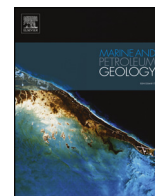




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Research paper

Authigenic carbonate formation influenced by freshwater inputs and methanogenesis in coal-bearing strata offshore Shimokita, Japan (IODP site C0020)

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ABSTRACT

Authigenic carbonate chemistry and mineralogy provide key insights into diagenetic pathways and microbial processes in sedimentary records. We characterize the mineralogy, elemental composition, and isotopic composition of 28 authigenic carbonate nodules recovered from coal-bearing forearc sediments offshore Shimokita Peninsula (Japan) to better understand fluid sources, diagenetic environments, and biogeochemical processes associated with subsiding sediments containing high organic carbon content. The carbonate nodules were collected at Integrated Ocean Drilling Program (IODP) Site C0020 from sediment cores between 1604 and 2460 m below seafloor (mbsf) spanning a transition from terrestrial to marine depositional environments. These carbonates range from < 1 cm to 9 cm thick and most commonly occur in sandstones/sands adjacent to the 2 km-deep early Miocene to late Oligocene lignite beds deposited within terrestrial to estuarine sediments. Almost all samples are composed of relatively pure siderite (74–92% FeCO₃) with two interlayered siderite and low Mg-calcite samples below 2400 mbsf. High δ¹³C (−1.2 to +12.8‰ VPDB) signatures indicate the carbonates were precipitated within the zone of methanogenesis, which was likely coupled to the weathering of silicate minerals, with no evidence of any carbonate produced as a result of anaerobic oxidation of methane. Low δ¹⁸O values (−7.4 to −1.6‰ VPDB) suggest the influence of meteoric waters during carbonate precipitation. High ⁸⁷Sr/⁸⁶Sr values in the carbonates within the main coal-bearing unit relative to the values in pore water suggest mineral precipitation began more shallowly than their current depth and was influenced by meteoric water and/or volcanic material weathering endmembers. Low rare earth element (REE) content and enrichment in heavier REEs suggest carbonate precipitation occurred mainly during early burial. Iron availability for siderite precipitation was likely influenced by microbial reduction of Fe (oxyhydr)oxides. The precipitation of these carbonates likely began within shallow freshwater/brackish aquifers and continued with burial, based on their composition and host strata.

1. Introduction

Precipitation of authigenic carbonates can occur during the early diagenesis of marine and terrestrial sediments, and is an important component in the modern and past global carbon cycle (Higgins et al., 2009; Schrag et al., 2013; Sun and Turchyn, 2014). Authigenic carbonate nodules and concretions formed in sediments during early diagenesis can be “considered ‘fossils’ of diagenetic environments” (Matsumoto and Matsuhisa, 1986), thus providing a snapshot of past

biogeochemical conditions in pore waters. Analyses of major elements (Ca, Fe, Mg, Mn) and isotopes (δ¹⁸O and δ¹³C) of authigenic carbonates can provide indication of past depositional environment (e.g., Veizer, 1983; Mozley, 1989; Mozley and Wersin, 1992; Rodrigues et al., 2015), diagenetic zonation within marine sediments (e.g., Matsumoto, 1983; Rodrigues et al., 2000; Bojanowski and Clarkson, 2012; Teichert et al., 2014), gas hydrate dissociation (e.g., Bohrmann et al., 1998; Matsumoto, 1989; Teichert et al., 2005), and methane seep dynamics (e.g., Greinert et al., 2001; Rietner et al., 2005; Formolo et al., 2004;

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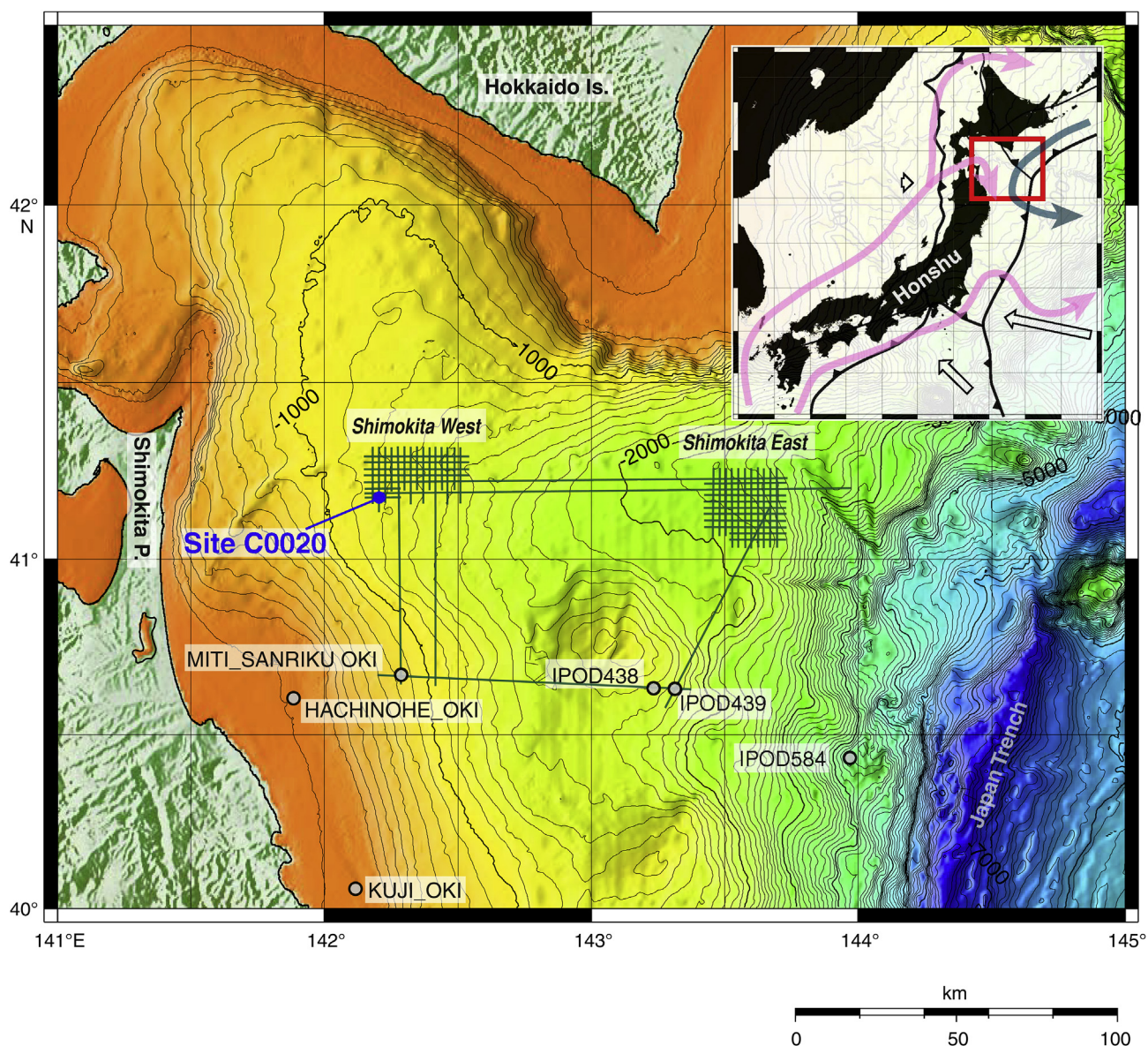
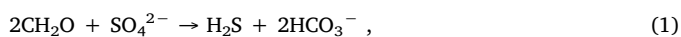


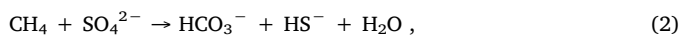
Fig. 1. Location map showing the location of IODP Site C0020 and showing sites drilled during the International Phase of Ocean Drilling (IOPD) during the Deep Sea Drilling Project (DSDP), as well as industry wells. Figure modified from Expedition 337 Scientists (2013).

Gieskes et al., 2005; Sauer et al., 2017). In addition trace metal (Ba, Sr, Mo, U) and rare earth elements (REE) analyses of authigenic carbonates can provide insight into fluid sources and paleo-redox conditions (Feng et al., 2009, 2010, Hu et al., 2014; Ge et al., 2010; Joseph et al., 2012, 2013).

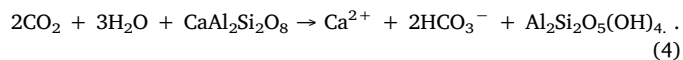
Authigenic carbonate formation occurs when increased alkalinity, often driven by microbially driven bicarbonate formation, favors the precipitation of solid-phase carbonate minerals. Three major biogeochemical processes that can induce increases in pore water bicarbonate/alkalinity include organoclastic sulfate reduction (Berner et al., 1970):



anaerobic oxidation of methane (AOM) (Reeburgh, 1976):



and methanogenesis neutralized by silicate weathering (Aloisi et al., 2004; Wallmann et al., 2008):



If suitable cations (e.g. Ca, Mg, Fe, Mn) are available in the pore-water, the excess alkalinity produced from these reactions can result in authigenic carbonate precipitation (Solomon et al., 2014). Most authigenic carbonates formed in marine or brackish water sedimentary environments vary in composition as their porewater geochemical conditions change through time; most often promoting end member compositions of aragonite, calcite, dolomite, and siderite (e.g. Matsumoto and Matsuhisa, 1986; Rodriguez et al., 2000; Malone et al., 2002; Teichert et al., 2014). Elemental, isotopic, and mineralogical characterizations of authigenic carbonates can help differentiate the processes responsible for their formation and thus help reconstruct the diagenetic history in sedimentary environments through time.

Authigenic carbonates composed of siderite (FeCO_3) are often associated with coal deposits, and can form during multiple stages of burial (e.g., Boardman, 1989; Botz et al., 1986; Curtis et al., 1986; Gould and Smith, 1979; Matsumoto and Iijima, 1981; Uysal et al., 2000). Siderite precipitation is observed in modern marsh and wetland

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