



The transfer of bomb radiocarbon and anthropogenic lead to the deep North Atlantic Ocean observed from a deep sea coral



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ABSTRACT

Deep-ocean, $\Delta^{14}\text{C}$, Pb concentrations, and Pb isotopes were reconstructed from a deep-sea coral *Enallopsammia rostrata* from 1410 m depth off of Bermuda. Our high-resolution time series is created from closely spaced radial cross sections, with samples taken from the center of concentric coral growth bands that we show to be the oldest portion of the section. Prebomb radiocarbon ages from the coral demonstrate that the vertical growth rate of the coral is linear, and the age of the coral is estimated to be 560–630 yr old based on the growth rate. Using this age model to reconstruct $\Delta^{14}\text{C}$ in deep seawater, we first detect bomb radiocarbon at the coral growth site around 1980, and show that $\Delta^{14}\text{C}$ increased from $-80 \pm 1\%$ (average 1930–1979) to a plateau at $-39 \pm 3\%$ (1999–2001). Pb/Ca of the coral ranges between 1.1–4.5 nmol/mol during the 16th and 17th centuries, and Pb isotope ratios ($^{206}\text{Pb}/^{207}\text{Pb} = 1.21$, $^{208}\text{Pb}/^{207}\text{Pb} = 2.495$) in this period agree with pre-anthropogenic values found in the pelagic sediments of the North Atlantic Ocean basin. Coral Pb/Ca is slightly elevated to 6.2 ± 0.9 nmol/mol between the 1740s and the 1850s and then increases to 25.1 ± 0.2 nmol/mol in the 1990s. The increase in coral Pb/Ca is accompanied by a decrease in coral $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{207}\text{Pb}$, indicating that the increase was caused by the infiltration of anthropogenic Pb to the coral growth site. Comparing our data to the surface coral $\Delta^{14}\text{C}$ and Pb records from Bermuda reveals a time scale of tracer transport from the surface ocean to the coral growth site. Some characteristic features, e.g., the bomb-derived $\Delta^{14}\text{C}$ increase, appear in the deep ocean approximately 25 yr later than the surface, but the overall increase of $\Delta^{14}\text{C}$ and Pb in the deep ocean is smaller and slower than the surface, showing the importance of mixing during the transport of these tracers.

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1. Introduction

Atmospheric radiocarbon and lead (Pb) have been perturbed by anthropogenic emissions over the past century. Major sources of the anthropogenic emissions are fossil fuel burning and nuclear weapon testing for radiocarbon, and leaded gasoline combustion and high-temperature industrial activities for Pb. As atmospheric radiocarbon and Pb transfer into the ocean, their oceanic distributions have been altered with time as well. The evolution of radiocarbon and Pb in the surface ocean has been documented by many studies through direct analysis of surface seawater samples (Linick, 1980; Schaule and Patterson, 1983; Broecker et al., 1985;

Boyle et al., 1986) or indirectly by analyzing these tracers in annually banded surface corals (Druffel and Suess, 1983; Druffel, 1989; Shen and Boyle, 1987; Kelly et al., 2009).

Because of their transient characteristics, monitoring the changes of radiocarbon and Pb in the ocean reservoir provides a large-scale geophysical experiment to understand the time scales of surface water ventilation, inter-basin mixing of water, and dispersal of anthropogenic inputs in the ocean interior. Moreover, knowing their oceanic distribution and evolution is necessary to assess the inventory of bomb radiocarbon and anthropogenic Pb in the modern ocean. Compared to the surface ocean, however, less is known about temporal changes of radiocarbon and Pb in deep ocean. Direct measurements of radiocarbon in the deep ocean began in the late 1950s (e.g., Broecker et al., 1960). Ocean-wide-scale surveys like GEOSECS, TTO, WOCE, and GEOTRACES provide deep ocean radiocarbon data with good spatial coverage over discrete time intervals, but high resolution records

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spanning the gaps in time between the surveys do not yet exist. Deep-ocean Pb data are even scarcer than radiocarbon because reliable measurement of Pb in deep seawater became available only recently, e.g., Pb concentrations in the late 1970s and Pb isotopes in the mid-1980s. In the western North Atlantic Ocean, only a few Pb isotope data (e.g., Shen and Boyle, 1988a; Reuer, 2002) existed for the waters below 1000 m prior to a recent US GEOTRACES survey in 2012. The lack of long, continuous data on the changes in deep ocean limits our understanding on the infiltration of bomb radiocarbon and anthropogenic Pb to the ocean interior and associated oceanic processes.

It may be possible to reconstruct changes of radiocarbon and Pb in the deep ocean on decadal to centennial time scales using deep-sea corals. Recently, deep-sea corals have shown to be important archives of past ocean variability. Fossil deep-sea corals have been successfully used to reconstruct changes of the ocean circulation over the last glacial and deglacial times (Adkins et al., 1997; Goldstein et al., 2001; Schroder-Ritzrau et al., 2003), and their potential uses as deep-sea biogeochemical (e.g., nutrient and temperature) proxies are being investigated (Gaetani et al., 2011; Ghosh et al., 2006; Shirai et al., 2005). It was shown that dissolved inorganic carbon in the ambient seawater is the primary source of carbon used for skeletogenesis of various species of calcareous deep-sea corals (Adkins et al., 2002; Frank et al., 2004; Griffin and Druffel, 1989; Roark et al., 2006). The source of Pb in deep-sea coral skeletons is unknown, but given that their surface relatives preserve the record of dissolved Pb in the ambient seawater, deep-sea corals might also be able to generate time series of Pb and Pb isotopes in the deep ocean.

In this study we reconstructed deep-ocean, radiocarbon, Pb, and Pb isotope histories at high resolution using a modern deep-sea coral. A coral specimen *Enallopsammia rostrata* that was collected alive from the deep North Atlantic Ocean was used for the study. *E. rostrata* is regarded as a major structure-forming species and has been found world-wide at depths between ~200 m to ~2000 m (Freiwald et al., 2004). It has been also found to live longer compared to other deep-sea coral species like *Desmophyllum dianthus* (Adkins et al., 2004), which allows us to attain geochemical records of several hundreds of years. One of the challenges in using deep-sea corals as a paleoceanographic tracer is estimating growth rate and age of the corals. As the banding of deep-sea corals is not necessarily controlled by the annual cycle for most species, the age of the corals is often estimated by absolute radiometric methods such as radiocarbon (Druffel et al., 1995; Adkins et al., 2002), ^{210}Pb (Druffel et al., 1990; Adkins et al., 2004), and ^{234}U – ^{230}Th (Cheng et al., 2000; Goldstein et al., 2001). In this study, we found that our coral has grown with a constant vertical growth rate. Using this growth rate, the coral was found to be ~560 yr old based on a vertical length of the coral, and radiocarbon, Pb/Ca, and Pb isotopes were also measured along the vertical growth axis of the coral to generate time-series. Radiocarbon has been previously measured in modern deep-sea corals in several studies (e.g., Roark et al., 2005, 2006), but these studies used bomb radiocarbon signals to constrain ages of the corals (e.g., assigning a certain year to the peak of $\Delta^{14}\text{C}$ in the coral) and did not focus on reconstructing the past ocean ^{14}C variability.

2. Sampling and analytical methods

2.1. Coral sampling

An *Enallopsammia rostrata* specimen (ALV-3701-8) was collected alive in September 2001 with the DSV Alvin from a depth of 1410 m on the north slope of Bermuda (64°W 32°N) (Fig. 1). The water mass at this location ($\sigma_\theta \sim 27.6$) is occupied by the

upper North Atlantic Deep Water (NADW) that is mainly formed by the ventilation of Labrador Sea Water (LSW) (Talley and McCartney, 1982). After collection, exterior contamination on the coral was mechanically cleaned with a Dremel tool and a diamond abrasion wheel, and sediment trapped inside the coral and the septa was removed with deionized water and a toothbrush.

Before cutting the coral, ^{14}C ages were measured in 5 different corallite tips in order to find a branch with the longest history. The result showed that the Tip 1 at the apex of the largest branch is the youngest one (Fig. 2). The excess ^{210}Pb study on the same specimen likewise showed that the corallite tip at the end of the same branch, adjacent to Tip 1, was the most recently precipitated (Adkins et al., 2004). Based on these results, we focused our time series analysis on the branch that terminates at Tip 1 because it is most likely to contain the deep ocean interval recording bomb radiocarbon and anthropogenic Pb infiltration.

From this branch, radial discs were cut perpendicular to the direction of coral growth in 3–5 mm intervals (Fig. 2). From each radial disc, concentric bands were identified under UV light. The banding in the radial sections was found to be asymmetric with the center of the concentric bands located close to the polyp side of the coral, as previously observed in other *E. rostrata* specimens (e.g., Houlbrèque et al., 2010). As a preliminary study, we measured ^{14}C ages from transects of three radial sections, from the center to the edge of each section along the longest possible transect (10–16 mm). In all three sections, the maximum ^{14}C age (the oldest age) was found within 1–4 mm of the section center, and the ^{14}C age decreased linearly with radial distance outside of the center, corresponding to the linear radial growth rate of 20–30 $\mu\text{m yr}^{-1}$ (Fig. 3). Based on this result, samples used for time series construction were cut from the central part of each radial disc (Fig. 2b). A sample mass of 11–44 mg was first cut from each radial disc and used for ^{14}C analysis, and the rest was used for Pb and Pb isotope analysis. Radial discs with younger ages often had limited amount of samples as the branch is thinner. The tip of the branch was analyzed for ^{14}C only, and a single Pb and Pb isotope measurement was made for most of these samples. For older samples (thicker branch), 2–3 replicate Pb and Pb isotope measurements were made with a sample mass of ~30 mg for each measurement.

Previous studies on modern deep-sea corals analyzed tracers along the radial section of the coral's thick base (e.g., Roark et al., 2006), while our time series reconstruction is based on the distribution of ^{14}C and Pb along the coral's vertical growth axis. Our sampling strategy provides two advantages over the previous approach. First, as the coral's vertical growth rate is more than an order of magnitude greater than the radial growth rate, it is easier to obtain samples with higher resolution when sampled along the vertical axis. Second, samples taken along the vertical axis are more likely to include recently precipitated coral skeleton given the growth pattern of *E. rostrata*. In the five *E. rostrata* specimens collected from the Central Pacific, Houlbrèque et al. (2010) found that the outermost parts of the coral bases are ~30 to 140 yr old, although their ages are expected to be near zero if the corals have been continuously grown to the radial direction. On the other hand, the age of an actively growing polyp at the end of a branch was found to be 6 ± 5 yr (U/Th age). Thus, Houlbrèque et al. suggested that *E. rostrata* stops calcifying at the base as the base becomes remote from the polyps (i.e., active areas of calcification), but keeps growing in its upper parts. Their observation also supports the assumption in our age model that the youngest tip of our coral was recently precipitated in the year of coral collection (section 3.1). Such age model cannot be used if the time series reconstruction is made from the radial section of the coral base.

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