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RESEARCH PAPER

Testing the Utility of Geochemical Proxies to Reconstruct Holocene Coastal Environments and Relative Sea Level: A Case Study from Hungry Bay, Bermuda

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On low-lying, tropical and sub-tropical coastlines freshwater marshes may be replaced by salt-tolerant mangroves in response to relative sea-level rise. Pollen analysis of radiocarbon-dated sediment cores showed that such a change occurred in Hungry Bay, Bermuda during the late Holocene. This well-established paleoenvironmental trajectory provides an opportunity to explore if geochemical proxies (bulk-sediment $\delta^{13}C$ and Rock-Eval pyrolysis) can reconstruct known environmental changes and relative sea level. We characterized surface sediment from depositional environments in Bermuda (freshwater wetlands, saline mangroves, and wrack composed of *Sargassum natans* macroalgae) using geochemical measurements and demonstrate that a multi-proxy approach can objectively distinguish among these environments. However, application of these techniques to the transgressive sediment succession beneath Hungry Bay suggests that freshwater peat and mangrove peat cannot be reliably distinguished in the sedimentary record, possibly because of post-depositional convergence of geochemical characteristics on decadal to multi-century timescales and/or the relatively small number of modern samples analyzed. Sediment that includes substantial contributions from *Sargassum* is readily identified by geochemistry, but has a limited spatial extent. Radiocarbon dating indicates that beginning at –700 CE, episodic marine incursions into Hungry Bay (e.g., during storms) carried *Sargassum* that accumulated as wrack and thickened through repeated depositional events until ~300 CE. It took a further ~550 years for a peat-forming mangrove community to colonize Hungry Bay, which then accumulated sediment rapidly, but likely out of equilibrium with regional relative sea-level rise.

Keywords: *Sargassum*; Rock-Eval pyrolysis; mangrove; radiocarbon; $\delta^{13}C$

1. Introduction

Relative sea-level (RSL) rise and transgression of the coast in tropical and sub-tropical regions causes freshwater wetlands to be increasingly inundated by storm tides and ultimately to either be replaced by salt-tolerant mangroves or lost to erosion. Organic coastal sediment that records this change in depositional environment can be used to reconstruct RSL and to describe the response (trajectory and rate) of coastal ecosystems to RSL rise (e.g., Ellison, 2005, 2008). This long-term perspective is a paleo analogue for environmental changes that are likely to occur as mangroves colonize newly-brackish zones under scenarios of 21st century sea-level rise and beyond (e.g., Mengel et al., 2018; Wong et al., 2014). In Bermuda, the background rate of late Holocene RSL rise is estimated at ~0.7 mm/yr (ICE6G_C VM5a Earth-ice model; Argus et al.,

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2014; Peltier, Argus & Drummond, 2015), compared to 2.17 mm/yr measured by tide gauges from 1932 to 2017. Continued acceleration in the rate of RSL rise under the RCP8.5 emission scenario results in a predicted 21st century RSL rise for Bermuda of 0.68 m (0.20–1.34 m, 95% credible interval; Kopp et al., 2014).

Reconstructing the botanical and environmental provenance of buried sediment relies on proxies such as plant macrofossils, counts of microfossil abundances (e.g., pollen; Berglund, 1987), and, more recently, geochemical analyses (e.g., Wilson, 2017). Since variable amounts and proportions of marine and terrestrial material (with different geochemical compositions) are deposited in sub-, inter-, and supra-tidal settings, geochemistry provides valuable tools for recognizing coastal environments in the stratigraphic record (e.g., Lamb, Wilson & Leng, 2006). The material used for geochemical analysis can be bulk (i.e., undifferentiated) sediment, or a specific subsample (e.g., a compound or size fraction) of the sediment. Measurements made on bulk sediment include total organic carbon (TOC), stable carbon isotopes ($\delta^{13}C$), the ratio of carbon to nitrogen (C:N), and a suite of values from Rock-Eval pyrolysis. Analysis of TOC, $\delta^{13}C$, and C:N (usually measured simultaneously) is an established technique in paleoenvironmental research including reconstructing Holocene RSL (e.g., Goslin et al., 2017; Lamb et al., 2006). Rock-Eval pyrolysis is an established technique for evaluating organic material in source rocks (and their modern analogs) for hydrocarbon characterization and exploration (e.g., Girkin et al., 2018; Lafargue, Marquis & Pillot, 1998), but is increasingly being used to generate paleoenvironmental reconstructions from recent sediments (e.g., Ariztegui et al., 2001; Lacey et al., 2018; Newell et al., 2016; Stein, 2008; section 3). By differentiating the relative contributions of marine- and terrestrial-derived organic matter, Rock-Eval pyrolysis has the potential to reconstruct Holocene coastal environments and RSL (e.g., Engelhart et al., 2013; Hare et al., 2014; Kemp et al., 2017).

The use of these proxies is underpinned by reasoning through analogy, in which the observable and measurable characteristics of modern sedimentary environments are used to recognize and interpret their equivalents preserved in the sedimentary record (Jackson & Williams, 2004). A first step in establishing the utility of a proxy is to test if it can reliably distinguish the environment in which modern samples accumulated, which is often achieved by objectively grouping samples (e.g., Goslin et al., 2017; Wilson, 2017). In studies seeking to reconstruct RSL, a key goal is to determine if a proxy (or multiple proxies used together) can distinguish sediment that accumulated above the influence of salt-water tides in a freshwater marsh from inter-tidal sediment that accumulated in a saline mangrove or salt marsh. Since modern samples are typically a slice of surface sediment (0–1 cm), they are time averaged (over a few years, depending on sedimentation rate) and incorporate early diagenesis. However, post-depositional change in sediment geochemistry on longer timescales (decades to centuries) can distort the relationship between sedimentary facies in the stratigraphic record and their modern counterparts (e.g., Benner, Fogel & Sprague, 1991). This longer-term change cannot be captured by analyzing only modern samples and is often overlooked in evaluating proxies, with the potential effect that diagenesis is misinterpreted as paleoenvironmental change (e.g., Disnar et al., 2008). Examining the ability of a proxy to reconstruct a known paleoenvironmental trajectory provides a practical test of its utility and insight into the influence of post-depositional change.

Previous work in Hungry Bay, Bermuda (Ellison, 1992, 1993, 1996) reconstructed late-Holocene paleoenvironmental change by analyzing pollen in radiocarbon-dated sediment cores and demonstrated that a freshwater marsh was replaced by a saline mangrove in response to RSL rise. To investigate the utility of $\delta^{13}C$ (in conjunction with TOC and C:N) and Rock-Eval pyrolysis as paleoenvironmental and sea-level proxies in Bermuda, we first examine if geochemical analysis of surface sediment can distinguish modern depositional environments; in particular, freshwater wetlands and saline mangroves that can both be dominated by *Rhizophora mangle*, red mangrove, in Bermuda (Thomas, 1993). Second, we apply these geochemical proxies to the sediment sequence of known provenance in Hungry Bay to test their practical utility as stand-alone and complementary paleoenvironmental proxies. This analysis, coupled with radiocarbon dating, further develops the paleoenvironmental and RSL record for Hungry Bay and provides insight into the advantages and limitations of these techniques for reconstructing RSL in Bermuda and locations with similar mangrove flora.

2. Study Area

Bermuda is a group of coral limestone islands that lie on an extinct seamount in the northwest Atlantic Ocean (**Figure 1A**). Karstic erosion of Bermuda created a landscape that supports internal ponds (e.g., Mangrove Lake; **Figure 1A**) with no surface connection to the ocean. However, the permeable nature of eolianite results in a water table that rises and falls in internal ponds with daily tides and with RSL change due to the Ghyben-Hertzberg relationship. Marine water may enter some ponds through this mechanism, while others are connected to the ocean by underwater cave systems (e.g., van Hengstum & Scott, 2012). Several of Bermuda's present-day ocean bays and sounds are former ponds that were submerged by RSL rise (e.g., Harrington Sound; **Figure 1A**). These settings accumulated sediment during the Holocene that records the transition from freshwater to saline conditions (e.g., Ashmore & Leatherman, 1984; Hatcher et al., 1982; Tackaberry, Rueger & Gastaldo, 2004).

Due to its proximity to the warm Gulf Stream, Bermuda has a sub-tropical climate that supports coral-algal reefs and the most northerly mangroves in the world (Spalding, Kainuma & Collins, 2010). The largest remaining area of mangrove (~0.03 km^2) on Bermuda is in Hungry Bay (Sterrer & Wingate, 1981; **Figure 1A**) and is representative of Bermuda's mangrove ecosystems. The bay is enclosed by eolianite and connected to the open ocean by a narrow, sub-tidal inlet (1.4–1.8 m below Ordnance Datum; Ellison, 1992) and a second, ephemeral inlet that today is overwashed by unusually high-water levels (**Figure 1B**).

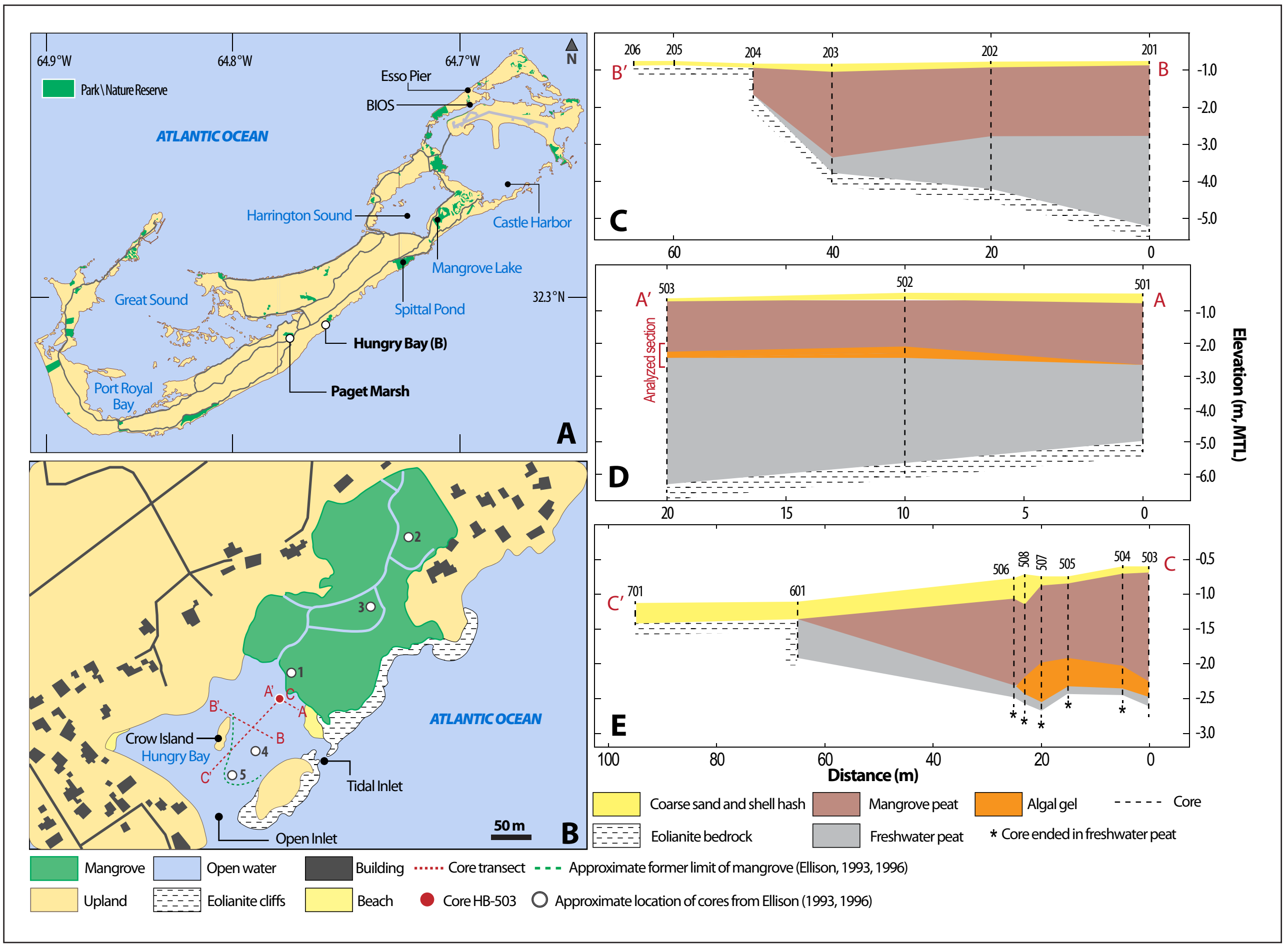

Figure 1: (A) Location of the Hungry Bay and Paget Marsh sites in Bermuda. Tide gauges are located at Esso Pier and the Bermuda Institute for Ocean Science (BIOS; formerly Bermuda Biological Station). **(B)** Position of core transects (A-A′, B-B′, C-C′) and core HB-503 in Hungry Bay. The deeper open inlet allows exchange of water with the Atlantic Ocean at all times. The shallow tidal inlet is currently only overwashed during unusually high tides and/or storms. Areas identified as upland are typically covered by soil and vegetation or developed, while areas identified as eolianite cliffs are exposed rock. **(C, D, E)** Stratigraphy underlying Hungry Bay described in the field from cores collected along transects. MTL = mean tide level.

Two arboreal species (*Rhizophora mangle* and *Avicennia germinans*, black mangrove) dominate the Hungry Bay mangrove which has retreated (**Figure 1B**) because of storm damage and RSL rise (Ellison, 1992). There are no understorey shrubs or herbs. This low diversity of mangrove species reflects Bermuda's position in the neotropical assemblage of mangrove species (Spalding et al., 2010; Woodroffe & Grindrod, 1991). More broadly, the floral diversity of mangroves in the western Atlantic Ocean (e.g., Bermuda, southern Florida, and the Caribbean) is markedly lower than in the Indo West Pacific (Ellison, Farnsworth & Merkt, 1999). Therefore, our results may not be representative of modern or paleoenvironmental conditions beyond the western Atlantic Ocean. Great diurnal tidal range (mean lower low water to mean higher high water) measured by tide gauges at Esso Pier (NOAA station ID 2695540; **Figure 1A**) and the Bermuda Biological Station (NOAA station ID 2695535) is 0.86 m and 0.83 m respectively. Mesoscale eddies can elevate or depress sea level around Bermuda for days to weeks (Siegel, McGillicuddy & Fields, 1999). Weather systems and storms overprint astronomical tides on timescales of hours to days by forcing water into, or out of, Hungry Bay through the sub-tidal inlet that is aligned with prevailing south-westerly winds (**Figure 1A**). Consequently, brown, pelagic macroalgae (*Sargassum natans* and *Sargassum fluitans*) are commonly blown into Hungry Bay from the open ocean (particularly during winter storms) and accumulate as a wrack line along the front edge of the mangroves (Ellison, 1992). Because the spatial distribution of modern wrack is restricted to a relatively narrow band, sediment formed from this material may also be limited in extent. The *Sargassum* wrack is commonly degraded by subaerial exposure and displaced by tides (Ellison, 1992), which can carry wrack ~10–20 m into the mangrove (S.R. Smith, personal observation). Prior to the creation of a man-made channel through the mangrove, penetration of wrack into the mangrove was likely less common.

Peat-forming, freshwater (<2‰ salinity) wetlands in Bermuda (e.g., Paget Marsh, Devonshire Marsh, and Pembroke Marsh; **Figure 1A**) are vegetated by ferns (e.g., *Osumunda cinnomemea*), sedges (e.g., *Carex bermudiana*), and sawgrass (*Cladium jamaicae*), with trees (*Juniperus bermudiana*, Bermuda cedar and *Sabal bermudana*, Bermuda

Palmetto) in some drier locations (e.g., Thomas, Eakins & Logan, 1991; Thomas et al., 1992). *Rhizophora mangle* communities live in freshwater at some sites (e.g., Thomas, 1993), which is notable because this species dominates saline mangroves. Since some freshwater and mangrove environments are characterized by the same species, it is an important challenge for reconstructing RSL to reliably distinguish them in the stratigraphic record. According to Thomas (2005), the total area of freshwater wetlands in Bermuda during the early 19th century was ~1.2 km^2, since when close to half of these ecosystems were lost (an estimated ~0.67 km^2 remained in 1997) and today there are relatively few (and small) sites at which to collect samples of modern, freshwater sediment.

Holocene paleoenvironments in Hungry Bay were previously established through a stratigraphic investigation and analysis of pollen preserved in five, radiocarbon-dated sediment cores (Ellison, 1992, 1993, 1996). This work identified two enclosed depositional basins that are separated by a shallow sub-surface high in the underlying eolianite. The western (lower) basin is in the seaward part of the bay, while the second (and slightly higher) basin is occupied by the current mangrove. Pollen analysis showed that the seaward (core HB-1) and landward (core HB-2) basins began to accumulate freshwater peat (as evidenced by abundant pollen from salt-intolerant taxa such as *Juniperus bermudiana* and *Sabal bermudana*) at ~5600 years before present (BP; 1950 CE). In response to RSL rise, a peat-forming mangrove community consisting of *Rhizophora mangle* and *Avicennia germinans* colonized the seaward basin at ~2000 years BP and then the landward basin at ~700 years BP (Ellison, 1992, 1993, 1996).

3. Rock-Eval Pyrolysis

Rock-Eval pyrolysis is a well-established and widely-used technique for characterizing the amount and type of hydrocarbons in potential source rocks (e.g., Lafargue et al., 1998), but is increasingly being used to generate paleoenvironmental and paleoclimate reconstructions using unconsolidated Quaternary sediments from, for example, lakes (e.g., Meyers & Lallier-vergés, 1999; Mills et al., 2018), freshwater wetlands (e.g., Upton et al., 2018), and coastal environments (e.g., Kemp et al., 2017). The analysis is performed by exposing rocks or sediment to progressive heating and subsequent cooling in an oxygen-free atmosphere and then further heating in the presence of oxygen. The response of the sample at specific times in this cycle is measured using flame ionization and infrared detectors. A broad appeal of using Rock-Eval pyrolysis is that measurements are made objectively and rapidly on relatively small samples with little pre-processing. In the specific context of reconstructing late Holocene RSL, this technique is effective at differentiating samples based on the relative proportions of marine and terrestrial organic material that they contain, which could enable recognition of sub-tidal, inter-tidal (e.g., mangroves), and freshwater environments in the stratigraphic record. Furthermore, Rock-Eval pyrolysis quantifies the maturity of organic carbon, which can provide insight into the burial history and diagenetic alteration of sediment (e.g., Roberts et al., 2018).

Measurements made at specific temperatures and times in the heating/cooling cycle provide insight on the type and maturity of organic material that is present, which may be diagnostic of depositional environments (e.g., Metcalfe et al., 2016). From these directly measured parameters, a series of additional, secondary parameters are calculated. S_1 is the concentration of free hydrocarbons (mainly small, volatile molecules; units of mg HC/g) liberated during initial heating to 300°C. The quantity of bound hydrocarbons (derived from kerogen) released by thermal cracking at temperatures of 300–500°C is S_2 (units of mg HC/g). The temperature at which S_2 reaches its maximum is T_{max}. The yield of CO_2 during breakdown of insoluble organic matter (kerogen) is termed S_3 (units of mg CO_2/g). The carbon remaining after pyrolysis (resistant and refractory compounds) is S_4 and measured in units of mg C/g, but typically expressed as a percentage by weight of the original sample. Residual carbon (RC) is calculated from the addition of two S_4 values (RC CO and RC CO_2). Hydrogen index ($[S_2 \times 100]$/TOC; units of mg/g TOC) and oxygen index ($[S_3 \times 100]$/TOC; units of mg/g TOC) are secondary parameters that can help to characterize the origin of organic material. Marine algae (e.g., *Sargassum*) typically have a higher H:C ratio and lower O:C than terrestrial plants. Consequently, sediment derived primarily from marine sources yields a higher hydrogen index and lower oxygen index than terrestrial sediment where organic matter was derived from vascular plants (e.g., leaf litter). Specific methods for Rock-Eval pyrolysis are presented in section 4.4.

4. Methods

4.1. Sampling of modern environments and sediment cores

To test if bulk-sediment geochemistry can distinguish among modern depositional environments, we collected samples of modern (surface; 0–1 cm) sediment from four freshwater-wetland communities at Paget Marsh that were vegetated by *Cladium jamaicae*, *Juniperus bermudiana*, *Myrica cerifera* (wax myrtle), and *Rhizophora mangle*. Salinity measured using a refractometer at the time of sample collection was <2‰. In Hungry Bay, we collected four modern samples of mangrove peat (two where only *Rhizophora mangle* was present and two where the plant community was *Rhizophora mangle* and *Avicennia germinans*). Notably, *Rhizophora mangle* can thrive in freshwater environments in Bermuda as well as being the dominant species in saline mangroves (e.g., Thomas, 1993). Therefore, it is important to test if fresh and saline environments vegetated by the same species can be distinguished from one another to ensure the accuracy of any resulting paleoenvironmental and RSL reconstructions. We also collected three samples of *Sargassum* wrack that were qualitatively judged to represent fresh, decaying, and decayed examples. All samples were collected in October 2011, sealed into bags and refrigerated for transportation to minimize desiccation, contamination, and degradation. On return to the laboratory, all samples were immediately freeze dried and stored in cool, dark conditions until analysis. All geochemical measurements were completed within ~18 months of sample collection.

Sediment beneath Hungry Bay was described from cores collected in October and November 2011 using a Russian-type peat corer (to prevent compaction and/or contamination) along transects that extended through the shallow bay and span the seaward depositional basin (**Figure 1**). Types of sediment in cores were recognized through comparison (color, texture, composition) to surface sediment in modern environments in Bermuda and our collective experience of coring and describing Holocene sediment in freshwater wetlands, salt marshes, and mangroves in Bermuda and elsewhere. Core HB-503 was selected for analysis because it is located close (~40 m) to core HB-1 of Ellison (1992, 1993, 1996), which showed a paleoenvironmental change from freshwater marsh to mangrove that is typical of the seaward depositional basin. This sedimentary sequence was correlated along multiple transects and cores HB-503 and HB-1 are representative of the stratigraphy underlying the seaward basin.

The elevation of all cores and samples in Hungry Bay was determined by deploying an automated water-level logger that measured water height at five-minute intervals for two weeks spanning the transition from spring to neap tides. The elevation of samples relative to the logger was measured using a total station. Measured water levels were correlated with the Esso Pier tide gauge to establish local tidal datums in Hungry Bay. A real-time kinematic satellite navigation survey linked the elevation of tidal benchmark “4” at Esso Pier to the water logger and samples in Hungry Bay. The sample elevations reported in this study (relative to mean tide level for the 1983–2001 tidal epoch; **Figure 1C–E**) differ systematically from those of Ellison (1992, 1993, 1996), who used Ordnance Datum (mean sea level in 1964).

4.2. Radiocarbon dating

We radiocarbon dated two sections of HB-503 using two different approaches. Across the freshwater to mangrove transition there were no identifiable plant macrofossils to reliably date past depositional surfaces. Therefore, we prepared samples for radiocarbon dating by sieving 1-cm thick sediment slices under running water at 500 μm to remove contaminating younger rootlets. All material <500 μm was retained in a sieve pan, extracted under a vacuum, placed in a borosilicate vial, and dried at 45°C. Material >500 μm was discarded after inspection. This approach is more likely to reliably estimate the age of depositional surfaces in mangroves than radiocarbon dating of unsieved bulk sediment (Woodroffe et al., 2015). At shallower depths in the mangrove peat, reliable surface markers were present (e.g., leaves lying horizontally) and these samples were prepared by cleaning them individually under a binocular microscope to remove adhered sediment and/or ingrowing rootlets and drying at 45°C (e.g., Kemp, Nelson & Horton, 2013). All samples were dated at the National Ocean Sciences Accelerator Mass Spectrometry laboratory following standard acid-base-acid pretreatment and conversion to graphite. Measured $\delta^{13}C$ values for radiocarbon dates are from an aliquot of combusted CO_2 and were used to correct for natural fractionation of carbon isotopes. Reported radiocarbon ages and uncertainties (**Table 1**) were calibrated (2σ range) individually using the IntCal13 or Marine13 dataset (Reimer et al., 2013) as appropriate for the type of sample analyzed (terrestrial or marine respectively). When using Marine13, we applied a correction (ΔR) for the region-specific difference between marine and atmospheric ages (section 6.2).

Table 1: Radiocarbon dates from core HB-503. Radiocarbon ages reported by the National Ocean Sciences Accelerator Mass Spectrometry (laboratory code OS) facility for core HB-503 in Hungry Bay, Bermuda. Ages and errors following the rounding conventions of Stuiver & Polach (1977). Sample $\delta^{13}C$ values are relative to the Vienna Pee Dee Belemnite (V-PDB) standard and were measured on an aliquot of CO_2 collected during sample combustion.

Depth (m)	ID	^{14}C Age	^{14}C Age Error	$\delta^{13}C$ (‰, V-PDB)	Dated Material
0.76	OS-96382	890	25	–28.89	Bark
0.76	OS-94842	935	25	–25.36	Wood
0.91	OS-96381	910	20	–27.22	Bark
1.13	OS-94841	1180	25	–24.05	Wood
1.26	OS-94830	1150	25	–26.75	Leaf
1.42	OS-94856	1230	30	–24.26	Leaf
1.60	OS-94838	1210	25	–24.32	Leaf
1.67	OS-103779	1330	25	–29.03	Bulk sediment (<500 μm)
1.70	OS-95330	1210	30	–26.16	Bulk sediment (<500 μm)
1.74*	OS-95398	2070	30	–20.21	Bulk sediment (<500 μm)
1.81*	OS-95329	2040	30	–19.40	Bulk sediment (<500 μm)
1.97*	OS-103780	2700	30	–20.31	Bulk sediment (<500 μm)
1.99*	OS-95399	2810	30	–20.27	Bulk sediment (<500 μm)
2.02	OS-95400	2410	25	–25.90	Bulk sediment (<500 μm)
2.06	OS-103781	1030	25	–27.20	Bulk sediment (<500 μm)

* Denotes samples calibrated using the Marine13 dataset.

4.3. Measurement of TOC, δ^{13}C, and C:N

Since marine, intertidal, and freshwater coastal environments receive, to varying degrees, different types of organic material they can often be distinguished from one another using measurements of TOC, δ^{13}C (relative to the Vienna Pee Dee Belemnite standard; V-PDB), and total nitrogen (from which C:N is calculated; e.g., Lamb et al., 2006). Samples were treated with 1 M HCl, left overnight, washed with deionized water, and oven dried at 60°C. This step removed carbonate from samples, which is particularly important in Bermuda as ongoing karstification of eolianite liberates carbonate that could be delivered to wetland sediment. Analysis was performed with a Europa Scientific Elemental Analyzer. The reference material used for δ^{13}C analysis was A-R001 (δ^{13}C = –26.43‰). For quality control purposes check samples of IA-R001, IA-R005 (δ^{13}C = –26.03‰), and IA-R006 (δ^{13}C = –11.64‰) were also analyzed during the batch analysis of the samples. IA-R001, IA-R005, and IA-R006 were calibrated against, and traceable to, IAEA-CH-6 (δ^{13}C = –10.43‰), an interlaboratory comparison standard distributed by the International Atomic Energy Agency. Replicate analysis of samples indicated an average standard deviation of 0.08‰. Total nitrogen of the original samples was analyzed using the above elemental analyzer. The reference material used during nitrogen analysis was IA-R001 (total nitrogen = 1.88%). All results are presented as a range and/or mean since standard deviations calculated across a small number of samples can be misleading.

4.4. Rock-Eval pyrolysis

Pyrolysis was performed on 60 mg of dry, powdered sediment using a Rock-Eval(6) analyzer (Vinci Technologies) in standard configuration (pyrolysis and oxidation as a serial process). Samples were placed in the pyrolysis oven and heated from 300°C (hold three minutes) to 650°C (hold three minutes) at 25°C/minute in an inert atmosphere of N_2. After transfer to the combustion oven, the remaining carbon was oxidized at 300–850°C at 20°C/minute (hold five minutes). Hydrocarbons released during this two-stage process were measured by a flame ionization detector. CO and CO_2 released during thermal cracking of bound organic matter was monitored using an infrared cell. The performance of the instrument was checked every ten samples against the accepted values of an Institut Français du Pétrole standard (IFP 160 000, S/N1 5-081840). All results are presented as a range and/or mean since standard deviations calculated across a small number of samples can be misleading.

5. Characterization and differentiation of modern depositional environments

5.1. TOC, δ^{13}C, and C:N

The TOC content of *Sargassum* (25.2–29.9%) was similar to values measured on mangrove sediment (23.7–27.9%). In contrast, freshwater environments are characterized by higher TOC values (42.2–45.4%), except for one sample from an environment vegetated by *Cladium jamaicae* that had a TOC content of 30.3% (**Figure 2A**). Measured δ^{13}C values from *Sargassum* ranged from –16.6‰ (sample identified as fresh) to –18.4‰ (sample identified as decayed), which indicates that ^{12}C is preferentially removed from *Sargassum* during initial decay (**Figure 2B**). Freshwater sediment yielded δ^{13}C values of –26.9‰ to –29.7‰ and bulk-sediment from saline mangroves had δ^{13}C values of –24.3‰ to –26.0‰.

Macroalgae tissue displays a wide range δ^{13}C composition that varies by species across a range of at least –4.9‰ to –30.9‰ (Chmura & Aharon, 1995). The δ^{13}C composition of *Sargassum* from Hungry Bay is similar to reported values elsewhere. For example, in Japan *Sargassum piluliferum* had an average δ^{13}C of –15.9‰ (Matsuo et al., 2010). Similarly, in the tropical Atlantic Ocean, Corbisier et al. (2006) reported values for living *Sargassum filipendula* of –15.1‰. In coastal waters near Hong Kong, Wai et al. (2008) measured mean δ^{13}C values on living *Sargassum* of –15.4 to –18.4‰. The δ^{13}C values from mangroves in Hungry Bay are similar to those reported from bulk-surface sediment elsewhere despite differences in mangrove flora, its diversity among regions, and bedrock type. For example, Tue et al. (2011) showed that mangrove sediment in Vietnam was characterized by δ^{13}C values of –22.5‰ to –28.2‰; in Indian mangroves δ^{13}C values ranged from –20.6‰ to –29.4‰ (Bouillon et al., 2003; Manju et al., 2016); and surface sediment from Brazilian mangroves had δ^{13}C values of approximately –26‰ (Sanders et al., 2010). Bulk-sediment δ^{13}C values from mangroves and freshwater environments both reflect the dominance of plants using the C_3 photosynthetic pathway coupled with sedimentation that occurs primarily through *in-situ* peat formation with limited allochthonous input (e.g., Chmura & Aharon, 1995; Khan, Vane & Horton, 2015; Lamb et al., 2006). The range of surface bulk-sediment δ^{13}C values from freshwater and mangrove environments in Bermuda do not overlap with one another (**Figure 2B**), although we caution that this could reflect the relatively small number of samples that we analyzed and which may not fully capture within-group variability (section 5.4). δ^{13}C values in *Sargassum* are clearly distinctive from those in freshwater and mangrove peat, which suggests that sediment enriched with macroalgae is likely to be identifiable in the stratigraphic record.

Of the three sample groups, *Sargassum* had the highest and most variable C:N values (from 28.5 for the fresh sample to 43.0 for the decaying sample; **Figure 2C**) and these values are comparable to those reported for living *Sargassum* from Hawaii and Australia (18–52; Atkinson & Smith, 1983). Freshwater environments had intermediate C:N values (17.7–24.9) and saline mangroves had the lowest C:N values (11.5–15.3; **Figure 2C**). Across these three parameters, sediment from a freshwater *Rhizophora mangle* community at Paget Marsh is more similar to freshwater environments dominated by other species (e.g., *Myrica cerifera*) than it is to sediment deposited in a saline mangrove where *Rhizophora mangle* is also dominant. We note that the difference between *Sargassum* tissue and bulk sediment at least in part reflects the early stages of diagenesis and mixing that characterize the formation of surface sediment.

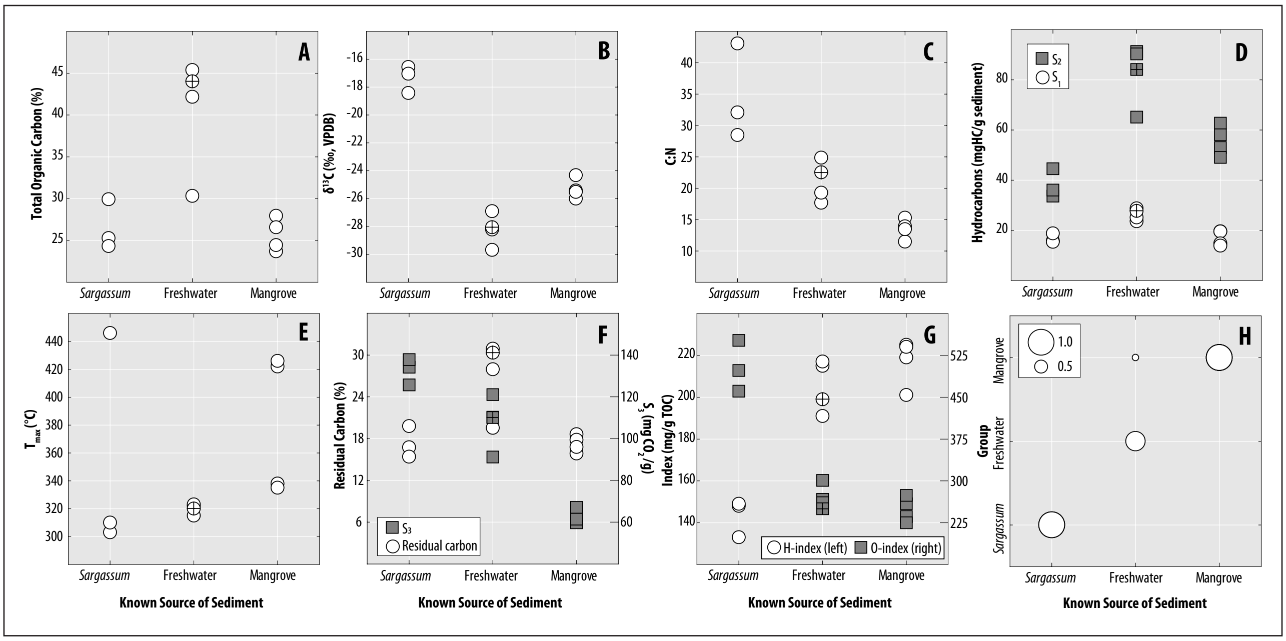

Figure 2: Rock-Eval pyrolysis and stable carbon isotope measurements made on bulk surface (0–1 cm) sediment from depositional and floral environments in Bermuda. Samples of mangrove sediment and *Sargassum* are from Hungry Bay. Samples of freshwater sediment are from Paget Marsh and include environments vegetated by *Cladium jamaicae*, cedar swamp, *Myrica cerifera* and *Rhizophora mangle*. Measurements from the freshwater, *Rhizophora mangle*-dominated environment are differentiated by symbols filled with crosses to enable comparison with saline mangroves. Note that hydrogen (H) and oxygen (O) indexes are shown in a panel (G) with two y-axes to accommodate differences in scale. In the panel (H) showing groups identified using partitioning around medoids, symbol size represents the proportion of samples assigned to each group. Absence of symbols indicates that no samples were assigned to a group. The single outlier was a sediment sample from a freshwater environment vegetated by *Cladium jamaicae* that was grouped with samples from saline mangroves.

5.2. Rock-Eval pyrolysis

Each of the primary and secondary parameters measured using Rock-Eval pyrolysis are described in section 3. *Sargassum* (15.4–18.8 mg HC/g) and mangrove sediment (13.9–19.6 mg HC/g) yielded relatively low concentrations of free hydrocarbons (S_1) compared to freshwater sediment (23.6–28.7 mg HC/g; **Figure 2D**). All environments yielded larger quantities of bound hydrocarbons (S_2) than free hydrocarbons. The highest S_2 concentrations came from freshwater samples (65–91 mg HC/g) and the lowest concentrations came from *Sargassum* (34–45 mg HC/g). T_{max} in the fresh and decayed *Sargassum* samples was relatively low (303°C and 310°C respectively), while the sample identified as decaying had a T_{max} of 446°C (**Figure 2E**). Although T_{max} values do not correspond to the qualitative and sequential descriptions of decay state of *Sargassum*, they may indicate that labile carbon was preferentially lost during the initial (pre-burial) stages of decay because the loss of labile carbon concentrates more stable carbon that is only liberated at higher temperatures during the heating cycle. Freshwater sediment was characterized by uniformly low T_{max} (315–322°C), while mangrove samples were split between two samples with high T_{max} (422 and 426°C where only *Rhizophora mangle* was present) and two samples with low T_{max} (335 and 338°C, where *Rhizophora mangle* and *Avicennia germinans* were present). Measured S_3 was highest in *Sargassum* (126–138 mg CO_2/g), intermediate in freshwater sediment (91–121 mg CO_2/g) and lowest in mangrove sediment (60–67 mg CO_2/g). Residual carbon in *Sargassum* was 17–20%, compared to 20–31% in freshwater sediment and 16–19% in mangrove sediment (**Figure 2F**). Samples of *Sargassum* had the lowest hydrogen index (133–149 mg/g TOC) and highest oxygen index (461–552 mg/g TOC; **Figure 2G**). There is considerable overlap between freshwater and mangrove peat when these environments are compared using hydrogen (191–217 mg/g TOC for freshwater sediment and 201–225 mg/g TOC for mangrove sediment) and oxygen indexes (250–301 mg/g TOC for freshwater sediment and 225–274 mg/g TOC for mangrove sediment). Rock-Eval pyrolysis measurements (**Figure 2D–G**) on freshwater sediment vegetated by *Rhizophora mangle* are more similar to other freshwater samples than they are to saline mangrove sediment where the dominant vegetation is also *Rhizophora mangle* (as noted for TOC, $\delta^{13}C$ and C:N values; section 5.1).

The similarity in Rock-Eval pyrolysis parameters (particularly hydrogen and oxygen indexes) between freshwater and mangrove sediment arises in Bermuda because mangroves are dominated by vascular plants that provide large quantities of *in-situ* organic carbon compared to the input of marine organic carbon. A similar pattern was observed in Guadeloupe where Rock-Eval pyrolysis could not distinguish between sediment from adjacent mangrove and freshwater environments (Lallier-Verges et al., 1998) and in sub-arctic Russia, where adjacent salt marshes and freshwater wetlands were not distinguishable (Kemp et al., 2017). We conclude therefore, that Rock-Eval pyrolysis alone is unlikely to differentiate between sediment deposited in mangrove and freshwater environments, but is likely to objectively recognize sediment that includes significant inputs from marine macroalgae.

5.3. Identifying groups of modern samples

The potential utility of a paleoenvironmental proxy can be tested by objectively placing samples of modern sediment into groups based on their measured geochemical characteristics. If group membership corresponds to the known depositional environment, then the measured parameters are potentially useful for reconstructing paleoenvironments and RSL. To objectively group samples that share common geochemical characteristics, we used partitioning around medoids (Kaufman & Rousseeuw, 2005; Rousseeuw, 1987). This technique calculates a silhouette width for each sample, which is a measure of how well it is classified (Rousseeuw, 1987). Positive values (maximum = 1) indicate that within-group variance is less than among-group variance and that a sample is well classified. Conversely, negative values (minimum = –1) are indicative of a poorly-classified sample. An average silhouette width is calculated across all samples within each group as a convenient means to evaluate how distinctive or similar they are to one another. Although there is no agreed-upon threshold for average silhouette width, values greater than ~0.3 are often considered as evidence that a group is distinctive.

The input for partitioning around medoids was measured values of $\delta^{13}C$, C:N, TOC, and the Rock-Eval pyrolysis primary parameters (S_1, S_2, S_3, residual carbon, and T_{max}). Exclusion of secondary (derived from others) parameters such as hydrogen index and oxygen index prevented bias. Prior to analysis, we standardized each variable to have mean = 0 and standard deviation = 1 because of differences in the units and magnitudes of measurements among variables. Based on *a priori* grouping of samples (freshwater sediment, mangrove sediment, and *Sargassum*), we specified that partitioning around medoids should place the modern samples into three groups. With the sole exception of the sample from a *Cladium jamaicae*-dominated freshwater wetland that was grouped with the saline mangrove samples, all samples were objectively placed into their *a priori* groups (**Figure 2H**). The average silhouette width calculated across all samples was 0.50 (0.08–0.81 for individual samples), compared to 0.30, 0.78, and 0.46 for the groups that exclusively contained *Sargassum* samples, freshwater samples, and mangrove sediment plus the sample of *Cladium jamaicae*, respectively. Therefore, a multi-proxy approach combining measurements of $\delta^{13}C$, C:N, TOC, and Rock-Eval pyrolysis can often distinguish between modern sediment that accumulated in freshwater and mangrove environments and could have utility in reconstructing RSL.

5.4. Influence of sample size

Our analysis is based on a small number of modern samples and comprises a single sample for each of four different sub-environments (as defined by the dominant vegetation) where freshwater peat is forming at Paget Marsh, four samples of mangrove sediment from Hungry Bay (two locations vegetated solely by *Rhizophora mangle* and two sites where *Avicennia germinans* and *Rhizophora mangle* were both present), and three samples of recently-deposited *Sargassum* in different stages of early decay (**Figure 2**). These samples were collected to systematically provide modern analogues for sediment in the stratigraphic record. At the time of sample collection each sample was judged to be representative of the environment from which they were collected and there was no observable evidence of disturbance. The measured values are similar to those reported in the literature (as discussed previously), which further suggests that despite the small sample size they are not outliers. Although analysis of more samples is desirable to accurately quantify variance and complexity, there are only a handful of small freshwater marshes and saline mangroves in Bermuda, which limits the geographic variability that is likely to exist among and within sites. The low-diversity nature of Bermuda's mangroves also limits variability in bulk sediment geochemistry and our sampling regime included the major vegetation and elevation zones within the Hungry Bay mangrove. Therefore, our conclusion that Rock-Eval pyrolysis alone cannot distinguish between freshwater and mangrove sediment in the sedimentary record is unlikely to be changed by analyzing more modern samples that would likely increase within-group variability. In contrast, we recognize that incorporating measurements from additional surface sediment samples may ultimately demonstrate that modern freshwater wetlands and saline mangroves in Bermuda cannot be reliably differentiated even with a multi-proxy approach as increased within-group variance could blur the geochemical boundaries between these two environments.

6. Characterization and differentiation of paleoenvironments

6.1. Sub-surface stratigraphy in Hungry Bay

At sub-tidal locations in the seaward depositional basin (**Figure 1**), surface sediment is comprised of a thin (<0.3 m) and dynamic veneer of coarse sand and shell hash that lies directly on top of eolianite bedrock at locations outside of a depositional basin (**Figure 1B–D**). A thicker sequence of sediment is present within the depositional basin that broadly includes three distinctive units. The lowermost unit (up to 3.5 m thick) is an amorphous, black peat with abundant fine roots that is visually similar in composition and texture to surface sediment samples from Paget Marsh. Pollen analysis confirms that this unit was deposited in a freshwater environment (Ellison, 1996).

Overlying the freshwater peat in some locations is a distinctive unit of orange-colored algal gel (up to ~0.3 m thick), which lacked structure and intact/identifiable plant material. It had a high moisture content and greasy texture. We interpreted the algal gel as possibly being derived from *in-situ* decay of *Sargassum* because it is abundant in the Atlantic Ocean around Bermuda, orange-brown in color, lacks woody material, and is frequently carried into Hungry Bay by tides and storms. The lower part of this unit (below ~1.88 m) was darker and slightly more structured than the upper part (above ~1.88 m). The contact between freshwater peat and algal gel is sharp, indicating a rapid environmental change and/or a hiatus in accumulation caused by erosion and/or a period of non-deposition. The algal gel is found only at locations close to the front edge of the modern mangrove, which our leveling survey indicated is the landward edge of the depositional basin. This unit was not identified in the stratigraphic investigation of Ellison (1992, 1993, 1996) because the cores did not intersect with the algal gel due to its limited extent.

Overlying the algal gel (or, in its absence, the freshwater peat) is mangrove peat (up to ~1.75 m thick) recognized by its distinctive red-brown color, fibrous texture, and identifiable plant remains including pieces of prop roots, bark, leaves, and fine roots. These characteristics are shared by modern sediment deposited in the Hungry Bay mangrove where *Rhizophora mangle* is dominant. The distribution of this unit indicates that mangroves once occupied the entire depositional basin (Ellison, 1993; **Figure 1B**) and pollen analysis confirms that it was deposited in a saline mangrove. *Rhizophora mangle* lives in some freshwater environments in Bermuda and *Rhizophora* spp. produces abundant and widely-distributed pollen (Engelhart et al., 2007; Muller, 1959), therefore the presence of *Rhizophora* spp. pollen does not conclusively mean that Hungry Bay was saline. In contrast, *Avicennia germinans* is an obligate halophyte (Nguyen et al., 2015) and produces relatively little pollen that is not widely distributed (Grindrod, 1985; Mao, Zhang & Bi, 2006; van Campo & Bengo, 2004). Therefore, the presence of *Avicennia germinans* pollen (Ellison, 1993) is evidence that Hungry Bay was occupied by a saline mangrove community, but is not definitive evidence that the bay had a surface connection to the ocean because *Avicennia germinans* is present in some modern Bermudan saline ponds (Thomas et al., 1992). However, the presence of marine sponge spicules in the mangrove peat quantified by Ellison (1993) indicates regular and persistent inundation by seawater.

6.2. Radiocarbon dating the freshwater to mangrove transition

A radiocarbon date at the top of the freshwater peat unit (2.02 m; **Table 1**) yielded a calibrated (IntCal13) age of –729 CE to –403 CE (**Figure 3**). Radiocarbon dates from the algal gel unit were calibrated using the IntCal13 and Marine13 datasets (Reimer et al., 2013). Due to the apparent age of carbon in the shallow ocean, the Marine13 dataset returns a younger age than calibration of the same samples using IntCal13. Since the algal gel was likely derived from *Sargassum*, we considered two different marine reservoir corrections. The first correction ($\Delta R_{Bermuda}$) of –129 ± 29 years is the sole value reported for Bermuda in the Marine Reservoir database of Reimer et al. (2013). It was derived from radiocarbon measurements made on a *Diploria strigosa* coral colony at ~5-m depth near North Rock, Bermuda (Druffel, 1997; ~16 km north of Esso Pier, **Figure 1A**). However, it may not be appropriate to use

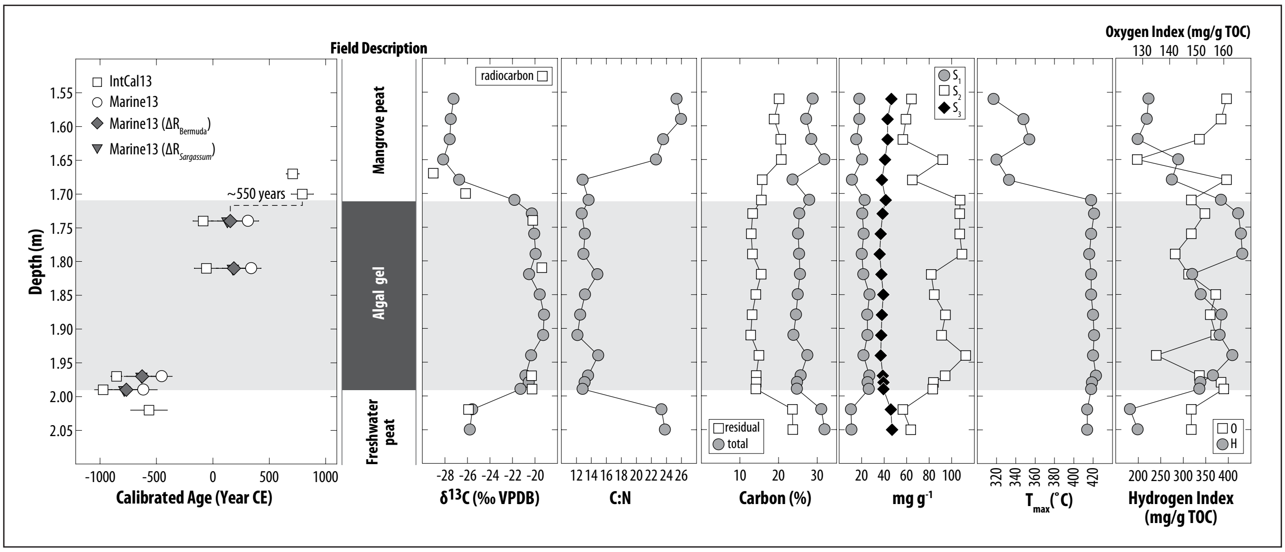

Figure 3: Radiocarbon ages, stable carbon isotope measurements, and Rock-Eval pyrolysis measurements made on 1-cm thick samples of bulk sediment from core HB-503. For calibrated radiocarbon ages (median and 2σ range) symbol shape and shading denotes the calibration curve and ΔR (where applicable) used. Each radiocarbon date from the unit interpreted as decayed *Sargassum* was calibrated four times because as a photosynthesizing macroalgae living in surface water in the western North Atlantic Ocean it may uptake carbon from regional ocean water and/or the atmosphere. The schematic sediment column shows the depths of units described in the field at the time of collection. The grey-shaded interval on other panels shows the depths over which the algal gel layer (interpreted as decayed *Sargassum*) was found and is presented solely for ease of correlating measured values to stratigraphic position.

$\Delta R_{Bermuda}$ for calibrating radiocarbon ages from *Sargassum* because it is a photosynthesizing macroalga. The second correction ($\Delta R_{Sargassum}$) was estimated from the radiocarbon age (320 years, no uncertainty provided) of a living *Sargassum thallus* (collected between Bermuda and the U.S. Atlantic coast at 36°24′ N, 69°37′ W) that was reported in Revelle & Suess (1957). The radiocarbon age of this sample was originally attributed to Suess (1954) and we assumed that it was physically collected in 1949 CE. Using the deltaR tool of Reimer & Reimer (2016) with an assumed uncertainty of ± 40 years, this yielded a value for $\Delta R_{Sargassum}$ of −148 ± 46 years (1σ). The similarity of $\Delta R_{Bermuda}$ and $\Delta R_{Sargassum}$ indicates that radiocarbon dates from the algal gel layer calibrated with either ΔR will return similar ages, and that $\Delta R_{Bermuda}$ is appropriate for *Sargassum* in the Atlantic Ocean around Bermuda.

Two dates (1.97 m and 1.99 m) in the lowermost part of the algal gel indicate that it began to accumulate at approximately −700 CE. The similarity of this age with that of the uppermost part of the freshwater peat suggests that the sharp stratigraphic contact represents a rapid environmental change rather than a hiatus. The uppermost part of the algal gel (1.74 m) was deposited at −13 to 269 CE. The similarity of this age to the one at 1.81 m (**Figure 3**) suggests that sedimentation was faster in the upper part of the algal gel than it was in the lower part. Radiocarbon dates at 1.70 m and 1.67 m show that accumulation of mangrove peat began at approximately 700–900 CE (IntCal13 calibration).

6.3. Geochemical analysis of HB-503

Bulk-sediment $\delta^{13}C$ values in the freshwater peat (2.05–1.99 m; **Figure 3**) were approximately −26‰ and became less depleted in the algal gel (−19.2‰ to −21.8‰; 1.98–1.71 m). The mangrove peat (1.68–1.56 m) had $\delta^{13}C$ values of −26.7‰ to −28.1‰. We note that additional $\delta^{13}C$ measurements from the bulk sediment used for radiocarbon dating are in close agreement with those that we measured. Comparison of these values with their presumed modern analogues indicates that $\delta^{13}C$ in bulk freshwater and mangrove sediment converge after incorporation into the stratigraphic record and are difficult to distinguish from one another. This interpretation includes a caveat that we analyzed a relatively small number of surface sediment samples and that an increase of within-group variance from expanding this dataset could render these environments indistinguishable in modern settings too, in which case post-depositional change would not be invoked as the reason for their similarity (section 5.4).

Due to preferential decay of cellulose over lignin, sediment $\delta^{13}C$ values usually become more negative through time (Benner et al., 1987) as observed when comparing modern mangrove sediment from Hungry Bay (**Figure 2B**) with mangrove peat in HB-503 (**Figure 3**). However, modern freshwater sediment in Bermuda yielded more negative $\delta^{13}C$ values (**Figure 2B**) than freshwater peat in HB-503 (**Figure 3**). This trend could arise from analyzing too few core and modern samples to adequately capture the full degree of variability (section 5.4) and/or a flux of material to the buried freshwater peat (at, or since, the time of its deposition) that was relatively rich in ^{13}C (e.g., marine algae). The algal gel was more depleted in ^{13}C (approximately −20‰) than *Sargassum* tissue (approximately −17‰; **Figure 2**), perhaps because decomposition concentrated ^{12}C and/or dilution of the gel with non-*Sargassum* material through physical mixing, or bioturbation moved carbon vertically in the stratigraphic column (e.g., downward penetration of mangrove roots). Despite these changes, the algal gel has a distinctive $\delta^{13}C$ signature that allows it to be easily differentiated from the sediment above and below. The $\delta^{13}C$ values we measured in the algal gel are comparable to those reported for organic marine gel (−21.6‰ to −19.5‰) in a sediment core from Mangrove Lake (Hatcher et al., 1982).

C:N values in the freshwater and mangrove sediment (22–26) are markedly higher than in the algal gel (12–16). The lack of similarity between C:N values from modern *Sargassum* tissue (**Figure 2C**) and the algal gel (**Figure 3**) indicates that decay of *Sargassum* involves preferential removal of carbon over nitrogen (e.g., Rice & Tenore, 1981), enrichment in nitrogen through microbial action (e.g., Sharma et al., 1994), and/or a sediment source other than, or in addition to, *Sargassum*. Although, similar trends occur soon after deposition of plant tissue in freshwater marshes and saline mangroves, our modern samples from these environments were comprised of time-averaged surface sediment that adequately captures the initial stages of decomposition (e.g., Fell & Master, 1980; Rice & Tenore, 1981). Although freshwater and mangrove sediment cannot be distinguished from one another using C:N alone, the algal gel unit is readily identified. Furthermore, C:N values in the algal gel are comparable to those reported for organic marine gel (11–12) in Mangrove Lake (Hatcher et al., 1982).

Residual carbon (13–24%) and TOC (24–32%) showed little variability in HB-503, although measured values in the algal gel were modestly lower than in the underlying freshwater peat and overlying mangrove peat (**Figure 3**). The concentration of bound hydrocarbons (S_2) was elevated in the algal gel (mean 96 mg HC/g) compared to the freshwater (mean 60 mg HC/g) and mangrove (mean 67 mg HC/g) peats. T_{max} was consistently high (mean 418°C) below 1.70 m and decreased markedly in the mangrove peat (mean 334°C). Due to little variability among sedimentary units in S_3 and TOC, there was no clear downcore trend in oxygen index. In contrast, the hydrogen index was higher in the algal gel (320–431 mg/g TOC) than in either the freshwater peat (182–199 mg/g TOC), or the mangrove peat (219–289 mg/g TOC). This trend mirrors the concentration of bound hydrocarbons (S_2) because TOC values showed little variability. Typically, marine macroalgae yield higher hydrogen index values than terrestrial plants because they are richer in lipids and proteins resulting in a higher H:C ratio. The labile nature of carbon in marine algae results in elevated concentrations of S_2 hydrocarbons. The increase in S_2 and hydrogen index between 1.99 m and 1.71 m in HB-503 (**Figure 3**) supports our interpretation that deposition of terrestrial, freshwater peat was ended by deposition of a significant input of macroalgae such as *Sargassum*. When mangroves colonized Hungry Bay, S_2 concentrations and hydrogen index values decreased because sedimentation returned to being dominated by terrestrial peat. These results indicate that the best parameter (of those we measured) for identifying buried sediment which received significant input from macroalgae is elevated hydrogen index values. The increase in hydrogen index from relatively lower values in the freshwater peat to relatively higher values in the algal gel occurs across 0.02 m, indicating an abrupt shift in depositional environment. In contrast, the decrease in hydrogen index from the algal gel to the overlying mangrove peat spans ~0.1 m. The more diffuse nature of this change could represent a gradual environmental transition (for example *Sargassum* being washed into the recently-established mangrove) and/or mixing of material between the two units.

Bulk surface sediment entering the stratigraphic record undergoes post-depositional change on decadal to multi-century timescales during which the less-polymerized (more labile) fraction of S_2 hydrocarbons is systematically removed, resulting in more resistant (less labile) material becoming increasingly concentrated in the sediment. Assuming a constant parent material, this trend would cause T_{max} to increase with depth/age unless burial rates varied through time. More rapid burial allows labile carbon to remain in the sedimentary record, while slower burial leaves sediment exposed to diagenesis (on or near the surface) for longer periods of time, which promotes removal of labile carbon. For example, modern freshwater peat from Paget Marsh (**Figure 2E**) yielded systematically and markedly lower T_{max} than its counterparts in HB-503. In contrast, mangrove peat in HB-503 (1.70–1.55 m depth) that is more than 1000 years old yielded lower T_{max} (310–340°C; **Figure 3**) values than two of the four samples of surface mangrove sediment (**Figure 2E**). This could indicate rapid burial, or alternatively a change in parent material because T_{max} of modern (surface) organic carbon in modern mangroves may have a bimodal distribution even over small distances. In the specific case of Bermudan mangroves, lower T_{max} in modern sediments occurred where *Avicennia germinans* was present, which possibly indicates that this species is a source of particularly labile carbon in peat formation, although expansion of the modern dataset to include more surface sediment samples from mangrove sub-environments is needed to fully evaluate this suggestion. Rock-Eval pyrolysis of mangrove sediment in French Guiana showed high T_{max} (430–450°C) at the surface and a step-wise decrease to lower T_{max} (340–360°C) at depth (Marchand et al., 2008), which indicates that the deeper (and presumably older) mangrove sediment is more labile than its modern counterparts, perhaps because rapid burial allowed the sediment to enter anoxic conditions and halt decomposition sooner than is currently the case. In contrast, downcore T_{max} measurements from *Rhizophora*- and *Avicennia*-dominated mangroves in Senegal are uniformly high (400–420°C) and show no trend over the 0–0.4 m depth interval (Sakho et al., 2015; it is unclear how much time this interval represents). The high and uniform T_{max} values suggests that mobilization of labile carbon occurred more quickly than burial. Notably, the input of litter from *Avicennia* in Senegal was not associated with low T_{max}, suggesting that the pattern observed in Bermuda may be associated with small-scale variability rather than a systematic trend caused by the species of litter. However, the mangroves in these examples are young and occupy dynamic coastlines, which results in low TOC (<5%) and makes them poor analogues for Hungry Bay, where protected mangroves accumulate sediment that (in HB-503 at least) has TOC of ~30%. In Panama, Upton et al. (2018) showed that T_{max} in a core of mangrove sediment was strongly bimodal with bulk sediment returning values of either ~300°C or ~400°C. Higher values were associated with units of peat that included shells that may indicate a dynamic environment where sediment is (re)exposed and labile carbon is readily removed. However, the near-binary nature of T_{max} in this core and in modern sediment from Hungry Bay warrants further investigation. While the measured Rock-Eval parameters were unable to distinguish between freshwater and mangrove sediment, they do provide valuable insight into the relative rates of diagenesis and burial. Furthermore, they can estimate the amount of CO_2 that could be emitted from re-exposure of buried sediment to the atmosphere, which is higher/lower for buried peat that is rich/depleted in labile carbon (as estimated by low/high T_{max}). Potential CO_2 emissions can therefore vary among depositional environments (higher for freshwater peat than for mangrove peat in Bermuda for example) and as a function of burial rate (Upton et al., 2018).

6.4. Identifying groups of samples in HB-503

We used partitioning around medoids to test if samples in HB-503 formed groups based on their geochemical characteristics that corresponded to their known environmental origin. This analysis followed the approach described for modern samples (section 5.3) and we again specified *a priori* that three groups were present because of the observed stratigraphic transitions from freshwater peat to algal gel to mangrove peat. Samples were allowed to form groups freely with no constraint imposed by stratigraphic order or relationships. The average silhouette width was 0.39 (–0.05 to 0.59 across individual samples). All freshwater samples and four of the five samples of mangrove peat were placed into the same group (**Figure 4**) with an average silhouette width of 0.38. The second group included the remaining sample of mangrove peat and seven of the twelve algal gel samples (average silhouette width of 0.22) and specifically, it included (with one exception) the samples from the lower part of the algal gel (1.99–1.82 m). The third group was comprised exclusively of samples from the upper part (1.79–1.71 m) of the algal gel (average silhouette width of 0.37). These results demonstrate that freshwater and mangrove peat beneath Hungry Bay are difficult to distinguish from one another using the geochemical variables that we measured (even when using a multi-proxy approach), but that the algal gel unit can be sub-divided. This sub-division is primarily driven by S_2 values, which are higher in the upper section (average 109 mg/g) than in the lower section (average 88 mg/g; **Figure 2**). If all samples in the algal gel were origi-

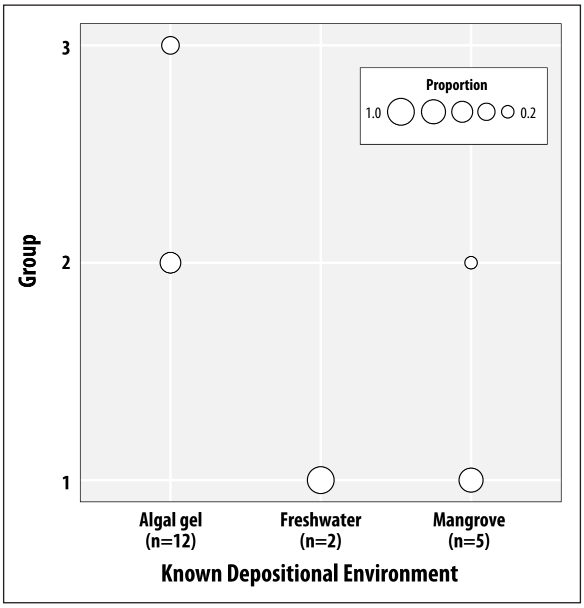

Figure 4: Results of partitioning around medoids of geochemical measurements made on sediment samples in core HB-503 that are known to represent three different depositional environments (X axis). Symbol size represents the proportion of samples from each of the three depositional environments that were assigned to each group. Absence of symbols indicates that no samples were assigned to a group.

nally composed of the same material, this difference probably indicates that the upper part of the unit accumulated more quickly (as tentatively suggested by sediment accumulation rates; **Figure 3**, section 6.2), resulting in faster burial and less decomposition of bound carbon than in the lower part of the unit.

6.5. Evolution of Hungry Bay

Like many of Bermuda's ponds and bays (e.g., Ashmore & Leatherman, 1984; Hatcher et al., 1982; Tackaberry et al., 2004), Hungry Bay was initially occupied by a freshwater community. Tackaberry et al. (2004) suggested that the accumulation of black, amorphous peat (which they termed *sapric*) reflects standing water in a marsh environment because the height of the groundwater table (controlled by RSL, since eolianite is porous) intersected with the elevation of the basin. From approximately –500 CE onward, algal gel accumulated at some locations in Hungry Bay (**Figure 1**). In Mangrove Lake, Hatcher et al. (1982) showed that a freshwater marsh was succeeded abruptly (as shown by a sharp stratigraphic contact) by a lake in which anoxic bottom waters allowed up to 12 m of organic gel to accumulate first from freshwater algae and then from marine algae. The switch from freshwater to salt water (and hence from freshwater to marine algae) in Mangrove Lake occurred through a sub-surface connection to the ocean. The organic gel in Mangrove Lake was red, brown, or green, displayed banding and laminations at some depths, and is present throughout the entire (albeit small) depositional basin. Its geochemical composition (Hatcher et al., 1982) shows some similarities with the algal gel identified in Hungry Bay such as high TOC (40–50%), relatively low C:N (11–17), and δ^{13}C of approximately –20‰. This could indicate that Hungry Bay was occupied by a (fresh or saline) lake with anoxic bottom waters. However, the algal gel in Hungry Bay is a different color, lacking in structure, thin (<0.4 m) and restricted to a narrow band along the landward edge of the depositional basin. This pattern is more consistent with delivery of *Sargassum* during episodic marine incursions than with the presence of a lake. Radiocarbon dates (**Figure 3**), geochemical measurements (in particular S_2 concentration), and grouping of samples in HB-503 (**Figure 4**) suggest that the upper part of this unit accumulated more quickly (and therefore experienced less decomposition) than the lower part and that it was the consequence of more than a single storm tide entering Hungry Bay. We propose two mechanisms that could explain this pattern. Firstly, the frequency of marine incursions (assuming a stationary storm climate) is controlled by the vertical relationship between the sill at the entrance to Hungry Bay and RSL. As RSL rose, more regular delivery of *Sargassum* into Hungry Bay likely occurred on smaller, and therefore, more frequent storm tides. This pattern is consistent with *Sargassum* being delivered at faster rates as RSL rose, making it easier for storm tides to enter Hungry Bay. Secondly, the amount *Sargassum* being delivered to coastal environments is observed to vary markedly through time as a product of ocean conditions including source region (e.g., Brooks et al., 2018; Franks, Johnson & Dong, 2016). For example, a sufficient quantity of *Sargassum* was delivered to sites in the Caribbean during a so-called "golden tide" in June 2011 to cover some beaches with as much as 1 m of macroalgae (Schell, Goodwin & Siuda, 2015) and this event was mirrored several months later in Bermuda. Similar events may explain the presence on the algal gel unit in the stratigraphic record below Hungry Bay.

By approximately 700 CE a saline mangrove community was present in Hungry Bay and sufficiently mature to accumulate peat. This could have been achieved by regular submergence of the sill by high tides, or by salt water water entering an inland pond through the porous eolianite. However, the increasing presence of marine sponge spicules (Ellison, 1996) suggests that there was likely some surface connection with the ocean. Our radiocarbon dates indicate that approximately 550 years (within uncertainty of calibrated ages at 1.67 m, 1.70 m, and 1.74 m the difference could be approximately 400–900 years) elapsed between deposition of the top of the algal gel and the detectable presence of peat-forming mangroves. This lag could represent the time it took for viable mangrove propagules to enter the bay through the restricted inlet. The colonizing propagules were carried by prevailing currents from as far away as southern Florida because Hungry Bay is located on the southern coast of Bermuda (Ellison, 1996) and prevailing currents approach from the open Atlantic Ocean making it less likely that propagules would be transported from elsewhere in Bermuda. The absence of *Rhizophora mangle* pollen in the freshwater peat (Ellison, 1996) shows that there was not a mangrove forest (either saline or fresh) nearby because this species produces large quantities of widely-dispersed pollen (e.g., Muller, 1959). However, in core HB-1 Ellison (1996) reported the presence of mangrove pollen (*Rhizophora* mangle *and Avicennia* germinans) by approximately 0 CE. Therefore, the radiocarbon date at 1.70 m could represent the first formation of mangrove peat rather than colonization by mangrove trees.

We radiocarbon dated surface markers in the mangrove peat (**Table 1**; **Figure 5**). A Bchron age-depth model (Parnell et al., 2008) developed for 1.60–0.76 m estimates that the average rate of mean annual sediment accumulation was ~1.7 mm/yr over the period 650–1230 CE. In the absence of a river catchment, a high proportion of this sediment was likely the *in-situ* accumulation and decay of mangrove litter and any material carried over the sill and into Hungry Bay by tides and/or storm tides. A high rate of sedimentation is supported by the low T_{max} values measured in the lowest part of the mangrove peat (**Figure 3**; see discussion in section 6.3), which indicates that burial occurred more rapidly than removal of labile carbon by diagenesis. In comparison, the rate of RSL rise for Bermuda predicted by the ICE–6G_C VM5a Earth-ice model (Argus et al., 2014; Peltier et al., 2015) over this time period is ~0.7 mm/yr. Since 1995, a permanent global positioning satellite receiver at the Bermuda Institute for Ocean Sciences (**Figure 1A**) has measured subsidence of 0.46 ± 0.21 mm/yr, which likely approximates the vertical land motion component of ongoing glacio-isostatic adjustment. These predicted and measured rates are broadly consistent with RSL reconstructions for the late Holocene (Ashmore & Leatherman, 1984; Neumann, 1971) prior to the onset of historic rates of rise. These results suggest disequilibrium between the rate of mangrove peat accumulation in Hungry Bay and regional-scale RSL rise.

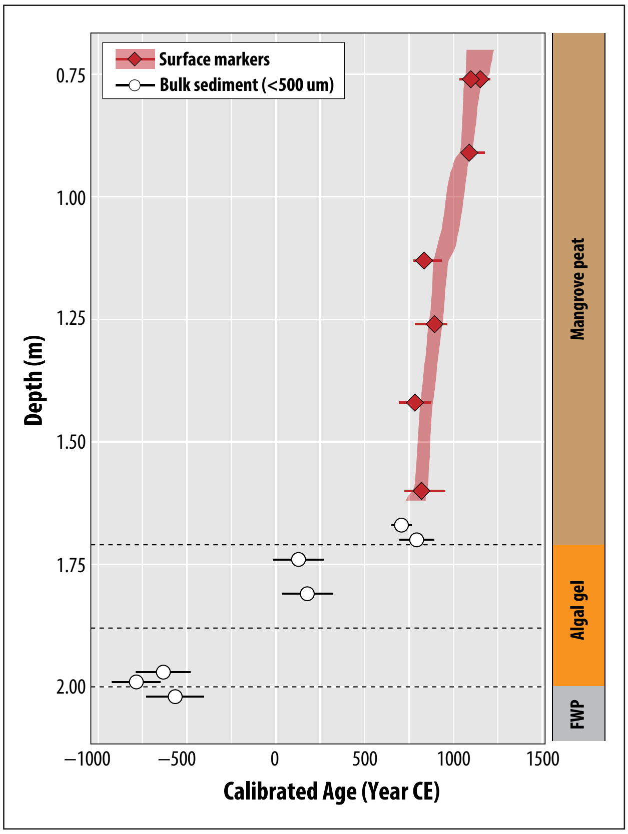

Figure 5: History of mangrove peat accumulation in Hungry Bay. The shaded envelope represents the 95% confidence of a Bchron age-model that used the radiocarbon dates on surface markers (filled symbols) as input. These dates were calibrated using the IntCal13 dataset as part of the Bchron analysis. Open symbols are the calibrated radiocarbon ages used to establish when the transition from freshwater peat to algal gel to mangrove peat occurred. Horizontal dashed lines represent the subdivision of the unit of algal gel that was noted at the time of sample collection. FWP = freshwater peat.

7. Conclusions

We investigated the efficacy of TOC, $\delta^{13}C$, C:N and Rock-Eval pyrolysis as proxies for coastal depositional environments and RSL in Bermuda. A multi-proxy approach indicates that surface sediment from freshwater wetlands, saline mangroves, and *Sargassum* wrack can likely be distinguished from one another using these geochemical proxies as evidenced by results from partitioning around medoids. However, the small number of samples in this modern dataset likely underestimates within and among group variability.

Examining only surface sediment overlooks the potentially significant effect of post-depositional diagenesis on the geochemical characteristics of buried sediment. Existing pollen analysis demonstrated that Hungry Bay was occupied by a freshwater wetland that was colonized by mangroves in response to RSL rise. Geochemical characterization of known freshwater and mangrove peats in the stratigraphic record beneath Hungry Bay indicates that these environments cannot be distinguished from one another using the geochemical techniques that we investigated, probably because post-depositional processes (e.g., preferential removal of more labile carbon) acting on timescales of decades to centuries caused convergence. In contrast, sediment that includes substantial contributions from marine macroalgae (e.g., *Sargassum*) is easily distinguished by its characteristic $\delta^{13}C$ signature and elevated hydrogen index in particular. Therefore, bulk-sediment geochemistry can yield paleoenvironmental insight at locations where this type of sediment is preserved in the coastal stratigraphic record. Rock-Eval pyrolysis can help to evaluate relative rates of diagenesis and burial in a stratigraphic unit, which is potentially valuable for drawing paleoenvironmental inferences.

We combined existing pollen data with additional transects of sediment cores, new radiocarbon dates, and geochemical measurements to add to understanding of the paleoenvironmental evolution of Hungry Bay in response to RSL rise. Of note is the spatially-limited presence of algal gel that likely formed from *Sargassum* being washed into Hungry Bay with increasing frequency by storms and high tides occurring on top of rising RSL. A variable rate of *Sargassum* supply from the open ocean may have also contributed to the formation of this unit. There is likely a disequilibrium between the accumulation rate of mangrove peat in Hungry Bay and regional-scale late Holocene RSL trends predicted by a widely-used Earth-ice model, global positioning satellite observations, and existing geological data.

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Competing Interests

Simon Engelhart is a guest editor for this special collection but was removed from all editorial duties for this submission. The other authors declare that they have no competing interests.

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