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Ocean acidification and molluscan shell taphonomy: can elevated seawater $p\text{CO}_2$ influence taphonomy in a naticid predator–prey system?

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Abstract

The size and frequency of gastropod drill holes in shells of their prey are common indicators of predator-prey ecology in the fossil record. Taphonomic processes occurring after predation, however, can influence the preservation of shells in a given fossil assemblage and can thus influence ecological inferences based on preserved shells. To determine if ocean acidification (OA) has the capacity to influence prey shell taphonomy in a gastropod drilling predation system, we tested for effects of elevated $p\text{CO}_2$ on dissolution rates, breakage force, and drill hole diameters in non-fragmented shells of two prey species of the cannibalistic naticid gastropod, *Euspira heros*. Drilled and non-drilled shells of *Littorina littorea* and *E. heros* were exposed to control ($\sim 300 \mu\text{atm}$) and elevated (~ 800 and $4000 \mu\text{atm}$) $p\text{CO}_2$ treatments for five weeks. Dry shell weight and drill hole diameter (outer and inner) were recorded for individual shells before and after exposure; the force required for shell breakage was recorded at the end of the exposure period. Shell mass loss in 800 and $4000 \mu\text{atm}$, respectively, were ~ 1 and 7% for *E. heros*, and ~ 0 and 4% for *L. littorea*, compared to $\sim 0\%$ in the control for both species. Shell breakage force was unaffected by elevated $p\text{CO}_2$, but was affected by species and drill hole presence, with *E. heros* shells requiring a force of ~ 220 and 269 Newtons in drilled and non-drilled shells, respectively, compared to ~ 294 and 415 Newtons in *L. littorea*. At $4000 \mu\text{atm}$, outer drill hole diameter significantly increased by $\sim 12\%$ for *E. heros*, while inner drill hole diameter significantly increased by $\sim 13\%$ in *E. heros* and $\sim 10\%$ in *L. littorea*. Ultimately, this study provides the first documentation of molluscan shell taphonomy in the context of OA for a gastropod drilling predation system and sets the stage for future research.

Keywords: carbon dioxide, global change, carbonate dissolution, palaeoclimate, pH, predation

1. Introduction

Gastropod drilling predation is recognized as an important ecological and evolutionary component in marine ecosystems, as drilling gastropods are (and have historically been) important predators of marine molluscs globally (Commito, 1982; Kabat, 1990; Kelley and Hansen, 2003; Harper, 2006; Clements and Rawlings, 2014). The fossil record of gastropod drilling predation is a commonly studied system in palaeoecology (Kabat, 1990; Kowalewski, 2002; Kelley and Hansen, 2003; Harper, 2006; Li et al., 2011), owing to the telltale drill holes that these predators make in the shells of their prey and the high degree of fossil preservation of calcium carbonate shells (Guerrero and Reyment, 1988; Kelley and Hansen, 2003; Grey et al., 2005; Dietl and Kelley, 2006; Clements et al., 2013; Kubika et al., 2017). Such studies use drill holes to infer aspects of predator-prey ecology between drilling gastropods and their prey, and often use relative frequencies of drilled and non-drilled shells to infer predator-prey ecology (Dudley and Vermeij, 1978; Hansen and Kelley, 1995; Kelley et al., 2001; Dietl et al., 2004; Kowalewski et al., 2005; Das et al., 2013). These studies have been central to understanding changes in predatory behaviour and predation intensity throughout evolutionary time, and are the basis for broad-scale hypotheses regarding macroevolutionary patterns and processes (Vermeij, 1987; Kelley, 1988, 1991; Kelley and Hansen, 1993, 1996; Leighton, 2002; Dietl, 2003; Dietl et al., 2004; Kowalewski et al., 2005; Huntley and Kowalewski, 2007; Chiba and Sato, 2012, 2016; Klompmaker et al., 2017).

Although the fossil record of drilling predation provides a tangible system to study predator-prey interactions throughout evolutionary time, taphonomic processes (i.e. physical, chemical and geological processes that biological remains endure post-mortem; see Kowalewski, 2002, pp. 27–28 for an overview of these processes) can preclude accurate inferences (although not

always; e.g. see Wainwright et al., 1982; Nebelsick and Kowalewski, 1999; Kelley, 2008). In the context of predator-prey systems involving drilling gastropods, a variety of taphonomic effects can obfuscate interpretations of predator-prey interactions. Such effects include shell resistance to pre- and post-burial taphonomic forces that can break shells (Roy et al., 1994; Kaplan and Baumiller, 2000; Zuschin and Stanton, 2001; Grey et al., 2006; Martinell et al., 2012), durophagous degradation and/or destruction by non-drilling animals (Vermeij et al., 1979), and differential hydrodynamic sorting of shells with different morphologies (Chattopadhyay et al., 2013a,b). For example, weaker shells and shells containing predatory drill holes may be preferentially broken and removed from an assemblage compared to non-drilled and stronger shells (Roy et al., 1994; Kaplan and Baumiller, 2000; Zuschin and Stanton, 2001; Grey et al., 2006; Martinell et al., 2012). Similarly, drilled shells can exhibit a higher entrainment velocity than non-drilled shells, which can affect shell sorting and result in flawed inferences of drilling intensity (Chattopadhyay et al., 2013b), and other shell attributes can also affect sorting (Chattopadhyay et al., 2013a). It is thus important to take taphonomic influences into account when inferring aspects of predator-prey ecology from assemblages of fossilized shells, as such influences may lead to spurious interpretations of drilling gastropod predator-prey ecology.

A well-studied family of drilling gastropods is the Naticidae. Naticid gastropods are major predators of marine molluscs and their predatory impacts have been documented as early as the Cretaceous (Sohl, 1969; although drill holes in molluscs from other drilling gastropod taxa [e.g., *Platyceratidae*] are thought to have originated as early as the Cambrian; Baumiller, 1990; Baumiller and Macurda, 1995; Brett and Walker, 2002). Throughout the evolutionary history of naticid predation, the global ocean has fluctuated between periods of low and high pH (Veron, 2008; Kump et al., 2009; Hönisch et al., 2012; Clarkson et al., 2015). Periods of low oceanic pH

are primarily driven by the dissolution (into seawater) of gases that modify seawater carbonate chemistry and disrupt carbonate system equilibrium (primarily carbon dioxide [CO₂], although other gases contribute as well; see Doney et al., 2007). In recent years, this process has been coined ‘ocean acidification’ (OA) (Mostafa et al., 2016). The global ocean is currently experiencing OA due to the burning of fossil fuels, as approximately one third of excess atmospheric CO₂ produced by anthropogenic activities is absorbed into the ocean (Caldeira and Wickett, 2003; Hoegh-Guldberg, 2014). OA events in the geologic past may have also been driven by increased CO₂ from other sources (e.g. volcanic activity), as well as from increases in other atmospheric gases being dissolved into the ocean (e.g. sulfur dioxide, SO₂) (Veron, 2008; Kump et al., 2009; Hönisch et al., 2012; Ohno et al., 2014; Clarkson et al., 2015; Tyrell et al., 2015).

One well-described consequence of OA is that increasing acidity increases the vulnerability of calcium carbonate material, and calcium carbonate-bearing organisms, to dissolution (Ries et al., 2009; Hoffmann et al., 2010; Gazeau et al., 2013). CO₂-induced OA can increase the rate of dissolution to varying degrees across calcifying taxa depending on a number of factors, including (among others) a species’ physiological capacity to precipitate shell, and the carbonate mineral composition of a given species’ shell (e.g. more soluble aragonite *vs.* less soluble calcite; Ries et al., 2016). Indeed, OA events in the geologic past are correlated with mass extinctions of calcified organisms (Ohno et al., 2014; Clarkson et al., 2015; Tyrell et al., 2015; although it is unclear if these “mass extinctions”, as evidenced by a lack of carbonate material at particular periods, represent a true extinction event or simply represent an increased loss of carbonate material post-mortem, rather than a mass die-off). With respect to post-mortem shells, those that differ in their condition (i.e., taphonomic state; e.g. weathered *vs.* non-weathered, broken *vs.*

unbroken) and mineral composition (e.g. aragonite vs. calcite as mentioned above) can dissolve at disproportionate rates under OA (Cubillas et al., 2005; Waldbusser et al., 2011; Abdulghani 2014). Shells with naticid drill holes (or any drill hole) may therefore dissolve at faster rates than those without drill holes due to a potentially weakened structure and/or increased shell-layer exposure to seawater, as more taphonomically-impacted (i.e., degraded) shells typically lose more shell mass than less impacted shells (Abdulghani 2014). Differences in shell structure and mineralogy across species may result in disproportionate losses of certain species from a given assemblage due to dissolution, which can also be impacted by OA, leading to inaccurate accounts of biodiversity (e.g. Cherns and Wright, 2000, 2009; Cherns et al., 2011). Consequently, OA could directly influence aspects of shell taphonomy such as dissolution rate, shell thickness and strength, shell colouration, and drill hole size. Furthermore, through increased dissolution, OA may indirectly affect the relative contribution of various taphonomic agents such as transportation and lithification (among others) by increasing the probability of shell breakage and/or other damages.

While OA may influence shell taphonomy and, thus, inferences from molluscan shell assemblages, studies have yet to directly quantify this potential. As such, this study tested for effects of elevated $p\text{CO}_2$ on dissolution rates and shell breakage force in drilled and non-drilled shells of two prey species of the northern moon snail (*Euspira heros*)–*Littorina littorea* (Linnaeus, 1758) and cannibalized *E. heros* (Say, 1822). We hypothesized that drilled shells would dissolve quicker and break more easily than non-drilled shells because of increased degradation from drilling (more degraded shells dissolve more readily than less degraded shells; Waldbusser et al., 2011; Abdulghani 2014), and that this effect would be more pronounced for shells exposed to elevated $p\text{CO}_2$ owing to increased dissolution under higher $p\text{CO}_2$ (Ries et al.,

2009; Hoffmann et al., 2010; Gazeau et al., 2013). We also hypothesized that drill hole diameters in shells exposed to elevated $p\text{CO}_2$ would increase more-so than those under low $p\text{CO}_2$ due to increased dissolution.

2. Methods

2.1 Shell collection and selection

Shells used in this experiment were beach-collected from Port Morien Bay (46°6'55" N, 59°53'10" W), located in Cape Breton Island, Nova Scotia, Canada. They were collected in 2010 as part of another project documenting moon snail predation in eastern Cape Breton (see Clements et al., 2013; Clements and Rawlings, 2014 for details). During the time between collection and the initiation of our experiment on 8 February, 2018, shells were dry-stored at room temperature. For the purposes of this experiment, we used two species of gastropod prey that are commonly consumed by the northern moon snail, *Euspira heros*, in Cape Breton: periwinkles (*Littorina littorea*; 18.9 ± 2.6 mm shell length [$\bar{x} \pm \text{SD}$]) and conspecific moon snails (*Euspira heros*; 18.9 ± 3.2 mm shell length).

Prior to the experiment, the shells of each species were sorted by presence/absence of drill holes (drill hole diameters: 4.2 ± 0.8 mm for *E. heros* shells; 3.5 ± 0.8 mm for *L. littorea* shells [$\bar{x} \pm \text{SD}$]; Table 1). Care was taken to exclude shells that showed signs of weathering and only non-fragmented (i.e., intact) shells were selected. Once sorted, the shells were individually labeled and weighed (dry shell mass to the nearest 0.1 mg). Using digital calipers, we recorded shell length, along with inner and outer drill hole diameters (IDD and ODD, respectively). Shell length was defined as the distance from the outermost tip of the apertural lip to the apex, while drill hole diameters were defined as the distance across the center of the drill hole between outer

and inner drill hole edges, with the shell apex oriented upward (caliper tips oriented perpendicular to and touching shell surface).

2.2 Experimental setup and design

Shells from each species \times drill hole category were transferred to one of three experimental units. The experimental units were semi-static systems filled with 7 L of artificial seawater (distilled water + Instant Ocean®; temperature $\sim 16\text{--}18^\circ\text{C}$, salinity ~ 17 psu; see Table 2) that received constant aeration over the course of the experiment. While the chosen salinity was approximately half that of full seawater, creating a higher salinity with Instant Ocean would have resulted in unrealistically high alkalinity conditions that would impact dissolution parameters. Furthermore, salinity conditions in the nearshore estuarine environments where these shells were collected can be as low as 10 psu. Each unit consisted of two plastic containers (8 L capacity; 25 cm depth), with one placed inside the other; the bottom of the inside container was removed and replaced with mesh prior to setup; shells sat on top of the mesh during exposure. This allowed shells to be situated ~ 5 cm off the bottom of the outer container and provided maximum surface area exposure to seawater. Full water changes were conducted twice weekly (every 3–4 days) to minimize alkalinity buildup and an increase in salinity over time due to evaporation (Table 1; see description in Section 2.3 *Carbonate system manipulation*); disruption of $p\text{CO}_2$ was negligible during water changes (fresh water equilibrated to experimental conditions within ~ 10 mins).

Each container corresponded to one of three $p\text{CO}_2$ treatments: 300 μatm (pre-industrial control), 800 μatm (RCP8.5 near-future scenario; Hoegh-Guldberg et al., 2014), and 4000 μatm (extreme conditions inferred in the geologic past and currently in nearshore coastal areas); details on carbonate system manipulation are found in the next section (2.3 *Carbonate system*

manipulation). While the high $p\text{CO}_2$ treatment exceeded average conditions estimated in the geologic past (e.g. Kump et al., 2009), such conditions are not uncommon for prolonged periods of time in current nearshore coastal and estuarine waters (e.g. Provoost et al., 2010). Eighteen replicate shells from each species \times drill hole category (with the exception of two groups which only had 17 replicate shells) were randomly assigned to each $p\text{CO}_2$ treatment ($n_{\text{total}} = 70\text{--}72$ shells per $p\text{CO}_2$ treatment). Shells were sorted to ensure an even size-frequency distribution across treatment combinations and were evenly positioned within each experimental unit so that they were not touching; drill hole orientation was not standardized.

Shells were exposed to their respective $p\text{CO}_2$ conditions for a total of five weeks. Measurements of shell mass (dry shell weight to the nearest 0.1 mg) and drill hole diameters (inner and outer) were taken again for each individual shell after exposure. Shells were dried at 70°C for 72h to ensure there was no residual moisture prior to weighing. The shells were subsequently crushed using a digital force gauge (Digital Measurement Metrology Inc.) to assess shell breakage force (in Newtons) for each individual shell. For the latter, shells were placed on a steel plate with the aperture facing down and compressive pressure was applied to the top of the shell with a flat surface until the shell broke (Figure S2); the maximum compressive force applied to break the shell was recorded.

2.3 Carbonate system manipulation

Seawater $p\text{CO}_2$ was controlled with CO_2 injection using a typical aquarium CO_2 control system (American Marine Inc.) to maintain the two elevated $p\text{CO}_2$ treatments (mid and high); the control treatment was maintained simply by not adding CO_2 . Two CO_2 cylinders, each fitted with a regulator and solenoid valve, were connected to individual pH controllers calibrated (two-point

calibration to pH 4.0 and 7.0 with potassium phosphate buffers; Hoskin Scientific) and adjusted to a pH setpoint of either 7.7 (mid pH, mid $p\text{CO}_2$) or 7.0 (low pH, high $p\text{CO}_2$). A hose equipped with an air stone was connected to each solenoid valve and supplied CO_2 to each seawater container when the pH increased 0.1 units above the respective setpoint; the CO_2 delivery ceased once the pH returned to the setpoint (see Table 2).

For each treatment we recorded seawater pH, temperature, and salinity every 1–2 days. Seawater pH was measured using a standard handheld pH meter and probe (Oakton® PC 450); probes were calibrated weekly as described above for the pH controllers. Seawater temperature and salinity were measured using a handheld multi-parameter sonde (YSI® Pro30 conductivity / TDS / salinity / temperature meter). Total alkalinity (A_T) was also measured at the beginning and end of each water change (every 3–4 days) to ensure that changes in shell mass for each treatment could be confidently attributed to shell dissolution (as increases in alkalinity would signify shell dissolution; Chisolm and Gattuso, 1991); A_T was measured manually using two-point Gran titrations with 0.01 N hydrochloric acid (HCl) on 5 mL seawater samples as per the methods of Edmond (1970). While A_T increased over the time interval between water changes (particularly in the high $p\text{CO}_2$ treatment), saturation states of calcite and aragonite between treatments never overlapped and A_T continued to increase linearly past 4 days in each treatment (i.e., shell dissolution occurred over the entirety of the 4 day periods). Seawater pH, temperature, salinity, and initial A_T were used to estimate additional carbonate system parameters ($p\text{CO}_2$, bicarbonate concentration, and the saturation states of calcite and aragonite) in CO2SYS (Pierrot et al., 2006) using the first and second dissociation constants of Mehrbach et al. (1973) refit by Dickson and Millero (1987).

2.4 Statistical analyses

All statistical analyses were conducted in R version 3.4.1 (R Core Team, 2017) with a significance threshold of $\alpha = 0.05$. Supplementary File 1 contains R-Script for all analyses and a general overview of the factors tested can be found in Table 1; raw data can be found in Supplementary Files 2–4. Q-Q plots and residual variance plots were used to visually assess assumptions of normality and homoscedasticity, respectively. We used linear mixed effects (LME) modelling to test for the effects of species (fixed factor with two levels), drill hole category (fixed factor with two levels), and $p\text{CO}_2$ (fixed factor with three levels) on shell dissolution (computed as the percentage of shell mass loss over the course of the experiment) and shell breakage force. LME was also used to test for effects of species and $p\text{CO}_2$ on % drill hole diameter change. For all models, shell length (in mm) and shell ID (i.e., individual shells) were included as random variables. Unlike ANOVA, this approach partitions the error term and therefore increases analytical power (Galecki and Burzykowski, 2013). Prior to analyses, percentage data (i.e., % shell mass loss and % drill hole diameter change) were arcsin transformed (Ahrens et al., 1990) and breakage force data were log transformed to remove initial violations of statistical assumptions (i.e., non-normality and heteroscedasticity). All LME analyses were run using the ‘nlme’ package (Pinheiro et al., 2018) and, where significant effects were detected, Tukey HSD post hoc tests were conducted using the ‘multcomp’ package (Hothorn et al., 2008).

3. Results

3.1 Shell dissolution

LME revealed a significant interaction between species and $p\text{CO}_2$ on shell mass loss. While shell mass loss under the low $p\text{CO}_2$ treatment was negligible for both *E. heros* and *L. littorea*

shells, *E. heros* shells lost more mass than *L. littorea* shells under the mid- and high $p\text{CO}_2$ treatments (Figure 1a). On average, *E. heros* and *L. littorea* shells, respectively, lost 0.2% and 0.0% of their shell mass in the low $p\text{CO}_2$ treatment, 0.8% and 0.1% in the mid $p\text{CO}_2$ treatment, and 6.8% and 4.0% in the high $p\text{CO}_2$ treatment. A significant interaction between drill hole category and $p\text{CO}_2$ was also evident; however, this effect was solely driven by the effect of elevated $p\text{CO}_2$, as there were no differences in mass loss between drilled and non-drilled shells within $p\text{CO}_2$ treatments (see Supplementary File 1). Shells from each $p\text{CO}_2$ treatment showed visual signs of dissolution as well, with increasing $p\text{CO}_2$ resulting in increased colour loss on the shell exterior; this was particularly apparent for *L. littorea* (Figure 1b). Differential increases in A_T across treatments over the 3–4 day periods between water changes support that shell mass loss was driven by shell dissolution (Table 2).

3.2 Shell breakage force

Species and drill hole category independently affected shell breakage force, while $p\text{CO}_2$ had no effect; all interactive effects were non-significant (Table 3). For *L. littorea*, it took approximately 146 more Newtons of force, on average, to break non-drilled shells (415.1 N) than it took to break drilled shells (293.9 N) (Figure 2a). In contrast, the force required to break drilled *E. heros* shells was approximately 49 more Newtons (269.2 N) than non-drilled shells (220.2 N) (Figure 2a). Shell breakage force was similar across $p\text{CO}_2$ treatments (Figure 2b).

3.3 Drill hole diameter

LME revealed a significant species \times $p\text{CO}_2$ interaction on ODD change (Table 4). On average, ODD increased with $p\text{CO}_2$ for both species. While the change in ODD did not differ across $p\text{CO}_2$ treatments for *L. littorea*, the increase in ODD was greater under higher $p\text{CO}_2$ for *E.*

heros shells (Figure 3a). For *L. littorea* shells, ODD increased, on average, by 2%, 8%, and 1% in the 300, 800, and 4000 $\mu\text{atm } p\text{CO}_2$ treatments, respectively. In contrast, ODD in *E. heros* shells had an average increase of 0%, 3%, and 12% in the 300, 800, and 4000 $\mu\text{atm } p\text{CO}_2$ treatments, respectively.

Comparatively, LME revealed a significant independent effect of $p\text{CO}_2$ on the change in IDD (Table 4). IDD increased with $p\text{CO}_2$ for both species and this increase was significantly different between the low and high $p\text{CO}_2$ treatments (Figure 3b). For *L. littorea* shells, IDD increased, on average, by 2%, 4%, and 10% in the 300, 800, and 4000 $\mu\text{atm } p\text{CO}_2$ treatments, respectively. In *E. heros* shells, IDD increased by 3%, 5%, and 13% in the 300, 800, and 4000 $\mu\text{atm } p\text{CO}_2$ treatments, respectively. Observer measurement error was 2.12% (range: 0.83% – 3.33%) for OBD and 2.27% (1.68% – 3.03%); we are therefore confident that significant increases in drill hole diameter (>10%, on average) were real and not an artifact of observer error.

4. Discussion

This experiment set out to test for effects of elevated $p\text{CO}_2$ on taphonomic aspects (dissolution rates, shell breakage force, and drill hole morphology) in post-mortem shells of two prey species (*Littorina littorea* and *Euspira heros*) of the northern moonsnail (*E. heros*). While the data were accompanied by substantial variability, perhaps due in part to the initial taphonomic state of individual shells despite our attempts to standardize this, clear yet complex patterns were evident. Our results indicated that elevated $p\text{CO}_2$ (and thus lower pH) consistent with near-future and historical conditions, and conditions often observed in nearshore coastal/estuarine waters, resulted in greater post-mortem shell dissolution rates for both species

(more-so for *E. heros*). Our results also revealed that shells with drill holes consistently had a lower breakage force than shells without drill holes, and that *E. heros* shells had a lower breakage force than *L. littorea*. While drilled shells were weaker than non-drilled shells, elevated $p\text{CO}_2$ did not affect the magnitude of difference in breakage force between drilled and non-drilled shells for either species. Finally, we observed contrasting effects of elevated $p\text{CO}_2$ on outer (ODD) and inner (IDD) drill hole diameters, whereby elevated $p\text{CO}_2$ resulted in increased IDD for both species and increased ODD for *E. heros* only, compared to lower $p\text{CO}_2$ treatments. These results ultimately suggest that effects of OA on shell characteristics in a naticid predator-prey system, although nuanced, have the potential to affect inferences of predator-prey ecology based on forensic assessments of post-mortem shells. Such effects could be applicable to both the fossil record and extant naticid predator-prey systems, although future research into this area is warranted.

4.1 Taphonomic effects of species and drill holes

Previous studies have reported that gastropod drill holes can reduce the force necessary to break shells. Using compressive force (similar to our methods), Roy et al. (1994) reported that naticid drill holes significantly weakened Recent bivalve shells (*Mulinia lateralis* (Say, 1822)) and suggested that drilled shells may be preferentially broken. Zuschin and Stanton (2001) observed similar results, reporting that drill holes significantly weakened *Anadara* shells, but that this weakening did not elicit a strong taphonomic bias in natural assemblages. Our results corroborate previous studies using bivalve shells and suggest that drill holes can significantly weaken gastropod shells. Studies have consistently reported, however, that the presence of drill holes does not necessarily result in the preferential breakage of shells from an assemblage under natural physical forces (Zuschin and Stanton, 2001; Kelley, 2008; Chojnacki and Leighton,

2013). Comparatively, compressive forces experienced during burial in nature are well below those that were required to break the shells in this study. Thus, while our results suggest that drill holes weaken post-mortem shells of two gastropods, it seems unlikely that this would result in significant taphonomic bias in fossil and Recent assemblages. Our results are thus more applicable to other sources of post-mortem compression such as crushing or hammering by live animals, and compression from heavy stones on top of the shells (Zuschin and Stanton, 2001). It should also be noted that drill hole presence and shell morphometry (including size and shape) can result in differential shell removal due to hydrodynamic forces (Chattopadhyay et al., 2013a,b). Given that elevated $p\text{CO}_2$ affected shell morphology herein (i.e., resulted in lighter shells), the influence of elevated $p\text{CO}_2$ on hydrodynamic sorting is warranted.

Relative degrees of shell breakage and fragmentation between species may also affect inferences of predator-prey interactions based on drilling frequency estimates. For example, Klompmaker (2009) reported that drilling frequency estimates based on non-fragmented bivalve shells (i.e., excluding fragmented shells) were significantly higher than frequencies estimated from a combination of non-fragmented and fragmented shells. In this study, we observed significantly different breakage forces between *E. heros* and *L. littorea* shells, with *E. heros* having a lower breakage force than *L. littorea*. While assessments of fragmentation under natural physical conditions for a broad range of species varying in shell strength are not readily available, natural shell fragmentation is likely greater for species with weaker shells. Given that the incorporation or dismissal of fragmented shells can influence estimates of drilling frequency (Klompmaker, 2009), differential fragmentation across species should be considered in such analyses. These considerations are particularly relevant given that naticid gastropods often forage on a wide range of prey species that can be differentially impacted by taphonomic effects (e.g.

Kabat, 1990; Huelsken, 2011; Clements et al., 2013; Clements and Rawlings, 2014; Villegas-Martín et al., 2016).

4.2 *The taphonomic role of ocean acidification*

While studies suggest that the presence of a naticid drill hole may not invoke taphonomic bias for drilling frequencies in shell assemblages (Zuschin and Stanton, 2001; Kelley, 2008; Chojnacki and Leighton, 2013), other factors may. For example, shell thickness can influence the relative preservation of shells within a given shell assemblage (Grey et al., 2006). The probability of shell preservation is further complicated by the cumulative effects of multiple taphonomic forces that may fragment or remove some shells at higher rates than others (Zuschin and Stanton, 2001). We saw substantially higher rates of shell dissolution under elevated $p\text{CO}_2$; however, there were no independent or interactive effects of $p\text{CO}_2$ on shell breakage force. This result suggests that the effects of physical taphonomic agents such as wave energy, shell transportation/tumbling, and lithification (among others) are unlikely to be influenced by OA for gastropod shells (though studies with a wider diversity of taxa are needed). This contradicts our initial prediction that elevated $p\text{CO}_2$ would result in weaker shells, but these results are not entirely surprising. For example, Landes and Zimmer (2012) reported that elevated $p\text{CO}_2$ did not significantly affect shell breakage resistance in live *L. littorea*, and indeed smaller snails (7 – 8 mm) were more difficult to break after exposure to combined elevated temperature and $p\text{CO}_2$ when compared to control conditions. It therefore seems unlikely that elevated $p\text{CO}_2$ would affect comparative breakage frequencies in gastropod shells across time and space, and is thus unlikely to impact the results of studies comparing drilling frequency estimates across geologic time (Kelley and Hansen, 1993; Huntley and Kowalewski, 2007; Mondal et al., 2017).

Furthermore, given that there was no interactive effect between $p\text{CO}_2$ and drill hole presence in

this study, elevated $p\text{CO}_2$ is unlikely to influence relative breakage frequencies between drilled and non-drilled shells within a given species. It is important to note here, however, that we only exposed shells to elevated $p\text{CO}_2$ for five weeks, and longer exposure durations may reveal different outcomes for shell breakage force and, thus, the influence of OA on other physical taphonomic forces (transportation, lithification, etc.); future studies employing longer exposure periods are needed to address this knowledge gap. Furthermore, the behaviour of predators and their prey can also be impacted by elevated $p\text{CO}_2$ (Briffa et al., 2012; Leduc et al., 2013; Clements and Hunt, 2015) and should be considered when inferring predator-prey behaviour in the fossil record.

The lack of $p\text{CO}_2$ effect on shell breakage in this study may be explained by a number of factors. For example, while differential rates of shell dissolution were clearly observed across $p\text{CO}_2$ treatments, only softer parts of the shells (e.g. periostracum, weaker shell material that is more susceptible to dissolution) may have dissolved over the five-week exposure, leaving behind the stronger and more robust shell material. Thus, if the stronger shell materials are largely responsible for shell strength and integrity, the dissolution of soft materials may not have been sufficient to result in increased shell weakness. Shell shape can also affect shell breakage (Vermeij, 1979; Zuschin and Stanton, 2001). While not empirically measured, we observed no indication of elevated $p\text{CO}_2$ altering shell shape, as has been observed in other taxa (e.g. foraminifera: Khanna et al., 2013; larval bivalves: Talmage and Gobler, 2010; Kim et al., 2016; juvenile gastropods: Ramajo et al., 2013). In the same vein, we did not assess $p\text{CO}_2$ effects on the internal shell architecture. Although dissolution occurred and was visible for the outer shell, the internal structure may not have been compromised, resulting in no $p\text{CO}_2$ effect on shell

breakage force. Ultimately, the mechanism(s) underpinning the consistent breakage forces across $p\text{CO}_2$ treatments observed in this study awaits further research.

Although we found no evidence that relative fragmentation frequencies between drilled and non-drilled gastropod shells would be affected by elevated $p\text{CO}_2$, other inferences from shell assemblages may be. Indeed, we observed effects of elevated $p\text{CO}_2$ on shell dissolution, which is known to play a major taphonomic role in shell preservation (Powell et al., 2011). With respect to shell dissolution, the most obvious taphonomic effect is in the overall preservation of shells. Under periods of high $p\text{CO}_2$, shells dissolve faster than under periods of lower $p\text{CO}_2$ and have a lower likelihood of preservation; this effect is most pronounced for aragonitic shells, given that aragonite is a highly soluble mineral form of calcium carbonate (e.g. compared to calcite; Cubillas et al., 2005; Tynan and Opdyke, 2011; Jordan et al., 2015). The observed disproportionate dissolution rates between the two species herein at mid- and high- $p\text{CO}_2$ treatments, but not the low $p\text{CO}_2$ treatment, offer potential implications for the assessment of shell abundance and preservation across geologic time (Cherns and Wright, 2000, 2009; Wright et al., 2003; Cherns et al., 2011), as high dissolution rates may lead to erroneous estimates. It is important to note here that our high $p\text{CO}_2$ treatment of 4000 μatm is higher than the average estimated value for both past and future conditions (e.g. Kump et al., 2009). Such a value is, however, not uncommon for prolonged periods of time in nearshore coastal and estuarine systems (e.g. Provoost et al., 2010). Nonetheless, future studies repeating our experiments should be sure to also assess dissolution rates of various shells under more modest $p\text{CO}_2$ conditions compared to our high (4000 μatm) treatment. Ultimately, inferences of predator-prey ecology of drilling gastropod systems (Chiba and Sato, 2016) could potentially reflect post-mortem

taphonomy due to climate-mediated alterations rather than ecological functioning, and further study is needed to tease apart these effects to enhance predictive capacity.

Alongside biological and ecological inferences, differential shell half-lives across taxa can result in significant influences in biological age estimates when using time-averaging techniques (Kosnik et al., 2009). Given the disproportionate and higher rates of dissolution observed under elevated $p\text{CO}_2$ in this study, it is possible that shell half-lives (i.e., time needed to destroy 50% of shells) within and across species could depend on the $p\text{CO}_2$ conditions during the time of preservation (although further research is needed to confirm or refute this hypothesis). Our results also revealed that the relative dissolution rate between two gastropod species was dependent on the $p\text{CO}_2$ level, as *E. heros* and *L. littorea* dissolution rates were statistically comparable at 300 μatm but different at 800 and 4000 μatm . As such, pooling specific taxa for time-averaging purposes may be appropriate for some time periods and not for others depending on the relative $p\text{CO}_2$ conditions (due to the differential effects of elevated $p\text{CO}_2$ on shell dissolution, and thus, potentially shell half-lives). A suite of other taphonomic processes such as abrasion, edge alteration, and changes to a variety of other distinguishing morphological characteristics may also be influenced by $p\text{CO}_2$ conditions during shell preservation. While our study does not provide sufficient evidence to answer such questions, future research in this direction is warranted.

Drill hole size is a direct indicator of predator size in forensic analyses of naticid drilling predation (Kitchell et al., 1981; Dietl and Alexander, 2000; Kowalewski, 2004; Beal 2006; Chiba and Sato, 2012; Clements and Rawlings, 2014; Klompaker et al., 2017). We observed a limited effect of elevated $p\text{CO}_2$ on drill hole diameters that were a function of species and diameter type (i.e., inner versus outer). For *E. heros* elevated $p\text{CO}_2$ conditions resulted in increases in both

inner and outer drill hole diameters compared to lower $p\text{CO}_2$ conditions. In contrast, only inner drill hole diameter increased in *L. littorea* under higher $p\text{CO}_2$. This is intuitive for *L. littorea*, as the outer shell is calcitic and thus has a greater ability to resist dissolution than aragonite (Harper, 2000; Cubillas et al., 2005) while the inner layer is aragonitic (Bandel, 1990). While a detailed documentation of the carbonate composition of *E. heros* shells is, to our knowledge, unavailable, it seems likely that *E. heros* shells would have a greater aragonite component, given that both drill hole diameters increased at similar rates and overall shell dissolution rates were higher (compared to *L. littorea*).

Importantly, these findings could potentially have implications for the use of drill hole size and morphology to infer predator size and identity. Herein, there exists a potential risk of overestimating a predator's size under conditions of elevated $p\text{CO}_2$. Given that shells were only exposed to elevated $p\text{CO}_2$ for five weeks, our data may be a conservative estimate for the magnitude of this effect. Furthermore, five weeks of exposure to elevated $p\text{CO}_2$ in nearshore coastal and estuarine systems (intertidal and subtidal) is not unexpected for Recent shells of extant systems, as current nearshore and estuarine systems often reach $p\text{CO}_2$ and pH conditions at or beyond those employed in this study and can exhibit substantial spatial and temporal variability on both small and large scales (Provoost et al. 2010; Duarte et al., 2013; Waldbusser and Salisbury, 2014; Wallace et al., 2014). Comparative inferences of predator sizes from drill holes at different times and/or in shells from different locations—even those within close proximity to one another—may thus be impacted by $p\text{CO}_2$ conditions, as increased drill hole sizes across individuals within a sample may increase mean estimates of predator size and their associated variance. Additionally, given that inner and outer drill holes did not change equally for *L. littorea* (but did for *E. heros*), elevated $p\text{CO}_2$ may also impact drill hole inferences of

drilling predator identity based on drill hole morphology (e.g. IDD:ODD ratios: Grey et al., 2005; Dietl and Kelley, 2006), although more research to this extent is required. It is important to note, however, that the increases in drill hole diameter under elevated $p\text{CO}_2$ were within the variability observed around average predator size estimates (e.g. Clements and Rawlings, 2014) and the effects of elevated $p\text{CO}_2$ on size inferences may be limited. Ultimately, while inferences of predator size and identity from drill hole size and shape across prey taxa in both Recent and fossil assemblages may be biased if the sampled shells were exposed to different $p\text{CO}_2$ conditions post-mortem, future studies explicitly testing this effect are needed.

4.3 Limitations of the current study

While the results herein highlight that OA can potentially influence taphonomy in gastropod drilling predation systems, our study does not come without limitations. Firstly, it is important to note a number of methodological limitations of our approach. For example, post-mortem shells don't usually float and instead lay on the sediment surface, and those shells preserved in the fossil record are buried quickly and deeply in sediment. Consequently, beach-collected shells like those used herein tend to have a lower probability of preservation due to a lack of sediment burial, and likely have more weathering/taphonomic damage. To this extent, Klompaker et al. (2017) reported gradually-increasing drill hole diameters across the Phanerozoic, which do not appear to correlate with oceanic $p\text{CO}_2$ changes (e.g. Hönisch et al., 2012). Thus, the shells used in our experiment may not be representative of shells in other areas (e.g. continental shelf) that have a higher probability of burial and preservation.

Placing shells on a porous mesh elevated off the bottom of our experimental chambers may also give inaccurate estimates of dissolution that would be observed under natural conditions due

to the lack of shell burial and no contact with a soft-sediment bottom. Indeed, porewater $p\text{CO}_2$ and pH conditions can often be different than that of the overlying water (porewater can be more acidic than overlying water when the overlying water pH is relatively high, or *vice versa*; Widdicombe et al., 2011; Clements and Hunt, 2017). Furthermore, our high $p\text{CO}_2$ treatment (where the majority of effects were observed) is higher than average conditions estimated in the geologic history of drilling predation (Veron, 2008; Kump et al., 2009; Hönisch et al., 2012; Clarkson et al., 2015), and we used artificial seawater with a relatively low salinity rather than natural seawater. As such, while our results suggest that OA may influence shell taphonomy and increase the susceptibility of post-mortem shells to taphonomic agents, far more research is needed to apply our results more precisely to drilling predation systems.

It is also important to recognize that breakage force does not directly translate into shell breakage or loss from an assemblage. In many cases, while gastropod drill holes resulted in weaker shells, the degree of weakness was not enough to result in preferential shell breakage in weaker shells under natural conditions (Zuschin and Stanton, 2001; Kelley, 2008; Chojnacki and Leighton, 2013). The shells in our experiment were also placed on a mesh and elevated from the bottom of our experimental chambers to ensure maximum exposure, which contrasts with the physical placement of shells on (or in) sediments under natural conditions. It is therefore important to note that factors affecting shell breakage force in this study may not translate directly into weaker shells being preferentially removed from an assemblage under natural conditions, and tumbling/wave energy experiments would serve well in more accurately testing for OA effects on shell fragmentation. It should also be noted that we used crushing force as an estimate for shell breakage strength, and that other types of physical force such as punctures and direct blows/hits may elicit different results regarding breakage and fragmentation than those

observed herein. Furthermore, we only used a five-week exposure period, and the effects of longer-term exposure to elevated $p\text{CO}_2$ are not interpretable from our short-term exposure. Nonetheless, given the differences in dissolution observed in our study with short-term (five week) exposure, elevated $p\text{CO}_2$ may impact the relative loss of shells from a given assemblage (Cherns and Wright, 2000, 2009; Wright et al., 2003; Cherns et al., 2011; Tynan and Opdyke, 2011; Jordan et al., 2015). Future studies assessing the taphonomic effects of elevated $p\text{CO}_2$ under more natural conditions (e.g. on top of sediment vs. buried and under natural hydrodynamic conditions) are also warranted.

In this experiment, we used gastropod shells to test hypotheses regarding the taphonomic roles of OA. Drilling predators are known to prey on a suite of taxa with varying shell morphometries (both presently and in the geologic past)—some of which may be more prone to dissolution/breakage than others, including bivalves and gastropods, brachiopods, tube-dwelling polychaetes, scaphopods, ostracods, decapods, chitons, and barnacles, among others (e.g. Kelley and Hansen, 2003; Hoffmeister et al., 2004; Morton and Harper, 2009; Klompmaker, 2012; Martinell et al., 2012; Harper, 2016; Klompmaker et al., 2015). Furthermore, while the presence of drill holes in molluscan prey may not result in the preferential fragmentation of drilled shells (Zuschin and Stanton, 2001; Kelley, 2008; Chojnacki and Leighton, 2013), high rates of breakage across drill holes in other taxa such as tube-dwelling polychaetes suggest that drill holes could result in preferential breakage for other species (Klompmaker, 2012). Consequently, our results may not be applicable to other prey taxa of drilling predators. Estimating the taphonomic effects of OA on other drilling gastropod prey should thus be considered for each species independently.

Finally, this study tested for the potential for OA to influence shell taphonomy while keeping other environmental parameters (i.e., temperature, salinity, oxygen, light) constant. It is important to note, however, that differences in our controlled environmental parameters can act independently or interactively with elevated $p\text{CO}_2$ to affect rates of dissolution. For example, molluscan shell dissolution rates under elevated $p\text{CO}_2$ are documented to differ depending on temperature (Ries et al., 2016). Thus, the environment or microenvironment where a post-mortem shell ends up may influence the taphonomic state of the shell. Future studies assessing the taphonomic roles of OA would benefit from testing such hypotheses in the context of multiple environmental parameters.

5. Conclusions

We tested for effects of OA on gastropod shell taphonomy in the context of a naticid predator-prey system. We found that OA impacted some aspects of shell taphonomy, but not others. Our results suggest that OA has the potential to influence shell dissolution, shell colouration, and drill hole size, and may thus potentially influence paleoecological inferences regarding naticid predator size, identity, and assemblage biodiversity. In contrast, however, we observed no independent or interactive effects of OA on shell breakage strength, suggesting that preferential post-mortem fragmentation, and therefore drilling frequency inferences, are unlikely to be affected by OA (although alternative experimental methods employing longer exposure times and tumbling experiments may yield different results). Ultimately, our study serves as a starting point for assessing the role of OA in molluscan shell taphonomy in gastropod drilling predation systems and paves the way for future research. Such research would serve to increase understanding about the role of OA in palaeoecological and palaeobiological inferences.

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Highlights

- Tested for taphonomic effects of elevated $p\text{CO}_2$ in a naticid predator-prey system
- High $p\text{CO}_2$ induced greater shell dissolution rates, which differed across species
- Breakage force differed across species and drill hole category
- No $p\text{CO}_2$ effect on shell breakage force
- Limited species-specific drill hole diameter increase under high $p\text{CO}_2$

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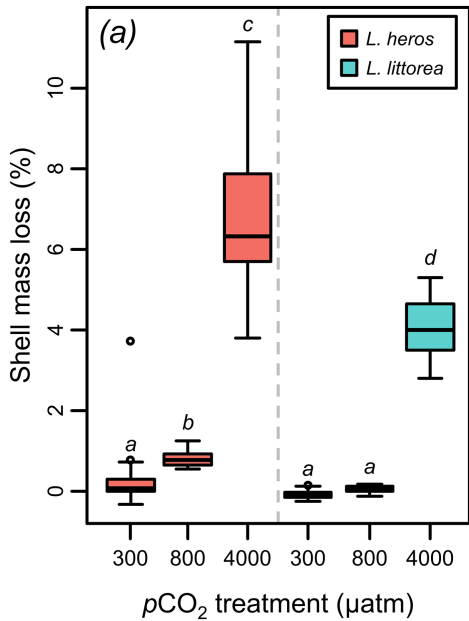


Figure 1

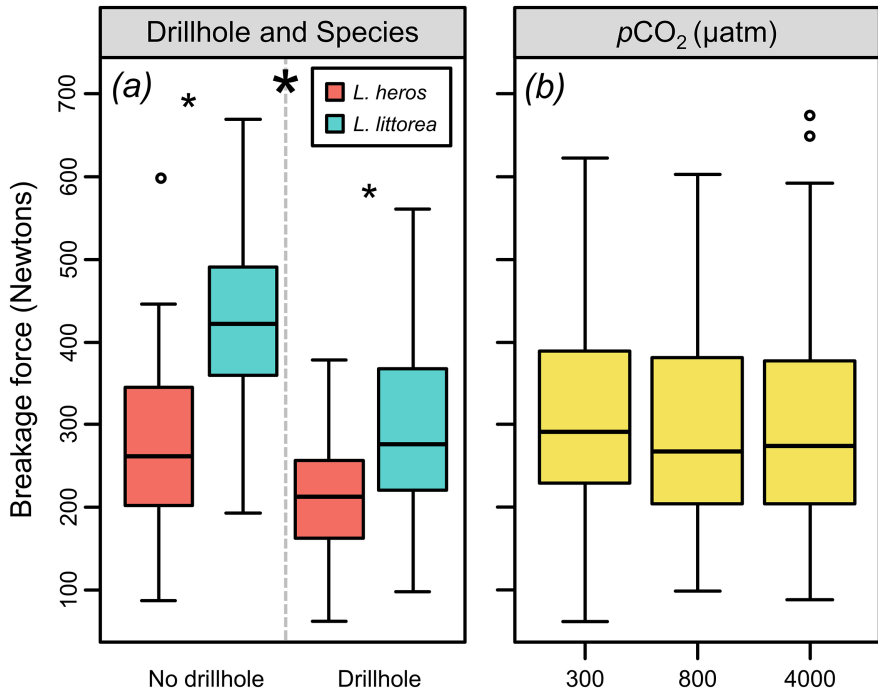


Figure 2

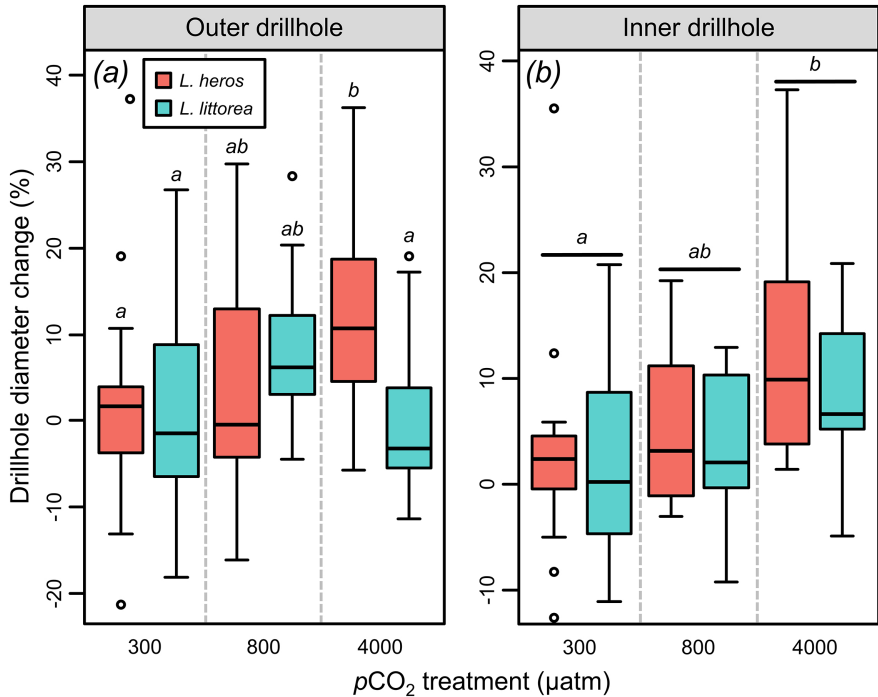


Figure 3

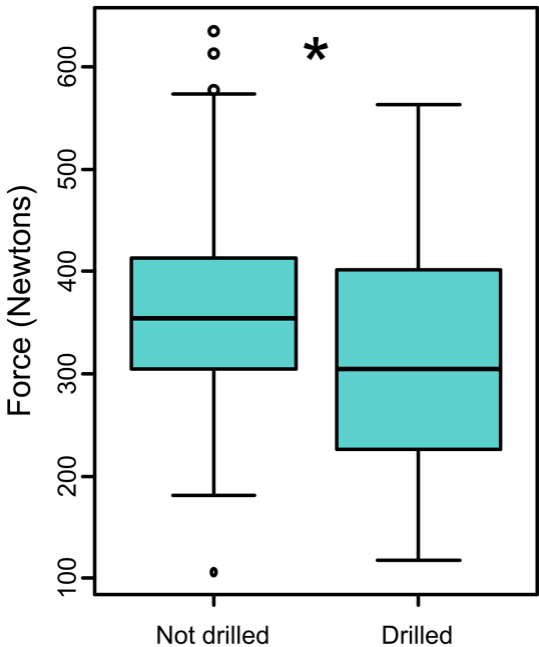


Figure 4

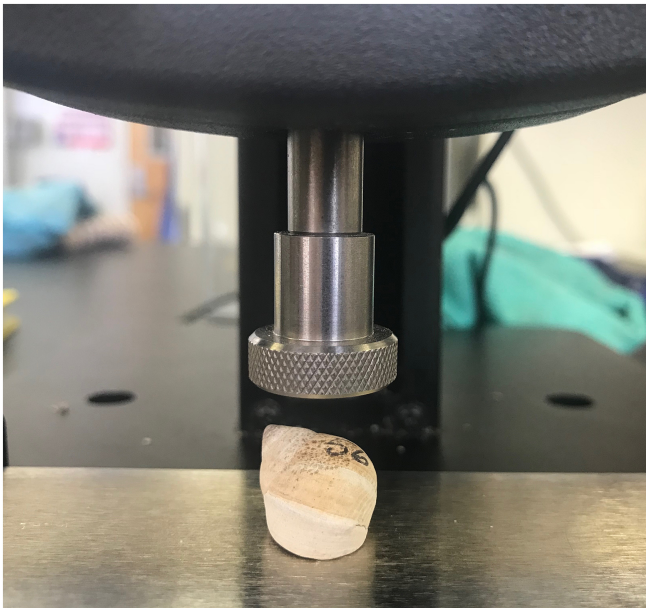


Figure 5