

Potential of submarine-cave sediments and oxygen isotope composition of cavernicolous micro-bivalve as a late Holocene paleoenvironmental record

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1 **Potential of submarine-cave sediments and oxygen isotope composition of**  
2 **cavernicolous micro-bivalve as a late Holocene paleoenvironmental record**

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21

22

23 **Abstract**

24 A sediment layer (43 cm thick) and surface sediments (5 cm thick) in a  
25 submarine limestone cave (31 m water depth) on the fore-reef slope of Ie Island, off  
26 Okinawa mainland, Japan, were examined by visual, mineralogical and  
27 geochemical means. Oxygen isotope analysis was performed on the cavernicolous  
28 micro-bivalve *Carditella iejimensis* from both cored sediments and surface  
29 sediments, and the water temperature within the cave was recorded for nearly one  
30 year. These data show that: (1) Water temperature within the cave is equal to that at  
31 30 m deep in the open sea; (2) the biotic and non-biotic environments within the  
32 cave have persisted for the past 2,000 years; (3) mud-size carbonate detritus is a  
33 major constituent of the submarine-cave deposit, and may have come mainly from  
34 the suspended carbonate mud produced on the emergent Holocene reef flat over the  
35 past two millennia; (4) the  $\delta^{18}\text{O}$ -derived temperature ( $T_{\delta^{18}\text{O}}$ ) of *C. iejimensis*  
36 suggests that the species grows between April and July; (5) the  $T_{\delta^{18}\text{O}}$  of *C.*  
37 *iejimensis* from cored sediments implies that there were two warmer intervals, at  
38 AD  $340 \pm 40$  and AD  $1000 \pm 40$ , which correspond to the Roman Warm Period and  
39 Medieval Warm Period, respectively. These suggest that submarine-cave sediments  
40 provide unique information for Holocene reef development. In addition, oxygen  
41 isotope records of cavernicolous *C. iejimensis* are a useful tool to reconstruct  
42 century-scale climatic variability for the Okinawa Islands during the Holocene.

43

44 **Key words:** submarine-cave sediments; Okinawa Islands; late Holocene;45 **Palaeoclimatology; micro-bivalve**46 **1. Introduction**

47 In recent years there has been an increased attention of late Holocene climate  
48 changes, including the Medieval Warm Period (800–1300 AD) and the Little Ice Age

49 (1400–1900 AD), because they have significantly influenced human activities and the  
50 geographic distribution of many organisms (e.g., DeMenocal, 2001; Mayewski et al.,  
51 2004). In tropical and subtropical seas, hermatypic corals with annual growth bands  
52 have proved to be excellent archives for the paleoclimate and environment (e.g., Druffel,  
53 1982, Dunbar et al., 1994; Linsley et al., 1994, 2000; Abram et al., 2001). As a result,  
54 several regions have climate records covering the past 400 yr (Gagan et al., 2000;  
55 Hendy et al., 2002).

56 Sheltered submarine caves near coral reefs are among the most valuable sites for  
57 studying cryptic and deep-sea organisms, and a number of workers have examined their  
58 evolutionary significance (Jackson et al., 1971; Jackson and Winston, 1982; Kobluk,  
59 1988, Kase and Hayami, 1992; Reitner and Gautret, 1996; Wörheide, 1998; Kano et al.,  
60 2002). Recently, the oxygen isotopic composition of coralline sponges and serpulid  
61 tubes within submarine caves has been studied as a proxy for palaeotemperature (Böhm  
62 et al., 2000; Antonioli et al., 2001; Haase-Schramm et al., 2003). Although  
63 submarine-cave sediments have been reported from many caves (Hayami and Kase,  
64 1993; Vacelet et al., 1994), there is no published study of cave sediments, except for  
65 infralittoral caves (Di Geronimo et al., 1997). Submarine-cave sediments may constitute  
66 a continuous depositional record, because they are largely sheltered from the physical  
67 mixing and erosion from fluid motion that occurs in the open sea. Therefore, these  
68 deposits may provide a Holocene record of the environmental variations around coral  
69 reefs. Furthermore, the oxygen isotopic composition of cavernicolous bivalves  
70 preserved in the cave deposits may have the potential to reconstruct paleoclimate trends.

71 The main goals in this study are to: (1) describe the lithological features of sediments  
72 in a submarine cave on the slope of a fringing coral reef northeast of Ie Island, off

73 Okinawa Island, Japan, (2) document the stratigraphic distribution of cavernicolous  
74 bivalves in the cored sediments, and (3) evaluate the potential of oxygen isotope  
75 analysis of the cavernicolous bivalve *Carditella iejimensis* as a tool for reconstructing  
76 the late Holocene paleoclimate.

77

## 78 **2. Submarine cave on Ie Island**

79 The northward-flowing, tropical Kuroshio Current allows coral reef formation in  
80 the Ryukyu Islands of southwestern Japan (Fig. 1), where a large number of  
81 submarine limestone caves exist (Hayami and Kase, 1993). We examined sediment  
82 samples from a submarine cave named Daidokutsu (“large cave”) on the  
83 northeastern coast of Ie Island (Fig. 1). The island is approximately 23 km<sup>2</sup> in area  
84 and covered by the Quaternary Ryukyu Limestone, with a fringing reef close to the  
85 Pleistocene limestone sea cliffs, which have been undercut at mean high-water level.  
86 The surface of the fringing reef is mostly flat and exposed at ebb tide (mean tidal  
87 range is 1.8 m). MacNeil (1950) inferred that the reef flat at Ie Island has been  
88 planed down from a reef that in comparatively recent times was at least 1.8 m  
89 higher. The sea level at Okinawa Island between 3,500 and 1,700 yr BP was stable  
90 and also was the highest of the Holocene (Koba et al., 1982), at less than one meter  
91 above the present sea level. This would have been caused by erosion and solution of  
92 the original reef after a subsequent fall of sea level after ca. 1,700 yr BP. The  
93 lowering of sea level is due to uplift with the rate of 0.7 mm per year (Koba et al.,  
94 1982).

95 The reef crest is about 200 m from the shoreline, and 100 m from Daidokutsu  
96 cave. The cave’s entrance lies about 20 m deep on the fore-reef slope, and is one  
97 meter high and two meters wide (Fig. 2). The cave deepens abruptly inward and  
98 there are flat, wide caverns in its middle and innermost parts (Tabuki and Hanai,

99 1999); the maximum depth of the cave is 31 m. Traces of biogenic activity are not  
100 evident in the cave's bottom sediments. Based on the sea-level curve of Fairbanks  
101 (1989), Bard et al. (1996) and Toscano and Macintyre (2003), the cave was  
102 submerged at ca 9,000 yr BP, and even the entrance might have been completely  
103 submerged at about 8,000 yr BP.

104

### 105 **3. Methods**

106

#### 107 *3.1. Field work*

108

109 We measured water temperature hourly in the innermost part of Daidokutsu cave,  
110 with a Nichiyu Giken Kogyo NWT-SN self-registering thermometer, from 26 July  
111 2003 to 6 July 2004. Cave sediment was collected by hand with a coring tube 5 cm  
112 in diameter, and a 43-cm core was recovered without reaching basement (Fig. 2). In  
113 addition, we collected surface sediments (5 cm thick) near the coring site in order to  
114 obtain abundant individuals of the cavernicolous bivalve *Carditella iejimensis*.

115

#### 116 *3.2. Sediment analysis*

117

118 The sediment core was split and described, and X-ray radiographs were taken of  
119 slab samples (6 cm wide X 20 cm long X 1 cm thick) from the split core. Thirty-six  
120 1.2-cm-thick samples were taken along the core to measure grain size, carbonate  
121 content and grain composition. Grain sizes were determined using standard sieves  
122 (class interval 1 $\phi$ ). The carbonate content of both sand-size (1 g) and mud-size (200  
123 mg) particles were determined by gentle removal in 10% acetic acid. To analyze

124 grain compositions of sand- and mud-size particles, we prepared two smear slides  
125 from one samples. Sand-size particles were examined on the smear slide by  
126 point-counting (200 counts per slide). For disarticulated shells of bivalve and  
127 ostracods, a separate valve was counted as one individual. We used the Comparison  
128 Chart for Visual Percentage Estimation of Terry and Chilingar (1955) for a  
129 semi-quantitative visual assessment of mud-size components, except for coccoliths,  
130 with the percentages corrected to compensate for the degree of grain dispersal on  
131 the smear slides. Since the coccoliths are very small and masked by the coarser  
132 grains, their evaluation is difficult using a semi-quantitative visual assessment. In  
133 this study, we examined whether or not the sediment samples yielded coccoliths.  
134 We analyzed thirteen samples for major minerals. Their mineral composition was  
135 analyzed by powder X-ray diffractometry (XRD) using a Rigaku RINT2500V and  
136 JDX3536.

137 Oxygen and carbon isotope analyses were performed on nine samples of  
138 mud-size carbonate, since grain-size analysis showed that mud-size grains  
139 predominant in the submarine-cave deposits (see below). Powdered samples were  
140 analyzed on a Finnigan MAT 251 mass spectrometer at Hokkaido University, Japan.  
141 Isotopic values are expressed relative to the isotopic ratio of carbon dioxide gas  
142 derived from the Pee Dee Belemnite (PDB) in conventional delta notation using the  
143 NBS-20 standard. Analytical precision for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values is 0.08‰ and  
144 0.05‰, respectively, based on multiple analysis of a laboratory standard.

145

### 146 *3.3. Analysis of micro-bivalves*

147

148 All micro-bivalves were picked and counted from the >0.5 mm fraction of  
149 thirty-six 1.2-cm-thick samples. Taxonomic identifications are based on Hayami  
150 and Kase (1993). For disarticulated shells, a separate valve was counted as one  
151 individual. Based on sediment density (here set to 1.25 g/cm<sup>3</sup> of grain density for  
152 all samples) and sedimentation rate, the accumulation rate of the dominant species  
153 is expressed as the number of specimens per cm<sup>2</sup>/yr.

154

#### 155 3.4. Analysis of palaeotemperature

156

157 As noted below, the micro-bivalvia *Carditella iejimensis* is suitable for  
158 reconstructing palaeotemperature using oxygen isotope analysis. There are no  
159 published studies of the oxygen isotope composition of *C. iejimensis*, so we  
160 analyzed 30 individuals (articulated and single valves) from surface sediments (5  
161 cm thick). The delicate prodissoconch of all individuals is present, although they  
162 lack soft parts. In addition, we recovered 13 individuals of *C. iejimensis* from the  
163 core.

164 Oxygen isotope data from bivalve mollusc shells are valid for climate  
165 reconstruction, because they reflect both surface temperature and the  $\delta^{18}\text{O}$  of  
166 ambient water. Therefore many workers have analyzed a number of shallow-water  
167 species (e.g., Chinzei, et al., 1987; Schöne et al., 2003, 2004, 2005; Mueller-Lupp  
168 et al., 2004; Watanabe et al., 2004; Jones et al., 2005). These studies estimated  
169 seasonal temperature variation from intra-annual oxygen isotope values. Such high  
170 temporal resolution requires bivalve specimens that are several centimeters in size.  
171 In contrast, the adult shell size of *C. iejimensis* is less than 4 mm, so that we used

172 the whole shell for oxygen isotope analysis. X-ray diffraction analyses show that *C.*  
173 *iejimensis* consists of 100% aragonite. Samples were not pretreated by roasting.  
174 Isotopic determinations were made at CEA/CNRS, France, and Hokkaido  
175 University. Isotopic values are expressed relative to the isotopic ratio of carbon  
176 dioxide gas derived from the PDB in a conventional delta notation using the  
177 NBS-20 standard, with precision better than 0.05‰.

178

### 179 *3.5. Radiocarbon dating*

180 The radiocarbon ages of eight well-preserved mollusc shells were conducted by Beta  
181 Analytic Inc., using accelerator mass spectrometry. Three samples were mixtures of  
182 several species, in order to provide enough volume for dating (Table 1). In addition, the  
183 radiocarbon ages of three carbonate-mud samples containing dated molluscs were  
184 obtained from the same method. We submitted 0.5 g of carbonate mud to each date.  
185 Calibrated age ranges were calculated according to Method A of Stuiver et al. (1998),  
186 after applying a local correction for the northwestern Pacific of 355 years ( $\Delta R = 35 \pm$   
187 25) (Hideshima et al., 2001).

188

## 189 **4. Results**

190

### 191 *4.1. Modern conditions*

192

193 Water temperatures within the cave range between 29.6°C (31 August, 2003)  
194 and 20.3°C (29 February, 2004). The pattern of the seasonal change in water  
195 temperatures within the cave is similar to the mean monthly temperatures at 30 m

196 deep ( $T_{30m}$ ) around Okinawa for 1874-2001 (Japan Oceanographic Data Center;  
197 J-DOSS) (Fig. 3a). Patterns of daily changes in temperature are synchronous with  
198 the tidal cycle (Fig. 3b). This implies that the alternation of water masses within the  
199 cave is caused by the tidal cycle.

200

#### 201 *4.2. Sedimentology and composition*

202

203 Traces of biogenic activity are lacking in the cave's bottom sediment, and  
204 burrowing macrobenthos such as polychaetes are absent from the core samples.  
205 Sedimentary structures such as laminae and trace fossils are absent. The mud  
206 content of the sediment is  $70 \pm 5\%$ , while particles larger than  $1 \phi$  range from 0.2 to  
207 6.1%, with an average of about 2% by weight (Fig. 4). The carbonate content of  
208 sand-size sediment ranges between 78 and 96%, while the content of mud-size  
209 sediment ranges from 90 to 97%. In both sand- and mud-size sediments, the  
210 carbonate content is significantly low in the lower portion (Fig. 4).

211 XRD analysis shows that bulk samples consist of 50-58% high-Mg calcite,  
212 30-37% aragonite and 9-19% calcite (Fig. 4), with relative percentages determined  
213 by peak-intensity analysis (Neumann, 1965). The magnesium concentrations  
214 determined by X-ray diffraction (Goldsmith et al., 1961) were  $13.9 \pm 1.6$  mole %  
215 Mg in high-Mg calcite. As noted below, benthic foraminifera are abundant in the  
216 cave sediments. It is likely that their shells contribute high-Mg calcite. The  $\delta^{18}\text{O}$   
217 and  $\delta^{13}\text{C}$  of carbonate mud are  $-1.19 \pm 0.08\text{‰}$  and  $1.32 \pm 0.11\text{‰}$  PDB, respectively  
218 (Fig. 4). There are no significant changes in the stratigraphic distribution of  
219 minerals, or of oxygen and carbon isotope ratios.

220 Both sand- and mud-size particles are dominated by carbonate debris (Figs. 4  
221 and 5). In sand-size particles, the identified component consists dominantly of

222 sponge spicules, benthic foraminifera, ostracods and fragments of echinoids (Fig. 4).  
223 In addition, many pumice grains occur in the lower part, where the carbonate  
224 content is relatively low (Fig. 4). In mud-size particles, the identified component  
225 consists dominantly of spicules of both sponges and didemnids (Fig. 5). Coccoliths  
226 are common in all samples. There are no systematic changes in the stratigraphic  
227 distribution of grain components, except for the pumice grains. The stratigraphic  
228 distribution of pumice grains exhibits a pattern similar to that for non-carbonate  
229 grains (Fig. 4). From this, we think that the deposition of pumice grains caused the  
230 relatively low carbonate content of the lowermost part. We estimate that the  
231 contribution of pumice grains to the total sedimentation was less than 20% by  
232 weight. The pumice grains were transported to this area by the Kuroshio Current,  
233 since there are no active volcanoes around Ie Island or Okinawa mainland.

234

#### 235 4.3. Radiocarbon age dating

236

237 Radiocarbon-dated mollusc shells and carbonate mud are shown in Table 1. The  
238 gastropod *Neritopsis radula* (2 cm in shell length) and bivalves *Bentharca*  
239 *irregularis*, *Paravamussium crypticum*, *Cyclopecten ryukyuensis*, *Carditella*  
240 *iejimensis* and *Coralliophaga hyalina* live in submarine caves or cryptic habitats  
241 (Hayami and Kase, 1993). All bivalves are very small in adult size, at less than 5  
242 mm in length and height and are epifaunal or semi-infaunal. The small gastropods  
243 *Stosicia (Isseliella) bourguignati*, *Emarginula crassicostata* and *Iniforis poecila*  
244 also occur in the intertidal zone and beach drift and are epifaunal or semi-infaunal.  
245 In contrast, *Tenagodus (Tenagodus) cumingi* occurs as clusters buried in sponges  
246 attached to cave walls, and its  $^{14}\text{C}$  age may be older than that of other molluscs in  
247 the same stratigraphic horizon (Kitamura et al., 2003).

248 The results of  $^{14}\text{C}$  dating are plotted in Fig. 6. In the depth interval from 35 cm

249 to 6 cm, all ages fall nicely along an age-depth line, implying continuous deposition  
250 and a constant sedimentation rate of 20.0 cm/kyr. Although pumice grains  
251 accumulated during the deposition of the lower part (43 to 35 cm in depth), the  
252 relative increase in sedimentation rate was less than 20%, based on their  
253 above-noted contribution to total sedimentation. Therefore, we use 20.0 cm/kyr as  
254 the sedimentation rate below 35 cm depth. Using this value, the age of the lowest  
255 sampled sediments is estimated as AD 100, which agrees closely with the  $^{14}\text{C}$  age  
256 of AD 0 of the lowest sample including *T. cumingi* (Fig. 6). The sedimentation rate  
257 of these submarine cave deposits is similar to that of postglacial deep-sea deposits  
258 (400-2,500 m depth) in the Okinawa Trough and Ryukyu Trench slope  
259 (Ujiié and Ujiié, 1999; Xu and Oda, 1999; Jian et al., 2000).

260 Two samples above the 6-cm level contain  $100.5 \pm 0.4$  pMC (percent modern  
261 carbon) and  $111.5 \pm 0.4$  pMC (Table 1). According to Stuiver and Polach (1977),  
262 these values approximate the  $\Delta^{14}\text{C}$  values of  $4.5 \pm 3.8\text{‰}$  and  $111.5 \pm 4.2\text{‰}$ . Such  
263 high  $\Delta^{14}\text{C}$  values are derived from  $^{14}\text{CO}_2$  that was created by atmospheric weapons  
264 testing in the 1950s and early 1960s. Based on coral records for the period  
265 1913-1979 from Okinawa,  $\Delta^{14}\text{C}$  values increased from  $-42$  to  $+175\text{‰}$  between  
266 1954-1956 and 1970, and then remained stable until 1979 (Konishi et al., 1982).  
267 This value is higher than  $80\text{‰}$  of the ocean, except for areas influenced by  
268 upwelling (Guilderson et al., 2000). Thus, the ages of the upper and lower samples  
269 (0-2.2 cm and 3.4-5.8 cm, respectively) correspond to ca. 1960 and the latter half of  
270 the 1960s, respectively (Fig. 6). The dated mollusc species are epifaunal or  
271 semi-infaunal suspension feeders (Hayami and Kase, 1993). Based on this, the  
272 reversed age relationship of the two samples was caused by bioturbation. Therefore,  
273 we think that the thickness of the mixed surface layer ranges from 2 to 6 cm.

274 Because of surface mixing, the temporal resolution of core data is calculated as 100  
275 to 300 years using a value of 20.0 cm/kyr for the sedimentation rate.

276 The radiocarbon ages of the dated carbonate mud are 1-2 kyr older than those of  
277 mollusc samples. Considering the presence of biogenic grains with high  $\Delta^{14}\text{C}$   
278 values, the true age differences in the upper 6 cm of sediment are significantly  
279 larger than the numerical ages. There is no doubt that the age differences between  
280 them tend to increase with time (Fig. 6).

281

#### 282 4.4. *Micro-bivalves*

283

284 We identified a total of 25 bivalves (Table 2), which exhibit excellent  
285 preservation despite being mostly disarticulated. The prodissoconch can be  
286 observed in many individuals. These factors imply that the fossils represent a life  
287 assemblage. The micro-bivalvia *Parvamussium crypticum*, *Cosa kinjoi*, *Carditella*  
288 *iejimensis* and *Cyclopecten ryukyuensis* predominate and are suspension feeders.  
289 They are endemic to cavernicolous environments (Hayami and Kase, 1993). The  
290 stratigraphic distribution of their accumulation rates is shown in Figure 7; the range  
291 for a given species is less than 0.03/cm<sup>2</sup>/yr. There is no significant temporal change  
292 in species composition or in the accumulation rate of the dominant species.

293 *P. crypticum*, *C. kinjoi* and *C. ryukyuensis* are not suitable for the reconstruction  
294 of palaeotemperature from oxygen isotope composition. This is because *C. kinjoi*  
295 and *C. ryukyuensis* are too small (<100 mg) for such analysis, while *P. crypticum*  
296 consists of a mixture of calcite and aragonite. The isotope composition of *C.*  
297 *iejimensis*, which consists of 100% aragonite, is alone useful for the reconstruction  
298 of palaeotemperature.

299

300 4.5. Oxygen isotopic analysis of *Carditella iejimensis*

301

302 McConnaughey (1989a) has shown that calcifying organisms with a fast growth  
303 rate may have lower  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values than would be expected based on  
304 equilibrium criteria alone. This depletion of the heavier stable isotopes of both  
305 elements is caused by kinetic isotope fractionation during the  $\text{CO}_2$  hydration and  
306 hydroxylation, with rapid calcification favoring strong kinetic effects  
307 (McConnaughey, 1989b; McConnaughey and Whelan, 1997; Zeebe and  
308 Wolf-Gladrow, 2001). Since kinetic effects result in simultaneous depletion of  $^{18}\text{O}$   
309 and  $^{13}\text{C}$ , the relationship between them can be used to check whether kinetic isotope  
310 effect influences  $^{18}\text{O}$  value of *C. iejimensis*. There is no significant correlation  
311 between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  ratios of the specimens from surface sediment (Fig. 8). We  
312 think that kinetic effects are not significant for the oxygen isotope composition of *C.*  
313 *iejimensis*. The composition is controlled by both the temperature and the isotopic  
314 composition of the ambient seawater in which the animals live.

315 As noted above, our temperature record indicates that the mean monthly water  
316 temperature within the cave is almost the same as the water temperature at 30 m  
317 depth ( $T_{30\text{m}}$ ) off the southwestern Okinawa Island (Fig. 2). We did not measure  
318 salinity within Daidokutsu cave. However, its value is likely equal with that of  
319 ocean surface water, since water has flowed continuously into and out of the cave  
320 for the past 2,000 years, as noted below.

321 All  $\delta^{18}\text{O}$  values were calculated using the following equation of Goodwin et al.  
322 (2001):

323  $T_{\delta^{18}\text{O}} = 20.6 - 4.34[\delta^{18}\text{O}_{\text{aragonite}} - (\delta^{18}\text{O}_{\text{water}} - 0.2)]$ , where  $\delta^{18}\text{O}_{\text{aragonite}}$  is  
324 measured relative to the PDB scale and  $\delta^{18}\text{O}_{\text{water}}$  is relative to the SMOW scale.

325 Oba (1988) reported the following linear relationship between water salinity ( $S$ )  
326 and  $\delta^{18}\text{O}$  of seawater in the region from the East China Sea to the Kuroshio Current  
327 off the southern Japanese Island:

$$328 \quad \delta^{18}\text{O}_{\text{water}} = -6.76 + 0.203S$$

329 Instrumental records from Sesoko Marine Science Center, 10 km from our  
330 sampling point, show that the salinity of the surface waters ranges from 34.5 p.s.u.  
331 in summer to 35.1 p.s.u. in winter (Nakano and Nakamura, 1993a, b). Based on  
332 these data, we infer the salinity to be 34.8 p.s.u. Changes in  $\delta^{18}\text{O}_{\text{water}}$  derived from  
333 seasonal variation of salinity are estimated at 0.12‰ and correspond to small  
334 difference in estimated temperature of 0.4°C.

335 The results of radiocarbon dating suggest that the duration of samples from the  
336 surface sediments is approximately the last 50 yrs. For specimens of *C. iejimensis*  
337 from surface sediment, there exists a statistically significant correlation between the  
338 shell sizes and  $\delta^{18}\text{O}$  values ( $r=-0.55$ ,  $n=30$ ,  $p<0.01$ ) (Fig. 9). Their  $T_{\delta^{18}\text{O}}$  ranges  
339 between 22.4 and 26.6°C (Fig. 10). For the same sized individuals, the difference of  
340  $T_{\delta^{18}\text{O}}$  is 2°C. The  $T_{\delta^{18}\text{O}}$  ranges between 23.5 and 26.5°C in samples of the cored  
341 sediments (Fig. 10).

342

## 343 **5. Discussion**

344

### 345 *5.1. Paleoenvironmental changes within the cave*

346

347 The occurrences of coccoliths and pumice indicate that sea water has flowed  
348 continuously into Daidokutsu cave. This is supported by occurrences of the  
349 cavernicolous bivalve species (Fig. 7). Since the level of predation pressure is low  
350 in a submarine cave, the biomass of suspension feeders approaches the carrying  
351 capacity (Hayami and Kase, 1993). In the case of micro-bivalves, food supply is a  
352 more important factor for the carrying capacity than is competition for space. Thus,  
353 the constant accumulation rate of the dominant bivalves implies that an amount of  
354 nutrition, which has been brought from outside the cave, was not varied over the  
355 past 2,000 years. It seems probable that this influx of water is related to the  
356 fluctuation of water masses within the cave related to the tidal cycle. The current  
357 velocity was not measured within the cave, although it is very low, because  
358 mud-size particles have deposited continuously over the past 2,000 yrs. In summary,  
359 it seems that the cave has remained sheltered, with low hydrodynamic energy, and  
360 we believe that the sediments constitute a continuous depositional record.

361

## 362 *5.2. Sources of carbonate debris*

363

364 Our analyses of grain distribution and grain compositions show that mud-size  
365 carbonate debris predominates in the submarine-cave deposits (mean of  $67.4 \pm$   
366  $5.5\%$  and range of  $55.2$  to  $76.5\%$ ). Both the  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  ratios of mud-size  
367 carbonate in the submarine cave deposit are nearly  $0\%$  PDB (Fig. 4). This indicates  
368 that the main source of carbonate mud was marine carbonate, rather than carbonate  
369 precipitated on the surface of the cave walls and ceiling (e.g., stalactites and  
370 stalagmites) before the cave was submerged. Because the latter has significant low  
371 values for both the  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  ratios (Allan and Matthews, 1982). As noted  
372 above, the radiocarbon ages of the carbonate mud sediments are 1-2 kyr older than  
373 those of the mollusc samples (Fig. 6). Considering the presence of calcareous shells

374 such as coccoliths and the spicules of didemnids, the ages of unidentified grains are  
375 older than the dated carbonate-mud sediments. These factors imply that these grains  
376 are detritus from the emergent reef flat that has been reduced to its modern level by  
377 erosion (MacNeil, 1950). This process also explains the expansion of age  
378 differences between the unidentified mud-size particles and the mollusc samples.  
379 The water movement has transported suspended carbonate detritus into the cave,  
380 and these fine grains (including coccoliths) settled in the cave and were aggregated  
381 into pellets by suspension feeders such as coralline sponges, bivalves and  
382 brachiopods. Our results imply that there were no significant changes in condition  
383 of the hinterland during the past 2,000 years, except for the drift of pumice grains.  
384 In summary, the submarine cave deposits provide high-resolution records of the  
385 development of coral reef and volcanic activity around Ryukyu and Philippine  
386 Islands.

387

388

### 389 *5.3. Potential of C. iejimensis as an archive of Holocene palaeotemperature*

390

391 For specimens of *C. iejimensis* from surface sediment, there is a significant  
392 correlation between  $\delta^{18}\text{O}$  values and the shell sizes (Fig. 9). We calculated the  $T_{\delta^{18}\text{O}}$   
393 during the precipitation of the outer shell portion, which is 3.0 mm or more from  
394 the umbo, based on the relationship between the size and weight of *C. iejimensis*  
395 (Fig. 11) and the equation of the regression line. The calculated value is 26.8°C and  
396 corresponds to  $T_{30\text{m}}$  during July ( $27.0 \pm 1.4^\circ\text{C}$ ). The average value of  $T_{\delta^{18}\text{O}}$  for  
397 small shells is between  $T_{30\text{m}}$  during May ( $23.6 \pm 1.0^\circ\text{C}$ ) and June ( $24.8 \pm 1.1^\circ\text{C}$ ).  
398 From these data, we believe that shell deposition began in early April and ended  
399 in July. Shell growth of bivalves is generally influenced by a number of

400 environmental factors such as water temperature, salinity, and food availability. As  
401 noted above, there are not significant seasonal changes in salinity within the cave.  
402 Around Okinawa Island, the phytoplankton becomes most abundant in winter and  
403 remains at low levels in the other seasons (Imai et al., 1988; Xu et al, 2005). From  
404 these, the growth pattern of *C. iejimensis* is mainly controlled by temperature.

405 As noted above, the environment within the cave has been maintained over the  
406 past two millennia. Therefore we infer that the  $\delta^{18}\text{O}$  values of sea water and the  
407 temperatures both within and outside of the cave were the same for the past 2,000  
408 yrs. Based on this assumption, we compare the  $T_{\delta^{18}\text{O}}$  values of fossil and recent  
409 shells of *C. iejimensis* (Fig. 10). Except for two shells, the values derived from  
410 fossil fall within the range for recent specimens. In contrast, two shells have values  
411 that are 2°C higher than the estimated temperature derived from the equation of the  
412 regression line. The ages of these horizons are estimated to be AD  $340 \pm 40$  ( $26.2 \pm$   
413  $0.1^\circ\text{C}$ ) and AD  $1000 \pm 40$  ( $26.1 \pm 0.2^\circ\text{C}$ ), respectively. According to the J-DOSS  
414 database, this value corresponds to a May and June  $T_{30\text{m}}$  of  $26.2^\circ\text{C}$  at Kume Island,  
415 which is located between the study site (100 km to the west) and the main path of  
416 the Kuroshio Current (Fig. 1).

417 According to Yang et al. (2002), warm conditions prevailed in eastern China at  
418 AD 0-300 and AD 800-1100. These two intervals correspond to the Roman Warm  
419 Period and Medieval Warm Period, respectively. However, we detected no evidence  
420 in the  $\delta^{18}\text{O}$  record of *C. iejimensis* of the cool conditions in the Little Ice Age (LIA;  
421 AD 1400-1900). Glacial advances in both hemispheres (Grove, 1988) and enhanced  
422 polar atmospheric circulation (Kreutz et al., 1997) suggest that the LIA was a global  
423 event. Unfortunately, there are no proxy climate data, or historical documents,

424 which allowed these workers to reconstruct the climatic conditions of Okinawa  
425 during the LIA. There is abundant evidence of LIA cool conditions in the terrestrial  
426 records of China (Qian and Zhu, 2002; Yang et al., 2002). However, Rind (1998,  
427 2000) has suggested that cooler LIA conditions were possibly restricted to higher  
428 latitudes. None of the few LIA reconstructions of the tropics identify cold periods  
429 synchronous with those in Northern Hemisphere composite reconstructions (Hendy  
430 et al., 2002; Gagan et al., 2004). It is therefore probable that the shallow seas  
431 around Okinawa Island did not experience cool conditions from AD 1400 to 1900.

432 In summary, paleoclimatic records based on oxygen isotope analyses of *C.*  
433 *iejimensis* are not inconsistent with other proxy series from the Northern  
434 Hemisphere, including China, except for the LIA. We thus believe that  $\delta^{18}\text{O}$   
435 analysis of *C. iejimensis* is a reliable source of information about paleoclimatic  
436 fluctuations in the Okinawa area. Reconstructing additional paleoclimate data  
437 would require improved sampling for millimeter-size shells of *C. iejimensis*.

438

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446

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659 Figure caption

660 Fig. 1 Location of Daidokutsu submarine cave on Ie Island, off Okinawa Island,  
661 Japan.

662 Fig. 2 Simplified transverse section and horizontal projection of Daidokutsu cave.

663 We observed air bubbles rising from the sea floor (2 m depth) above Daidokutsu  
664 during sediment sampling (duration of sampling was about 20 minutes).

665 Fig. 3 Observed temperatures within the cave. (a) Comparison of observed temperatures  
666 with mean monthly water temperature at 30 m depth (the Japan Oceanographic Data  
667 Center), (b) Comparison of observed temperatures with tidal records through  
668 September 2003.

669 Fig. 4 Columnar diagrams of submarine-cave sediment core, showing stratigraphic  
670 change in grain distribution, carbonate content, mineral composition, isotopic ratios  
671 of mud-size carbonate and components of sand-size particles. M-C: Mg calcite, C:  
672 calcite, A: aragonite.

673 Fig. 5 Columnar diagrams of submarine-cave sediment core, showing stratigraphic  
674 change in component of mud-size particles.

675 Fig. 6 Results of AMS  $^{14}\text{C}$  dating of molluscs and mud-size carbonate.

676 Fig. 7 Stratigraphic distribution of the accumulation rate of the four dominant  
677 bivalves.

678 Fig. 8  $\delta^{18}\text{O}$ - $\delta^{13}\text{C}$  plot for the cavernicolous micro-bivalve *C. iejimensis* in 5-cm-thick  
679 surface sediments.

680 Fig. 9 Relationship between the  $\delta^{18}\text{O}$  and shell height of *C. iejimensis* in surface  
681 sediments.

682 Fig. 10 Relationship between the  $\delta^{18}\text{O}$ -derived temperature and shell height of the

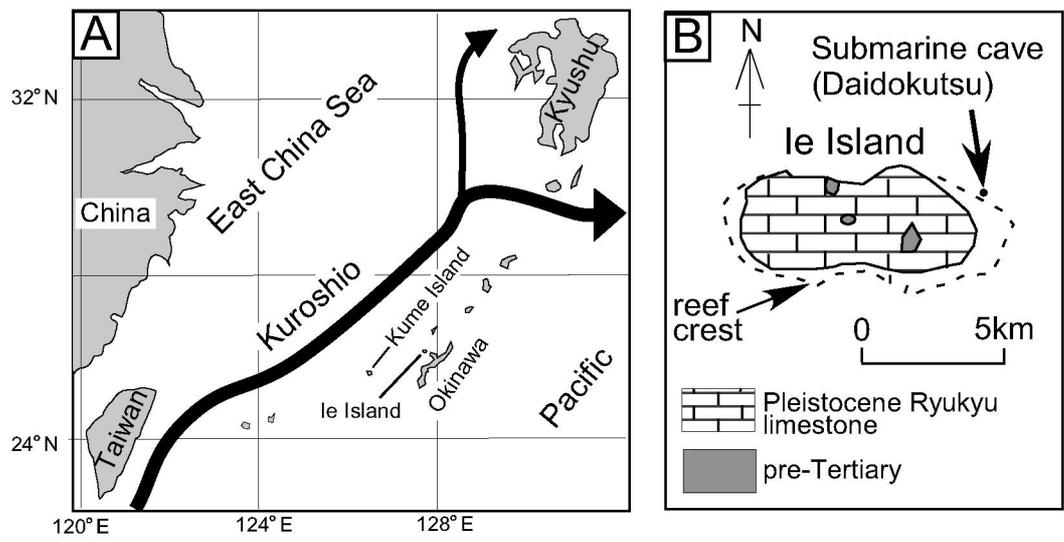
683 cavernicolous micro-bivalve *C. iejimensis* in 5-cm-thick surface sediments and the  
684 cored sediments. The regression line is calculated from the data of specimens from  
685 surface sediments.

686 Fig. 11 Relationship between the shell height and shell weight of *C. iejimensis*.

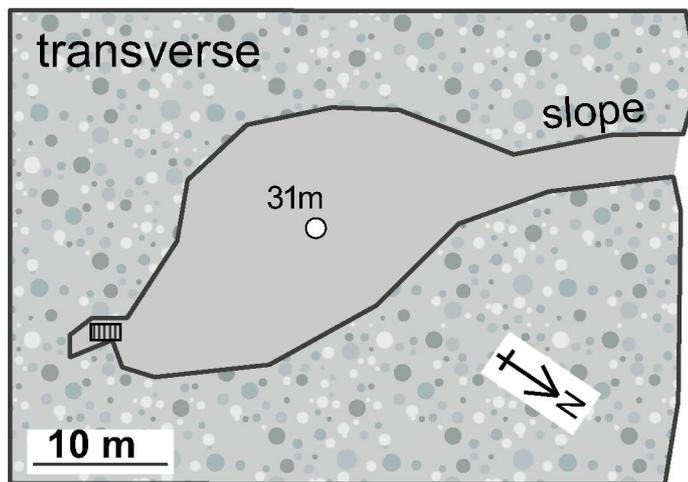
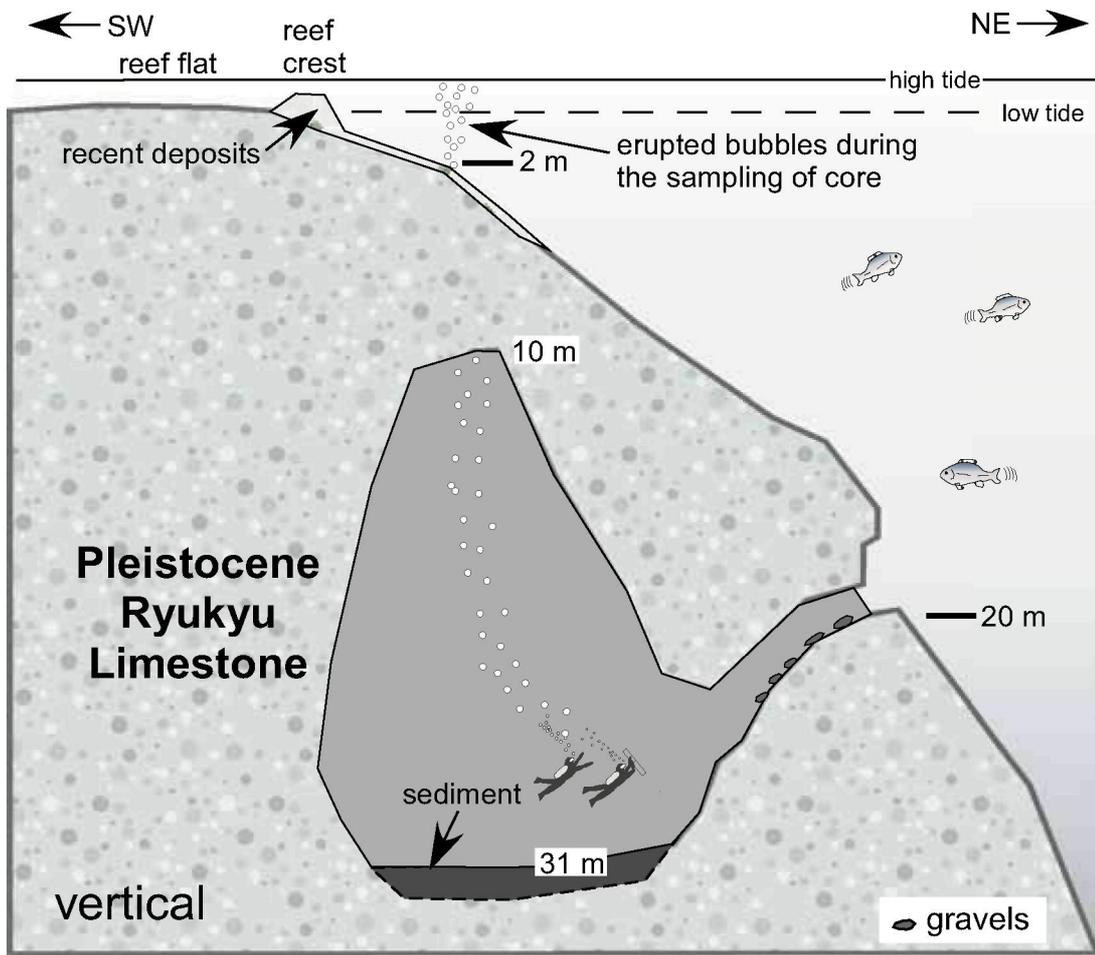
687 Table 1 Result of mollusc <sup>14</sup>C-dating. All samples were analyzed with an accelerator  
688 mass spectrometry by Beta-Analytic Corporation.

689 Table 2 List of bivalve species from Daidokutsu. A: articulated shells, R: right  
690 valve, L: left valve.

691

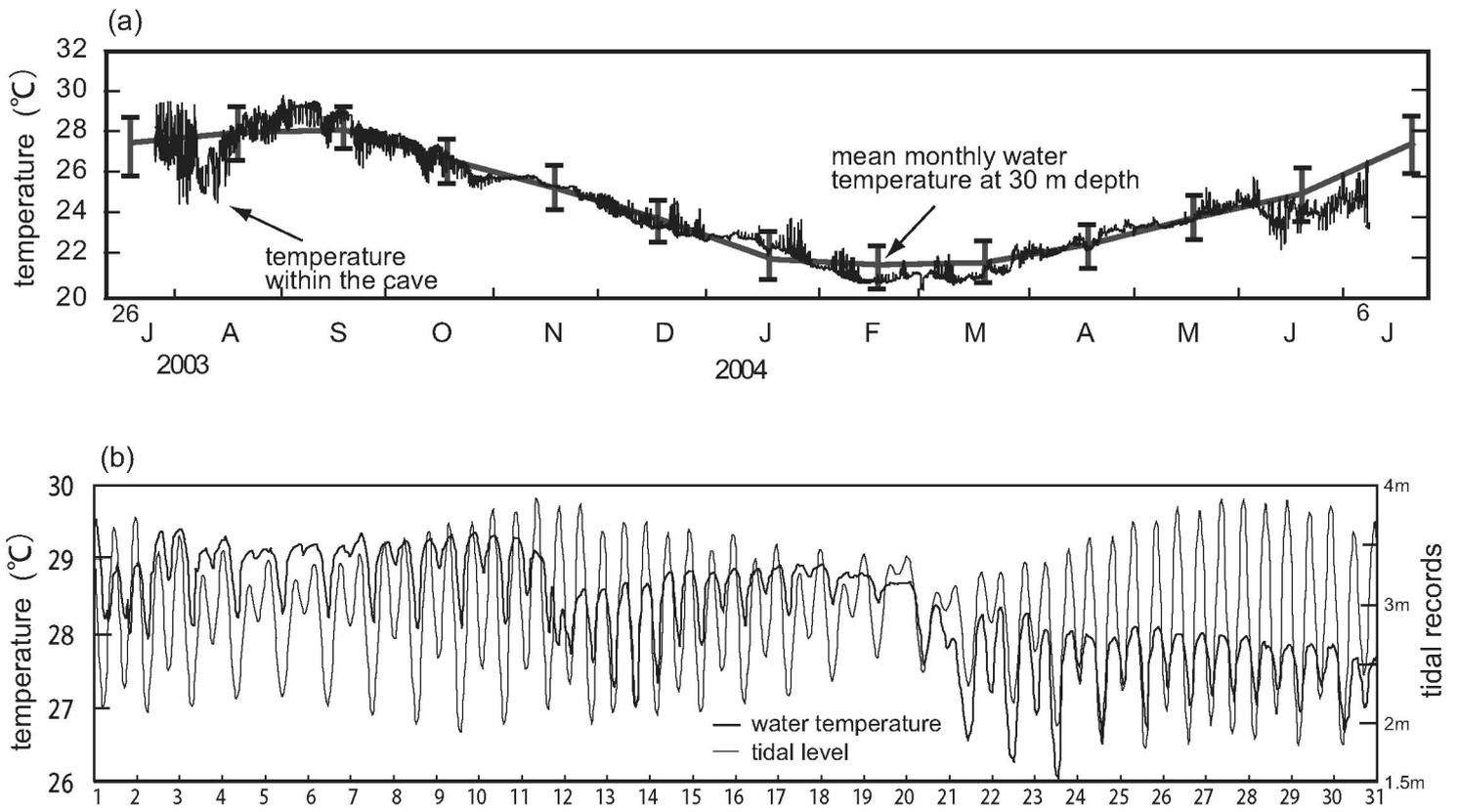


Kitamura et al. Fig. 1



- sample site of cored sediments
- ▣ thermometer

Kitamura et al. Fig. 2



Kitamura et al. Fig. 3

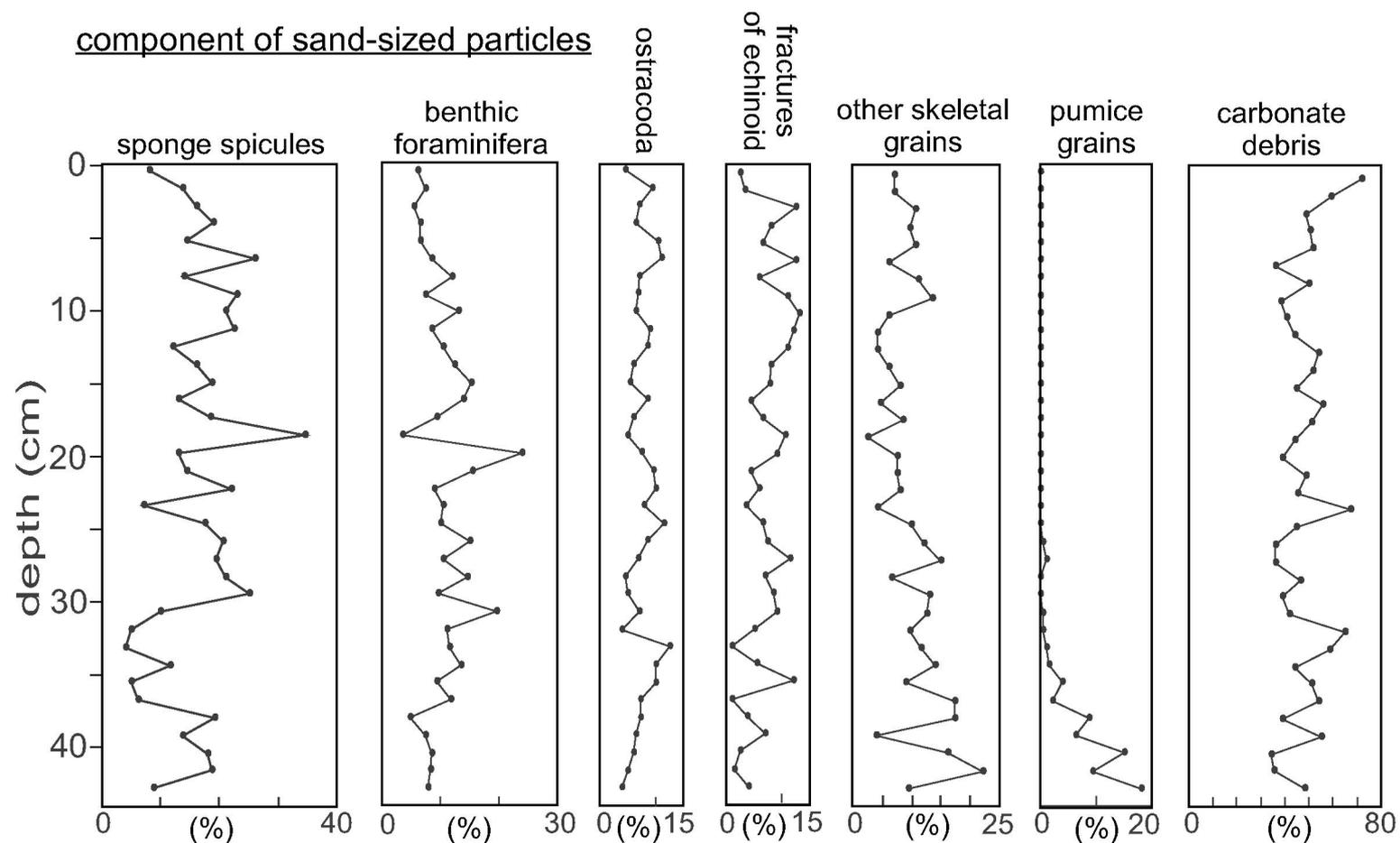
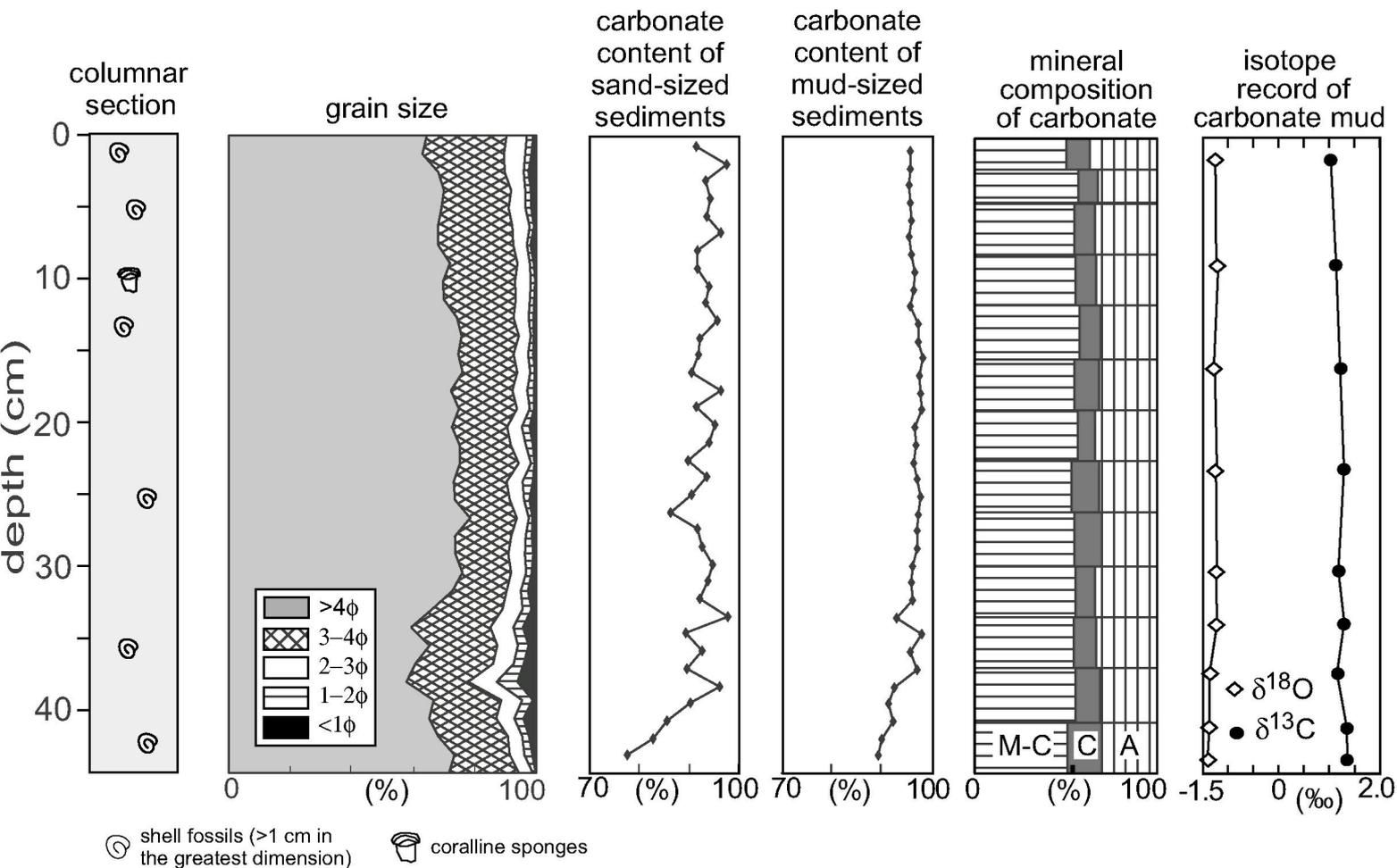


Fig. 4 Kitamura et al

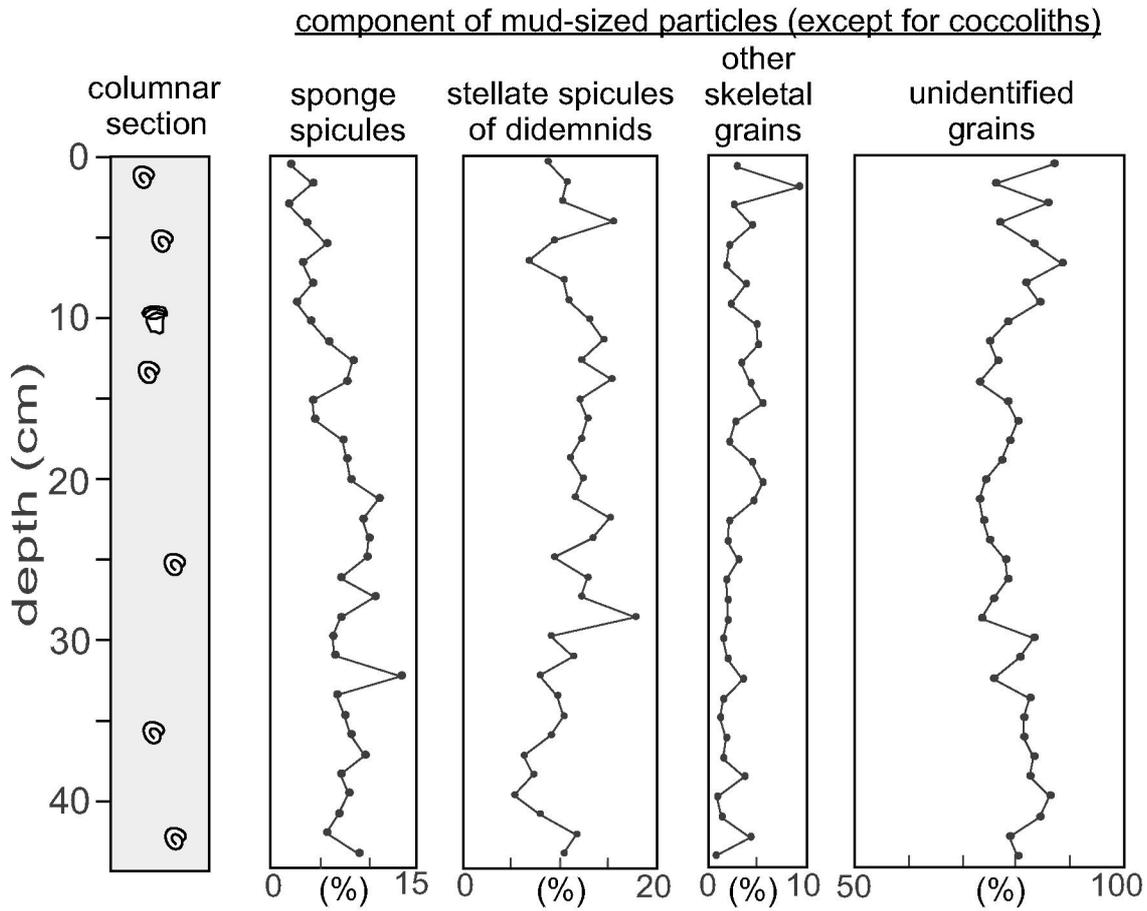


Fig. 5 Kitamura et al

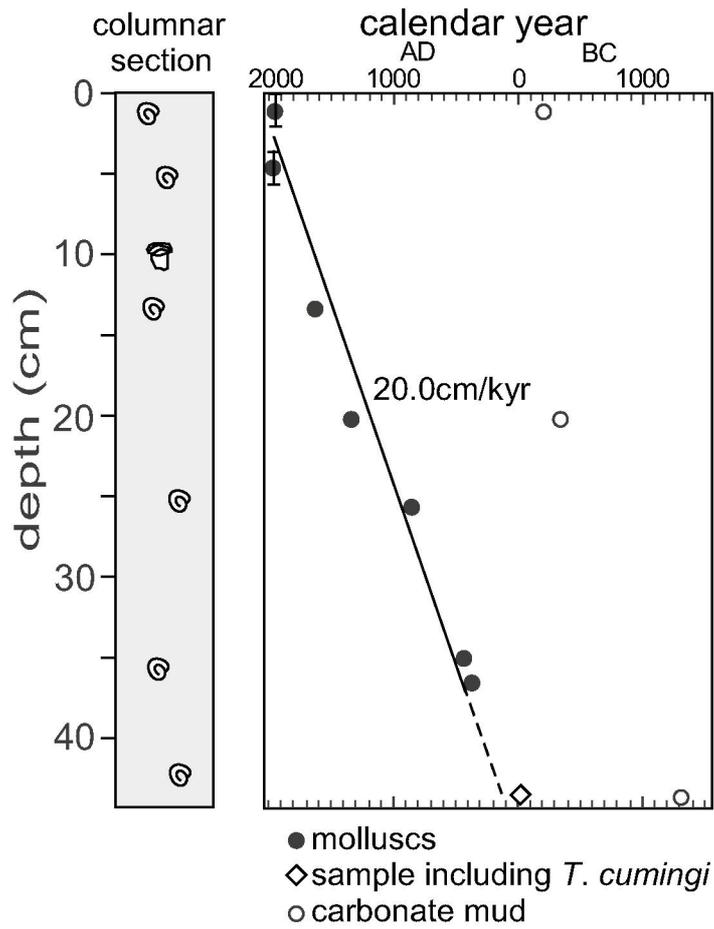
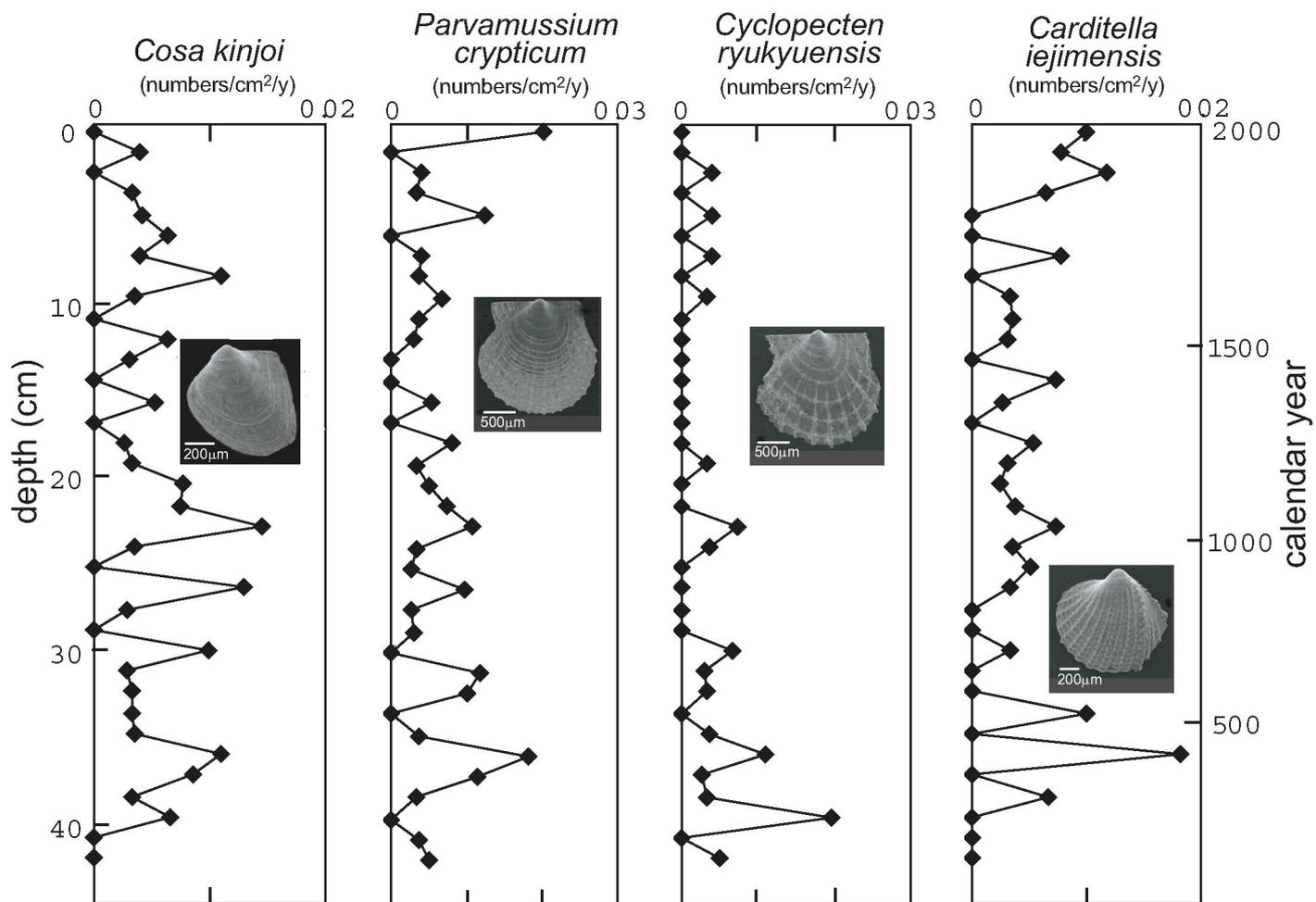
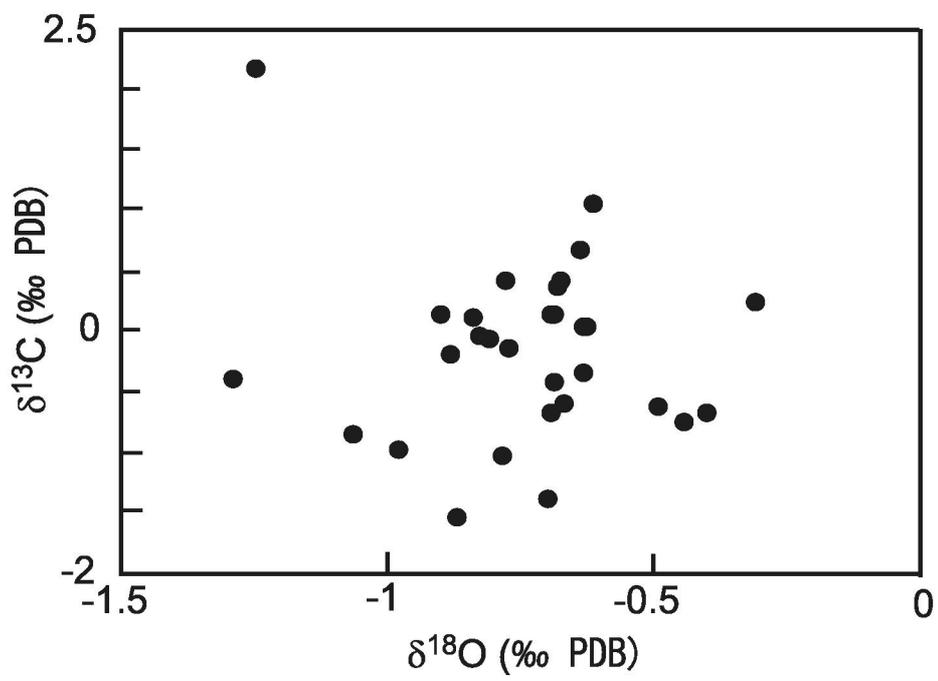
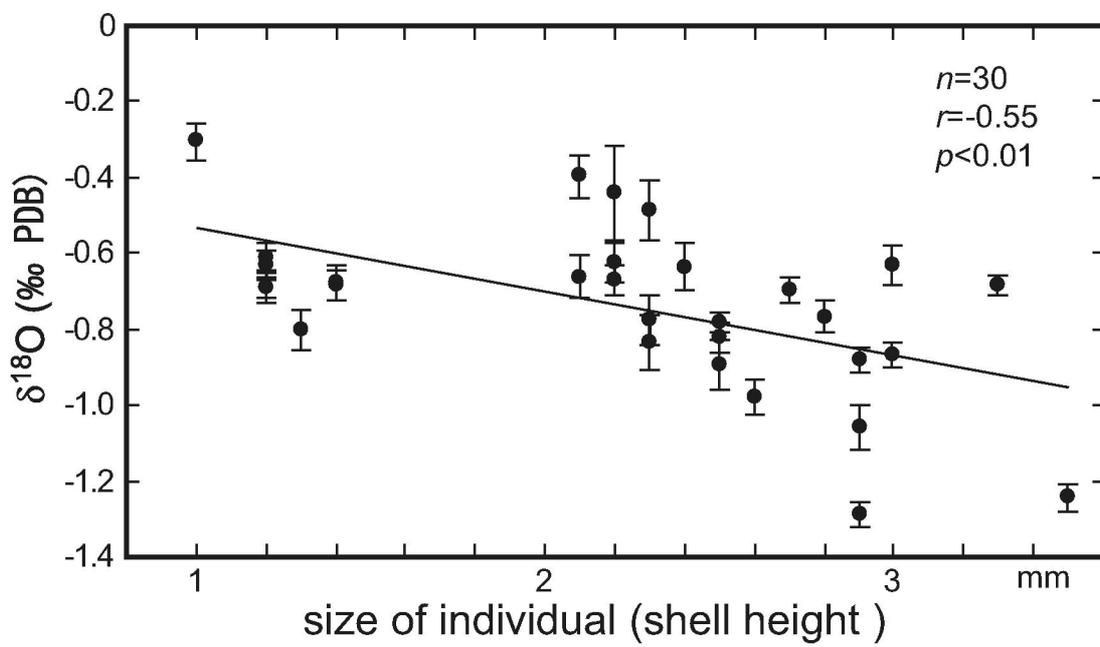


Fig. 6 Kitamura et al



Kitamura et al Fig. 7



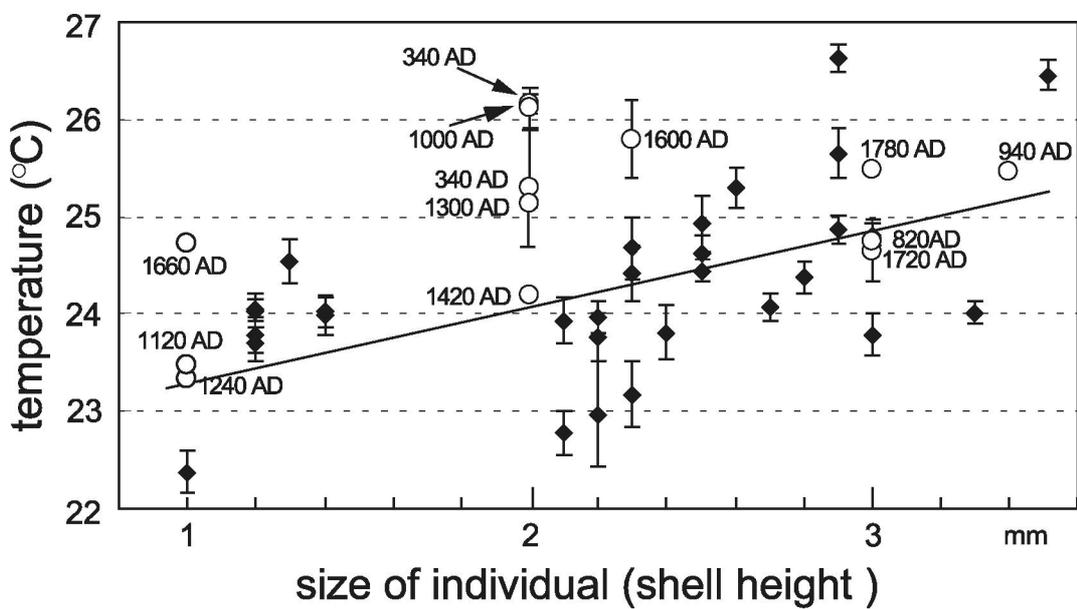


Kitamura et al Fig. 9

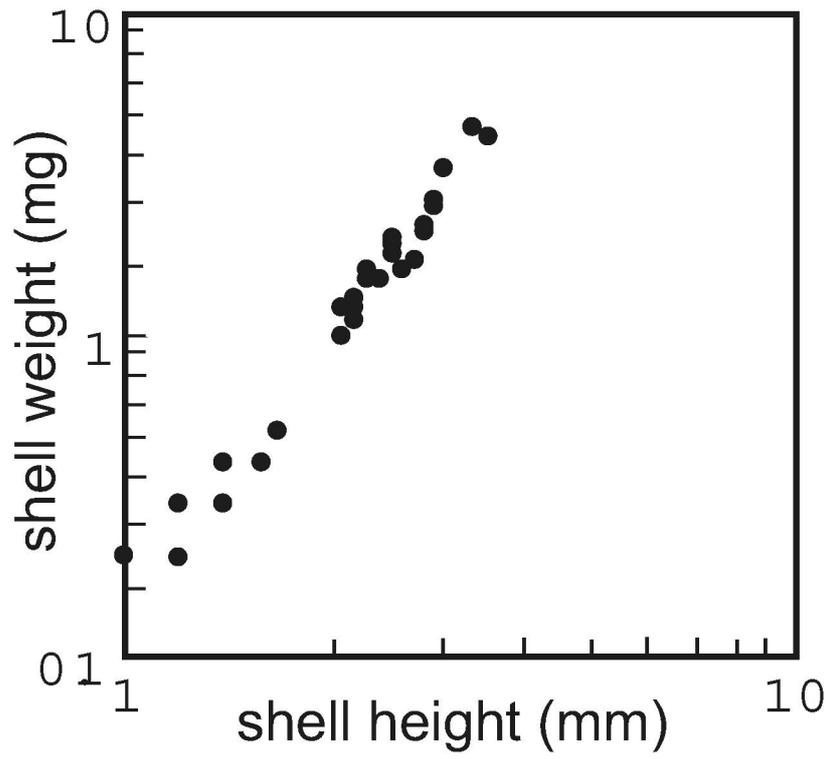
◆ specimens from surface sediments

○ specimens from cored sediments

The regression line is calculated from the data of specimens from surface sediments



Kitamura et al Fig. 10



Kitamura et al Fig. 11

depth (cm)	laboratory number	sample	measured <sup>14</sup> C age (BP)	δ <sup>13</sup> C	conventional <sup>14</sup> C age* (BP)	Calibrated age	
						intercept** (cal BP)	age ranges (cal BP)
0–2.2	Beta-185831	mix of <i>Parvamussium crypticum</i> (one individual), <i>Carditella iejimensis</i> (one individual) and <i>Iniforis poecila</i> (one individual)	106±0.4 pMC	+1.4	100.5±0.4 pMC	–	–
3.4–5.8	Beta-185832	mix of <i>Parvamussium crypticum</i> (one individual), <i>Cyclopecten ryukyuensis</i> (one individual), <i>Carditella iejimensis</i> (one individual) and <i>Coralliophaga hyalina</i> (one individual)	118±0.4 pMC	+2.5	111.5±0.4 pMC	–	–
13	Beta-167418	<i>Neritopsis radula</i> (one individual)	220±40	+1.5	650±40	320	300–250
19.6	Beta-180894	<i>Acar</i> sp. aff. <i>A. plicata</i> (one individual)	590±40	+2.7	1040±40	610	630–540
25	Beta-167416	<i>Stosicia (Isseliella) bourguignati</i> (one individual)	1080±40	+2.9	1540±40	1100	1172–1056
34	Beta-185221	<i>Septifer</i> sp. (one individual)	1530±40	+2.8	1530±40	1520	1560–1460
35.5	Beta-167415	<i>Mitrella</i> sp. (one individual)	1570±40	+1.7	2010±40	1580	1640–1540
43	Beta-167414	mix of <i>Bentharca irregularis</i> (one individual), <i>Emarginula crassicostata</i> (one individual) and <i>Tenagodus (Tenagodus) cumingi</i> (one individual)	1920±40	+3.2	2380±40	2000	2050–1940
0–0.8	Beta-194743	Carbonate mud	2090±40	+0.5	2510±40	2150	2250–2120
19.0–20.2	Beta-194742	Carbonate mud	2160±40	+0.9	2580±40	2290	2310–2200
41.8–43.0	Beta-194741	Carbonate mud	2960±40	+0.8	3380±40	3250	3310–3200

\*reservoir correction was not applied.

\*\*intercepts between the conventional <sup>14</sup>C age and the calibration curve of Stuiver *et al.* (1998).

All errors are 1σ.

pMC: percent modern carbon

Species \ Sample No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	
<i>Acar</i> sp. aff. <i>A. plicata</i>		R1	L1		L1														R1	L2																	
<i>Bentharca excavata</i>													R1																								
<i>Bentharca</i> sp.						L1																												R1			
<i>Cosa kinjoi</i>			R1/L1	A1	A1/R1/L1	R2/L1	R1	R1	R1	R1	R2/L1		L1	R3/L1		R1	L4	R1/L1	A1/R1/L1	L1	R1		A1/R1		R1	R1/L1		R1	R3	L1	A1/R1	L1	A1		L1		
<i>Cosa uchimae</i>			R1	R2				L1		L1	R2												L1			R1		R1									
<i>Limopsoidea?</i> gen. et sp. Indet.					R1									R1																						R1	
<i>Brachidontes</i> sp.					R1			R1/L1	L1						R1						L1																
<i>Septifer</i> sp.						R1																															
<i>Dacrydium zebra</i>			R1																																	R1	
<i>Parvamussium crypticum</i>	R1	L1		L1	A2/L1/R1	R2/L3	R1		L3	R1/L3		R1	R1	R2/L1	L1	L1	L3	R1/L1	R2	L1	R2/L1		R2			L1	R1	R1/L1	L1	R1		R3	L1	R1		R2/L2	
<i>Cyclopecten ryukyuensis</i>	R1		R4/L2	L1	A1	L3	R1		L1	L1	L2						L1	L2																			
<i>Chlamydeffa incubata</i>																																					
<i>Chlamydeffa tenuissima</i>											L1																										R1
<i>Divarilima elegans</i>	R1				R1/L1	R1/L2	R1		L1		R1	A1						L2		R1	R1													R1		L1	
<i>Limaria</i> sp.																																					L1
<i>Cardita uruma</i>																																					
<i>Carditella iejimensis</i>				L2		R4/L1		L3			R1			L1	R1/L1	L1	R1/L1	A1	L1	L1	R1/L1		R1	R1/L1		R1	R1	L1		R2				R2	L3	R2	A1/L1
<i>Salaputium unicum</i>										L1																											
<i>Kelliella japonica</i>			L1		L1											L1																					A1
<i>Periglyta chemnitzii</i>						L1																															
<i>Gafrarium pectinatum</i>		R1																																			
<i>Coralliophaga hyalina</i>																																					R1
<i>Hiatella</i> sp. aff. <i>H. orientalis</i>																																					L1
<i>Poromyda</i> sp.													R1	L1				R1																			R1
<i>Halonympha asiatica</i>													L1																								L1
Depth (cm)	43.0-41.8	41.8-40.6	40.6-39.4	39.4-38.2	38.2-37.0	37.0-35.8	35.8-34.6	34.6-33.4	33.4-32.2	32.2-31.0	31.0-29.8	29.8-28.6	28.6-27.4	27.4-26.2	26.2-25.0	25.0-23.8	23.8-22.6	22.6-21.4	21.4-20.2	20.2-19.0	19.0-17.8	17.8-16.6	16.6-15.4	15.4-14.2	14.2-13.0	13.0-11.8	11.8-10.6	10.6-9.4	9.4-8.2	8.2-7.0	7.0-5.8	5.8-4.6	4.6-3.4	3.4-2.2	2.2-1.1	1.1-0	

Kitamura et al Table 2