

## **Cold seep carbonate mounds with *Vesicomya* (*Calyptogena*) *kawamurai* (*Bivalvia*: *Vesicomyidae*) in slope-mud fades of the Pliocene forearc basin of the Sagara-Kakegawa area, central Japan**

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Source: Paleontological Research, 7(4) : 313-328

Published By: The Palaeontological Society of Japan

URL: <https://doi.org/10.2517/prpsj.7.313>

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# Cold seep carbonate mounds with *Vesicomya (Calypptogena) kawamurai* (Bivalvia: Vesicomysidae) in slope-mud facies of the Pliocene forearc basin of the Sagara-Kakegawa area, central Japan

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Received December 24, 2002; Revised manuscript accepted June 30, 2003

**Abstract.** Cold-seep carbonate mounds containing *Vesicomya (Calypptogena) kawamurai* paleocommunity occur in massive siltstones of the Pliocene Tamari and Hijikata Formations, both deposited on the upper to middle slope of a forearc basin in the Sagara-Kakegawa area, central Japan. The shell-rich carbonate mounds vary from lenticular (2 m in diameter and 0.5 m in maximum thickness) to barrel-shaped (0.8 m in diameter and 1.5 m in length). One lenticular shelly concretion consisted of densely crowded vesicomysid shells lying parallel to bedding, and overlies a brecciated siltstone with dolomicritic cement. A barrel-shaped concretion contained abundant articulated valves of *V. (C.) kawamurai* in life orientation, and preserves a three-dimensional view of a chemosymbiotic habitat maintained over several generations of these large clams. In addition, the carbonate mounds contain various void spaces (open burrows and dissolved-shell molds) which are fringed by authigenic carbonates (splayed fibrous aragonites and dolosparites). The aragonite burrow-linings in the carbonate mounds and calcite/Mg-calcite micronodules in the siltstone matrix have very  $^{13}\text{C}$ -depleted isotopic signatures ( $\delta^{13}\text{C} = -43.59\text{‰}$  to  $-54.54\text{‰}$  PDB).

The taphonomic, petrographic, and stable isotopic evidence confirms that the vesicomysid paleocommunities formed due to biogenic methane seepage. Brecciation of the siltstone shows an explosive collapse of the sediment fabric, possibly triggered by decomposition of gas hydrates in near-surface sediments. The subsequent biological architecture of burrows and large, dead-shell cavities acted as efficient conduits that facilitated continued seepage. The chemosymbiotic bivalves were able to colonize slope mudstone during the methane-seepage associated with gas hydrate decomposition, and their biological activities altered the muddy substrate to promote the methane seepage that sustained several generations of clams.

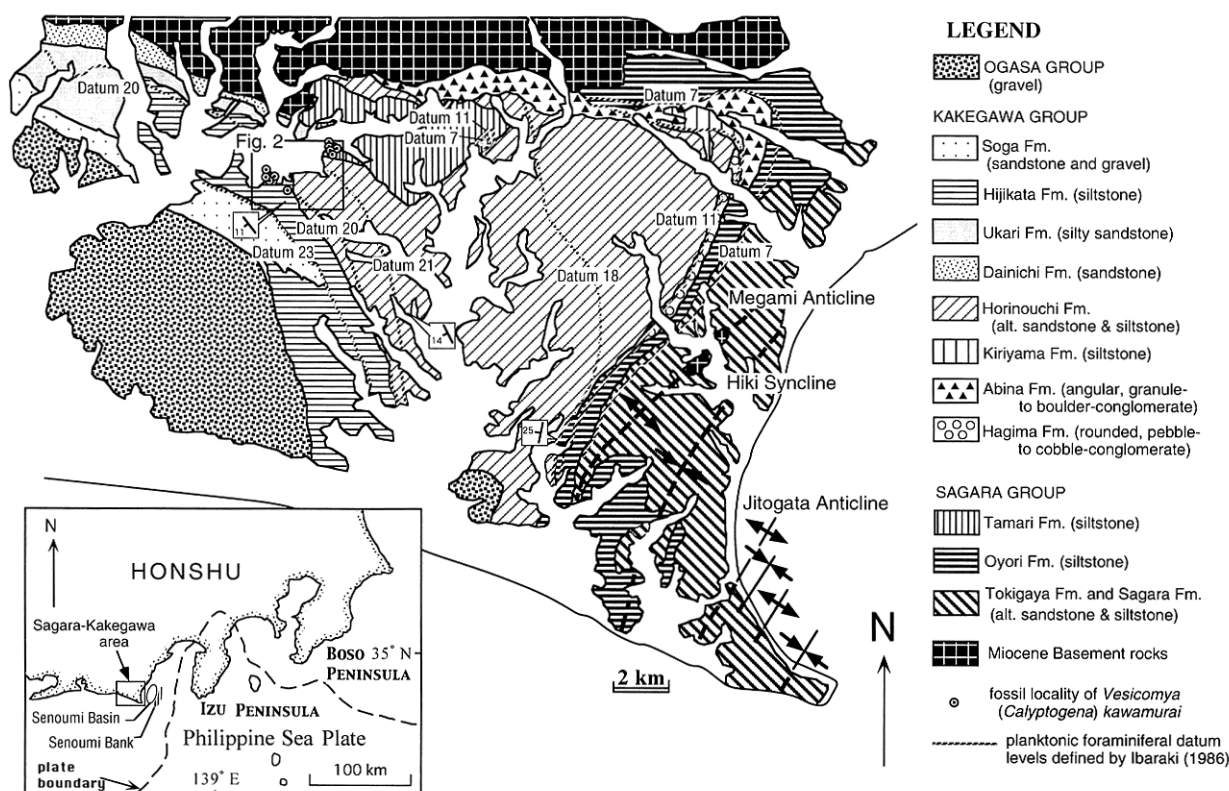
**Key words:** chemosymbiosis, carbonate mounds, Kakegawa, Pliocene, upper to middle slope, vesicomysids

## Introduction

Clams of the vesicomysid subgenus *Calypptogena* are one of the best-known chemosymbiotic taxa, relying on endosymbiotic sulfide-oxidizing bacteria for their nutritional requirements. Since the discovery of dense clusters of the giant white clam, *Vesicomya (Calypptogena) magnifica* Boss and Turner, at the hydrothermal vent sites on the Galapagos Rift in 1977 (Corliss *et al.*, 1979), numerous other examples of vesicomysid communities have been found in various sulfide-rich deep-sea environments, including cold-seepage associated with plate subduction (e.g., Okutani and Egawa, 1985; Suess *et al.*, 1985), oil and gas seepage (Kennicutt *et al.*, 1985), submarine fan sediments rich in organic materials (Mayer *et al.*, 1988), and accumu-

lations of organic debris (e.g., whale carcasses; Smith *et al.*, 1989). The biogeography, ecology, and evolution of chemosymbiotic bivalves have been reviewed by Tunnicliffe *et al.* (1998), Sibuet and Olu (1998), Little (2001), and Kojima (2002).

*Vesicomya (Calypptogena) kawamurai* (Kuroda) was originally described as the type species of the genus *Akebiconcha* by Kuroda (1943) from a depth of about 100 fathoms in Sagami Bay, central Japan. Since then, shell specimens of the species have been recovered from shelf to slope depths (around 200–400 m) off the Pacific coast of central and southwestern Japan by longlines for demersal fish, and by trawlers (Shikama, 1962; Tsuchida, 1986; Numanami *et al.*, 2000). The depth range of *V. (C.) kawamurai* is shallower than for other Japanese *Calyppto-*



**Figure 1.** Schematic geologic map of the Sagara-Kakegawa area, showing *Vesicomya (Calypptogena) kawamurai* fossil localities (modified from figures 1A and 1B of Nobuhara and Takatori (1999), © The Geological Society of Japan). New fossil localities and geological structures (strike and dip, Hiki Syncline and Jitogata Anticline) are added on figure 1B of Nobuhara and Takatori (1999).

*gena* species (Okutani *et al.*, 2000; Fujikura *et al.*, 2000), but other aspects of its ecology are little known because live specimens have not yet been observed.

Fossil *V. (C.) kawamurai* specimens have been found in the Pliocene siltstones in the Sagara-Kakegawa area of central Japan (Majima *et al.*, 1990; Nobuhara and Tanaka, 1993). These siltstones were deposited on the landward slope (ca. 300–1000 m deep) of a forearc basin. Majima *et al.* (1990) illustrated three juvenile valves identified as *Calypptogena* sp. from the upper Pliocene Hijikata Formation, but did not undertake a paleoecological study because of limited outcrop. Nobuhara and Tanaka (1993) reported *Vesicomya (Calypptogena) kawamurai* paleocommunities from the upper Miocene to upper Pliocene Tamari Formation. The abundant, large shells of *V.(C.) kawamurai* are lying parallel to siltstone bedding planes and form shell beds with other chemosymbiotic bivalve species, *i.e.*, *Lucinoma* sp. aff. *L. acutilineata*, *Conchocele bisecta*, and *Acharax johnsoni*. Nobuhara and Tanaka (1993) concluded that this fossil community was chemosymbiotic, supported by a reducing mud bottom at bathyal hypoxic depths, rather than hydrocarbon seepage,

because the fossil shell beds are enclosed by massive, fine siltstone of low permeability without any tectonic or sedimentary structures such as faults and slumping escarpments suggesting past seepage.

In contrast, Majima (1999) suggested that these *V. (C.) kawamurai* paleocommunities depended on the cold seepage triggered by a decomposition of methane hydrate, just like the lucinid and thyasirid bivalve paleocommunities from Pleistocene lower-shelf muddy sediments of the Miura and Boso Peninsulas, central Japan (Shibazaki and Majima 1997; Tate and Majima, 1998). These Pleistocene lucinid and thyasirid paleocommunities are associated with carbonate mounds with extreme  $^{13}\text{C}$  depletion. Carbonates with light  $\delta^{13}\text{C}$  signatures are a characteristic of modern cold-seep sites (e.g., Sakai *et al.*, 1992; Lalou *et al.*, 1992) and are noted as good indicators of ancient cold seeps and their carbon sources (Paull *et al.*, 1992; Beauchamp and Savard, 1992). This paper describes carbonate mounds containing *V. (C.) kawamurai* specimens from the Tamari and Hijikata Formations, and reassesses the paleoecology of the fossil bivalve communities using carbonate petrography and stable isotopic analysis.

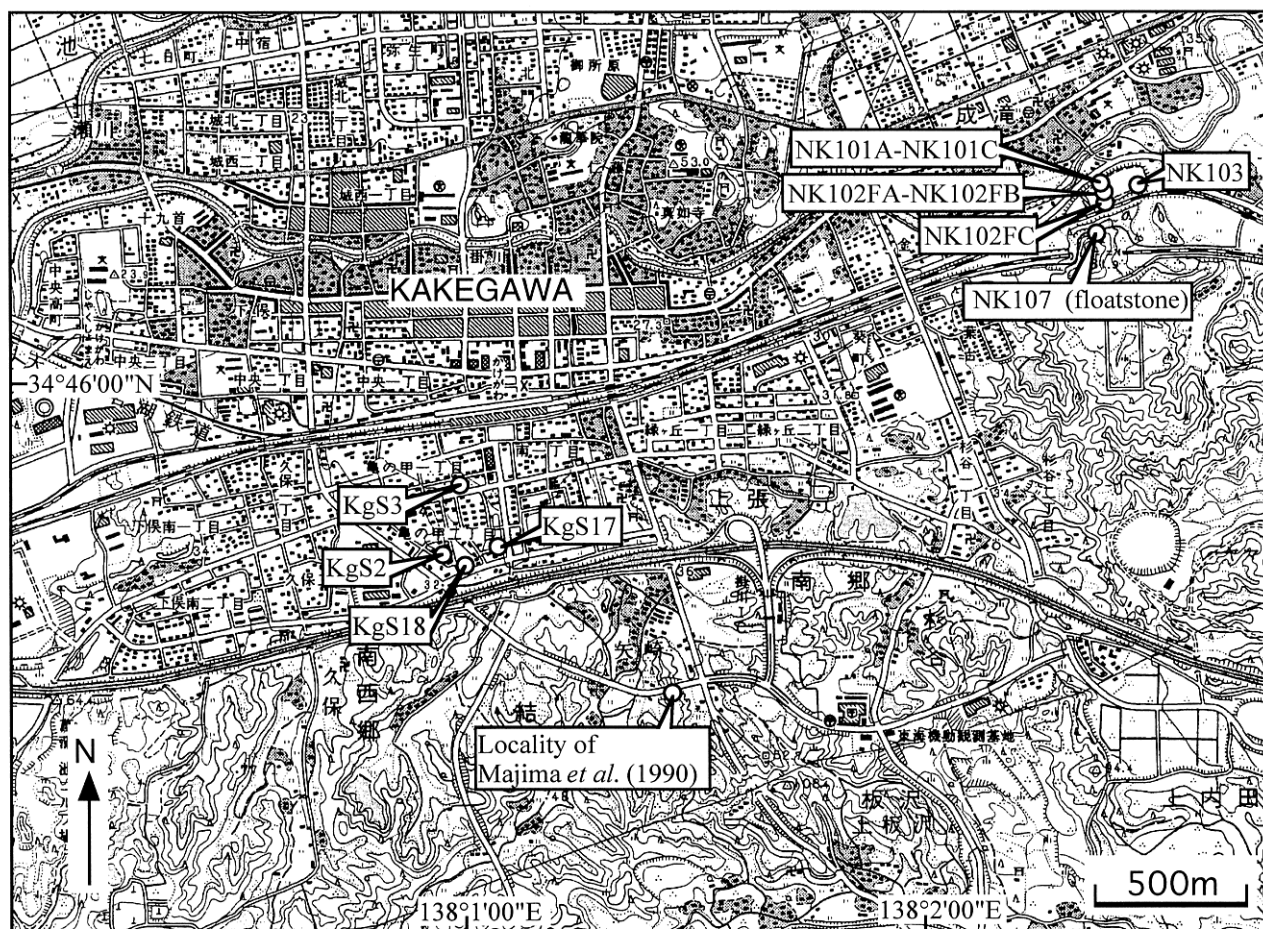


Figure 2. *Vesicomya (Calyplogena) kawamurai* fossil localities on 1:25,000-scale topographic map Quadrangle "Kakegawa".

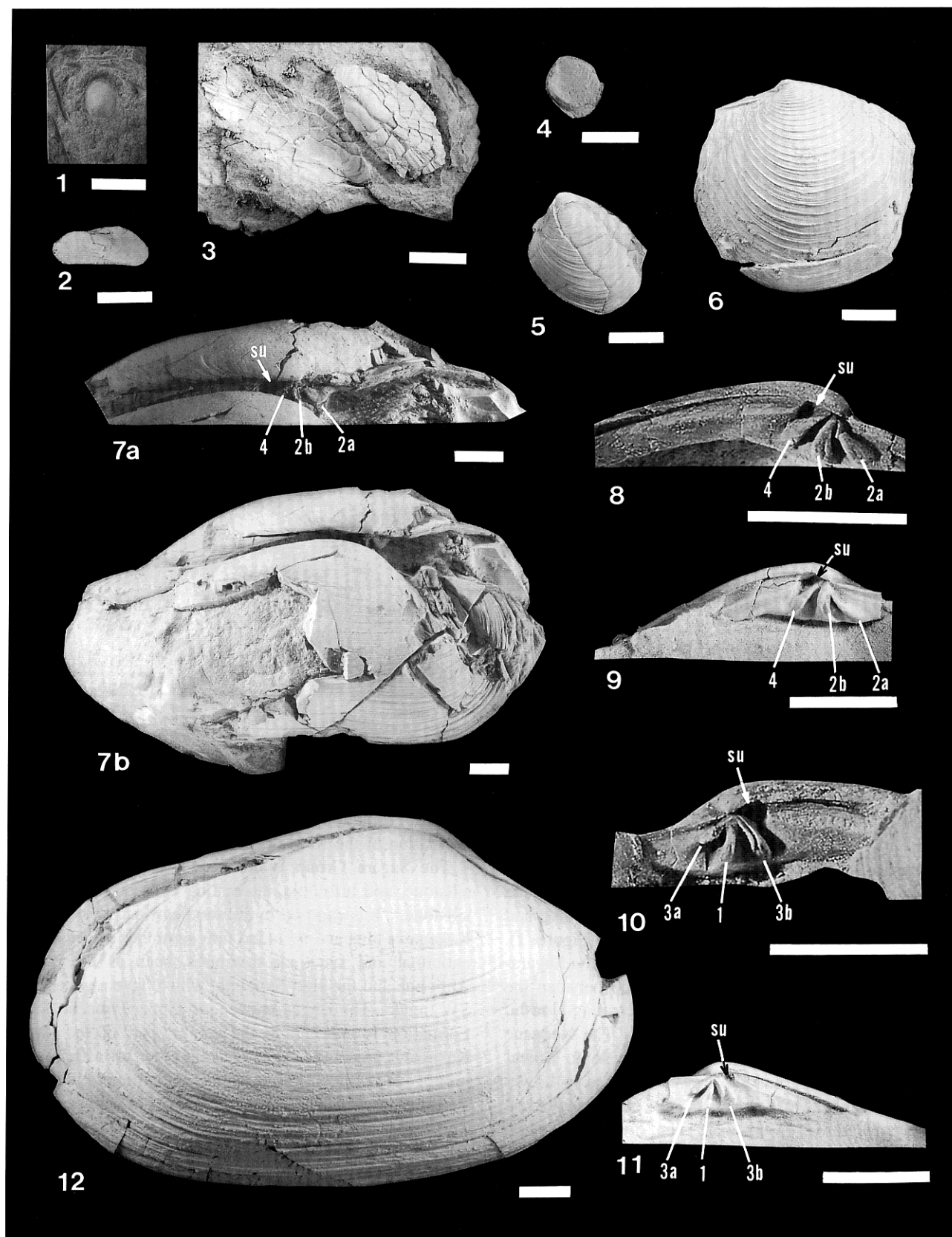
### Tectonic and geologic setting

The Sagara-Kakegawa area is situated along the Pacific margin of central Honshu, Japan. The upper Neogene stratigraphic sequence is widely distributed in the area and consists mainly of marine clastic sediments (Figure 1). These strata were deposited in a forearc basin (the Sagara-Kakegawa Basin) lying in an arc-trench gap between the subducting Philippine Sea Plate and the overriding Honshu Arc (Sugiyama, 1992; Tsukawaki, 1994). The Neogene basin fill is divided into the Middle Miocene to Lower Pliocene Sagara Group and the Lower Pliocene to Lower Pleistocene Kakegawa Group. The geochronological framework for these groups has been established using biostratigraphy (planktonic foraminifera: Saito, 1960; Oda, 1971; Ibaraki, 1986, calcareous nannofossils: Kameo, 1998), magnetostratigraphy (Yoshida and Niitsuma, 1976; Ishida *et al.*, 1980), and fission track dating (Shibata *et al.*, 1984). Specimens of *V. (C.) kawamurai* are found in the

Tamari Formation of the Sagara Group and the Hijikata Formation of the Kakegawa Group (Figures 1, 2).

The Sagara Group is composed mainly of sandstone, siltstone, and turbiditic, alternating beds of sandstone and siltstone. It is exposed in the southeast part of the Sagara-Kakegawa area and is folded into a number of conspicuous anticlinal and synclinal structures, such as the Megami Anticline and the Hiki Syncline, which have a general NE-SW trend. The Upper Miocene to Upper Pliocene Tamari Formation is exposed in the central part of the Sagara-Kakegawa area and is isolated from other formations of the group. It is folded into a monoclinical structure with an E-W trend dipping gently to the south (Tsukawaki, 1994). The Tamari Formation (Makiyama, 1928) is approximately 600 m thick and composed mainly of massive siltstone (Ibaraki, 1986). It is exposed as a partial inlier surrounded by the Horinouchi Formation of the Kakegawa Group. Calcareous nannofossil biostratigraphy (Kameo, 1998) indicates that the upper part of the formation is temporally





equivalent to the lower part of the Horinouchi Formation. *Vesicomya (Calypptogena) kawamurai* occurs in the uppermost part of the Tamari Formation. The horizons are situated between Datum 11 (3.7 Ma) and Datum 18 (3.0 Ma) of Ibaraki's (1986) planktonic foraminiferal biostratigraphy, but the calcareous nannofossil biostratigraphy by Kameo (1998) assigned the horizons to a younger age, between the initiation horizon of the acme of small *Gephyrocapsa* spp. (2.02 Ma) and the last appearance datum of *Discoaster broweri* (2.00 Ma). The upper part of the Tamari Formation was deposited at upper- to middle-bathyal depths according to benthic foraminiferal data by Shikama *et al.* (1972). The fossil localities of Nobuhara and Tanaka (1993) yielded benthic foraminiferans such as *Bolivinita quadrilatera* that live at approximately 1000 m depth (personal communication with Hiroshi Kitazato, JAMSTEC), and mollusks that live at bathyal depths under the influence of cool intermediate water, such as *Neptunea constricta* and *Acharax johnsoni* (Nobuhara, 1992; Nobuhara and Tanaka, 1993).

The Kakegawa Group consists of a lower unit of marginal conglomerates and flysch-type alternating beds of sandstone and siltstone and an upper unit of shelf-to-slope clastic facies (conglomerate, sandstone, and siltstone). The Kakegawa Group crops out only to the west of the Megami Anticline, which corresponds to the outer ridge of the sedimentary basin. This ridge was uplifted by a change in the subducting direction of the Philippine Sea Plate from north to northwest at 3.6 Ma (Tsukawaki, 1994). The Kakegawa Group is folded into a monocline with a NW-SE trend and southwestward dip, but the trend turns gradually to NE-SW in the southeastern part of the area along the outer ridge. The Hijikata Formation (Makiyama, 1963) consists mainly of the massive siltstone, and is about 400 m thick (Tsukawaki, 1994). It conformably overlies the Horinouchi Formation and grades, westward, into the Ukari Formation (outer-shelf silty sandstones). The horizons with *V. (C.) kawamurai* are situated between Datum 20 (1.95 Ma) and Datum 23 (1.60 Ma) of the planktonic foraminiferal biostratigraphy of Ibaraki (1986), and close to

the last appearance datum of *Discoaster broweri* (2.00 Ma) according to the calcareous nannofossil biostratigraphy of Kameo (1989). The paleobathymetry of the fossil localities is approximately 400–500 m (upper bathyal) based on benthic foraminiferal assemblages (Aoshima, 1978). Mollusks living at bathyal depths, under the influence of cool intermediate water, such as *Fusitriton oregonensis*, were also obtained at Loc. KgS3.

### Mode of fossil occurrence and carbonate mounds

*Vesicomya (Calypptogena) kawamurai* occurs in the massive siltstones of the Tamari and Hijikata Formations in three ways: 1) shell beds, in which all the valves lie parallel to the bedding (Nobuhara and Tanaka, 1993), 2) widely dispersed shells in concave-up positions (Majima *et al.*, 1990), 3) densely crowded shells in carbonate mounds. Kanie *et al.* (1992) noted this third mode of occurrence in the Tamari Formation, but did not describe the details of the carbonate lithology. Carbonate mounds yielding vesicomyyid shells were also found in the Hijikata Formation during the construction of a residential area in Kakegawa City, but until now have not been described. Unfortunately none of the Hijikata Formation carbonate mounds remain, and the material described here comes from the following museums and private collections: Natural History Museum and Institute, Chiba, Mizunami Fossil Museum, and the private fossil collections of Yasumitsu Kanie (Yokosuka City), Masaharu Suzuki and Tsumoru Tanabe (Fukuroi City).

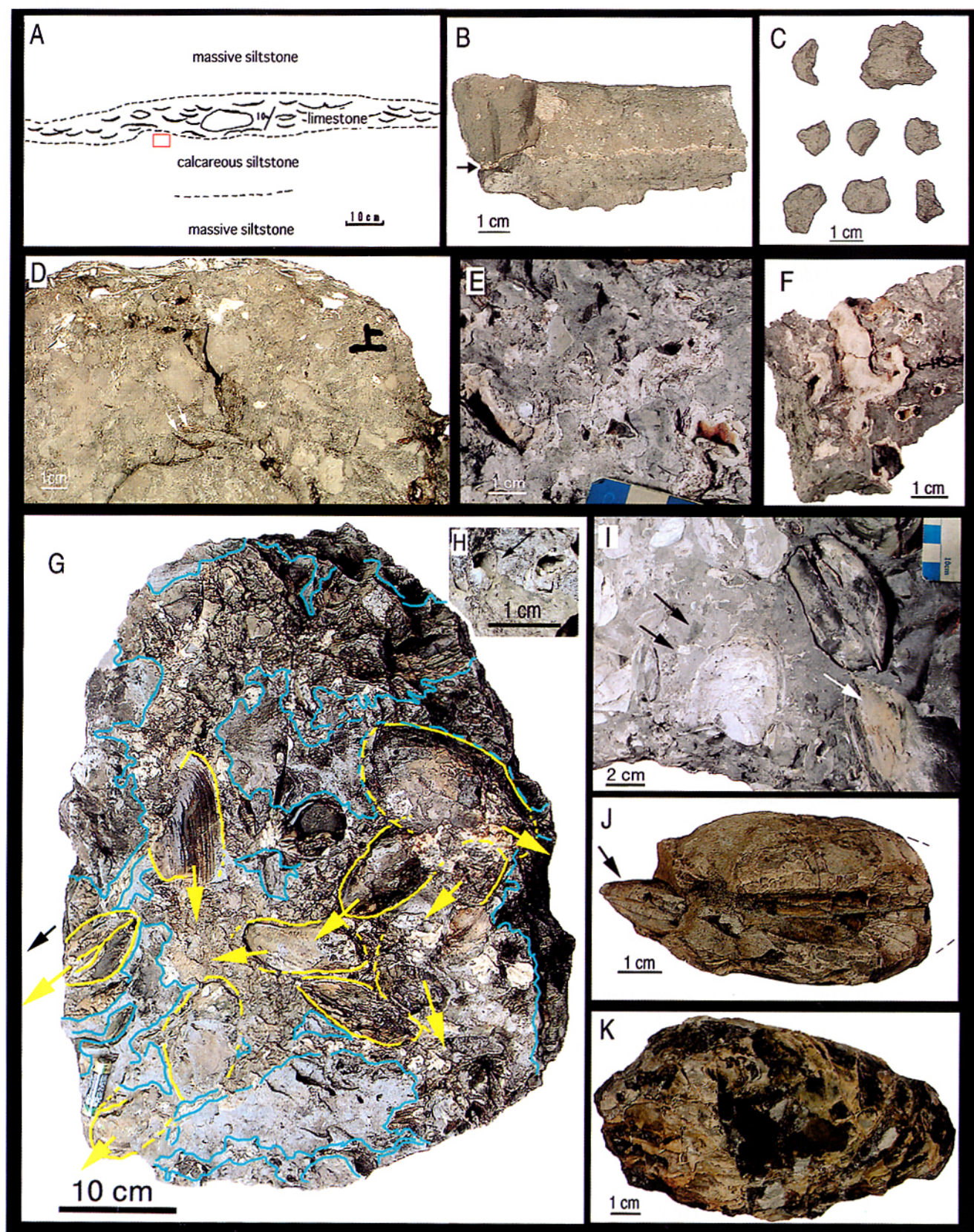
The following is a review of the mode of occurrence previously reported by Majima *et al.* (1990) and Nobuhara and Tanaka (1993), and new descriptions of chemosymbiotic paleocommunities from carbonate mounds in each formation.

### Tamari Formation

Nobuhara and Tanaka (1993) described the *V. (C.) kawamurai* paleocommunity from the uppermost part of the Tamari Formation. They found that large and thick shells

◀ **Figure 3.** *Vesicomya (Calypptogena) kawamurai* paleocommunity from the Tamari and Hijikata Formations. 1. Patellogastropod. Dorsal view of internal mold, SUM-CM-G0001, Loc. NK101 (floatstone), Tamari Formation. 2. "*Bathymodiolus*" sp. External view of left valve, SUM-CM-B0007, Loc. NK102FB, Tamari Formation. 3. *Acharax johnsoni* (Dall). External view of an open, but articulated specimen (right) and its external mold (left), ESN50007, Loc. NK103, Tamari Formation. 4. Lucinoid bivalve. External cast of left valve, SUM-CM-B0008, Loc. KgS18, Hijikata Formation. 5. *Conchocele bisecta* (Conrad). External view of left valve, ESN50006, Loc. NK103, Tamari Formation. 6. *Lucinoma* sp. aff. *L. acutilineata* (Conrad). External view of left valve, ESN 50005, Loc. NK103, Tamari Formation. 7–12. *Vesicomya (Calypptogena) kawamurai* (Kuroda). su. = subumbonal pit. The other symbols of dentition follow Horikoshi (1987). 7a–b. CBM-PS1409, Loc. KgS2, Hijikata Formation. 7a. Dentition of left valve. 7b. Right lateral view of articulated specimen, rather compressed. 8. Dentition of left valve, IGSU-M-002, Hijikata Formation. Dentition symbols are added on figure 3–Ca of Majima *et al.* (1990), © The Geological Society of Japan. 9. Dentition of left valve, ESN50009, Loc. NK10? (floatstone), Tamari Formation. 10. Dentition of right valve, IGSU-M-003, Hijikata Formation. Dentition symbols are added on figure 3–Da of Majima *et al.* (1990), © The Geological Society of Japan. 11. Dentition of right valve, ESN50008, Loc. NK101C, Tamari Formation. 12. Right lateral view of articulated specimen, ESN50012, Loc. NK103, Tamari Formation. Illustrated specimens are stored in the collections of Furukawa Museum of Nagoya University (registration numbers prefixed by ESN), Shizuoka University (IGSU and SUM), and Natural History Museum and Institute, Chiba (CBM). Scale bar = 10 mm.





of *V. (C.) kawamurai* are densely concentrated in several horizons (Locs. NK101A, NK101B, NK101C, NK102FA, NK102FB, NK102FC, NK103; see figures 2 and 3 in Nobuhara and Tanaka, 1993). The shell beds also commonly contain *Lucinoma* sp. aff. *L. acutilineata*, *Conchocele bisecta*, and *Acharax johnsoni* and are rarely associated with "*Bathymodiolus*" sp. and small limpets (Figure 3). Most of the bivalves lie horizontally, with their commissure plane parallel to bedding, and not in life position. However, the shells have not been transported far from their original habitat, because 1) approximately 50% of the specimens are still articulated, 2) the shell beds contain juveniles as well as large adults, and 3) some specimens are found articulated, but in a "butterfly" position.

Nobuhara and Tanaka (1993) could not find any carbonate mounds in their survey, but Kanie *et al.* (1992) briefly mentioned a shelly limestone containing vesicomiid fossils temporarily exposed during road construction just west of Loc. NK102FC. Figure 4A is a redrawn outcrop sketch by Y. Kanie of this exposure. The shelly limestone is a lenticular mound, ca. 2 m in diameter and ca. 0.25 m maximum thickness. Just below this mound is a silty calcareous concretion, ca. 0.25 m thick. A block sample (20 cm × 20 cm × 10 cm) was collected by Y. Kanie across the boundary of the upper shelly carbonate mound and the lower calcareous siltstone (Figures 4A, D). This sample is described below.

The upper shelly carbonate mound is composed of densely packed vesicomiid shells supported by a matrix of calcareous siltstone. Most of the shells are disarticulated and fragmented. The siltstone just below the shelly carbonate mound is brecciated and cemented by carbonate (Figure 4D). The siltstone breccia fragments are irregular in shape, ca. 2 to 10 cm in length, light gray in color, and matrix-supported in calcareous siltstone which is dark gray in color and contains numerous yellow authigenic carbonate spots. The calcareous siltstone has cavities formed by void spaces in the matrix, fractures within the siltstone rub-

ble, and dissolved bivalve shell molds (Figure 4D). The inner walls of these cavities were lined by yellow authigenic carbonate.

Similar carbonate mounds associated with siltstone breccia are probably common in the area around the outcrop described in Nobuhara and Tanaka (1993). Susumu Tomida collected a calcareous concretion floatstone containing abundant vesicomiid shells at Loc. NK107 (about 75 to 100 m south of Loc. NK102FB) (Figure 2). The block has been broken to remove the shells, but the remains deposited in the Mizunami Fossil Museum have the following lithological and faunal characters. The matrix commonly contains brecciated siltstone fragments. The vesicomiid shells are densely packed and supported by a matrix of carbonate-cemented siltstone, and limpets are also frequently contained in the block sample.

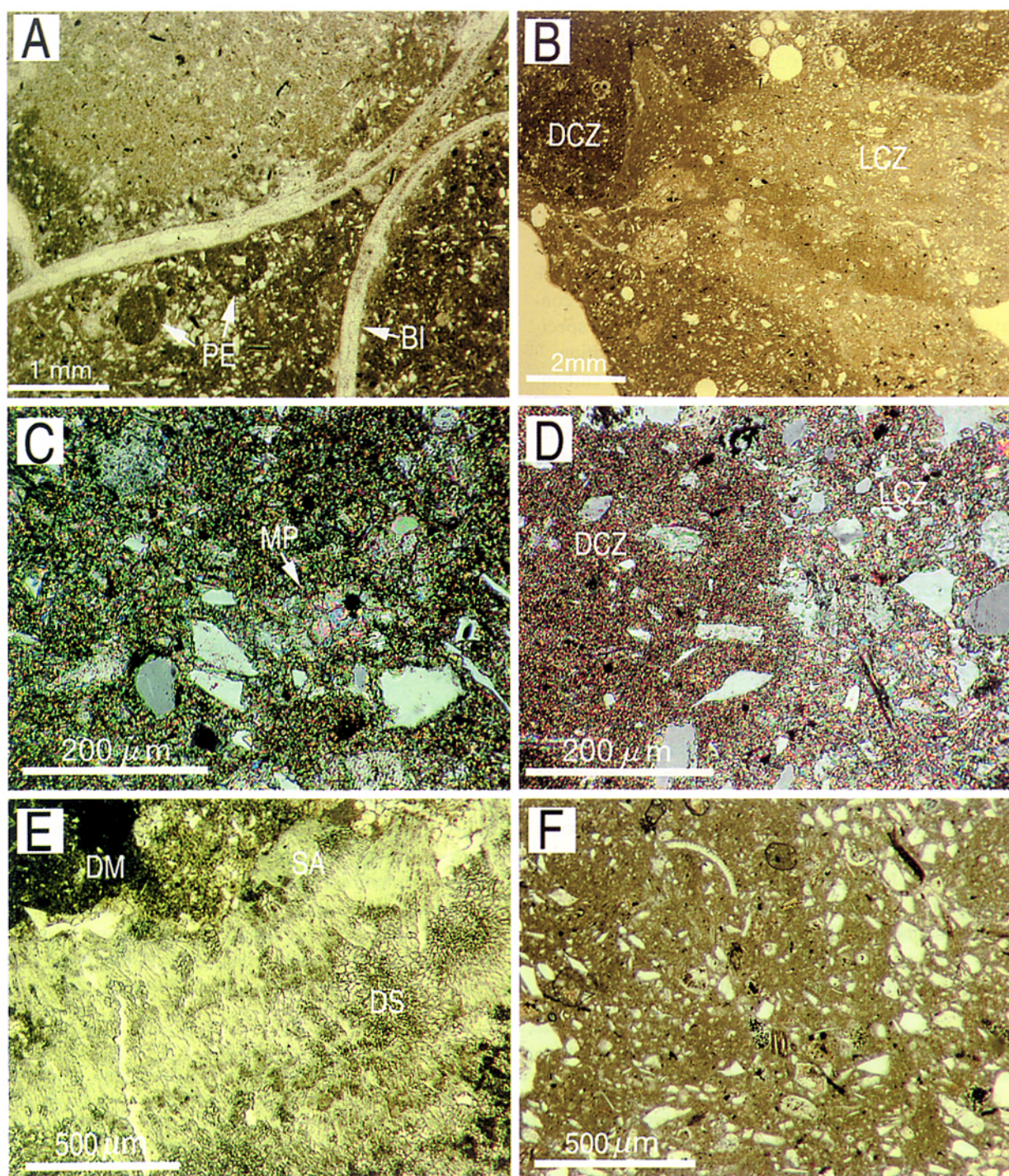
Authigenic carbonates have been recently found in a siltstone block from Loc. NK102FB. These carbonates occur as small, irregularly shaped blebs (Figure 4C) and burrow linings (Figure 4B). The blebs are scattered as micronodules (ca. 1 cm in diameter) in the siltstone containing abundant vesicomiid shells. The burrow-lining carbonate is white, about 2–3 mm thick, and covers the full relief of the cemented burrow-fill sediments (ca. 3 cm in diameter, >10 cm in length) which are weathered out of the outcrop.

### Hijikata Formation

Majima *et al.* (1990) reported the occurrence of *Calypptogena* sp. shells from siltstones of the Hijikata Formation (Figures 3–8, 10). These specimens can be assigned to *Vesicomya (Calypptogena) kawamurai* on the basis of the dentition, which is identical to that of shells from the Tamari Formation (Nobuhara and Tanaka, 1993). All the vesicomiid shells are from juveniles (maximum 65 mm in length), and form a small loose cluster, in which disarticulated and gaping articulated valves are stacked in a concave-up position (see figure 2 in Majima *et al.*, 1990).

← **Figure 4.** Lithofacies of carbonate blocks and mode of fossil occurrence of *Vesicomya (Calypptogena) kawamurai*. **A.** Outcrop appearance of the Tamari Formation carbonate mounds, just west of Loc. NK102FC (a redrawn outcrop sketch by Yasumitsu Kanie). Red rectangle shows the position of block sample shown in Figure 4D. **B.** Aragonite burrow linings (black arrow), Loc. NK102FB, Tamari Formation. **C.** Irregular-shaped blebs of calcite/Mg-calcite, Loc. NK102FB, Tamari Formation. **D.** Brecciated calcareous siltstone just below the shelly limestone shown in the red rectangle in Figure 4A. White arrows show cavities left by dissolved bivalve shells which were lined by yellow authigenic dolomite. **E–I.** Carbonate blocks sampled from barrel-shaped carbonate concretion at Loc. KgS18, Hijikata Formation (collected by Masaharu Suzuki). **E.** Irregular veins and variably sized holes are lined or filled by white and yellow authigenic carbonates. **F.** Aragonite burrow lining. **G.** 3–D view of a chemosynthetic habitat observed in a carbonate block. Blue lines: boundary between network of irregular veins and massive calcareous siltstone. Veins are lined or filled by white and yellow authigenic carbonates. Yellow lines: Outline of articulated vesicomiid valves. Yellow arrows: Shell anterior direction. Note that the articulated vesicomiid valves are concentrated in the veins with their anterior margins pointing in the same direction, which corresponds to the downward direction at the time of deposition (black arrow) shown by geopetal structures. **H.** Geopetal structure in the carbonate block shown by Figure 4G. Lower half of void space in the articulated bivalve was filled by calcareous siltstone (black arrow). **I.** Sharp-edged shell fragments and brecciated siltstone (black arrows) and yellow authigenic dolomite fringing cavities of dissolved shell (white arrow). **J, K.** Molds of dissolved vesicomiid shells encrusted with yellow dolomite. **J.** A young articulated valve (black arrow) nested inside an adult valve, Loc. KgS18, Hijikata Formation (collected by Masaharu Suzuki). **K.** Yellow dolomites have filled in shell cracks caused by sedimentary compaction. Loc. KgS 3, Hijikata Formation (collected by Tsumoru Tanabe).





**Figure 5.** **A.** Impure dolomicrite composed of microcrystalline dolomite, which cements clay- and silt-sized minerals, peloids (PE), and bioclasts including bivalves (BI) and foraminiferans. Loc. KgS18, Hijikata Formation. Transmitted light. **B.** Flow structure in impure dolomite. A light-colored zone (LCZ) flows into a surrounding dark-colored zone (DCZ). Just west of Loc. NK102FC, Tamari Formation. Transmitted light. **C.** A light-colored zone contains micro-patches (MP) of coarse-grained dolomite crystals. Loc. KgS18, Hijikata Formation. Cross nicols. **D.** A light-colored zone (LCZ) contains coarser-grained dolomite microcrystals than a dark-colored zone (DCZ). Just west of Loc. NK102FC, Tamari Formation. Cross nicols. **E.** Splayed fibrous aragonites (SA) fringe inner wall of a burrow space in the dolomicritic matrix (DM), and are covered by anhedronal dolosparite (DS) with corrosion surfaces. Loc. KgS18, Hijikata Formation. Transmitted light. **F.** Impure micrite composed of calcite to Mg-calcite, which cements clay- and silt-sized minerals and bioclasts. Loc. NK102FB, Tamari Formation. Transmitted light.



This mode of occurrence suggests that the shells have not been transported very far from the original habitat.

Calcareous concretions containing vesicomiid shells have also been found at four localities (Locs. KgS2, 3, 17, and 18) situated about 800 m northwest along strike from the locality of Majima *et al.* (1990) (Figure 2). All of the carbonate outcrops are gone now, but the available information suggests that the carbonate mounds occurred in a variety of sizes and shapes, including a barrel-shaped calcareous concretion (1.5 m in length and 0.8 m in diameter) at Loc. KgS18 (personal communication with Masaharu Suzuki and Yoshitsugu Okumura), and irregularly shaped concretions in a siltstone block (about 5 m–10 m) at Loc. KgS2 (personal communication with Hiroshi Kitazato). Materials remaining from these are three boulder-sized blocks from the barrel-shaped concretion at Loc. KgS18, cemented interior molds of articulated vesicomiid shells from Locs. KgS2, 3, and 17, and two siltstone blocks containing vesicomiid shells from Loc. KgS2.

Almost all of the shells in the calcareous concretions have been dissolved (Figures 4I, J, K), but the siltstone around the concretions at Loc. KgS2 contains well preserved specimens (Figure 3–7). An articulated adult shell in a siltstone block from here (Collection Number CBM-PS1409 deposited in Natural History Museum and Institute, Chiba) was referred to *Vesicomya (Calypptogena) kawamurai* on the basis of its dentition (Figure 3–7a).

The boulder-sized block samples and fossils at Loc. KgS18 offer the most significant data necessary to elucidate the lifestyle of the vesicomiid paleocommunities (Figures 4E, F, G, H, I, J). These blocks are composed of carbonate-cemented siltstone and contain abundant vesicomiid shells. The lithology is characterized by a network of irregular veins and variably sized holes, up to 1 cm across, both of which are lined or filled by white and yellow authigenic carbonates (Figures 4E, G). The authigenic carbonates also fringe burrows (Figure 4F) and the cavities of dissolved shells (Figures 4G, I). Most of the vesicomiid shells are articulated, but sharp-edged shell fragments also occur, together with brecciated siltstone (Figure 4I). The articulated vesicomiid valves tend to be concentrated in the veins with their anterior margins pointing in the same directions, which correspond to the downward direction at the time of deposition shown by geopetal structures (Figures 4G, H). Moreover, juvenile articulated valves are frequently nested into the larger older valves (Figure 4J). These indicate that the articulated shells are preserved in life position. Limpets are also frequently contained, and minute lucinoid bivalves formed shell clusters, less than 10 cm in diameter, in the blocks. The fossil data suggest that the calcareous block preserves an *in situ* chemosymbiotic paleocommunity of several bivalve generations.

The interior molds of the articulated vesicomiid valves

from Locs. KgS2, 3, and 17 are formed of carbonate-cemented siltstone and their surfaces are covered with white and yellow authigenic carbonates (Figure 4K), just like the interior molds in the boulder-sized block at Loc. KgS18. The material and outcrop information mentioned above suggest that carbonate mounds at localities KgS2, 3, and 17 may also preserve *in situ* chemosymbiotic paleocommunities.

### Carbonate petrography

Authigenic carbonates in and around the mounds in the Tamari and Hijikata Formations have four modes of occurrence: 1) impure dolomicrite as a matrix of the carbonate mounds, 2) yellow to yellowish brown, pure dolosparite as void-space fills and coatings in the carbonate mounds, 3) white, splayed fibrous aragonite within burrow linings in and around the carbonate mounds, 4) impure micrite composed of calcite to Mg-calcite as small-sized blebs scattered in the silty sediments. The petrography of these carbonate facies were determined by X-ray refractometry, and the details are below.

#### Impure dolomicrite

This forms the matrices of the carbonate mounds and sediment infill of vesicomiid articulated valves. It is also found in the interstitial materials between fragments of the siltstone breccia, just below the vesicomiid bed in the Tamari Formation. The impure dolomicrite is composed mainly of microcrystalline dolomite, which cements clay- and silt-sized minerals, peloids, and bioclasts including bivalves and foraminiferans. In transmitted light, the dolomicrite appears as light- and dark-colored zones (Figures 5A, B). These are irregularly mixed, but sometimes show a flow structure. Figure 5B shows a light-colored zone flowing into a surrounding dark-colored zone. Under crossed nicols, light-colored zones can be seen to contain coarser-grained anhedral crystals than do the dark-colored zones (Figures 5C, D).

#### Yellow to yellowish-brown pure dolosparite

This lines and fills a variety of void spaces, including cracks, pores, and the cavities left by dissolved shells in the carbonate mounds (Figure 5E). The dolosparite is composed of a mosaic of anhedral crystals and is devoid of organic matter and bioclasts. The anhedral crystals grew from the inside walls of the void spaces and covered the impure dolomicrite and splayed fibrous aragonite. The anhedral crystals commonly form small patches, up to 5 mm in diameter, in the matrix of dolomicrite. These patches appear as yellow-colored spots in hand specimens (Figure 4D).



**Table 1.** Stable carbon ( $\delta^{13}\text{C}$ ) and oxygen ( $\delta^{18}\text{O}$ ) isotope analyses of authigenic carbonates. Delta ( $\delta$ ) values are given in per mil (‰), relative to the PDB standard. The experimental error ranges from 0.02 to 0.04‰. Both carbon and oxygen isotopic values for shell material are compiled after Khim *et al.* (1996). Precision is  $\pm 0.1\text{‰}$ .

| Material analyzed                          | Locality                | Sample No. | $\delta^{13}\text{C}$ | $\delta^{18}\text{O}$ |
|--|-------------------------|------------|-----------------------|-----------------------|
| <b>Authigenic Carbonate</b>                |                         |            |                       |                       |
| Irregular blebs<br>(calcite to Mg calcite) | NK102FB<br>(Tamari Fm.) | 1a         | -43.59‰               | +3.61‰                |
|  |                         | 1b         | -45.92‰               | +3.07‰                |
|  |                         | 1c         | -46.03‰               | +3.30‰                |
| Burrow lining<br>(aragonite)               | NK102FB<br>(Tamari Fm.) | 2a         | -44.00‰               | +2.75‰                |
|  |                         | 2b         | -45.91‰               | +2.27‰                |
| Burrow lining<br>(aragonite)               | KgS18<br>(Hijikata Fm.) | 3a         | -48.01‰               | +2.99‰                |
|  |                         | 3b         | -54.06‰               | +2.85‰                |
|  |                         | 3c         | -54.54‰               | +3.06‰                |
|  |                         | 3d         | -54.18‰               | +3.25‰                |
|  |                         | 3e         | -52.60‰               | +2.87‰                |
| Fossile Shell Carbonate*                   | NK103                   |            | -0.86‰ to             | +2.08‰ to             |
| <i>Calyptogenia kawamurai</i>              | (Tamari Fm.)            |            | +1.18‰                | +2.63‰                |

### White splayed fibrous aragonite

This fringes the inner walls of 1 to 3 cm-wide burrows and minute bubble-shaped void spaces (Figures 4E, F). The splayed fibrous aragonite is pure and devoid of organic matter (Figure 5E). The fibrous crystals rarely have the rhombic terminations in the free space of the cavities, as most of them are covered by dolomicrite or anhedral dolosparite with corrosion surfaces.

### Impure micrite (calcite to Mg calcite)

Small, irregularly shaped nodules, around 1 cm in diameter, are in the siltstone that contains abundant vesicomid shells at Loc. NK102FB (Figure 4C). The nodules are composed of homogeneous micritic calcite and Mg-calcite with a high content of clay- and silt-size clastics and bioclasts including bivalves and foraminiferans (Figure 5F).

### Stable isotopic analyses

Ten samples of authigenic carbonates were analyzed for carbon and oxygen isotopes (Table 1). Three samples (samples 1a to 1c) are irregular blebs of calcite/Mg-calcite from Loc. NK102FB in the Tamari Formation. The other seven are aragonite burrow linings from Loc. NK102FB in the Tamari Formation (samples 2a, 2b) and from Loc. KgS18 in the Hijikata Formation (samples 3a to 3e). The samples were cleaned and powder samples were obtained using an art knife under a binocular scope. The isotopic analyses were carried out using a MAT 250 mass spectrometer with a special gas inlet system for ultrasmall gas samples (Wada *et al.*, 1982, 1984). The isotopic values

are reported in delta notation ( $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ ) relative to the PDB standard. The experimental error of the measurements ranges from 0.02 to 0.04‰ for both the oxygen and carbon isotopes.

Oxygen isotope measurements from the aragonite samples show a narrow range of  $\delta^{18}\text{O}$  values (+2.27‰ to +3.25‰). The calcite/Mg-calcite bleb samples have more positive values of +3.07‰ to +3.61‰. The  $\delta^{18}\text{O}$  values from the authigenic aragonite samples are close to the  $\delta^{18}\text{O}$  values (+2.08‰ to +2.63‰) from the aragonite of vesicomid shells at Loc. NK103, a horizon near Loc. NK102FB (Khim *et al.*, 1996). Khim *et al.* (1996) measured a  $\delta^{18}\text{O}$  profile along the axis of maximum shell growth, and suggested that the hydrographic conditions were stable and similar to those at modern bathyal depths (6 to 8 °C, calculated by using the aragonite isotope temperature equation of Horibe and Oba, unpublished). Burrow-fill carbonates from both the Tamari and Hijikata Formations were formed at water temperatures similar to those in which the vesicomid shells were secreted.

Carbon isotope measurements from the authigenic carbonates have very negative  $\delta^{13}\text{C}$  values of -43.59‰ to -54.54‰. The  $^{13}\text{C}/^{12}\text{C}$  ratio can be used as a tracer back to the original carbon source reservoir, because characteristic ranges of  $\delta^{13}\text{C}$  values are known for most carbon sources that are accessible to modern seep organisms. These include total dissolved inorganic carbon in seawater ( $\sim 0\text{‰}$ ), marine organic carbon (particulate and dissolved) ( $\sim -20\text{‰}$  to  $-25\text{‰}$ ), terrestrial plant material ( $\sim -10\text{‰}$  to  $-30\text{‰}$ ), thermogenic methane (+ values to  $\sim -40\text{‰}$ ), and biogenic methane ( $\sim -50\text{‰}$  to  $< -80\text{‰}$ ) (Paull *et al.*, 1985, and references therein). The remarkably depleted  $\delta^{13}\text{C}$  values from

the authigenic carbonates in the Tamari and Hijikata Formations indicate derivation from a biogenic methane source. This contrasts with the  $\delta^{13}\text{C}$  values of +1.0‰ to -1.0‰ obtained by Khim *et al.* (1996) from a vesicomiid shell at Loc. NK103. This indicates that the semiinfaunal vesicomiid shell was probably formed from seawater bicarbonate rather than biogenic methane-derived bicarbonate.

### Discussion

The autochthonous fossils in the carbonate mounds, and the negative  $\delta^{13}\text{C}$  values (-43.59‰ to -54.54‰) of the authigenic carbonates show that the *Vesicomya* (*Calyptragenia*) *kawamurai* paleocommunities recognized in the Pliocene Tamari and Hijikata Formations were associated with biogenic methane seepage. Moreover, the patchy distribution of carbonate mounds suggests localized percolation of methane-enriched pore fluids through the slope muds. Recent methane seeps have not yet been found in the outer shelf to slope, muddy environments around the Japanese Islands (Majima, 1999), in contrast to the numerous living vesicomiid communities on permeable sediments associated with submarine erosion features (fault scarps, canyon walls, fan valleys, and slide escarpments) in trenches and accretionary prisms in the Japanese forearc regions (Fujioka and Taira, 1989). Below I discuss how carbonate mounds in the Tamari and Hijikata Formations were formed within muddy slope sediments.

#### Methane-rich fluid origin

Sibuet and Olu (1998) suggested that the percolation of methane-rich fluids in active margins is mainly caused by 1) upward migration of underground fluids *via* active thrust faults or diapiric mud volcanoes due to plate convergence compressional forces; and 2) submarine erosion in large-scale slides, fault scarps and canyons, which expose methane-rich layers and keep fluid conduits open. These geological processes, however, cannot explain the Pliocene carbonate mounds. Most Recent chemosymbiotic communities are associated with active thrust faults and diapirs and are found in the deformational front of accretionary prisms (Sibuet and Olu, 1998); however, the Tamari and Hijikata Formations were deposited in shallower and more landward settings and lack large-scale thrust faults and diapiric structures which could have provided direct fluid percolation. Slump scars are frequently observed in the slope deposits of the Tamari and Hijikata Formations, but these small-scale marine erosional features were quickly filled by sand and silt and not associated with chemosymbiotic bivalves.

Majima (1999) proposed that fossil chemosymbiotic bivalve communities in outer-shelf to slope mudstone are re-

lated to methane-gas eruption *via* destabilization of methane hydrate. Gas hydrate deposits are quite common in deep-sea marginal basins on both active and passive margins (Kvenvolden *et al.*, 1993). The outer-shelf to slope setting near the coast is generally under a rich supply of organic matter, and this leads to high production of biogenic methane in the subsurface. Majima (1999) figured core samples 3.0 m and 2.35 m deep from under an outcrop of a carbonate mound in the Pleistocene Koshiba Formation, Miura Peninsula, central Japan. The sediment matrix in the cores was entirely replaced by white carbonate (3.0 m core depth), and large, thick shells were fragmented *in situ* and disseminated in the muddy sand (2.35 m core depth). Majima (1999) suggested that these lithological characters were formed by explosive methane-gas eruption.

Similar shell fragmentation and brecciated siltstone also occur in the carbonate mounds of the Tamari and Hijikata Formations. For example, the calcareous siltstone just below the methane-derived limestone of the Tamari Formation is composed of siltstone breccia surrounded by a dolomitic matrix (Figure 4D). The lithology of the calcareous siltstone indicates that a semiconsolidated silt bed was broken up by an explosive collapse of the sediment fabric. If the explosive event was caused by destabilization of methane hydrate, then the outcrop section shown in Figure 4A suggests that the methane hydrate was formed and destabilized near the sediment-water interface.

Typically, gas hydrates are considered to be formed tens to hundreds of meters below the seafloor, depending on methane availability and conditions of temperature and pressure. However, recent surveys have found gas hydrates in near-surface sediments at water depths of between 500 and 800 m in the Gulf of Mexico (MacDonald *et al.*, 1994) and between 600 and 700 m at Hydrate Ridge on the Cascadia subduction zone (Kastner *et al.*, 1998; Suess *et al.*, 1999). Gas hydrates should be stable under the temperature and pressure conditions on the bathyal seafloor (Torres *et al.*, 1998), but the thickness of hydrated sediments is relatively thin at bathymetrical depths shallower than 1000 m and the upper limits of the free-gas stable zone is near the seafloor (Nakamura, 2001). In these situations, hydrate destabilization may be rapid and will be sensitive to tectonic, eustatic or marine environmental changes. The Tamari and Hijikata Formations were deposited at similar bathymetric depths and may have had similar gas-charged mudstone.

The petrography of the carbonate-mound matrices with flow structures (Figure 5B) indicates that unconsolidated mud with a high water content can be a conduit for methane-rich fluid flow. If the origin of the methane is destabilized gas hydrates in near-surface sediments, then fluid flow does not need to be channelized, but is dispersed



diffusively and provides an energy source for vesicomylids. A modern example of vesicomylid beds associated with such diffusive methane-rich fluid flow was reported from muddy talus deposits above a fault scarp on the Nankai accretionary prism (Ashi, 1997).

It is unclear, at present, what caused the hydrate destabilization during deposition of the Pliocene siltstones. In the Cascadia subduction zone, destabilization is thermally triggered by warm fluids ascending along lateral faults (Suess *et al.*, 1999). However, similar faults have not been recognized in the Tamari and Hijikata Formation outcrops. Majima (1999) suggested that methane hydrates can also be destabilized by seawater warming or eustatic sea-level falling. These scenarios cannot explain the Pliocene examples because 1) the chemosymbiotic paleocommunities are associated with cool-water deep-sea mollusks such as *Neptunea constricta* and *Fusitriton oregonensis*, and 2) the fossil localities in the Hijikata Formation formed in the transgressive phase of the stratigraphic sequence framework of Sakai and Masuda (1995).

#### Maintenance of chemosymbiotic community

Vesicomylid species have a symbiotic relationship with chemoautotrophic sulfide-oxidizing bacteria (Reid, 1990; Fisher, 1990). During the deposition of the Tamari and Hijikata Formations, seep-derived methane was probably oxidized by microbial sulfate reducers in the sediments just below the *Vesicomya* (*Calyplogena*) *kawamurai* communities, and this generated sulfide from pore-water sulfate of seawater origin. This process occurs at living vesicomylid communities (e.g., Boulègue *et al.*, 1987; Masuzawa *et al.*, 1992; Wallmann *et al.*, 1997; Boetius *et al.*, 2000).

Tryon and Brown (2001) suggested that the maintenance of elevated levels of sulfide in the shallow subsurface is more essential to vesicomylid communities than is the direct outflow of methane-rich fluid onto the seafloor. They reported that the vesicomylid clams on Hydrate Ridge are not associated with strong outflow sites, but instead with oscillatory and inflow settings. The transport mechanism of seawater sulfate is problematic as diffusion of sulfate into the sediments would be limited to the very surface of the sediments where fluids are being expelled. Tryon and Brown (2001) pointed out that oscillating flow promotes dispersive transport and mixing of seawater sulfate with shallow sources of methane-derived gas hydrates. Moreover, Wallmann *et al.* (1997) suggested that the pumping activity of giant vesicomylids rapidly brings the sulfate into the reaction zone with advecting methane.

The abundant, large *V. (C.) kawamurai* could also cause

a bioirrigation effect by their pumping activity. In addition, dead vesicomylid shells and the burrows of other infaunal animals formed the porous biological architecture in the substrate and would have allowed increased seepage. The cavities formed by large articulated vesicomylid valves may also have acted as drains of subsurface sulfide-rich water. Some young individuals are nested within the dead valves (Figure 4J). Moreover, the abundant, large shells were dissolved out after burial and the cavity spaces remaining also permitted seepage (Figure 4G). The burrows were active seawater pumps into the sediments during the life of the host animals, and often remained open after the hosts died, allowing increased passage of methane-rich fluids (Figures 4E, F).

#### Process of carbonate mound formation

Precipitation of authigenic carbonates associated with cold seepage is caused by the oxidation of methane via microbial sulfate reducers. This generates  $\text{HCO}_3^-$  and can produce porewater oversaturated with respect to calcite ( $\text{CH}_4 + \text{SO}_4^{2-} = \text{HCO}_3^- + \text{HS}^- + \text{H}_2\text{O}$ ) (Wallmann *et al.*, 1997). Petrographic analysis of authigenic carbonates together with megascopic lithological characters, modes of fossil occurrences, and stable isotopic signatures, provide keys to understanding the details of the carbonate mound formation (e.g., Matsumoto, 1990; Beauchamp and Savard, 1992; Takeuchi *et al.*, 2001).

The carbonate mounds of the Pliocene Tamari and Hijikata Formations are characterized by matrices composed of impure dolomicrite. Splayed fibrous aragonite often grew on the inner wall of burrows, but was later covered by dolomicrite or pure dolosparite with corrosion surfaces. Dolomitization can take place in marine and lacustrine environments under two conditions: 1) low dissolved sulfate concentration, or 2) insubstantial contemporaneous silica diagenesis (Baker and Kastner, 1981). Among these conditions, the former is important to dolomitic carbonate formation in cold-seepage sites. Takeuchi *et al.* (2001) found a dolomitic carbonate chimney at a modern cold-seep site on the Kuroshima Knoll, Ryukyu Islands Arc, and suggested that the chimney was formed in an environment where interstitial-water sulfate was diluted by strong inflow of seep water and by microbial sulfate reducers oxidizing the methane.

Using all the data presented above, a process of formation of the carbonate mounds in the Pliocene slope silt sediments is proposed as follows (Figure 6).

1. A gas-hydrate zone in near-surface sediments was destabilized (Figures 6a, b). The semiconsolidated siltstone

← **Figure 6.** Schematic diagrams of the development of Tamari and Hijikata Formation carbonate mounds and the *Vesicomya* (*Calyplogena*) *kawamurai* paleocommunities.

was brecciated by the explosive collapse and fluid venting. Open burrows of unknown hosts acted as efficient conduits for methane-rich fluids, and aerobic methane oxidation in the burrows caused precipitation of splayed fibrous aragonite on their inner walls. Calcite to Mg-calcite micro-nodules also formed during  $^{13}\text{C}$ -depleted seepage close to the sediment surface.

2. Chemosymbiotic clams, *Vesicomya* (*Calyptogena*) *kawamurai*, colonized the seepage site (Figure 6c). Seawater flushing through the clam's body cavity extended into the sediment and brought sulfate into the reaction zone with methane. Microbial sulfate reducers in the sediments just below the vesicomyid community oxidized the methane and generated sulfide, which was in turn utilized by the symbionts in the clams. Below the reaction zone, sulfate was microbially consumed and diluted by the seepage flow. This led to formation of dolomitic concretions (Figure 6d).

3a. If the methane seepage was ephemeral, the vesicomyid community died (Figure 6e), and winnowing of the abundant dead shells led to the formation of a shelly limestone just above the calcareous concretion formed of brecciated siltstone. This is seen in the Tamari Formation outcrop section (Figure 4A).

3b. If the methane source was large enough to sustain long-lasting continued seepage, multiple-generation chemosymbiotic communities would have formed in the small place where background mud accumulation took place (Figures 6f, g). This is seen in the large carbonate blocks containing several generations of the vesicomyid paleocommunities in the Hijikata Formation (Figure 4G). Colonization of chemosymbiotic communities caused the mud substrate to become heterogeneous with macrobenthos burrows, abundant dead shells, and mosaic development of calcareous concretions (Figures 6f, g). Passage of methane-rich fluids was possibly promoted not only by a network of interstitial voids between the concretions but also by burrow cavities and dead clams.

In conclusion, the chemosymbiotic bivalves colonized during the mud-substrate methane seepage that was caused by hydrate decomposition, and the biological architecture of burrows and large dead shells altered the seepage, which continued over several generations of clams. The dolomitic carbonate mounds were formed by giant clam bioirrigation, grew upwards in the process of sediment accumulation, and preserved a three-dimensional view of the chemosymbiotic community habitat in the Pliocene Tamari and Hijikata Formations of the forearc Sagara and Kakegawa area of Japan.

### Acknowledgments

I express my gratitude to Hideki Wada and Kazuhiro Kato (Shizuoka University) for stable isotopic analyses,

and Tomowo Ozawa (Nagoya University) for valuable comments on the lithology and petrography of the carbonate mounds, and Hiroshi Kitazato (JAMSTEC) for providing outcrop information and valuable comments on seepage system and benthic foraminiferal assemblages. I am much indebted to Yasumitsu Kanie (former curator of Yokosuka City Museum) for providing important outcrop information and carbonate samples. My special thanks are also due to the following persons and institutes for providing and loaning samples of carbonate and molluscan fossils: Kiichiro Hachiya, Yoshiaki Mizuno, Tsumoru Tanabe (The Tokai Fossil Society), Masaharu Suzuki (Fukuroi City), Manabu Muto and Takao Sugino (Kakegawa City), Mizunami Fossil Museum, and Natural History Museum and Institute, Chiba. Valuable information on the outcrops was also provided by Kiyotaka Chinzei (Osaka Gakuin University), Yasuo Kondo (Kochi University), Yoshitsugu Okumura (Mizunami Fossil Museum), and Susumu Tomida (Chyukyo Gakuin University). I would like to thank Crispin T. S. Little (Leeds University) for critical reading of the first draft. Helpful comments by James L. Goedert (University of Washington) and an anonymous reviewer are also appreciated. This study was partly supported by a Grant-in-Aid for Scientific Research (A) from the Japan Society for the Promotion of Science (No. 13740298).

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