>14</sup>C yr for the entire Mediterranean. The Mediterranean  $\Delta R$  appears to be relatively constant within this uncertainty for the past 6000 or 7000 yr, but beyond that time frame variations in  $\Delta R$  should be considered when calibrating <sup>14</sup>C ages for marine samples from this region.

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