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Variability of ^{14}C reservoir age and air–sea flux of CO_2 in the Peru–Chile upwelling region during the past 12,000 years

Matthieu Carré [a,*], Donald Jackson [b], Antonio Maldonado [c], Brian M. Chase [a], Julian P. Sachs [d]

[a] Institut des Sciences de l'Evolution, Université de Montpellier, CNRS, IRD, EPHE, place Eugène Bataillon, 34095 Montpellier, France
[b] Departamento de Antropología, FACSO, Universidad de Chile, Ignacio Carrera Pinto 1045, Ñuñoa, Santiago, Chile
[c] Centro de Estudios Avanzados en Zonas Aridas (CEAZA), Universidad de La Serena, Casilla 599, La Serena, Chile
[d] School of Oceanography, University of Washington, Box 355351, Seattle, WA 98195, USA

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ABSTRACT

The variability of radiocarbon marine reservoir age through time and space limits the accuracy of chronologies in marine paleo-environmental archives. We report here new radiocarbon reservoir ages (ΔR) from the central coast of Chile (~32°S) for the Holocene period and compare these values to existing reservoir age reconstructions from southern Peru and northern Chile. Late Holocene ΔR values show little variability from central Chile to Peru. Prior to 6000 cal yr BP, however, ΔR values were markedly increased in southern Peru and northern Chile, while similar or slightly lower-than-modern ΔR values were observed in central Chile. This extended dataset suggests that the early Holocene was characterized by a substantial increase in the latitudinal gradient of marine reservoir age between central and northern Chile. This change in the marine reservoir ages indicates that the early Holocene air–sea flux of CO_2 could have been up to five times more intense than in the late Holocene in the Peruvian upwelling, while slightly reduced in central Chile. Our results show that oceanic circulation changes in the Humboldt system during the Holocene have substantially modified the air–sea carbon flux in this region.

Introduction

Extending over 5000 km from the equator to ~50°S, the Peru–Chile coastal upwelling region is the longest eastern boundary upwelling system in the world. The Peru–Chile coastal upwelling plays a significant role in the global carbon cycle, being a highly productive area (Chavez et al., 2008) as well as one of the most intense carbon sources of the global coastal ocean (Laruelle et al., 2010). There is growing evidence that eastern boundary upwelling systems are intensifying with global warming (Bakun, 1990; McGregor et al., 2007; García-Reyes and Largier, 2010; Narayan et al., 2010; Gutiérrez et al., 2011). In addition, climate model simulations recently projected a change in upwelling spatial structure with future intensification being larger at high latitudes than at low latitudes (Wang et al., 2015). Assessing current changes in upwelling systems requires knowledge of their natural temporal and spatial variability.

The Humboldt Current system is complex, involving water masses from the Pacific Equatorial undercurrent in the north and subantarctic surface and intermediate waters in the South, which are characterized by different $\Delta^{14}C$ values (Toggweiler et al., 1991; Strub et al., 1998). The difference in the ^{14}C age of dissolved inorganic carbon (DIC) in marine surface waters relative to the ^{14}C age of contemporaneous terrestrial carbon in equilibrium with the atmosphere is referred to as the marine radiocarbon reservoir age (R) and is due to the residence time of carbon in the ocean. Today, the average marine radiocarbon reservoir age in the ocean mix layer is assumed to be 400 yr by convention. For the past 10,500 years, the Marine13 radiocarbon calibration dataset (Reimer et al., 2013) includes a radiocarbon reservoir age calculated using the atmospheric ^{14}C calibration curve IntCal13 and an ocean–atmosphere box diffusion model (Reimer et al., 2013). Although the model is simplified, the calculated marine radiocarbon curve is consistent with independent estimates from marine archives (Reimer et al., 2013). From 10.5 to 13.9 cal ka BP, the marine radiocarbon calibration includes data from Cariaco Basin varved sediments and from corals (Reimer et al., 2013). Local deviations from the global reservoir age (ΔR), however, vary in space and time with oceanic circulation.

Surface waters off Peru and northern Chile are typically characterized by large marine reservoir ages owing to the upwelling of ^{14}C-depleted deep waters. ΔR values may change on seasonal (Jones et al., 2007, 2010) to multi-millennial time scales as a function of variations in upwelling intensity and/or the origin of the upwelled waters (Toggweiler et al., 1991; Fontugne et al., 2004; Ortlieb et al., 2011). Modern and past ΔR estimates are scarce in the southeast Pacific. On the Chilean coast south of 24°S, only two estimates of pre-bomb ΔR are available today in the 14CHRONO marine reservoir database (http:// http://calib.qub.ac.uk/marine/). A few estimates of Holocene ΔR are available from sediment cores collected off southern Chile

* Corresponding author.
E-mail address: matthieu.carre@umontpellier.fr (M. Carré).

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(De Pol-Holz et al., 2010, Van Beek et al., 2002; Siani et al., 2013) and none from the coast. Therefore, the uncertainty in ΔR is considerable along ~3600 km of coast, which is a significant limitation on the accurate ^{14}C-dating of ancient sedimentological, biological, or archeological materials of marine origin.

Here we provide new estimates of ΔR over the past 12,000 years in central Chile (31°S–33°S) from paired charcoal and mollusk shells collected in archeological shell middens in the area of Los Vilos. The coast in this area is open to the ocean and does not show any local oceanographic feature or any large river system, so we can consider it as representative of the coastal Humboldt system at this latitude. A comparison with reconstructions from southern Peru and northern Chile compiled by Ortlieb et al. (2011) provides new insights into the spatial structure variability of the globally significant Peru–Chile coastal upwelling system. Based on an empirical relationship between ^{14}C reservoir age and pCO_2 in the southeast Pacific, we discuss the implications for past variability of air–sea CO_2 exchange in this region.

Material and methods

A pre-bomb radiocarbon reservoir age value was estimated from a *Mesodesma donacium* shell collected in Valparaiso and deposited in Paris at the National Museum of Natural History in 1837. This modern reservoir age value might be slightly overestimated since this shell had probably been collected a few years earlier. Holocene ΔR values were estimated from ^{14}C dates of paired mollusk shells and charcoal fragments collected in seven archeological shell middens close to Los Vilos (31.9°S, 71.5°W), on the central coast of Chile (Fig. 1, Table 1). Details on hunter-gatherer archeological occupations around Los Vilos can be found in Jackson (2002) and Méndez and Jackson (2004, 2006). Some authors recommend dating multiple pairs so that contemporaneity of samples can be statistically tested (Russell et al., 2011), which was not possible here. However, the risk of non-contemporaneity was here minimized by a careful control of the archeological context and the stratigraphy. Shell middens used in this study, except for one site (Ñague, Table 1), were thin lenses resulting from ephemeral occupations lasting approximately one season. We selected these sites to ensure a contemporaneous deposition of charcoal and shells. In the Ñague middens, contemporaneity was ensured by selecting shells that were collected in the stratigraphy between two charcoal fragments that yielded statistically undistinguishable radiocarbon dates (Table 1).

The estimate of a marine reservoir age can be biased if the charcoal fragment comes from a tree that died centuries before being used as fuel. This issue, referred to as the "old wood" effect, can be relatively common in the hyper arid coast of Peru (Kennett et al., 2002). While charcoal fragments are sometimes older than associated shells in northern Chile due to the old wood effect and must thus be discarded (Ortlieb et al., 2011), this was not observed in any pair analyzed in central Chile. This risk is mitigated in the central coast as compared to the Atacama desert because central Chile is much less arid, and so dead trees are not as well preserved as in the Atacama desert. Plant species could not be determined from charcoal fragments. However, we minimized the risk of old wood bias by analyzing two charcoal fragments when possible.

We used shell fragments from the same species, *M. donacium*, to minimize variability related to microhabitat or to biological effects. *M. donacium* is a filter-feeder bivalve living in the intertidal to subtidal zone of high energy sand beaches of Peru and Chile (Tarifeño, 1980). Filter feeders are considered to be in equilibrium with dissolved inorganic carbon and thus well suited for ΔR reconstructions (Petchey and Ulm, 2012). Seasonal changes in coastal upwelling can result in substantial

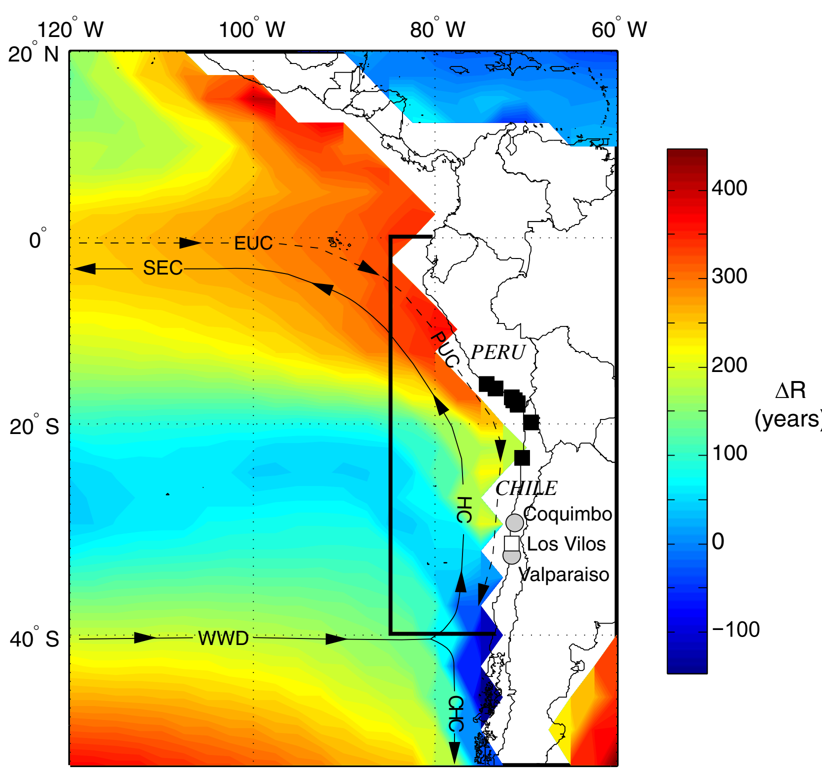

Figure 1. Map of the study region showing simulated marine reservoir age deviations (ΔR, years) of surface water (Butzin et al., 2012) in modern pre-bomb conditions. A simplified representation of ocean circulation (continuous arrows for surface currents and dashed arrows for undercurrents) based on Strub et al. (1998) indicates the South Equatorial current (SEC), the Equatorial undercurrent (EUC), the Peruvian undercurrent (PUC) (which feeds coastal upwelling), the Humboldt current (HC) (also called the Peruvian current), the West Wind drift (WWD), and the Cape Horn current (CHC). We show the sites for modern pre-bomb reservoir age estimates (gray circles), published Holocene reservoir age estimates in southern Peru and northern Chile (black squares) (Southon et al., 1995; Kennett et al., 2002; Owen, 2002; Fontugne et al., 2004; Ortlieb et al., 2011), and Los Vilos, the site for Holocene reservoir age estimates in central Chile (this study, open square). The thick black line shows the area considered for the calculation of the relationship between pCO_2 and ΔR (Fig. 3).