



U/Th-dating and post-depositional alteration of a cold seep carbonate chimney from the Campos Basin offshore Brazil

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ABSTRACT

Dating authigenic cold seep carbonates allows determining the timing of active seepage of hydrocarbon-rich fluids at continental margins and ultimately the detection of the geological factors affecting it. U/Th-dating has been selected in this study, the method having been successfully used before. However, incorporation of initial thorium in substantial amounts of detrital material, due to the precipitation of carbonates within the seabed, and post-depositional alteration potentially pose a major problem, violating the basic assumptions of the method. Here we report results from several samples from the Campos Basin off the coast of Brazil. They are unusual in their high $\delta^{13}\text{C}$ values (mean of -10.0% VPDB), high detrital contents and being dominantly dolomite. Elevated $^{230}\text{Th}/^{238}\text{U}$ activity ratios indicate that most of the carbonates lost uranium during diagenesis. Isochrones are obtained from the unaffected samples yielding an age of 140.4 ± 3.1 ka. Vertical growth of the chimney must have been short and rapid since results from the top and bottom parts of the chimney concur within analytical uncertainties. Furthermore, profiles of U/Th radioisotope specific activity, $\delta^{234}\text{U}$ and ^{232}Th specific activity on two quarter slices of a carbonate chimney reveal the occurrence of three distinct diagenetic processes: U loss, U exchange with seawater and Th gain.

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1. Introduction

Cold seeps, where fluids seep out of the ocean floor, have been discovered at both active and passive continental margins, widely distributed around the globe: in the North Sea (Hovland et al., 1987), Mediterranean Sea (Bayon et al., 2009a), Black Sea (Peckmann et al., 2001), Gulf of Mexico (Paul et al., 1984; Roberts and Aharon, 1994), Gulf of Cadiz (Diaz-del Rio et al., 2003), Gulf of Guinea (Rongemaille et al., 2011) and off the coasts of Oregon (Kulm et al., 1986), Japan (Lalou et al., 1992), New Zealand (Orpin, 1997) and Costa Rica (Karaca et al., 2010) among others.

Ecosystems based on consortia of bacteria and archaea that are capable to anaerobically oxidize methane usually develop around these sites (Boetius et al., 2000). As a consequence of the thereby increased carbonate alkalinity, authigenic carbonates precipitate at cold seeps (Aloisi et al., 2002). Since the timing of carbonate

formation through this process critically depends on the availability of hydrocarbon-rich fluids (Luff and Wallmann, 2003; Luff et al., 2004), dating these carbonates provides an excellent archive of active seepage in the past. Determining this timing is of interest, on the one hand considering that methane itself potentially has a big influence on global climate as a strong greenhouse gas, but also to understand the geological factors influencing hydrocarbon seepage. In this regard, U/Th dating proved to be the most fruitful technique, because the incorporation of carbon from geologically old hydrocarbon reservoirs inhibits obtaining valid ^{14}C ages for the authigenic carbonates (Aharon et al., 1997; Bayon et al., 2009a; Feng et al., 2010; Lalou et al., 1992; Peckmann et al., 2001; Teichert et al., 2003; Watanabe et al., 2008). Only in some cases can radiocarbon dating be applied successfully to calcareous shells cemented in the carbonates (Aharon et al., 1997).

A considerable source of uncertainty for U/Th dating is the correction for thorium that is not in-grown by radioactive decay, but is incorporated during carbonate formation. Isochrone techniques are powerful tools to minimize these uncertainties because they assume samples to be composed of two distinct phases. In the case of U/Th dating, one phase is supposed to contain all the initial thorium (detrital fraction) while the other carries only thorium produced by radioactive decay (authigenic fraction). Measurements of several cogenetic

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subsamples consisting of varying proportions of the two components then allow separation of detrital and authigenic fractions. Isotopic values determined for the authigenic fraction are subsequently used to calculate a robust age (Bischoff and Fitzpatrick, 1991; Luo and Ku, 1991).

Isochrone methods rest on three basic assumptions (Allègre, 2008): a) the subsamples must be coeval, b) they must consist of only two different fractions to be separated and c) the sample must have behaved as a closed system since its deposition. In U/Th dating, assumption b) corresponds to the requirement of a homogenous detrital fraction regarding its $^{230}\text{Th}/^{232}\text{Th}$ activity ratio. In fact, more than one source of initial thorium with different $^{230}\text{Th}/^{232}\text{Th}$ activity ratios is allowed, as long as they are distributed evenly among the detritus.

Post-depositional alterations potentially pose a substantial problem to U/Th dating of carbonates associated with cold seeps. The topic has been largely ignored in published data of other study sites. Bayon et al. (2009a) discussed the influence of thorium scavenged from seawater, but only as an additional component of initial thorium, not as a process happening post-depositionally (addressing assumption b), not c)). Recently, Torres et al. (2010) investigated diagenetic alterations of foraminiferal shells in cold seep settings. They concluded, from stable isotope and elemental ratio data, that secondary mineralization happens in foraminifera and affects their isotopic composition. Because of depletion in Sr/Ca ratios, they also suspect that the studied carbonates were altered after their deposition.

Here we present U/Th, XRD and stable isotope results of several authigenic carbonates from the Campos Basin, off the coast of Brazil. $\delta^{234}\text{U}$ and $^{230}\text{Th}/^{238}\text{U}$ activity ratio are used to identify samples displaying post-depositional alterations. Moreover, an analysis of uranium and thorium isotopic compositions, along two cross-sectional profiles of carbonate M1 Chimney, enables the detailed investigation of the diagenetic processes affecting it. Finally, in order to determine robust ages, isochrones are obtained from the samples that do not show any signs of post-depositional alteration.

2. Regional setting

The study area is located in the middle to lower portions of the continental slope of the Campos Basin, offshore Brazil (Fig. 1A), where the hemipelagic drape is thin or absent and deep-seated faults reach the shallow section. The samples found at the shallowest water depths (~1100 m below sea level (mbsl)) occur where the slope steepens and becomes convex due to the shape of the underlying upper Miocene clinoforms (Kowsmann et al., 2002). In this portion of the slope, older sediments are exposed by slide scars which are

mainly controlled by fault geometry. Some of these faults reach underlying hydrocarbon reservoirs (Kowsmann et al., 2002). Towards the base of the slope (~1370 to 1760 mbsl), the carbonates occur on an exposed terrace formed by the collapse of a fault-bounded inter-canyon nose and also along the face of an uplifted fault block (Miller, 2006).

3. Materials and methods

Authigenic carbonate samples were collected by a SCV 3000 Remotely Operated Vehicle (ROV) under contract with PETROBRAS. The chimneys range in length from 24 to 40 cm, with diameters varying from 2 to 16 cm (Fig. 1B). Their shapes vary from cylindrical to conical and sometimes vortex-like, with single narrow internal venting conduits. Bifurcations are present in some of the cylindrical chimneys. Slabs are sub-rounded to blocky with a maximum length of 24 cm. Internally the carbonates exhibit yellowish gray color with an oxidized pale yellowish orange rim and massive fine grained texture. Their surfaces are covered by a dark gray to black iron oxide coating and generally display irregular to abundant vuggy textures. The chimneys were found laying horizontally on the seafloor.

M1 Chimney was examined in detail. It is a 40 cm tall carbonate chimney (Fig. 1B). Three separate samples of M1 Chimney were analyzed: a fragment from 6 cm from the top and two quarter slices from 11 cm and 35 cm from the top. On each quarter slice, several samples were taken creating a profile perpendicular to the growth axis. In contrast, pieces available for U/Th measurements of the other samples consisted of small fragments broken off the exterior of the chimney or slab.

Bulk mineralogy was determined by X-ray diffraction (XRD) on powdered samples using a Rigaku D/MAX-2200/PC diffractometer with a CuK α radiation of the PETROBRAS Research Center (CENPES). True dolomite was identified by characteristic peaks which match the dolomite standard in the ICDD data file (036-0426) such as: 2.89 Å (100% intensity); 2.19 Å (20%); 1.79 Å (10%); 1.80 Å (10%); 2.02 Å (10%); and 2.40 Å (10%). The full width at half maximum (FWHM) of these peaks varies between 0.2 and 0.3 Å, indicating good crystallinity, in spite of the dolomite presenting a microcrystalline texture.

Stable isotope measurements were carried out at CENPES by extracting carbon dioxide from the carbonates, using 100% phosphoric acid, which is analyzed by a Thermo Finnigan MAT 252 mass spectrometer against the VPDB standard with a precision of 0.05‰ for carbon and 0.08‰ for oxygen. Online acidification took 200 s at 70 °C and offline (more appropriate for dolomites) 72 h at 25 °C.

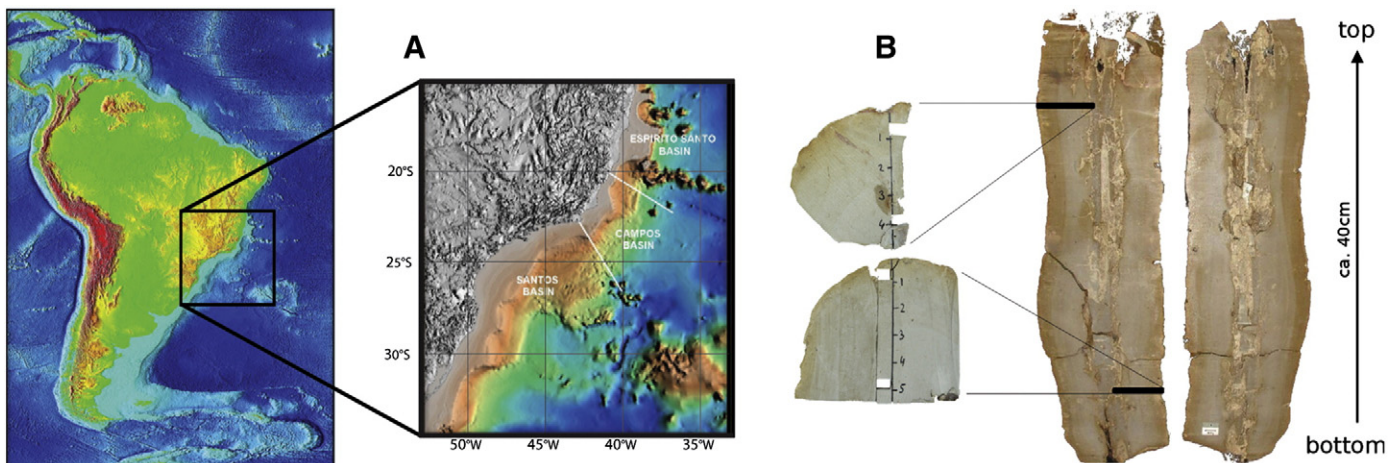


Fig. 1. A) Location map showing the study area: Campos Basin, offshore Brazil. B) Sample M1 is a 40 cm tall carbonate chimney. Cross sections on two quarter slices at 11 cm and 35 cm from the top were analyzed.

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