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RADIOCARBON RESERVOIR AGES IN THE MEDITERRANEAN SEA AND BLACK SEA

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ABSTRACT. We measured apparent marine radiocarbon ages for the Mediterranean Sea, Black Sea, and Red Sea by accelerator mass spectrometry radiocarbon analyses of 26 modern, pre-bomb mollusk shells collected living between AD 1837 and 1950. The marine reservoir (R(t)) ages were estimated at some 390 ± 85 yr BP, 415 ± 90 yr BP and 440 ± 40 yr BP, respectively. R(t) ages in the Mediterranean Sea and Black Sea are comparable to those for the North Atlantic Ocean (<65°N), in accordance with the modern oceanic circulation pattern. The ΔR values of about 35 ± 70 yr and 75 ± 60 yr in the Mediterranean area show that the global box-diffusion carbon model, used to calculate R(t) ages, reproduces the measured marine ^{14}C R(t) ages in these oceanic areas. Nevertheless, high values of standard deviations, larger than measurement uncertainties are obtained and express decadal R(t) changes. Such large standard deviations are indeed related to a decrease of the apparent marine ages of some 220 yr from 1900 AD to 1930 AD in both the Mediterranean Sea and the western North Atlantic Ocean.

INTRODUCTION

Species living in ocean surface waters show depleted radiocarbon ages with respect to those living contemporaneously under atmospheric conditions. This difference, or marine ^{14}C apparent age, is due to 1) oceanic circulation processes that tend to advect intermediate and deep ^{14}C-depleted water masses to the surface, 2) atmospheric ^{14}C changes, and 3) air-sea CO_2 exchange processes.

Reservoir ages of the global mixed marine surface layer may be estimated from a global box-diffusion carbon model, which reproduces depth dependant marine ^{14}C variations in response to atmospheric ^{14}C changes (Oeschger et al. 1975; Stuiver et al. 1986; Stuiver and Braziunas 1993). Nevertheless, deviations (ΔR) from these modeled marine reservoir ages in response to local oceanic conditions are widely observed, as for example, in upwelling areas (Taylor and Berger 1967; Mangerud and Gulliksen 1975; Robinson and Thompson 1981; McFadgen and Manning 1990; Domack 1992; Dye 1994; Heier-Nielsen et al. 1995; Berkman and Forman 1996; Ingram and Southon 1997; Goodfriend and Flessa 1997).

Here we present measurements of the reservoir ages of surficial waters of the Mediterranean Sea, Black Sea, and Red Sea, which are still poorly known (Broecker and Olson 1961; Delibrias 1985; Cember 1988; Pelc 1995). In the Mediterranean area, such determinations are important to calibrate the ^{14}C ages of marine materials for accurate comparison of marine and continental geological, climatological records, together with human settlement, which are intimately tied in this region.

Oceanic changes in the Mediterranean Sea are mainly connected to those in the North Atlantic Ocean. Inflowing Atlantic Ocean surface waters at the Gibraltar Strait rapidly overturn at intermediate depths from the extreme Eastern Mediterranean basin into the North Atlantic Ocean with a residence time of some 100 years (Broecker and Gerard 1969; Stuiver and Ostlund 1983). Hence, in the modern Mediterranean Sea, coastal upwellings do not affect greatly the ^{14}C marine reservoir ages. Thus, circulation changes in the North Atlantic Ocean, freshwater input from the Mediterranean

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coastal rivers, and subsequent hardwater effects, together with ^{14}C atmospheric changes could contribute to changes of the ^{14}C apparent ages of the Mediterranean Sea surface waters.

MATERIAL AND METHODS

^{14}C reservoir ages were first measured by Broecker and Olson (1961) from one pre-bomb shell from the Algerian continental shelf at 360 ± 80 yr with a regional deviation ΔR at −133 yr. Three other ages of pre-bomb mollusks shells from the French and Algerian continental shelf averaged at some 350 ± 35 yr (Delibrias 1985). Nevertheless, recent ^{14}C datings of several shells collected along the French continental shelf show highly variable Mediterranean Sea reservoir ages (Pelc 1995).

In this study, 26 modern mollusk bivalves from the Mediterranean Sea, Black Sea, and Red Sea coasts were obtained from the collection of the Muséum National d'Histoire Naturelle, Paris (Table 1). The year of collection represents the date of addition to the collection, which is supposed to be that of collection of living mollusks. Shells include the three epifaunal species (*Chlamys glaber, Irus irus, Arca nodulosa*) and eight infaunal species (*Cerastoderma glaucum, Ruditapes decussatus, Venerupis aurea, Leda commutata, Glycymeris glycymeris, Nucula nucleus*, *Nucula margaritacea, Chamelea gallina*). Datings, collection sites, and dates of collection are reported in Table 1 and Figure 1.

^{14}C analyses were performed at the Gif-sur-Yvette Tandetron accelerator (Arnold et al. 1987). Shell samples (about 15 mg of carbonate) were etched with 0.5N hydrochloric acid and, after a 50% loss, were rinsed with deionized water to remove surface contaminations. Subsequently, 0.5 ml of phosphoric acid was added under vacuum and the sample was hydrolyzed to generate CO_2. The final step was to reduce the CO_2 to graphite using hydrogen with iron powder (Arnold et al. 1987). Two targets of iron-carbon powder, 2 mm in diameter, were then prepared and measured to improve the ^{14}C precision. Results are given as conventional ^{14}C ages in yr BP, based on the measured $^{14}C/^{12}C$ ratio, corrected for the natural isotopic fractionation by normalizing the results to the standard $\delta^{13}C$ value of −25‰ PDB (Stuiver and Polach 1977).

RESULTS AND DISCUSSION

The ^{14}C ages, R(t) and ΔR expressed in yr BP and, $\Delta^{14}C$ (‰) are listed in Table 1, including Gif measurements and data from the literature. The ^{14}C age reservoirs R(t) for the modern shells were calculated by subtracting the atmospheric ^{14}C value at the date of collection (Stuiver and Becker 1993) from the measured apparent ^{14}C ages of the mollusks.

Among all the data, three samples (GifA 96702, GifA 96719, and GifA 96723) show ^{14}C ages larger than 1500 yr BP (Table 1). These ages are considered too old, and these samples were not collected alive, likely from ancient cliffs.

In the Mediterranean Sea, the average R(t) age is calculated at 420 yr BP, and the dispersion (±110 yr) largely exceeds measurement uncertainty. The ^{14}C reservoir age of the Black Sea, extended to the Dardanelles Strait, calculated from six modern shells, is similar, and shows a slightly lower variability at 415 ± 90 yr BP over the same time period. Finally, one determination in the Red Sea yielded a ^{14}C reservoir age of 440 ± 40 yr (GifA 96703), in agreement with previous measurements at about 470 yr on recent corals (Cember 1988).

Table 1 AMS ^{14}C dates of modern pre-bomb mollusk shell samples in the Mediterranean Sea, Black Sea, and Red Sea, and their reservoir ages

Site nr	Lab code	Species	Collection site	Year of collection	Sample ^{14}C age (BP)	±1 σ	$\Delta^{14}C$ (‰)	±1 σ	Tree-ring ^{14}C age (BP)	±1 σ	Reservoir age R(t) (yr)	Model age	ΔR (yr)	$\delta^{18}O$ (PDB)
Liguro-Provençal														
1	GifA 96726	*Nucula nucleus*	Antibes	1873	450	40	−54.48	−5.0	121	5	329	482	−32	—
2	GifA 96724	*Nucula nucleus*	St Raphael	1892	455	35	−55.07	−4.3	93	9	362	470	−15	1.41
3	GifA 96699	*Chlamys glaber*	La Seyne	1892	470	40	−56.83	−5.0	93	9	377	470	0	0.94
4	GifA 96711	*Nucula margaritacea*	Marseille	1873	510	35	−61.51	−4.3	121	5	389	482	28	—
5	GifA 96709	*Venerupis aurea*	Marseille	1874	550	40	−66.18	−5.0	122	5	428	481	69	0.89
6	GifA 96716	*Arca nodulosa*	Banyuls	1906	570	35	−68.50	−4.3	84	8	486	464	106	1.91
7	GifA 96705	*Ruditapes decussatus*	Sète	1892	685	45	−81.74	−5.6	93	9	592	470	215	0.31
8	GifA 96706	*Ruditapes decussatus*	Marseille	1892	720	40	−85.73	−5.0	93	9	627	470	250	−0.79
9	Gif 3314[a]	*Cardium echinatum*	Beaulieu	1907	700	60	−83.45	−7.4	87	8	613	463	237	—
10	Ly 6872	*Acanthocardia tuberculata*	Sète	1900	720	45	−85.73	−5.6	84	9	636	467	253	—
11	Gif 4068[a]	*Arca noé*	Toulon	1837	405	35	−49.17	−4.3	118	6	287	504	−99	—
12	Ly 6900[b]	*Turritella sp.*	Banyuls	1900	565	55	−67.92	−6.8	84	9	481	467	98	—
Adriatic Sea														
13	GifA 96707	*Irus irus*	Dalmatia	1873	380	35	−46.20	−4.3	121	9	259	482	−87	−3.19
14	GifA 96718	*Leda commutata*	Rovigne	1926	390	50	−47.39	−6.2	148	6	242	466	−76	—
15	GifA 96722	*Glycymeris glycymeris*	Adriatic Sea	1867	540	30	−65.01	−3.7	120	6	420	486	54	1.36
16	GifA 96712	*Chamelea gallina*	Venice lagoon	1950	785	35	−93.10	−4.3	199	7	586	483	302	−1.78
Algero-Tunisian														
17	GifA 96710	*Nucula margaritacea*	Alger	1881	620	35	−74.28	−4.3	111	5	509	477	143	—
18	GifA 96720	*Leda commutata*	Oran	1900	740	35	−88.00	−4.3	84	9	656	467	273	—
19	Gif 4067[a]	*Turbo rugosus*	Cherchel	1905	460	35	−55.66	−4.3	81	8	379	464	−4	—
20	Ly 6948[b]	*biological limestone*	Mahdia	1948	500	50	−60.35	−6.2	199	7	301	481	19	—
Tyrrhenian Sea														
21	GifA 96704	*Cerastoderma glaucum*	Bastia	1921	495	40	−59.76	−5.0	133	6	362	465	30	−0.99
22	GifA 96717	*Arca tetragona*	Naples	1873	535	40	−64.43	−5.0	121	5	414	482	53	2.19
23	GifA 96725	*Nucula nucleus*	Naples	1892	610	110	−73.13	−13.6	93	9	517	470	140	1.8
24	Ly 6863[b]	*C. corrugatum*	Sicily	1900	525	50	−63.27	−6.2	84	9	441	467	58	—
Black Sea														
28	GifA 96714	*Chamelea gallina*	Istanbul	1951	540	40	−65.01	−5.0	199	7	341	483	57	−2.46
29	GifA 96713	*Chamelea gallina*	Istanbul	1900	545	40	−65.59	−5.0	84	9	461	467	78	−2.04
30	GifA 96708	*Irus irus*	Dardanelli Strait	1900	605	40	−72.55	−5.0	84	9	521	467	138	−2.70
31	GifA 96698	*Chlamys glaber*	Black Sea	1837	480	40	−58.00	−5.0	118	6	362	504	−24	—
32	GifA 96701	*Cerastoderma glaucum*	Black Sea	1843	615	40	−73.70	−5.0	119	6	496	499	116	−1.59
33	[c]	*Mytilus galloprovincialis*	Crimea	1931	460	35	−55.66	−4.3	153	6	307	486	−26	—
Red Sea														
34	GifA 96703	*Cerastoderma glaucum*	Red Sea	1904	525	40	−63.27	−5.0	82	8	443	465	60	3.08
Others														
25	GifA 96715	*Chamelea gallina*	Malaga	1929	430	35	−52.12	−4.3	152	6	278	467	−37	0.87
26	GifA 96700	*Chlamys glaber*	Mediterranean Sea	1887	585	35	−70.24	−4.3	101	5	484	473	112	1.10
27	GifA 96721	*Mactra corallina*	Port Saïd	1904	715	40	−85.16	−5.0	82	8	633	465	250	−1.93
Rejected														
35	GifA 96702	*Cerastoderma glaucum*	Zouara	1904	8870	100	−668.52	−12.4						
36	GifA 96719	*Leda commutata*	Palermo	1906	1630	60	−183.65	−7.4						
37	GifA 96723	*Nucula nucleus*	Sfax	1892	2020	60	−222.34	−7.4						

[a]From Delibrias (1985); [b]From Pelc (1995); [c]From Jones and Gagnon (1994).

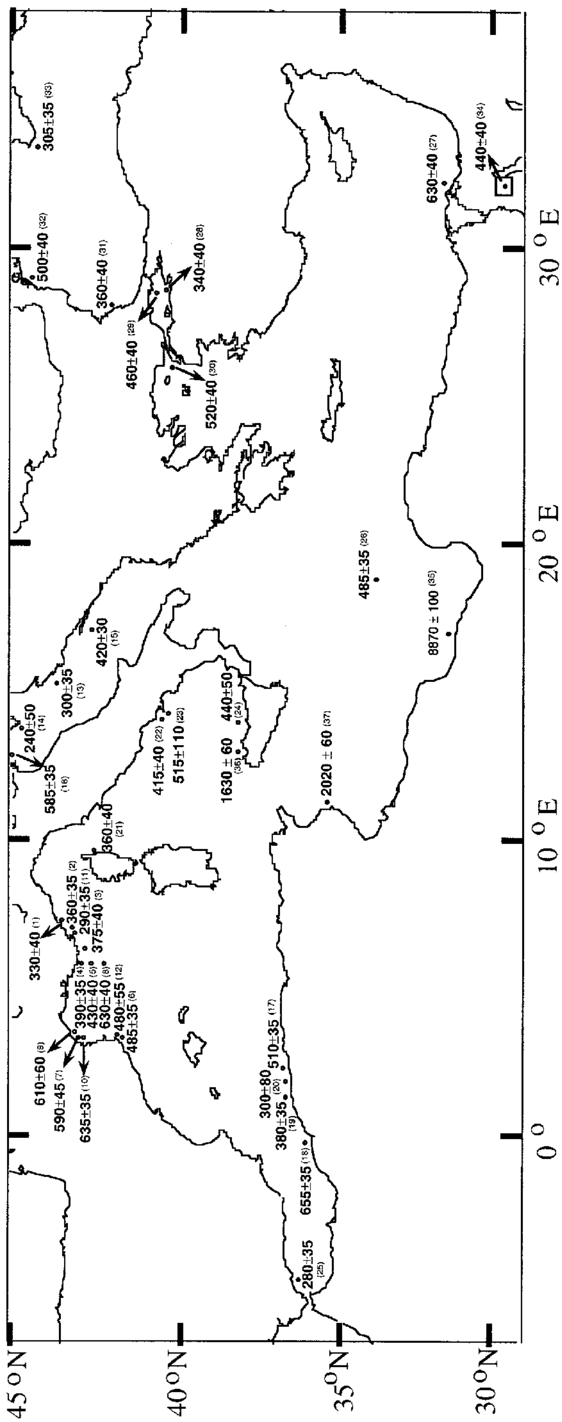

Figure 1 Reservoir ages R(t) and locations of marine shell samples. Numbers in parentheses refer to data list in Table 1.

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Variability as a Function of Sample Locations

The R(t) age distribution in the Mediterranean Sea over the considered time interval 1830–1950 AD ranges from 240 to 660 yr (Figure 2a). It shows three significant distinct populations at the 99% confidence interval (Student test), the main group centered at 420 ± 65 yr BP. The observed variability of the Mediterranean Sea R(t) cannot be attributed to a specific region, as it is observed in the Liguro-Provençal region, as well as along the Algero-Tunisian-Egyptian coasts and in the North Adriatic Sea region (Figures 1,2b; Table 1).

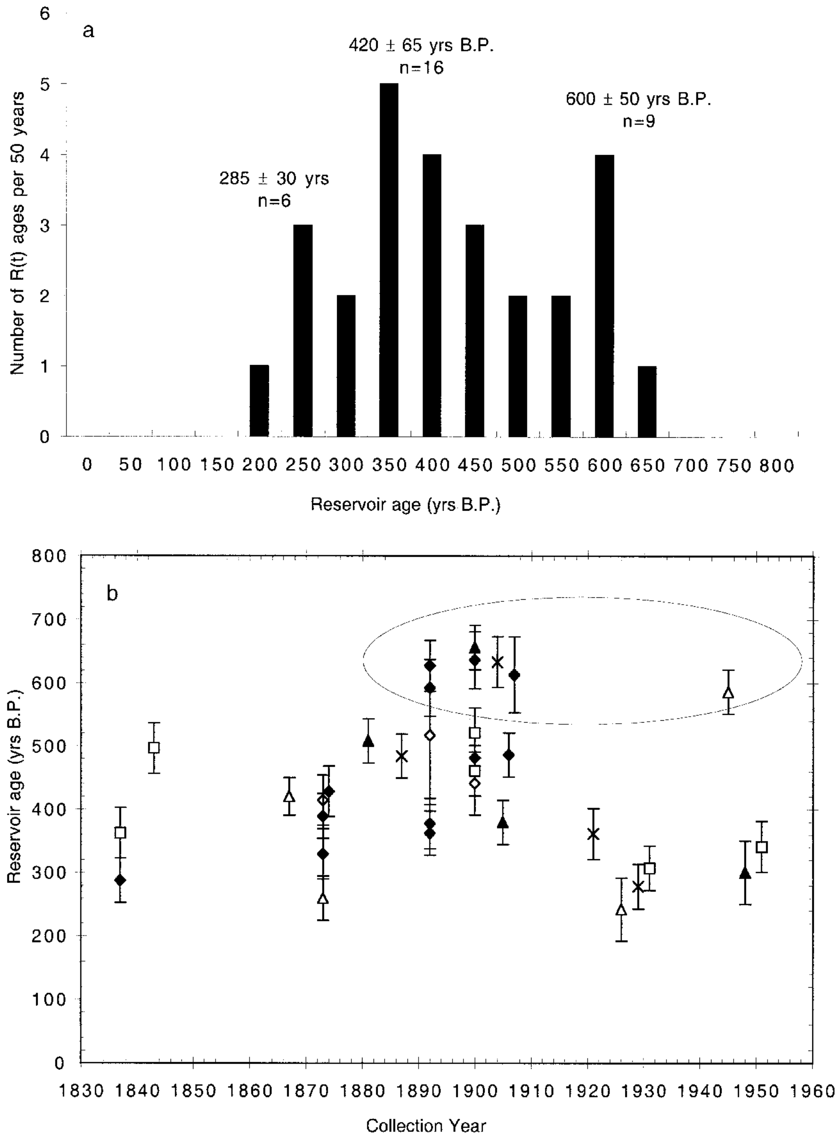

Figure 2 a) Histogram of the R(t) ages frequency as a function of a 50-yr step increase of R(t) for the Mediterranean Sea mollusk shell apparent ages in Table 1. b) R(t) ages as a function of collection year for samples from the Mediterranean Sea: i) Liguro-Provençal region (black diamonds), ii) Adriatic Sea (open triangles), iii) Algero-Tunisian coasts (black triangles), iv) Tyrrhenian Sea (open diamonds) and v) isolated points (crosses), from the Red Sea (open circles), and from the Black Sea (open squares).

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Among the seven samples of high reservoir ages at around 600 ± 50 yr BP, four mollusk shells come from the Liguro-Provençal coast where several rivers, such as the Rhone, Hérault and Aude, discharge after draining through limestones via several brackish lagoons, before reaching the Mediterranean Sea: 1) Ly 6872 at 636 ± 45 yr BP, collected at Sète, close to the Hérault River and Thau lagoon, and 2) Gif 33-14 at 613 ± 60 yr BP collected from the Rhone delta at Beaulieu. Moreover, the two other samples, GifA96705 and GifA96706, collected at Marseille and Sète, are of the same lagoonal species, *Ruditapes decussatus*. Thus, hardwater effects as well as biological processes may be suspected for this last sample (Figure 1; Table 1). ^{14}C-depleted freshwater contribution during growth of the mollusks could also explain the high reservoir ages of the samples: 1) GifA 96721 at 633 ± 40 yr BP, originating from Port-Saïd at the Nile River mouth, and 2) GifA 96712, dated at 586 ± 35 yr BP from the Lido near the Venice Lagoon. Finally, a sample GifA 96720 collected in Oran (Algeria) shows a high reservoir age at 656 ± 35 yr BP. The presence of submarine freshwater sources along the coast (Khereci and Messadi 1992) allows us to suggest that this sample may be influenced by hardwater effects.

Recrystallization processes, and subsequent contamination of the mollusk carbonate by modern carbon, would explain the R(t) group of younger ages centered at around 285 ± 30 yr BP (Figure 2a). But, on one hand, these mollusks do not present a specific depth habitat with respect to the others. On the other hand, such processes are difficult to observe because all the mollusks used have calcitic shells (Table 1). Therefore, we estimated the mean ^{14}C reservoir age of the Mediterranean Sea surface waters at 390 ± 85 yr BP (n=20), excluding only local/biological ^{14}C ageing phenomena. The mean ΔR value is calculated at 35 ± 70 yr by subtracting the Mediterranean Sea apparent age from the model age of the mixed oceanic layer (Stuiver and Braziunas 1993). This value is statistically similar to that of the Black Sea at 75 ± 65 yr and the Red Sea at 60 ± 40 yr.

We compared the Mediterranean Sea and Black Sea reservoir ages to the North Atlantic Ocean ones (Figure 3a), using published ^{14}C data on supposed live shells collected south of 65°N, and mainly in the eastern North Atlantic Ocean (Broecker and Olson 1961; Mangerud 1972; Mangerud and Gulliksen 1975; Heier-Nielsen et al. 1995). Using the same procedure for recent Mediterranean shells, the mean R(t) age for the North Atlantic Ocean is estimated at 380 ± 90 yr BP (n=33), and the mean ΔR at 20 ± 80 yr. The very similar reservoir ages of the surficial waters in both oceanic regions are in agreement with their oceanic circulation relationships. Moreover, the insignificant ΔR difference between the measured and modeled R(t) estimates suggest that the global box-diffusion carbon model mainly reproduces the marine ^{14}C variations in response to atmospheric ^{14}C changes in these areas (Oeschger et al. 1975; Stuiver et al. 1986; Stuiver and Braziunas 1993). However, the R(t) age variability in the two oceanic regions is 1.5 to 2 times higher than the dating uncertainties.

Variability as a Function of the Sampling Date

Rapid oscillations of the Δ^{14}C values of oceanic surface waters as a function of time were recently revealed by ^{14}C measurements on banded corals (Druffel 1997; Guilderson and Schrag 1998). In the western North Atlantic Ocean, the observed biennial to decadal ^{14}C variations were tied to changes in the vertical mixing of the water column related to the North Atlantic Oscillation (NAO) (Druffel 1997). To discriminate possible short-term fluctuations, which would explain the observed R(t) age dispersion, we averaged the R(t) ages obtained for a single year AD or two close year AD, when the standard deviation does not exceed the dating uncertainties (Table 2; Figure 3b). This procedure was applied to R(t) ages in the North Atlantic Ocean and in the Mediterranean Sea, extended to the Black Sea during the statistically most represented time interval (1860 AD to 1950 AD).

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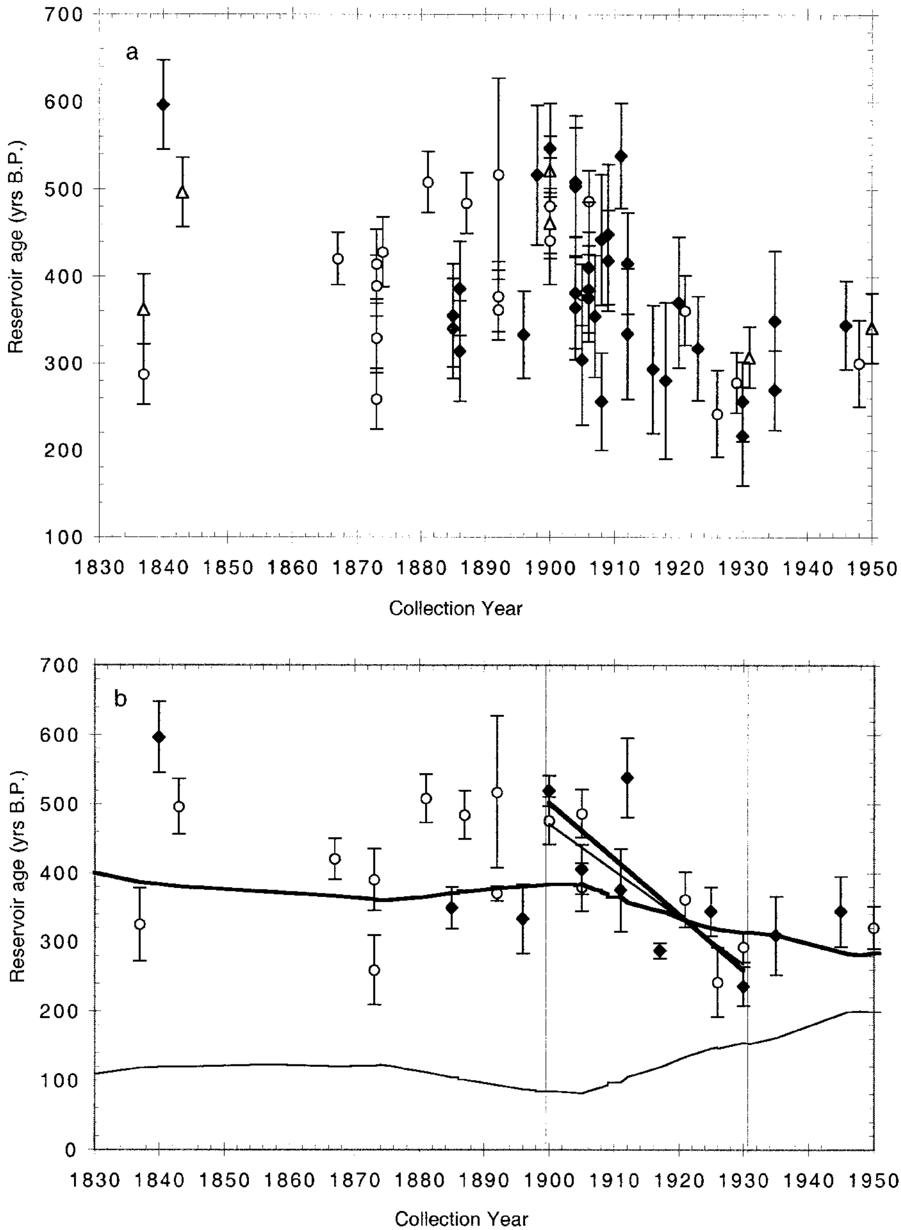

Figure 3 a) R(t) ages of mollusk samples from the Mediterranean Sea (open circles), Black Sea (open triangles) and from North Atlantic Ocean (<65°N) (black diamonds) vs. collection year. b) Same R(t) ages as Figure 3a, averaged for single or two close yr (see text) in the Mediterranean Sea (closed circles) and in the North Atlantic Ocean <65°N (black diamonds). Regression lines are shown between 1900 AD and 1930 AD in the Mediterranean Sea (R= 0.88; n=6) (thin straight line) and in the North Atlantic Ocean (R=0.77; n=7) (thick straight line), respectively. Atmospheric radiocarbon ages (thin line) and global modeled marine mixed layer radiocarbon ages (thick line) are also represented (Stuiver and Becker 1993; Stuiver and Braziunas 1993).

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Table 2 Mean AMS ^{14}C datings per year AD or two closed-year AD for the Mediterranean Sea/Black Sea shells and for the North Atlantic Ocean (<65°N) shells. References are given in the text.

Collection year	Sample ^{14}C age (BP)	±1 σ	Reservoir age R(t) (yr)	ΔR (yr)	Number of shell samples
Mediterranean and Black Seas					
1837	443	53	325	–62	2
1843	615	40	496	9	1
1867	540	30	420	54	1
1873	532	20	410	50	3
1881	620	35	508	143	1
1887	585	35	484	112	1
1890	463	11	370	–8	2
1900	560	34	476	93	4
1905	460	35	379	–4	1
1905	570	35	486	106	1
1921	495	40	361	30	1
1926	390	50	242	–76	1
1930	445	22	293	–22	2
1950	520	31	321	38	2
North Atlantic Ocean < 65°N					
1840	715	51	596	213	1
1885	452	30	349	–22	4
1896	420	50	333	–47	2
1900	602	22	519	137	3
1905	493	36	405	30	7
1911	479	60	375	17	2
1912	635	57	538	173	1
1917	406	11	287	–59	2
1925	479	35	344	14	2
1930	390	28	236	–77	2
1935	470	57	309	–1	2
1945	543	51	344	61	1

From 1860 AD to 1900 AD, the R(t) ages do not show significant variations (Figure 3b). From 1900 AD to 1930 AD, the R(t) values show a significant tendency to decrease, at the 99% confidence interval, by some −220 ± 40 yr (R=0.88; n=6). Such a feature is also observed in the North Atlantic Ocean during the same period, with a R(t) age decrease of similar amplitude at approximately −240 ± 25 yr (R=0.77; n=7). Both R(t) age differences are significant at 2σ according to the mean uncertainties (Table 2). In the interval 1900 AD to 1930 AD, the atmospheric ^{14}C ages inversely vary by an increase of about 80 yr (Stuiver and Becker 1993), due to atmospheric ^{14}C dilution by free $^{14}CO_2$ injected from combustion of old carbon (Suess effect). ^{14}C measurements on banded corals from the western North Atlantic Ocean show that such an effect could lag by some 25 yr in the marine environment (Druffel 1982, Figure 6). Consequently, the decreasing marine ^{14}C

trend between 1900 AD and 1930 AD could be overestimated. Nevertheless, the amplitude of this trend remains significant, without considering the atmospheric ^{14}C changes at 140 ± 40 yr and 160 ± 30 yr in the Mediterranean and Atlantic shells. Thus, the high dispersion of the mean R(t) ages is relevant to short-term marine ^{14}C fluctuations.

The interval 1900 AD to 1930 AD was previously recognized as a period of an unusually high NAO, with high winter pressure in Açores and anomalously low winter pressure in Iceland, leading to stronger westerlies over Europe (Hurrell 1995). Wind strengthening over the North Atlantic Ocean would tend to favor an increase of the air-sea CO_2 exchange, leading to ^{14}C renewal in the surficial water masses and decrease of the R(t) ages in the North Atlantic Ocean and the Mediterranean Sea. Previous calculations showed that the marine reservoir age would decrease by about 130 yr with a 50% change of mean wind speed (Bard 1988; Bard et al. 1994).

CONCLUSION

We have established a ^{14}C marine reservoir age correction for the Mediterranean Sea, Black Sea, and Red Sea surface waters, from AMS measurements of modern pre-bomb calcareous marine shells. Over the interval 1830 AD to 1950 AD, the marine reservoir age (R(t)) of the Mediterranean Sea is estimated at 390 ± 85 yr BP. This result is very similar to that for the North Atlantic Ocean (R(t) age (<65°N) at 380 ± 90 yr BP), in agreement with the modern ocean circulation pattern. Shells of live-taken mollusk provide a reliable material to analyze short-term marine ^{14}C changes, which are expressed by the large uncertainty on the R(t) estimates. Although there is agreement between the measured and modeled R(t) ages over the whole period considered, the global box-diffusion carbon model partially tends to smooth the short-term marine ^{14}C R(t) variations. During the interval 1900 AD to 1930 AD, the apparent marine ages decrease by some 150 yr in the Mediterranean Sea and Black Sea as well as in the western and eastern North Atlantic Ocean. This may account for the influence of the Suess effect together with an increase of air-sea CO_2 exchanges, favored by the NAO within stronger and frequent wind storms over the North Atlantic Ocean.

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