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AUSTRALIAN MARINE RADIOCARBON RESERVOIR EFFECTS: ΔR ATLAS AND ΔR CALCULATOR FOR AUSTRALIAN MAINLAND COASTS AND NEAR-SHORE ISLANDS

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ABSTRACT. Studies of pre-bomb mollusks live-collected around the Australian coastline have concluded that near-shore marine radiocarbon reservoir effects are small and relatively uniform. These studies are based on limited samples of sometimes dubious quality representing only selective parts of Australia's lengthy coastline. We systematically examine spatial variability in the marine radiocarbon reservoir effect (ΔR) through analysis of 292 live-collected mollusk samples across the Australian mainland coasts and near-shore islands subject to strict selection criteria. This study presents 233 new ΔR values combined with an evaluation of 59 previously published values. Results demonstrate significant spatial variability in marine radiocarbon reservoir effects across the study region. ΔR values range from 68 ± 24 ^{14}C years off the Pilbara region of Western Australia to –337 ± 46 ^{14}C years in the southern Gulf of Carpentaria in Queensland. Most sets of local values exhibit internal consistency, reflecting the dominant influence of regional oceanography, including depletion in ΔR values southwards along the eastern Australian coastline coincident with the East Australian Current. Anomalous values are attributed to inaccurate documentation, species-specific relationships with the carbon cycle and/or short-term fluctuations in marine radiocarbon activities. To account for the heterogeneous distribution of marine ^{14}C, we recommend using a location specific ΔR value calculated using the Australian ΔR Calculator, available at: https://delta-r-calc.jcu.io/.

KEYWORDS: marine ^{14}C, marine radiocarbon reservoir effects, mollusks, MRE, radiocarbon, ΔR.

INTRODUCTION

Radiocarbon (^{14}C) ages obtained on contemporaneous terrestrial and marine samples are not directly comparable. On average, Holocene subtropical marine samples are ca. 400 ^{14}C years older than contemporaneous terrestrial samples (Stuiver et al. 1986; Bard 1988). The difference is caused by marine radiocarbon reservoir effects (MRE) that reflect the different residence time of carbon in marine versus atmospheric reservoirs. While ^{14}C produced in the upper atmosphere is rapidly and relatively evenly incorporated throughout the atmosphere, long residence times for carbon in the deep ocean and uneven mixing of upwelling deep ocean waters cause significant spatial variability in global MRE (Mangerud 1972). A range of other factors

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can impact local MRE, including local upwelling along with temperature, wave, wind, and current regimes influencing air-sea gas exchange (Alves et al. 2018). Local differences can be particularly marked in estuaries and lagoons where local geology, terrestrial freshwater inputs, and incomplete exchange with the open ocean can accentuate MRE (Little 1993; Ulm 2002; Ulm et al. 2009; Petchey et al. 2023).

Successive marine calibration curves (Marine04, Marine09, Marine13, Marine20) have modeled global-scale surface ocean ^{14}C activity (Hughen et al. 2004; Reimer et al. 2009, 2013; Heaton et al. 2020). The difference between local and modeled global-scale surface ocean ages is expressed as ΔR (Stuiver et al. 1986). Negative ΔR values reflect lower MRE for the studied region compared with the global marine model and vice versa. Local ΔR values can be determined from a variety of approaches (the most common include ^{14}C dating known-age marine samples; ^{14}C dating paired contemporaneous marine/terrestrial samples; and paired ^{14}C/U-series dating of corals) (Stuiver and Braziunas 1993; Hua et al. 2020). To avoid ambiguity, here we use the term ΔR when discussing local correction values in general and ΔR_{XX} when denoting offsets to a specific calibration curve (i.e., ΔR_{20} when referring to a ΔR value derived from and for use with the Marine20 marine calibration curve) (see Heaton et al. 2023). ΔR and ΔR variability are usually small in areas where surface waters are well-mixed (e.g., east coast of Australia) but can be large and highly variable in areas with strong upwelling of deep waters with long residence times (e.g., Southern California, Antarctica) (e.g., Culleton et al. 2006; Hall et al. 2010).

To date, relatively few MRE studies have been undertaken in Australia compared with other areas of the globe (see Reimer and Reimer's 2001 14CHRONO Centre Marine Reservoir Correction Database, http://calib.org/marine/). This is at least partly attributable to the enduring impact of Gillespie's (1975, 1977; Gillespie and Temple 1977; Gillespie and Polach 1979) pioneering studies that demonstrated relatively little variability in MRE based on the dating of six marine shell specimens from four locations around the Australian coastline. Subsequent studies proposed only minor deviations based on local studies that largely supported the values proposed by Gillespie (e.g., Rhodes et al. 1980; Bowman and Harvey 1983; Gill 1983; Head et al. 1983; Bowman 1985a, 1985b). For decades it was a common practice in Australia to adjust ^{14}C ages on marine samples for MRE by simply subtracting 450 or 450 ± 35 ^{14}C years to make them comparable to coeval terrestrial samples. These ages were often reported as "corrected shell dates" (e.g., Godfrey 1989; O'Connor 1989; Bird and Frankel 1991; Sim 1998). Conventional radiocarbon ages (cf. Stuiver and Polach 1977) were often not listed, making the use of the published marine shell ^{14}C ages and their calibration problematic. Despite several studies suggesting variability in MRE (e.g., Hughes and Djohadze 1980; Woodroffe et al. 1986; Woodroffe and Mulrennan 1993; Murray-Wallace 1996; Spennemann and Head 1996; Ulm 2002; Ulm et al. 2009), no systematic study of Australian MRE has been undertaken (cf. Ulm 2006).

Studies in Australian coastal archaeology and geomorphology are highly dependent on ^{14}C ages obtained on marine samples, particularly mollusks. Mollusk remains are the dominant component of many coastal deposits, often with limited or no representation of other material (e.g., charcoal) suitable for radiocarbon dating. Mollusks may be preferred for dating owing to their relatively short lifespan and often larger surface area, limiting movement in deposits. In contrast, charcoal samples (where available) may have potentially large in-built ages (e.g., "old-wood effect" for charcoal, see Schiffer 1986), and even "short-lived" plant materials might have inbuilt age of 50–150 years (Anderson 1991; Allen and Wallace 2007). Individual small charcoal pieces can also move more readily across the matrix, distorting local chronologies.

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Mollusks are represented in archaeological deposits in Australia dating from at least 42,000 cal BP (Veth et al. 2017), making marine mollusk samples central to the chronology-building that underpins our understanding of the human history of the continent. The SahulArch dataset (Saktura et al. 2023) hosted on the Octopus database (Codilean et al. 2022) indicates that more than 23% of ages from archaeological deposits across the continent are on marine mollusk samples (26.4% of ages from Holocene archaeological deposits). Determining ΔR values and developing an understanding of ΔR variability through space and time is therefore crucial to refining chronologies in Quaternary science.

This study specifically examines spatial variability in ΔR around the Australian coastline measured in mollusks live-collected between AD 1841 and AD 1956. By providing a large-scale assessment of ΔR values and variability across the continent, this study improves the calibration of radiocarbon ages based on marine materials, which is essential for assessing the antiquity of, and changes in, Aboriginal and Torres Strait Islander social practices, cultures, and technologies through the archaeological record and the impacts of coastal change on human populations (e.g., Williams et al. 2018). Results also have implications for geomorphology, providing new information on modeling rates and impacts of sea-level change (e.g., Sloss et al. 2018), coral reef development and evolution of reef islands (e.g., Chivas et al. 1986; Woodroffe et al. 2007), as well as the evolution of the Australian coastline and the impact of environmental change on coastal landscapes. The results and their application have important implications for coastal management and conservation efforts, as understanding the history of coastal environments is crucial for making informed decisions about their protection and management.

METHODS

Synthesis of Previous Studies

A systematic review was undertaken of all previously published ΔR values available from live-collected mollusks around the Australian coast using the 14CHRONO Centre Marine Reservoir Correction Database (Reimer and Reimer 2001) as well as an extensive literature search to achieve comprehensive data collection (Tables S1–2). This review is restricted to mollusks, which are the most commonly ^{14}C-dated materials in Australian coastal archaeological and geomorphological studies. There are a small number of studies of recent marine radiocarbon reservoir effects based on coral records that are not included here (Druffel and Griffin 1993, 1999; Squire et al. 2013; Hua et al. 2015; Komugabe-Dixson et al. 2016; Wu et al. 2021). However, a comparison between the ΔR values based on mollusks reported here and those based on these corals within the time frame of our study is presented in Results and Discussion below.

A total of 59 ΔR values obtained on live-collected mollusks have been published prior to the current study (Table S2; Figure 1). Full details are presented in Table S1. Laboratory reports were checked, where available, and original primary publications were accessed to confirm details for legacy ages.

Identification of Samples in Museum Collections

To expand the geographical coverage of live-collected mollusk samples, all major Australian museum malacology collections were systematically evaluated for appropriate samples, including the Australian Museum, Queensland Museum, Western Australian Museum, South Australian Museum and Museums Victoria.

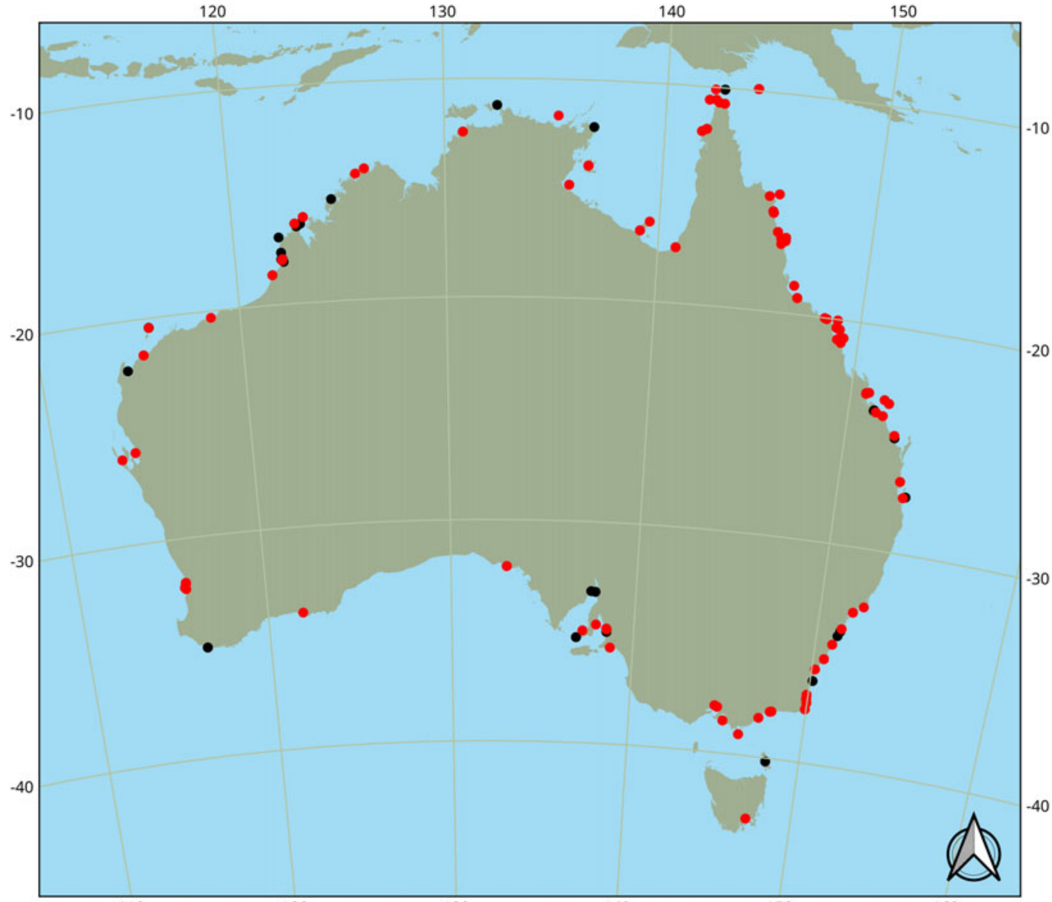

Figure 1 Map of Australia showing locations of ΔR samples discussed in this study. Black dots represent ΔR values reported in previous studies. Red dots represent new values reported in this study. (See online version for color figures. See Tables S1–3 for full details and Figures S1–18 for full-size figures.)

Evaluation of museum collections commenced with a review of mollusks identified on museum databases as live-collected in or before AD 1950. These specimens were then located in collections and visually inspected for suitability against the sample selection criteria (see below). Many of these specimens were rejected at this stage, typically because no physical or documentary evidence clearly demonstrated live-collection. Often accession or donation dates were recorded on museum databases rather than live-collection dates. Another limitation of relying on museum database searches to identify samples stems from the large proportion of materials held in museum malacology collections that are not accessioned. Furthermore, much of the critical information recorded in documentary material accompanying specimens or on hand-written museum accession registers has not been migrated to online databases.

The second stage involved a systematic visual examination of all holdings and associated documentation of taxa (especially bivalves) targeting taxa commonly selected in radiocarbon dating of archaeological deposits. Williams et al. (2014) report that *Anadara* spp./*Tegillarca* spp., *Ostrea* spp./*Saccostrea* spp. and *Latona* spp. (syn. *Donax* spp., *Plebidonax* sp.) are the

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dominant marine bivalves dated in Australian archaeology. Where no suitable target taxa were identified, the evaluation was broadened to include taxa well-represented in museum collections guided by curatorial and collection management staff.

Selection Criteria

Expanding and refining the criteria outlined in O'Connor et al. (2010), rigorous sample protocols were developed and implemented for use in this study. Samples had to meet all these conditions to be included in subsequent analyses. The test for inclusion was applied sequentially down the list 1–9 below (i.e., a sample might be excluded on the basis of multiple criteria, but was listed as excluded on the basis of its highest-ranked criterion for exclusion).

1. *Phylum: Mollusca. Class: Bivalvia:* Samples must be bivalves. Mollusks are the most dated marine materials in archaeology and Quaternary studies in Australia (Williams et al. 2014; Saktura et al. 2023). This study excludes values based on gastropods and non-molluscan marine exoskeletal material (e.g., coral). Gastropods have been shown to be more problematic for MRE studies as their detrital, carnivorous, herbivorous, algal grazing and/or omnivorous feeding systems provide pathways for the incorporation of non-dissolved inorganic carbon (non-DIC) sources into mollusk shell structures (Tanaka et al. 1986). ^{14}C activity in suspension-feeding (or filter-feeding) bivalves should reflect dissolved inorganic carbon (DIC) of the ambient waters in which they lived (Petchey et al. 2013).

2. *Live-Collected:* Samples must have been collected as live specimens. Many specimens in museum dry collections exhibit bleaching, edge-rounding, marine growth on inside surfaces and/or bore holes indicating a time-lag between death and collection. Only specimens with definitive evidence of live-collection, such as unambiguous documentation, the retention of the desiccated animal or the presence of residual ligament, muscle and/or periosteum were accepted for this study.

3. *Live-Collected ≤AD 1950:* Samples must have collection dates before or in AD 1950. Atmospheric nuclear weapon testing resulted in enriched atmospheric and oceanic ^{14}C levels after this time. This 'bomb effect' has been detected in coral cores from the Pacific Ocean and Indonesian Throughflow from c.AD 1954 (e.g., Andrews et al. 2016; Wu and Fallon 2020). Ascough et al. (2005) recommend use of samples live-collected before AD 1890 to avoid the combined effects of atmospheric nuclear weapon tests and the Suess effect caused mainly by industrial-scale burning of fossil fuels. Unfortunately, very few live-collected specimens in Australian museum collections (and only five samples in this study) pre-date AD 1890. Although this is an important consideration for the calculation of the marine reservoir age, ΔR values are calculated from the offset with the marine calibration curve which is modeled with atmospheric ^{14}C as input thereby including the Suess effect (Heaton et al. 2020).

4. *Collection Date Known to Within ≤1 Year:* Samples must have specified collection dates that confine their collection to a single calendar year. Many of the amateur mollusk collectors who donated to museums collected over decades and often collections were acquired by the museums many years subsequent to their collection (e.g., after the death of the collector). In some cases, the date of specimen collection can only be confidently bracketed by the active collecting years of a particular collector. For example, Bowman (1985a) used samples from the Bernard Bardwell Collection whose collection dates could only be determined to be between AD 1902 and AD 1950 using biographical information.

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Only specimens with a firm collection date or where collection can be constrained within a single year were accepted for this study. Although the time resolution of Marine20 is 10 years (Heaton et al. 2020), this criterion is designed to identify samples with ambiguous collection dates. Individual museum lots of mollusks collected over a period of more than a year have an elevated risk of containing mixed samples from different years and different locations. This criterion is designed to remove those samples.

Even where the year of collection appears unambiguous, sources of error can occur. For example, samples Wk-43560, Wk-43561, and Wk-43562 were dated on the basis that documentation accompanying the samples indicated a collection year of AD 1950. The specimens all conjoin and exhibit ligament and color suggesting live-collection. Subsequent radiocarbon dating revealed that the mollusks lived post-AD 1960s, after ^{14}C derived from atmospheric nuclear bomb tests entered the oceans ($F^{14}C$ values of 1.055, 1.023, and 0.989, respectively) (Reimer et al. 2004). Subsequent examination of museum records and other sources (Wilson and Stevenson 1977:98) indicated that nearly all specimens from the Broome area sourced from Anthony Kalnins were collected in the mid-to-late AD 1960s, indicating likely mislabelling of the year of collection and/or mixing of specimens in the lot.

5. *Known Collection Location:* Samples must have reliable provenance data (i.e., geographic location). There are many complexities that affect samples at the local level, such as upwelling, tidal flushing, terrestrial runoff effects from freshwater input, and local geology (Dye 1994; Stuiver and Braziunas 1993; Ulm 2002; Ulm et al. 2009). As some species have wide tolerance levels, detailed collection provenance is essential.

6. *Species Identified:* Samples must be reliably identified to species. Different species, even those belonging to the same family, may exhibit differences in diets and relationships with the carbon cycle, contributing to variability in ΔR values (see Criterion #1 above) (Petchey et al. 2012, 2013). Only samples identified to species are included in this study.

7. *Suspension-Feeder:* Samples must be suspension-feeding species. ΔR values will vary as a result of species-specific feeding habits (i.e., carnivores, deposit feeders, algal grazers, omnivores or suspension feeders). Carnivores (e.g., *Syrinx aruanus*, *Melo amphora*), deposit feeders (e.g., *Terebralia* spp., *Telescopium* spp.) and algal grazers (e.g., *Rochia* spp.) are likely to have a greater uptake of carbon from the other animals, sediments and geology they feed upon (Tanaka et al. 1986; Hogg et al. 1998; Petchey et al. 2012, 2013). Some grazers with magnetite-toughened teeth remove and ingest the surface of the rock with the algae, potentially exacerbating this problem. Although most bivalves are suspension-feeders, several families including Cyrenidae and Tellinidae appear to be able to switch to other feeding pathways (Snelgrove and Butman 1994:151; Beesley et al. 1998:342; Twaddle et al. 2017). These taxa are excluded from the analyses presented here.

8. *Ages from Same Lot, Pair or Valve have ^{14}C ages that are Statistically Indistinguishable:* Ages from samples assumed to be collected from the same location and same time, but not returning similar ages, are excluded. Dissimilar ages from the same lot (i.e., ages on multiple individuals) suggest confounding problems, for example, mixed mollusks in the lots (see Criterion #4 above), pointing to possible mixing of specimens within the lot, potentially from different times and locations. Dissimilar ages from the same pair (i.e., ages on both valves of a single bivalve) could indicate laboratory errors, contamination, or sampling of different years of growth. Where individual specimens of the same taxa from the same lot have more than one radiocarbon determination (e.g., both valves of a bivalve, or multiple valves sampled from the

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same lot), ages were subject to a chi-squared test to test if they are coeval following the procedures outlined in Ward and Wilson (1978). Specimens with ages that failed the chi-squared test were excluded. For specimens and lots with two or more ^{14}C ages, a chi-squared test was undertaken to determine whether the ages were statistically indistinguishable. For lots, pairs or valves with only two ages that failed the test, all ages were excluded from further analysis as it was not possible to determine which age accurately reflected the true age of death of the specimen. For specimens with three or more ages, the T statistic was used to identify non-contemporaneous age/s and exclude the affected samples from subsequent analyses (Ward and Wilson 1978). Details are presented in Table S1.

9. *Only a Single Mollusk Dated:* Samples are only included where a single mollusk is dated, avoiding samples comprising multiple individual mollusks with different life histories. Some studies using conventional radiocarbon dating methods (e.g., liquid scintillation counting) combined multiple mollusks, sometimes from different taxa, to reach minimum sample sizes (e.g., Bowman 1985a).

Physical Collection of Samples

After photographing each specimen, an 8–10 mm-long and ~4–5 mm-wide sample was taken parallel to the margin of each shell using a Dremel® 3000 Rotary Tool fitted with a diamond wheel. This sample size is designed to achieve adequate quantities for ^{14}C, ^{18}O, and ^{13}C analyses, minimise damage to samples, avoid seasonal variation and give an average value approximating the ^{14}C age of death of the mollusk (Culleton et al. 2006; Petchey et al. 2008).

Radiocarbon Dating

Accelerator mass spectrometry (AMS) ^{14}C age determinations were undertaken at the University of Waikato Radiocarbon Dating Laboratory and the Australian Nuclear Science and Technology Organisation (ANSTO).

Waikato samples were pre-treated following standard AMS protocols (UCI KCCAMS Facility 2011a, 2011b). Shell (<3 mm fragments, 35–45 mg) were etched in 0.1M HCl at 80ºC to remove ~45% of the surface, then dried. Cleaned shells were then tested for recrystallization by Feigl staining (Friedman 1959) to ensure either aragonite, or a natural aragonite/calcite distribution was present in the shell. CO_2 was collected from shells by reaction with 85% H_3PO_4 under vacuum at 70ºC for 30 min. Cryogenically separated CO_2 was reduced to graphite with H_2 at 550°C using an iron catalyst. $\delta^{13}C$ was measured on a LGR Isotope analyser CCIA-46EP. Pressed graphite was analysed at the Keck Radiocarbon Dating Laboratory, University of California on a NEC 0.5MV 1.5SDH-2 AMS system (Beverly et al. 2010).

At ANSTO, after visual inspection for the presence of any powdery, potentially extraneous calcite deposition, shell surfaces were physically cleaned by abrasion with a Dremel® rotary tool. 20–100 mg of shell was cut from the shell and the surface etched using 0.5M HCl for 3–5 min under sonication at room temperature, removing ~10–50% of the surface (Hua et al. 2001). The Feigl staining test (Friedman 1959) was undertaken on cleaned aragonite shells (OZM111–OZM116) to confirm removal of calcite. Hydrolysis was performed with 85% H_3PO_4 at 60ºC overnight and the resulting CO_2 was collected and purified cryogenically. The purified CO_2 was reduced to graphite using H_2 over an Fe catalyst at 600ºC (Hua et al. 2001) and measured for ^{14}C on the STAR 2MV HVEE Tandetron (AMS) at ANSTO (Fink et al. 2004). ^{14}C measurements were normalised to NBS Oxalic Acid I (HOxI) as primary standard

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and corrected for process blanks using IAEA C-1 marble (Rozanski 1991) and $\delta^{13}C$ isotopic fractionation which was determined on residual graphite targets using an elemental analyser Elementar vario MICRO cube coupled to a Micromass Isoprime IRMS.

The removal of 10–50% of the sample surface by physical cleaning and/or acid etch removed any surficial contaminants. ^{14}C ages are reported without rounding, following the recommendations of Russell et al. (2011). $F^{14}C$ is calculated according to Reimer et al. (2004).

Calculation of ΔR

ΔR_{20} was calculated using Reimer and Reimer's (2017) online *deltar* program (http://calib.org/deltar/), where the collection year of each shell sample (i.e., year of death) was converted to an equivalent global marine modeled ^{14}C age using the Marine20 calibration dataset and then this age was subtracted from the mean of the measured ^{14}C age of the sample. Uncertainty of each ΔR value is the uncertainty of the sample ^{14}C measurement, without including the uncertainty associated with the equivalent marine modeled ^{14}C age. This practice aims to avoid the inclusion of this uncertainty twice in the final calibrated ages (first in the determination of ΔR and second during age calibration using Marine20 and the estimated ΔR value). Reimer and Reimer's (2017) online *deltar* calculation tool only returns values on calendar ages up to AD 1949. For samples collected ≥AD 1950, an equivalent global marine modeled ^{14}C age was derived from the Marine20 calibration curve data and subtracted from the measured ^{14}C age. Note that any updates of the marine calibration curve beyond Marine20 (Heaton et al. 2020) would require recalculation of the ΔR values presented here using the primary data presented in Table S1.

Stable Isotope Analyses

$\delta^{13}C$ and $\delta^{18}O$ values were measured on solid shell samples or CO_2 gas prepared on AMS vacuum lines using a CO_2 isotope analyser (CRDS) (Los Gatos Research model CCIA-46). Phosphoric acid (102%) was added to each ground shell sample (0.42–0.5 mg) to evolve CO_2. Samples were heated (72°C, ≥1 hr) to promote hydrolysis before analysis of the $\delta^{18}O$ and $\delta^{13}C$ values. International Atomic Energy Agency (IAEA) standards NBS-18 (calcite) and NBS-19 (limestone) were used to construct a two-point isotope calibration curve ($\delta^{13}C = -5.014‰$, $\delta^{18}O = -23.2‰$ and $\delta^{13}C = 1.95‰$, $\delta^{18}O = -2.20‰$ respectively) and further evaluated using BDH ($\delta^{13}C = -24.95‰$, $\delta^{18}O = -13.99‰$) and Sigma ($\delta^{13}C = -14.18‰$, $\delta^{18}O = -20.07‰$) synthetic $CaCO_3$ standards (Beinlich et al. 2017). A drift correction was made after every two samples using 1500 ppm CO_2 reference gas. $\delta^{13}C$ and $\delta^{18}O$ values are reported as ‰ V-PDB. Routine precision of 0.3‰ or better is typical, as determined using sample reproducibility of duplicate measurements.

Modeling ΔR Variability around the Australian Coast

To model spatial variability of ΔR values around the coast of Australia, 182 records of ΔR measurements that met the inclusion criteria were processed. Kriging, a Gaussian process regression, was used to interpolate ΔR_{20} values around the Australian coast at unsampled locations, using Python tools from the SciKit GStat package (Mälicke et al. 2021). Different models were fit against the variogram data, with the spherical variogram returning the lowest error chosen (Figure S19). A semi-variogram was produced plotting the semi-variance against lag for the point pairs in the data set. The spherical model was applied to the kriging function and kriging was performed for all pixels with a size of 10 km within a rectangular region

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bounding the Australian mainland and Tasmania. The Australian Equal Albers projection EPSG:3577 was adopted to minimise distortion. Results were masked to a 300 km border around the coast. The outputs resulted in two raster images, one representing the kriged ΔR values, and the other the Error Variance, of which the square root represents the standard error of the interpolation process. To account for both the standard errors of the original ΔR measurements, and those of the interpolation process, a further interpolation of measurement standard errors was performed, creating a measurement standard error surface. The two error surfaces were combined by taking the square root of the sum of their squares to produce an overall standard error surface.

RESULTS AND DISCUSSION

The 292 samples in this study, including 59 previously published values, were live-collected between AD 1841 and AD 1956 (a single legacy sample collected in AD 1956 from Key Island, Tasmania, by Gill 1983, is the only sample in the dataset post-dating AD 1950). The samples derive from 114 unique locations around the Australian coastline, spanning from Mer (Murray Island) in the north (10ºS) to Hobart (lutruwita) in the south (43ºS), and from Shark Bay (Gutharraguda) in the west (113°E) to North Stradbroke Island (Minjerribah) in the east (153°E) (Tables S1–3; Figure 1). Full details are presented in Table S1 and all data points are plotted by laboratory number in Figures S1–9. Specimens from the Veneridae and Arcidae families comprise almost half (47%, n=136) of the dataset (Table 1).

In total, 110 samples did not meet the selection criteria and were excluded from the analysis (Table 2). Fifty-nine percent of ΔR values obtained prior to this study were excluded. Most values in the full dataset (see Table S1) were excluded as they were not bivalves (n=52; Criterion #1); because ^{14}C ages from shells in the same lot failed the chi-squared test (n=21; Criterion #8); or because the period of collection could not be confidently limited to ≤1 year (n=20; Criterion #4). Smaller numbers of samples were rejected where determined $F^{14}C$ values showed that mollusks were live-collected after AD 1950 (n=7; Criterion #3), where samples were not live-collected or where there was uncertainty about live-collection status (n=5; Criterion #2), where the sample was a bivalve but not a suspension feeder (n=3; Criterion #7), where the collection location was unknown or where there was uncertainty about the collection location (n=1; Criterion #5), and where the mollusk species was not identified or where there was uncertainty over species identification (n=1; Criterion #6).

Statistically different ages derived from shells in the same museum lot may have several possible explanations. Culleton et al. (2006) and Jones et al. (2007) have documented significant intrashell ^{14}C variability. These impacts are usually associated with areas of near-shore upwelling. However, other short-term impacts may influence ^{14}C in individual growth bands, such as heightened storm activity resulting in higher rates of mixing of atmospheric carbon into marine waters (Goodfriend and Flessa 1997). This could be a particular problem for older legacy marine reservoir values obtained on whole mollusk valves, potentially averaging different ^{14}C abundances across the growth of the mollusk. The precise provenance of samples could also explain some variability/unexpected values. The level of museum documentation available for most specimens was not specific enough to determine whether collection occurred from enclosed or semi-enclosed water bodies (e.g., estuary or bay versus adjacent open coasts). Specimens living in enclosed water bodies with incomplete exchange with the open ocean could return ΔR values at variance with adjacent well-mixed surface ocean values (e.g., Ulm 2002).

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Table 1 Mollusk families represented in the dataset.

Family	# samples	# species
Veneridae	100	24
Arcidae	36	10
Neritidae	27	3
Donacidae	25	2
Mesodesmatidae	22	3
Glycymerididae	15	1
Pectinidae	15	7
Cardiidae	9	3
Volutidae	5	1
Pteriidae	4	3
Mactridae	3	2
Patellidae	3	1
Potamididae	3	2
Turbinidae	3	2
Cerithiidae	2	1
Cypraeidae	2	2
Cyrenidae	2	1
Limidae	2	1
Muricidae	2	1
Tellinidae	2	2
Buccinidae	1	1
Carditidae	1	1
Chitonidae	1	1
Crassatellidae	1	1
Haliotidae	1	1
Mytilidae	1	1
Phasianellidae	1	1
Pinnidae	1	1
Placunidae	1	1
Tegulidae	1	1
TOTAL	292	82

Table 2 Primary reason samples were excluded from analysis, following the inclusion criteria hierarchy.

#	Criterion	# samples	%
1	Not Bivalvia	52	47.3
8	Ages from Same Lot, Pair, or Valve have ^{14}C ages that are Statistically Different	21	19.1
4	Collection Date Not Confined to ≤1 Year	20	18.2
3	Not Live-Collected ≤AD 1950	7	6.4
2	Not Live-Collected	5	4.5
7	Not Suspension-Feeder	3	2.7
5	Unknown Collection Location	1	0.9
6	Species Not Identified	1	0.9
	TOTAL	110	100

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The remaining 182 accepted ΔR_{20} values range from 68 ± 24 ^{14}C years at Port Hedland (Marapikurrinya) in Western Australia to –337 ± 46 ^{14}C years at Mornington Island (Gununa) in the southern Gulf of Carpentaria in Queensland (see ΔR_{20} Atlas in Figures S10–18). Most sets of local values are internally consistent, reflecting the dominant influence of regional oceanography. Most Australian coasts are adjacent to broad and shallow areas of the continental shelf (<75 m deep), with only a few localities like Cape Range in Western Australia <10 km from the edge of the crustal continental shelf.

Mean ΔR values for major regions in Australia are shown in Figure 2 and Table S4. There are three areas where published coral ^{14}C data are available in the current study time frame (AD 1841–1950) including southeast Queensland, Bass Strait, and southeast Tasmania. For the first region, a mean ΔR value of –166 ± 27 ^{14}C years (see Table S5) was derived from 116 coral data points from Heron Island and Abraham Reef (Druffel and Griffin 1993, 1999), Masthead Island (Wu et al. 2021) and Heron Reef (Hua et al. 2015). This value agrees well with our mean ΔR value of –167 ± 12 ^{14}C years for this region (Figure 2). The mean ΔR value for the Bass Strait based on U-Th dated deep-sea corals (n=9 from AD 1865–1946; Komugabe-Dixson et al. 2016) of –128 ± 33 ^{14}C years (see Table S6) also agrees well with our value for this area of –120 ± 13 ^{14}C years (Figure 2). For southeast Tasmania, there is a coral datum for North Sister at AD 1856 indicating a ΔR value of –117 ± 41 ^{14}C years (Komugabe-Dixson et al. 2016; see Table S6), which overlaps with our value for this region of –111 ± 14 ^{14}C years within 1σ uncertainties (Figure 2). The strong concordance between our shell-based and published coral-derived ΔR values gives us confidence in our mollusk-derived ΔR values.

There is a notable depletion in ΔR values southwards along the eastern Australian coastline coincident with the East Australian Current (EAC) (Figure 2). Although not a major difference (~40 ^{14}C years from –190 years in northern Queensland to –152 years for southern New South Wales), it is significant and somewhat surprising given the well-equilibrated nature of waters of the EAC along the length of the eastern Australian seaboard. In terms of simple oceanographic conditions, we would expect very similar ΔR values along the entire east coast south to southern New South Wales where the Tasman Front breaks off (Wijeratne et al. 2018). It is possible that the enriched ΔR values reported here for areas of northeast Australia impacted by the EAC result from the extended period of atmospheric-ocean surface exchange during transport along the northeast seaboard with limited mixing with older subsurface waters. Enriched ΔR values have been associated with ^{14}C enrichment of ocean waters in shallow marine environments subject to active wave and wind action (Forman and Polyak 1997). In contrast, much of the southeast Australian coastline is impacted by intermittent upwelling when northerly winds and eddies from the EAC bring water from offshore into near-shore waters (CSIRO 2012).

The ΔR values along the west coast of Australia are substantially depleted compared with the values for the east and north coast of Australia (Figure 2). There is a confluence of source waters on the west coast: in the north the Holloway Current is fed by the Indonesian Throughflow and Eastern Gyral Current whereas the South Indian Counter Current feeds the Leeuwin Current on the mid-Western Australian coast and the southwest coast is impacted by the South Indian Counter Current from the west as well as the Leeuwin Current from the north (Wijeratne et al. 2018). The northeast coast area is also subject to upwelling where tidal motions bring deeper nutrient-rich waters to the surface (CSIRO 2012). Although the west coast is fed by different water sources, their average ΔR values are very similar, overlapping with each other within 1σ (see Figure 2).

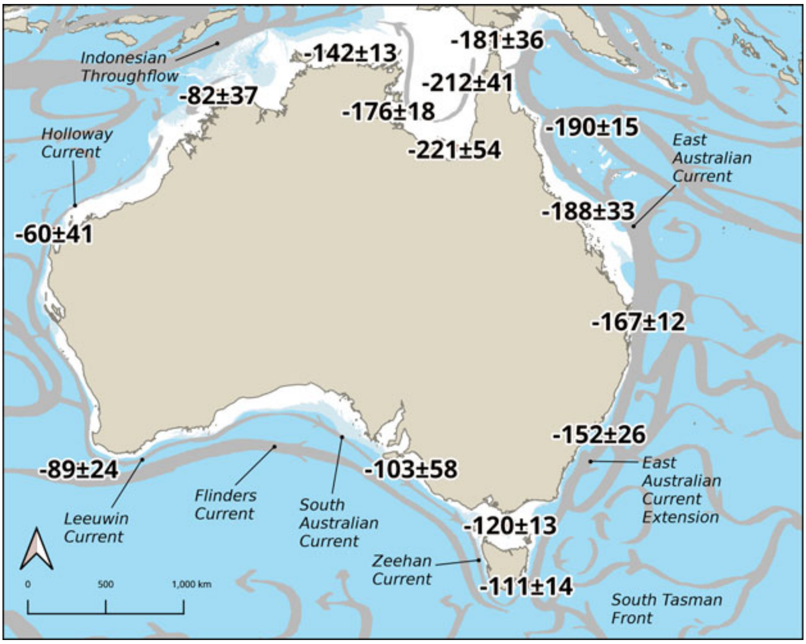

Figure 2 Map of Australia, showing determined pooled ΔR values and major surface ocean currents (after Wijeratne et al. 2018). Individual ΔR values selected for pooling were all accepted values within a contiguous area, with pooled ΔR value groupings separated by large lengths of coastline with no values, but within a dominant marine surface current. Average ΔR values are used here to broadly characterize ΔR variability around the Australian coastline. We recommend researchers use a location specific ΔR value calculated using the Australian ΔR Calculator, available at: https://delta-r-calc.jcu.io/ (see below). See Table S4 for pooled ΔR methods and statistics.

The variability in ΔR noted above for the east and west coasts of Australia is at odds with Ulm's (2006) previous assumption, based on fewer data points, that ΔR values were very similar along both coastlines. Ulm (2006) also suggested that ΔR values in southeast Australia (extending from southern New South Wales, Victoria and to around Tasmania), would be very difficult to predict owing to localised variation in currents and local upwelling. However, the data reported here (albeit with limited data available for Tasmania) indicate that the general magnitude of marine reservoir variability is similar across this region.

These new data suggest general uniformity in the magnitude of ΔR across the Torres Strait and northern Cape York Peninsula, with more negative values in the southern Gulf of Carpentaria. The enriched ΔR values documented here are associated with the shallow waters of Torres Strait and the Gulf of Carpentaria where there are high rates of atmospheric-ocean surface ^{14}C exchange and less mixing with older subsurface waters (cf. Petchey 2009; Ulm et al. 2009). As noted previously, such ^{14}C enrichment of ocean waters may occur in shallow marine environments subject to active wave and wind action (Forman and Polyak 1997), exacerbated in this region by input of monsoon runoff combined with the less open circulation of the Gulf.

The new values broadly spanning most of the Australian coastline provide more confidence in characterisation of regional MRE, with high-resolution values derived from a wider range and larger number of samples, and from a range of geographic contexts. The large error estimates

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associated with previous ΔR values may have masked our appreciation of variability in the MRE around Australia.

Australian ΔR Calculator

The interpolated kriged ΔR surface is shown in Figure 3 and the overall standard error surface in Figure 4 (combining both the standard errors of the original ΔR measurements, and those of the interpolation process). Figure 5 shows modeled predictions at selected locations around the coast of Australia.

ΔR values and the overall standard errors for any location around the Australian coastline can be calculated using the online Australian ΔR Calculator available at: https://delta-r-calc.jcu.io/. ΔR values produced by the calculator can be used with the Marine20 calibration dataset (Heaton et al. 2020) and common radiocarbon calibration programs including OxCal (Bronk Ramsey 1995) and CALIB (Stuiver and Reimer 1993) for age calibration of radiocarbon ages on Australian marine samples.

Future Research Directions

This study aimed to provide a robust understanding of the marine radiocarbon reservoir effects in Australia and improve the accuracy of radiocarbon dating of marine materials. A more complete understanding of ΔR variability would benefit from broadening this study to include more locations and more suspension-feeding mollusk species, particularly a broader range of species than currently represented in ^{14}C age datasets. Species-specific ΔR predictions can be further refined with larger numbers of samples representing robust stratification of species (e.g., Petchey et al. 2012, 2013), to further refine knowledge and application of ΔR variability. Live-collected mollusks held in museums outside Australia and in private collections are important potential additional sampling sources. Additional specimens may be identified in Australian collections as they are progressively electronically databased.

Samples used in this study represent the majority of Australian coastlines, with notable concentrations around population centres where late nineteenth and early twentieth century shell collecting was focussed. Some stretches of coastline are poorly represented in the accepted ΔR dataset, such as the Great Australian Bight between Spencer Gulf and Esperance (ca. 1650 km of coastline), between Esperance and Perth in southwest Australia (ca. 950 km), between Melbourne and Adelaide (ca. 750 km), between the Mitchell Plateau and Darwin (ca. 750 km), and between Perth and Shark Bay in central Western Australia (ca. 700 km). Other locations with documented concentrations of archaeological shell deposits also have no local ΔR values, such as the north coast of New South Wales and the north and west coasts of Tasmania.

The values recommended in this study should only be considered reliable for the recent past, with samples collected in the nineteenth and early twentieth centuries. ΔR fluctuates through time, responding to ^{14}C activity in source waters. Hua et al.'s (2015, 2020) study of corals from the Great Barrier Reef demonstrates large ΔR variations of ca. 490 ^{14}C years between 5500 and 7000 cal BP, while ΔR has been relatively stable in this region over the past ca. 5500 years (see also Komugabe-Dixson et al. 2016; cf. Petchey 2020; Petchey et al. 2023). Further studies of temporal variability are needed to characterize the magnitude and spatial variability of MRE in the past. Even more substantial changes in ΔR are likely during periods of sea-level change associated with glaciation/deglaciation (Heaton et al. 2023). Dating of shell/charcoal paired samples from archaeological sites in Australia has often yielded ambiguous results, largely

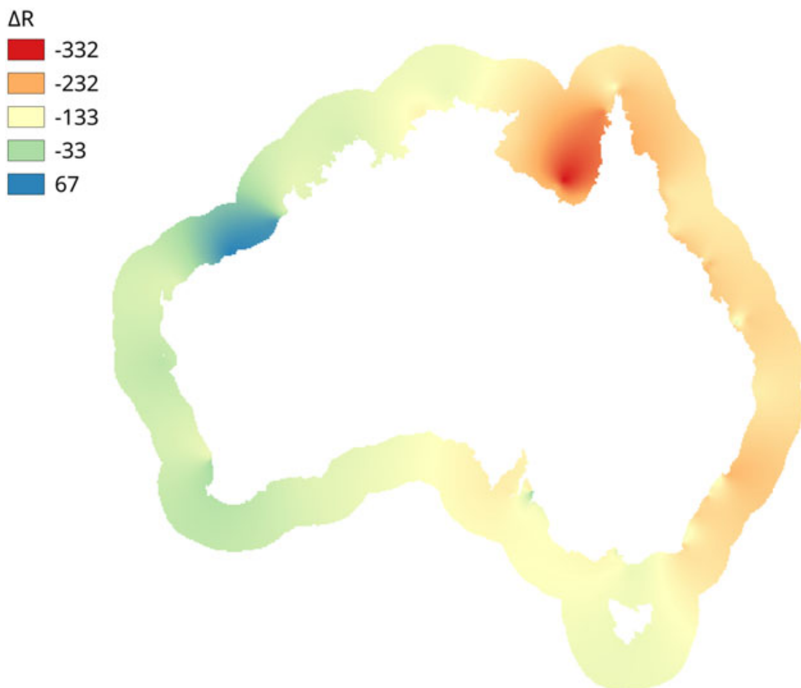

Figure 3 Interpolated kriged ΔR (^{14}C years) surface. Color scale shows range of ΔR values from enriched in red to depleted in dark blue. The kriging layer extends 300 km offshore from the closest mainland point.

attributable to the low degree of confidence in the temporal association of many paired samples (Gillespie and Temple 1977; Hughes and Djohadze 1980; Ulm 2002). One way to address this challenge is to develop local marine calibration curves by expanding regional sample sizes to examine ΔR through time in particular localities (e.g., Petchey and Schmid 2020). Large-scale U-Th dating programs of long-lived and fossil corals are likely to provide some of the most robust records using current techniques. Another way is to ensure that paired shell/charcoal samples are obtained from high-integrity contexts of well-stratified sites employing fine-grained excavation methods and ensuring that the charcoal (or unburnt plant matter) is from short-lived plant parts such as seeds or leaves (Petchey et al. 2012, 2013).

Future studies could also shed further light on short-term variability in ΔR through implementing intrashell dating programs paired with sclerochronological analysis (Culleton et al. 2006). Similarly, stable isotope analysis may help shed light on intraspecies ΔR variability in the same taxa living in different environments.

CONCLUSION

This study represents the largest regional assessment of ΔR variability in the world. Although equatorial, tropical, and subtropical waters exhibit low reservoir ages, this study demonstrates significant variability around the Australian landmass that is closely related to regional hydrological conditions. These data suggest relative geographical uniformity in open ocean

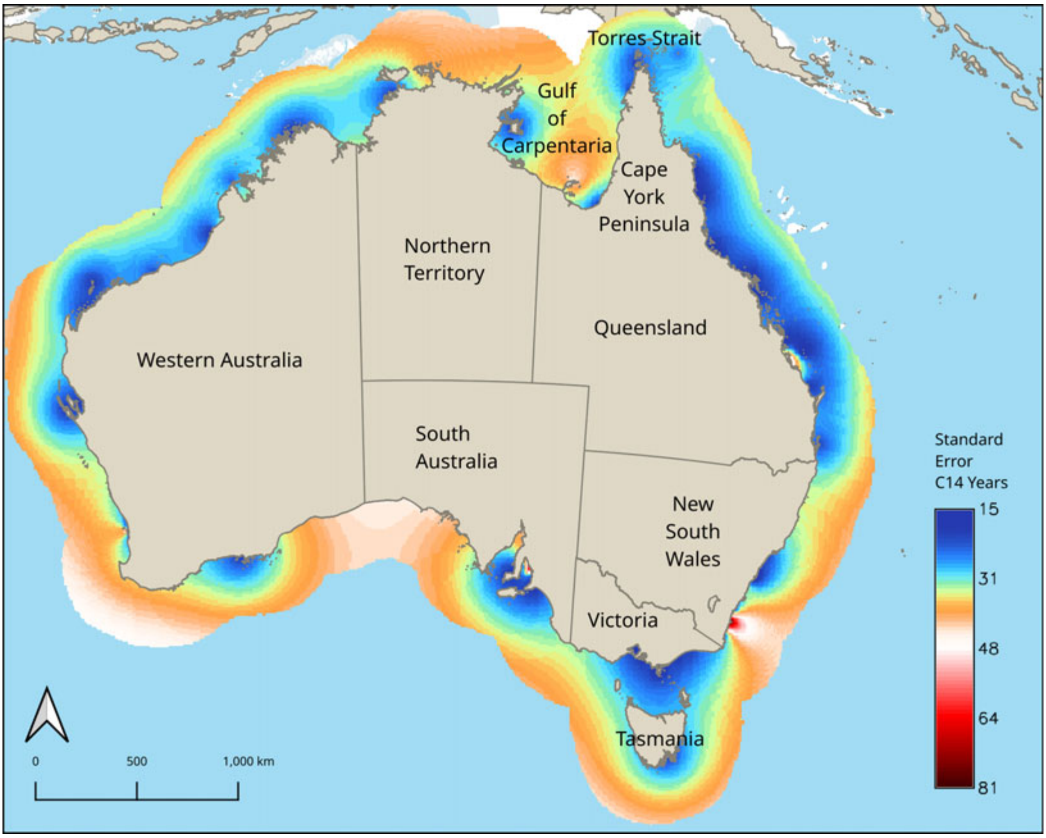

Figure 4 Overall standard error (^{14}C years) surface (combining both the standard errors of the original ΔR measurements, and those of the interpolation process). Color scale shows range of ΔR standard errors from small in blue with large in red. The kriging layer extends 300 km offshore from the closest mainland point.

marine carbon reservoir variability across east, southeast, south, west, northwest, and north Australia, in the late nineteenth and early twentieth centuries. The new values provide more confidence than previous studies, with the more precise values derived from a wider range of species and from a wider range of geographic contexts. Systematic evaluation of legacy MRE studies using strict sampling criteria highlights systemic problems with sample selection and provenance as well as species identification. Legacy marine reservoir data points should only be used with caution and following appropriate evaluation. There is a need to broaden the study to include more locations and marine materials and expand regional sample sizes to examine ΔR through time in particular localities, to address challenges in developing local marine calibration curves. Understanding ΔR through time is significant because accurate marine reservoir corrections for mollusks and other marine taxa are central to debates concerning changes in Aboriginal and Torres Strait Islander societies, cultures, and technologies through the archaeological record, refining sea-level curves and the geomorphological development of coastal environments. The results reported here will support research in many fields by providing a more secure characterisation of local marine reservoir conditions.

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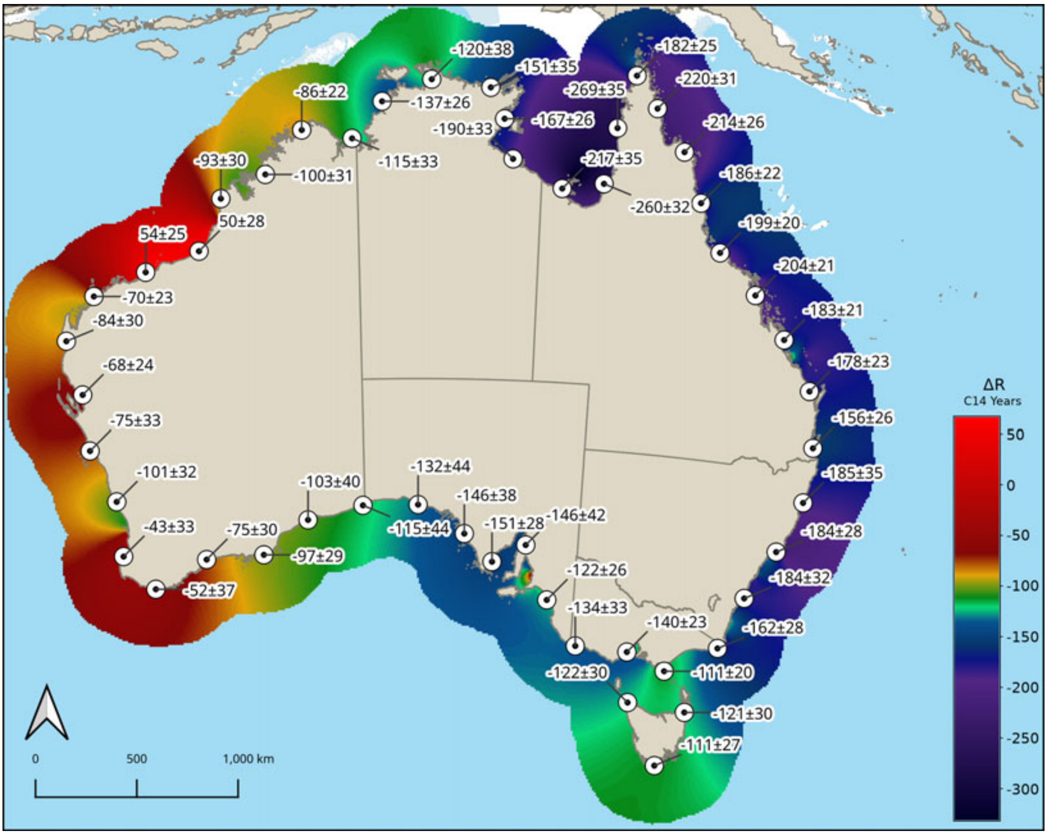

Figure 5 Australian ΔR (^{14}C years) Calculator modeled predictions at selected locations around the coast of Australia. Kriging was performed for all pixels with a size of 10 km. The kriging layer extends 300 km offshore from the closest mainland point. Color scale shows range of ΔR values from enriched in dark blue to depleted in red.

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SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit https://doi.org/10.1017/RDC.2023.95

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