Micro-CT Scanning of Tests of Three Planktic Foraminiferal Species to Clarify Dissolution Process and Progress

S. Iwasaki1, K. Kimoto1, Y. Okazaki2, and M. Ikehara3

1Research Institute for Global Change, JAMSTEC, Yokosuka, Japan, 2Department of Earth and Planetary Sciences, Graduate School of Sciences, Kyushu University, Fukuoka, Japan, 3Center for Advanced Marine Core Research, Kochi University, Nankoku, Japan

Abstract Evaluation of foraminiferal test dissolution in deep-sea sediments facilitates reconstruction of seawater chemistry. Here we observed test dissolution processes of the planktic foraminifera Trilobatus sacculifer, Globigerinoides ruber, and Neogloboquadrina dutertrei from midlatitudes of the western North Pacific; in these three species, we tested the ability of a new dissolution index using data from X-ray micro-computed tomography scanning. Although the dissolution process of foraminiferal tests differed slightly among species, dissolution of all species was equally assessed by the calcite density distribution (%Low-CT-number calcite volume) calculated from the CT number histogram. In T. sacculifer and G. ruber, the test area density, a conventional proxy for assessing test condition based on weight measurement, is affected by variations in the thickness of the outermost chamber wall; thus, this conventional proxy can be affected by sea surface conditions during test calcification. In contrast, the relationship between the %Low-CT-number calcite volume of tests and the deep seawater calcite saturation state suggests that X-ray micro-computed tomography scanning is applicable for evaluating the intensity of foraminiferal test dissolution at the undersaturated deep sea floor in this area and is an invaluable proxy for detecting deep seawater carbonate ion concentration changes on glacial-interglacial timescales.

1. Introduction

The North Pacific Ocean has great carbon storage capacity; the oldest CO2-rich deep seawaters reach the northern end of this area. Therefore, deep seawater of the North Pacific Ocean has been considered to store poorly ventilated water containing substantial amounts of carbonate dioxide respired during the Last Glacial Maximum (LGM) and to play a significant role in deglacial CO2 outgassing (e.g., Galbraith et al., 2007; Gray et al., 2018). In the subarctic North Pacific, reconstruction of deep seawater ventilation suggests the existence of stratified deep seawater during the LGM, and well-ventilated deep seawater and formation of North Pacific Deep Water (NPDW) during the last deglaciation (17–16 ka). This reconstruction implies that the North Pacific deep seawater plays a significant role in the global carbon cycle (Okazaki et al., 2010; Rae et al., 2014). The deep seawater carbonate ion concentration ([CO3²⁻]) is governed primarily by the concentration of dissolved inorganic carbon and alkalinity, and its reconstruction can provide valuable insights into the changes in the carbon cycle between glacial and interglacial periods; such reconstruction is generally difficult for higher latitudes of the North Pacific due to high carbonate dissolution and insufficient amount of foraminiferal test in deep sediment. Although benthic foraminiferal B/Ca ratios have been developed as a deep seawater [CO3²⁻] proxy (Yu & Elderfield, 2007) and applied in the eastern equatorial Pacific (Umling & Thunell, 2018), this proxy requires single-epifaunal species of benthic foraminifera. Thus, sediment core samples available for B/Ca ratio measurement are limited in the sediment samples that contain sufficient fossils of epifaunal benthic foraminiferal species.

Planktic foraminiferal tests are the major component of deep-sea carbonate in the deep-sea sediment (Schiebel, 2002). Dissolution and preservation of these tests is intimately associated with the carbonate saturation state ([CO3²⁻]) in the surrounding seawater, which is defined as the difference between the measured carbonate ion concentration ([CO3²⁻]IN SITU) and the calculated theoretical value of calcite saturation ([CO3²⁻]SATURATION) (e.g., Berger et al., 1982). Assuming constant [Ca²⁺] on a time scale shorter than a
10^5 year, carbonate saturation state is controlled by the [CO₃²⁻] of seawater. Therefore, dissolution and preservation of planktic foraminiferal tests in the sediment can provide valuable information about past deep seawater [CO₃²⁻]. Numerous studies have estimated carbonate dissolution in the ocean by using data from analysis of planktic foraminiferal tests. In particular, the size-normalized weight (SNW) of foraminiferal tests is one of the most common proxies to estimate carbonate dissolution (e.g., Broecker & Clark, 2001; Lohman, 1995; Qin et al., 2017). Based on calibration between SNW in core-top sediments and bottom-water ∆[CO₃²⁻] in the equatorial Pacific, size-normalized test weights have served as a proxy for deep seawater [CO₃²⁻] (e.g., Broecker & Clark, 2001; Qin et al., 2017). In contrast, the test area density (μg μm⁻²) of foraminifera, which is the ratio of the weight of a test to its projected area, is a more rigorous size-standardized proxy than SNW, and it is also employed as a proxy of calcification intensity controlled by sea surface conditions (e.g., Moy et al., 2009; Weinkauf et al., 2013; Zarkogiannis et al., 2019). Sea surface conditions where foraminifera calcify can influence the thickness of foraminiferal test walls (Marshall et al., 2013) and bulk density (Iwasaki et al., 2019), which implies that the initial weights of foraminiferal tests differ depending on growth conditions as suggested in Barker and Elderfield (2002); thus, the test weight proxies SNW and test area density may be affected by not only dissolution at the deep seafloor but also calcification at the sea surface. The assessment of carbonate dissolution based on foraminiferal test weight measurements has been performed on the assumption that the sea surface conditions were uniform during glacial-interglacial periods, despite these uncertainties in factors affecting initial test growth. Therefore, we need a more precise method to estimate foraminiferal test dissolution intensity at the deep sea floor.

The internal structure of foraminiferal tests has been observed by X-ray micro-computed tomography (XMCT) scanning. This technique has also been used to evaluate test dissolution intensity (Iwasaki et al., 2015; Johnstone et al., 2010). Johnstone et al. (2010) have observed the dissolution of planktic foraminiferal tests (Globigerinoides ruber, Trilobatus sacculifer, Neogloboquadrina dutertrei, and Pulleniatina obliquiloculata) from core-top sediments: these observations have revealed that the inner calcite of T. sacculifer, N. dutertrei, and P. obliquiloculata tests become porous with dissolution and the outer calcite is well preserved. On the other hand, the whole test of G. ruber, which lacks an outer crust and has a thin wall, becomes porous with dissolution. Subsequently, Iwasaki et al. (2015) have performed dissolution experiments under pH-controlled conditions and have evaluated the dissolution process of Globigerina bulloides by using the CT number, which is an indication of calcite density. They have suggested that G. bulloides tests consist of early-developed calcite (EDC) formed during the juvenile stage, and inner and outer calcite layers in the outermost chamber. Iwasaki et al. (2015) have revealed that G. bulloides tests start to dissolve from the vicinity of the EDC, and dissolution then spreads to the inner calcite layer, which has a porous microgranular crystalline structure; in contrast, the outer calcite layer, with a dense, euhedral crystalline structure, is resistant to dissolution. They used variations of mean CT number (bulk density of test) and CT number histograms, which are measured by XMCT scanning, in order to evaluate the dissolution intensity of G. bulloides tests: the mean CT number of G. bulloides tests decreases and the CT number histogram shifts from a unimodal to a bi-modal distribution with the progression of test dissolution. Thus, in order to quantify the shift of the CT number histogram with test dissolution, Iwasaki et al. (2015) suggested %Low-CT-number calcite volume, which is calculated based on the CT number histogram, as a new index of test dissolution intensity. Because the CT number histogram is independent of variations of test size and test wall thickness due to differences of initial test condition, this index is expected to be useful as a new proxy of carbonate dissolution at the deep seafloor; in the future, it might be developed as a quantitative proxy of deep seawater [CO₃²⁻]. No study, however, has investigated whether the shift of the CT number histogram revealed by XMCT scanning is applicable to assess dissolution intensity of foraminiferal species other than G. bulloides. Therefore, evaluation of the relationship between the variation in CT number histogram of dominant species in the North Pacific and deep seawater ∆[CO₃²⁻] is necessary to establish this method as a new paleo-deep seawater [CO₃²⁻] proxy applicable in the North Pacific.

Here we employed XMCT scanning to evaluate the progression of test dissolution of multiple species of planktic foraminifera in the seafloor sediments in midlatitudes (~30°N) of the western North Pacific, around the northern limit of the wide distribution of carbonate sediment. We verified the applicability of this test dissolution index using the CT number histogram suggested by Iwasaki et al. (2015) with three additional species (T. sacculifer, G. ruber, and N. dutertrei) that are dominant species in this study area. The dissolution process of these three species of planktic foraminiferal tests was investigated with cross-sectional isosurface
images and CT number histograms of well-preserved tests from sediment trap samples and partially dissolved tests from core-top sediments in the western North Pacific. Afterward, we investigated the relationships between the dissolution index by XMCT scanning and the deep seawater Δ[CO$_3^{2-}$] where sediment cores were sampled, and we discuss the potential of the new dissolution index as a proxy for deep-seawater [CO$_3^{2-}$] in midlatitudes of the western North Pacific.

2. Material and Methods

2.1. Multiple Core Samples and Sediment Trap Samples

Planktic foraminiferal tests were obtained from the core tops (0–1 cm depth from the seafloor) of eight core samples that were collected during cruises of the R/V Shinsei-maru KS-15-4 from 1 to 10 June 2015 and the R/V Hakuho-maru KH-16-6 from 11 to 28 November 2016 between 29 and 34°N in the western North Pacific (Figure 1). Water depths (m) of sampling sites ranged from 2,221 to 4,006 m. Deep seawater Δ[CO$_3^{2-}$] was calculated with CO2calc software (Robbins et al., 2010) from data for seawater parameters (temperature, salinity, total alkalinity, total inorganic carbon, and concentrations of phosphate and silicate) that were obtained for nearby Global Ocean Data Analysis Project (GLODAP) sites (Key et al., 2004) (Table 1). Data from water bottles collected from three stations from the WOCE-P09 line were used for calculation of deep seawater Δ[CO$_3^{2-}$] at core KH16-6 St. 2, St. 3, St.5, and St. 6. Data from one station from the WOCE-P09 line were used for the deep seawater Δ[CO$_3^{2-}$] calculation at KH16-6 St. 9 and at KS15-4 St. 1, St. 2, and St. 3. The estimated sea floor seawater Δ[CO$_3^{2-}$] at core sites ranged from −3.67 to −24.09 μmol kg$^{-1}$.

From the seafloor sediments, tests of Trilobatus sacculifer, formerly named Globigerinoides sacculifer (Spezzaferri et al., 2015), and Neogloboquadrina dutertrei (D’Orbigny) from the 355- to 500-μm size fractions were hand-picked. In this study, T. sacculifer test without a sac-like final chamber were selected. In addition, tests of Globigerinoides ruber (D’Orbigny), which is the sensu stricto morphotype, from the 200- to 355-μm size fractions were handpicked (Figure 2).

In addition to sediment samples, we used foraminiferal tests from sediment traps moored in the western equatorial Pacific (Site MT3: 00° 00.843’S, 145°01.580’E; collection depth: 1020 m; collecting period: from 1 to 15 October 1999; Yamasaki et al. (2008)) to investigate the conditions of foraminiferal tests of the same species before long-term dissolution on or in the deep seafloor sediment. The seawater Δ[CO$_3^{2-}$] at the mooring depth of the sediment trap was 15 μmol kg$^{-1}$, supersaturated with respect to calcite, based on the nearby bottle sampling data derived from WOCE P10 transect. The sediment trap collecting cups were filled with filtered seawater containing formalin (final concentration 3% v/v) neutralized with sodium tetraborate to prevent bacterial activity. After the cruise, sediment trap samples were sieved through a 1-mm–mesh screen to remove large particles. Then, tests of T. sacculifer, G. ruber, and N. dutertrei were handpicked from the same size fractions used for the sediment samples. Thereafter, three tests of each species were randomly selected and scanned by XMCT.

2.2. X-ray Micro-CT Scanning

The XMCT system (ScanXmate-D160TSS105/11000, Comscantecno Co., Ltd., Kanagawa, Japan) at the Japan Agency for Marine-Earth Science and Technology, Yokosuka, Japan, was used to investigate the physical characteristics of the foraminiferal tests (Iwasaki et al., 2015). For 3-D observation of the foraminifer tests, we used a high-resolution setting (X-ray focus spot diameter, 0.8 μm; X-ray tube voltage, 80 kV;
Table 1  
Multiplecore Sample Depths (m), Locations and Deep Seawater Carbonate Saturation State (Δ[CO₃²⁻]), and Number of Scanned Tests

| Cruise | Site | Depth  | Latitude | Longitude | Δ [CO₃²⁻] | Scanned Number | %Low-CT number calcite volume | Scanned Number | %Low-CT number calcite volume | Scanned Number | %Low-CT number calcite volume |
|--------|------|--------|----------|-----------|-----------|----------------|--------------------------|----------------|--------------------------|----------------|--------------------------|
| KH16-6 | St. 2 | 2221   | 33°46′ N | 137°36′ E | -3.67     | 6              | 31.2 ± 8.3              | 10              | 30.2 ± 5.9              | 13              | 22.5 ± 6.5              |
| KH16-6 | St. 3 | 3079   | 32°40′ N | 138°34′ E | -10.18    | 8              | 30.5 ± 8.0              | 7              | 28.9 ± 6.0              | 8              | 33.6 ± 9.8              |
| KH16-6 | St. 5 | 3267   | 31°07′ N | 138°41′ E | -9.68     | 8              | 41.4 ± 13.6             | 7              | 37.2 ± 6.5              | 6              | 41.0 ± 13.6             |
| KH16-6 | St. 6 | 4006   | 32°28′ N | 137°26′ E | -24.09    | 9              | 51.2 ± 9.7              | 6              | 52.6 ± 9.7              | 8              | 57.0 ± 4.7              |
| KH16-6 | St. 9 | 2922   | 29°20′ N | 133°31′ E | -12.06    | 10             | 46.7 ± 16.4             | 6              | 42.5 ± 5.2              | 8              | 55.3 ± 8.9              |
| KS15-4 | St. 1 | 2863   | 29°25′ N | 133°31′ E | -11.62    | 8              | 48.4 ± 8.1              | 6              | 37.0 ± 12.7             | 8              | 33.2 ± 8.7              |
| KS15-4 | St. 2 | 3218   | 29°13′ N | 133°28′ E | -13.74    | 8              | 42.3 ± 10.9             | 8              | 49.3 ± 4.9              | 8              | 48.3 ± 6.7              |
| KS15-4 | St. 3 | 2792   | 29°27′ N | 133°33′ E | -11.10    | 5              | 43.9 ± 8.4              | 6              | 42.6 ± 11.2             | 8              | 45.1 ± 10.9             |

Figure 2. Reconstructed CT images of T. sacculifer, G. ruber, and N. dutertrei tests observed in this study. Test images of each species are rotated views of the same specimens. Displayed samples are nondissolved tests obtained from a sediment trap. The cross-sectional isosurface images to the right show the internal structure of a test, the color scales show the distribution of calcite density, and the double-headed white arrows show the thicknesses (μm) of the outermost chamber walls.
detector array size, 2,000 × 1,336; 1,500 projections/360°; 0.5 s/projection). After XMCT scanning, ConeCTexpress software (Comscantecno Co., Ltd.) was used to correct and reconstruct the tomography data. Image cross sections were reconstructed from filtered back projections following the general principle of Feldkamp cone beam reconstruction. The scanning and data processing methods were those of Iwasaki et al. (2015).

The CT number, indicating calcite density, was calculated based on the X-ray attenuation coefficient of each sample. In this study, a calcite standard crystal (a particle of NBS-19) was used to standardize the CT number of each test sample: the mean CT numbers of air and standard crystal were defined to be 0 and 1,000, respectively, and the CT numbers of foraminiferal test samples were calculated according to the following equation:

\[
\text{CT number} = \left( \frac{\mu_{\text{sample}} - \mu_{\text{air}}}{\mu_{\text{calcite STD}} - \mu_{\text{air}}} \right) \times 1000
\]

where \(\mu_{\text{sample}}, \mu_{\text{calcite STD}},\) and \(\mu_{\text{air}}\) are the X-ray attenuation coefficients of the sample, calcite standard crystal, and air, respectively.

Isosurface CT images of foraminiferal tests were obtained by Molcer Plus 3-D imaging software (WhiteRabbit Corp., Tokyo, Japan). The mass of voxels with a size of 0.8 \(\mu\text{m}\) on each side, which have a specific CT number, was calculated on the basis of isosurface images of foraminiferal tests. In the cross section of isosurface images, distribution of CT number, which indicates the submicron scale porosity of calcite, are represented by color contours.

2.3. Test Dissolution Index Based on MXCT Scanning

To estimate carbonate dissolution intensities of foraminiferal tests, we employed variations of CT number histograms obtained by XMCT scanning. Iwasaki et al. (2015) have found that CT number histograms of dissolved \(G. \text{bulloides}\) tests show bimodality of the dissolved and preserved calcite. Based on this result, they proposed the relative volume of low-CT-number calcite to the volume of calcite in the whole shell (%Low-CT-number calcite volume) as a more quantitative proxy for carbonate dissolution than the conventional test weight proxy. Based on this concept, we used the following equations to calculate %Low-CT-number calcite volume, a measure of the dissolution intensity of \(T. \text{sacculifer}, G. \text{ruber},\) and \(N. \text{dutertrei}\) tests:

\[
\%\text{Low-CT-number calcite volume} = \left( \frac{V_{\text{low-CT-number calcite}}}{V_{\text{whole shell}}} \right) \times 100
\]

where \(V_{\text{low-CT-number calcite}}\) indicates the volume of low-CT-number calcite in an individual test and \(V_{\text{whole shell}}\) indicates the volume of the whole individual test. In this study, based on the results of XMCT scanning, we classified low CT-number and high CT-number calcite as calcite with CT number values of 200–500 and >500, respectively.

In order to estimate the required sample number for XMCT scanning of foraminiferal tests from sediment samples, the Shapiro-Wilk test and Ballet’s test were used to check for normality and equality of variance of %Low-CT-number calcite volume data, and the assumption of normality and equality were satisfied. Thereafter, the required minimum sample size was statistically estimated based on the following equation using by %Low-CT-number calcite volume data of \(N. \text{dutertrei}\), \(N = (2k/CI)^{\frac{1}{2}}s^2\), where \(k\) indicates a critical value (1.65) under the significance level of 10%, \(CI\) indicates the confidence interval and \(s^2\) indicates variance of the data from the sediment sample. The \(CI\) and \(s^2\) of sample were calculated using by %Low-CT-number calcite volume data of \(N. \text{dutertrei}\), and the minimum sample size was estimated as 6 specimens under the significance level of 10%. In this study, more than 6 specimens (mostly more than 8 specimens) were scanned to satisfy the required sample number, except for five specimens of \(T. \text{sacculifer}\) form KS15-4 St. 3 due to the low abundance in the sediment sample.

2.4. Measurement of Outermost Chamber Wall Thicknesses

The mean thickness of the outermost chamber wall of foraminiferal tests was measured by using Molcer Plus 3-D imaging software. For accurate measurement of chamber wall thickness, we selected cross sections normal to and halfway along the chamber outer surface, and the wall thicknesses were measured at 5 points for each chamber on the selected isosurface image of the chamber walls. In the tests of \(T. \text{sacculifer}\) and \(G. \text{ruber},\)
the thicknesses of the final and f-1 chambers were measured. In N. dutertrei, the thicknesses from the final to f-3 chambers were measured. Note that chamber wall thicknesses from the core KH16-6 St. 6 (water depth 4,006 m) might be underestimated because of significant etching of the inner calcite and difficulty in identifying the inner surface of the outermost chamber. Based on these results, the mean values of all the outermost chamber wall thicknesses were calculated for each core site.

2.5. Size-Normalized Weight and Test Area Density Measurement

For SNW measurement of planktic foraminifer tests, which is a conventional proxy for test dissolution intensity, 20–50 individual tests each of T. saccularifer, G. ruber, and N. dutertrei were picked from narrow size fractions (T. saccularifer: 300–355 μm, G. ruber: 250–300 μm, N. dutertrei: 300–355 μm). The total weights (μg) of tests were measured with an ultramicrobalance (Cahn C-35, Thermo Electron Corp., Round Rock, TX, USA), and data of SNW (μg) were calculated as average test weight. The analytical precision of the weight measurement was ± 0.3 μg (± 1 σ) based on 15 repeated measurements. Note that we could not measure SNW at the following core sites due to small amount of test samples from the narrow size fraction (T. saccularifer, KH16-6 St. 2 and 3; G. ruber, KH16-6 St. 2, 3, 5, 6, 9 and KS15-4 St. 1; N. dutertrei, KH16-6 St. 2, 3 and 5).

In addition, we measured the test area density of a planktic foraminifera; test area density has been employed as a proxy for calcification intensity at the sea surface. The weight of each individual test of T. saccularifer, G. ruber, and N. dutertrei that was scanned by XMCT was measured with an ultra-microbalance (Cahn C-35, Thermo Electron Corp., Round Rock, TX, USA). After the measurements of individual test weights, 2-D projected areas of individual tests were measured with image analysis software (Motic Image Plus 2.1S, Shimadzu Rika Corp., Tokyo, Japan). The area density of each T. saccularifer, G. ruber, and N. dutertrei test was calculated using the following equation based on the method of previous studies (Marshall et al., 2013).

\[
\text{Test area density (μg μm}^{-2}\) = \text{test weight (μg)/projected area (μm}^2\)
\]

In order to evaluate the difference in test area density between sampling sites, a normality test and two-tailed Student’s t tests were performed with JMP statistical software (SAS Institute, Cary, North Carolina, USA).

3. Results

3.1. Change in Internal Structure of Foraminiferal Tests With Dissolution

In order to understand the dissolution process of the tests of three species of planktic foraminifera, variations in the internal structure of tests from sediment trap samples and from three deep seafloor sediment samples from different water depths are shown in cross-sectional isosurface images (Figure 3). The seawater at the mooring depth of the sediment trap was supersaturated with respect to calcite; furthermore, we could not find any etching or apparent reduction of CT number on the inside of tests from the cross-sectional isosurface images (Figure 3). The confirmation by this finding that the condition of the observed foraminiferal tests from the sediment trap sample was pristine suggests that they were not affected by inorganic dissolution during the process of settlement to 1,020 m. In the cross-sectional isosurface images of T. saccularifer tests in the seafloor sediment from core KH16-6 St. 3 (water depth 3,079 m), the decrease in test density began on the EDC and inner calcite layer of the outermost chamber. In the deepest T. saccularifer tests from the core KH16-6 St. 6 (water depth 4,006 m), the EDC and middle calcite layer of the outermost chambers were selectively dissolved; in contrast, the edges of the outer and inner calcite layers of the outermost chambers remained, while there was gradual smoothing of the outer surface (Figure 3a). The G. ruber tests from the cores at KH16-6 St. 2 (water depth 2,221 m) and St. 3 (water depth 3,079 m) showed an apparent decrease in the calcite density in the outermost chambers, except for the final chamber (Figure 3b). In the well-dissolved G. ruber tests from core KH16-6 St. 6 (water depth 4,006 m), we observed selective dissolution of the EDC and middle layer of the outermost chambers, except for the final chamber, as in the tests of T. saccularifer. Finally, in the case of N. dutertrei, selective dissolution of the EDC was found in the tests from core KH16-6 St. 2 (water depth 2,221 m), and the dissolved area (low-CT number area) then spread to the inner calcite layer of the outermost chambers in the tests from core KH16-6 St. 3 (water depth 3,079 m). Interestingly, unlike the tests of G. ruber, the final chamber of N. dutertrei tests more easily dissolved than did the other parts of the outermost chambers. In the well-dissolved N. dutertrei tests from core KH16-6 St.6 (water depth 4,006 m), the densities of the EDC and inner calcite layer of the outermost chambers were much reduced (Figure 3c). However, unlike the
3.1. Tests of T. sacculifer and G. ruber, in the outermost chambers of N. dutertrei, there was no selective dissolution of the middle calcite layer.

3.2. Change in Density Distribution in Tests

We used CT number histograms to compare the calcite density distributions in the tests of the three species of planktic foraminifera from sediment trap samples and the three seafloor sediment samples of different water depth and Δ[CO₃²⁻] (Figure 3). Among species, the characteristics of the changes in the CT number histogram with progression of test dissolution with depth were similar. First, the CT number histograms for the initial conditions of all three foraminiferal species from the sediment trap sample were unimodal, with a single mode at high CT number (~900). Second, in the CT number histograms of tests in the deep seafloor where water was undersaturated with respect to calcite (e.g., KH16-6 St.3, water depth 3,079 m), a new peak appeared with a mode at a CT number less than 500. Finally, in the CT number histograms of T. sacculifer and G. ruber at the most undersaturated site (KH16-6 St.6, water depth 4,006 m), the peak with a mode at a CT number exceeding 500 disappeared, and a sharp peak with a mode at a CT number less than 500 became prominent. In contrast, the CT number histogram of N. dutertrei at this site had two sharp peaks with modes at CT numbers less than and greater than 500, and the former peak was higher than the latter. Therefore, the process of changes in the CT number histogram of the three foraminiferal species with progression of test dissolution with increasing depth was characterized by a shift from a unimodal density distribution with a high (>500) CT number to a bimodal density distribution, followed by a trend toward a unimodal density distribution with a low (<500) CT number.

3.3. Relationship Between Shift of CT Number Histogram and Deep Seawater Δ[CO₃²⁻]

CT number histograms of planktic foraminiferal tests measured by XMCT scanning provide valuable information about carbonate dissolution intensity (Iwasaki et al., 2015), which we propose as a new proxy for estimating deep seawater Δ[CO₃²⁻]. Because the high and low CT number peaks of CT number histograms can be distinguished at the threshold CT number of 500, and because an area of extremely low CT number (i.e., <200) could include artificial noise from voxels other than those for test calcite, we defined low-CT-number calcite areas as those with CT numbers of 200–500. We calculated %Low-CT-number calcite volume and plotted it and mean CT number (bulk density) of foraminiferal tests versus deep seawater Δ[CO₃²⁻] at each core (Figure 4). The %Low-CT-number calcite volume, which is calculated from the CT number histogram, was significantly and negatively correlated with deep seawater Δ[CO₃²⁻]: T. sacculifer. $R^2 = 0.52, p = 0.044$;
$G.\ ruber$: $R^2 = 0.65$, $p = 0.016$; $N.\ dutertrei$: $R^2 = 0.59$, $p = 0.027$. In contrast, mean CT number only weakly, positively correlated with deep seawater $\Delta[CO_3^{2-}]$: $T.\ sacculifer$, $R^2 = 0.45$, $p = 0.069$; $G.\ ruber$, $R^2 = 0.50$, $p = 0.052$; $N.\ dutertrei$, $R^2 = 0.55$, $p = 0.035$. These results suggest that carbonate dissolution at the deep sea floor caused by undersaturated bottom water should be a significant factor controlling the dissolution intensity of planktic foraminiferal tests in the sediment.

### 3.4. Variations in Conventional Test Weight Proxies

In this study, SNW and test area density of $T.\ sacculifer$, $G.\ ruber$, and $N.\ dutertrei$ tests were measured and compared with deep seawater $\Delta[CO_3^{2-}]$ at each site (Figures 5a and 5b). Neither SNW nor area density of these foraminiferal tests was positively correlated with the deep seawater $\Delta[CO_3^{2-}]$, the suggestion being

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**Figure 4.** %Low-CT-number calcite volume (left) and mean CT number of foraminiferal tests (right) versus deep seawater $\Delta[CO_3^{2-}]$ (μmol kg$^{-1}$) at each sampling site. The square of the correlation coefficient ($R^2$) and the type I error rate ($p$-value) are also shown.
that these test weight proxies are controlled not only by the deep seawater $\Delta[\text{CO}_3^{2-}]$ but also by other factors in this study area. The test area density is a more rigorous size-standardized proxy of $\Delta[\text{CO}_3^{2-}]$ and is also often used as a proxy of calcification intensity at the sea surface. Thus, in order to identify the factors that control the test area density proxy in the study area, we compared the test area density of the three species at each site with two possible controlling factors: the mean thickness of the outermost chamber wall (μm) and %Low-CT-number calcite volume of scanned foraminiferal tests. The solid lines are linear regression of each species. The square of the correlation coefficient ($R^2$) and the type I error rate (p-value) are also shown in (c) and (e).
The test area densities of *G. ruber* were significantly correlated only with the mean thickness of the outermost chamber wall (*G. ruber*: $R^2 = 0.89$, $p = 0.0005^*$) (Figure 5c), the suggestion being that the thickness of the outermost chamber wall is a significant factor controlling the area density of *G. ruber* tests. The test area densities of *T. sacculifer* and *N. dutertrei* did not show significant correlation with the thickness of the outermost chamber wall, although they have a weak positive relationship. On the other hand, there were no negative relationships between test area density and %Low-CT-number calcite volume, which demonstrates the difficulty in identifying the specific factor controlling of test area density of *T. sacculifer* and *N. dutertrei*. In addition, the relationship between the mean thickness of the outermost chamber wall and %Low-CT-number calcite volume was investigated to evaluate the effect of initial test wall thickness on dissolution sensitivity of test (Figure 5e): the relatively high correlation ($R^2 = 0.52$, $p = 0.04$) was found in *T. sacculifer*, suggested that test wall thickness may affect the dissolution sensitivity of *T. sacculifer* test.

In this study, we investigated the difference in the test area density of three species due to the differences in sea surface condition. The sites of multiple core samples in this study can be divided into a northern area located from 31 to 33°N (KH16-6 St. 2, St. 3, St. 5, and St. 6) and a southern area located at 29°N (KH16-6 St. 9, KS15-4 St. 1, 2, and 3). After confirming normality of each data set, a two-tailed Student’s *t* test between each area was performed, and the results showed that the individual test area densities of *T. sacculifer* and *G. ruber* were significantly higher in the southern area than in the northern area ($p < 0.05$): *T. sacculifer* (northern area, $0.12 \pm 0.02 \mu g \mu m^{-2}$, $N = 31$; southern area, $0.14 \pm 0.03 \mu g \mu m^{-2}$, $N = 31$; *t* test, $t = 2.84$, degrees of freedom [df] = 60, $p = 0.003$) and *G. ruber* (northern area, $0.12 \pm 0.02 \mu g \mu m^{-2}$, $N = 30$; southern area, $0.14 \pm 0.03 \mu g \mu m^{-2}$, $N = 25$; *t* test, $t = 3.42$, df = 53, $p = 0.0006$), and no significant difference was found in *N. dutertrei* (northern area, $0.15 \pm 0.04 \mu g \mu m^{-2}$, $N = 35$; southern area, $0.16 \pm 0.03 \mu g \mu m^{-2}$, $N = 32$; *t* test, $t = 0.26$, df = 65, $p = 0.80$) (Figure 6).

**4. Discussion**

4.1. Relationship Between the Dissolution Process and Chamber Wall Structure

As described in Iwasaki et al. (2015), crystalline structure of test walls is closely related to the test dissolution process. The tests of most planktic foraminiferal species are layered with primary calcite (e.g., ontogenetic calcite) around the primary organic sheet, and secondary calcite, which is added on the outer side of the primary calcite (e.g., crust or gametogenic calcite) (Erez, 2003; de Nooijer et al., 2014). Crust generally comprises blocky euhedral crystals that thicken the test wall. Gametogenic calcite, on the other hand, is a
The test formation process with variation of calcite structure is closely related to the distribution of trace elements like Mg, an important paleo-temperature proxy. Based on high-resolution trace element mapping on a cross section of the outermost chamber, it has been suggested that the structure of the spinose species *T. sacculifer* adds several calcite crust layers over the ontogenetic calcite layer, *N. dutertrei* adds several calcite crust layers over the ontogenetic calcite layer (Fehrenbacher et al., 2017). Therefore, as shown by CT scanning in a previous study (Johnstone et al., 2010), the selective dissolution of inner calcite and preservation of outer calcite observed in this study suggest that *N. dutertrei* tests comprise dissolution-prone ontogenetic calcite layers formed during an early life stage and dissolution-resistant calcite crust layers added after the formation of the ontogenetic calcite layer of each chamber; thus, the thinner width of the calcite crust on the final chamber (Steinhardt et al., 2015) may well facilitate the dissolution of the final chamber prior to the younger outermost chambers.

The test formation process with variation of calcite structure is closely related to the distribution of trace elements like Mg, an important paleo-temperature proxy. Based on high-resolution trace element mapping on a cross section of the outermost chamber, it has been suggested that the structure of the spinose species *T. sacculifer* is multi-layered with thin high Mg/Ca bands. This banding has been attributed to intrashell Mg/Ca variability initially resulting from ontogenetic effects (i.e., chamber formation; Sadekov et al., 2005). Comparison of Mg/Ca distributions across chamber walls between non-dissolved and dissolved *T. sacculifer* tests revealed slight selective dissolution of Mg-rich calcite (Sadekov et al., 2010), the suggestion being that test dissolution can slightly influence the bulk Mg/Ca ratios of *T. sacculifer*, and supposedly *G. ruber* as well (Rongstad et al., 2017). Thus, although it is complicated by differences in Mg distribution between individuals and each chambers, dissolution prone calcite may be that in Mg-rich areas in *T. sacculifer* and *G. ruber*. In contrast, across chamber walls of *N. dutertrei* tests, the distribution of Mg/Ca ratios differed significantly between the inner and outer sides, with significantly higher Mg/Ca ratios in the ontogenetic calcite layer and lower Mg/Ca ratios in the calcite crust (Pena et al., 2008; Sadekov et al., 2005; Steinhardt et al., 2015). Of the tests of the three species in this study, the parts of *N. dutertrei* tests characterized by high Mg/Ca ratios are considered to be the most sensitive to dissolution (Dekens et al., 2002). The selective dissolution of the ontogenetic calcite layer on the outer side of the chamber wall with higher Mg/Ca ratios and preservation of calcite crust on the outer side of the chamber wall with lower Mg/Ca ratios seems to effectively reduce the Mg/Ca ratio of *N. dutertrei* as dissolution progresses as shown in Rongstad et al. (2017). If the high Mg/Ca layer in the test was prone to dissolution, the variation in Mg/Ca ratios due to calcification temperature could influence the process and sensitivity of test dissolution. As for *T. sacculifer* and *G. ruber*, the ratio of Mg/Ca at each calcite layer is controlled by sea surface water temperature. However, because the Mg/Ca ratios of low-Mg layers are approximately 20% lower than the whole test average, the average Mg/Ca ratio of a test is considered to be influenced by the initial conditions of the layered structure of test, such as number of layers or crust thickness (Sadekov et al., 2009). This implies that dissolution processes and the sensitivity of foraminiferal tests may be associated with the initial layered structures that are caused by differences in calcification temperature or other biomineralization conditions (e.g., diurnal pH variation); in turn, such structural variations may influence the test dissolution proxy based on XMCT scanning.
Therefore, the extent to which calcification temperature contributes to the variations of foraminiferal test dissolution intensity in sediment samples should be evaluated in the future work with XMCT scanning.

4.2. Foraminiferal Test Dissolution Index Based on CT Number Histogram

Although our 3-D, high-resolution observations based on visualized calcite density distributions in planktic foraminiferal tests showed slightly different dissolution processes among species, changes in the CT number histogram showed a similar pattern among the three species as follows. As dissolution progressed, the CT number histogram of foraminiferal tests shifted from a unimodal density distribution with a high CT number (>500) peak to a bimodal density distribution with high (>500) and low (<500) CT number peaks, and finally toward a unimodal density distribution with a low CT number (<500) peak. Such changes in histograms from unimodal to bimodal density distributions represent selective dissolution of porous and dissolution-prone calcite and preservation of euhedral, dissolution-resistant calcite in the tests (Iwasaki et al., 2015). This characteristic of histogram change was found equally in all three species in this study; we therefore consider that %Low CT-number calcite volume is equally applicable to all three species studied here (T. sacculifer, G. ruber, and N. dutertrei) as a proxy of test dissolution intensity. In the plots of %Low CT-number calcite volume and deep seawater Δ[CO₃²⁻] (Figure 4), %Low CT-number calcite volume of T. sacculifer and G. ruber showed relatively higher values at the shallowest site (KH16-6_St.2) than they are predicted. This suggests that excess dissolution of foraminiferal tests has occurred at this site. This excess dissolution can be caused by the variation in growth conditions at the sea surface, decomposition of organic matter during settlement (Milliman, 1993), or the influence of undersaturated porewater after burial in sediment (Berger, 1970; Hales, 2003), none of which are not controlled by deep seawater Δ[CO₃²⁻]. Because KH16-6 St.2 is located on a continental slope and is closest to the land of all sites in this study, input of terrestrial material is considered to be high. Such a different sedimentary environment than the other sites may have contributed to excess dissolution of foraminiferal tests in the sediment. Furthermore, such sedimentary environment can influence the variability of individual test dissolution intensity in each sediment sample. The standard deviations of %Low CT-number calcite volume between individual tests in each sample set were ±8.0% to 16.4% for T. sacculifer, ±5.2% to 12.7% for G. ruber, and ±4.7% to 13.6% for N. dutertrei (Table S1); thus, we conclude that G. ruber and N. dutertrei, which were characterized by relatively low variability of %Low CT-number calcite volume in each sample set, are more appropriate for carbonate dissolution assessment than is T. sacculifer. In addition, the relatively high correlation between the thickness of the outermost chamber wall and %Low CT-number calcite volume of T. sacculifer implies that variability in test wall thickness between individuals of T. sacculifer, probably caused by differences in intrashell layers number or gametogenic calcite thickness, affects the sensitivity of test dissolution and may cause the larger variability of %Low CT-number calcite volume of T. sacculifer than of the other species. Furthermore, variation between individuals in calcite crust formation may affect the sensitivity of foraminiferal test dissolution. The presence calcite crust and differences in thickness may affect the intrashell density distribution shown by the CT number histogram and sensitivity to dissolution of test, although we suppose that this influence is less than that on test weight. Because crust formation is controlled by several factors (Steinhardt et al., 2015), variation in crust formation between individuals may contribute to higher variability of %Low CT-number calcite volume of T. sacculifer in sediment samples. On the other hand, we found no significant effect of test size (long axis) on %Low CT-number calcite volume, the suggestion being that %Low CT-number calcite volume is independent of test size variation in sample sets. Although several factors that may affect the test dissolution sensitivity should be recognized and the quality of regression between the proxy and deep seawater Δ[CO₃²⁻] must be improved by analyzing samples from additional cores in future works, this proxy is already sufficiently robust to detect glacial-interglacial change in deep seawater [CO₃²⁻], which has been estimated to only approximately ±10 to 20 μmol kg⁻¹ (Qin et al., 2017) or ~28 μmol kg⁻¹ (Fehrenbacher & Martin, 2011) variation in the equatorial Pacific.

4.3. Factors affecting Test Area Density

SNW (μg) and test area density (μg μm⁻²) based on foraminiferal test weight measurements are conventional proxies of dissolution intensity at the deep seafloor and calcification intensity at the sea surface, respectively, although both proxies may be affected by dissolution and calcification intensity. In particular, the test area density, which is the most rigorous size-standardized proxy, is considered to be controlled by both sea-surface and deep seawater conditions: this is because the test area density is normalized by the 2-
D projection test area observed from outside, which means there is no information about internal structure or initial test wall thickness. In contrast, XMCT scanning facilitates observation of the internal structure of foraminiferal tests and measurement in individual foraminiferal tests of physical characteristics, like chamber wall thickness, that affect test area density. The results of this study suggest that the initial thickness of the outermost chamber wall seems to be a principal factor that controls the test area density of *T. sacculifer* and *G. ruber* in the study area (Figure 5c). The calcification intensity of *T. sacculifer* and *G. ruber* has been considered to be principally controlled by seawater carbonate ion concentration where they grow (e.g., Gonzalez-Mora et al., 2008; Marshall et al., 2013; Naik et al., 2010). However, multiple factors other than carbonate ion concentration, like seawater temperature and productivity, have been suggested to control the calcification intensity of these species (e.g., Mohan et al., 2015; Weinkauf et al., 2016). Although this study did not aim to identify specific environmental factors that control test area densities of foraminiferal test, test area densities, in particular *G. ruber*, were correlated with initial chamber wall thickness (Figure 5c) and they were significantly heavier in the southern area (Figure 6), the suggestion being that test area densities of these species are affected by test initial condition due to the differences in the sea surface environment.

The CTD and bottle sampling data from GLODAP stations near these areas (northern area: 29.0°N, 137.0°E; southern area: 32.3°N, 137.0°E) suggest that seawater temperature and [CO$_3^{2–}$] at ~10 m depth in summer are approximately 0.5 °C and 5.5 μmol kg$^{-1}$ higher in the southern area than in the northern area. Furthermore, the shift of the Kuroshio current on decadal to orbital timescales directly influences the surface conditions in this area. Therefore, we consider that changes in sea surface conditions on glacial-interglacial timescales at this study area can influence the test wall thickness during the growth stage, and that such changes are not negligible in assessments of test dissolution intensity of *T. sacculifer* and *G. ruber* based on test area density. In contrast, in this study we did not find a specific factor that controls the area density of *N. dutertrei*. The calcification intensity of *Neogloboquadrina* foraminifera is known to be affected by seawater condition (i.e., carbonate ion concentration or temperature) where they grow (e.g., Barker & Elderfield, 2002; de Villiers, 2003; Mohan et al., 2015). Furthermore, the crust on *N. dutertrei* tests is known to have been thicker during the LGM than in modern times (Jonkers et al., 2012), the suggestion being that the initial test condition of *N. dutertrei* should be affected by sea surface conditions. However, the assessment of *N. dutertrei* dissolution processes in this study revealed significant loss of calcite from EDC and the inner surface of the outermost chamber, which shows the thinning of the outermost chamber wall with test dissolution. The thinning of chamber walls could lead to underestimation of measurement of initial chamber wall thickness. Thus, we speculate that the test area density of *N. dutertrei* obtained from this study area seems to be controlled by both initial test condition and dissolution intensity; if so, then it is difficult to identify the specific controlling factor. From these results, therefore, to assess the carbonate dissolution intensity at the deep seafloor, we suggest that XMCT scanning, which can independently assess the effects of initial test condition, is a more appropriate proxy than is test weight measurement.

5. Conclusions

On the basis of observations of internal structure and estimation of calcite density in tests of three species of planktic foraminifera obtained from multiple core samples in the midlatitudes of the western North Pacific, we inferred characteristics of the test dissolution process of each species based on 3-D isosurface images and CT number histograms as follows:

1. Selective dissolution of the intermediate calcite layer of the outermost chamber wall was found in tests of *T. sacculifer* and *G. ruber*. In contrast, we found selective dissolution of the inner calcite layer of the outermost chamber wall in tests of *N. dutertrei*. These characteristics of the test dissolution process seem to be related to chamber wall structure associated with the chamber calcification process.

2. The changes in the pattern of CT number histogram with test dissolution are common to all three species of planktic foraminifera. These changes include a shift from a unimodal to bimodal density distribution with increasing depth and dissolution. This characteristic of CT number histogram change is quantified by %Low-CT-calcite volume, which facilitates evaluation of the intensity of test dissolution.

3. The %Low-CT-number calcite volume calculated by histogram correlated with the deep seawater Δ[CO$_3^{2–}$]. This correlation implies that identification of changes in deep seawater [CO$_3^{2–}$] on glacial-interglacial timescales is possible. However, the test area density based on weight measurement is affected by factors other than deep seawater Δ[CO$_3^{2–}$], such as variability of chamber wall thickness.
In addition to the tests of *G. bulloides* investigated by a previous study (Iwasaki et al., 2015), we conclude that the %Low-CT-number calcite volumes based on XMCT scanning is applicable to other planktonic foraminiferal species reported in this study and for assessing carbonate dissolution intensity at the deep seafloor in the midlatitudes of the western North Pacific, where foraminiferal test weight measurements provide no records of carbonate dissolution fluctuation.

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