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Reconstruction of ocean environment time series since the late nineteenth century using sclerosponge geochemistry in the northwestern subtropical Pacific

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Abstract

The geochemistry of calcifying marine organisms is an excellent proxy for reconstructing paleoceanographic history, but studies of hypercalcified demosponges (sclerosponges) are considerably fewer than those of corals, foraminifers, and bivalves. For this study, we first generated near-annual resolved stable carbon and oxygen isotope (δ^{13}C and δ^{18}O) and element/Ca ratios (Sr/Ca, Ba/Ca, Pb/Ca, U/Ca) time series for 1880–2015 from sclerosponge samples (*Acanthochaetetes wellsi*) collected at Miyako Island and Okinawa Island in the Ryukyu Islands of southwestern Japan. The δ^{13}C records exhibited a typical variation of anthropogenically derived Suess effects, demonstrating that the rates of decrease of −0.0043‰/year before 1960 and − 0.024‰/year after 1960 in the northwestern subtropical Pacific were respectively similar to and about 1.4 times higher than those of the Caribbean Sea in the tropical Atlantic. Spectral analysis of the δ^{18}O time series revealed significant periodicity of approximately 2, 3, 6.5, 7–10, and 20–30 year/cycle, indicating that sea surface conditions in the southern Ryukyu Islands had been dominated by interannual and decadal variations in temperature and seawater δ^{18}O since the late nineteenth century. The Sr/Ca and U/Ca ratios for the species *A. wellsi* (high-Mg calcite) might not be a robust proxy for seawater temperatures, unlike *Astrosclera willeyana* and *Ceratoporella nicholsoni* sclerosponges (aragonite). An evident increasing Pb/Ca trend after 1950 found in the samples is probably attributable to Pb emissions from industrial activities and atmospheric aerosols in eastern Asian countries. The Ba/Ca variations differ greatly among sampling sites, which might be attributable to the respective local environments. This evidence demonstrates that more high-resolution age determinations and geochemical profilings enable delineation of secular variations in ocean environments on annual and interannual timescales. Results of our study suggest that if sclerosponges living in deeper ocean environments are collected, spatial and vertical oceanographic variations for the last several centuries will be reconstructed along with coral proxy records.

Keywords: Sclerosponge, *Acanthochaetetes wellsi*, Oxygen and carbon isotopic composition, Minor and trace elements, Isotope equilibrium, Paleoenvironmental proxy, Ryukyu Islands

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1 Introduction

Hypercalcified demosponges ("sclerosponges" hereinafter), members of the earliest branching metazoan taxon (Porifera), persisted as dominant reef-building marine organisms throughout the Phanerozoic (Vacelet 1985; Reitner and Engeser 1987; Wood 1990; Reitner 1992). The genus *Acanthochaetetes* Fischer 1970 is present in the fossil record from the Early Cretaceous (Reitner 1992). In modern coral reefs, sclerosponges can be found in cryptic niches of dark environments, unlike zooxanthellate scleractinian corals (Wörheide 1998). Sclerosponges, commonly mushroom-shaped, deposit a calcium carbonate skeleton at a very slow growth rate of mostly fewer than about 2 mm/year; they can grow for up to several hundred years (Benavides and Druffel 1986; Böhm et al. 1996; Reitner and Gautret 1996; Wörheide et al. 1997; Wörheide 1998; Swart et al. 2002; Fallon et al. 2003; Grottoli et al. 2010).

To elucidate climate change and global warming, environmental proxies such as trees, sediments, and corals are strongly needed for demonstrating temperature variations over the last millennia (PAGES 2k Consortium 2013; Abram et al. 2016). The stable carbon and oxygen isotope records ($\delta^{13}C$ and $\delta^{18}O$) of sclerosponges are a proxy for reconstructing dissolved inorganic carbon $\delta^{13}C$ of seawater (e.g., Böhm et al. 1996; Swart et al. 2010) and seawater temperature and salinity (e.g., Wörheide 1998; Böhm et al. 2000; Moore et al. 2000; Rosenheim et al. 2005a) for the past. Earlier studies have revealed that similarly to corals, the $\delta^{13}C$ and $\delta^{18}O$ values of sclerosponges respectively reflect the increase of fossil fuel CO_2 in seawater (Böhm et al. 2002; Swart et al. 2010) and interannual climate variation (Swart et al. 2002; Grottoli 2006; Grottoli et al. 2010; Wu and Grottoli 2010). Other minor and trace elements and isotopes can be a proxy for monitoring ocean environments and chemistry (Lazareth et al. 2000; Fallon et al. 2005; Rosenheim et al. 2005b; Ohmori et al. 2014). Particularly, some studies have shown that sclerosponge strontium/calcium (Sr/Ca) presents some potential for determining seawater paleotemperatures (e.g., Rosenheim et al. 2004; Waite et al. 2018). Its fidelity as a paleoenvironmental proxy has been verified by evaluation and calibration studies of sclerosponges (Böhm et al. 2000; Rosenheim et al. 2005b, 2009; Asami et al. 2020a).

Importantly, sclerosponges can live in environments with less sunlight (e.g., reef caves and overhangs) and at greater water depths of up to several hundred meters in Indo-Pacific tropical regions (e.g., Hartman 1980) and in the Atlantic and Caribbean regions (e.g., Druffel and Benavides 1986; Böhm et al. 1996), unlike zooxanthellate corals and giant clams living in shallow coral reefs. For that reason, their skeletal chemistry can be useful for elucidating the vertical variations of physical and chemical parameters of seawater and for generating their several-centuries-long time series that overlap with and extend beyond instrumental oceanographic data. However, sclerosponge-based climate reconstructions over a hundred years are extremely few relative to those of corals. Moreover, they have not yet been extracted from the northwestern subtropical Pacific.

We first generated > 100-year-long multiple proxy records of $\delta^{13}C$, $\delta^{18}O$, and elements/Ca ratios, Sr/Ca, barium/Ca (Ba/Ca), lead/Ca (Pb/Ca), and uranium/Ca (U/Ca), in living sclerosponges (*Acanthochaetetes wellsi* Hartman and Goreau 1975) collected from the Ryukyu Islands of southwestern Japan. Based on radiocarbon and $\delta^{13}C$ results, near-annually resolved time series of the geochemical records were established for the last one hundred years and more, which are expected to be useful for paleoclimate archives such as corals. The study results emphasize that the geochemical time series show secular decade-scale variations of ocean environments in the subtropical Northwest Pacific.

2 Materials and methods

2.1 Study site and modern environment

The study sites are located off the southern coast of Miyako Island and at Manza Cape around the western coast of Okinawa Island in the Ryukyu Islands of southwestern Japan (Fig. 1). The northward-flowing warm Kuroshio current (the western North Pacific current) passes along the Ryukyu Islands, which has allowed the development of coral reefs with high biodiversity in the islands. The Kuroshio current, the strong northwestern component of the subtropical North Pacific gyre, advects a large amount of heat from the tropics to northern mid-latitudes. The climate is subtropical, with atmospheric temperatures of 17.6–28.5 °C and 16.5–28.6 °C and an annual mean of 23.4 °C and 22.8 °C at Miyako and Okinawa Islands for 1961–2015 (Japan Meteorological Agency; http://www.jma.go.jp/jma/indexe.html). The mean annual precipitation is greater than 2000 mm/year, with a rainy season (May–June) and typhoon season (August–September) at the two sites. Sea surface temperatures (SST) with a 2°–2° grid for 1961–2015, available at the study sites (NOAA NCDC ERSST ver. 3b, http://iridl.ldeo.columbia.edu/), show variation of 22.9–29.1 °C and 21.8–28.9 °C with annual means of 26.1 °C and 25.4 °C in Miyako (23°–25° N, 125°–127° E) and Okinawa (25°–27° N, 127°–129° E), respectively. According to monthly average data available from the Japan Oceanographic Data Center (JODC) for 1906–2003 (http://www.jodc.go.jp/), the sea surface salinity (SSS) varies 34.3–34.9 and 34.1–35.0, respectively, with annual means of 34.7 and 34.6 at Miyako (24°–25° N, 125°–126° E) and Okinawa (26°–27° N, 127°–128° E).

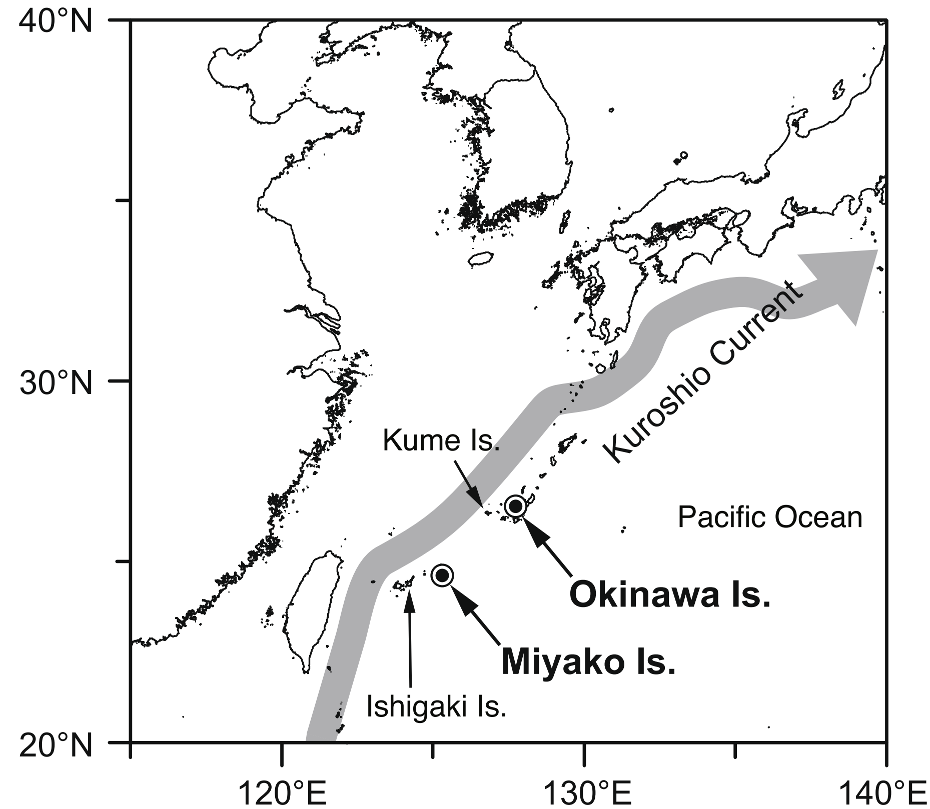

Fig. 1 Map of eastern Asian and the northwestern Pacific regions showing the locations of Miyako Island and Okinawa Island (this study) and Ishigaki Island (Abe et al. 2009; Hirabayashi et al. 2017) and Kume Island (Ohmori et al. 2014) discussed in the text

2.2 Samples

The sclerosponges (*Acanthochaetetes wellsi*) examined in this study were growing on the wall in a dark submarine crevasse (Miyako Island) and cave (Okinawa Island) of outer reef edges. In June 2012, a living sclerosponge sample (SMJ-2-S1) was collected at 10.2 m water depth from Miyako Island (24° 42′ 24.2″ N, 125° 19′ 02.6″ E) (Figs. 1 and 2). The sample SMJ-2-S1 is 7 cm in height, 21 cm in length of the major axis, and 18 cm in length of the minor axis. In October 2015, a living sclerosponge sample (MNZ-S1) was collected at 2 m water depth from Okinawa Island (26° 30′ 14.9″ N, 127° 50′ 52.5″ E) (Figs. 1 and 2). The sample MNZ-S1 is 9 cm in height, 19 cm in length of the major axis, and 18 cm in length of the minor axis. The skeletal surfaces (tissue layer zones) of samples were mostly light brown or orange, which has a thickness of less than 2 mm. The samples were well rinsed ultrasonically with milli-Q water (resistance = 18.2 MΩ·cm; Millipore Corp.). They were then dried.

The sclerosponge samples were slabbed to 3 mm thickness parallel to the axis of skeletal growth. X-radiographs were taken under exposure conditions of 30 kV, 3 mA, and a distance of 53 cm between a lamp and a film with exposure time of 15–60 s, using soft X-ray film shooting equipment (Soft X-Ray Sofron SRO-405A; Soken Co., Ltd.) and a computed radiographic reader (FCR PRIMA T2; FUJI FILM Corp.) at the University of the Ryukyus (UR). The X-radiographic images of the samples showed skeletal banding comprising alternating high-density (dark) and low-density (light) bands (Fig. 2). Powder subsamples for geochemical analyses were taken every 1 mm along the skeletal growth direction manually using a pre-cleaned diamond drill, roughly corresponding to the annual sampling resolution. To determine the time series of geochemical records, radiocarbon values ($\Delta^{14}C$, ‰) of 13 subsamples along the growth direction of the SMJ-2-S1 sample were analyzed by Paleo-Lab Co. Ltd., Japan (Supplementary Table S1). The data are reported in terms of deviation from the activity of the pre-industrial wood standard (Stuiver and Polach 1977), which is corrected for isotope fractionation to a $\delta^{13}C$ value of −25.0‰. In addition, powder subsamples were taken from different portions of the skeleton surface to evaluate within-skeleton variations in the geochemistry, following the method described by Asami et al. (2020a).

2.3 Skeletal structures

To check the mineralogy and preservation state of the skeleton, the fragments were examined using X-ray

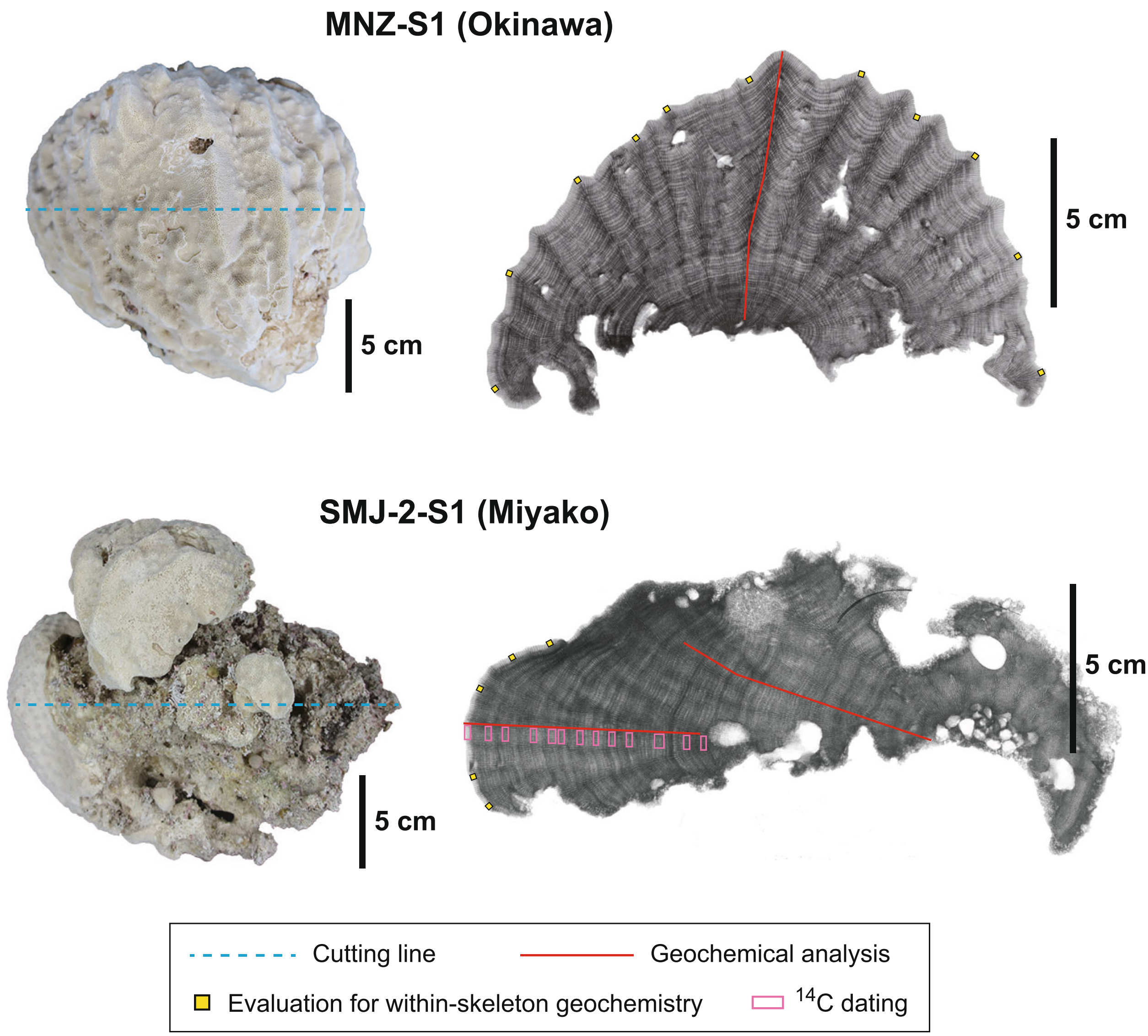

Fig. 2 Photos and X-radiographic positive images of the two modern sclerosponges show skeletal growth bandings comprising alternating high-density (dark) and low-density (light) bands. Slab samples were extracted by cutting along the blue lines. Geochemical analyses were conducted along sampling transects for generating the time series (red lines) and at selected portions of the coeval skeleton for evaluating within-skeleton variation (yellow squares). Radiocarbon measurements were taken at selected skeletal portions (pink squares)

diffraction (XRD) analysis and scanning electron microscopy (SEM), following the methods reported by Asami et al. (2015, 2020a). At UR, XRD analysis was conducted using a multipurpose XRD device (RINT Ultima/PC; Rigaku Corp.) to test for mineralogy. After powdered samples were mounted on well-cleaned glass slides with HCl, ethanol, and milli-Q water, they were analyzed using Cu Kα radiation (40 kV, 30 mA) by scanning from 20° to 37° at a 2θ angle at 0.02° steps. A microscope (TM3030; Hitachi High-Tech. Corp.) was used for SEM imaging of the skeleton. Also, XRD analysis revealed that the *A. wellsi* samples consisted of high-Mg calcite with 20.1 mol% Mg for SMJ-2-S1 and 17.3 mol% Mg for MNZ-S1, consistent with data reported from earlier studies (Reitner and Engeser 1987; Reitner and Gautret 1996; Grottoli et al. 2010). The SEM observations revealed that skeletal aspects of our sclerosponge samples (Fig. 3) closely resemble those found in earlier studies (Reitner and Engeser 1987; Reitner et al. 1997; Gilis et al. 2013), clearly showing a typical macrostructure organization with vertical calicles subdivided by horizontal tabulae and a microstructure of high-Mg calcite skeleton. Additionally, results confirmed that the samples had a well-preserved skeleton without physical evidence of diagenetic alteration, such as overgrowth of inorganically precipitated secondary cement, traces of dissolution,

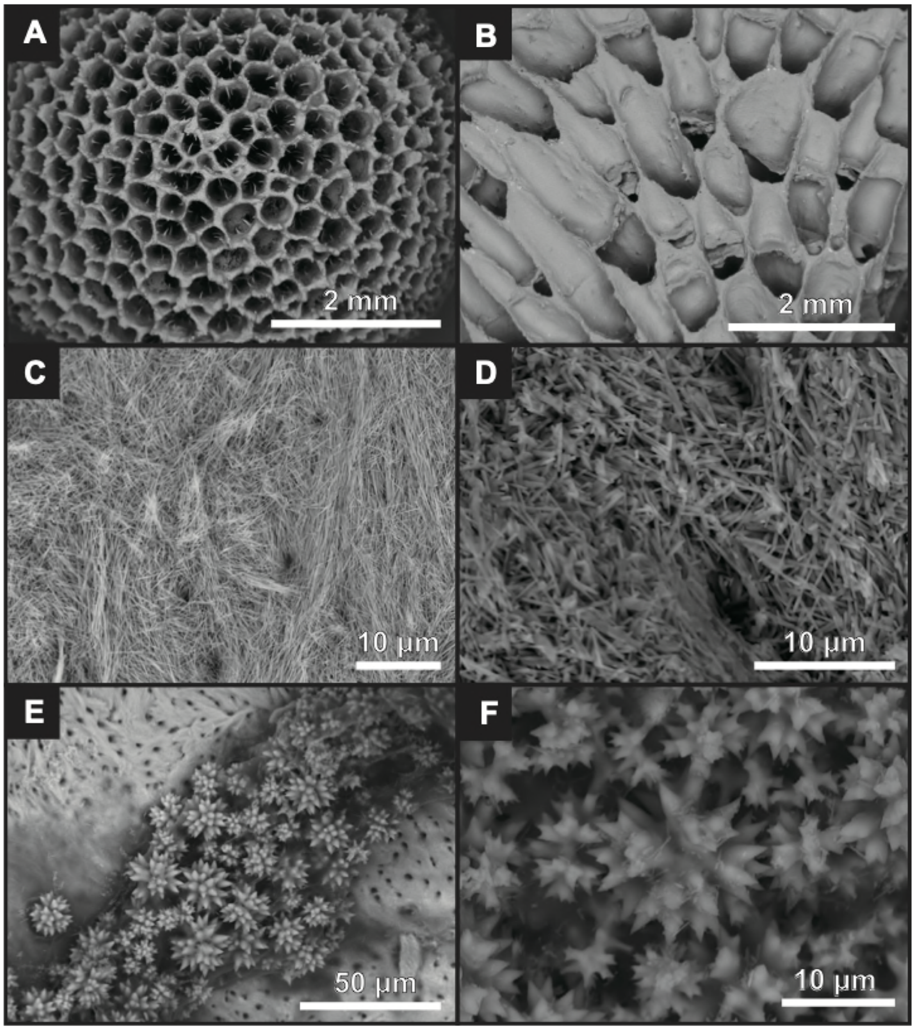

Fig. 3 Scanning electron micrograph images of sclerosponges. A well-preserved pristine calcite skeleton is visible. **A** and **B** Macrostructural organization of high-Mg calcite skeleton with vertical calicles subdivided by horizontal tabulae. **C** and **D** Lamellar microstructure of high-Mg calcite skeleton, the secondary structure of the tabulae and coatings of the tubes, randomly oriented single crystals. **E** and **F** Spicular skeleton and spiraster microscleres

or bioerosion. Consequently, these results indicate that *A. wellsi* samples used for this study can retain pristine calcite skeleton with original values of isotope composition and elemental concentrations.

2.4 Stable isotope analyses

Stable carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) isotope analyses of calcium carbonate (0.14–0.16 mg) were conducted using a continuous-flow isotope ratio mass spectrometer attached to a Gasbench II and a GC-PAL auto-sampler (Delta V Advantage; ThermoFisher Scientific Inc.) at UR, following the method presented by Asami et al. (2015, 2020a). Isotopic ratios were reported in conventional δ notation relative to Vienna Pee Dee Belemnite (VPDB). External precision (1σ, N = 190) throughout the entire analysis was ± 0.04‰ and ± 0.06‰ for $\delta^{13}C$ and $\delta^{18}O$. Accuracy of the measurements of sclerosponge samples was evaluated based on replicates of IAEA CO-1 calcite standard, yielding average $\delta^{13}C$ and $\delta^{18}O$ values of 2.46 ± 0.05‰ and −2.41 ± 0.06‰ (N = 27), which respectively show excellent agreement with recommended values.

2.5 Elements/Ca analyses

Elements/Ca ratios (Sr/Ca, Ba/Ca, Pb/Ca, U/Ca) were analyzed using an inductively coupled plasma-mass spectrometer (XSeries II; ThermoFisher Scientific Inc.) at UR, fundamentally following the method reported by Asami et al. (2009, 2020a). Each sclerosponge powder sample (approx. 0.15 mg) was dissolved in 4.5 mL of 0.5 mol/L high-purity HNO_3 diluted with ultrapure Milli-Q water. Each seawater sample of 125 µL was diluted 80 times with high-purity HNO_3 and ultrapure Milli-Q water. Internal standard elements (Sc, Y, and Yb) were added to all the solutions to produce equal concentrations to control matrix effects and to correct for instrumental noise. Solutions were analyzed for ^{43}Ca, ^{44}Ca, ^{45}Sc, ^{87}Sr, ^{88}Sr, ^{89}Y, ^{137}Ba, ^{138}Ba, ^{172}Yb, ^{208}Pb, and ^{238}U. Measurements were conducted in triplicate. Calibrations of the five gravimetric standard solutions yielded high correlation coefficients of $r > 0.99998$ for Ca and Sr and of $r > 0.99995$ for Ba, Pb, and U. A reference solution, matched gravimetrically to the Ca concentration of the average sclerosponge sample solutions, was measured at intervals of three samples to correct instrumental drift. Based on replicate measurements of the solution of carbonate reference material JCp-1 (Okai et al. 2002), external precisions for Sr/Ca, Ba/Ca, Pb/Ca, and U/Ca determinations were better than 0.30%, 1.7%, 4.1%, and 0.77% relative standard deviations (N = 166). The measurement values of samples were calibrated using the widely accepted values of Hathorne et al. (2013).

3 Results and discussion

3.1 Within-skeleton variations and comparison with equilibrium calcite

The within-skeleton variations of geochemical components in an *A. wellsi* specimen were estimated at the coevally precipitated skeleton (Fig. 4A). The standard deviations (1σ) of geochemical analyses of SMJ-2-S1 and MNZ-S1 samples were, respectively, ± 0.049‰ and ± 0.039‰ for $\delta^{13}C$, ± 0.056‰ and ± 0.093‰ for $\delta^{18}O$, ± 0.061 and ± 0.045 mmol/mol for Sr/Ca (RSD, 3.9% and 3.6%), ± 0.54 and ± 0.17 μmol/mol for Ba/Ca (RSD, 7.7% and 9.9%), ± 172 and ± 253 nmol/mol for Pb/Ca (RSD, 8.1% and 7.8%), and ± 0.021 and ± 0.012 μmol/mol for U/Ca (RSD, 8.5% and 4.3%). The variations of sclerosponge geochemical records are small among all skeletal portions, with no marked trend, except for Ba/Ca and Pb/Ca, which accord well with those of *Astrosclera willeyana* sclerosponge reported by Asami et al. (2020a). These results reflect that within-skeleton variations in $\delta^{13}C$, $\delta^{18}O$, Sr/Ca, and U/Ca are slight for the *A. wellsi* species.

To evaluate the fidelity of *A. wellsi* as a paleoenvironmental proxy, we compared our data with the $\delta^{13}C$ and $\delta^{18}O$ values of calcite that is precipitated inorganically in isotopic equilibrium with seawater at the sclerosponge growth site: so-called equilibrium calcite. The $\delta^{13}C$ values of the equilibrium calcites were estimated using DIC $\delta^{13}C$ of seawater, the carbon-isotope fractionation factors among CO_2 species (Zhang et al. 1995), and the calcite–HCO_3^- enrichment factor (1.0 ± 0.2‰; Romanek et al. 1992). Because of the lack of data on DIC $\delta^{13}C$ of seawater around the study sites, the annual mean DIC $\delta^{13}C$ value is assumed to fall within a range of 0.58‰ in November to 0.78‰ in August, estimated respectively using two significant relations from west of Okinawa Island (Takayanagi et al. 2012) and southeast of Okinawa Island (Suzuki et al. 2009). Consequently, the $\delta^{13}C$ values of the equilibrium calcites are expected to be roughly 1.6–2.2‰ (Fig. 4B). The $\delta^{18}O$ values were calculated using an equation derived from laboratory synthesis experiments of calcite (Kim and O'Neil 1997), with correction for $MgCO_3$ in calcite (Tarutani et al. 1969). The annual mean seawater temperatures of 26.3 °C for 2010–2012 in Miyako Island and 25.2 °C for 2013–2015 in Okinawa Island were used from the ERSST v3b gridded dataset. The seawater $\delta^{18}O$ values of 0.35–0.52‰ and 0.26–0.56‰ were estimated respectively for Miyako Island and Okinawa Island from

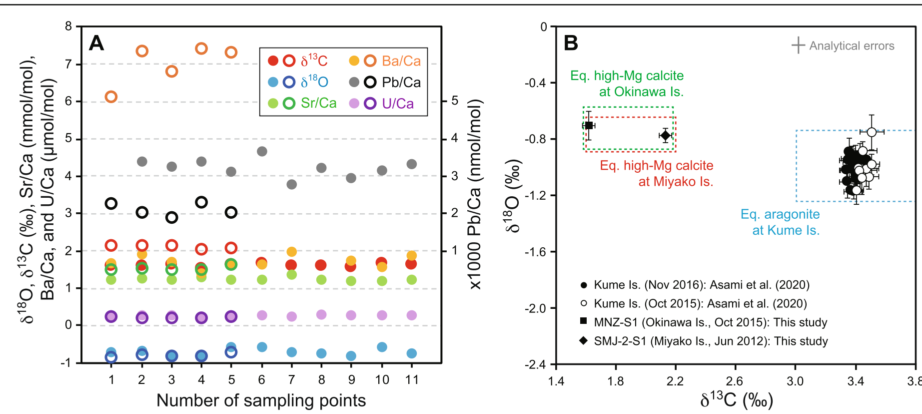

Fig. 4 **A** Within-skeleton variations in the geochemistry of the SMJ-2-S1 (open circles) and MNZ-S1 (dotted circles) samples. The Ba/Ca and Pb/Ca data at the respective sampling point of X = 8 and X = 1 for the MNZ-S1 sample were evaluated as outliers based on the Smirnov-Grubbs test (p < 0.025). **B** Scatter plot of $\delta^{13}C$ and $\delta^{18}O$ averages with 1σ standard deviations of coeval skeletal portions in individual samples. Red and green areas are shown respectively as estimated ranges of isotopically equilibrium calcite precipitated at the study sites of Miyako and Okinawa Islands. For comparison, aragonite sclerosponge (*Astrosclera willeyana*) data and an equilibrium aragonite estimate are also shown (Asami et al. 2020a)

JODC salinity data using a relation for coral reef environment in Ishigaki Island in the southern Ryukyus (Abe et al. 2009). Results show that the $\delta^{18}O$ values of equilibrium high-Mg calcites precipitated at the study sites are expected to fall within a range of −0.85‰ to −0.67‰ in Miyako Island and from −0.88 to −0.58‰ in Okinawa Island (Fig. 4B). Consequently, the $\delta^{13}C$ and $\delta^{18}O$ values of coeval skeletal portions of our *A. wellsi* samples are consistent with those of equilibrium calcites, as same as the findings for *A. wellsi* (Böhm et al. 1996; Grottoli et al. 2010) and *A. willeyana* (Asami et al. 2020a). These lines of evidence document that our *A. wellsi* samples can be suitable for the paleoenvironmental reconstruction study because they probably have a little vital or biological effect on skeletal geochemistry.

3.2 Time series determination

To convert sclerosponge geochemical records from the depth domain to the time domain, an age model was established using our $\Delta^{14}C$ results of 13 subsamples along the growth direction of the SMJ-2-S1 sample (Table S1 and Fig. 2) and earlier reported high-resolution data of accurately dated coral skeleton collected at a 5 m water depth from Ishigaki Island (Hirabayashi et al. 2017) in the Ryukyu region (Fig. 1). It is assumed for this study that a difference in local ^{14}C reservoir effects between Miyako Island and Ishigaki Island can be ignored because the two sites are separated by only a short distance (approx. 120 km). Moreover, they are located at almost equal water depth in the same oceanographic setting in the Ryukyu Islands. Figure 5 depicts a comparison showing good consistency between the two $\Delta^{14}C$ profiles for 1947–1997, which yields an age model with a pretty high correlation (r = 0.999) under the condition that the skeletal growth rate of sclerosponge is constant at 1.02 mm/year. Our growth rate estimate accords with earlier studies for the same species: *A. wellsi* (1.355 mm/year, Grottoli et al. 2010; 0.80–0.85 mm/year, Ohmori et al. 2014). Applying the age model to the data before 1940, a 130-year-long (1880–2012 AD) time series of the geochemistry was generated from the SMJ-2-S1 sample. For age determination of MNZ-S1 sample, the $\delta^{13}C$ profiles were used for this study, the method of which is based on evidence that the $\delta^{13}C$ records of our two samples clearly exhibited a typical decreasing trend associated with the anthropogenically derived Suess effects as well as those shown in corals and other sclerosponges (see details presented in the next section). The decreasing trend of the depth domain MNZ-S1 $\delta^{13}C$ record under the condition that the skeletal growth rate is constant at 1.42 mm/year corresponded well with that of the SMJ-2-S1 $\delta^{13}C$ time series since 1960, yielding a high correlation coefficient of 0.99 (Supplementary Figure S1). Based on those results, it can be estimated that the sclerosponge MNZ-S1 has grown for about 55 years. We identified 65 ± 5 pairs of skeletal density bandings in selected three transects near the geochemical profile of the X-radiographic image of MNZ-S1 using software (ImageJ64), possibly implying that the growth bandings might have been formed annually (Fig. 2). However, the overestimation of 10 ± 5 pairs of density bandings may indicate that the MNZ-S1 sclerosponge occasionally has seasonally formed growth bandings. Therefore, the skeletal bandings of sclerosponges cannot be used for age determination in this study, like corals, because further validation studies using ultra-high resolution laser ablation analyses are needed.

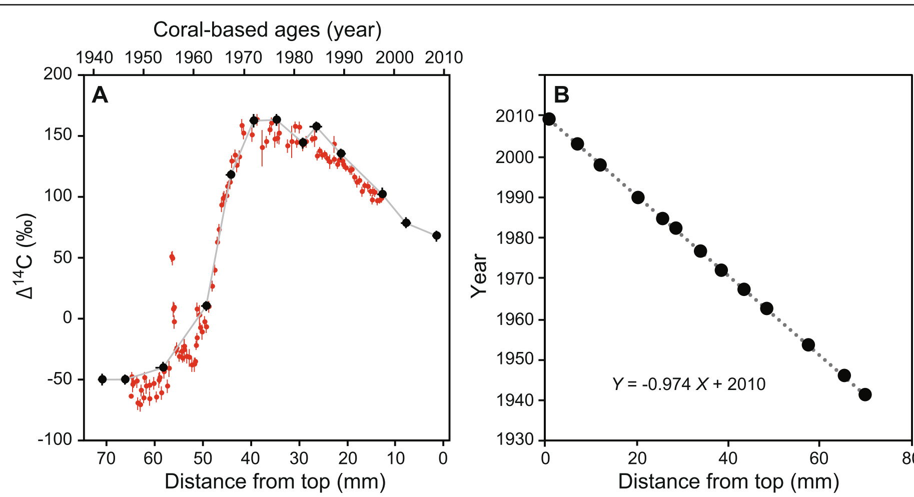

Fig. 5 A Comparison of the depth domain profile of radiocarbon ($\Delta^{14}C$) data of the SMJ-2-S1 sclerosponge with a coral-derived time series from Ishigaki Island in the southern part of Ryukyu Islands (Hirabayashi et al. 2017). **B** Established age model in this study at the assumption that sclerosponge skeletal rate has been constant

3.3 Anthropogenic signatures: carbon isotope

The $\delta^{13}C$ values of SMJ-2-S1 and MNZ-S1 samples were 2.06‰ to 3.49‰ for 1880–2012 and were 1.54‰ to 3.01‰ for 1960–2015 (Fig. 6). The near-annual resolved $\delta^{13}C$ time series of the SMJ-2-S1 sample showed decreasing trends at low (−0.0043‰/year) and high (−0.023‰/year) rates, respectively before and after 1960 (Table 1; Fig. 6). The latter rate is identical to that of the MNZ-S1 sample (−0.024‰/year). Such a typical decreasing trend found in this study probably reflects the Suess effect, which is widely accepted as a result of the burning of ^{12}C-enriched fossil fuels (Keeling et al. 1979; Quay et al. 1992). The effect has decreased $\delta^{13}C$ of dissolved inorganic carbon (DIC) in seawater since the mid-twentieth century because of the accelerated increase of anthropogenically derived CO_2 with isotopically low $\delta^{13}C$ values into the ocean from the atmosphere, results of which are recorded in corals and sclerosponges in the Pacific and Atlantic Oceans (e.g., Böhm et al. 2002; Asami et al. 2005; Swart et al. 2010). The post-1960 rates of decrease of our sclerosponge $\delta^{13}C$ records are close to that of the northwestern tropical Pacific coral for 1960–2000 (Asami et al. 2005) (Fig. 6) and of atmospheric CO_2 at Hawaii for 2000–2016 (derived from the Scripps CO_2 Program initiated in 1956 by Keeling CD). It is fascinating that the rate of decrease of our $\delta^{13}C$ records (−0.0043‰/year) for 1880–1960 was similar to those (−0.0033 to −0.0060‰/year) of the tropical Atlantic sclerosponges (Lazareth et al. 2000; Böhm et al. 2002; Swart et al. 2002), but our results (−0.019 and −0.020‰/year) were slightly higher than those (−0.006 to −0.017‰/year) for 1960–1992 (Table 1; Fig. 6). These results indicate that the accelerated increase in anthropogenic CO_2 with low $\delta^{13}C$ values has been about 1.4 times greater in the surface ocean around Miyako and Okinawa Islands of the subtropical Northwest Pacific than in the Caribbean Sea of the tropical Atlantic since the mid-twentieth century. The difference in the decreasing rates of sclerosponge $\delta^{13}C$ records between the two regions is consistent with that in annual mean sea-air CO_2 flux (Takahashi et al. 2009), showing that CO_2 sink is significantly higher in the northwestern Pacific region, especially during winter than in the Caribbean Sea.

3.4 Seawater temperature variations: oxygen isotope, Sr/Ca, and U/Ca

The respective $\delta^{18}O$, Sr/Ca, and U/Ca values of the SMJ-2-S1 sample for 1880–2012 were −1.11‰ to −0.72‰, 1.32 to 1.70 mmol/mol, and 0.22 to 0.27 μmol/mol and averaged −0.90‰, 1.52 mmol/mol, and 0.24 nmol/mol (Fig. 7). The respective values of the MNZ-S1 sample for 1960–2015 were −1.19‰ to −0.48‰, 1.32 to 1.51 mmol/mol, and 0.26 to 0.34 μmol/mol and averaged −0.75‰, 1.39 mmol/mol, and 0.29 μmol/mol. The $\delta^{18}O$, Sr/Ca, and U/Ca time series accord well with long-term variations and trends of the seawater temperatures on decadal and multi-decadal timescales. Some inconsistency exists between the SSTs and sclerosponge geochemical records on annual-to-interannual timescales, the reason of which is explainable by the errors (probably less than ± 2 years) of age determination on the assumption that the skeletal growth rate is constant. Therefore, to discuss seasonal, annual, and interannual resolved climate signals, specifying annually formed growth layers in sclerosponge skeleton in combination with high-resolution ^{14}C measurements will be useful.

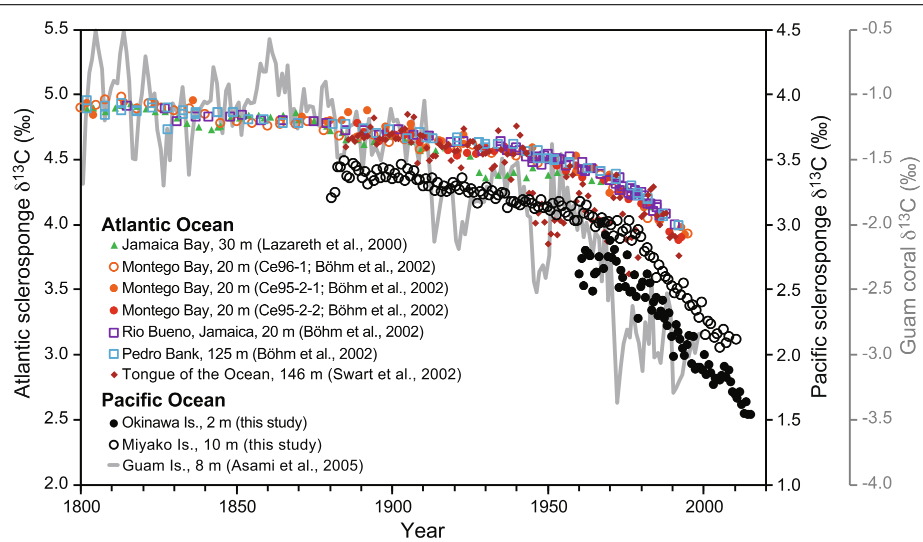

Fig. 6 Time series of sclerosponge $\delta^{13}C$ from Miyako and Okinawa Islands in the northwestern subtropical Pacific compared to earlier published data from sclerosponges in the tropical Atlantic (Lazareth et al. 2000; Böhm et al. 2002; Swart et al. 2002) and a *Porites* sp. coral in the western tropical Pacific (Asami et al. 2005)

Table 1 Decreasing trends estimated from sclerosponge $\delta^{13}C$ records since 1800

	Locality	Water depth (m)	Before 1960			After 1960		
			Period	$\delta^{13}C$ slope for 1880-1960		Period	$\delta^{13}C$ slope for 1960-1992	
				(‰/year)	(‰/year)		(‰/year)	(‰/year)
***Pacific Ocean* (*this study*)**								
MNZ-S1	Okinawa Is.	2				1960-2015	−0.024	−0.019
SMJ-2-S1	Miyako Is.	10	1880-1960	−0.0043	−0.0043	1960-2012	−0.023	−0.020
***Atlantic Ocean* (*Carribean Sea*)**								
Lazareth et al. (2000)	Jamaica Bay	30	1800-1960	−0.0037	−0.0049	1960-1983	−0.010	
Böhm et al. (2002) (Ce96-1)	Montego Bay	20	1800-1960	−0.0031	−0.0033	1960-1995	−0.015	−0.015
Böhm et al. (2002) (Ce95-2-1)	Montego Bay	20	1800-1960	−0.0030	−0.0048	1960-1994	−0.016	−0.016
Böhm et al. (2002) (Ce95-2-2)	Montego Bay	20	1889-1960	−0.0040		1960-1993	−0.017	−0.017
Böhm et al. (2002)	Rio Bueno, Jamaica	20	1827-1960	−0.0040	−0.0060	1960-1986	−0.013	
Böhm et al. (2002)	Pedro Bank	125	1800-1960	−0.0026	−0.0038	1960-1992	−0.017	−0.017
Swart et al. (2002)	Tongue of the Ocean	146	1886-1960	−0.0079		1960-1992	−0.006	−0.006

Since 1960, SSTs have increased in the Miyako and Okinawa Islands with respective rates of +0.0162 °C/year and +0.0124 °C/year (Table 2). Applying a linear regression, the decreasing trends of sclerosponge $\delta^{18}O$, Sr/Ca, and U/Ca from Miyako and Okinawa Islands are estimated respectively as -3.62×10^{-3} and -3.81×10^{-3} ‰/year, -1.55×10^{-3} and -1.18×10^{-3} mmol/mol/°C/year, and -0.271×10^{-3} and -0.727×10^{-3} μmol/mol/°C/year (Table 2). The $\delta^{18}O$ values of biogenic carbonates are well known to reflect the temperature and $\delta^{18}O$

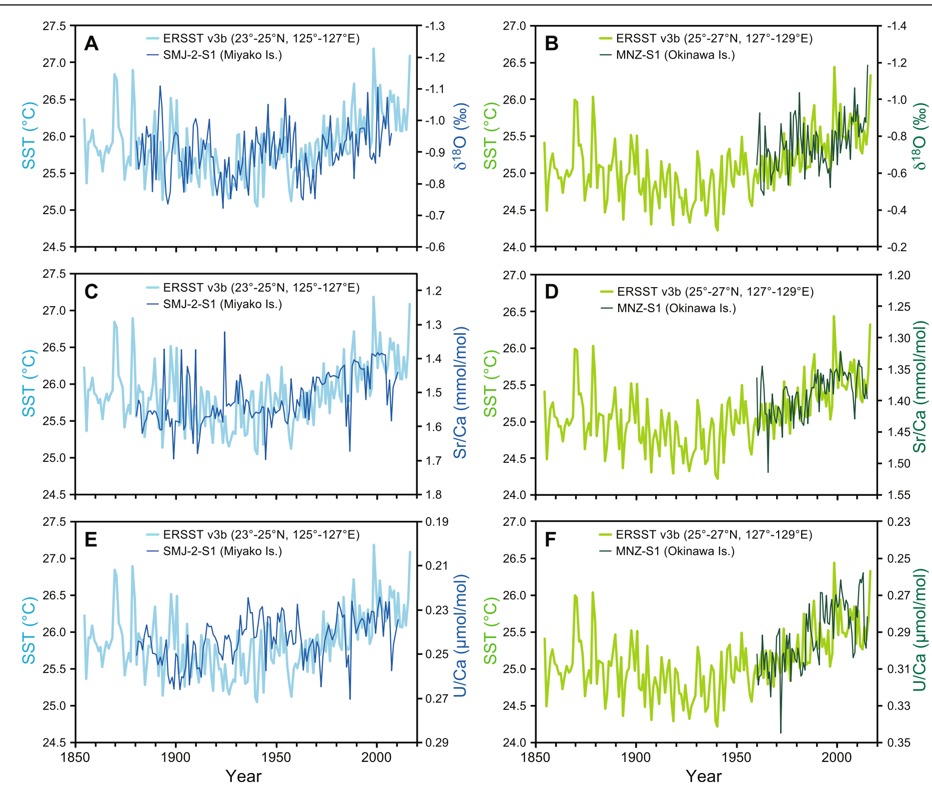

Fig. 7 Comparison of time series of sclerosponge $\delta^{18}O$ (**A** and **B**), Sr/Ca (**C** and **D**), and U/Ca (**E** and **F**) from Miyako Island (left panels) and Okinawa Island (right panels) with instrumental SST for 1880–2016 (ERSST v3b, http://iridl.ldeo.columbia.edu/)

Table 2 Post-1960 trends estimated from SST and sclerosponge $\delta^{18}O$, Sr/Ca, U/Ca records

ID	Locality	SST (°C/year)	$\delta^{18}O$ slope (‰/year)	Dependence (‰/°C)	Sr/Ca slope (mmol/mol/year)	Dependence (mmol/mol/°C)	U/Ca slope (μmol/mol/year)	Dependence (μmol/mol/°C)
MNZ-S1	Okinawa Is.	+0.0124	−0.00381	−0.307	−0.00118	−0.095	−0.000727	−0.059
		R = 0.605	*R* = −0.444		*R* = −0.551		*R* = −0.683	
SMJ-2-S1	Miyako Is.	+0.0162	−0.00362	−0.223	−0.00155	−0.096	−0.000271	−0.017
		R = 0.672	*R* = −0.622		*R* = −0.360		*R* = −0.380	

The *R* value for the linear regression between age and variable (SST, $\delta^{18}O$, Sr/Ca, and U/Ca) is statistically significant at the 95% confidence limits

of seawater, the evidence of which was also confirmed for the species *A. wellsi* sclerosponge (Grottoli et al. 2010). The SST dependence of SMJ-2-S1 $\delta^{18}O$ is almost identical to those of the same species from Saipan (ca. −0.24‰/°C) (Grottoli et al. 2010) and of inorganic calcite (ca. −0.22‰/°C) (Kim and O'Neil 1997). The value (−0.307‰/°C) of MNZ-S1 $\delta^{18}O$ is much higher than expected; implying that seawater $\delta^{18}O$ at the sclerosponge living site in Okinawa has decreased by approximately 0.1‰ since 1960. Although the SST dependencies of Sr/Ca for the long-term trend are almost identical between the two sclerosponge samples, several abrupt excursions in the Sr/Ca signals did not track SST variations (Fig. 7). For *A. wellsi*, such inconsistency between Sr/Ca and SST is explainable by an ICP-MS interference with accurate Sr/Ca measurement of high-Mg materials and by the interference of Mg with Sr incorporation into the high-Mg calcite (Morse and Bender 1990; Grottoli et al. 2010). For U/Ca, the mechanisms of uranyl complexes incorporation into carbonates are very complicated (Langmuir 1978; DeCarlo et al. 2015). The post-1960 decreasing trend and SST dependency for U/Ca are largely different between the two samples (Table 2). Furthermore, the U/Ca and Sr/Ca values correlated with moderately positive *r* values of 0.66 for SMJ-2-S1 and 0.67 for MNZ-S1, but no significant correlations were found with $\delta^{18}O$ values (Table 3). Considering results of earlier reports (Grottoli 2006; Grottoli et al. 2010; Wu and Grottoli 2010), one can infer that the high-Mg calcite Sr/Ca and U/Ca values of *A. wellsi* sclerosponges might not be a useful indicator for temperature, unlike those of aragonite sclerosponges (*Astrosclera willeyana* and *Ceratoporella nicholsoni*) (Rosenheim et al. 2004; Fallon et al. 2005; Haase-Schramm et al. 2005; Waite et al. 2018; Asami et al. 2020a).

Unlike the Sr/Ca and U/Ca records, the $\delta^{18}O$ time series of the SMJ-2-S1 sample clearly showed decade-scale variation for 1880–2012 (Fig. 7). Spectral analyses of the 5-year moving average time series using a multi-taper method (see the methodology and climatic applications in earlier studies (Mann and Lees 1996; Ghil et al. 2002; Asami et al. 2020b)) revealed a marked concentration of its variance in periodicity of approximately 2, 3, 6.5, 7–10, and 20–30 years/cycle for 1880–2012, which exceed the 90% confidence level (Fig. 8A). The similar periodicity roughly corresponds to the significant spectra of instrumental SST in Miyako Island for 1880–2012 (Fig. 8B). These results indicate that the sea surface condition around Miyako Island had been highly dominated by interannual and decadal variations in temperature and seawater $\delta^{18}O$ since the late nineteenth century. It is conceivable that the interannual variations with

Table 3 Correlations between sclerosponge geochemical records[a]

	$\delta^{18}O$	Sr/Ca	Ba/Ca	Pb/Ca	U/Ca
MNZ-S1 (Okinawa) 1960-2015					
$\delta^{13}C$	**0.51**	**0.55**	*0.06*	**−0.78**	**0.69**
$\delta^{18}O$		*−0.08*	*−0.19*	**−0.32**	*0.09*
Sr/Ca			**0.48**	**−0.59**	**0.67**
Ba/Ca				*−0.27*	*0.18*
Pb/Ca					**−0.58**
SMJ-2-S1 (Miyako) 1960-2012					
$\delta^{13}C$	**0.63**	*0.31*	**−0.41**	**−0.64**	**0.36**
$\delta^{18}O$		*0.19*	*−0.08*	*−0.24*	*0.26*
Sr/Ca			*0.24*	*−0.30*	**0.66**
Ba/Ca				**0.37**	*0.15*
Pb/Ca					*−0.25*

[a]Coefficent values in italics and bold fall outside ($p > 0.01$) and inside ($p < 0.01$) the 99% confidence intervals, respectively

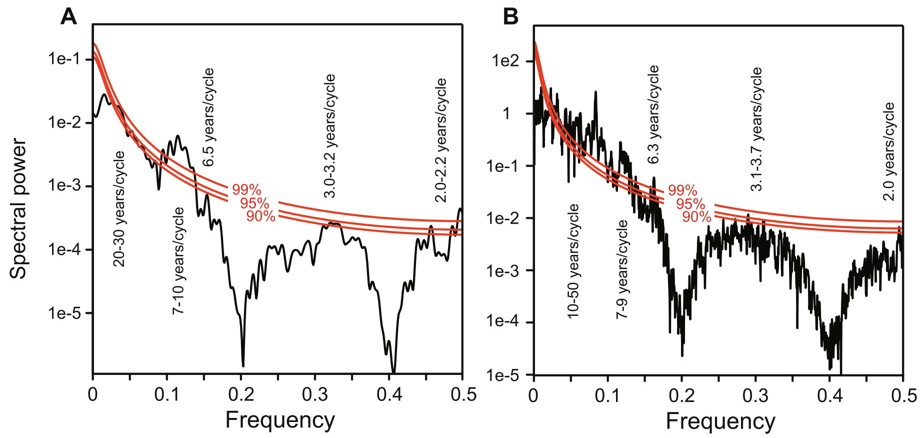

Fig. 8 Results of multi-taper method spectral analyses of the SMJ-2-S1 sclerosponge $\delta^{18}O$ (**A**) and instrumental SST (ERSST v3b) (**B**) for 1880–2012. The 90%, 95%, and 99% confidence levels are shown as red lines. Numbers denote interannual to decadal periodicity with significant peaks at the confidence level

relatively shorter and longer periodicity of about 2 years and 3–8 years are linked respectively to the East Asian Monsoon and El Niño/Southern Oscillation, which is consistent with climatic signatures in coral $\delta^{18}O$ records from the Ryukyu Islands (Mishima et al. 2010; Asami et al. 2020b). The Pacific Decadal Oscillation (PDO), a dominant climate mode in the Pacific Ocean, was found to be significant based on modern long-coral $\delta^{18}O$ records kept for the northwestern tropical-to-temperate Pacific region for the last two centuries (Asami et al. 2005; Watanabe et al. 2014). Therefore, it is likely that the decadal-scale variations with the periodicity of 20–30 years detected in the sclerosponge $\delta^{18}O$ record might be associated with the PDO variations.

3.5 Other environmental signals: Ba/Ca and Pb/Ca

The Ba/Ca values of the SMJ-2-S1 and MNZ-S1 samples were 0.19–10.3 μmol/mol for 1880–2012 and 0.00–2.80 μmol/mol for 1960–2015 (Fig. 9). Considering the within-skeleton Ba/Ca variations of ± 0.54 μmol/mol (RSD, 7.7%) for SMJ-2-S1 and ± 0.17 μmol/mol for MNZ-S1 (RSD, 9.9%) (Fig. 4), the Ba/Ca values and temporal variations differ greatly between the two samples and also from a different species *C. nicholsoni* from the

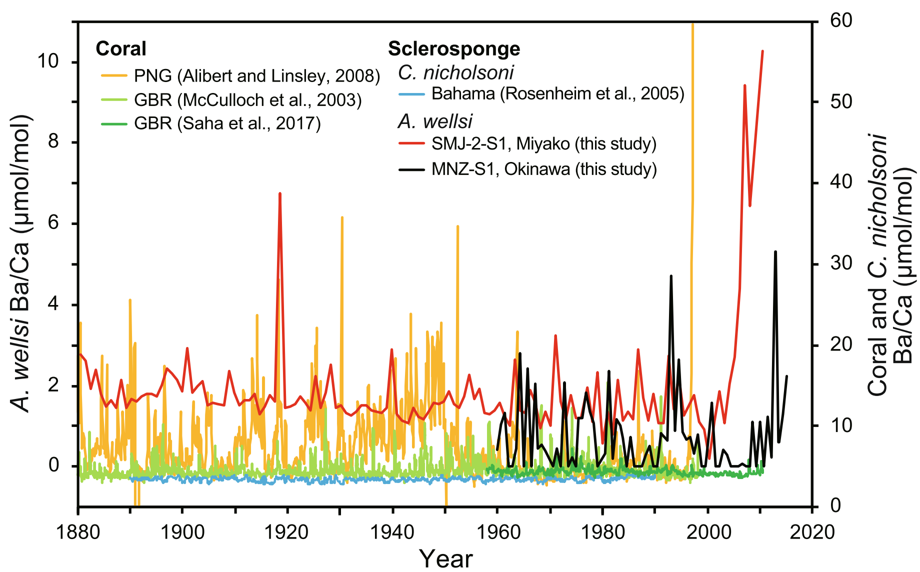

Fig. 9 Secular Ba/Ca variations in sclerosponges and corals from the Pacific and Atlantic Oceans

Atlantic (Rosenheim et al. 2005b). These can be partially attributable to wide interspecific, intraspecific, and within-skeleton variations in Ba/Ca, as reported from an earlier study (Asami et al. 2020a) and as portrayed in Fig. 4. Another reason for the different Ba/Ca variations might be related to local environmental effects. Earlier studies of annually banded corals in the Pacific Ocean demonstrated that increasing short-term signals in coral Ba/Ca time series generally corresponded to river discharge and terrestrial runoff associated with heavy rainfall and land-use practices (McCulloch et al. 2003; Saha et al. 2018) and to ocean currents (Alibert and Kinsley 2008a, 2008b) (Fig. 9). It is noteworthy that the SMJ-2-S1 Ba/Ca values have increased markedly since 2000: its signature is not found in the MNZ-S1 values. That large difference cannot be explained by regional differences in precipitation and ocean currents because the two sites are located in almost identical climate settings (Fig. 1). One possibility for the abrupt Ba/Ca increase in the SMJ-2-S1 record would be related to a land-use change: large-scale roads had been constructed from the late 1990s to the early 2010s, and resort facilities with hot spring, golf course, and hotel had developed from the 1990s to the 2010s around the study site in the southern part of Miyako Island (Geospatial Information Authority of Japan, https://mapps.gsi.go.jp/) (Supplementary Figure S2). Some earlier studies demonstrated that skeletal Ba/Ca ratios of coral aragonite can be a proxy for the amount of land-based sediment input to the reef environment because Ba is sourced from the fine-grained components of terrestrial soils (McCulloch et al. 2003; Fleitmann et al. 2007; Prouty et al. 2010). It is, therefore, likely that the temporal Ba/Ca variations in our sclerosponge SMJ-2-S1 sample might have recorded the history of terrigenous sediment flux associated with land development since 1880. Nevertheless, we cannot clarify the reason from the present study results because the physiological process and the mode of barium incorporation into the sclerosponge skeleton remain unknown (Rosenheim et al. 2005b; Allison et al. 2012; Asami et al. 2020a).

The Pb/Ca values of the SMJ-2-S1 and MNZ-S1 samples were 228–3349 nmol/mol for 1880–2012 and 1699–4146 nmol/mol for 1960–2015 (Fig. 10). These Pb concentrations are much higher than those of corals: a fact which agrees with results described in earlier reports (Benavides and Druffel 1986; Shen and Boyle 1987). The relatively high Pb/Ca records are attributable to a biological characteristic by which sclerosponges concentrate Pb in their skeleton by containing nutrition via filter feeding particulates with absorbed heavy-metal species (Hartman and Goreau 1966; Willenz and Hartman 1989). The temporal Pb/Ca variations of our sclerosponges showed a similar increasing trend during the late twentieth century, but the values were significantly different (Fig. 10), considering the within-skeleton variations of ± 172 nmol/mol (RSD, 8.1%) for SMJ-2-S1 and ± 253 nmol/mol (RSD, 7.8%) for MNZ-S1 (Fig. 4). Moreover, our Pb/Ca ratios of *A. wellsi* samples (high-Mg calcite) are higher than that found from Kume Island (Ohmori et al. 2014), despite being close to our study site (Fig. 1), and are higher than those of *C. nicholsoni* (aragonite) (Lazareth et al. 2000; Rosenheim et al. 2005b). That difference indicates that the mechanisms of Pb incorporation might have large interspecific, intraspecific, and within-skeleton variations. Those indications can be partially supported by the results of an evaluation study of sclerosponge (Rosenheim et al. 2005b; Asami et al. 2020a) and our results portrayed in Fig. 4. An increasing trend was identifiable in our sclerosponge Pb/Ca records since 1950 (Fig. 10). The Pb/Ca increase primarily accords with those of corals and sclerosponges

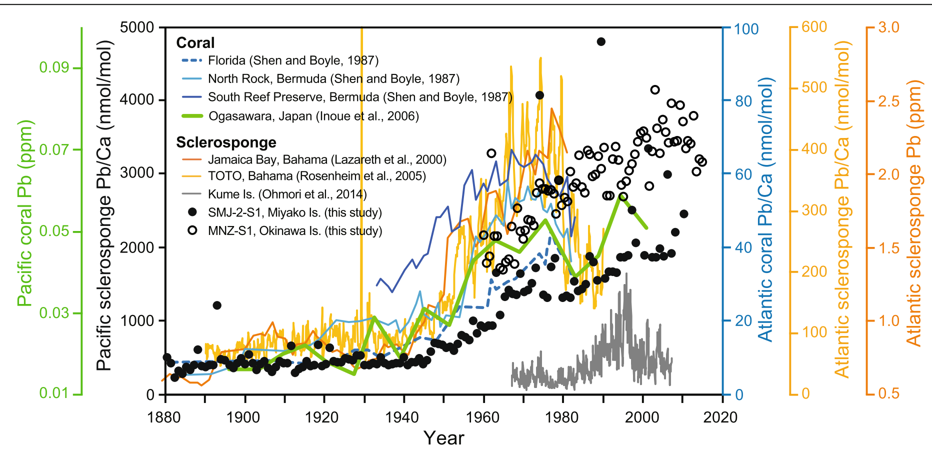

Fig. 10 Secular Pb/Ca and Pb concentration variations in sclerosponges and corals from the Pacific and Atlantic Oceans

from the Pacific and Atlantic oceans, which is attributed to Pb emission from industrial activities. The increases in the Atlantic coral and sclerosponge records were followed by a drop occurring in approximately the 1970s, caused by decreased use of lead alkyl additives in gasoline, mainly in the USA (Boutron et al. 1991; Rosman et al. 1993), but those in the Pacific records have been maintained since the 1950s (Inoue et al. 2006; this study). The anthropogenic Pb/Ca signature found in our sclerosponge records is probably associated with industrial activities in eastern Asian countries and Chinese atmospheric aerosols (Zheng et al. 2004; Hao et al. 2008). The predominant source of Pb was identified by lead isotope ratios of the western Pacific corals (Inoue and Tanimizu 2008). The increasing Pb/Ca trends of our sclerosponges from Miyako and Okinawa Islands accord well with the time series of Pb contents in an Ogasawara coral (Fig. 4), indicating that the continued increase of Pb/Ca ratios after the 1980s can be caused by a mixture of Pb emitted from Asian countries with heavy industrialization. However, the sclerosponge Pb/Ca record from Kume Island (Ohmori et al. 2014) has maintained low until the 1980s and decreased since the late 1990s, the variation of which is not consistent with those of the Ogasawara coral and our sclerosponge records. One possible explanation of the difference is that the sclerosponges lived at different water depths of 2–10 m (this study) and 23 m (Ohmori et al. 2014). That is, it is likely that our sclerosponges and the Ogasawara coral, living in very shallow waters, may record the temporal variations in Pb concentration sensitively because aerosols with anthropogenically generated lead are transported to the sea surface through the atmosphere (Inoue and Tanimizu 2008). Further investigations of Pb isotope ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) must be conducted of our samples to clarify the potential source of Pb in the Miyako and Okinawa Islands.

4 Conclusions

From *Acanthochaetetes wellsi* sclerosponge samples for 1880–2012 and 1960–2015, collected respectively at Miyako Island and Okinawa Island in the Ryukyu Islands of southwestern Japan, we first generated near-annual resolved time series of skeletal $\delta^{13}C$, $\delta^{18}O$, and element/Ca ratios (Sr/Ca, Ba/Ca, Pb/Ca, U/Ca). The $\delta^{13}C$ values have decreased at an increasing rate since the mid-twentieth century, indicating the anthropogenically derived Suess effect. Our estimations demonstrated that the $\delta^{13}C$ decrease rate in the northwestern subtropical Pacific was similar to the Caribbean Sea before 1960 but about 1.4 times higher after 1960. The $\delta^{18}O$ records were mostly accorded with long-term variations in seawater temperatures. Using spectral analysis, we detected significant periodicity of 2, 3, 6.5, 7–10, and 20–30 year/cycle in the $\delta^{18}O$ time series, which indicates that sea surface conditions around the southern Ryukyu Islands had been dominated by interannual and decadal variations in temperature and seawater $\delta^{18}O$ since the late nineteenth century.

In contrast to aragonite sclerosponges (*A. willeyana* and *C. nicholsoni*), the validity of Sr/Ca and U/Ca records of *A. wellsi* with high-Mg calcite skeleton for a robust proxy of seawater temperature cannot be determined from data used for this study. Compared to previously published data, the increasing Pb/Ca trends after 1950 found in our sclerosponge records are probably caused by Pb emissions from industrial activities in eastern Asian countries and Chinese atmospheric aerosols. A great difference in the Ba/Ca variations is apparent between our two sclerosponges, which might be related to local environments. However, large intraspecific and within-skeleton variations in Ba/Ca and Pb/Ca prevent the interpretation of environmental signals recorded in the sclerosponge samples. This study suggests that, with more high-resolution age determinations, sclerosponge-based reconstruction studies can elucidate secular variations in ocean environments on seasonal, annual, and interannual time scales.

The Kuroshio Current allows the development of widely distributed reefs with a highly diversified biota of the Ryukyu Islands during the late Quaternary (Iryu et al. 2006). In combination with the geochemistry of calcified marine organisms such as corals and giant clams, if long-lived sclerosponges are collected from deeper ocean environments, we can generate long time series reflecting spatial and vertical changes in coral reef environments of the Ryukyu Islands that have taken place over the past few hundred years, extending far beyond the limits of meteorological observation data, and over the last deglaciation.

Abbreviations

JODC: Japan Oceanographic Data Center; VPDB: Vienna Standard Pee Dee Belemnite

5 Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40645-021-00434-7.

Additional file 1: Table S1. Radiocarbon data reported as Δ14C of the sample series extracted from the sclerosponge SMJ-2-S1 with the estimated date of skeletal formation. **Figure S1.** Comparison of the depth domain $\delta^{13}C$ profile of the sclerosponge MNZ-S1 with the $\delta^{13}C$ time series of SMJ-2-S1 determined by radiocarbon data. The decreasing trend of MNZ-S1 $\delta^{13}C$ record under the condition that the skeletal growth rate is 1.42 mm/year shows good agreement with that of SMJ-2-S1 for 1960–2012, yielding a high correlation between the two regression lines (r = 0.99). **Figure S2.** Aerial photos of the southern part of Miyako Island in 1995 and 2019 (modified from Geospatial Information Authority of Japan, https://mapps.gsi.go.jp/), showing the locations of the sclerosponge sampling site and large-scale road and resort constructions.

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Authors' contributions
RA designed the study, analyzed the samples and data, and wrote the manuscript. YM, MM, and TS carried out the fieldwork and sampling. TM, RS, RU, and YI collaborated with RA in the experimental study. The authors read and approved the final manuscript.

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Availability of data and materials
Sclerosponge data generated in this study will be available at NOAA NCDC World Data Center for Paleoclimatology (https://www.ncdc.noaa.gov/data-access/paleoclimatology-data). Please contact the corresponding author for data requests.

Declarations

Competing interests
The authors declare that they have no competing interests.

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