- 1 Climatic and hydrologic variability in the East China Sea during the last 7,000 years based on
- 2 oxygen isotope records of the submarine cavernicolous micro-bivalve Carditella iejimensis
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### 12 Abstract

13The micro-bivalve Carditella iejimensis inhabits the sediment surface within submarine caves at Ie Island, Okinawa, Japan. A comparison of the  $\delta^{18}O$  values ( $\delta^{18}O_{aragonite}$ ) of empty and living 1415shells indicates that the shell is formed over several seasons, and that the main cause of mortality is low water temperature. According to this hypothesis, it is likely that the shells with heaviest 16 $\delta^{18}O_{aragonite}$  values (-0.4‰) among the recent dataset formed under or close to the lower limit of 1718 growth temperature for the species. Assuming an unchanging temperature tolerance of the species 19during the Holocene, samples with  $\delta^{18}O_{aragonite}$  values heavier than -0.4‰ indicate unusually low temperatures and enrichment of  $\delta^{18}$ O of sea water ( $\delta^{18}O_{seawater}$ ). The  $\delta^{18}O_{aragonite}$  record of C. 2021*iejimensis* from sediment cores recovered from Daidokutsu cave shows no clear long-term trend in sea surface temperature or  $\delta^{18}O_{\text{seawater}}$  in the East China Sea during the past 7,000 years, and 22indicates anomalously cool and dry events (enrichment in  $\delta^{18}O_{seawater}$ ) at around 6,300 and 5,550 2324cal. years BP. These events may have been related to changes in the activity of the East Asian 25monsoon, related in turn to weakening solar activity. In contrast, these anomalies appear to be 26obscured during the last 1,000 years, including a weak Asian summer monsoon event during the 27Little Ice Age, thereby indicating that the mode of the East China Sea climatic and hydrologic 28response to decadal- to centennial-scale variability in the intensity of East Asian monsoon has 29varied over the past 7,000 years.

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*Key words:* East China Sea, Middle-Late Holocene, submarine cavernicolous micro-bivalve,
oxygen isotope, East Asian monsoon

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## 34 **1. Introduction**

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36 The magnitude of natural climate change during the Holocene is one of the key factors in 37 estimating the current and future effects of anthropogenic climate change (e.g., Bond et al., 1997, 382001; deMenocal, 2001; Cronin et al., 2003; Mayewski et al., 2004; Wang et al., 2005; Wanner et 39 al., 2008). It is necessary to undertake such studies in many regions of the world, not only because 40 of regional differences in ecosystems and the lifestyle of human populations, but because of spatial 41 variations in the nature and degree of climate change. East Asia, which is home to approximately 42one-third of the world's population, is strongly influenced by the East Asian monsoon (Fig. 1), 43which is driven by differential heating between the Asian continent and the surrounding seas and 44 Northwest Pacific.

45Precisely dated stalagmite oxygen isotope records from southern China reveal that Holocene 46 weakening in the summer monsoon corresponds to an orbitally induced reduction in summertime 47solar insolation in the Northern Hemisphere (e.g., Dykoski et al., 2005; Wang et al., 2005). On a 48decadal-century scale, weak Asian summer monsoon events correspond to periods of reduced 49solar activity (e.g., Neff et al., 2001; Jung et al., 2002; Gupta et al., 2003; Wang et al., 2005). However, Maher (2008) suggested that stalagmite  $\delta^{18}$ O records reflect the declining Holocene 5051influence of isotopically lighter, Indian-monsoon-sourced moisture over China and consequent 52increase in the proportion of isotopically heavy rainfall, sourced from the relatively oceanic East 53Asian monsoon. This view was proposed because stalagmite records do not match other East 54Asian proxy rainfall records based on the magnetic properties of loess/palaeosol (Maher and Hu, 552006) and intercomparisons of cave oxygen isotope data (Hu et al., 2008).

The winter monsoon has no such signature in the hydrological cycle, and is reconstructed mainly from changes in dust flux. Wang et al. (1999) and Lim et al. (2005) reported that the winter monsoon weakened at 4,000–3,000 years BP in response to increasing winter insolation. Yancheva et al. (2007) claimed that the strengths of the summer and winter monsoons are anti-correlated on a decadal time scale; however, this hypothesis has been challenged by Zhang and Lu (2007) and 2 Zhou et al. (2007). Other studies have argued that centennial-scale variations in the winter monsoon (intensification) are influenced by solar activity (weakening) (Lim et al., 2005; Xiao et al., 2006; Yancheva et al., 2007). Although there is intense debate regarding the nature of Holocene changes in the Asia monsoon, analyses of continental climate records have greatly improved our understanding in this regard. In contrast, few studies have considered the relationship between the East Asian monsoon and the oceanography of the East China Sea, which borders China and Japan.

67 Previous studies have examined Holocene oceanographic changes in the East China Sea based 68 on geochemical analysis of the planktonic foraminifera Globigerinoides ruber recovered from 69 deep-sea sedimentary cores (Jian et al., 2000; Ijiri et al., 2005; Sun et al., 2005; Lin et al., 2006). 70 Jian et al. (2000) proposed that a decrease in sea surface temperature (SST) during the period 714,600–2,700 cal. years BP was related to an intensification of the winter monsoon. In addition, the 72authors argued that this decrease in SST may have caused a remarkable decrease in the abundance 73 of the planktonic foraminifera *Pulleniatina obliquiloculata* (i.e., the *Pulleniatina* Minimum Event), 74which is regarded as the Kuroshio Current indicator species by Jian et al. (1996), Li et al. (1997), 75and Ujiié and Ujiié (1999). In contrast, Lin et al. (2006) reported no change in SST or sea surface 76 salinity during the Pulleniatina Minimum Event. This discrepancy among different studies may 77reflect the varying quality of data obtained from many different individuals of G. ruber.

78Globigerinoides ruber remains at water depths of 2–50 m during its life cycle (Fairbanks et al., 79 1982; Hemleben et al., 1989; Lin et al., 2004), and thrives in the East China Sea during autumn, 80 spring, and summer when SST is high and the water column is well stratified (Xu et al., 2005). 81 During the summertime development of thermal stratification, sea temperatures vary from 82 28–29°C at 0 m depth to 25°C at 50 m (Xu et al., 2005). Because a single sample for geochemical 83 analysis requires 20–40 individuals of G. ruber (Jian et al., 2000; Ijiri et al., 2005; Sun et al., 2005; 84 Lin et al., 2006), each sample represents a mixture of individuals that lived during different years 85 and at different depths.

To reconstruct climatic and hydrologic variability in the East China Sea during the Holocene, there exists the need for other long, continuous records of paleoceanographic change. To this end,

Kitamura et al. (2007b) and Yamamoto et al. (2008) measured the  $\delta^{18}O_{aragonite}$  values of empty 88 (dead) and whole shells of the cavernicolous micro-bivalve C. iejimensis, which lives in surface 89 90 sediment within the submarine cave Daidokutsu (meaning large cave in Japanese; 29 m water depth; 26°43'N, 127°44'E) of Okinawa, Japan (Fig. 2). Their data reveal that the  $\delta^{18}$ O-derived 9192temperature represents the springtime water temperature for each year, and that there exists no significant millennial-scale trend in the  $\delta^{18}O_{aragonite}$  record over the past 3,000 years. Because the 93 94mixed surface layer was less than 2 cm thick during the deposition of calcareous mud in the cave, 95 time-averaging of the fossil specimens was less than 100 years (Yamamoto et al., 2008). Thus, the  $\delta^{18}O_{aragonite}$  records of *C. iejimensis* provide unique data for reconstructing Holocene oceanographic 96 97 changes in the East China Sea. However, Yamamoto et al. (2009b) found no systematic ontogenetic variations in the  $\delta^{18}O_{aragonite}$  values of empty shells from surface sediment within 98 99 Daidokutsu cave, which is inconsistent with previous interpretations (Kitamura et al., 2007b; 100 Yamamoto et al., 2008).

In the present study, we re-evaluate the potential of  $\delta^{18}O_{aragonite}$  values of *C. iejimensis* as a tool for reconstructing Holocene oceanographic change. Based on our revised interpretation, we reconstruct climatic and hydrologic variability in the East China Sea during the last 7,000 years based on new data. The results provide information on the evolution of the mode of the climatic and hydrologic response of the East China Sea to the East Asian monsoon.

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## 107 **2. Study area and water-temperature conditions**

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The entrances to the submarine caves Daidokutsu and Shodokutsu (meaning *small cave* in Japanese) lie in about 20 m water depth on the fore-reef slope of Ie Island, off the Motobu Peninsula of Okinawa Island (Fig. 2a). Daidokutsu is 40 m long, dark inside, and deepens inward to its deepest point at 29 m below sea level, while Shodokutsu cave is more than 30 m long, dark inside, and has a horizontal cave floor (Fig. 2b and 2c). The floors of both caves are covered by calcareous mud (Hayami and Kase, 1993; Kitamura et al., 2003, 2007a). 115Kitamura et al. (2007b) and Kitamura and Yamamoto (2009) measured hourly water temperature data in Daidokutsu cave from 26 July 2003 to 6 July 2004, and from 27 August 2007 116 117to 19 September 2008 (Fig. 3), revealing seasonal changes from 29°C in August–September to 21°C in February. Yamamoto et al. (2009b) reported no significant difference in salinity and 118  $\delta^{18}O_{\text{seawater}}$  value inside the cave (salinity = 34.3 psu,  $\delta^{18}O_{\text{seawater}} = 0.3 \pm 0.1\%$ ) and at 30 m depth 119 outside the cave (salinity = 34.2 psu,  $\delta^{18}O_{seawater} = 0.4 \pm 0.1\%$ ) during early July, which is the 120121summer monsoon season. These data reveal that underground water did not flow into Daidokutsu 122 cave during the observation period and that patterns of seasonal change in water temperature and salinity within the cave are similar to those measured outside the cave at 30 m water depth (mean 123124monthly values) around Okinawa for the period 1906–2003 (Japan Oceanographic Data Center; J-DOSS;  $1^{\circ} \times 1^{\circ}$  grid cells, 26–27°N latitude and 127–128°E longitude) (Fig. 3a and 3d). We did 125not measure water conditions in Shodokutsu cave. 126

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#### 128 **3. Material**

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#### 130 *3.1. Shells*

Carditella iejimensis, which measures less than 3.5 mm in height and length (Fig. 5b), is a shallow infaunal suspension feeder with a shell of 100% aragonite. The species is only found in Daidokutsu and Shodokutsu caves (Fig. 2) among many caves in the Ryukyu Islands, Bonin Islands, Philippine Islands, Saipan, Palau, and Guam (Hayami and Kase, 1993, 1996). No previous study has investigated the ecology of the species. Our preliminary study found only four living specimens in Daidokutsu cave, whereas numerous living specimens were found on the surface sediment within Shodokutsu cave (Hayami and Kase, 1993).

138 Kitamura et al. (2007b) measured the  $\delta^{18}O_{aragonite}$  values of 30 empty and whole shells of *C*. 139 *iejimensis* picked from the >1 mm fractions of surface sediments (uppermost 5 cm of sediment, 140 representing the past 250 years) within Daidokutsu cave, and obtained a statistically significant 141 correlation between shell size and  $\delta^{18}O_{aragonite}$  (r = -0.55, p < 0.01). Yamamoto et al. (2008) added to this dataset the  $\delta^{18}O_{aragonite}$  values of 17 empty and whole shells collected from surface sediments within Daidokutsu cave (n = 47, r = -0.37, p < 0.05) (Fig. 5a), and calculated the following relationship:

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$$\delta^{18}O_{aragonite} = -0.10H - 0.63$$
 (1)

146 Using the above equation and the palaeothermometry equation presented by Grossman and Ku

147 (1986) and Goodwin et al. (2001), we have

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$$T(^{\circ}C) = 20.6 - 4.34 [\delta^{18}O_{aragonite/VPDB} - (\delta^{18}O_{water/VSMOW} - 0.2)].$$
 (2)

Kitamura et al. (2007b) and Yamamoto et al. (2008) estimated the  $\delta^{18}$ O-derived temperature for the 149150period during the precipitation of small shells (shell height of 1 mm) and the outer portions of large shells (3.0 mm or more from the umbo), yielding values of 23.8°C and 26.1°C, respectively. 151Given that these values correspond to monthly mean water temperatures during May and July, 152respectively, Kitamura et al. (2007b) and Yamamoto et al. (2008) proposed the following 153interpretation: the  $\delta^{18}$ O-derived temperature obtained from the shells of C. *iejimensis* indicates 154155water temperature between May and July of each year. Because the mixed surface layer was less 156than 2 cm thick during the deposition of calcareous mud in Daidokutsu cave (thereby indicating that the time-averaging of the fossil specimens was less than 100 years; Yamamoto et al., 2008), 157the  $\delta^{18}O_{aragonite}$  records of C. *iejimensis* provide high-resolution data for reconstructing 158millennial-scale oceanographic change in the East China Sea. A previous analysis of  $\delta^{18}O_{aragonite}$ 159values of C. iejimensis from three sediment cores from Daidokutsu cave (Cores 02, 04 and 05; Fig. 160 1616) showed no significant millennial-scale trend over the past 3,000 years, suggesting that both springtime temperature and  $\delta^{18}O_{\text{seawater}}$  at 30 m depth around the Okinawa Islands (Yamamoto et al., 1622008). 163

However, Yamamoto et al. (2009b) found no systematic ontogenetic variations in the  $\delta^{18}O_{aragonite}$  values of 19 empty shells, which is inconsistent with previous interpretations that the  $\delta^{18}O$ -derived temperature obtained from the shells of *C. iejimensis* indicates water temperature between May and July of each year (Kitamura et al., 2007b; Yamamoto et al., 2008). The authors proposed that the species may grow over several seasons, but did not speculate on the reason for 169 the size dependence of  $\delta^{18}O_{aragonite}$  in the empty shells.

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# 171 *3.2. Cored sediments*

As noted above, Yamamoto et al. (2008) obtained  $\delta^{18}O_{aragonite}$  values for *C. iejimensis* over the past 3,000 years, from three sediment cores recovered from Daidokutsu cave. To extend this earlier study, in the present study we collected additional measurements from whole shells within core 19 (6 cm in diameter and 233 cm in length), which was collected from Daidokutsu cave during September 2007 (Yamamoto et al., 2009a).

177The sediment in the core shows a fining-upward trend, and is divided into a lower, gray 178calcareous sand (233–178 cm depth) and an overlying gray calcareous mud (178–0 cm depth) (Fig. 6) (Yamamoto et al., 2009a). Based on <sup>14</sup>C age data, the cored sediments preserve a record of 179180fossils and sedimentation over the past 7,000 years. The sedimentation rate is estimated to be 28.6 cm/kyr between 233 and 211 cm depth, 49.2 cm/kyr between 211 and 153 cm depth, and 28.9 181 182cm/kyr between 153 and 0 cm depth (Fig. 6). Based on temporal changes in the abundance of 183debris derived from a red soil layer and temporal changes in cavernicolous bivalve assemblages, 184 Yamamoto et al. (2009a) proposed that the filling of cavities within the reefal foundations of the 185cave has continued over time, resulting in a progressive decrease in the flux of water between the 186 interior and exterior parts of the cave over at least the past 6,500 years.

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# 188 **4. Methods**

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190 4.1. Water conditions and analysis of  $\delta^{l8}O_{seawater}$ 

We used an Alec Electronics ACT-HR self-registering temperature–salinity meter to measure water temperature and salinity every hour in the innermost part of Shodokutsu cave, from 3 June 2007 to 31 July 2009. Unfortunately, records of water temperature within the cave were not obtained between 25 September 2007 and 19 September 2008 due to malfunction of the temperature–salinity meter (Fig. 2c). Water samples for oxygen isotope analysis were collected 196 within Shodokutsu cave and upon the reef slope at 30 m water depth on 2 July 2007. 197  $\delta^{18}O_{seawater}$  analyses were performed at the Geo-Science Laboratory in Nagoya, Japan, using a 198 Finnigan MAT delta S.  $\delta^{18}O_{seawater}$  values are reported relative to VSMOW; analytical precision 199 (1SD) was better than  $\pm 0.1\%$ .

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# 201 4.2. Analyses of $\delta^{18}O_{aragonite}$ of micro-shell specimens

To examine the cause of the size dependence of  $\delta^{18}O_{aragonite}$  values for empty shells of *C*. *iejimensis*, we compared  $\delta^{18}O_{aragonite}$  between living and empty specimens from Shodokutsu cave. For this purpose, samples of 5-cm-thick surface sediment were collected from the middle part of Shodokutsu cave (Fig. 2c) on 3 June 2009 and 29 July 2009.

Twenty-nine living individuals and 54 empty specimens of *C. iejimensis* were picked from the >1 mm fractions. We also picked 160 specimens of *C. iejimensis* from the >1 mm fractions of 1-cm-thick samples from core 19 (i.e., from 233 sediment samples).

All samples received no additional thermal or chemical treatment prior to stable isotope analysis.  $\delta^{18}O_{aragonite}$  values of whole shells of *C. iejimensis* were determined using a Finnigan MAT 251 mass spectrometer at Hokkaido University, Japan. Individual samples were reacted with 100% phosphoric acid at 60°C. Isotope ratios are reported relative to VPDB, and analytical precision (1SD) was better than ±0.08‰.

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215 5. Results
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217 5.1. Water conditions and \delta^{I8}O_{seawater}
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The results show that water temperature within Shodokutsu cave varied between 20°C and 219 29°C (Fig. 4a). Salinity within the cave showed seasonal change, ranging from 33.5 to 34.5 (Fig. 220 4b). The salinity values from 3 June 2007 to 24 September 2007 are the same as mean monthly 221 salinity measured at 30 m depth in the open sea (J-DOSS, Fig. 4b). On the other hand, the salinity 222 values from October 2008 to July 2009 are 0.4 lower than the mean monthly salinity. For the

measurement day (2 July 2007), we found no significant difference in salinity or  $\delta^{18}O_{\text{seawater}}$ 223between inside the cave (salinity = 33.9 psu,  $\delta^{18}O_{\text{seawater}} = 0.3 \pm 0.1\%$ ) and outside the cave at 30 m 224depth (salinity = 34.2,  $\delta^{18}O_{\text{seawater}} = 0.4 \pm 0.1\%$ ). The obtained  $\delta^{18}O_{\text{seawater}}$ -salinity relationship is 225similar to the linear relationship reported by Abe et al. (2009), who measured  $\delta^{18}O_{\text{seawater}}$  and 226salinity within surface water at Ishigaki Island (24°20.2'N, 124°9.8'E), Okinawa, Japan, every 10 227 days from 1998 to 2004 ( $\delta^{18}O_{\text{seawater}} = -10.3 (\pm 0.5) + 0.31 (\pm 0.01) S$ , where S = salinity). Based on 228the  $\delta^{18}O_{seawater}$  –salinity relationship and our salinity data,  $\delta^{18}O_{seawater}$  values within the cave are 229230estimated to be  $0.3 \pm 0.1\%$  throughout the year.

The mean daily temperatures within Shodokutsu cave were higher by <2°C than those in Daidokutsu cave during summer, while the water temperatures in other seasons were similar between the caves.

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# 235 5.2. $\delta^{i8}O$ values of living and empty specimens from surface sediments

The shell heights of living specimens from surface sediments in Shodokutsu cave range from 1.2 to 3.5 mm (Fig. 5b, Table 1). The  $\delta^{18}O_{aragonite}$  values of living specimens collected in early June and late July ranged from -1.0 to -1.5‰ and from -1.1 to -1.5‰, respectively (no significant difference). There was no significant correlation between shell size and  $\delta^{18}O_{aragonite}$ . The mean and 1 $\sigma$  deviation for  $\delta^{18}O_{aragonite}$  values were -1.28 ± 0.13‰.

241  $\delta^{18}O_{aragonite}$  values obtained for empty specimens ranged between -1.3 and -0.6‰, and show a 242 size dependence (n = 54, r = -0.42, p = 0.002) (Fig. 5c, Table 2). The obtained relationship is as 243 follows:

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$$\delta^{18}O_{aragonite} = -0.11H - 0.69$$
 (3)

where *H* is shell height. The slope of the regression line is similar to that obtained for empty shells from Daidokutsu cave (-0.10, Fig. 5a; Yamamoto et al., 2008).

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248 5.3.  $\delta^{18}O_{aragonite}$  values of specimens from cored sediments

We obtained the  $\delta^{18}O_{aragonite}$  values of 160 specimens from cored sediments (Table 3). Kitamura et al. (2007b; core 01) and Yamamoto et al. (2008; cores 02, 04, and 05) reported the  $\delta^{18}O_{aragonite}$  values of *C. iejimensis* from cored sediments from Daidokutsu cave. We sorted these previous data, and our data, into right and left valves (Fig. 7). The  $\delta^{18}O_{aragonite}$  values range from -2.1 to 1.7 ‰, with most being between -1.0 and -0.5‰ (Fig. 5d). The  $\delta^{18}O_{aragonite}$  records show that the long-term trend in  $\delta^{18}O_{aragonite}$  has been stable for the past 7,000 years, although anomalously heavy  $\delta^{18}O_{aragonite}$  values are observed at 6,500 and 5,000 cal. years BP (Fig. 7).

256 We found a statistically significant correlation between  $\delta^{18}O_{aragonite}$  and shell height for empty 257 shells from core 19 (Fig. 5d), as follows:

$$258 \qquad \delta^{18}O_{\text{aragonite}} = -0.35H + 0.06 \quad (\text{right valves}) \tag{4}$$

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$$\delta^{18}O_{\text{aragonite}} = -0.48H + 0.30 \quad (\text{left valves}) \tag{5}$$

The slopes of the regression lines for right and left valves (-0.35 and -0.48, respectively) are much steeper than those for empty shells within surface sediments from Daidokutsu and Shodokutsu caves. Most of the shells with heavy  $\delta^{18}O_{aragonite}$  values are less than 1.5 mm in height and are concentrated in periods dated between 6,500 and 5,000 cal. years BP.

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#### 265 **6.** Discussions

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## 267 6.1. Potential of C. iejimensis as an archive of Holocene palaeoceanography

268 There is no significant difference in salinity or  $\delta^{18}O_{seawater}$  between inside Shodokutsu cave 269 and outside the cave at 30 m depth, thereby indicating that  $\delta^{18}O_{aragonite}$  of *C. iejimensis* in the cave 270 is not influenced by the inflow of underground water.

In Shodokutsu cave, the mean water temperature during June is 24.9°C, and about 2°C lower than values during July (Fig. 4a). Based on the temperature equations for aragonitic molluscan shells presented by Grossman and Ku (1986) and Carré et al. (2005), a difference in water temperature of 2°C would cause a 0.5‰ variation in the  $\delta^{18}O_{aragonite}$  value of shell. However, in the living specimens, there is no significant difference in  $\delta^{18}O_{aragonite}$  among datasets obtained in early June and late July. Moreover, small individuals are found in late July. These findings are consistent with Yamamoto et al.'s (2009b) hypothesis that the species may be able to grow over several seasons, but are inconsistent with a previous interpretation that shell deposition begins in early April and ends in July (Kitamura et al., 2007b; Yamamoto et al., 2008). We propose that the size dependence of  $\delta^{18}O_{aragonite}$  of empty shells arises mainly because of the temperature dependence of mortality rate during the juvenile stage.

282Mortality in bivalves is caused by biotic and abiotic factors. Kase and Hayami (1994) 283suggested that predators are sparse in Shodokutsu cave and that their activities are infrequent, 284based on an analysis of the frequency of drilling predation on cavernicolous micro-bivalves. 285Hayami and Kase (1993) noted that C. iejimensis makes up more than 90% of living individuals 286from the bottom sediments of Shodokutsu cave. In fact, we found several tens of living individuals 287of the species, but few living individuals of other bivalve species. This dominance of C. iejimensis 288indicates that intraspecific competition, rather than interspecific competition, is the main cause of 289mortality in the species.

290The main abiotic agents of mortality are temperature, salinity, oxygen concentration, siltation, 291and waves (e.g., Dame, 1996). Based on our data and observations, we consider that temperature is 292the only important abiotic agent of mortality for C. iejimensis. Consequently, among all the agents 293of mortality for this species, we propose that temperature is especially important, and we consider that the size dependence of  $\delta^{18}O_{aragonite}$  of empty shells of *C. iejimensis* is caused mainly by the 294295high mortality rate of juveniles during exceptionally cold periods. During such periods, the 296probability of finding a dead immature individual (empty, small shell) within the death assemblage 297would be much higher than that during periods of normal water temperature. As a result, the proportion of specimens in the death assemblage with high  $\delta^{18}O_{aragonite}$  would be lower in the 298299subgroup of larger specimens.

300 If the above interpretation is correct, it is likely that shells with the heaviest  $\delta^{18}O_{aragonite}$  value 301 (-0.4‰) among the recent dataset grew under or close to the lower limit of growth temperature. 302 Many  $\delta^{18}O_{aragonite}$  values heavier than -0.4‰ are found within sediment deposited between 6,500 and 5,000 cal. years BP. Given the widely accepted view that the environmental tolerance of marine invertebrate species is likely to remain largely constant over a period of several thousand years, we consider that the  $\delta^{18}O_{aragonite}$  values heavier than -0.4‰ indicate unusually low temperature and enrichment of  $\delta^{18}O_{seawater}$  (dry conditions).

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# 308 6.2. Reconstruction of paleoceanographic variations over the past 7,000 years

We found no clear long-term trend in the  $\delta^{18}O_{aragonite}$  records during the last 7,000 years, 309 310except for frequent centennial-scale fluctuations. The absence of a long-term trend is consistent with the findings of Sun et al. (2005) and Lin et al. (2006), who estimated SST and  $\delta^{18}O_{seawater}$ 311based on an analysis of the Mg/Ca and  $\delta^{18}$ O values of the planktonic foraminifera G. ruber from 312313 deep-sea sediment cores recovered from the East China Sea (Figs. 2 and 6). The authors found no 314 clear insolation-related trend of decreasing intensity in the summer monsoon since the 315early-middle Holocene, which is characteristic of terrestrial records from monsoon regions (An, 316 2000; Fleitmann et al., 2003; Wang et al., 2005). This observation of a lack of any long-term trends in SST,  $\delta^{18}O_{\text{seawater}}$ , or sea surface salinity (SSS) over the past 6,000 years is also supported by 317318 other paleoceanographic studies (Ijiri et al., 2005) and simulations of paleoclimate (Kitoh et al., 319 2006; Oppo et al., 2007). Sun et al. (2005) and Lin et al. (2006) proposed that the absence of 320 long-term trends reflects the complexity of hydrographic processes, as hydrographic conditions 321were influenced by water originating from both high and low latitudes, which experienced 322 contrasting SST histories during the Holocene. Our study, as well as that of Lin et al. (2006), 323 detected no distinctive, consistent anomalies in the climatic and hydrologic variability associated 324 with the Pulleniatina Minimum Event.

As noted above, it is likely that  $\delta^{18}O_{aragonite}$  values heavier than -0.4‰ indicate occurrences of unusually low temperatures and dry conditions. Thus, our records show anomalously cool and dry events at about 6,300 and 5,550 cal. years BP. These events closely coincide with weak Asia summer monsoon events (Neff et al., 2001; Wang et al., 2005; Selvaraj et al., 2007), and the older event corresponds to one of three summertime cold events (which occurred at about 1,700, 2,300–4,600, and 6,200 cal. years BP) inferred from the planktonic foraminiferal record of the East
China Sea (Xiang et al., 2007). Therefore, we consider that unusually cool and dry events in the
East China Sea may have been related to weak summer monsoon events.

333 Abe et al. (2009) detected interannual variation of  $\delta^{18}O_{seawater}$  of surface water (+0.007‰year<sup>-1</sup>) based on continuous records from 1998 to 2004 at Ishigaki Island, and proposed 334 335 that both a reduction in wintertime northeasterly winds and summertime southwesterly winds caused a decrease in precipitation and consequent enrichment of  $\delta^{18}O_{seawater}$  around the island. This 336 337 interpretation is based on the fact that the vapor source for the island is evaporation from the 338 surface of the northern East China Sea in winter and the tropics in summer (Fig. 1). If these 339 processes are applicable to decadal- to centennial-scale variability in the intensity of East Asian monsoon, a weak summer monsoon may result in enrichment of  $\delta^{18}O_{seawater}$  around the present 340 341study area. A strong winter monsoon (anomalously low air temperature) may cause a decrease in  $\delta^{18}O_{\text{seawater}}$  by transporting large amounts of vapor and producing anomalous low SSTs, resulting in 342heavy  $\delta^{18}O_{aragonite}$  values. As noted above, although there has been considerable debate regarding 343 344 the nature of variability in the winter monsoon, Yancheva et al. (2007) reported that the strengths 345of the winter and summer monsoons are anti-correlated on a decadal time scale. In such a case, a 346 strong winter monsoon may have contributed to the anomalously cool and dry events in the East 347 China Sea at about 6,300 and 5,550 cal. years BP.

As noted above, a weak summer monsoon and strong winter monsoon occur during times of weak solar activity (Neff et al., 2001; Fleitmann et al., 2003; Lim et al., 2005; Wang et al., 2005; Xiao et al., 2006; Yancheva et al., 2007). Two anomalously cool and dry events in the East China Sea took place during the interval of Bond event 4, which is an ice-rafting event in the North Atlantic (Bond et al., 2001) (Fig. 7). These findings indicate that these cool and dry events may be related to weak solar activity.

It is noteworthy that in the present data, the occurrence of heavy  $\delta^{18}O_{aragonite}$  values appears to be obscured after 1,000 cal. years BP, even though a weak Asian summer monsoon event occurred during the Little Ice Age (Wang et al., 2005). Sixty  $\delta^{18}O_{aragonite}$  values fall between 6,500 and 5,500

cal. years BP, while 32 values fall in the period between 1,000 years BP and the present. Therefore, 357 it appears to be difficult to explain the absence of anomalously heavy  $\delta^{18}O_{aragonite}$  values after 1,000 358cal. years BP in terms of sampling intervals. In addition, Lin et al. (2006) reported that 359summertime  $\delta^{18}O_{seawater}$  values in the East China Sea during the early Holocene oscillated between 360 0.7% and -0.2%, but the magnitudes of the oscillations decreased to ~0.5\% in the middle 361362 Holocene and  $\sim 0.2\%$  during 2–1 ka (Fig. 7). These observations may indicate that the mode of the 363 East China Sea climatic and hydrological response to decadal- to centennial-scale variability in the 364 intensity of East Asian monsoon has varied during the past 7,000 years. An examination of the cause of this mode shift is required to further understand natural climate change around the East 365366 China Sea and East Asia.

367

#### 368 **7. Conclusions**

369

1. This study found no relationship between shell size and the  $\delta^{18}O_{aragonite}$  values of living specimens of the submarine cavernicolous micro-bivalve *C. iejimensis*, indicating that the species forms its shell over several seasons. Accordingly, the size dependence of the  $\delta^{18}O_{aragonite}$  values of empty shells is well explained by the high mortality rate of juveniles during exceptionally cold periods.

2. Based on our interpretation that the main cause of mortality in the species is exceptionally low temperature, we consider that the shells that formed at the lower limit of growth temperature represent the heaviest  $\delta^{18}O_{aragonite}$  values (-0.4‰). It is therefore likely that the  $\delta^{18}O_{aragonite}$  values heavier than -0.4‰ indicate occurrences of unusually low temperatures and enrichment of  $\delta^{18}O_{seawater}$  (i.e., dry conditions).

3. The obtained  $\delta^{18}O_{aragonite}$  values indicate no clear long-term trend in SST or  $\delta^{18}O_{seawater}$  in the 381 East China Sea during the middle–late Holocene, and reveal the occurrence of unusually cool and 382 dry events at around 6,300 and 5,550 cal. years BP. These events were probably related to decadal-383 to centennial-scale variability in the intensity of the East Asian summer monsoon, which was 384 linked in turn to weakened solar activity.

- 4. The absence of heavy  $\delta^{18}O_{aragonite}$  values after 1,000 cal. year BP indicates that the mode of the East China Sea climatic and hydrologic response to decadal- to centennial-scale variability in the intensity of East Asian summer monsoon has varied over the past 7,000 years.
- 388

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557

558 Figure Captions

559

560 Figure 1 Spatial distributions of observed mean surface wind (vectors) in East Asia for the periods

561 June–August and December–February. Modified from Kitoh (2005).

562

Figure 2 (a) Location maps of Daidokutsu and Shodokutsu submarine caves at Ie Island, off
Okinawa Japan, the locations of drill cores A7 (Sun et al., 2005; Xiang et al., 2007) and MD403
(Lin et al., 2006), and Dongge cave, China. NEC: North Equatorial Current; KC: Kuroshio Current.

566 (b) Simplified cross-section of Daidokutsu cave, showing the location of core sample 19. (c)

- 567 Simplified cross-section of Shodokutsu cave.
- 568

Figure 3 Observed temperatures within Daidokutsu cave. The observation period was from 26 July
2003 to 6 July 2004, and from 27 August 2007 to 19 September 2008.

571

Figure 4 (a) Observed temperatures within Shodokutsu cave. The observation period was from 3 June 2007 to 24 September 2007 and from 20 September 2008 to 31 July 2009. (b) Observed salinities within Shodokutsu cave. The observation period was from 3 June 2007 to 24 September 2007 and from 20 September 2008 to 31 July 2009. Seasonal variations in salinity (monthly mean values) at 30 m depth for the period 1906–2003 ( $1^{\circ}\times1^{\circ}$  grid cells; 26–27°N latitude and 127–128°E longitude). Error bars represent one standard deviation ( $1\sigma$ ).

578

Figure 5 Relationship between  $\delta^{18}$ O and the shell height of *C. iejimensis*. Error bars represent one standard deviation of a single  $\delta^{18}$ O analysis. Note that the vertical axes are inverted so that higher temperatures (lower  $\delta^{18}$ O) are toward the top. (a) Empty shells from surface sediment within Daidokutsu cave; (b) living shells from surface sediment within Shodokutsu cave; (c) empty shells from surface sediment within Shodokutsu cave; (d) empty shells from core 19 within Daidokutsu cave.

Figure 6 Stratigraphic columns compiled for cores 01, 02, 04, 05, and 19 recovered from within 586587Daidokutsu cave, showing the stratigraphic distribution of Daidokutsu pumice. Also shown is a 588map of the distribution of surface sediment facies within the cave. Facies 1, gray calcareous sand; Facies 2, gray calcareous mud; Facies 3, calcareous sand containing the skeletons of partly 589590encrusted coralline sponges.

591

Figure 7 Comparison of  $\delta^{18}$ O values of C. *iejimensis* from cored samples (present day) with the 592 $\delta^{18}O_{seawater}$  record from the East China Sea (Lin et al., 2006), stalagmite  $\delta^{18}O$  record from Dongge 593594cave in southeast China (Wang et al., 2005), and record of ice-rafted debris from the North Atlantic (Bond et al., 2001). Note that the axes are inverted so that higher temperatures (lower  $\delta^{18}$ O) are 595596toward the top. 597

598Table 1 Results of oxygen isotope analyses of living specimens of C. iejimensis from Shodokutsu 599cave.

600

Table 2 Results of oxygen isotope analyses of empty specimens of C. iejimensis from surface 601sediments in Shodokutsu cave. 602

603

Table 3 Results of oxygen isotope analyses of empty specimens of C. iejimensis from core 19 in 604 605Daidokutsu cave.



Yamamoto et al Fig. 1



# Yamamoto et al Fig. 2



Yamamoto et al Fig. 3



Yamamoto et al. Fig. 4





Yamamoto et al. Fig 6



Yamamoto et al Fig. 7

collected in		collected in		
2009/6/3			2009/7/29	
Height (mm)	δ <sup>18</sup> Ο		Height (mm)	δ <sup>18</sup> Ο
3.5	-1.25		2.7	-1.32
3.3	-0.98		2.6	-1.33
2.9	-1.37		2.5	-1.25
2.7	-1.28		2.3	-1.40
2.5	-1.46		2.3	-1.18
2.1	-1.31		2.2	-1.50
1.7	-1.51		2.2	-1.41
1.7	-1.25		2.2	-1.25
1.5	-1.22		2.2	-1.17
1.5	-1.08		2.1	-1.44
1.4	-1.34		2.1	-1.17
1.3	-1.36		2.0	-1.14
1.3	-1.22		1.9	-1.29
1.2	-1.25		1.6	-1.30
1.2	-1.08			

# Yamamoto et al. Table 1

Height	δ <sup>18</sup> 0	Height	δ <sup>18</sup> 0
(11111)		(1111)	
1.2	-1.02	2.2	-1.16
1.2	-0.76	2.2	-1.03
1.2	-0.69	2.2	-0.99
1.3	-0.96	2.2	-0.88
1.3	-0.91	2.3	-1.03
1.3	-0.85	2.3	-1.00
1.3	-0.76	2.3	-0.86
1.4	-1.03	2.4	-1.33
1.4	-0.95	2.4	-1.07
1.4	-0.91	2.4	-0.80
1.4	-0.91	2.5	-1.01
1.4	-0.77	2.5	-0.95
1.4	-0.65	2.5	-0.90
1.5	-0.87	2.6	-0.96
1.5	-0.84	2.6	-0.95
1.5	-0.80	2.7	-1.20
1.5	-0.79	2.8	-1.17
1.5	-0.63	2.8	-0.80
1.5	-0.62	2.9	-1.03
1.5	-0.59	2.9	-0.99
1.6	-0.82	3.0	-1.13
1.6	-0.64	3.0	-1.10
1.7	-0.78	3.0	-0.95
2.1	-1.14	3.0	-0.93
2.1	-1.07	3.0	-0.91
2.1	-0.85	3.0	-0.76
2.2	-1.21	3.5	-0.75

Yamamoto et al. Table 2

Та	bl	е	3
		_	_

$\begin{array}{c c} \text{Depth} \\ (cm) \end{array} Valve \begin{array}{c} \text{Height} \\ (mm) \end{array} \delta^{18} O \end{array} \xrightarrow[cm) \end{array} \begin{array}{c} \text{Depth} \\ (cm) \end{array} Valve \begin{array}{c} \text{Height} \\ (cm) \end{array} \delta^{18} O \end{array} \xrightarrow[cm) \end{array} \begin{array}{c} \text{Depth} \\ (cm) \end{array} Valve \begin{array}{c} \text{Height} \\ (cm) \end{array} \delta^{18} O \end{array}$	Depth Valve Height δ (cm) Valve (mm) δ	<sup>18</sup> 0
3.5 R 2.5 -0.73 118.5 R 1.9 -1.02 154.5 R 2.1 -0.88	185.5 L 2.5 -0	).59
4.5 L 3.3 -0.84 119.5 R 2.9 -0.59 155.5 L 1.0 1.08	186.5 L 1.1 -0	08.0
17.5 L 3.0 -2.10 120.5 R 1.8 -0.44 156.5 R 1.1 -0.63	186.5 R 1.2 0	.30
18.5 R 2.0 -0.64 121.5 R 2.1 -1.27 156.5 L 1.7 -0.84	187.5 R 2.5 -0	.88
26.5 L 3.0 -0.69 122.5 L 2.8 -0.71 156.5 L 2.4 -0.99	188.5 L 2.5 -0	).84
33.5 R 2.3 -0.24 122.5 L 3.0 -0.65 157.5 L 1.4 -0.69	189.5 R 2.3 -1	.05
37.5 L 2.5 -0.74 124.5 R 1.3 -0.68 158.5 L 1.3 -0.81	190.5 R 1.8 -0	).70
38.5 L 1.4 -0.48 124.5 R 2.9 -0.42 158.5 L 1.4 -0.87	190.5 R 2.6 -0	).73
49.5 R 1.1 0.03 125.5 R 1.2 0.04 159.5 R 2.5 -1.14	191.5 L 1.7 -0	).97
50.5 R 2.7 -0.75 126.5 R 1.4 -0.86 159.5 R 2.8 -0.72	192.5 R 1.6 -0	).65
57.5 R 1.2 -0.40 126.5 R 2.7 -0.61 160.5 L 2.7 -0.83	192.5 R 2.8 -0	).89
59.5 R 1.0 0.16 127.5 R 1.9 -1.07 162.5 R 1.5 -0.06	193.5 R 2.2 -1	.12
64.5 L 2.2 -0.73 127.5 L 2.3 -0.79 163.5 L 1.1 0.39	194.5 L 1.4 -0	).78
67.5 R 1.9 -1.17 127.5 R 3.0 -0.64 164.5 R 1.2 0.08	194.5 L 1.5 -0	).94
69.5 L 2.9 -0.59 128.5 L 3.0 -0.93 165.5 R 1.0 0.89	195.5 L 2.5 -0	).84
70.5 R 2.7 -0.67 130.5 R 1.9 -0.69 167.5 R 2.6 -0.76	196.5 R 1.3 -0	).29
78.5 R 1.3 0.43 130.5 R 2.3 -0.95 168.5 L 1.4 -0.40	196.5 L 2.3 -0	).74
84.5 R 2.0 -1.03 132.5 L 1.1 0.15 169.5 L 1.6 -0.70	197.5 L 2.3 -0	).96
85.5 L 1.4 -1.86 133.5 L 2.1 -0.78 170.5 L 2.2 -0.89	198.5 L 2.0 -0	).85
85.5 R 2.5 -0.87 134.5 R 2.4 -0.77 171.5 L 0.7 0.29	199.5 L 1.1 0	.65
89.5 L 2.5 -0.69 135.5 L 2.1 -0.56 171.5 R 2.2 -0.97	199.5 R 2.0 -2	2.00
90.5 R 1.2 -0.81 136.5 R 1.0 -0.25 171.5 R 2.7 -0.74	200.5 L 2.3 -0	08.0
90.5 L 2.2 -1.02 137.5 R 1.2 -0.72 172.5 R 1.7 -0.59	201.5 L 1.3 -0	).37
91.5 R 1.7 0.14 137.5 R 3.0 -0.74 173.5 R 2.4 -0.90	202.5 R 1.7 -0	).74
103.5 R 2.6 -0.90 137.5 R 3.1 -0.55 174.5 L 1.2 -0.61	203.5 L 1.0 0	.40
104.5 L 2.8 -0.91 138.5 R 1.1 -0.35 174.5 R 2.2 -0.83	203.5 L 2.2 -0	).85
106.5 L 1.2 0.21 139.5 R 2.0 -0.66 175.5 L 1.4 -0.75	204.5 R 1.0 1	.34
107.5 L 3.1 -0.89 143.5 L 2.0 -0.79 177.5 R 1.2 -0.45	204.5 R 2.2 -0	).93
109.5 R 2.7 -0.80 146.5 R 1.7 -0.53 178.5 R 1.3 -0.62	205.5 L 1.1 1	.72
110.5 L 1.4 -1.08 146.5 L 2.4 -1.06 178.5 R 1.4 -0.97	205.5 L 1.4 -0	.88
111.5 L 2.3 -0.84 147.5 R 1.0 -1.17 178.5 R 1.9 -1.00	206.5 L 1.4 0	.20
112.5 R 1.9 -0.82 147.5 L 1.2 0.12 179.5 L 1.2 -0.71	208.5 L 2.2 -0	08.0
113.5 R 2.0 -0.53 147.5 R 2.8 -0.56 179.5 R 1.2 -0.44	209.5 L 1.8 -1	.11
114.5 L 1.2 -0.61 148.5 R 2.4 -0.88 179.5 R 1.3 0.54	210.5 R 1.5 -0	).64
114.5 L 1.3 -0.22 149.5 R 1.7 -0.57 179.5 R 2.0 -0.96	212.5 L 1.5 -0	).63
114.5 R 1.6 -0.84 150.5 L 0.9 0.75 179.5 R 2.2 0.19	215.5 R 2.2 -0	).76
115.5 L 2.2 -0.71 150.5 R 2.2 -0.91 180.5 L 2.8 -0.88	216.5 L 1.1 1	.03
117.5 L 1.4 -0.79 150.5 R 2.9 -0.71 181.5 L 1.5 -0.65	217.5 L 2.1 -0	).72
117.5 R 1.5 -0.48 151.5 L 2.0 -0.90 181.5 R 1.5 -0.60	218.5 R 2.4 -0	).77
<u>118.5 L 1.7 -0.59 152.5 L 2.7 -0.67 183.5 L 1.4 -1.70</u>	<u>226.5 R 1.3 -0</u>	).49

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