Inorganic and black carbon hotspots constrain blue carbon mitigation services across tropical seagrass and temperate tidal marshes

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Abstract

Total organic carbon (TOC) sediment stocks as a CO₂ mitigation service require exclusion of allochthonous black (BC) and particulate inorganic carbon corrected for water–atmospheric equilibrium (PIC_{eq}). For the first time, we address this bias for a temperate salt marsh and a coastal tropical seagrass in BC hotspots that represent two different blue carbon ecosystems of Malaysia and Australia. Seagrass TOC stocks were similar to the salt marshes with soil depths < 1 m (59.3 ± 11.3 and 74.9 ± 18.9 MgC ha⁻¹, CI 95% respectively). Both ecosystems showed larger BC constraints than their pristine counterparts did. However, the seagrass meadows' mitigation services were largely constrained by both higher BC/TOC and PIC_{eq}/TOC fractions ($38.0\% \pm 6.6\%$ and $43.4\% \pm 5.9\%$, CI 95%) and salt marshes around a third ($22\% \pm 10.2\%$ and $6.0\% \pm 3.1\%$ CI 95%). The results provide useful data from underrepresented regions, and, reiterates the need to consider both BC and PIC for more reliable blue carbon mitigation assessments.

Introduction

Blue carbon refers to the carbon sequestered and stored in salt marsh, mangrove, and seagrass beds (Nellemann, Corcoran, & M., 2009). There has been a growing interest in measuring, mapping, and valuing blue carbon stocks to promote their importance in climate change mitigation (Macreadie et al. 2019). However, the use of total organic carbon (TOC) for sequestration and stock assessments has recently been in question. Traditional assessments have largely failed to remove allochthonous organic recalcitrants from the equation (Gallagher 2014; Gallagher 2015; Gallagher 2017; Chew and Gallagher 2018; Chuan et al. 2020; Gallagher et al. 2020). These stable organic forms are not produced by the blue carbon ecosystem; deposition within the ecosystems' sediments does not afford additional protection from remineralization. Consequently, their presence is not a measurable storage or sequestration service in the mitigation of greenhouse gas emissions. While the argument is unequivocal and recognised by the IPCC as an important blue carbon constraint (Bindoff et al. 2019), data are scarce on their contributions to blue carbon ecosystems.

Arguably, black carbon (BC) is the most ubiquitous of these recalcitrant organic forms. Black carbon also referred to as pyrogenic carbon, comes from the burning of biomass and fossil fuels (Gustafsson et al. 2009). Their recalcitrance to microbial attack comes from the charring effect, the extent of which results in greater degrees of recalcitrance (Binh Thanh et al. 2010). As previously implied, as a constraint on blue carbon mitigation BC production cannot be caused or initiated within the ecosystem. This is certainly the case for seagrasses. For mangroves, historical satellite records of combustion sites indicated that this was unlikely, at least outside deliberate attempts during periods of drought to ignite a build-up of dumped trash adjacent to or inside the mangrove forest (Chew and Gallagher 2018). For saltmarsh, fire can be more common and deliberate but largely restricted to the tall canopies of relatively dry reed and grass-dominated ecosystems (Nyman and Chabreck 1995). As result, BC delivery to these blue carbon ecosystems can come from soil from a history of catchment fires and pollution, or more immediately and directly from atmospheric deposition (Chew and Gallagher 2018).

In addition to BC, attention has been given to the role of the particulate inorganic carbon (PIC) fractions. These calcareous products can be either produced by the ecosystems' biota or geogenic carbonates, namely coral sands or dolomitic soils that makeup part of the sedimentary matrix. The biotic carbonates originate from the ecosystem's plant epibionts, benthic macrofauna (e.g. crabs, snails, and bivalves), and more recently found in seagrass leaves of *Thalassia* spp. (Enriquez and Schubert 2014). There is also evidence for sedimentary carbonate formation under anoxia (Chuan et al. 2020), however, the extent of this process is currently not known. Importantly, PIC has been regarded as the remnants of not a carbon sink but a carbon source process. During the production of PIC, CO₂ is formed and expelled into the atmosphere after a chemical redistribution with other dissolved inorganic forms of carbonate and bicarbonate. The extent of which is determined by water body salinity and the atmospheric CO₂ (Ware et al. 1992; Saderne et al. 2018).

For recalcitrant organic forms such as BC, the coverage in the literature is still embryonic. However, PIC measurements are gaining traction where distinctions can be made between ecosystems that largely support biogenic PIC (Saderne et al. 2019). Yet, compilations of both BC and PIC remain rare (Gallagher et al. 2020). To increase data coverage, and within the limits of resources, measurements of BC and PIC were made across targeted blue carbon ecosystems of Malaysia and Australia to investigate the sum of black and calcareous blue carbon bias. The wetlands were chosen to expect a large bias in traditional stock estimates from the cumulated fractions of BC and PIC to the TOC stocks. To increase generality, the examples were separated by very different geographic and climatic regions, as well as categories of blue carbon ecosystems where the BC content has not been specifically addressed. That is, examples from the southern limit of Australian rural temperate salt marshes (Tasmania), and a tropical urban seagrass meadow within Malaysia. Both regions are in BC hot spots and underrepresented in our understanding of BC and PIC. Furthermore, we discuss our findings to identify ways in which our study can be extended to motivate further investigations of blue carbon bias as a stock mitigation service by accounting for the sum of both BC, other autochthonous recalcitrants, and PIC contributions.

Methods

Site description

The temperate salt marsh examples are located in Southeast Tasmania, along the margins of Blackmans Bay (Fig. 1b). The surrounding areas are largely rural with agricultural and forested lands punctuated by small settlements, namely Murdunna, Boomer Bay, Marion Bay, and the village of Dunalley (Fig. 1b). The salt marsh vegetation is largely comprised of fire-resistant succulent herbs, which support an abundant calcareous benthic fauna (Prahalad et al. 2020). The succulents are mainly *Sarcocornia quinqueflora*, mixed with the succulent shrub *Tecticornia arbuscula*, occasionally interspersed with the sedge *Gahnia filum* (Prahalad et al. 2020). The sea rush *Juncus kraussii* is also notable in places and forms large stands in the case of the Marion Bay salt marsh, the largest marsh in southern Tasmania (Prahalad et al. 2020). Their surrounding catchments have a history of fire events. Such fire events are ubiquitous across

the large temperate regions of Australia (Attiwill and Adams 2013). Furthermore, their shallow soils (< 0.5 m) are less likely to dilute the BC fraction over older and deeper soils that have seen less extensive fire events (Doerr and Santín 2016). Of particular note are two pivotal fires that engulfed the salt marshes. The 1967 Black Tuesday bush fires across most of Southeast Tasmania (Tasmania, 1967). A more recent bush fire in 2013 (Hyde 2013) spread through the catchment and immediate surroundings of Blackman Bay and northern parts of the Forestier Peninsula.

The tropical seagrass meadow example occupies Middle Bank within the Penang Straits (Fig. 1a) and is Malaysia's second-largest (50.7 ha). The bank and the channels to its immediate west and east have been a persistent feature, as recorded from old naval charts since the early 19th century (Chee et al. 2017). The meadow is of a patchy configuration dominated by the large leaf *Enhalus acrooides*, mixed with the small-leaved spoon grass *Halodule ovalis*, which supports abundant calcareous benthic fauna (Shau Hwai et al. 2007). The Penang Straits harbors a major port, a light industrial complex, and a relatively large urban population density (Chee et al. 2017). Along with industrial and vehicle BC combustion products, the region has been regularly affected by the peat fires of Indonesia from the late 20th century (Gaveau et al. 2014). Other possible sources of BC may emanate from the Penang River to the north of the Middle Bank, and the Jelutong municipal waste landfill on the peninsula to the west of the bank (Fig. 1a). Furthermore, its patchy seascape and coastal exposure to fine organic clay particles can elevate BC fractions from greater amounts of sorption and exported allochthonous litter (Gallagher et al. 2019). Although, supply from soil washout and export from the bank may be constrained. The river does not appear to flow directly over the bank (Asadpour et al. 2012), and sharp resuspended sediment boundaries within the confines of the bank (Lim et al. 2009) indicate its isolation from surrounding coastal waters.

Sampling

Salt marsh sampling sites ($n = 22$) for integrated stock assessments were selected across five locations, with a minimum of 3 sites per location (Fig. 1b). In each location, sites were selected at random towards the seawater edge at low tide at least > 100 m apart. To identify any confounding of BC dependence with soil stock depths < 1 m, sediment cores were also taken for macro char profiles in the following year (May 2018) where the surrounding fires were at their most and least intense across the salt marsh sampling zone. The position of the macrochar peaks down sediment cores have been validated as a means to date and determine the rates of accretion from the known pivotal fire event of 1967 (Gallagher and Ross 2017). It is expected that the pivotal fire experienced by the first author (JBG) in 2013 across the Blackmans Bay region (Fig. 1b) would also leave a definitive near-surface sedimentary signal. Sampling sites for the Middle Bank seagrass meadow were located in the northerly two-thirds of the meadow. The area was bordered by two possible sources of BC pollution other than from atmospheric fallout, namely the Penang River and the Jelutong municipal tip (Fig. 1a). Sites were selected randomly with > 200 m separation in regions stratified on the east and west side of the meadow facing the surrounding navigation channels. The final number of sites ($n = 15$) was determined by the time of exposure at low tide, and an ability to traverse the soft sediments. Nevertheless, the extent of the sampling region is

representative of a sparse tropical seagrass meadow at the landscape scale (Habeeb et al. 2007). All sites sampled in both salt marsh and seagrass were assumed to be sufficiently distant (> 20 to 30 m) as to be depositional independent of each other (Zhao et al. 2006).

An open-faced 'Russian Peat Corer' was used to take 50 cm cores of uncompressed sediment for macro char profiles in vegetated salt marsh soils and stock measurements within seagrass stands. The remaining salt marsh stock samples were exhumed carefully with a PVC pipe and shovel from the base of the soil, where quaternary sands mark the depth of the salt marsh accumulation. Samples were immediately put on ice before transport. The sample cores were mixed well within sealed plastic bags to physically represent the stock variables average; after which a measured volume ($\geq 5 \text{ cm}^3$) was taken for dry bulk density with a cut-off 20 cm^3 syringe, in the manner of a piston corer. After drying at 60°C the remaining mixed samples were shaken and sieved through a $200 \mu\text{m}$ mesh to remove both roots and large shells pieces before further processing for organic matter and elemental carbon analysis (Chew and Gallagher 2018).

Sample Analysis

Subsamples of more than 2 cm^3 for stock analysis were ground to $< 63 \mu\text{m}$ before organic gravimetric and elemental analysis. The calcareous carbon content as PIC was analysed gravimetrically (Heiri et al. 2001) using a molecular conversion coefficient (0.273). Particulate inorganic carbon as a source of CO_2 (PIC_{eq}) was adjusted for salinity and atmospheric equilibrium in shallow waters by a coefficient fraction of 0.63. The coefficient was determined for the calcite phase in seawater by Ware et al. (1992) as applied to other blue carbon ecosystems (Howard et al. 2018). The coefficient of variation (CV) for PIC analysis was found to be typically around 0.14% ($n = 6$). All carbon and organic matter contents as dry weight was corrected for salinity (Lavelle et al. 1985). Salinity and pH measurements were taken from the pore water with a handheld refractometer and portable pH meter respectively (Chew and Gallagher 2018).

Chemothermal isolation followed by combustion was used to measure black organic carbon (Elmquist et al. 2004). The method is well tested and covers a wide spectrum of black carbon forms, from chars to soots (Hammes et al. 2007). Black carbon was first isolated after a slow ramping speed to 360°C and acidified with HCl to inhibit self charring before analysis of the remaining organic carbon (Elmquist et al. 2004), according to modified protocols devised and tested by Chew and Gallagher (2018). For the Middle Bank sediments, total organic carbon and BC analysis were first determined gravimetrically after combustion as total organic matter (TOM) and after the black organic matter was isolated at $360 \pm 1^\circ\text{C}$ (BM). Their respective carbon contents were then calculated from previously constructed seagrass sediment calibration curves that used a CHN analyser as a measure of carbon from thermogravimetric organic matter determinations (Fig. S1 Supplementary material and details of method). Identical conditions of weight, temperature, ramping speeds, combustion time, and laboratory furnace were used to isolate the BM ($100 \pm 1^\circ\text{C}$ to $360 \pm 1^\circ\text{C}$ over 24 hrs). Combustion times and conditions for the TOM ($100 \pm 1^\circ\text{C}$ to $550 \pm 1^\circ\text{C}$) and the isolated BM ($100 \pm 1^\circ\text{C}$ to $550 \pm 1^\circ\text{C}$) were those used by Heiri et al. (2001). The curve was constructed from a Malaysian lagoon that included the sediment type and seagrass genera assemblages

found at Middle Bank. In the curve construction, the square root of the sum of the regressions' residual mean square error and the square of the standard deviations of the in-house mangrove sediment standards (mean TOM 16.71% and BM 2.83%) indicated the resultant CV for TOC and BC contents were typically around 0.12 and 0.10 and respectively. For the salt marsh samples, TOC, and BC after isolation using Chew and Gallagher (2018) protocols, were measured directly with a Thermo Finnigan EA 1112 Series Flash Elemental Analyser after HCl acidification within silver cups (0.7 and 1.7 mg with an accuracy of 0.1 μg) at the Central Science Laboratories, University of Tasmania, and calibrated using a certified sulphanilamide standard (standard deviation 0.08%, $n = 5$). In addition to BC contents, their BC stable isotope signatures were taken from finely ground dry subsamples and analyzed after HCl acidification at the Water Studies Centre, School of Chemistry (Monash University). The laboratory used an NCA GSL2 elemental analyzer interfaced to a Hydra 20–22 continuous-flow isotope ratio mass spectrometer (IRMS; Sercon Ltd., UK). The precision was $\pm 0.2\text{‰}$ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (standard deviation; $n = 5$). To ensure the accuracy of the isotopic results, internal standards (i.e. ammonium sulfate, sucrose, gelatine, and bream) were run concurrently with the sediment samples. These internal standards have been calibrated against internationally recognised reference materials (i.e. USGS 40, USGS41, IAEA N1, USGS 25, USGS 26, and IAEA C-6).

The two salt marsh cores taken from the proximity of Marion Bay and King George Sound salt marsh (Fig. 1a) were used for macrochar particle profiles. Before the particles were identified and counted, humic acids and organic matter were removed with KOH followed by H_2O_2 on $\approx 2 \text{ cm}^{-3}$ of wet sediment and passed through a 100 μm sieve before being washed into a petri dish (Stevenson and Haberle 2005). The volume was estimated by KOH reagent displacement. The remaining black particles posing as a fractured appearance amongst the now the bleached sediment particles were counted automatically with Image J™ on an enhanced black and white image taken through a magnifying glass (x 5) underlying a white base (Stevenson and Haberle 2005). A maximum entropy filter was used to isolate the particle images, and any particles in contact with each other were separated with a bone shape function.

Data Analysis

Ordinary least squared regressions (OLS) and a one-way analysis of variance for the difference between means (ANOVA), and Student t-test between mean pairs were carried out in Sigmaplot 12™. Where normality was rejected, a Mann–Whitney test replaced ANOVA (Sigmaplot 12™). Comparisons between OLS parameters were determined using an analysis of covariance of their slopes (PAST™). Differences between OLS intercepts of the dependent variable were performed using a Student t-test taken from their standard errors using the 2 degrees of freedom that supports an OLS regression. The precision as coefficient of variations (CV) were $< 10\%$ for all carbon variables: TOC ($\pm 6.3\%$, $n = 14$); BC ($\pm 9.0\%$ ($n = 4$); PIC (± 2.8 , $n = 14$). For $\delta^{13}\text{C}$ ($\pm 0.15\%$ ($n = 5$)). The coefficient of variation ($n = 5$) for char counts was typically 5.3% after repeating the image analysis after remixing the sample identified as the 1968 peak (the numbers ranging between 670 to 750 counts cm^{-3}). We recognize that this may be statistically underestimated by not repeating the cleaning stage with a separate subsample. However, the order of magnitude differences in what appears to be the background counts over and the recognizable peaks are

not consistent with noise or variance. This contention was supported by the results of a smoothed first derivative with an amplitude threshold of 0.6 and a slope threshold of 0.2 which separated the peaks from baseline variance (O'Haver 2020) (Supplementary material Peak Detection Template). Data, location coordinates, and species descriptions related to the figures presented in the article can be found in the Institute of Marine and Antarctic Studies Metadata open access (<https://doi.org/10.25959/J4PJ-AD26>), and Supplementary materials Table S1 and Table S2.

Results

Sediment Parameters

All the salt marsh sampling sites had full coverage of succulent plant species (Table S1 Supplementary material). Only the coastal wetland strip surrounding King George Sound showed evidence of macrofaunal bioturbation. Numerous burrows were observed over the surface, the effect could otherwise reduce and broaden macrochar peaks deposited by fire events. Salinities and pH did not significantly correlate with each other ($P < 0.05$, Fig. S2 Supplementary material) and ranged from 1.4‰ to 30.4‰ and 4.83 to 7.83 respectively (Table S1 Supplementary material). Moderate but significant correlations ($P < 0.05$) of pH and salinity with other soil parameters were restricted to peat content with bulk density, soil depth, and other biological soil structures (Fig. S2 Supplementary material). All salt marsh data were combined to investigate relationships between variables over landscape scales as one statistical population. Individual carbon parameters across salt marshes displayed equal variances with no differences in their means or median contents (mean TOC $P = 0.45$, mean BC $P = 0.84$, median PIC $P = 0.67$; equal variance TOC $P = 0.92$; BC $P = 0.60$, PIC $P = 0.15$). Middle Bank seagrass sediment pore water salinities were found to be invariant at around 30‰ across the sampling region. Spatial distributions using a kriging model indicated that bulk densities were the smallest and therefore muddier (Tolhurst et al. 2005) towards the NE quarter of the sample area with increasing saltiness (Tolhurst et al. 2005) towards the WSW to SW of the sampling area (Fig. S3 Supplementary material). Furthermore, the average BC $\delta^{13}\text{C}$ signatures across all salt marsh sites (-24.86 ± 0.63 , CI 95%) was consistent with allochthonous sources. The signatures are typical C3 tree and shrub vegetation signatures (-32‰ to -22‰ , average -24‰) and not consistent with the fire susceptible C4 salt marsh reed and cord grasses (-17‰ to -9‰ , average -14‰) (Krull et al. 2007).

Organic and Inorganic Carbon Variability

The differences between all salt marsh and seagrass carbon forms were striking (Fig. 2a,b). On average, the salt marsh TOC content was around 35 fold greater than for seagrass. For the remaining BC and PIC_{eq} content variables, this was reduced to around 15 and 4.3 fold respectively from the larger PIC and BC fractions within seagrass sediments. When expressed as a concentration, the differences were reduced considerably. On average salt marsh [TOC] was only 8.3 fold greater than seagrass, with the remaining concepts falling in proportion accordingly (Fig. 2c,d). This reflects the smaller dry bulk

densities found in the salt marsh ($0.39 \pm 0.10 \text{ g cm}^{-3}$, CI 95%) over that of the sandier seagrass sediments ($1.50 \pm 0.07 \text{ g cm}^{-3}$, CI 95%).

In contrast to content and concentration, the TOC stocks between these two ecosystems did not show any significant differences (Fig. 2e,f, $P = 0.36$ Mann-Whitney Rank Sum Test). The median salt marsh and seagrass TOC stocks were 70.3 MgC ha^{-1} (39.1 to 103.5 as 25% and 75 % quartiles) and 59.5 Mg ha^{-1} (43.2 Mg ha^{-1} to 67.0 Mg ha^{-1} as 25% and 75% quartiles). This convergence was the result of temperate salt marshes' shallow soils of $< 30 \text{ cm}$ (Table S1 Supplementary material). Stocks are currently defined by a depth of disturbance of at least 1 m. This is a depth where organic matter is subject to oxidation and remineralization (Pendleton et al. 2012). However, this convergence with TOC stocks between the salt marsh and seagrass did not run to the remaining BC and PIC_{eq} stocks (Fig. 2e,f). Salt marsh BC stocks diverged to less than half of that found in seagrass (median $13.1 \text{ MgC ha}^{-1} \pm 3.7$, CI 95% and $25.2 \text{ MgC ha}^{-1} \pm 4.9$, CI 95% respectively, t-test $P < 0.001$). And salt marsh PIC_{eq} stocks were an order of magnitude smaller than seagrass (a median of 2.3 MgC ha^{-1} 1.2 to 2.4 for respective 25% and 75% quartiles, and a median of 21.0 MgC ha^{-1} 19.2 to 21.3 as 25% and 75% quartiles, respectively, Mann-Whitney Rank Sum Test $P < 0.001$).

The shallow and variable soil depths across the salt marshes $< 1 \text{ m}$ implies that their carbon stocks are also a function of depth. However, no dependence was found for salt marsh BC/TOC fractions with soil depth (Fig. 4a), despite a similar average accretion rate over 50 years of deposition of around 0.25 cm yr^{-1} and 0.21 cm yr^{-1} . It appeared that any BC/TOC soil depth dependence may have been confounded by an inability for all sites to record all pivotal fire events. King George Sound salt marsh did not record the 2013 fire that destroyed the nearby Dunalley village (Fig. 4b). Overall the average accretion rate across the Southeast Tasmanian salt marsh sites is likely represented as around 0.23 cm yr^{-1} . The two accretion sites likely represent the extremes of the sampling region and a type. The similarities also support the contention that recent salt marsh accretion from the mid 20th century around Tasmania is determined by changes in sea level (Gehrels et al. 2012). Given the average TOC across all sampling sites of around 49.3 mgC cm^{-3} , as the sum of the average dry bulk density and TOC content, equates to rates of carbon sequestration across the Southeast Tasmanian sites of around $113.4 \text{ gC m}^{-2} \text{ yr}^{-1}$ ($1.13 \text{ MgC ha}^{-1} \text{ yr}^{-1}$). Indeed, this average from a relatively small number of cores is likely to be representative of the region. Single-core sites have been found to represent the mean in carbon accretion within a salt marsh and between salt marshes of the same region (Callaway et al. 2012). In terms of carbon sequestration services corrected for BC deposition, and BC, through subtraction from the TOC content, and the PIC_{eq} content as a source of CO_2 , this equates on average to around $88.5 \text{ gC m}^{-2} \text{ yr}^{-1}$ and $81.7 \text{ gC m}^{-2} \text{ yr}^{-1}$ respectively, a total mitigation bias of around 18.3%.

In summary, irrespective of the similarity between the tropical seagrass meadow and temperate salt marsh TOC stocks (Fig. 2g,h), mitigation corrections for BC and PIC were very different. For seagrass, the BC/TOC fraction was around $43.3\% \pm 5.9\%$ (CI 95%) with nearly matching constraints from PIC_{eq} of 38%

$\pm 6.6\%$ (CI 95%). In total, there was an $81.3\% \pm 9.0$ (CI 95%) overestimate in sedimentary carbon stocks. This is in contrast to the salt marshes, where the smaller but moderate BC/TOC fraction of $22.0\% \pm 10.2\%$ (CI 95%) was supplemented by a significant but minor $\text{PIC}_{\text{eq}}/\text{TOC}$ fraction of $6.0\% \pm 3.1$ (CI 95%). Nevertheless, taken together this is a significant overestimate of more than a third of carbon stocks as a mitigation service ($28.0\% \pm 10.8\%$ CI 95%). For the average salt marsh carbon sequestration services, which were independent of soil depth the bias was less extensive (18.3%).

Dependence of Carbon Concepts with Total Organic Carbon

Ordinary least squares regressions across the seagrass meadow produced a strong positive correlation of sedimentary BC contents with its TOC counterpart closely trending from the origin (Fig. 3a, $r = 0.84$, $\bar{r} = 0.99$). In contrast, the salt marsh BC–TOC content regression model displayed only a moderate correlation, with good power but with poor predictive value (Fig. 3c, $r = 0.62$, $\bar{r} = 0.88$). The poor prediction is reflected in the relatively larger variance around the regression line. However, this result may have been leveraged by a single large sample value (Fig. 3c ■, Cook's distance = 1.2). After the removal of this likely outlier, the resultant regression became weaker and exhibited low power ($r = 0.39$, $\bar{r} = 0.43$). For the seagrass meadow's sedimentary PIC contents, we found a significant dependence on its TOC counterpart ($r = 0.83$, $\bar{r} = 0.99$), but with relatively little variation in PIC with TOC contents (Fig. 3b). This invariance with TOC content was similar across the salt marsh (Fig. 3d).

Discussion

Measuring and comparing values of carbon concepts (e.g., stocks, concentrations, and contents of TOC, BC, and PIC) across underrepresented blue carbon ecosystems and environs are useful measures of variance towards global assessments. However, additional insight and predictive power are also possible by their relationship with each other (Chew and Gallagher 2018; Gallagher et al. 2019). To this end, the following discussion examines the statistical ranges and relationships between those carbon concepts, as a measure of their importance for mitigation service assessment and insights on the nature of the supply of BC and PIC variance

Black Carbon Fraction Variance and Sources

It has been argued that the structure of BC–TOC regressions is the result of the addition of two possible allochthonous supply paths, namely, aeolian deposition and soil washout. With aeolian deposition, sedimentary contents of BC across a meadow or wetland is independent of its TOC. The variance in BC content is likely affected by different accretion processes for largely similar rates of aeolian supply over larger areas than embayments (Chew and Gallagher 2018; Gallagher et al. 2019; Sun et al. 2008). Whereas sedimentary BC supplied from soil washout increases with TOC, where the BC intercept represents the underlying remnant of BC invariance from additional aeolian deposition (Chew and Gallagher 2018). In the case of Tasmanian salt marshes, the relative invariance of BC with TOC appears to be consistent with a supply of BC dominated by aeolian deposition (Fig. 3c). As BC emitted to the atmosphere is undiluted by soil organics, the aeolian deposition would be expected to elevate the

BC/TOC sedimentary fractions ($22\% \pm 10.2\%$, CI 95%, and median 14.7%) over a salt marsh supplied largely from BC diluted with soil organics. Indeed, it was noted that the bush fires of 2013 immediately surrounding the Tasmanian salt marsh were quickly deposited as large macrochar particles. This was not only felt by the author (JBG) at the time of the fire but also observed as heavy deposits washed up on the beaches and the local seagrass surface sediments after the 2013 fire (Fig. S4 Supplementary material). Nevertheless, the dilution of relatively pure aeolian BC deposits and their resultant BC/TOC fractions are likely to be less than another salt marsh around the globe. The average Tasmanian salt marsh carbon sequestration rate ($113.4 \text{ gC m}^{-2} \text{ yr}^{-1}$) is close to half of the global average ($244.7 \text{ gC m}^{-2} \text{ yr}^{-1}$) (Ouyang and Lee 2013). Although, they were notably larger than the tidal marshes across the neighbouring state of Victoria ($86.86 \text{ gC m}^{-2} \text{ yr}^{-1} \pm 4.79 \text{ SE}$) (Macreadie et al. 2017). The differences appear to be largely the result of a higher TOC content across the Tasmanian salt marsh ($14.41\% \text{ dry wt} \pm 1.63 \text{ SE}$ and $7.86\% \text{ dry wt} \pm 0.59 \text{ SE}$ respectively). Furthermore, it is unlikely that the larger TOC content in Tasmanian systems comes from additional BC inputs. The state of Victoria has not been immune from pivotal fire events (Forest Fire Management Victoria, 2020).

Comparisons of allochthonous BC content with other salt marshes do not appear to be directly available. Nevertheless, data exists for autochthonous refractory carbon (C_{rf}) fractions for two humid subtropical climate salt marshes across the USA and two temperate oceanic climate salt marshes across the Iberian Peninsula (Leorri et al. 2018). The authors consider that this fraction contained an old pyrogenic component supplied from the upper catchments. Although it was not clear what were the origins of the remaining fractions if any, we also noted that the analytical method was a slight modification of an older BC methodology. Nevertheless, their reported C_{rf} /TOC fractions values were still smaller than our temperate BC fractions, ranging from 6.4–13.1% and 13.1–17% as dry wt for the USA and Iberian pairs, respectively. Either way, their study indicates that while BC is an important part of an allochthonous recalcitrant carbon, there maybe are other important allochthonous recalcitrants to consider. Possibilities have been suggested. These are the kerogens associated with shale deposits, phytolith occluded carbon and the more recent inputs from microplastics (Chew and Gallagher 2018; Rillig 2018).

In contrast with the Tasmanian salt marsh, the BC contents across Middle Banks' tropical seagrass meadow appear to show an unexpected dependence with TOC (Fig. 4a). This is consistent with a supply of BC largely from soil or adjacent sedimentary washout (Chew and Gallagher 2018; Gallagher et al. 2019). It appears that Middle Bank may not have been as isolated from the shoreline as first hypothesised. Although, there is a small BC positive intercept to suggests an additional aeolian deposition. The effect of which would have elevated the meadows' BC/TOC fractions. Interestingly, the proportional response of BC with TOC is statistically identical to that found across a rural meadow dominated by the same seagrass species (*Enhalus* spp.) at Limau Limauan ($P = 0.64$, reanalysed from Supplementary material attached to Gallagher et al., 2019). Although, Middle Bank supported a larger overall BC/TOC fraction than Limau Limauan ($26.1\% \pm 4.9 \text{ CI } 95\%$). This may be a combination of a smaller fraction of BC aeolian deposition, based on a statistically smaller BC intercept ($P = 0.003$ of the two intercepts being equal) and the extent of black carbon pollution between these regions. Limau

Limauan is adjacent to the shoreline and located within a North Borneo marine park. The park being located within the penumbra of the Southeast Asian BC hotspot (Chew and Gallagher, 2018; Permadi et al. 2018). Nevertheless, the differences between Limau Limauan and the Middle Bank seagrass meadows did not reflect the larger categorical differences in BC emissions between their respective regions' (Permadi et al. 2018). The apparent normalization of this difference may reflect their different tidal niches. The Middle Bank seagrass meadow is intertidal and the Limau Limauan seagrass meadow is subtidal. It has been noted that tidal exchange can remove remobilise particulate BC from intertidal wetlands to coastal waters as its dissolved fraction (Dittmar et al. 2012). Indeed, such a process is also consistent with the smaller concentration of BC in the Tasmanian salt marsh from the more extensive 1967 'Black Tuesday fire' over the larger surface BC peak from the 2013 fire (Fig. 3a).

Particulate Inorganic Carbon Variance

The relative invariance of PIC with TOC content, more so for salt marsh than the seagrass meadow, may simply reflect their canopy as a distinct calcareous faunal niche. Indeed, extensive and relatively constant fractions of PIC with near-identical TOC stocks have also been reported for Merambong, another Malaysian intertidal coastal seagrass meadow that supports an *Enhalus* spp. (Rozaimi Jamaludin et al. 2017). However, PIC stocks were more than double that of the Middle Bank meadow. The reasons for this difference are unclear, other than due to the difference in sample preparation. There was no indication that there has been a need to separate any larger calcareous pieces < 2 mm that would otherwise elevate the PIC content (Rozaimi Jamaludin et al. 2017).

Carbon Mitigation Services

The results indicate that carbon stock services in mitigating greenhouse gas emissions may not be significant for some tropical intertidal coastal seagrass meadows, particularly those which appear to support an abundant calcareous benthic fauna and occupy BC hotspots. In the same vein, current carbon stock mitigation for some temperate salt marshes estimates may be required to be reduced by up to a third ($28.0\% \pm 10.8\%$ CI 95%). This is largely by their BC contributions in relative terms and absolute terms by their shallow soil depths < 1 m.

Limitations of this study and further implications

It has been accepted that despite dissolution (Dittmar and Koch 2006) BC remains largely unmineralised over climatic scales (Kuzyakov, Bogomolova, & Glaser, 2014). In contrast, accounting calcareous mitigation services are not as straightforward. Current blue carbon conceptual models caution the presence of geogenic carbonates (Saderne et al. 2018). Their production and associated emissions of CO₂ have been produced outside the ecosystem. Such a geogenic constraint is not necessary for the Middle Bank seagrass and the Tasmanian salt marsh sediments. It would then seem that only the ecosystems' biogenic carbonates play a role as an ecosystem carbon source. However, a more considered ecosystem service logic could argue for a different set of considerations. These are the services that account for the source and stability of all carbonates relative to those services, and emergent services within a replacement non-vegetated ecosystem. For example, the dissolution of coral

rubbles by anthropogenic ocean acidification outside vegetated canopies (Eyre et al. 2014; Sulpis et al. 2018). Such a process adds to the pool of coastal dissolved inorganic carbon (DIC) which turns over at millennial scales (Maher et al. 2018). The removal of this carbon sink service could be conceivably be constrained by the presence of a vegetated coastal canopy. Canopy photosynthesis is known to elevate its water column pH (Kowek et al. 2018; Krause-Jensen et al. 2016) and would thus constitute a reduction in mitigation services over the calcareous surface sediments of the non-vegetated alternative. Alternatively, the presence of PIC below the surface sediments is only a remnant of its original deposit. Where the sediments are acidic, usually associated with sulphate reduction, the PIC can dissolve and sequester carbon as DIC to the coastal and oceanic pool with a millennial turnover rate (Maher et al. 2018). On the other hand sulphate reduction will produce sufficient bicarbonate for CaCO_3 precipitation (Mucci et al. 2000), or with sufficient iron and acidity there is evidence to show FeCO_3 precipitation and associated production of CO_2 (Chuan et al. 2020). Additional consideration may also be required on the origin of the ecosystem's biogenic carbonates. The production of calcium carbonates as part of a photosynthetic carbon concentrating mechanism has been noted for some seagrass (Enriquez and Schubert 2014) and macroalgae that can inhabit seagrass meadows (Borowitzka and Larkum 1987). As carbonate precipitates, the emission of CO_2 within the lacunae their thalli and leaves are in large part recycled to RuBisCO for carbon fixation. Consequently, their presence is the remnant of an additional organic carbon sink and not a source. Indeed such plant carbonates can be a major contributor to calcareous sediments (Enriquez and Schubert 2014; Perry et al. 2019). For salt marsh plants, this process as far as we are aware has not been tested. Nevertheless, mitigation services are also measured relative to the replacement ecosystem. For PIC stock it could be expected that in a non-vegetated state, its subsequent oxidation and dissolution would be sequestered to the DIC pool (Howard et al. 2018). However, the extent of this has not been as tested.

Conclusions

It was found that allochthonous BC was likely to be a major contributor to the TOC sedimentary stocks (around 43.3%) for coastal seagrass meadows that occupy BC hotspots, such as Middle Bank (Peninsula Malaysia). Although, the extent of this fraction may be reduced by circumstances of remobilisation from intertidal flushing of its dissolved BC fraction. When the BC constraint was pooled with a similar biogenic PIC_{eq} fraction (around 38%), there was very little remaining of the carbon stock as a mitigation service (around 18.7% as sequestered organic carbon. In contrast, the temperate salt marshes subject to pivotal fire events likely supported a moderate allochthonous BC fraction of around 22% of its TOC stocks, with minor contributions from the PIC_{eq} (around 3.2%), such as Southeast Tasmania. This BC fraction appears to be elevated over another less temperate salt marsh with a similar allochthonous carbon concept. The reasons are unclear, other than salt marshes in Tasmania supports a shallower sediment profile, lower than the average global rate of carbon sequestration, and has been impacted by nearby large forest fire events. These appear to have supplied BC largely undiluted by aeolian deposition. These examples of extremes if not ubiquitous, by themselves represent large areas across climatic extremes of the temperate and the tropical that can support such extremes. Either way, it would be prudent to address the sources,

recalcitrance, and function of particulate carbon types to address what can be considered bias in carbon stocks, and sequestration mitigation assessments. This will assist in avoiding both local and global overestimates and ensure that greenhouse gas emitters do not exceed their capacity within a management scheme for carbon sinks.

Declarations

Funding sources

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Conflict of interest/competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval

Not applicable, no animal or plant materials were disturbed or manipulated during sampling or analysis

Guidelines on ethical review or waiver

Malaysia: <http://www.nccr.gov.my/index.cfm?menuid=26&parentid=17> (accessed 22 February 2021).

Australia: <https://www.arc.gov.au/policies-strategies/policy/codes-and-guidelines> (accessed 22 February 2021).

Consent to participate

Not applicable.

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CRedit authorship contribution statement

JBG: Conceptualization Validation, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Visualization, Resources. VP: Visualisation, Resources, Writing - original draft, Writing - review & editing, Funding acquisition. JA: Investigation, Resources, Writing - review & editing. All authors read and approved the final manuscript.

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Figures

Figure 1

Location of coring sites across the Southeast Tasmanian Salt marshes (Australia) (a), and the Middle bank seagrass meadow within the Penang Straits (Malaysia) (b).

Figure 2

Sediment carbon concepts across the Middle Bank seagrass meadow (left hand charts (a),(c),(e),(g)) and Southeast Tasmania Salt marshes (right hand charts (b),(d),(f),(h)). Error bars refer to confidence limits (CI) 95% about the mean; and TOC, BC, and PIC_{eq} refer to total organic carbon, black carbon, and that fraction of particulate inorganic carbon available for atmospheric exchange in seawater respectively.

Figure 3

Above (a), the relationship between the fraction of black carbon (BC) within the total organic carbon (TOC) pool with soil depth across Southeast Tasmanian salt marshes, the dotted lines refer to 95% confidence limit boundaries either side of an ordinary least squares regression. Below (b) illustrate the macro char profiles down the soil profiles at the northern and southern boundaries of the sampled salt marshes. 1968 and 2013 mark the positions of the two pivotal fires of the period.

Figure 4

Above (a), the relationship between the fraction of black carbon (BC) within the total organic carbon (TOC) pool with soil depth across Southeast Tasmanian salt marshes, the dotted lines refer to 95% confidence limit boundaries either side of an ordinary least squares regression. Below (b) illustrate the macro char profiles down the soil profiles at the northern and southern boundaries of the sampled salt marshes. 1968 and 2013 mark the positions of the two pivotal fires of the period.

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