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1	Assessing the impact of diagenesis on foraminiferal geochemistry from a low latitude,
2	shallow-water drift deposit
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31	IODP
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#### 34 ABSTRACT

35 Due to their large heat and moisture storage capabilities, the tropics are fundamental in modulating both regional and global climate. Furthermore, their thermal response during past 36 37 extreme warming periods, such as super interglacials, is not fully resolved. In this regard, we 38 present high-resolution (analytical) for a miniferal geochemical ( $\delta^{18}$ O and Mg/Ca) records for 39 the last 1800 kyr from the shallow (487 m) Inner Sea drift deposits of the Maldives 40 archipelago in the equatorial Indian Ocean. Considering the diagenetic susceptibility of these 41 proxies, in carbonate-rich environments, we assess the integrity of a suite of commonly used 42 planktonic and benthic foraminifera geochemical datasets (Globigerinoides ruber (white), 43 Globigerinita glutinata (with bulla), Pulleniatina obliquiloculata (with cortex) and Cibicides 44 mabahethi) and their use for future paleoceanographic reconstructions.

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46 Using a combination of spot Secondary Ion Mass Spectrometer, Electron Probe Micro-47 Analyzer and Scanning Electron Microscope image data, it is evident that authigenic 48 overgrowths are present on both the external and internal test (shell) surfaces, yet the degree 49 down-core as well as the associated bias is shown to be variable across the investigated 50 species and proxies. Given the elevated authigenic overgrowth Mg/Ca (~12–22 mmol/mol) and  $\delta^{18}$ O values (closer to the benthic isotopic compositions) the whole-test planktonic G. 51 52 ruber (w) geochemical records are notably impacted beyond ~627.4 ka (24.7 mcd). Yet, 53 considering the setting (i.e. bottom water location) for overgrowth formation, the benthic 54 for a for a single the single test in the single test of t 55 beyond ~790.0 ka (28.7 mcd). Even though only the top of the G. ruber (w) and C. mabahethi 56 records (whole-test data) would be suitable for paleo-reconstructions of absolute values (i.e. sea surface temperature, salinity, seawater  $\delta^{18}$ O), the long-term cycles, while dampened, 57 58 appear to be preserved. Furthermore, planktonic species with thicker-tests (i.e. P.

59	obliquiloculata (w/c)) might be better suited, in comparison to thinner-test counter-parts (i.e.
60	G. glutinata (w/b), G. ruber (w)), for traditional whole-test geochemical studies in shallow,
61	carbonate-rich environments. A thicker test equates to a smaller overall bias from the
62	authigenic overgrowth. Overall, if the diagenetic impact is constrained, as done in this study,
63	these types of diagenetically altered geochemical records can still significantly contribute to
64	studies relating to past tropical seawater temperatures, latitudinal scale ocean current shifts
65	and South Asian Monsoon dynamics.
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#### 84 1. INTRODUCTION

85 In comparison to deep-sea records, high-resolution (both temporal and analytical)

86 paleoclimate reconstructions for shallow-intermediate, equatorial regions are limited. This is 87 in part due to (i) the drive to reconstruct and model past global ocean circulation patterns, 88 which predominantly use high-temporally resolved deep-sea archives, and which are difficult 89 to recover from lower latitudes and (ii) the increased susceptibility of calcifying organisms to 90 diagenetic overprint in shallow, carbonate-rich environments. The tropics are an integral 91 component of both regional and global climatic systems due to their heat and moisture storage 92 capabilities (Gupta et al., 2010 and references within; Hastenrath et al., 1993; Schott et al., 2009). Yet, notably there is a lack of reliable sea surface temperature (SST) and  $\delta^{18}$ O records 93 94 from these equatorial regions, particularly from past extreme warming periods, which would 95 further facilitate the understanding of tropical climatic systems. In this regard, we present a 96 new high-resolution (analytical) tropical record from the Inner Sea of the Maldives 97 archipelago in the equatorial Indian Ocean. We test the suitability of this unique tropical 98 record for paleo-reconstructions given its shallow location, proximity to carbonate banks and 99 the anticipated diagenetic alteration down-core.

100

101 Innumerable studies have shown that the geochemistry of foraminiferal tests (shells) are 102 invaluable paleoceanographic, -climatic and -ecological archives (e.g. Birch et al., 2013; 103 Kroon and Ganssen, 1989; Lisiecki and Raymo, 2005; Groeneveld et al., 2008; Raddatz et al., 104 2017; Stainbank et al., 2019). Yet, many of these geochemical proxies (e.g. Mg/Ca and  $\delta^{18}$ O) 105 are susceptible to diagenetic alteration, which calls into question their fidelity (Edgar et al., 106 2015, 2013; Groeneveld et al., 2008; Panieri et al., 2017). According to Edgar et al. (2015) the 107 foraminifera test wall, texture and morphology are prone to at least three diagenetic processes: 108 overgrowth, partial dissolution and recrystallization. All three have the potential to bias the

109 original test geochemistry and affect the accuracy of paleo-reconstructions. As such, prior to 110 undertaking paleo-reconstructions, it is paramount to establish the diagenetic state of this 111 foraminiferal calcite. This is particularly important within carbonate-rich environments which 112 have high sediment porosity and permeability and thus a higher "diagenetic potential" in 113 comparison to the more impermeable clay-rich sediments (Edgar et al., 2013). Additionally, 114 in comparison to the more robust, heavily calcified benthic foraminifera, planktonic tests are 115 more susceptible to alteration (Edgar et al., 2013). Moreover, as there is a large array of 116 planktonic foraminifera test wall textures; this influence is not necessarily uniform between 117 species. Bé (1977) correlated increased diagenetic resistance for species with smaller pores, 118 thicker walls and larger test size. These considerations are important to aid in the selection of 119 less diagenetically predisposed for aminiferal species, particularly for depth stratified (i.e. 120 studies assessing discrete living and calcification depths within the water column) thermocline 121 reconstructions.

122

123 To contribute to an improved understanding of ocean-atmospheric interactions within the 124 tropical Indian Ocean, and the associated foraminiferal diagenetic processes, we present high 125 analytically resolved foraminiferal datasets from the shallow, Maldives Inner Sea. 126 Specifically, our study addresses the following issues: (i) the overall preservation state of the 127 individual long-term geochemical records and their suitability for future paleoceanographic 128 interpretations and (ii) the differential susceptibility to diagenesis for three planktonic 129 foraminiferal species, with discrete wall textures, and a benthic counterpart using a 130 combination of imaging and elemental/isotopic geochemical approaches. 131

132 **2. METHODS** 

133 This study uses samples retrieved during the International Ocean Discovery Program (IODP) 134 Expedition 359 in 2015. All samples come from Site U1467 (4°51.0274'N, 73°17.0223'E) 135 drilled at a water depth of 487 m within the drift deposits of the Inner Sea of the Maldives 136 archipelago in the northern equatorial Indian Ocean (Fig. 1) (Betzler et al., 2017). The base of 137 these drift deposits have been biostratigraphically dated to 12.9-13.0 Ma, coinciding with the 138 establishment of the South Asian Monsoon (Betzler et al., 2018, 2016). As such, the drift 139 deposits represent an important paleoceanographic and -monsoon archive due to their unique 140 ability to preserve continuous sediment records by bottom current deposition (Lüdmann et al., 141 2013). The top 61 meters composite depth (mcd) from Hole U1467B, C, composed of 142 unlithified foraminifer-rich wackestone to packstone, were used in this study. The shipboard 143 splice was adjusted after the Expedition based on X-ray fluorescence core scanner data using 144 Fe intensity (counts per second, cps) (Kunkelova et al., 2018). 145 146 Fig. 1. Study location and position of IODP Expedition 359 Site U1467 in the Inner Sea 147 of the Maldives (blue circle), located in the northern equatorial Indian Ocean (GEBCO

148 **Compilation Group, 2019).** 

149

150 Long-term, high-resolution (analytical) whole-test (whole-shell) stable isotopic ( $\delta^{18}$ O) records

151 are compiled for three planktonic species, *Globigerinoides ruber* (white), *Globigerinita* 

152 glutinata (with bulla), Pulleniatina obliquiloculata (with cortex) and one benthic

153 representative *Cibicides mabahethi*. In a few cases when the target benthic species is rare,

- 154 *Cibicides wuellerstorfi* tests are included to ensure enough calcite for the isotopic
- 155 measurements. Complementary whole-test Mg/Ca ratios are measured for *G. ruber* (w).
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157 A combination of methods is utilized to constrain the influence and susceptibility of 158 diagenetic alteration on the foraminiferal test geochemistry. A visual Scanning Electron 159 Microscope (SEM) check is implemented, on selected samples, for both the target planktonic 160 and benthic species. This visual check is then used to define a qualitative Diagenesis Rank 161 (DR) to ascertain and track the down-core change in the preservation state of the foraminiferal 162 tests. Supplementary spot-geochemical (isotopic and elemental) measurements are obtained 163 using a Secondary Ion Mass Spectrometer (SIMS) and Electron Probe Micro-Analyzer 164 (EPMA).

165

#### 166 2.1. Site U1467 mineralogy and geochemistry

167 Site U1467 sediment core interstitial water chemistry and sediment mineralogy are presented in Betzler et al. (2017). Concentrations of  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Sr^{2+}$  were measured from interstitial 168 169 water (IW) samples either by shipboard Inductively Coupled Plasma Atomic Emission 170 Spectroscopy (ICP-AES) or shore-based ion chromatography (see Betzler et al., 2017; Blättler 171 et al., 2019 for further details). Relative concentrations of aragonite, high-Mg calcite (HMC), 172 low-Mg calcite (LMC), dolomite, and quartz were measured using X-ray powder diffraction 173 (XRD) (see Betzler et al., 2017 for a full overview of the data collection methods). For 174 simplicity we show only the data, reported as mbsf (meters below seafloor), from Hole 175 U1467B as it yielded the most extensive and intact records at this site (Fig. 2). 176 177 Fig. 2. IODP Expedition 359 Hole U1467B sediment mineralogy and pore water

178 **geochemistry** (Betzler et al., 2017).

179

180 **2.2. Species selection criteria** 

181	To explore the susceptibility of different species to diagenetic alteration and the biases on
182	their respective whole-test geochemical compositions, we assess three shallow/intermediate
183	dwelling planktonic foraminiferal species: G. ruber (w), G. glutinata (w/b) and P.
184	obliquiloculata (w/c). These species have reported apparent calcification depths for the
185	northern equatorial Indian Ocean of ~70 m, 81 m and 74–104 m, respectively (Stainbank et
186	al., 2019) and have discrete wall textures, thicknesses and porosities (Supplementary Material
187	1). Globigerinoides ruber (w) is chosen as it is the predominant foraminiferal species used for
188	surface/near-surface seawater reconstructions (e.g. Birch et al., 2013; Bunzel et al., 2017;
189	Rebotim et al., 2017; Stainbank et al., 2019). Globigerinoides ruber (w) is a spinose,
190	shallow-dweller with a cancellate wall texture and a porosity of ~5 pores/25 x 25 $\mu m$ (Bé,
191	1968) (Supplementary Material 1). Globigerinita glutinata (w/b) is included as a second
192	shallow-dweller, as it is non-spinose with a smooth, microperforate wall texture with a
193	porosity of ~112 pores/25 x 25 µm (Bé, 1968; Birch et al., 2013; Stainbank et al., 2019)
194	(Supplementary Material 1). Lastly, the shallow-intermediate dweller P. obliquiloculata (w/c)
195	is included as it is non-spinose, with a smooth wall texture (Birch et al., 2013; Stainbank et
196	al., 2019). While the original test has a porosity of ~9 pores/25 x 25 $\mu$ m, the formation of the
197	smooth, glossy cortex layer drastically reduces the diameter and spherical nature of the pores
198	resulting in a reduction in the overall porosity (Bé, 1968; Steinhardt et al., 2015)
199	(Supplementary Material 1). Cibicides mabahethi, a calcareous hyaline epibenthic species, is
200	chosen as the benthic representative for this site as it has been shown to accurately record
201	bottom water conditions for the first ~200 kyr (time interval of the study by Bunzel et al.,
202	2017) for the Maldives Inner Sea (Bunzel et al., 2017).
203	

#### 204 2.3. Traditional whole-test for aminiferal geochemical analysis

Sediment samples were air dried, weighed and washed through a 32 µm sieve to remove the finer silts and clay fraction. They were then oven-dried at 30°C for 48 hours, weighed and dry sieved into discrete fractions for foraminiferal picking. For all geochemical analyses, species were picked from pre-defined, narrow sieve size fractions, to minimize intra-specific ontogenetic isotopic fractionation effects (Stainbank et al., 2019). Additionally, to remove any adhering particles, and to prevent carbonate contamination, all picked specimens were precleaned in Milli-Q water for a few seconds by ultrasonication.

In total, 3802 traditional whole-test stable oxygen isotopic ( $\delta^{18}$ O) measurements were made 213 214 (data is available on Pangaea). From 0–61 mcd a measurement for G. ruber (w: 212–250 µm) 215 was obtained every  $\sim 3$  cm. Isotopic analyses of G. glutinata (w/b: 125–150 µm) and P. 216 obliquiloculata (w/c: 355–400 µm), were carried out across three selected intervals with 217 differential diagenetic influences (see Section 3.2 for more details) at a resolution of 3-12 cm; 218 Interval 1 (15.2–20.5 mcd), Interval 2 (31.4–34. 5 mcd) and Interval 3 (53.3–56.1 mcd). 219 Benthic isotopic data (C. mabahethi > 180  $\mu$ m) were only generated at a ~3 cm resolution 220 between 0–40.0 mcd as below this interval they become scarce. The G. ruber (w) and C. 221 *mabahethi*  $\delta^{18}$ O measurements for the top 6.7 mcd (n = 441) were analyzed at Rutgers 222 University on a MicroMass (FISONS) Optima Isotope Ratio Mass Spectrometer with an 223 attached multi-prep device. Samples were reacted with 100 % phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) at 224 90°C for 15 min, and the evolved CO<sub>2</sub> gas collected in a liquid nitrogen coldfinger and 225 analyzed compared to a reference gas. The samples are corrected using an internal laboratory standard (RGF1), which is routinely run against NBS-19 and NBS-18. All other  $\delta^{18}$ O data (n 226 = 3361) were obtained at the Grant Institute of the University of Edinburgh on a Thermo 227 228 Electron Delta+ Advantage Mass Spectrometer integrated with a Kiel carbonate III automated 229 extraction line and corrected using the laboratory's internal standard. Approximately 0.05 mg

230 was required for each measurement with each expressed as parts per mil (‰) relative to

231 Vienna Pee Dee Belemnite (VPDB). Replicate measurements of the standards give the

instruments an analytical precision (1 $\sigma$ ) of ~0.05 ‰ for  $\delta^{18}$ O and  $\delta^{13}$ C. The reproducibility,

based on inter-laboratory replicate measurements (n = 6), was ~0.20 ‰.

234

235 Mg/Ca ratios were measured on 30 pooled G. ruber (w: 212–250 µm) tests from a sample 236 every  $\sim 3$  cm, for the interval 14.8–31.4 mcd (n = 535; data is available on Pangaea). All 237 samples were cleaned according to the standard oxidative protocol of Barker et al. (2003). 238 Samples were then leached with a very weak 0.001N HNO<sub>3</sub> acid solution prior to dissolving 239 in 0.075M HNO<sub>3</sub>. The reductive cleaning step was excluded as it has been shown to cause the 240 dissolution of the foraminiferal tests resulting in subsequently lower Mg/Ca values (Barker et 241 al., 2003; Elderfield et al., 2006). Analyses were conducted at the Institute of Geosciences of 242 the Goethe University of Frankfurt and at the School of GeoSciences at the University of 243 Edinburgh. The former measurements (n = 232) were carried out by Inductively Coupled 244 Plasma Optical Emission Spectrometry (ICP-OES) on a Thermoscientific iCap 6300 (dual 245 viewing) and the latter (n = 303) on a Vista Pro ICP-OES. Inter-laboratory replicates (n = 11)246 were run to assess potential differences and the reproducibility which was determined to be 247 better than 0.30 mmol/mol (mean difference). In Frankfurt, the final centrifuged sample 248 solutions were diluted with an yttrium solution (1 mg/l) prior to measurement to allow for the 249 correction of matrix effects. Whereas, in Edinburgh a set of six standard solutions (with 250 increasing Ca concentrations from 0 to 100 %) were used for matrix and background 251 correction (Sadekov et al., 2013). At both facilities, five calibration solutions were measured 252 before each analysis to allow for intensity ratio calibrations. Element/Ca measurements were 253 standardized using an internal consistency standard (ECRM 752-1, 3.761 mmol/mol Mg/Ca). 254 Replicates of the ECRM at the Edinburgh (n = 61) and Frankfurt (n = 45) facilities yielded  $2\sigma$  255 Mg/Ca uncertainties (SD) of 0.15 and 0.17 mmol/mol, respectively. During all Mg/Ca

256 measurements, the elements Al, Fe, and Mn were screened to check for Mn-Fe oxide coatings

and clay mineral contamination. Procedural blanks were routinely run to monitor for potential

258 contamination during the cleaning process.

259

#### 260 **2.4. Electron Probe Micro-Analyzer (EPMA) analysis**

261 Samples for both EPMA and SIMS (Section 2.5) analyses were selected to provide 262 representatives from the entire length of core including altered versus non-altered portions 263 (see Section 3.2 for sample locations). Polished cross-sections of G. ruber (w) (n = 36) and P. 264 *obliquiloculata* (w/c) (n = 6) tests were prepared by embedding the tests in Struers Epothin. 265 Briefly, selected tests were (i) cleaned according to the same oxidative protocol (Barker et al., 266 2003) as for the traditional whole-test Mg/Ca geochemical analyses and (ii) placed umbilical 267 side-down on adhesive film within 1-inch Struers mounting cups, then (iii) embedded in 268 Epothin and degassed under high vacuum for 5 minutes and once set (iv) polished using a 269 successively finer Struers water-based diamond suspension from 9 µm down to 0.25 µm to 270 expose a cross-section of each test (Supplementary Material 2). All EPMA samples were then coated in 5 nm of carbon. 271

272

273 Quantitative Mg and Ca spot analyses and semi-quantitative elemental maps were measured 274 using a JEOL JXA-8530F Electron Probe Micro-Analyzer, equipped with a Schottky field 275 emission gun, at the Faculty of Geosciences and Environment of the University of Lausanne. 276 Calcite, CaCO<sub>3</sub> (Ca = 39.98 wt%) and dolomite, CaMg(CO<sub>3</sub>)<sub>2</sub> (Mg = 13.18 wt%) were used 277 as standards for Ca and Mg, respectively. Spot measurements were acquired using a 10 kV 278 accelerating voltage and probe size of  $1-2 \mu m$ . Overview elemental maps were carried out 279 with a 0.50 µm step size, an image resolution of 720 × 600 pixels, 7 kV accelerating voltage,

an electron beam current of 10 nA and a dwell time of 50 ms. Additional higher resolution elemental maps were obtained with a  $0.23-0.29 \mu m$  step size and an image resolution of  $256 \times 192$  pixels.

283

#### 284 2.5. Secondary Ion Mass Spectrometer (SIMS) analysis

285 Polished cross sections, of selected G. ruber individuals (n = 27) were prepared for SIMS 286 analyses following the same protocol as for the EPMA samples (Section 2.4), with the 287 exclusion of the initial oxidative cleaning step. In situ oxygen isotope analyses were carried 288 out using a Cameca IMS 1280-HR Secondary Ion Mass Spectrometer (SIMS) at the 289 SwissSIMS ion probe facility within the University of Lausanne (Supplementary Material 3). 290 A primary Cs+ ion beam with an intensity of 1.2nA was focused to a size of  $\sim 10 \,\mu\text{m}$ . The 291 secondary <sup>16</sup>O and <sup>18</sup>O were detected simultaneously on two faraday cups, which were 292 calibrated at the beginning of the session. The mass resolving power was set to 2400 (entrance slit set at 121  $\mu$ m and multicollection slit 1), in order to fully remove possible <sup>17</sup>OH 293 294 interference on mass 18. A single analysis took ~4 min, including 30 sec. of pre-sputtering, 295 automatic centering of secondary deflectors and 20 cycles of 5 sec. of measurements. 296 Calibration was made using an in-house pure calcite standard (UNIL-C1). Six analyses of the 297 standard were made at the beginning of the measurements for each mount, after which UNIL-298 C1 was measured once every four analyses. Reproducibility of the standard (spot to spot) was 299 0.4 and 0.3 % (2 $\sigma$ ) on the two mounts, respectively. Internal uncertainty (single analysis) was 300 0.3 to  $0.4 \% (2\sigma)$ .

301

#### 302 **2.6. Scanning Electron Microscopy (SEM)**

Thirty-five samples from the top 61 mcd of the core are used to constrain the preservationstate of the target planktonic and benthic foraminiferal species. Samples are positioned at

305	regular intervals down-core and include representatives from glacial and interglacial peaks
306	(see Section 3.2 for sample locations). From each sample, three to four G. ruber (w), G.
307	glutinata (w/b), P. obliquiloculata (w/c) and C. mabahethi/wuellerstorfi tests were picked
308	from the same size fractions as used for the geochemical analyses. Tests were cleaned for a
309	few seconds by ultrasonication in Milli-Q water, cracked open and mounted on carbon based
310	adhesive discs on standard SEM stubs. Stubs were coated, under vacuum, in 40 nm of gold
311	using a Bal-tec SCD 050 sputter coater and imaged on a Thermo Fisher SEM FEIXL30SFEG.
312	The preservation state of each species for each sample is ascertained, using SEM images,
313	according to three categories (i) external test surface, (ii) internal test surface and (iii) test
314	wall texture (Table 1). A ranking is assigned for each category and the sum (total out of 7)
315	used to define the overall qualitative Diagenesis Rank (DR) for each species, with 0 defining
316	'pristine tests' and 7 the most 'diagenetically altered'.

317

318 **Table 1. Diagenesis Rank categories and classifications.** 

Danking		Categ	gory		
Kalikilig	1. External surface	2. Internal surface	3. Wall texture		
	Pristine unaltered sur	rface texture with no	Pristine microgr	anular wall texture with	
0	authigenic overgrow	th crystals present	well-defined por	es	
	both on the test surface	ce or within the pores			
	Small overgrowths	present (up to ~0.5	Visibly altered v	vall texture with	
1	$\mu$ m) with a patchy-uniform distribution		authigenic crystals within the wall cross		
1			sections either d	ue to overgrowth	
			precipitation or p	partial recrystallization	
2	Mostly medium sized	overgrowths present			
2	(~0.5-2 $\mu$ m) with a u	niform distribution			
2	Mostly large sized	overgrowths present			
5	$(>2 \ \mu m)$ with a unifo	rm distribution			

319

320 Using the MATLAB v2018a software, a color-map is generated for each species to visualize

321 the down-core evolution in preservation state (i.e. the change in DR). The 35 samples are used

322 to define an initial matrix, and the rankings interpolated between each point using a linear

323 function.

324

#### 325 **2.7. Age model**

326 The stratigraphic framework for the upper 61 mcd of Site U1467 is established by graphically correlating both our planktonic G. ruber (w) and benthic C. mabahethi  $\delta^{18}$ O records to the 327 328 stacked reference curve of Ahn et al. (2017) (Prob-stack). Prob-stack was selected as it is 329 based on a compilation of 180 global benthic isotope records and includes the datasets from the frequently used LR04  $\delta^{18}$ O stack of Lisiecki and Raymo (2005). The age model for our 330 331 datasets is obtained using Analyseries v2.0.4 (Paillard et al., 1996). To facilitate the tuning 332 process, initially all datasets were linearly detrended and our planktonic and benthic records 333 smoothed (3 pt. running average) to eliminate some of the noise. The main glacial and 334 interglacial peaks are used as the initial tie-points, with additional prominent maxima and 335 minima subsequently correlated to optimize the visual alignment of the datasets 336 (Supplementary Material 4).

337

#### **338 3. RESULTS**

#### 339 **3.1.** Carbonate mineralogy

340 At IODP Expedition 359 Hole U1467B, LMC is ubiquitous, generally contributed by 341 planktonic foraminifera and coccoliths, making up the bulk of the sediment mineralogy from 342 the core-top down to 700 mbsf (Fig. 2, Betzler et al., 2017). Aragonite is contributed mainly 343 by Halimeda fragments, coral fragments and pteropods, and decreases from ~46 % between 344 0-10 mbsf to only 9 % at 200 mbsf. From 200 to 500 mbsf aragonite still has a small, variable 345 contribution (0-9 %). However, a marked increase in the aragonite contribution is noted from 500-600 mbsf (up to 23 %) with again a decrease down to 700 mbsf. HMC (with Mg<sup>2+</sup> 346 347 concentrations >4 mol%; Morse, 2003), which is contributed predominantly by red coralline algae, benthic foraminifera, bryozoans, and echinoderms, is only present (4–7 %) in the 348

sediments in the top 10 mbsf. Minor peaks in dolomite mainly occur within the top 400 mbsf. The pore-fluid species ( $Ca^{2+}$ ,  $Mg^{2+}$  and  $Sr^{2+}$ ) are visibly correlated with the aragonite concentrations (Betzler et al., 2017). Both  $Ca^{2+}$  and  $Mg^{2+}$  respectively, increase and decrease concomitantly with a reduction in aragonite percent contribution.  $Sr^{2+}$  increases rapidly to peak concentrations around 100 mbsf where it plateaus until ~500 mbsf below which there is a gradual decline.

355

The study interval (0 to ~61 mbsf), corresponds to Unit I (0–110 mbsf) of the logged lithostratigraphic units (a total of 6 units were identified) at Site U1467 (Betzler et al., 2017). It consists of an unlithified foraminifer-rich wackestone to packstone (very fine to fine grained) with a calcareous ooze matrix (Betzler et al., 2017). As noted, the abundance of aragonite is high in the study interval with HMC present in the first 10 mbsf. The species Ca<sup>2+</sup> and Sr<sup>2+</sup> are present in their lowest concentrations (mean Ca<sup>2+</sup>: 10.61 mM, Sr<sup>2+</sup> 111.42 mM) whereas, Mg<sup>2+</sup> is in its highest concentration (mean Mg<sup>2+</sup>: 53.58 mM) (Betzler et al., 2017).

#### 364 3.2. Traditional whole-test foraminiferal geochemistry and Diagenesis Ranks

All geochemical records ( $\delta^{18}$ O and Mg/Ca) show clear cyclicity down-core (Figs. 3-4). Based on the alignment of the stable isotope stratigraphies, the planktonic  $\delta^{18}$ O *G. ruber* (w) record from 0–61.0 mcd represents the time interval 0–1800.0 kyr covering Marine Isotope Stage (MIS) 1-64 (Figs. 3a, 4a-b). The benthic record, spanning 0–40.0 mcd, encompasses the interval 0 to 1179.0 kyr (Figs. 3c, 4a,c), MIS1-35. The sedimentation rate varies down-core between 0.4–16.3 cm/kyr (Fig. 4d).

371

**Fig. 3. Long-term geochemical records from IODP Expedition 359 Site U1467 against** 

373 core depth with (a) the *G. ruber* (w)  $\delta^{18}$ O, (b) Mg/Ca records and (c) the *C. mabahethi* 

δ<sup>18</sup>O record. Visual SEM check (grey circles) and EPMA/SIMS (red and black circles,
respectively) samples are shown for reference. A 3-pt moving average smoothing was
applied to our datasets (thick lines) to highlight the cycles.

377

Fig. 4. (a) The benthic foraminifera  $\delta^{18}$ O stack (prob-stack) of Ahn et al. (2017) is used to obtain the age model for (b) IODP Expedition 359 Site U1467 *G. ruber* (w) and (c) *C. mabahethi*  $\delta^{18}$ O records. A 3-pt moving average smoothing was applied to our datasets (b, c: thick lines) to highlight the cycles. Sedimentation rates (Sed. Rate) are referenced as cm/kyr in (d). Marine Isotope Stages (MIS) are shown (vertical dashed lines) and form the main tie-points for the age model (Supplementary Material 4).

384

385 SEM observations reveal diagenetic alterations are primarily due to the precipitation of

authigenic calcite (overgrowth) (Fig. 5). Furthermore, the diagenetic susceptibility of the four

387 investigated species, G. ruber (w), G. glutinata (w/b), P. obliquiloculata (w/c) and C.

388 mabahethi, differs down-core (Figs. 5-6). The resultant Diagenesis Rank (DR) values reflect a

389 gradual decline, in the overall preservation state from 0 to 1800.0 kyr (0–61.0 mcd) (sample

390 locations noted on Figs. 3, 6). While this down-core decline is variable across all species, all

391 have pristine, unaltered tests in the core-top samples (359-U1467A, B-Mudline) (Fig. 5).

392 Further down-core, authigenic overgrowths occur both on the internal and external test

393 surfaces and within pores, resulting in a reduction in pore-size until complete infilling is

394 achieved (Fig. 5, Supplementary Material 1). The size of these authigenic overgrowths varies

from small and patchy to large (>  $2 \mu m$ ) well-developed, euhedral crystals growing

396 perpendicular to the test wall and covering the entire inner and outer test surfaces

397 (Supplementary Material 1).

398

399 Fig. 5. Scanning Electron Microscope (SEM) images of (a) G. ruber (w), (b) G. glutinata 400 (w/b), (c) P. obliquiloculata (w/c) and (d) C. mabahethi/C. wuellerstorfi showing the outer 401 (1, 2 and 4) and inner test surfaces (3, 5). Images of pristine IODP Expedition 359 Site 402 U1467 tests from the core-top (Sample at 0 ka: A, B-Mudline) are shown in 1-3 and 403 images of diagenetically altered tests (Sample at 1743.8 ka, 58.7 mcd: B-7H-3, 75-76 cm) 404 are shown in 4-5. Representative examples of overgrowth size and distribution are 405 shown for the inner test surfaces of G. ruber (w) in (e-h) (Samples: e. B-Mudline; f. B-406 3H-4, 99-100 cm; g. B-4H-5, 36-37 cm; h. C-6H-5, 57-58 cm). White bars show test wall 407 thicknesses and red arrows indicate representative examples of overgrowth crystals. 408 Whole-test scale bars =  $100 \mu m$  and close-up image scale bars =  $20 \mu m$ . 409 410 Fig. 6. Long-term stable isotope ( $\delta^{18}$ O) records for IODP Expedition 359 Site U1467 for 411 (a) the planktonic species G. ruber (w) with intervals 1-3 comparing G. glutinata (w/b) 412 and P. obliquiloculata (w/c) records (note: Selected Marine Isotope Stages (MIS) are 413 shown for reference); Diagenesis Rank (DR) color maps are shown for (b) G. ruber (w), 414 (c) G. glutinata (w/b), (d) P. obliquiloculata (w/c) and (e) C. mabahethi with (f) the long-415 term  $\delta^{18}$ O record for the benthic species *C. mabahethi*. The samples used for the visual SEM check are also shown as grev circles on (a). Red and teal curves show  $\delta^{18}O$  *G. ruber* 416 417 (w) and C. mabahethi data, respectively from Bunzel et al. (2017). A 3-pt moving average 418 smoothing was applied to the datasets (thick lines) to highlight the cycles. 419 420 *Pulleniatina obliquiloculata* (w/c) appears to be the most susceptible species to authigenic

421 overgrowth with medium sized, well-developed crystals already observed at ~274.5 ka (11.6

422 mcd) on the external test surfaces. In contrast, these are first observed on *G. ruber* (w) at

423 ~790.0 ka (28.7 mcd), G. glutinata (w/b) at ~558.3 ka (22.2 mcd) and the benthic tests further

424 down core at ~807.9 ka (29.1 mcd). Large well-developed authigenic crystals are evident on 425 tests of G. ruber (w), G. glutinata (w/b), P. obliquiloculata (w/c) and C. mabahethi from 426 ~1190.9 ka (40.6 mcd), ~1015.1 ka (36.0 mcd), ~864.7 ka (31.3 mcd) and ~872.0 ka (31.7), 427 respectively (Supplementary Material 1). In addition to size, differences in overgrowth 428 distributions are noted between the planktonic and benthic species. While all species show 429 roughly similar development of overgrowth crystals across their outer test surface, with 430 concentrations of overgrowths in the pores, distinct differences are observed on their inner 431 test surfaces. For the planktonic species, authigenic overgrowths are more or less 432 homogeneous across the surface whereas for the benthic species, they are generally 433 concentrated within the pores and along the sutures with patchy growth across the inner 434 surfaces (Supplementary Material 1).

435

436 Intervals 1-3 were specifically selected, with reference to the G. ruber (w) DR data, as 437 representative segments with differential diagenetic influences. Variability between the three 438 planktonic  $\delta^{18}$ O records does occur across the intervals (Fig. 6). Interval 1, which covers 439 MIS10-13 (354.0–505.0 kyr), has the lowest DR values for all species (0–3). Interval 2, which 440 covers MIS21-26 (866.7–974.0 kyr), is located in the section with a notable increase in the 441 respective DR values (2-4). Finally, Interval 3 covers MIS54-59 (1573.8-1690.9 kyr) in the 442 most altered portion of the records with the highest DR values (>4). The glacial-interglacial 443 cycles are more prominent across all records in Interval 1. Both G. ruber (w) and P. 444 *obliquiloculata* (w/c) display a large amplitude change from the glacial peak MIS12 across 445 the deglaciation to the interglacial peak MIS11 ( $\Delta \sim 2.20$  ‰ and 2.50 ‰, respectively). 446 Globigerinita glutinata (w/b) shows a smaller change across MIS12 to MIS11 ( $\Delta$  1.41 ‰), 447 and less prominent glacial-interglacial peaks in Interval 2 compared to G. ruber (w) and P. 448 obliquiloculata (w/c). Interval 3 shows notably higher isotopic values with the glacial-

interglacial peaks markedly reduced, although still clearly visible, for all species. Despite the variation and extent of diagenetic alteration, Pleistocene glacial-interglacial cycles can be readily identified in all long-term  $\delta^{18}$ O records (Fig. 6). However, beyond ~1300 ka (46.1 mcd), the *G. ruber* (w) data shows a notable down-core trend towards higher  $\delta^{18}$ O values (Fig. 6).

454

#### 455 **3.3.** Comparison of biogenic calcite and authigenic overgrowth geochemistry

456 SIMS spot analyses (~10 µm diameter) are similar to the G. ruber (w) test wall thickness (~9-457 12 µm) such that a large proportion of the measurements partly overlapped onto epoxy. The 458 location of these compromised analyses (n = 35) was confirmed by SEM observation, and 459 only those measurements which were located solely on the foraminifera test were included in 460 the reported analyses (Fig. 7, Supplementary Material 3). Overall, 16 SIMS and 36 EPMA 461 spot measurements from the F-0 (final), F-1 (penultimate) and F-2 (antepenultimate) 462 chambers were measured on seven samples for G. ruber (w), (Fig. 7, Supplementary 463 Materials 2-3). An additional five EPMA spot analyses of large authigenic overgrowths were 464 measured (Fig. 7, Supplementary Material 2). Limited SIMS and EPMA sample sets were 465 obtained due to the limitations of spot size (for SIMS) and epoxy stability (for EPMA). Yet, 466 the datasets are still deemed sufficient to assess overall trends and the application of these 467 micro-analytical techniques in foraminiferal diagenetic studies. Mean values and standard 468 deviations of the measurements along with sample locations (with reference on the  $\delta^{18}$ O G. 469 ruber (w) curve) are shown in Figure 7.

470

471 Fig. 7. (a) SIMS and (b) EPMA spot data versus whole-test ICP-MS/ICP-OES

472 measurements for *G. ruber* (w) with the location of the samples (circles with number ID:

473 black = SIMS and red = EPMA samples) referenced on (c) the long-term *G. ruber* (w)

 $\delta^{18}$ O curve; (d) G. ruber (w) whole-test Mg/Ca data and (e) the G. ruber (w) Diagenesis 474 475 Rank (DR) color map. IODP Expedition 359 Site U1467 sample IDs: (1) A, B-Mudline; 476 (2) B-3H-3, 9-10 cm; (3) B-4H-5, 36-37 cm; (4) B-6H-2, 75-76 cm; (5) B-6H-3, 75-76 cm; 477 (6) B-6H-4, 75-76 cm; (7) B-7H-3, 75-76 cm. Sea surface temperature (SST) calculations 478 are based on the G. ruber (w) equation of Anand et al. (2003). Red curve shows  $\delta^{18}$ O G. 479 ruber (w) data from Bunzel et al. (2017). A 3-pt moving average smoothing was applied 480 to all datasets (thick lines) to highlight the cycles. Note: F-0 (final), F-1 (penultimate) 481 and F-2 (antepenultimate) chambers, WT= whole-test and OG = overgrowth 482 measurements. WT data in a) and b) for A, B-Mudline Sample 1 is from Stainbank et al. 483 (2019).

484

485 Whole-test  $\delta^{18}$ O and Mg/Ca values are comparable to SIMS and EPMA spot measurements 486 for core-top Sample 1 at 0 ka (0 mcd) and to EPMA data for Sample 2 at 409.0 ka (17.4 mcd). 487 From Sample 3 (= 864.7 ka, 31.3 mcd) to Sample 7 (= 1743.8 ka, 58.7 mcd) the whole-test G. 488 *ruber* geochemical values are consistently higher than the SIMS (z = 1.5-5.2; p = 0.9-1) and 489 EPMA (z = 2.7-8; p = 1) data obtained from individual chambers (Fig. 7a-b). Moreover, the 490 difference varies with the highest values and largest offsets in the deepest, oldest sample. 491 Interestingly, one F-1 SIMS data point for Sample 7 hit substantial diagenetic overgrowths 492 and has a relatively high  $\delta^{18}$ O value of -1.24 ‰ (Supplementary Material 3). The EPMA 493 overgrowth data (Samples 6 and 7) reveals a HMC composition with 3-6 times higher Mg/Ca 494 values than original test compositions. Samples 3-7 cover the interval corresponding to an observed change in the slope of the long-term G. ruber (w)  $\delta^{18}$ O record with a down-core 495 496 trend towards higher values. A marked increase in the slope of the long-term G. ruber (w) 497 Mg/Ca record is, however, already noted prior to Sample 3 (Fig. 7d). Similar to the EPMA 498 spot analyses (Fig. 7b), the elemental maps of both G. ruber (w) and P. obliquiloculata (w/c)

499	show clear Mg enrichment in the authigenic overgrowths (Fig. 8). Both species have a LMC
500	test (Figs. 8.a1, 8.b1), whereas P. obliquiloculata (w/c) has natural HMC growth bands
501	indicated on Figure 8 (yellow arrows). Overgrowths, on both the inner and outer test surfaces,
502	have 4–10 times higher Mg intensities than the biogenic calcite (Figs. 8.a2, 8.b2).
503	
504	Fig. 8. Mg (intensity) EPMA maps. Examples of IODP Expedition 359 Hole U1467B
505	pristine specimens (Mudline) are shown for (a1) G. ruber (w) and (b1) P. obliquiloculata
506	(w/c) with diagenetically altered examples shown for (a2) G. ruber (w) (7H-3, 75-76 cm)
507	and (b2) <i>P. obliquiloculata</i> (w/c) (6H-4, 75-76 cm). T = test, OG = overgrowth, EE = edge
508	effect. Reference to sample locations (ID1, 6 and 7) are shown on Fig. 7. Note the high-
509	Mg (yellow arrows) and low-Mg banding in the <i>P. obliquiloculata</i> (w/c) tests. Scale bars
510	= 10 μm.
511	
512	4. DISCUSSION
513	4.1. Preservation state of the individual long-term geochemical records
514	The stable oxygen isotope composition ( $\delta^{18}$ O) of the original foraminiferal test is a function of
515	the ambient seawater temperature and $\delta^{18}O$ composition during its initial precipitation
516	(Pearson, 2012). Similarly, biogenic test Mg/Ca ratios reflect ambient seawater temperatures
517	(e.g. Raddatz et al., 2017; Rippert et al., 2016). Authigenic overgrowth is anticipated to occur
518	near the sediment/bottom-water interface and down-core (e.g. Regenberg et al., 2007). As
519	such, its oxygen isotope composition is controlled by a variety of factors including
520	thermodynamic fractionation, the $\delta^{18}O$ and pH of the precipitating fluid as well as mineralogy
521	and kinetic effects during the precipitation (Kim and O'Neil, 1997; O'Neil et al., 1969; Swart,
522	2015). The elemental ratio compositions are related to site mineralogy and pore water
523	geochemistry.

524

525 From 0 to ~200 kyr (0 to ~7.3 mcd), the cycles and amplitudes of IODP Expedition 359 Site 526 U1467  $\delta^{18}$ O G. ruber (w) and C. mabahethi records, are comparable to the records produced 527 by Bunzel et al. (2017) (Fig. 6). Bunzel et al. (2017) compiled stable isotopic datasets from 528 Core SO236-052-4 retrieved from the Maldives Inner Sea (03°55.09'N; 73°08.48'E) at a water 529 depth of 382 m (Note: the slight amplitudinal difference between the benthic datasets is due to 530 differences in study site depths). Notably, Bunzel et al. (2017) used a lower analytical 531 resolution and a larger test size (250–350 µm) for their G. ruber (w) record, yet visually the 532 two datasets are nearly identical. This attests to the good preservation state of Site U1467 samples for the first ~200 kyr (7.3 mcd). Furthermore, we can consider the G. ruber (w)  $\delta^{18}$ O 533 534 and Mg/Ca records from 0 to ~627.4 kyr (0 to ~24.7 mcd) to be pristine/near-pristine. This is 535 reflected by the low DR values (0-2) and is manifest in the coherence between the SIMS and 536 EPMA spot analyses data (Samples 1 and 2) and the whole-test geochemical datasets. In 537 particular, EPMA data for Sample 2 (Fig. 7b) illustrates that at least up until 409.0 ka (17.4 538 mcd) the whole-test Mg/Ca values closely reflect the original test geochemistry. While small, 539 patchy HMC overgrowth is present on the internal surfaces (DR = 1), it does not appear to be 540 sufficient to bias the whole-test geochemistry. Calculated SSTs are also notable for being 541 within the modern, seasonal seawater temperature range (Fig. 7d) (Krahmann and Krüger, 542 2018; Quadfasel, 2017; Reolid et al., 2017).

543

The degree of influence of the authigenic overgrowth on whole-test geochemical records can vary significantly and is not necessarily consistent across the various proxies (i.e.  $\delta^{18}$ O versus Mg/Ca). A change in the preservation state of the *G. ruber* (w) tests occurs between ~627.4 to 790.0 kyr (24.7–28.7 mcd) onwards (DR values > 2). Discrepancies are also recorded by the SIMS and EPMA data from individual chambers which are significantly lower than respective

549 whole-test geochemical values from Sample 3 ( $\Delta \sim 1.1$  % and  $\sim 1.5$  mmol/mol, respectively) to 550 Sample 7 ( $\Delta \sim 1.8$  ‰ and  $\sim 4.1$  mmol/mol, respectively) (Fig. 7a, b). There is a notable down-551 core increase in the slope of the  $\delta^{18}$ O G. ruber (w) record. This is captured by SIMS Samples 552 3-5, which show a  $\sim 1$  ‰ increase in the whole-test values and Sample 7, the most 553 diagenetically altered, which shows a  $\sim 2$  ‰ increase. The compositional trends observed in the  $\delta^{18}$ O data are notably amplified in the long-term Mg/Ca record, with a steeper down-core 554 555 slope increase. Considering the original test geochemistry is usually < 6 mmol/mol, only a 556 limited amount of HMC overgrowth (~12-22 mmol/mol) would be required to bias the 557 whole-test Mg/Ca towards significantly higher values (Fig. 7). This is reflected in EPMA 558 Sample 3 (= 864.7 ka, 31.3 mcd), which has minimal overgrowth present on the external test surface (external category Diagenesis Rank = 1), yet small-medium well-developed euhedral 559 560 crystals across the entire inner test surface (internal category Diagenesis Rank = 2). This 561 overgrowth, although small, is sufficient to elevate the whole-test Mg/Ca values and increase 562 the SST reconstruction by  $\sim 2^{\circ}$ C. This is further substantiated by EPMA Samples 4, 6 and 7, 563 which have higher DR values due to poorer preservation states and an increased presence 564 and/or size of overgrowth crystals. All have lower spot F-0 and F-1 chamber Mg/Ca values in 565 comparison to the whole-test data, resulting in progressively larger discrepancies in SST 566 estimates of ~4–7°C.

567

The precipitation of HMC overgrowths, as opposed to the more stable LMC, could be a function of both the composition and content of the bulk sediment as well as the pore water geochemistry (e.g. Panieri et al., 2017; Regenberg et al., 2007). At Site U1467, HMC is only present in the top 10 mbsf (Fig. 2). Where HMC in the sediment has dissolved, Mg<sup>2+</sup> is released and may be incorporated (elsewhere) in crystalline overgrowths on biogenic carbonates. Aragonite dissolution and active sulphate reduction were noted below 50 mbsf at

574 this site by Betzler et al. (2017). The former had been noted earlier in a pteropod study by 575 Sreevidya et al. (2019) who reported intense dissolution of aragonitic pteropods beyond ~800 ka. As such, an increased saturation state of pore waters, with respect to Mg<sup>2+</sup>, in combination 576 with active sulphate reduction may have favored HMC over LMC authigenic precipitates 577 578 (Panieri et al., 2017; Swart, 2015). Higher resolution sediment mineralogy together with 579 petrography and pore water profiles are needed to conclude as to the exact processes behind 580 the depth occurrence and Mg-content of the HMC overgrowths. However, the combined 581 influence of HMC with naturally elevated  $\delta^{18}$ O values (Tarutani et al., 1969), with thermodynamic fraction ( $\Delta \sim 19^{\circ}$ C between the surface seawater and at 487 m depth) and 582 583 related fluid geochemistry changes would further justify the precipitation of authigenic overgrowths with elevated  $\delta^{18}$ O values. These processes could thus explain the notable biases 584 585 (stable isotopic and elemental) on the planktonic whole-test geochemical data from 586 SIMS/EPMA Sample 3 (= 864.7 ka, 31.3 mcd) to 7 (= 1743.8 ka, 58.7 mcd).

587

588 Although no SIMS or EPMA data was collected for the benthic C. mabahethi species, a 589 change in preservation state (DR > 2) occurs between  $\sim$ 790.0 to 807.9 kyr (28.7–29.1 mcd). 590 As such, we can surmise the records beyond this interval to have some degree of diagenetic 591 overprint (Figs. 6e-f). This is supported by observations of large, well-developed overgrowth 592 on the internal test surfaces at 872.0 ka (31.7 mcd). Even though minimal overgrowth is 593 present on the external surfaces, it is presumed that the large internal crystals would be 594 sufficient to affect the whole-test geochemical records. However, although the Mg/Ca biases would be similar to that on the G. ruber (w) records, the  $\delta^{18}$ O influences should be minor 595 596 considering diagenetic precipitation is anticipated to occur near the sediment/bottom water 597 interface in close proximity to where the original biogenic calcite precipitated (Edgar et al., 598 2013). This would imply similar seawater temperatures; however, differences in pH,

599 mineralogy and kinetic effects during the authigenic overgrowth precipitation cannot be

600 excluded. This hypothesis is supported by the prominent glacial-interglacial cycles with their

amplitudes seemingly well preserved for the entirety of the benthic  $\delta^{18}$ O record (0-1179.0 kyr)

602 (Figs. 3-4, 6).

603

#### 604 **4.2. Diagenetic susceptibility of individual planktonic species**

605 We found distinct differences in the preservation state of the three investigated planktonic 606 species. As such, the diagenetic bias on their respective whole-test geochemical compositions 607 varies. This is illustrated in Intervals 1-3 in Figure 6, with a variable preservation of the 608 glacial-interglacial amplitudinal differences. The whole-test G. ruber (w), G. glutinata (w/b) and *P. obliquiloculata* (w/c)  $\delta^{18}$ O records all reflect clear, large amplitude cycles within 609 610 Interval 1. However, relative to the thicker-walled P. obliquiloculata (w/c), the former two 611 species record dampened signals with the peaks notably less pronounced across Interval 2. 612 Across Interval 3, in the most diagenetically altered portion of the record, all datasets are notably affected with reduced glacial-interglacial maxima and minima and all  $\delta^{18}$ O records 613 614 converge because of increasing whole test  $\delta^{18}$ O values.

615

616 It might seem contradictory that the species, which appears to be most diagenetically

617 susceptible, would have the best whole-test geochemical preservation across Interval 2.

618 However, as *P. obliquiloculata* (w/c) has the thickest test (~24 μm, Figs. 5, 9), the rationale is

619 that a thicker test would equate to more of the whole-test composition being weighted by the

620 biogenic calcite. Therefore, in proportion to the primary signal, the authigenic overgrowth

621 would contribute a smaller percentage and result in a smaller bias compared to G. ruber (w)

and G. glutinata (w/b) which have significantly thinner tests (~9–12 and ~6–10  $\mu$ m,

623 respectively; Figs. 5, 9). This is in accordance with observations by Bé (1977) and Kucera

624 (2007). Bé (1977) correlated increased diagenetic resistance for species with smaller pores,

625 thicker walls and larger test size, while Kucera (2007) reports an increase in dissolution

626 resistance for species with a lower porosity (Fig. 9).

627

Fig. 9. Interpretations of diagenetic susceptibilities for the whole-test geochemistry of the
three investigated planktonic species. Interpretations are based on Diagenesis Rank
(DR) values, spot EPMA/SIMS and whole-test geochemistry data from IODP Expedition
359 Site U1467. Interpretations of dissolution susceptibilities after Kucera (2007).

632

#### 633 5. CONCLUSIONS

634 Foraminiferal geochemical records from shallow, carbonate platforms are integral for 635 understanding global paleoceanographic and -climatic trends but are subject to diagenetic 636 influences. This is the case at IODP Expedition 359 Site U1467, which represents a shallowintermediate water archive from the equatorial Indian Ocean of importance for South Asian 637 638 Monsoon (SAM) climatic studies and latitudinal scale shifts of ocean currents. Prior to 639 utilizing our high-analytically resolved geochemical records for paleo-reconstructions, it is 640 important to constrain the influence of diagenetic alteration on both the preservation of the 641 original geochemical compositions and long-term cyclicity.

642

The presence and influence of authigenic overgrowths on the inside and/or outside of the foraminiferal test as well as through the partial or full recrystallization of the test wall has long been recognised. Importantly, this authigenic calcite is not ubiquitous or uniform in terms of presence, size, distribution or geochemical composition and as such should be assessed on an individual study site basis. For simplicity, ease of use and to facilitate reproducibility in other studies, the qualitative DR detailed and used in this study only

649 constrains the presence of authigenic overgrowths on the test surfaces as well as within the 650 wall structure. However, it is noted that both the primary and authigenic calcite can be 651 susceptible to dissolution, which would further alter the geochemical compositions. These 652 dissolution features, which can include etched surfaces as well as lattice-like calcite 653 structures, as well as partial recrystallization of the wall texture are noted in the deepest, 654 oldest studied samples.

655

656 The degree of influence of the authigenic overgrowth on the whole-test geochemistry varies 657 across the foraminiferal species (G. ruber (w), G. glutinata (w/b), P. obliquiloculata (w/c), C. *mabahethi*) and proxies (i.e.  $\delta^{18}$ O versus Mg/Ca). Considering the site mineralogy, whole-test 658 Mg/Ca compositions are notably more impacted than  $\delta^{18}$ O records. However, taking into 659 660 consideration our individual spot analyses (SIMS and EPMA) and DR data, the top ~627.4 kyr of the G. ruber (w) Mg/Ca and  $\delta^{18}$ O records are well preserved and largely reflect the 661 662 primary geochemical compositions. Given the setting of overgrowth precipitation, the benthic *C. mabahethi*  $\delta^{18}$ O record from 0–790.0 kyr (DR = 0–2), if not the entire benthic record, is 663 664 considered to have pristine/near-pristine preservation of the primary  $\delta^{18}$ O signal. As such, the 665 top portions of both the G. ruber (w) (0 to ~627.4 kyr; 0-24.7 mcd) and C. mabahethi (0 to 666 ~790.0 kyr; 0-28.7 mcd) records (whole-test data) are suitable for paleoceanographic reconstructions of absolute seawater temperature, salinity and  $\delta^{18}$ O values. Notwithstanding 667 668 the down-core diagenetic influences, the glacial-interglacial cyclicity, while dampened, is 669 maintained as attested by the coherence of both the G. ruber (w) and C. mabahethi  $\delta^{18}$ O 670 records with Prob-stack. As such, the complete geochemical records could still be used for 671 cyclostratigraphy to assess long-term orbital scale properties.

672

673 Overall, we show foraminiferal species (e.g. P. obliquiloculata) with thicker tests are better 674 suited for whole-test geochemical analyses, in diagenetically susceptible settings, as they have 675 a higher proportion of biogenic calcite relative to authigenic overgrowth. Moreover, while 676 only a portion of our whole-test geochemical records reflect the primary compositions, SIMS 677 and EPMA spot measurements can be utilized to overcome any biases in diagenetically 678 impacted intervals. Considering the importance of these types of records for tropical 679 temperature studies, it is vital to employ an approach, whether it be spot versus whole-test 680 analyses, the utilization of specimens with thicker tests or a visual DR, which will allow the 681 preservation state to be assessed to obtain more accurate and reliable paleo-reconstructions. 682

002

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689

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873	Supplementary Material 1. Representative visual SEM images used to define the
874	Diagenesis Rank (DR) values for the individual species from IODP Expedition 359 Site
875	U1467.
876	
877	Supplementary Material 2. Electron Probe Micro-Analyzer (EPMA) data.
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879	Supplementary Material 3. Secondary Ion Mass Spectrometer (SIMS) data.
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881	Supplementary Material 4. Age model data for IODP Expedition 359 Site U1467.



Fig. 1.



Fig. 2.



Fig. 3.



Fig. 4.



Fig. 5.



Fig. 6.



Fig. 7.



Fig. 8.



Fig. 9.

# Supplementary Material 1. Representative visual SEM images used to define the Diagenesis Rank (DR) values for the individual species from IODP Expedition 359 Site U1467.

#### Supplementary Figs. 1a-b.

IODP Expedition 359 Site U1467 cross section and inner test surface SEM images of (a) *G. ruber* (w); (b) *G. glutinata* (w/b); (c) *P. obliquiloculata* (w/c) and (d) *C. mabahethi/wuellerstorfi* for Samples: (1) A, B-Mudline; (2) C-1H-1, 99-100 cm; (3) B-3H-3, 9-10 cm; (4) B-3H-3, 102-103 cm; (5) B-4H-5, 36-37 cm; (6) C-4H-3, 87-88 cm; (7) C-5H-4, 30-31 cm; (8) B-6H-2, 75-76 cm; (9) B-6H-3, 75-76 cm; (10) B-6H-4, 75-76 cm; (11) C-6H-5, 57-58 cm; (12) B-7H-3, 75-76 cm. For reference, SIMS and EPMA Samples 1-7 are indicated in the black and red circles, respectively. White bars indicate test thickness. Scale bars =  $20 \mu m$ .

#### Supplementary Figs. 1c-d.

IODP Expedition 359 Site U1467 external test surface SEM images of (e) *G. ruber* (w); (f) *G. glutinata* (w/b); (g) *P. obliquiloculata* (w/c) and (h) *C. mabahethi/wuellerstorfi* for Samples: (1) A, B-Mudline; (2) C-1H-1, 99-100 cm; (3) B-3H-3, 9-10 cm; (4) B-3H-3, 102-103 cm; (5) B-4H-5, 36-37 cm; (6) C-4H-3, 87-88 cm; (7) C-5H-4, 30-31 cm; (8) B-6H-2, 75-76 cm; (9) B-6H-3, 75-76 cm; (10) B-6H-4, 75-76 cm; (11) C-6H-5, 57-58 cm; (12) B-7H-3, 75-76 cm. For reference, SIMS and EPMA Samples 1-7 are indicated in the black and red circles, respectively. Scale bars =  $20 \mu m$ .



Supplementary Fig. 1a.



Supplementary Fig. 1b.



Supplementary Fig. 1c.



Supplementary Fig. 1d.



Supplementary Material 2. Electron Probe Micro-Analyzer (EPMA) data.

**Supplementary Fig. 2a.** IODP Expedition 359 Site U1467 *G. ruber* (w) SEM images showing examples of EPMA spot measurements for (a) the foraminifera test (T) and (b) large authigenic overgrowths (OG). Samples: (a1) B-4H-5, 36-37 cm; a2) B-6H-2, 75-76 cm; (b1-2) B-6H-4, 75-76 cm (Approximate location of the EPMA spots are shown for reference with the red dots with the red text indicating their individual EPMA IDs).

**Supplementary Table 2a.** IODP Expedition 359 Site U1467 *G. ruber* (w) EPMA test measurements used in the study. Measurements with total counts < 97 and > 102 were excluded. Note: SrO was measured for all samples and was zero in all instances.

Sample	EPMA ID	Sample ID	CaO	MgO	С	Total	Chamber	Mg/Ca (mmol/mol)
359-U1467B-Mudline	IODP359 I shell 1.1.1		56.45	0.21	44.53	101.19	F-0	5.17
359-U1467B-Mudline	IODP359 I shell 1.1.2		56.00	0.26	44.24	100.50	F-0	6.49
359-U1467B-Mudline	IODP359 I shell 1.2.3		55.23	0.19	43.55	98.97	F-0	4.87
359-U1467B-Mudline	IODP359 I shell 1.2.4	1	55.17	0.13	43.44	98.74	F-0	3.25
359-U1467B-Mudline	IODP359 I shell 1.1.3		55.86	0.13	43.98	99.97	F-1	3.17
359-U1467B-Mudline	IODP359 I shell 1.1.4		55.40	0.12	43.61	99.13	F-1	3.11
359-U1467B-Mudline	IODP359 I shell 1.2.2		55.74	0.15	43.91	99.80	F-1	3.70
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.1.1		55.55	0.18	43.79	99.52	F-0	4.51
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.1.2		56.30	0.26	44.47	101.03	F-0	6.43
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.2.1		55.37	0.18	43.65	99.20	F-0	4.45
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.2.2	2	55.41	0.16	43.66	99.23	F-0	3.90
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.1.3	2	54.98	0.29	43.46	98.73	F-1	7.36
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.1.4		54.82	0.24	43.28	98.34	F-1	6.14
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.2.3		55.31	0.14	43.56	99.01	F-1	3.54
359-U1467B-3H-3, 9-10 cm	IODP359 I shell 2.2.4		54.71	0.23	43.18	98.12	F-1	5.83
359-U1467B-4H-5, 36-37 cm	IODP359 I shell 4.1.2		54.96	0.21	43.36	98.53	F-0	5.20
359-U1467B-4H-5, 36-37 cm	IODP359 I shell 4.2.4		55.13	0.19	43.47	98.79	F-0	4.81
359-U1467B-4H-5, 36-37 cm	IODP359 I shell 4.1.3	3	55.01	0.20	43.39	98.60	F-1	5.15
359-U1467B-4H-5, 36-37 cm	IODP359 I shell 4.2.2		55.60	0.14	43.79	99.53	F-1	3.51
359-U1467B-4H-5, 36-37 cm	IODP359 I shell 4.2.3		55.75	0.18	43.95	99.88	F-1	4.48
359-U1467B-6H-2, 75-76 cm	IODP359 I shell 6.1.1		55.08	0.14	43.39	98.61	F-0	3.64
359-U1467B-6H-2, 75-76 cm	IODP359 I shell 6.1.3		55.17	0.13	43.44	98.74	F-1	3.32
359-U1467B-6H-2, 75-76 cm	IODP359 I shell 6.1.4	4	55.90	0.12	44.00	100.02	F-1	2.96
359-U1467B-6H-2, 75-76 cm	IODP359 I shell 6.1.5		55.75	0.15	43.92	99.82	F-1	3.79
359-U1467B-6H-2, 75-76 cm	IODP359 I shell 6.2.4		55.10	0.21	43.47	98.78	F-1	5.41
359-U1467B-6H-4, 75-76 cm	IODP359 I shell 8.1.2		56.90	0.13	44.79	101.82	F-0	3.11
359-U1467B-6H-4, 75-76 cm	IODP359 I shell 8.3.1		55.22	0.15	43.50	98.87	F-0	3.74
359-U1467B-6H-4, 75-76 cm	IODP359 I shell 8.1.4	6	55.32	0.16	43.59	99.07	F-1	4.08
359-U1467B-6H-4, 75-76 cm	IODP359 I shell 8.3.3		55.49	0.19	43.76	99.44	F-1	4.77
359-U1467B-6H-4, 75-76 cm	IODP359 I shell 8.3.4		54.92	0.22	43.34	98.48	F-1	5.53
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.1.1		56.78	0.14	44.72	101.64	F-0	3.49
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.1.2		55.74	0.15	43.90	99.79	F-0	3.66
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.1.6	7	55.13	0.12	43.40	98.65	F-1	3.13
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.2.3	/	55.09	0.18	43.43	98.70	F-1	4.43
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.2.4		55.42	0.17	43.68	99.27	F-1	4.18
359-U1467B-7H-3, 75-76 cm	IODP359 I shell 9.2.5		54.66	0.17	43.08	97.91	F-1	4.33

**Supplementary Table 2b.** IODP Expedition 359 Site U1467 *G. ruber* (w) EPMA overgrowth measurements used in the study. Measurements with total counts < 97 and > 102 were excluded. Note: SrO was measured for all samples and was zero in all instances.

Sample	EPMA ID	Sample ID	CaO	MgO	С	Total	Mg/Ca (mmol/mol)
359-U1467B-6H-4, 75-76 cm	K_1_1	6	54.70	0.55	43.52	98.77	13.87
359-U1467B-6H-4, 75-76 cm	K_1_1_4	0	56.29	0.41	44.62	101.32	10.13
359-U1467B-7H-3, 75-76 cm	K_5_2		56.19	0.53	44.67	101.39	13.12
359-U1467B-7H-3, 75-76 cm	K_5_2_b	7	54.80	1.05	44.15	100.00	26.71
359-U1467B-7H-3, 75-76 cm	K_5_1		54.76	1.01	44.08	99.85	25.76

Sample	Sample ID	Mg/Ca (mmol/mol)
359-U1467B-Mudline	1	5.66*
359-U1467B-Mudline	1	5.65*
359-U1467B-3H-3, 9-10 cm		6.30
359-U1467B-3H-3, 9-10 cm	2	6.00
359-U1467B-3H-3, 9-10 cm		5.97
359-U1467B-4H-5, 36-37 cm	3	6.18
359-U1467B-4H-5, 36-37 cm		6.22
359-U1467B-6H-2, 75-76 cm	4	6.05
359-U1467B-6H-2, 75-76 cm		5.72
359-U1467B-6H-4, 75-76 cm	6	6.62
359-U1467B-6H-4, 75-76 cm	0	6.10
359-U1467B-7H-3, 75-76 cm	7	8.45
359-U1467B-7H-3, 75-76 cm	] /	7.32

## **Supplementary Table 2c.** IODP Expedition 359 Site U1467 whole-test *G. ruber* (w) Mg/Ca data used in EPMA comparison.\*Data is taken from Stainbank et al. (2019).

#### Supplementary Material 3. Secondary Ion Mass Spectrometer (SIMS) data.



**Supplementary Fig. 3a.** Compilation of IODP Expedition 359 Site U1467 (a) stereomicroscope images of embedded and polished *G. ruber* (w) SIMS Sample H showing approximate SIMS spot locations and (b) transmitted light microscope images of *G. ruber* (w) SIMS Sample E showing approximate SIMS spot locations. Sample IDs are indicated on the image.



Supplementary Fig. 3b. IODP Expedition 359 Site U1467 SEM images showing examples of *G. ruber* (w) SIMS spots which (a) hit only the foraminiferal test (T) and (b) which partly hit the epoxy (E). IODP Expedition 359 Site U1467 Samples: (a1) B-4H-5, 36-37 cm (SIMS ID:  $\delta^{18}$ O\_foram\_H3\_1@1); (a2) B-6H-2, 75-76 cm (SIMS ID:  $\delta^{18}$ O\_foram\_H4\_1@1); (b1) B-Mudline (SIMS ID:  $\delta^{18}$ O\_foram\_H1\_2@1); (b2) B-4H-5, 36-37 cm (SIMS ID:  $\delta^{18}$ O\_foram\_H3\_1@3).

**Supplementary Table 3a.** IODP Expedition 359 Site U1467 *G. ruber* (w) SIMS measurement spots, which hit no epoxy and were used in the study. (Session H-1 standard yield: 1.10E+09; Session E-2 standard yield: 9.30E+08). Bold indicates the SIMS spot, which hit substantial authigenic overgrowth.

Sample	SIMS ID	Sample ID	Session	Yield	δ <sup>18</sup> Ο (‰)	2se (‰)	Chamber
359-U1467B-Mudline	$\delta^{18}O_foram_E7@4$	1	E-2	1.021E+09	-2.08	0.35	F-1
359-U1467B-4H-5, 36-37 cm	δ <sup>18</sup> O_foram_H3_1@1	3	H-1	1.093E+09	-2.70	0.38	F-0
359-U1467B-4H-5, 36-37 cm	$\delta^{18}O_{foram}E5@6$		E-2	8.728E+08	-2.22	0.29	F-1
359-U1467B-4H-5, 36-37 cm	$\delta^{18}O_{foram}E5@7$		E-2	9.644E+08	-2.14	0.39	F-2
359-U1467B-4H-5, 36-37 cm	$\delta^{18}O_{foram}E5@8$		E-2	9.468E+08	-2.81	0.30	F-2
359-U1467B-4H-5, 36-37 cm	δ <sup>18</sup> O_foram_H3_1@2		H-1	1.043E+09	-2.82	0.50	F-2
359-U1467B-6H-2, 75-76 cm	$\delta^{18}O_{foram}H4_1@1$	4	H-1	1.049E+09	-1.75	0.35	F-0/F-2
359-U1467B-6H-2, 75-76 cm	δ <sup>18</sup> O_foram_H4_1@2		H-1	1.065E+09	-2.63	0.38	F-2
359-U1467B-6H-3, 75-76 cm	$\delta^{18}O_{foram}E3@1$	5	E-2	1.078E+09	-2.15	0.34	F-1
359-U1467B-6H-3, 75-76 cm	$\delta^{18}O_{foram}E3@2$		E-2	9.996E+08	-2.51	0.43	F-1
359-U1467B-7H-3, 75-76 cm	$\delta^{18}O_{foram}E2@1$	7	E-2	9.859E+08	-1.55	0.44	F-1
359-U1467B-7H-3, 75-76 cm	$\delta^{18}O_{foram}E2@2$		E-2	9.881E+08	-2.18	0.39	F-1
359-U1467B-7H-3, 75-76 cm	δ <sup>18</sup> O_foram_H6_1@1		H-1	1.045E+09	-2.00	0.26	F-1
359-U1467B-7H-3, 75-76 cm	δ <sup>18</sup> O_foram_H6_2@1		H-1	9.555E+08	-1.24	0.41	F-1
359-U1467B-7H-3, 75-76 cm	δ <sup>18</sup> O_foram_H6_2@2		H-1	1.025E+09	-3.15	0.38	F-2
359-U1467B-7H-3, 75-76 cm	δ <sup>18</sup> O_foram_H6_2@3		H-1	1.035E+09	-2.12	0.27	F-2

359-U1467B, C	Prob-stack (Ahn et al., 2017)	Sedimentation	
Depth (mcd)	Age (kyr)	rate (cm/kyr)	
0.01	0	8.1	
0.58	7	3.9	
0.85	14	3.0	
1.00	19	2.0	
1.06	22	2.3	
1.15	26	3.0	
1.18	27	1.5	
1.63	57	2.2	
1.87	68	3.0	
2.32	83	15.0	
2.77	86	9.5	
3.15	90	4.9	
3.54	98	4.5	
3.81	104	2.6	
4.02	112	15.0	
4.17	113	4.5	
4.44	119	6.0	
4.50	120	9.0	
4.59	121	9.5	
4.78	123	13.5	
5.05	125	3.6	
5.41	135	3.6	
5.59	140	2.6	
6.16	162	2.3	
6.70	186	3.9	
7.21	199	4.5	
7.30	201	9.0	
7.39	202	9.8	
7.78	206	11.0	
8.11	209	12.0	
8.47	212	3.7	
8.62	216	5.6	
9.07	224	5.0	
9.72	237	16.3	
10.21	240	2.7	
10.48	250	4.4	
11.35	270	5.1	
12.12	285	7.5	
12.27	287	5.1	
12.78	297	2.5	
13.05	308	2.1	
13.20	315	12.6	
13.83	320	6.4	
14.28	327	10.5	
14.49	329	9.0	
14.67	331	3.0	

**Supplementary Material 4. Table 4a. Age model data for IODP Expedition 359 Site U1467** Tie-points

14.73	333	2.4
14.85	338	4.5
14.94	340	9.0
15.03	341	0.9
15.09	348	2.3
15.18	352	3.0
15.24	354	1.2
15.30	359	3.3
15.60	368	5.1
16.47	385	1.5
16.53	389	4.6
17.13	402	3.0
17.25	406	6.0
17.31	407	3.0
17.37	409	3.0
17.76	422	5.0
18.21	431	10.5
18.42	433	2.3
18.79	449	2.6
18.97	456	2.1
19.30	472	3.3
19.60	481	5.4
19.87	486	8.2
20.20	490	1.8
20.56	510	1.5
20.59	512	1.1
20.68	520	5.6
21.85	541	3.0
22.06	548	1.2
22.27	566	4.1
22.96	583	4.5
23.14	587	6.0
23.38	591	6.8
23.92	599	15.0
24.07	600	7.5
24.22	602	1.2
24.34	612	2.3
24.76	630	3.3
25.09	640	1.5
25.87	692	3.5
26.08	698	2.5
26.68	722	8.7
27.55	732	4.3
28.15	746	1.2
28.60	784	2.0
28.96	802	2.5
29.47	822	4.2
29.89	832	2.6
30.31	848	6.8
31.12	860	4.5

31.30	864	4.5
31.66	872	3.0
32.14	888	3.3
32.80	908	2.8
33.13	920	1.7
33.67	952	4.2
34.18	964	3.0
34.60	978	3.4
34.87	986	5.7
35.44	996	4.1
35.77	1004	2.2
36.16	1022	2.4
36.55	1038	3.0
36.67	1042	1.5
36.70	1044	0.9
36.82	1058	1.5
36.97	1068	6.8
37.24	1072	1.2
37.57	1100	1.5
37.69	1108	11.3
38.14	1112	2.7
38.41	1122	2.3
38.50	1126	1.7
39.10	1162	5.3
41.11	1200	5.7
42.94	1232	11.5
43.63	1238	1.1
43.81	1254	2.5
44.26	1272	9.6
45.22	1282	6.0
45.58	1288	4.1
46.57	1312	8.2
46.90	1316	5.3
47.11	1320	1.3
47.32	1336	2.1
47.65	1352	2.5
48.10	1370	2.6
48.31	1378	3.0
48.73	1392	7.5
49.03	1396	1.2
49.24	1414	1.5
49.72	1446	1.5
49.87	1456	10.9
50.74	1464	5.2
51.16	1472	2.6
51.58	1488	5.4
52.12	1498	0.7
52.27	1520	4.2
52.48	1525	3.0
52.78	1535	0.4

52.84	1550	1.9
53.08	1563	1.6
53.32	1578	1.9
53.56	1590	6.0
53.71	1593	2.6
54.04	1605	0.4
54.07	1613	0.9
54.16	1623	1.8
54.25	1628	0.9
54.34	1638	1.4
54.52	1650	1.8
54.61	1655	2.3
55.18	1680	8.5
57.10	1703	2.8
57.52	1718	5.6
57.94	1725	4.6
58.51	1738	2.4
58.75	1748	6.8
59.26	1755	9.0
60.16	1765	1.7
60.55	1788	3.0
60.70	1793	4.0
61.00	1800	3.4