Palaeogeography, Palaeoclimatology, Palaeoecology 455 (2016) 1–15

Contents lists available at ScienceDirect

Palaeogeography, Palaeoclimatology, Palaeoecology

journal homepage: www.elsevier.com/locate/palaeo

A late Miocene methane-seep deposit bearing methane-trapping silica minerals at Joetsu, central Japan

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ARTICLE INFO

Article history:
Received 8 December 2015
Received in revised form 20 April 2016
Accepted 4 May 2016
Available online 6 May 2016

Keywords:
Late Miocene
Methane seep
Aragonite
Silica

ABSTRACT

The modern Japan Sea is characterized by active methane seeps associated with gas hydrates, but their ancient counterparts are not fully understood. This study describes a newly discovered methane-seep carbonate block, the 'Nakanomata Seep Deposit', from the upper Miocene Nodani Formation in Joetsu City, central Japan. The age of this deposit is constrained to 7.5–6.5 Ma based on its fossil diatom assemblage. The deposit contains molluscan fossils typical of methane seeps, including vesicomyid and bathymodiolin bivalves, and provannid gastropods, and it retains an almost entirely aragonitic mineralogy, despite its Miocene age. It is composed of clotted microcrystalline aragonite containing nodules and intraclasts, and is crosscut by vein-like networks of voids and cavities rimmed with acicular aragonite. The δ^{13}C values of the carbonate phases are as low as −41.1‰ and the presence of lipid biomarkers (pentamethylicosane and crocetane) suggests that the deposit originated from the anaerobic oxidation of methane. It is suggested that an initially diffuse methane seepage formed the micritic nodules, followed by a more rapid and intense methane seepage that led to the development of abundant voids in the sediment; finally, the sediment was cemented by microcrystalline aragonite and void-lining acicular aragonite. The seep deposit also contains peculiar globular silica minerals and authigenic quartz. During their precipitation, these globular silica minerals may have trapped methane gas bubbles, and the minerals may be pseudomorphs after silica clathrate. Sufficient increase in pH and supersaturation of silica, which led to the dissolution and subsequent precipitation of these silica minerals, could have resulted from the degassing of carbon dioxide, promoted by an effective supply of methane, and its supersaturation, thus forming gas bubbles.

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1. Introduction

Hydrocarbon seeps (or cold seeps) are known from both active and passive continental margins of the world, such as the northeast, northwest, and southeast Pacific, the Gulf of Mexico, the Mediterranean Sea, the Black Sea, the East Atlantic, New Zealand, and the Japan Sea (e.g., Kennicutt et al., 1985; Kulm et al., 1986; Roberts and Aharon, 1994; Sibuet and Olu, 1998; Peckmann et al., 2001; Han et al., 2004; Teichert et al., 2005; Sahling et al., 2008; Matsumoto et al., 2009; Campbell et al., 2010; Watanabe et al., 2010 and references therein). In the Japan Sea, bacterial mats and active methane seepages are known from the Okushiri Ridge and off Joetsu (Takeuchi et al., 1992; Matsumoto et al., 2005). Methane seeps off Joetsu are of particular interest to scientists because they are characterized by a high methane flux, emanating gas bubbles (or methane plumes), and are associated with gas hydrate formation (Hiruta et al., 2009; Matsumoto et al., 2009). It has been suggested that the accumulation of organic-rich Neogene sediments, combined with high heat flow over the Japan Sea, produced thermogenic methane and that the faults that developed in these Neogene strata acted as migration pathways for the methane into the shallow sediments where gas hydrates form (Matsumoto et al., 2009).

At methane seeps, methane-charged fluids seep out toward the seafloor through conduits such as faults, and methane is oxidized anaerobically in the sediment. This reaction, known as the anaerobic oxidation of methane (AOM), occurs as follows:

$$CH_4 + SO_4^{2-} \rightarrow HS^- + HCO_3^- + H_2O$$

and is performed by microbial consortia of anaerobic methanotrophic archaea, ANME-1, -2, or -3 groups, and sulfate-reducing bacteria (e.g., Reeburgh, 1980; Masuzawa et al., 1992; Boetius et al., 2000; Orphan et al., 2001; Niemann and Elvert, 2008). Methane and sulfide

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http://dx.doi.org/10.1016/j.palaeo.2016.05.002
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ions are converted to energy and nutrients by chemosynthetic bacteria, and sustain unique biological communities (e.g., Sibuet and Olu, 1998; Levin, 2005; Campbell, 2006). Anaerobic oxidation of methane causes an increase in alkalinity and promotes the precipitation of carbonates, which inherit methane-derived carbon and its characteristic ^{13}C-depleted isotopic composition (e.g., Ritger et al., 1987; Kulm and Suess, 1990; Sakai et al., 1992; Stakes et al., 1999). Methane-seep carbonates derived from AOM are composed mainly of aragonite, calcite, and dolomite. They typically precipitate as microcrystalline concretions or form unique, extensive mounds characterized by highly porous fabrics and complex paragenetic sequences of cement and mineral phases (Campbell et al., 2002; Peckmann and Thiel, 2004; Reitner et al., 2005; Teichert et al., 2005). The δ^{13}C values of AOM-derived carbonates are often lower than −30‰ (relative to PDB); specific values depend on fluid composition and the degree of mixing with bicarbonate derived from other sources, which is thought to be related to fluid flow intensity (Joye et al., 2004). In addition, these carbonates preserve lipid biomarkers of AOM consortia such as the isoprenoid hydrocarbons, pentamethylicosane (PMI), and crocetane, along with their unsaturated derivatives, which originate from the cell membranes of ANMEs (Elvert et al., 1999; Blumenberg et al., 2004; Niemann and Elvert, 2008; Blumenberg, 2010).

These signatures of methane-seep carbonates (i.e., low δ^{13}C values and lipid biomarkers of AOM-performing microorganisms) have been recognized in marine sedimentary strata dating back as far as the Carboniferous (e.g., Beauchamp and Savard, 1992; Peckmann et al., 1999, 2002; Peckmann and Thiel, 2004; Majima et al., 2005; Birgel et al., 2006, 2008; Campbell, 2006; Himmler et al., 2008; Jenkins et al., 2008; Tsuboi et al., 2010). Ancient methane-seep carbonates also contain fossils of invertebrates such as vesicomyid bivalves and bathymodiolin mussels (e.g., Peckmann et al., 2002; Campbell, 2006; Amano et al., 2010, 2013a; Kiel, 2010; Amano and Jenkins, 2013), whose extant counterparts include those living at hydrothermal vents and hydrocarbon seeps, and which host chemoautotrophic endosymbiotic bacteria (Fisher, 1990; Taylor and Glover, 2010). One can use the geochemical and petrographic characteristics of ancient methane-seep deposits to infer certain properties of ancient methane-seep environments, for example fluid flow intensity and composition (Peckmann and Thiel, 2004; Peckmann et al., 2009).

However, ancient carbonates are commonly affected by late burial diagenesis, resulting in a change in mineralogical composition and alteration of primary oxygen isotope signatures. For this reason, most ancient seep carbonates are composed of calcite and dolomite, rather than aragonite (e.g., Beauchamp and Savard, 1992; Nyman et al., 2010). Furthermore, where Cenozoic (and a few Mesozoic) seep carbonates do retain their aragonite mineralogy, the aragonite is often only preserved as acicular or fibrous crystal aggregates or botryoids in void spaces or cavities (Terzi et al., 1994; Savard et al., 1996; Peckmann et al., 1999, 2002; Campbell et al., 2008; Amano et al., 2010; Smrzka et al., 2015). Such diagenetic alteration of seep carbonates hampers accurate interpretation of the physical and chemical environment of ancient hydrocarbon seeps.

Ancient seep carbonates are also often characterized by authigenic silica phases that may be present as replacement minerals (e.g., for methane-derived carbonate phases, molluscan shells, worm tubes) or as cement in cavities (Peckmann et al., 2002; Himmler et al., 2008; Kuechler et al., 2012; Amano et al., 2013a; Smrzka et al., 2015 and references therein). The precipitation of these silica phases is considered to postdate early diagenetic AOM-derived carbonate phases. Based on the results of numerical experiments, Smrzka et al. (2015) suggested that the dissolution and precipitation of silica phases are closely related to methane seepage and AOM processes.

Many vesicomyid fossils have been found in outcrops of Neogene strata in the Japan Sea region, although their descendants do not appear to be present around Recent methane seeps in the Japan Sea (Kanno et al., 1989; Kanno et al., 1998; Amano et al., 2001, 2013b; Amano, 2003; Amano and Kanno, 2005; Majima et al., 2005; Amano et al., 2010; Amano and Jenkins, 2011). Based on faunal analysis, and isotopic and petrographic examinations, three of the vesicomyid fossil localities in the Japan Sea region have been confirmed as ancient methane-seep sites: the middle Miocene Bessho Formation in Nagano Prefecture (Tanaka, 1959; Sato et al., 1993; Kanno et al., 1998; Nobuhara, 2010); the uppermost middle Miocene Ogaya Formation in Niigata Prefecture (Ueda et al., 1995; Amano et al., 2010); and the upper Miocene Morai Formation in Hokkaido (Amano, 2003; Ishimura et al., 2005). However, such geochemical studies, including lipid biomarker investigations, have not been carried out for many other fossil localities in this region. Furthermore, it is not known whether compositions of Neogene hydrocarbon seepage fluids in the Japan Sea region were mainly thermogenic in origin, like Recent methane seeps in this area.

This paper describes a newly discovered methane-seep carbonate from the upper Miocene Nodani Formation in Joetsu, Niigata Prefecture. Despite its Miocene age, this carbonate has retained its aragonitic mineralogy in both its microcrystalline matrix and void-lining acicular crystals, and has relatively ^{18}O-rich oxygen isotopic compositions. It also contains peculiar white to translucent, globular silica minerals that probably contain trapped hydrocarbon gases. The processes leading to the formation of these seep carbonate and silica minerals are inferred based on the paleontological, petrographical, mineralogical, isotopic, and biomarker properties of the rock. The discovery of hydrocarbon-trapping silica minerals directly hosted by a methane-seep carbonate, along with inferences about their processes of formation, provides evidence of a close relationship between methane seepage and silica formation.

2. Geological setting

At the eastern margin of the Japan Sea, the Niigata–Shin'etsu sedimentary basin was formed by rapid subsidence caused by rifting during the opening of the Japan Sea in the early to middle Miocene (e.g., Iijima and Tada, 1990; Jolivet and Tamaki, 1992; Takano, 2002). In the Japan Sea, the stress field changed from tensional to compressional between the late Miocene and Pliocene, causing basin inversion (Jolivet and Tamaki, 1992; Sato, 1994; Okamura, 2000). The specific timing of this change in the Niigata–Shin'etsu basin is estimated to be late Miocene (~7–6 Ma; Takeuchi, 1977; Okamura et al., 1995; Takano, 2002). Thick sequences of marine sediment were deposited in this basin throughout the Neogene, and these strata are well exposed in Nagano and Niigata prefectures (Fig. 1A). Many fossil vesicomyid bivalves have been reported from these units (Tanaka, 1959; Seki, 1983; Kanno et al., 1989, 1998; Amano and Kanno, 2005), some of which are from methane-seep deposits (Amano et al., 2010; Nobuhara, 2010).

In the western area of Joetsu City in Niigata Prefecture, which represents the northwestern part of the Niigata–Shin'etsu basin (Fig. 1B), middle Miocene to Pliocene strata are divided, in chronological order, into the Nambayama, Nodani, Kawazume, Nadachi, and Tanihama formations (Akahane and Kato, 1989). Northeast–southwest-trending anticline and syncline axes in this area were formed by E–W compressional tectonics in the late Miocene to Pliocene (Fig. 1B, Kano et al., 1991; Sato, 1994). Fossil vesicomyids in this area have been reported from the middle Miocene Nambayama, upper Miocene Nodani, and Pliocene Kawazume and Nadachi formations (Kanno et al., 1989; Amano and Kanno, 1991, 2005). The Nodani Formation is composed mainly of alternating gray fine-grained sandstones and dark gray siltstones that were deposited as submarine fan turbidites (Endo and Tateishi, 1990). The depth of deposition of this formation is considered to be lower sublittoral to upper bathyal based on molluscan fossil assemblages (Amano, 2002). A tuff bed (the Kanaya Tuff) is intercalated with the middle part of the Nodani Formation and has been dated as late Miocene (7.13 ± 0.42 Ma) in age using fission track dating methods (Muramatsu, 1989).

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