



## Sedimentary blue carbon dynamics based on chrono-sequential observation in a tropical restored mangrove forest

Raghab Ray<sup>1\*</sup>, Rempei Suwa<sup>2</sup>, Toshihiro Miyajima<sup>1</sup>, Jeffrey Munar<sup>3</sup>, Masaya Yoshikai<sup>4</sup>, Maria Lourdes San Diego-McGlone<sup>3</sup>, Kazuo Nadaoka<sup>4</sup>

5 <sup>1</sup> *Atmosphere and Ocean Research Institute, The University of Tokyo, Kashiwa*  
raghab.ray@aori.u-tokyo.ac.jp; miyajima@aori.u-tokyo.ac.jp

<sup>2</sup> *Japan International Research Center for Agricultural Sciences, Tsukuba*  
swrmp2005@yahoo.co.jp

10 <sup>3</sup> *Marine Science Institute, University of the Philippines, Diliman*  
[mlmcglone@msi.upd.edu.ph](mailto:mlmcglone@msi.upd.edu.ph); [jcmunar1@up.edu.ph](mailto:jcmunar1@up.edu.ph)

<sup>4</sup> *School of Environment and Society, Tokyo Institute of Technology, Tokyo*  
[yoshikai.m.aa@m.titech.ac.jp](mailto:yoshikai.m.aa@m.titech.ac.jp); [nadaoka.k.aa@m.titech.ac.jp](mailto:nadaoka.k.aa@m.titech.ac.jp)

15 *Correspondence to:* Raghab Ray ([raghab.ray@aori.u-tokyo.ac.jp](mailto:raghab.ray@aori.u-tokyo.ac.jp), [raghab.ray@gmail.com](mailto:raghab.ray@gmail.com))

15

**Abstract.** Among many ecosystem services, macro-climate regulation via the storage of large amounts of organic carbon (OC) in marine sediments (also known as ‘blue carbon’) has given mangroves enormous global attention due to their role in climate change mitigation. While there are many studies on blue carbon potential of intact mangroves (i.e. naturally growing), there have been very few on restored mangroves (i.e. planted). This study aims to address this knowledge gap by examining sediment development process during early colonization (rehabilitation) of mangroves in an OC-poor estuary in the Panay Island, Philippines. Based on endmember source apportionment in sedimentary organic matter, the contribution of mangrove plant material was higher at the older sites compared to the younger settings or bare sediment where there is more contribution of riverine input. A clear increasing gradient according to mangrove development was observed for bulk OC (0.06 to 3.4  $\mu\text{mol g}^{-1}$ ), porewater OC (292 to 2150  $\mu\text{mol L}^{-1}$ ), sedimentary OC stock (3.13 to 77.4  $\text{Mg C ha}^{-1}$ ) and OC loading per surface area (7 to 223  $\mu\text{mol m}^{-2}$ ). The estimate of carbon accumulation rates (6 to 33  $\text{mol m}^{-2} \text{yr}^{-1}$ ) based on chrono-sequence are within the global ranges and show an increasing pattern with mangrove age. Although a differential yet systematic pattern of increasing OC sink based on short-term chrono-sequence can define the role of a mangrove rehabilitation program, there is a need for long-term monitoring to verify the consistently elevated OC with mangrove growth.

20

25

### 30 **1. Introduction**

The term ‘Blue Carbon’ was first introduced more than a decade ago to describe the large quantity of organic carbon (OC) present in shallow coastal habitats like mangroves (Nellemann et al., 2009). Mangroves, located around the tropical and subtropical coastal regions are known for storing significant amounts of OC in the sediment and vegetation biomass. Out of typical total carbon stock of  $739 \pm 28 \text{ Mg C ha}^{-1}$  in mangrove ecosystems, sediment OC accounts for 73-79 % (1 m depth) while above- (AGB) and below-ground root biomass (BGB) account for 14-15% and 8-9%,

35



respectively (Alongi, 2020). Mangrove sediment is the largest depository of OC owing to their efficiency in trapping suspended sediments and associated sedimentary organic matter (SOM), high algal OM (benthic algae and phytoplankton) and vascular plant-derived OM and low decomposition rates of SOM under anoxic conditions in the sediment (0.5m to 3m depth core generally represent ~ 49 to 98% of ecosystem OC stock, Donato et al., 2011). The high C sink capacity of mangroves make them one of the key ecosystems that contribute to climate change mitigation by capturing large amount of atmospheric CO<sub>2</sub> (Howard et al., 2017). However, there has been a rapid loss of mangroves due to land use and deforestation which has resulted the release of OC stored in the sediments back to the atmosphere as CO<sub>2</sub> (Valiela et al., 2001). For instance, conversion of mangroves to aquaculture ponds, paddy fields, pastures, and tree removal have resulted in OC stocks per unit area becoming 2 to 8 times lower than the intact mangrove forests (Salmo et al., 2013; Kauffman et al., 2017; Sharma et al., 2020). Therefore, quantification of blue C provides added value for mangrove protection to ecosystem services and serves as a useful management tool when implementing plans for mangrove sustainability and productivity (Sheehan et al., 2019). During the last three decades, several countries have implemented mangrove rehabilitation and restoration programs effectively to reverse mangrove forest cover loss. Mangrove restoration efforts such as Reducing Emissions from Deforestation and forest Degradation (REDD+) have reduced C emissions by considering loss from the above ground biomass part only (Pendleton et al., 2012). Country like the Philippines have started to address REDD+, loss of mangroves, and degradation of blue C habitats through their policies and rehabilitation management plans. To assess effectiveness of such efforts, there should be a comparison between the intact and restored mangrove forest in terms of sediment OC stock and accumulation rates. Restored mangrove forests are rarely explored globally with some notable exceptions in the subtropical coastal regions of China (Ren et al., 2010; Lunstrum and Chen, 2014; Wang et al., 2021), and Vietnam (Van Hiew et al., 2017; Dung et al., 2016). In this study, an evaluation based on chrono-sequential observation (a.k.a. “space-for-time-substitution” or SFT; Pickett, 1989) is made for sites in the Philippines where restoration is a viable option. The judicious use of chrono-sequential observation or SFT has already advanced our understanding of short-term temporal dynamics of carbon in naturally expanding mangroves (e.g., 66-year by Walcker et al., 2017; 70 years by Kelleway et al., 2016). Chrono-sequence is an indirect approach or a type of ‘natural experiment’ (Pickett, 1989) that can be applied to the relatively younger sites (e.g., Bakhawan Ecopark, Philippines, examined in this study) where all environmental and biological conditions of the experimental sites must be identical except for the age, and species diversity is low (Nilsson and Wilson, 1991; Walker et al., 2010).

The capacity of nearshore vegetated habitats as blue carbon sink is controlled by geophysical constraints such as sediment supply rate, depositional conditions, and tidal elevation (Miyajima et al., 2017; Jiménez-Arias et al., 2020). Based on chrono-sequential studies of intact mangroves, OC accumulation in sediments increased with tree age, and OC sources change with mangrove development (Lovelock et al., 2010; Marchand et al., 2017; Walcker et al., 2018). Most of the OC stored in mangrove sediment change from plant-derived OM to algal OM at a proximal tidal flat (Gontharet et al., 2014; Prasad et al., 2017; Ray et al., 2018). With the development of mangroves, higher vascular plant or mangrove-derived OC sources may dominate OM pool (Marchand et al., 2006). A significant fraction of mangrove-derived OC that has accumulated on top of the bare sediment can be washed away to the nearshore waters by tidal action (Brown et al., 2021; Ray et al., 2020). By considering bare sediment and old growth mangrove stands



as two extreme ends of a transect that consists of mangroves with different ages, a systematic overview of sedimentary blue C dynamics can be captured for restored mangroves. Stable isotope ratio of carbon ( $\delta^{13}\text{C}$ ) is frequently used to evaluate the relative contributions of end member sources to the OM pool through mixing models with either  $\delta^{15}\text{N}$  or C:N ratios (Ray and Shahraki 2016; Sasmito et al., 2020). The use of such biogeochemical controls on blue C dynamics has rarely been reported for the restored mangroves (*Kandelia* dominated, Van Hiew et al., 2017).

In this study, we hypothesize that the restored mangroves increase sediment C storage in accordance with maturity of vegetation. To test this, we (1) calculated total OC (TOC), dissolved OC (DOC), and OC accumulation rate along a chrono-sequence of restored mangroves forests located in the Philippines, and (2) examined how blue C varies with sedimentary geochemical properties (OC, bulk density, specific surface area). The isotopic signatures such as  $\delta^{13}\text{C}$  which allows for an efficient provenance analysis of SOM was examined. Additionally, particulate OC (POC) present in the surface water was examined to assign different end member sources in the SOM pool (e.g., plant organ, riverine and pelagic algae).

## 85 2. Material and Methods

### 2.1. Study area

Sampling was conducted in a planted mangrove forest, locally known as Bakhawan Ecopark, located in Kalibo city on Panay Island in Central Philippines during the wet season (September 2018 and 2019) and dry season (February 2019) (Fig. 1,  $11^{\circ} 43' 12'' \text{ N}$ ,  $122^{\circ} 23' 39.12'' \text{ E}$ ). The Bakhawan Ecopark is the remnant area of a former deltaic mangrove at the mouth of the Aklan River (Duncan et al., 2016). Aklan River which has a drainage area of  $852 \text{ km}^2$  flows into the northwestern coastal area of Kalibo, continuously depositing high sediment yield to form the alluvial plain down the river. Sediments entrained by the longshore current formed sandbars, beach ridges, and coalesced mouthbar deposits. To prevent the damages by coastal flooding, a large portion of the sea facing mudflat were planted with 45 ha of *Rhizophora apiculata* and 5 ha of *Nypa fruticans* in 1990 by a cooperative comprised of local families (Kalibo Save the Mangrove Association or KASAMA). An additional 20 ha of *Rhizophora* spp. were planted in 1993 (Primavera, 2004) for the purpose of stabilizing the shoreline, decreasing sedimentation to the offshore, and increasing fish stocks and wood production (Department of Environment and Natural Resources or DENR, Philippines). Insect damage of the plantation in 1997 was followed by infilling of naturally-recruited *Avicennia marina* and *Sonneratia alba*. The seafront area was replanted in 2006 with *Rhizophora apiculata*, and subsequently recolonized naturally by *A. marina* and *Sonneratia alba* (Duncan et al., 2016). New recruitment of both *A. marina* and *R. apiculata* took place on the mud bank in May/June 2019. The inland part of the Ecopark is dominated by naturally growing mangroves. Natural growth of mangrove trees and planting efforts since the 90's at the Aklan River mouth stabilized and enlarged the mangrove forest by at least 627%, to a flourishing 121 hectares today. Based on remotely-sensed data, the land area of the forest has increased by 52.42% on average every five years since 1985. The Food and Agriculture Organization of the United Nations has cited the Bakhawan mangrove for excellence in forest management (Cadaweng & Aguirre 2005).



Tide in the Bakhawan Ecopark is semidiurnal microtidal with the highest amplitude of around 2m. The mangrove forest floor is fully inundated during the high tide. At the mouth of the Aklan River, water meanders along a small channel between the sandbar and mangrove-lined coast. The climate of Aklan is categorized as Type III (according to the Philippines Atmospheric, Geo-Physical, and Astronomical Services Administration) with no pronounced maximum rain period except for short dry periods of 1–3 months (December to February or March to May). The rest of the year represents wet season with total annual rainfall of  $3200 \pm 775$  mm and mean temperature of  $27.2$  °C (2017-2018, JRA-55 reanalysis).

Sediment sampling locations were categorized according to mangrove ageing as bare sediment (BS, 0-yr), pioneer mangroves (PM, 0.25-yr), young mangrove (YM, 10-yr), adult mangrove (AM, 20-yr) and mature mangroves (MM, 30-yr). These categories are partly influenced from Marchand et al. (2003) who examined chrono-sequential sedimentary OC in the naturally growing *Avicennia* dominated mangroves in the French Guiana muddy coast. The center of the mangrove forest is dominated by AM and the sea facing edge of the Ecopark has decreasing mangrove age from MM to PM. Both BS and PM are inundated during the high tide. *Rhizophora apiculata* is the dominant species at YM, AM and MM, while mixed mangroves (*Avicennia* and *Rhizophora* sp.) are found at PM. Among two sites of bare sediment, BS1 which is closer to YM, was sampled during the wet season, while BS2 which is isolated from the mangrove sites, was sampled in the dry season, (Fig. 1).

## 2.2. Sampling procedure

Sediment thickness was measured at each site using a tool for cone penetration test (KS-159, Kansaikiki Inc.) (Yoshikai et al., 2021). Single core was collected at each site during the low tide by manually pushing a Eijkelkamp peat sampler (DIK-105A, Ø 52mm, length 50cm) into the sediment. A total of 8 cores were retrieved during the survey period with seasonal collection made at AM, YM and MM sites. The GPS coordinates at all sampling sites were recorded to locate them again for duplicate sampling in a different season (Garmin MAP64s). Immediately after sampling, each core was sectioned into 2 cm interval up to the first 10 cm and 5 cm interval beyond 10 cm depth. Average sample depth at each site was consistently around 50 cm. *In situ* pH (NBS scale), temperature and oxidation reduction potential (ORP, AgCl electrode) were recorded for each section using hand-held multiparameter probes (HORIBA pH-conductivity sensors, WTW redox sensor). About 0.8 to 1 kg cores were collected from each site. Visible root material, decaying plant matter and dead wood were removed from the sediment sample in the field. Samples were kept in styrofoam box and brought to the laboratory within 3-4 hours after collection.

Additional sediment cores for porewater sampling were collected inserting PVC corer manually at each site (6.5cm inner diameter, 70 cm length). A total of 7 cores were retrieved on seasonal basis (single core from each site per season). Immediately after retrieval, the top and bottom ends of the corer were closed using rubber caps. Rhizon tubes (Rhizosphere Research Products) were inserted to the holes drilled at specified intervals (0.5cm interval till the first 4cm followed by 2 cm interval up to the 10cm, and finally 5 cm interval). A Rhizon tube is a small microporous polymer tube (2.5 mm diameter, < 0.2 µm pore size of the membrane) connected to a plastic syringe (25 ml capacity) by a standard luer-lock connector. Around 8–10 ml of porewater were extracted in ~1 hour. Salinity of the extracted



porewater was measured using a refractometer. Porewater was transferred to pre-combusted amber vials (20 mL) for dissolved organic carbon (DOC) analyses.

145 Tidal water sampling was conducted along the salinity gradient (0 to 33) in the Aklan River. For this study, three representative sites were chosen, the upstream of the Aklan River and outer-shore as potential endmember sources of SOM, and the river channel very close to BS site during the high tide. More details of water sampling techniques can be found in the supplementary material. A global tide prediction model (NAO.99b, Matsumoto et al., 2000) was used to correct water depth data to relative elevation at each site from Mean Sea Level or MSL.

150 In the laboratory, pre-weighed wet sediment subsamples were oven dried at 60°C for 48 hours to allow for calculation of dry bulk density (BD) and water content. For sediment subsampling, open mouth plastic syringe was used (2 cm id and 1.5cm length). Bulk density ( $\text{g cm}^{-3}$ ) was determined as the dry sediment weight (g) divided by the initial volume ( $\text{cm}^3$ ). Rest of the wet sediment samples were freeze dried using a Benchtop Freeze Dry System (Labconco). The freeze-dried samples were gently crushed using a mortar and pestle and passed through a 1 mm mesh stainless steel sieve to remove the large gravel (refer as coarse fraction). The sieved samples were stored in tightly capped glass vials  
155 under <40% relative humidity.

### 2.3 Chemical analyses

The dried and homogenized sediment samples were subjected to acid treatment to remove inorganic carbon. Approximately 1 g of dried sample was placed into screw-capped glass tubes (10 mL), and 2.0 N hydrochloric acid (HCl) solution was added to the sediment dropwise until all the carbonate was converted to  $\text{CO}_2$ . After centrifugation  
160 for 15 min at 2000 rpm, followed by washing with deionized water and decantation, the final residue in the tube was dried at 60 °C overnight. Once cooled, the dried samples were weighed into a tin capsule (10 x 10 mm), folded and kept temporarily in a 48-well microtiter plate until analysis.

The concentrations and isotope ratios of OC and total nitrogen (TN) in the treated samples were determined simultaneously by EA-IRMS (FLASH 2000/Conflo IV/DELTA V Advantage, ThermoFisher Scientific, Bremen,  
165 Germany). Two standard materials of different  $\delta^{13}\text{C}$  (-26.4 to -19.6‰) and  $\delta^{15}\text{N}$  (2.5 to 5‰) values (SI Science Ltd., Saitama, Japan) were used for calibration. The measured isotope ratios were represented using conventional  $\delta$ -notation ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ , in ‰) with Vienna Pee-Dee Belemnite and atmospheric  $\text{N}_2$  as the reference materials. The instrumental analytical precision was normally within  $\pm 1\%$  for the OC and TN concentrations and  $\pm 0.1\%$  for  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ .

Measurement of specific surface area (SSA) in sediment was performed by the multipoint Brunauer–Emmett–Teller  
170 (BET) method based on  $\text{N}_2$  gas adsorption under reduced pressure (details refers to Miyajima et al., 2017). The dried and homogenized sediment samples were heated at 350 °C for 12 h followed by calculation of weight loss on heating after 1 hour. Between 0.5 g and 2.0 g of the treated samples were weighed into glass flasks and desiccated in vacuo at 350 °C for 3 h. Immediately after cooling, a multipoint BET measurement was performed with  $\text{N}_2$  (purity > 99.99%) as the adsorbate using the BELSORP mini II (MicrotracBEL, Osaka, Japan) surface area analyzer. The slope of the  
175 BET plot in the linear region was used for estimating the SSA.



## 2.4 Data analyses

The results of the OC and TN concentrations, and the SSA were expressed as  $\mu\text{mol}$  and  $\text{m}^2$  per unit dry weight of bulk sediment, respectively. Carbon stock ( $\text{Mg C ha}^{-1}$ , top 50 cm, referred to as  $S$ ) of each core at 5 sites were calculated on seasonal basis as

$$180 \quad S = \sum_{i=0}^n C_n \times \rho_n \times l \dots \dots \dots (1)$$

Where  $C_n$  is C concentration (mass %) and  $\rho_n$  is the bulk density ( $\text{g cm}^{-3}$ ) of the sample, and  $l$  (cm) is length of sample section. The amount of OC preserved per unit surface area of sediment particles is referred to as OC loading (OC/SSA in  $\mu\text{mol m}^{-2}$ , Mayer 1994).

185 In a chrono sequence study, it is common to apply linear regression model between mangrove age and C stock and derive slope, i.e., carbon accumulation rate (CAR) (Alongi, 2004; Walcker et al., 2018) or individually following the formula such as  $\text{CAR} = [(\text{C stock of the stand}) - (\text{C stock of the previous stand})] / [(\text{age of the stand}) - (\text{age of the previous stand})]$  (Marchand et al., 2017). However, our results showed best-fit with exponential function for the relationship.

$$\text{OC stock}(\text{mol/m}^2) = a \cdot e^{b \cdot \text{yr}} \dots \dots \dots (2)$$

190 where  $a$  and  $b$  are constants determined from the field data as 100.34 and 0.058, respectively, and  $\text{yr}$  is the age of mangrove stands (years). Here we assume that with exponential increase of plant above ground mass with early mangrove growth, below ground root mass also increases that contribute significantly to OC accumulation with early mangrove development. However, because exact maturity stage of these mangroves is unknown, typical logistic curve equation that is otherwise used for the matured forest, was not applied, instead best-fitted exponential trend was used.

195 Based on exponential model, slope was derived for the individual sample following the equation:

$$\text{Slope or CAR} \left( \frac{\text{mol}}{\text{m}^2 \cdot \text{yr}} \right) = \left( \frac{d\text{OC}}{dt} \right) = a \cdot b \cdot e^{b \cdot \text{yr}} \dots \dots \dots (3)$$

Accordingly, CAR for the age class of mangroves (0.25, 5, 10, 15, 20, 25 and 30 year) was calculated and fitted in the exponential curve and compared with the slope derived from the linear regression model.

200 To evaluate the effects of sampling Depth, Season and stand Types (i.e., chronology based) on  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and C:N ratio of SOM, the general additive linear mixed model (GAM) was constructed with a *gam* function in a R package (mgcv). Depth, Season, Types and an interaction between Season and Types were treated as fixed effects where Depth was incorporated into the model. Using the results of *gam*, ANOVA was used to evaluate the significance of each variable with a significance level at  $p = 0.05$ .

205 For subsequent provenance analysis, a binary source mixing model was applied (Parnell and Inger, 2016) based on mean values of  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and C:N ratio of sediment samples from different depth in each core. The samples obtained in different seasons were treated as independent samples. In total, seven mean values were used for the analysis of Bayesian mix model using multiple R packages (RGtk2, MixSIAR, splanc). The endmember sources of OM chosen



for this study were green leaf of *Rhizophora apiculata*, POM (particulate organic matter) of marine and river water, and microphytobenthos (MPB). Among these endmembers except for POM, the  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and OC:TN ratio for green  
210 leaf and MPB were obtained from the literature (refers to Table S1). Although OC:TN were shown to be largely  
variable between green (~30) and yellow/senescent leaves on sediment floor (~50) of the *Rhizophora apiculata*, their  
 $\delta^{13}\text{C}$  values were the same (-28.5‰) (Nordhaus et al., 2017). For similar reason mangrove root is also not considered,  
instead leaf singularly presents as mangrove-derived OC source. Hence it is reasonable for this study to choose green  
leaves and avoid redundant increase of endmembers for the model. Microphytobenthos located on tidal flat between  
215 the interface of sea and land can also be variable depending on  $\delta^{13}\text{C}$  of dissolved inorganic carbon (photosynthesis  
substrate for MPB). This is similar to the riverine settings in French Guiana ('mixed MPB' -20.9‰, Ray et al., 2018),  
and contrast to oceanic setting in Red Sea ('pure MPB' -17.9‰, Shahraki et al., 2014). Number of endmembers were  
chosen carefully keeping small standard deviation of  $\delta^{13}\text{C}$  and OC:TN within each endmember and clearly separated  
with significant differences from each other (ANOVA,  $p < 0.05$ ). We refrained from considering a large number of  
220 endmembers, and omitted less important endmembers or treated collectively some endmembers whose isotopic and  
elemental compositions are similar (e.g. plant parts such as leaf, root, litter). Since there was no seagrass recorded at  
or around the sites, its potential endmember contribution to the mangrove SOM was ignored. On the contrary, although  
the isotopic composition of MPB and marine POM were not very different, we selected MPB as endmember due to  
its visible presence on the exposed tidal flat (BS2) during the low tide.

### 225 3. Results

Maximum elevation of 0.45 m from the MSL was measured at the mature mangrove (MM) site, and minimum of -  
1.3 m was recorded for the bare sediment (BS) (Fig. 2). Mean porewater salinity, sediment temperature and pH did  
not vary significantly between the various stages of mangrove development (Fig. 2). Sediment thickness was higher  
in mangrove sediment (140-265cm) than the tidal flat (<100 cm). Sediment bulk density at the top 50 cm increased  
230 with decreasing mangrove age tending to be maximum at BS (0.6 to 1.3 g/cm<sup>3</sup>,  $n = 93$ , Table 1). Specific surface area  
of the sediments varied from 9.8 to 21.2 g/m<sup>2</sup> ( $n = 19$ ) with lowest value for the coarser bare sediment. Fraction of  
large gravels (>1 mm mesh size) tend to be higher going seaward. A consistent increment in sediment OC  
concentration (top 50 cm,  $n = 93$ ), TOC:TN molar ratio ( $n = 92$ ), and porewater DOC ( $n = 64$ ) was observed with the  
mangrove evolution (BS: 0.06  $\mu\text{mol/g}$ , 21.9 mol/mol, and 292  $\mu\text{mol L}^{-1}$ ; at MM: 3.4  $\mu\text{mol g}^{-1}$ , 19.7 mol/mol, and 2150  
235  $\mu\text{mol L}^{-1}$ , respectively). The mean value of bulk  $\delta^{13}\text{C}$  became more negative with mangrove age ( $\delta^{13}\text{C} = -25.07\text{‰}$  at  
BS and -28.9‰ at MM,  $n = 93$ ). The mean  $\delta^{15}\text{N}$  varied from -1.15 at the BS to 1.06 ‰ at the adult mangrove (AM)  
site ( $n = 90$ ). Organic carbon (OC) stock was determined to be maximum at the mature stand (77.4 Mg C/ha) and  
lowest at the bare sediment (3.13 Mg C/ha). Similar to the trend of OC stock, OC loading varied widely between 4  
and 380  $\mu\text{mol/m}^2$  ( $n = 19$ ), with mangrove sites having higher OC loading (mean: 152  $\mu\text{mol/m}^2$ ) than the bare sediment  
240 (25  $\mu\text{mol/m}^2$ ).

Vertical profile of sediment OC and  $\delta^{13}\text{C}$ , specific surface area (SSA) and OC loading, and porewater DOC on seasonal  
basis are shown in Figs. 3–5. Sediment surface values (0-10 cm) of porewater salinity, pH and ORP changed  
significantly with mangrove ages in the wet season, ( $p < 0.05$ , Fig. S1), except for porewater salinity at bare sediment



245 during the dry season. The deeper layers of each sediment core (>10 cm) did not exhibit much variability in the  
properties. Minimum pH was recorded at 10cm depth of the mature mangroves (5.23). Vertical profiles of TOC and  
 $\delta^{13}\text{C}$  showed wide variations than the physico-chemical properties (Fig. 3a-d). Notable peaks for TOC were observed  
in mature mangrove (6 cm and 25 cm in dry and wet season, respectively) and adult mangrove (20 cm; dry season).  
Total organic carbon and  $\delta^{13}\text{C}$  in bare sediment changed very little with depth especially in the dry season. Significant  
differences in OC/TN molar ratios between sediment from different depth of MM and other sites were observed (Fig.  
250 3e,f). At the non-vegetated site (BS), the ratios varied slightly with seasonal changes. The vertical profile of SSA  
based on the values at three specific depths of each core showed a mild decreasing trend from the surface to deepest  
layer (50 cm) in both dry and wet season (Fig. 4a,b). The depth-specific values of SSA were always higher at the adult  
mangrove sites than the other two (except at 45-50 cm in dry season). Similar to SSA, OC loading also decreased with  
core depth and mature mangrove exhibited maximum loading in both seasons (Fig. 4c,d). All stages of mangrove  
255 development showed an overall increase of DOC concentration with depth regardless of the seasons (Fig. 5a,b). DOC  
concentrations of deeper layers reached up to four to five times higher than those of the upper sediment in the wet  
season and two to three times higher in the dry season. For example, our data demonstrated a change from 600 to  
around 4000  $\mu\text{mol L}^{-1}$  for the adult mangrove in both dry and wet season. A wide range in DOC of 128 to 920  $\mu\text{mol}$   
 $\text{L}^{-1}$  was also noticed for the non-vegetated bare sediment site.

260 On the basis of generalized additive model (GAM), results of ANOVA for the main parameters showed significant  
dependence of TOC over mangrove types ( $F = 46.37$ ,  $p < 0.001$ ), and depth ( $F = 9.80$ ,  $p < 0.001$ ) (Table 1). Porewater  
DOC varied significantly with mangrove types ( $F = 4.42$ ,  $p < 0.005$ ), similar to the  $\delta^{13}\text{C}$  ( $F = 28.3$ ,  $p < 0.001$ ) and  $\delta^{15}\text{N}$   
( $F = 3.92$ ,  $p < 0.05$ ). The coefficients in each parameter based on the GAM for TOC, DOC and  $\delta^{13}\text{C}$  are given in Table  
S2 and their changes with depth as smooth term are shown in Fig. S2.

265 The OC/TN and  $\delta^{13}\text{C}$  of the five mangrove types were plotted with the four end-member sources (River POM, Marine  
POM, Leaf and MPB) to rectify sources of SOM (Fig. 6), and subsequently calculate end-member contribution to  
each type (Fig. 7, Table S3). Except for MPB and marine POM, the relative contributions of mangrove leaf and river  
POM were significantly different for each site (one-way ANOVA, p-value mangrove leaf <0.05, p-value river POM  
<0.05). At the bare sediment and pioneer mangrove sites, river POM dominated the SOM pool with maximum  
270 contribution of 80% and 75%, respectively, whereas at the young and mature stands, mangrove leaf was the main  
potential contributor (73 and 82%, respectively). On the other hand, SOM remained more as a mixture of river POM  
and leaf material (~60%) at the adult mangrove sites. The contribution of benthic and pelagic algae as SOM sources  
was not very significant for the entire mangrove sediment in the area studied.

275 In the water part, results from three locations (upstream-channel-offshore) showed salinity changing from 0 to 33, pH  
increasing from 7.6 to 8.1, and DO (%) increasing from 89 to 105% to the downstream (Table S4). Surface water POC  
was 4–5 fold lower than the DOC in the upstream and channel water (~20  $\mu\text{mol/L}$  versus 90  $\mu\text{mol/L}$ ), and lowest in  
offshore water (10  $\mu\text{mol/L}$ ). The  $\delta^{13}\text{C}$ -POC was highest in offshore water (-22.8‰) and lowest upstream (-25.9‰).

#### 4. Discussion



#### 4.1 Sediment condition and organic matter

280 The results of dry bulk densities and granulometry (coarse fraction, SSA,) of the tidal flat and mangroves in Bakhawan  
Ecopark indicate that soils are relatively homogeneous with fine-grained fractions relatively prevalent towards the  
upper older mangrove region (mainly towards AM), while towards the shore, coarse-grained sands are more common  
on the tidal flat and younger mangroves (BS and YM, PM) (Fig. 2). Bulk densities (BD) at the sampling sites (0.3 to  
1.3 g cm<sup>-3</sup>) were higher than the reported BD values across mangrove soils of the Indo-Pacific regions (0.20 to 0.92 g  
285 cm<sup>-3</sup>, Donato et al., 2011), with sand fractions dominating the lower intertidal zone. Specific surface area is primarily  
constrained by grain size and its vertical profile is presumably related to finer upward trend of sediment grain (Fig.  
4a, c). The latter might have been a result of sediment-stabilizing function of mangroves or influenced by some land-  
use change in recent years (e.g., land use change can increase clay content via deposition of pollen, Shen et al., 2020)  
Furthermore, carbonate bearing minerals can influence SSA more on the tidal flat than the organic rich mangrove  
290 sediment. Among the older stands (MM and AM), higher SSA and sediment thickness, and low BD at the adult stands  
suggest dominance of finer grained clay material because of the long-term presence of mangrove itself, and the narrow  
sandbar halfway down the Ecopark (Fig. 1) that may close off the older mangroves from higher wave energy and shift  
deposits of finer grains. Total organic carbon is higher at the older sites than the younger ones due to such fine-grained  
(silt + clay) sediments that tend to have higher TOC than the coarse sands (Canfield, 1994). Fine-grained silt has larger  
295 SSA that create higher capacity to adsorb OM (Mayer, 1994).

Among the physicochemical properties, lower porewater salinity at the BS than the mangrove sites indicate greater  
dilution from direct input of river water, which become less with increasing elevation level inside the forest floor (Fig.  
2). On temporal scale, overall low porewater salinity during the wet season is more likely linked to rainwater dilution  
(350–450 mm in September, 75–150 mm in February, JRA-55 Reanalysis) and elevated groundwater level. Because  
300 of shading effect, surface sediment temperature at the mangrove vegetated sites was lower than the sun-exposed BS  
or PM. Lowest recorded ORP at BS strongly suggests that exchange of porewater with overlying water is limited, and  
this may have implication on the interpretation of porewater chemistry (i.e., DOC, section 4.3).

It has been observed that mangrove sediments in the river-dominated estuary accumulate larger proportion of the  
mangrove-derived OM than those in the tide-dominated estuaries/oceanic mangroves dominated by marine algae (e.g.,  
305 Indonesian mangroves, Kusumaningtyas et al., 2019; Latin American, Gontharet et al., 2014; Middle-East, Ray and  
Shahraki, 2016). In the microtidal riverine setting of the Bakhawan Ecopark, OM input from land sources is more  
dominant than the marine sources at the mangrove sites (Fig. 6). End-member mixing model suggests that there is a  
clear gradient with respect to the relative proportion of OM sources along the tidal flat-mangrove continuum (Fig. 7).

Sediments from cores at bare sediment and pioneering mangrove sites show the predominance of OM probably derived  
310 from fluvial transport of eroded organic material within the catchment. Upon transport into the coastal area, fine  
sediment and POM accumulate in calm shallow water channel at the topographically lowest elevation zone, (Fig. 2)  
and consequently the longer inundation period facilitates sedimentation and deposition of the suspended matter.  
Similar values of  $\delta^{13}\text{C}$  in surface water POC upstream ( $-25.9\%$ , Table S3) and sediment OC at the bare sediment and  
pioneering mangroves (mean  $-26\%$ ) support their upstream allochthonous sources. A sizeable contribution from MPB



315 may not be ruled out at bare sediment despite its minimal contribution from the mixing model (Fig. 7). In a companion  
study, other than the visible evidence of green algal patches, we measured day-time CO<sub>2</sub> uptake flux on the tidal flat,  
which contrasts with emission flux at the forested sites (tidal flat: -4 to -2 mmol m<sup>-2</sup> hr<sup>-1</sup>, mangrove: 5–12 mmol m<sup>-2</sup>  
hr<sup>-1</sup>, Ray et al., unpublished). Strikingly low OC:TN ratio at the surface sediment and minimum δ<sup>15</sup>N suggests presence  
320 of N<sub>2</sub> fixing bacteria on the tidal flat during exposed tide condition. The slightly higher mangrove contribution to SOM  
in pioneering mangroves (up to ~54%) clearly indicates additional OM input from the small growing plants along the  
channel.

At the topographically higher mangrove sites, greater contributions of autochthonous sources (i.e., mangrove plant  
materials) are correlated with lower δ<sup>13</sup>C and higher OC/TN (>12). This is favored by relatively high bed elevation  
and decrease in submersion time promoting retention of detrital OM in the sediment layer (shown as leaf OM, Fig. 7).  
325 Although such evidence of greater plant input to SOM at interior mangrove sites compared to mudflats are not new  
for the intact forests (Marchand et al., 2003; Sanders et al., 2010; Matos et al., 2020), it is rare for restored mangroves  
considering an extended gradient from mudflat to mangrove appearance (except in Vietnam, Van Hieu et al., 2017).

Terrestrial C<sub>3</sub> vascular plants, like mangrove plant organ have C/N ratios of around 12 or higher (Prahl et al., 1980)  
as it is composed predominantly of lignin and cellulose, which are nitrogen poor. The significant positive correlation  
330 between TOC and TN (r<sup>2</sup> = 0.96 at BS, PM, YM, and AM; r<sup>2</sup> = 0.42 at MM, figure not shown) in the sediments  
indicate that the C and N in the samples are predominantly associated with the organic pool. This suggests the  
protective role of early mangrove recruitment on sedimentary organic matter as seen in European coastal wetland  
(Jiménez-Arias et al., 2020). It is noteworthy that such correlation is relatively poor at the mature stands. Although  
the exact reason for this is unknown, the abundance of benthic animals during another complimentary experiment  
335 (burrow density at MM: 150±55, YM: 70±62, BS: 5±2 individual/m<sup>2</sup>, Ray et al., unpublished) might suggest intense  
bioturbation and sediment remobilization.

#### 4.2 Vertical profile of organic carbon

The difference in the sediment physicochemical and OC profile at the upper 50 cm of the layers may result from  
multiple factors that promote OM decomposition, such as immediate exposure of litter in surface layers (e.g., managed  
340 *Rhizophora* in Malaysia, Ashton et al., 1999), while coarse and fine roots contribute to carbon and nutrients at  
shallower depth (reported down to 52 cm for 27-yrs planted *Rhizophora* in Vietnam, Arnaud et al., 2021). Multiple  
mid-layer peaks of TOC are sometimes observed which presumably reflects the influence of root biomass (Fig. 3).  
This study showed that root activity within the sediment column is essentially dominant when comparing vegetated  
sites with bare sediment. Assuming a mixture of algae (marine POM plus MPB, δ<sup>13</sup>C = -21.07‰), and mangrove  
345 root/plant organ (δ<sup>13</sup>C = -28‰) as two major end-member sources of the sediment OM pool, provenance analysis  
confirms that roots contribute maximum at the old stands (75-100%) and minimum at BS and PM (52-72%)  
corroborating to the TOC peaks observed at different depths (particularly between 10-25cm, Fig. 3a,b).

Organic carbon profile was almost uniform in both seasons suggesting a dominant OM source prevailed down the  
cores. Root exudates like sugars, amino acids are suggested to be the main carbon source for the localized microbes



350 (Bouillon and Boschker 2006; Jiang et al., 2017). The role of burrowing crabs as carbon sink has also been reported  
at deeper soil layers of *Avicennia* stands in Kenya (40–80 cm, Andreetta et al., 2013) where burrowing sesarmids  
provide a continuous supply of fresh organic matter down the profile. At low tide, presence of water with low oxygen  
saturation, covering the bottom of the burrows avoids oxidation by creating an extension of sediment-air interface  
favoring greater OC accumulation (Kristensen, 2004; Smith III et al., 1991). The same study in Kenya reported greatest  
355 OC concentration where crab population were maximum., otherwise hypo- or anoxic, and create an extension of costal  
marshes sediment-air interface favoring greater OC accumulation. In this study, bioturbation might play an important  
role in carbon accumulation at the sediment horizon of the older stands where higher benthic communities were found  
compared to the tidal flat and younger mangrove sites.

The depth-wise relationships between sediment OC and porewater DOC are less obvious (Fig. 3, 5). Porewater DOC  
360 varies disproportionately with OC. The distinct feature of vertical DOC profile is the non-uniform distribution of  
concentration with mangrove age in contrast with the relatively uniform profile at bare sediment. Porewater DOC in  
non-vegetated sediment is known to be primarily controlled by oxygen availability and the presence of  
microphytobenthos that could drive porewater dynamics via OC leaching compared to mangrove sediments. At the  
vegetated sites, fluorescence and hydrophobic DOC of high molecular weight is known to drastically increase in  
365 anoxic coastal porewaters (Komada et al., 2004; Marchand et al., 2006). At the vegetated sites, porewater DOC  
concentrations shows higher concentrations with depth that is most likely a reflection of the net effect of diagenesis,  
subsurface transport and partial control via root uptake and release. Though salinity profile is not exactly like DOC,  
yet the salinity peaks at the sub surface depth, thereby a uniform pattern followed at AM and MM. This may lead us  
to hypothesize that water absorption by roots at the upper sediment may be augmented by the presence of radial  
370 mangrove roots leading to an increase of salinity at some sites and gravitational percolation of salt water and DOC to  
greater depths. However, unlike porewater salinity that is lower in wet season than dry season due to rainwater dilution,  
DOC did not vary seasonally at adult mangrove and mature mangrove sites suggesting perennial source and retention  
in the sediment. The porewater profile of salinity and DOC in mangrove sediments is very rare in the literature. One  
such comprehensive dataset by Marchand et al. (2006) in French Guiana reported similar findings of higher DOC at  
375 greater depth with mangrove ageing but no direct correlation with other co-variables. Like shown many years ago by  
Marchand et al. (2006), the influence of seasonal mangrove productivity and root activity on DOC vertical profile  
may be evident also for this study, however, detailed research is necessary to understand such relationship.

#### 4.3. Increase of organic carbon with mangrove development

The average TOC concentration in the top 50 cm mangrove sediment (PM to MM) in Bakhawan Ecopark is lower  
380  $(2.5 \pm 1.8 \mu\text{mol mg}^{-1})$  than the Indo-Pacific regions  $(9.9 \pm 5.2 \mu\text{mol mg}^{-1})$ , Donato et al., 2011), but comparable with the  
global mean  $(1.7 \mu\text{mol mg}^{-1})$ , Kristensen et al., 2008) and other restored mangroves in SE Asia (e.g.,  $2.2 \pm 0.05 \mu\text{mol}$   
 $\text{mg}^{-1}$  in Vietnam, Dung et al., 2016) and China  $(4.2 \pm 0.3 \mu\text{mol mg}^{-1})$ , Nam et al., 2016). Absence of peat organic layer  
at the sampled sites and fast decomposition observed in separate  $\text{CO}_2$  emission measurement (benthic emission:  $8$   
 $\text{mmol m}^{-2} \text{hr}^{-1}$  at upper limit of global range  $0.25$  to  $10.4 \text{mmol m}^{-2} \text{hr}^{-1}$ , Bouillon et al., 2008, Ray et al., unpublished)  
385 are the possible reasons behind such low to moderate OC in sediment. Besides that, rapid flushing out of POC favored



by the low-lying gentle elevation could account for the OC-poor state of the system. Isotope evidence of POC further indicate that it was sourced from the eroded mangrove soil composed of litter debris (mean around  $-25.0\%$  for salinity 0–25, Table S4) coming from the upstream and mangrove sites that flushed away via the channel to offshore ( $-25.0\%$  at salinity 33) at ebb time.

390 The present result shows that the development of restored mangrove forests could improve sediment OC as indicated by a clear progression in sediment TOC among the planted mangroves of different developmental stages leading towards soil maturity. The accumulation of OC in the mangrove sediment may be attributed to the increases in belowground root expansion with stand age (Salmo et al., 2014). Because of restricted water exchange with the seawater, mature *Rhizophora* stands accumulate higher mangrove derived OM (leaf litter and decomposing fine roots)

395 than the young and pioneering stand, therefore shows higher TOC. For the adult mangrove stands (AM) despite their moderate biomass, oxic conditions (positive ORP) may have favored OM decomposition in the wet season resulting into lower TOC than at MM. Such non-linear correlation of OC with mangrove chrono-sequence is in line with other works in the restored (Lunstrum and Chen, 2014; Van Hieu et al., 2017) and intact forests (Lovelock et al., 2010, Marchand et al., 2017).

400 Like TOC concentration, OC stock in sediment gradually increased with chrono-sequence which is consistent with other recent studies on mangrove plantations (Lunstrum and Chen, 2014; Van Hieu et al., 2017; Wang et al., 2021). Salmo et al., (2013) previously reported higher above ground biomass (AGB) in 17-yr old *Rhizophora* than the 12-yr old stands ( $101.8$  versus  $51.4$  Mg ha<sup>-1</sup>) at the Bakhawan Ecopark. Fine root production has also been found to increase with AGB and contribute to sediment OC stock more in mature mangroves (Zhang et al., 2021). On a global basis,

405 OC stock is mostly reported for sediment cores of 1 m depth. If core depth is normalized to 50 cm, our results still give comparable estimates ( $3$  to  $77$  Mg C ha<sup>-1</sup>) to the limited assessments of restored mangrove park, such as in north central Vietnam ( $54$  to  $84$  Mg C ha<sup>-1</sup>, 0 to 27-yr of *Kandelia obovats*; Van Hieu et al., 2017), Pichavaram ( $41$  to  $94$  Mg C ha<sup>-1</sup>, 12 to 21-yr of *Rhizophora spp.* (Gnanamoorthy et al., 2019) and Bhitarkanika Conservation in India, ( $38$  Mg C ha<sup>-1</sup>, 5-yr *Kandelia candel*, Bhomia et al., 2016). However, our stock estimate is lower than the organic soil of

410 oceanic setting in Sulawesi, Indonesia (c.a.  $150$ - $300$  Mg C ha<sup>-1</sup>, more than 10 years of *R. apiculata*, Cameron et al., 2019). Sediment OC stock is largely dependent on vegetation biomass and extent of litter input. Salmo et al., (2014) reported that 17- and 18-year old *Rhizophora* stands at the Bakhawan Ecopark had 30–40 % lower AGB compared with the natural mangroves ( $150$  ton ha<sup>-1</sup>) while 50-year old stands had similar AGB ( $132$  ton ha<sup>-1</sup>) with natural mangroves. Another reason might be the river dominated mangrove settings which are known to transport high allochthonous input and deposit mineral sediment that dilutes OM and lowers OC stocks than the marine settings

415 where mangrove-derived OM increase carbon stock in the sediment (Jennerjahn, 2020). Another reason could be sediment reworking during restoration or plantation work that can mobilize OC at least from the top 10 cm of the sediment. The rapid turnover may lead to reduction in OC stock. From a more general perspective, sediment OC stock depends on the distribution of fine roots (Noguchi et al., 2020), and efficient redox-dependent decay processes like

420 sulfate reduction that promote OM preservation and increase OC stock in older stands isolated from the sea (Lambis et al., 2008).



Increase in porewater DOC concentrations observed from tidal flat to pioneer mangrove then to older mangrove seems to reflect the increasing pattern of bulk OC (Fig. 2). With greater biomass, (mean tree height: PM <1m, YM 3-4m, AM 6-8m, MM 10-15m, data not shown), sediment TOC and porewater DOC concentration successively increased.

425 A similar trend for bulk and dissolved OC was observed in the naturally growing *Avicennia* dominated mangroves in the Amazonian coastline (Marchand et al., 2006). With progression of chrono-sequence, we may infer that the higher the biomass, the higher the TOC, and the higher the DOC concentrations in the shallow sediments.

Organic carbon preservation in marine sediments can be influenced by the physical factors such as association of OC with the surface of sediment mineral particles, also known as OC loading (Kiel and Mayer, 2014). Sediment with high

430 SSA tend to be rich in clay, iron and aluminum and store greater amounts of OM than low-SSA soils due to intimate organo-mineral association (Mayer, 1994). Therefore, OC loading can be considered as a proxy for blue carbon preservation and supply in marine system, although there has been no account of such result for mangroves worldwide. Our estimates of OC loading for the mangrove sediment and tidal flats ( $152$  and  $25 \mu\text{mol C m}^{-2}$ ) are within the range of continental margin sediment ( $40$ - $80 \mu\text{mol C m}^{-2}$ , Mayer, 1994), vegetated marine sediment ( $56$ - $67 \mu\text{mol C m}^{-2}$ ,

435 Miyajima and Hamaguchi, 2017), and floodplains ( $16$ - $42 \mu\text{mol C m}^{-2}$ , Goni et al., 2014), but much lower than in the high altitude soils where mineral phases are fully covered by rich-OM with high OC% ( $300$ - $780 \mu\text{mol C m}^{-2}$ , 5 to 10%; Wagai et al., 2009). At the Bakhawan Ecopark, variability of OC loading between mangrove sediment and tidal flat can be explained by the difference in spatial extent of individual sampled site. For example, when SSA was greater than  $15 \text{ m}^2 \text{ g}^{-1}$  (at AM and MM), significant negative correlation was observed between OC and SSA ( $r^2 = 0.95$ ,  $p$

440  $< 0.01$ ,  $n = 7$ , figure not shown) indicating that net accumulation rate of OC in these sites are not dependent on mineral particles but on mangrove-derived supply of OM. Whereas at BS with mean SSA  $< 15 \text{ m}^2 \text{ g}^{-1}$ , a quasi-significant positive relationship between these two variables suggests the possible role of physical sorption of OC in the riverine sediment mineral matrices for stabilization and sequestration of organic carbon ( $r^2 = 0.50$ ,  $n = 6$ ). Therefore, sediment in the mangroves does not share a common OC sequestration mechanism as with the pelagic continental shelf and

445 seagrass sediments.

#### 4.4 Accumulation of organic carbon

##### 4.4.1 Relevance of chrono-sequence approach

Several studies have relied on direct measurement of carbon accumulation rates (CAR) by combining sedimentary C content and soil accumulation rates estimated from radioisotopes,  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  or natural markers like volcanic

450 ashes (Sanders et al., 2010). These CAR estimations are based on the assumption that sediment and OC accumulation are in a steady state during the period of accumulation/deposition. However, the "indirect way" or the chrono-sequential theory assumes that the temporal variations in soil properties in different aged sites fall into the same time trajectory of OC accumulation. This assumption requires a condition that these different sites had experienced similar driving factors of OC accumulation processes following tree growth after the restoration. While hydrological

455 processes such as hydroperiod and tidal regime are considered to be one of the important drivers of OC accumulation, it is considered that these conditions do not largely vary among the sites and have been relatively stable over the time window in concern ( $\sim 30$  years after restoration) given the same level of ground elevations which locate in the same



forest (Fig. 2). Also, given the significant fraction of mangrove contribution to SOC (Fig. 7), the influence of C inputs from external systems may not be a significant factor in shaping the OC accumulation trajectories in the sites. Only  
460 the vegetation structures vary significantly among sites that may characterize the evolution of OC stock with forest age in our study site, Bakhawan Ecopark (Fig. 8). In addition, we consider that the AGB and BGB development in the sites follow a similar trajectory given the same level of soil salinity (Fig. 2) – one of the most important regulators of mangrove growth, and the same plantation spacing and species. Therefore, it is reasonable to consider that the OC accumulations in the different aged sites also follow the similar time trajectory, and thus Eq. (3) can be applied to  
465 estimate CAR in our study site.

#### 4.4.2 Carbon sink capacity

Buried OC in the sediment strongly depends on many environmental conditions such as mangrove forest productivity, deforestation and degradation rate, sediment accretion, topography, tidal regime, and bioturbation activities (Alongi, 2014, Pérez et al., 2018). Significant exponential and/or linear increase of OC stock with early mangrove age (till YM)  
470 probably suggests that primary productivity might be key controlling factor for C accumulation (Fig. 8a). However, the correlation between the standing stock of sediment OC with ageing at the adult/matured stands are essentially not very significant, indicating that the size of the sediment OC pool at these two sites are not determined simply by the primary production of the vegetation but is also constrained significantly by some other biological or geophysical factors. Rates of OC accumulation defined by their respective slopes in the exponential curve (BS to MM) and linear  
475 curve (YM to MM) at individual site of 50 cm depth ( $5.9 - 33 \text{ mol m}^{-2} \text{ yr}^{-1}$ , and  $15.93 \text{ mol m}^{-2} \text{ yr}^{-1}$ , respectively, Fig. 8a,b) are well within the ranges by chrono-sequential analysis of 1m cores in restored ( $12.7 \text{ mol m}^{-2} \text{ yr}^{-1}$ , Lunstrum and Chen, 2014), intact ( $6 \text{ to } 40 \text{ mol m}^{-2} \text{ yr}^{-1}$ , Marchand et al., 2017), or encroached mangroves ( $19 \text{ mol m}^{-2} \text{ yr}^{-1}$ , Kelleway et al., 2016). System specific variabilities such as sedimentation rate, decomposition rate, rate of litterfall and root production may cause such differences in CAR among the reported mangroves. It is noted that OC% in other  
480 mangrove soils were on average 2 times higher than the Bakhawan sites due to high terrestrial input to this forest driven by historical land use changes.

#### 4.5 Implication of blue carbon chrono-sequence

There is a crucial need to improve scientific understanding on blue C dynamics and to develop an appropriate framework for blue C assessment and monitoring mechanisms for future policy development. Achieving improved  
485 scientific understanding on C sources, stocks and to monitor the changes in accumulation rate at the early development stage and adult stages would require practical tools and guidance to enable the conduct of proper C analyses. In this study, supply of OM from the mangrove vegetation, benthic algae and upstream sediment transport are recognized as controls of blue C at the mangrove sites and tidal flat. Impression of more mangrove-derived C input is evident with mangrove development. Such apportionment of OC sources at different mangrove stand ages should be useful to  
490 improve our future knowledge on origin of blue C in REDD+ accountings that is yet to register sedimentary OC within the reduced C emission scheme despite sediment being recognized as largest C pool in the mangrove ecosystem (Duarte et al., 2013). A prior knowledge on sources and character of OC (generally refractory or mangrove-derived and labile or algal-derived) would be beneficial for fostering mangrove plantation program. Greater mangrove-derived



OC accumulation with ageing at the Bakhawan Ecopark might suggest long term storage of the refractory fraction,  
495 hence an effective return to REDD+ strategy. Another perspective is that any attempt of quantifying OC stock and  
accumulation rate following a plantation program should be well recorded in the relevant carbon accounting programs.  
For example, the Verified Carbon Standard Methodology ([www.verra.org](http://www.verra.org)) existing for mangrove restoration projects  
that is certified under the Cleaner Development Mechanism (CDM) program of the United Nations Framework  
Convention on Climate Change (UNFCCC, 2011) assumes CAR for 0–20 years mangrove as  $4 \text{ mol m}^{-2} \text{ y}^{-1}$  after  
500 plantation (Lunstrum and Chen, 2014), which is 4 times lower than our chrono-sequence based estimate ( $\sim 16 \text{ mol m}^{-2} \text{ y}^{-1}$ , linear slope for YM to MM stands). In IPCC 2013 supplementary (Tier 1, chapter 1), default  $\text{CO}_2$  emission factor  
from tidal flat was set to be 0, while it was  $-14 \text{ mol m}^{-2} \text{ y}^{-1}$  (negative means accumulation) for the planted/rehabilitated  
mangroves, latter is in line with the older mangroves at Bakhawan Ecopark, but not with each developmental stage.  
Such assumption may lead to severely underestimated actual blue carbon sink capacity of the mangroves and  
505 consequently carbon emission value. Therefore, a differential yet steady trend of blue C potential based on short-term  
chrono-sequence can help define the utility of a mangrove restoration efforts.

## 5. Conclusion

This study is a first attempt to apply a chrono sequence (or space-for-time substitution) approach to evaluate the  
distribution and accumulation rate of carbon in a 30 year (max age) restored mangrove forest. From this study, it is  
510 clearly seen that mangrove tree development coincided with sediment OC concentration and accumulation, hence  
mangrove plantations are expected to accelerate OC sequestration at early plantation stage. Source apportionment of  
sedimentary OM suggests higher contribution of mangrove vegetation upland and riverine POM plus benthic algae  
down the tidal flat. The accumulation of OC in the sediment may be attributed to the increases in belowground root  
expansion with stand age. This chrono-sequence based estimates of OC stock and accumulation rate can be useful  
515 reference for setting up carbon accounting in the *Rhizophora*-dominated mangrove restoration projects.

## Acknowledgement

We are grateful to the Japan International Cooperation Agency (JICA) and Japan Science and Technology Agency  
(JST) through the Science and Technology Research Partnership for Sustainable Development Program (SATREPS)  
520 for financially supporting the Project “Comprehensive Assessment and Conservation of Blue Carbon Ecosystems and  
their Services in the Coral Triangle (BlueCARES)”. We thank Jesus Abad, John Michael Aguilar, Dominic Bautista,  
Bryan C. Hernandez and Mr. Tsuyoshi Kanda for their assistance during field surveys. We are grateful for the overall  
support given by the University of the Philippines, Diliman and Aklan State University to the project.

## 525 Competing interests

The authors declared no potential conflict of interests.



## References

- 530 Alongi, D.M.: Carbon cycling and storage in mangrove forests, *Ann. Rev. Mar. Sci.* 6, 195–219, doi: 10.1146/annurev-marine-010213-135020, 2014.
- Alongi, D.M.: Global Significance of Mangrove Blue Carbon in Climate Change Mitigation, *Sci.*, 2, 67, doi:10.3390/sci2030067 2020.
- Alongi, D.M., Sasekumar, A., Chong, V.C., Pfitzner, J., Trott, L.A., Tirendi, F., Dixon, P., and Brunskill, G.J.:  
535 Sediment accumulation and organic material flux in a managed mangrove ecosystem: estimates of land–ocean–atmosphere exchange in peninsular Malaysia, *Mar. Geo.*, 208, 383–402, doi:10.1016/j.margeo.2004.04.016, 2004.
- Ashton, E.C., Hogarth, P. J. and Ormond, R.: Breakdown of mangrove leaf litter in a managed mangrove forest in Peninsular Malaysia, *Hydrobiologia*, 413: 77–88, 1999.
- 540 Arnaud, M., Morris, P.J., and Baird, A.J.: Fine root production in a chronosequence of mature reforested mangroves, *New Phytologist*, doi: 10.1111/nph.17480., 2021.
- Andreetta, A., Fusi, M., Cameldi, I., Cimò, F., Carnicelli, S., and Cannicci, S.: Mangrove carbon sink, Do burrowing crabs contribute to sediment carbon storage? Evidence from a Kenyan mangrove system, *J. Sea Res.*, 85: 524–533, doi: 10.1016/j.seares.2013.08.010, 2014.
- 545 Bhomia, R.K., MacKenzie, R.A., Murdiyarsa, D., Sasmito, S.D., and Purbopuspito, J.: Impacts of land use on Indian mangrove forest carbon stocks: implications for conservation and management, *Ecol. Appl.*, 26, 1396e1408, doi: 10.1890/15-2143, 2016
- Brown, R.D., Marotta, H., and Peixoto, R.B.: Hypersaline tidal flats as important “blue carbon” systems: a case study from three ecosystems, *Biogeosciences*. 18, 2527–2538. doi: 10.5194/bg-18-2527-2021, 2021.
- 550 Boto, K.G., and Wellington, J.T.: Soil characteristics and nutrient status in a Northern Australian mangrove forest, *Estuaries*, 7, 61–69, doi: 10.2307/1351957, 1984.
- Bouillon, S., and H. T. S. Boschker.: Bacterial carbon sources in coastal sediments: a cross-system analysis based on stable isotope data of biomarkers, *Biogeosciences*, 3, 175–185, doi: 10.5194/bg-3-175-2006, 2006.
- Cadaweng, E. A. and Aguirre, J. A.N.: Forest from the mud: the Kalibo experience. in: *In Search of Excellence: Exemplary Forest Management in Asia and the Pacific*, edited by: P.B. Durst, C. Brown, H.D. Tacio and M. Ishikawa, Bangkok, Thailand: FAO, 39–48, 2005.
- 555 Cameron, C., Hutley, L.B., Friess, D.A., et al.: Community structure dynamics and carbon stock change of rehabilitated mangrove forests in Sulawesi, Indonesia, *Ecol. Appl.*, 29, e01810, doi: 10.1002/eap.1810, 2019.



- 560 Chen, J., Wang, D., Li, Y., Yu, Z., Chen, S., Hou, X., White, J.R., and Chen, Z.: The carbon stock and sequestration rate in tidal flats from coastal China, *Global Biogeochem. Cy.*, 34(11), p.e2020GB006772, doi: 10.1029/2020GB006772, 2020.
- Donato, D.C., Kauffman, J.B., Murdiyarso, D., Kurnianto, S., Stidham, M., and Kanninen, M.: Mangroves among the most carbon-rich forests in the tropics, *Nature Geosci.*, 4: 293-297, doi: 10.1038/ngeo1123, 2011.
- 565 Dung, L.V., Tue, N.T., Nhuan, M.T., and Omori, K.: Carbon storage in a restored Mangrove forest in Can Gio Mangrove Forest Park, Mekong Delta, Vietnam, *Forest Ecol. Manag.*, 380, 31–40, doi: 10.1016/j.foreco.2016.08.032, 2016.
- Duarte, C.M., Losada, I.J., Hendriks, I.E., Mazarrasa, I., and Marbà, N.: The role of coastal plant communities for climate change mitigation and adaptation, *Nature Climate Change*, 3(11): 961-968, doi: 10.1038/nclimate1970, 2013.
- 570 Duncan, C., Primavera, J.H., Pettorelli, N., Thompson, J.R., Loma, R.J.A., and Koldewey, H.J.: Rehabilitating mangrove ecosystem services: A case study on the relative benefits of abandoned pond reversion from Panay Island, Philippines, *Mar. Pollut. Bull.*, 109(2): 772-782, doi: 10.1016/j.marpolbul.2016.05.049, 2016.
- Gnanamoorthy, P., Selvam, V., Ramasubramanian, R., Chakraborty, S., Prमित, D., and Karipot, A.: Soil organic carbon stock in natural and restored mangrove forests in Pichavaram south-east coast of India, *Indian J. Geo-Mar. Sci.*, 48, 2019, 801-808, 2019.
- 575 Gontharet, S., Mathieu, O., Lévêque, J., Milloux, M.J., Lesourd, S., Philippe, S., Caillaud, J., Gardel, A., Sarrazin, M., and Proisy, C.: Distribution and sources of bulk organic matter (OM) on a tropical intertidal mud bank in French Guiana from elemental and isotopic proxies, *Chemical Geology*, 376: 1-10, doi: 10.1016/j.chemgeo.2014.03.009, 2014.
- 580 Goni, M.A., Moore, E., Kurtz, A., Portier, E., Alleau, Y., and Merrell, D.: Organic matter compositions and loadings in soils and sediments along the Fly River, Papua New Guinea, *Geochim. Cosmochim. Ac.*, 140: 275-296, doi: 10.1016/j.gca.2014.05.034, 2014.
- Hatje, V., Masqué, P., Patire, V.F., Dórea, A., and Barros, F.: Blue carbon stocks, accumulation rates, and associated spatial variability in Brazilian mangroves, *Limnol. Oceanogr.*, 9999: 1–14, doi: 10.1002/lno.11607, 2020.
- 585 Howard, J., Sutton-Grier, A., Herr, D., Kleypas, J., Landis, E., Mcleod, E., Pidgeon, E., and Simpson, S.: Clarifying the role of coastal and marine systems in climate mitigation, *Front. Ecol. Environ.*, 15(1): 42-50, doi: 10.1002/fee.1451, 2017.
- Jennarjahn, T.C.: Relevance and magnitude of 'Blue Carbon' storage in mangrove sediments: Carbon accumulation rates vs. stocks, sources vs. sinks, *Estuar. Coast. Shelf S.*, 247, 107027, doi:10.1016/j.ecss.2020.107027, 2020.



- 590 Jiménez-Arias, J.L., Morris, E., Rubio-de-Inglés, M.J., Peralta, G., García-Robledo, E., Corzo, A., and Pappaspyrou, S.: Tidal elevation is the key factor modulating burial rates and composition of organic matter in a coastal wetland with multiple habitats, *Sci. Total Environ.*, 724: 138205, doi: 10.1016/j.scitotenv.2020.138205, 2020.
- Kauffman, J., Arifanti, V.B., Hernandez Trejo, H., del Carmen Jesús García, M., Norfolk, J., Cifuentes, M., Hadriyanto, D. and Murdiyarso, D.: The jumbo carbon footprint of a shrimp: carbon losses from mangrove deforestation, *Front. Ecol. Environ.*, 15(4): 183-188, doi: /10.1002/fee.1482, 2017.
- 595 Kelleway, J.J., N. Saintilan, P.I. Macreadie, C.G. Skilbeck, A. Zawadzki, and P.J. Ralph.: Seventy years of continuous encroachment substantially increases ‘blue carbon’ capacity as mangroves replace intertidal salt marshes, *Glob. Change Biol.*, 22 (3): 1097–1109, doi: 10.1111/gcb.13158, 2016.
- Keil, R. G., and Mayer, L.M.: Mineral matrices and organic matter, in: *Treatise on Geochemistry: Organic Geochemistry 2nd Edition Volume 12*, edited by: Falkowski, P.G., Freeman, K.H., Holland, H. and Turekian, K., Elsevier, Amsterdam, Netherlands, doi:10.1016/B978-0-08-095975-7.01024-X, 337-359, 2014.
- 600 Komada, T., Reimers, C.E., Luther III, G.W., and Burdige, D.J.: Factors affecting dissolved organic matter dynamics in mixed-redox to anoxic coastal sediments, *Geochim. Cosmochim. Ac.*, 68(20): 4099-4111, 2004.
- Kristensen, E.: Mangrove crabs as ecosystem engineers; with emphasis on sediment processes, *J. Sea Res.*, 59, 30–43, doi: 10.1016/j.seares.2007.05.004, 2008.
- 605 Kusumaningtyas, M.A., Hutahaean, A.A., Fischer, H.W., Pérez-Mayod, M., Ransby, D., and Jennerjahn, T.C.: Variability in the organic carbon stocks, sources, and accumulation rates of Indonesian mangrove ecosystems, *Estuar. Coast. Shelf S.*, 218, 310-323, doi: 10.1016/j.ecss.2018.12.007, 2019.
- Lamb, L., Muller, E., and Fromard, F.: Mangrove trees growing in a very saline condition but not using seawater, *Rapid Commun. Mass Spectrom.*, 22: 2835–2843, doi: 10.1007/s10021-010-9329-2, 2008.
- 610 Lovelock, C.E., Sorrell, B.K., Hancock, N., Hua, Q., and Swales, A.: Mangrove forest and soil development on a rapidly accreting shore in New Zealand, *Ecosystems*, 13(3): 437-451, doi: 10.1007/s10021-010-9329-2, 2010.
- Lunstrum, A., and Chen, L.: Soil carbon stocks and accumulation in young mangrove forests, *Soil Biol. Biochem.*, 75, 223-232, doi: 10.1016/j.soilbio.2014.04.008, 2014.
- 615 MacKenzie, R.A., Foulk, P.B., Klump, J.V., Weckerly, K., Purbospito, J., Murdiyarso, D., Donato, D.C. and Nam, V.N.: Sedimentation and belowground carbon accumulation rates in mangrove forests that differ in diversity and land use: a tale of two mangroves, *Wetl. Ecol. Manag.*, 24(2): 245-261, doi: 10.1007/s11273-016-9481-3, 2016.
- Marchand, C.: Soil carbon stocks and burial rates along a mangrove forest chronosequence (French Guiana), *Forest Ecol. Manag.*, 384: 92-99, doi: 10.1016/S0272-7714(02)00134-8, 2017.



- 620 Marchand, C., Lallier-Vergès, E., and Baltzer, F.: The composition of sedimentary organic matter in relation to the dynamic features of a mangrove-fringed coast in French Guiana, *Estuar. Coast. Shelf S.*, 56: 119–130, doi: 10.1016/S0272-7714(02)00134-8, 2003.
- Marchand, C., Albe´ric, P., Lallier-Verge`s, E., and Baltzer, F.: Distribution and characteristics of dissolved organic matter in mangrove sediment pore waters along the coastline of French Guiana, *Biogeochemistry*, 81:59–75, 625 doi:10.1007/s10533-006-9030-x, 2006.
- Matsumoto, K., Takanezawa, T., and Ooe, M.: Ocean tide models developed by assimilating TOPEX/POSEIDON Altimeter Data into hydrodynamical model: a global model and a regional model around Japan, *J. Oceanogr.*, 56: 567–581, 200.
- Matos, C.R., Berrêdo, J.F., Machado, W., Sanders, C.J., Metzger, E., and Cohen, M.C.: Carbon and nutrient 630 accumulation in tropical mangrove creeks, Amazon region, *Mar. Geo.*, 429: 106317, doi:10.1016/j.margeo.2020.106317, 2020.
- Mayer, L.M.: Relationships between mineral surfaces and organic carbon concentrations in soils and sediments, *Chem. Geol.*, 114: 347–363, doi:10.1002/Ino.10478, 1994.
- Miyajima, T., Hori, M., Hamaguchi, M., Shimabukuro, H., and Yoshida, G.: Geophysical constraints for organic 635 carbon sequestration capacity of *Zostera marina* seagrass meadows and surrounding habitats, *Limnol. Oceanogr.*, 62(3): 954-972, doi:10.1002/Ino.10478, 2017.
- Miyajima, T., Y. Tsuboi, Y. Tanaka, and I. Koike.: Export of inorganic carbon from two Southeast Asian mangrove forests to adjacent estuaries as estimated by the stable isotope composition of dissolved inorganic carbon, *J. Geophys. Res.*, 114: G01024, doi:10.1029/2008JG000861, 2009.
- 640 Nellemann, C., Corcoran, E., Duarte, C.M., Valdés, L., De Young, C., Fonseca L, and Grimsditch, G.: Blue carbon: a rapid response assessment. United Nations Environment Programme, GRID-Arendal, 2009.
- Nam, V.N., Sasmito, S.D., Murdiyarso, D., Purbopuspito, J., and MacKenzie, R.A.: Carbon stocks in artificially and naturally regenerated mangrove ecosystems in the Mekong Delta, *Wetl. Ecol. Manag.*, 24:231–244, doi: 10.1007/s11273-015-9479-2, 2016.
- 645 Nilsson, C. and Wilson, S.D.: Convergence in plant community structure along a disparate gradient: are lakeshores inverted mountainsides?, *The American Naturalist*, 137: 774-790, doi: 10.1086/285193, 1991.
- Nordhaus, I., Salewski, T., and Jennerjahn, T.C.: Interspecific variations in mangrove leaf litter decomposition are related to labile nitrogenous compounds, *Estuar. Coast. Shelf S.*, 192:137-148, doi:10.1016/j.ecss.2017.04.029, 2017.



- 650 Noguchi, K., Pongparn, S., Umnouysin, S., Patanaponpaiboon, P., Duangnamol, D., Yoneda, R., Utsugi, H., Sato, T., and Tabuchi, R.: Biomass and Production Rates of Fine Roots in Two Mangrove Stands in Southern Thailand, *JARQ*, 54(4): 349-360, doi:10.6090/jarq.54.349, 2020.
- Parnell, A.C., Inger, R., Bearhop, S. and Jackson, A.L.: Source partitioning using stable isotopes: coping with too much variation, *PloS one*, 5(3): e9672, doi:10.1371/journal.pone.0009672, 2010.
- 655 Pe´rez, A., Libardoni, B.G., Sanders, C.J.: Factors influencing organic carbon accumulation in mangrove ecosystems, *Biol. Letters*, 14: 20180237, doi:10.1098/rsbl.2018.0237, 2018.
- Pickett, S.T.A.: Space-for-time substitution as an alternative to long-term studies, in: *Long-term studies in ecology*, edited by: Likens G. E., Springer, 110-135, 1989.
- Primavera, J.H., Sadaba, R.S., Lebata, M.J.H.L., and Altamirano, J.R.: *Handbook of Mangroves in the Philippines – Panay*, SEAFDEC Aquaculture Department, Iloilo, Philippines, 106, 2004.
- 660 Salmo, S.G., Lovelock, C., and Duke, N.C.: Vegetation and soil characteristics as indicators of restoration trajectories in restored mangroves, *Hydrobiologia*, 720: 1–18, doi:10.1007/s10750-013-1617-3, 2013.
- Prahl, F.G., Bennett, J.T., and Carpenter, R.: The early diagenesis of aliphatic hydrocarbons and organic matter in sedimentary particulates from Dabob Bay, Washington, *Geochimic. Cosmochim. Ac.*, 44, 1967–1976, 1980.
- 665 Prasad, M.B.K., Kumar, A., Ramanathan, A.L., and Datta, D.K.: Sources and dynamics of sedimentary organic matter in Sundarban mangrove estuary from Indo-Gangetic delta, *Ecological Processes*, 6:8, doi:10.1186/s13717-017-0076-6, 2017.
- Pendleton, L., Donato, D.C., Murray, B.C., Crooks, S., Jenkins, W.A., Sifleet, S., Craft, C., Fourqurean, J.W., Kauffman, J.B., Marbà, N. and Megonigal, P.: Estimating global “blue carbon” emissions from conversion and degradation of vegetated coastal ecosystems, *PLoS One*, 7(9): e43542, doi: 10.1371/journal.pone.0043542, 2012.
- 670 Ranjan, R.K., Routh, J., Ramanathan, A.L., and Klump, J.V.: Elemental and stable isotope records of organic matter input and its fate in the Pichavaram mangrove-estuarine sediments (Tamil Nadu, India, *Mar. Chem.*, 126, 163–172, doi: 10.1016/j.marchem.2011.05.005, 2011.
- Ray, R., Michaud, E., Aller, R.C., Vantrepotte, V., Gleixner, G., Walcker, R., Devesa, J., Le Goff, M., Morvan, S. and Thouzeau, G.: The sources and distribution of carbon (DOC, POC, DIC) in a mangrove dominated estuary (French Guiana, South America), *Biogeochemistry*, 138(3), 297–321. doi:10.1007/ s10533-018-0447-9, 2018.
- 675 Ray, R., Thouzeau, G., Walcker, R., Vantrepotte, V., Morvan, S., Devesa, J., Le Goff, M., and Michaud, E.: Mangrove-derived organic and inorganic carbon export to the Atlantic Ocean? Time series study in the Guianese estuarine system, *Latin America, J. Geophys. Res. Biogeo.*, 125: 8, e2020JG005739, doi: 10.1029/2020JG005739, 2020.
- 680



- Ren, H., Chen, H., Li, Z., and Han, W.: Biomass accumulation and carbon storage of four different aged *Sonneratia apetala* plantations in Southern China, *Plant. Soil*, 327: 279–291, doi: 10.1007/s11104-009-0053-7, 2010.
- Shahraki, M., Fry, B., Krumme, U., and Rixen, T.: Microphytobenthos sustain fish food webs in intertidal arid habitats: a comparison between mangrove-lined and un-vegetated creeks in the Persian Gulf, *Estuar. Coast. Shelf Sci.*, 685 149: 203–212, doi: 10.1016/j.ecss.2014.08.017, 2014.
- Shen, H., Tu, Z., You, G., Shi, X.: Grain Size and Pollen of Sediments in Wanghu Lake (Central China) Linked to Hydro-Environmental Changes, *Water*, 12, 45; doi:10.3390/w12010045, 2020.
- Sasmito, S.D., Kuzyakov, Y., and Lubis, A.A.: Organic carbon burial and sources in soils of coastal mudflat and mangrove ecosystems, *Catena*, 187: 104414, doi: 10.1016/j.catena.2019.104414, 2020.
- 690 Sanders, C.J., Smoak, J.M., Naidu, A.S., Sanders, L.M., and Patchineelam, S.R.: Organic carbon burial in a mangrove forest, margin and intertidal mud flat, *Estuar. Coast. Shelf S.*, 90: 168–172, doi: 10.1016/j.ecss.2010.08.013, 2010.
- Sharma, S., MacKenzie, R.A., Tieng, T., Soben, K., Tulyasuwan, N., Resanond, A., Blate, G. and Litton, C.M.: The impacts of degradation, deforestation and restoration on mangrove ecosystem carbon stocks across Cambodia, *Sci. Total Environ.*, 706, 135416, doi: 10.1016/j.scitotenv.2019.135416. 2020.
- 695 Sheehan, L., Sherwood, E., Moyer, R.P., Radabaugh, K.R., and Simpson, S.: Blue carbon: an additional driver for restoring and preserving ecological services of coastal wetlands in Tampa Bay (Florida, USA), *Wetlands*, 39: 1317–1328, doi: 10.1007/s13157-019-01137-y, 2019.
- Smith III, T. J., Boto, K. G., Frusher, S. D., and Giddins, R. L.: Keystone species and mangrove forest dynamics: the influence of burrowing by crabs on soil nutrient status and forest productivity, *Estuar. Coast. Shelf. S.*, 33: 419–700 432, doi:10.1016/0272-7714(91)90081-L, 1991.
- Soper, F.M., MacKenzie, R.A., Sharma, S., Cole, T.G., Litton, C.M., and Sparks. J.P.: Non - native mangroves support carbon storage, sediment carbon burial, and accretion of coastal ecosystems, *Glob. Change Biol.*, 25:4315–4326, doi: 10.1111/gcb.14813, 2019.
- Van Hieu, P., Dung, L.V., Tai Tue, N., and Omori, K.: Will restored mangrove forests enhance sediment organic carbon and ecosystem carbon storage?, *Regional Studies in Marine Science*, 14: 43–52, doi: 705 10.1016/j.rsma.2017.05.003, doi: 10.1016/j.rsma.2017.05.003, 2017.
- Valiela, I., Bowen, J. L. and York, J. K.: Mangrove forests: one of the world’s threatened major tropical environments, *Bioscience*, 51:807–815, doi: 10.1641/0006-3568(2001)051[0807:MFOOTW]2.0.CO;2, 2001.
- Wagai, R., Mayer, L.M., and Kitayama, K.: Extent and nature of organic coverage of soil mineral surfaces assessed 710 by a gas sorption approach, *Geoderma*, 149: 152–160, doi: 10.1016/j.geoderma.2008.11.032, 2009.



- Wang, G., Yu, C., Singh, M., Guan, D., Xiong, Y., Zheng, R. and Xiao, R.: Community structure and ecosystem carbon stock dynamics along a chronosequence of mangrove plantations in China, *Plant Soil*, 464: 605–620, doi: 10.1007/s11104-021-04973-2, 2021.
- Walker, L. R., Wardle, D. A., Bardgett, R. D., and Clarkson, B. D.: The use of chronosequences in studies of ecological succession and soil development, *J. Ecol.*, 98, 725–736, doi:10.1111/j.1365-2745.2010.01664.x, 2010.
- Walcker, R., Gandois, L., Proisy, C., Corenblit, D., Mougin, É., Ray, R., and Fromard, F.: Control of ‘blue carbon’ storage by mangrove ageing: evidence from a 66-year-chronosequence in French Guiana, *Glob. Change Biol.*, 24: 2325–2338, doi: doi.org/10.1111/gcb.14100, 2018.
- Xiong, Y., Liao, B., and Wang, F.: Mangrove vegetation enhances soil carbon storage primarily through in situ inputs rather than increasing allochthonous sediments, *Mar. Pollut. Bull.*, 131: 378–385, doi:10.1016/j.marpolbul.2018.04.043, 2018.
- Yoshikai, M., Nakamura, T., Suwa, R., Argamosa, R., Okamoto, T., Rollon, R., Basina, R., Primavera-Tirol, Y.H., Blanco, A.C., Adi, N.S. and Nadaoka, K.: Scaling relations and substrate conditions controlling the complexity of Rhizophora prop root system, *Estuar. Coast Shelf S.*, 248:107014, doi:10.1016/j.ecss.2020.107014, 2021.
- Zhang, Y., Xiao, L., Guan, D., Chen, Y., Motelica-Heino, M., Peng, Y. and Lee, S.Y.: The role of mangrove fine root production and decomposition on soil organic carbon component ratios, *Ecol. Indic.*, 125: 107525, doi:10.1016/j.ecolind.2021.107525, 2021.

#### Figure legends

- Figure 1: Map of the study area. BS (Bare sediment); PM (Pioneer mangrove, 3-month); YM (Young mangrove, 10-yr); AM (Adult mangrove, 20-yr); MM (Mature mangrove, 30-yr). The circle shows presence of long-tailed sandbar between AM and YM site. The north-west inland part and bank of the Aklan River are the dominant places for the naturally occurring mangroves.
- Figure 2: Variation of physical and biogeochemical parameters in the sediments according to mangrove development.
- Figure 3: Vertical profiles of sedimentary carbon properties during wet and dry season.
- Figure 4: Vertical distribution of sediment surface area and OC loading during wet and dry season
- Figure 5: Vertical profiles of porewater DOC during wet and dry season
- Figure 6: Source identification of sedimentary organic matter using endmember carbon stable isotope ratio and OC:TN.
- Figure 7: Source apportionment of sedimentary organic matter at different mangrove stages by applying bayesian mixing model with  $\delta^{13}\text{C}$  and OC:TN.
- Figure 8: Slopes derived from linear and exponential relationship between (a) mangrove age and carbon stock (b) and carbon accumulation rate (CAR).



745

**Table 1: The significance of effects of season, type and depth to TOC, DOC, bulk OC/TN ratio  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  using ANOVA on the basis of the generalized additive model (GAM). In the parameter of depth, the approximate significance of smooth term is shown.**

Parameters	Season	Type	Depth
TOC	0.0855	< 0.001	< 0.001
DOC	0.1986	0.003	0.127
OC:TN	< 0.001	< 0.001	0.033
$\delta^{13}\text{C}$	0.0656	< 0.001	0.411
$\delta^{15}\text{N}$	0.0148	0.0061	0.737

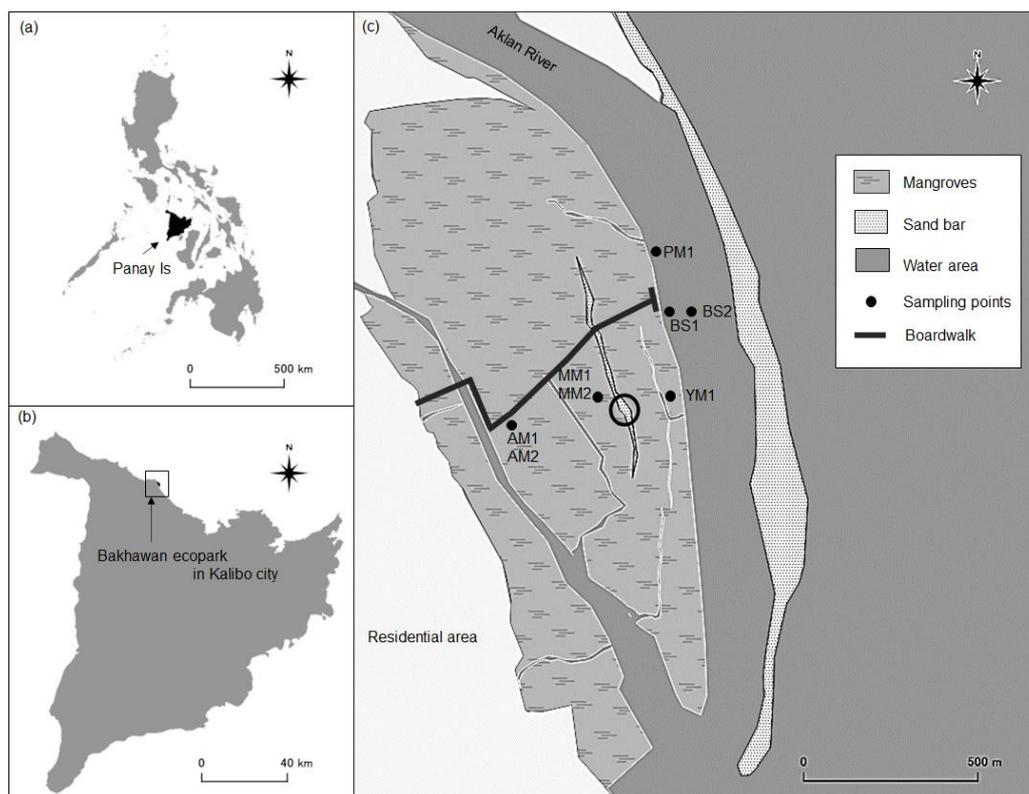
750

755

760

765

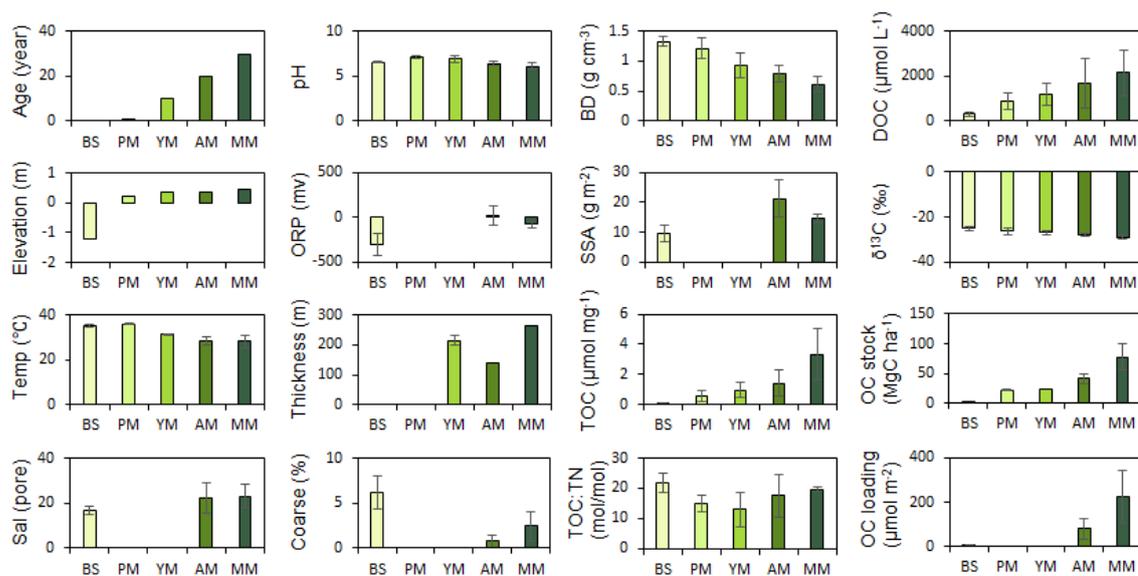
770



**Fig. 1.**

775

780



**Fig. 2.**

785

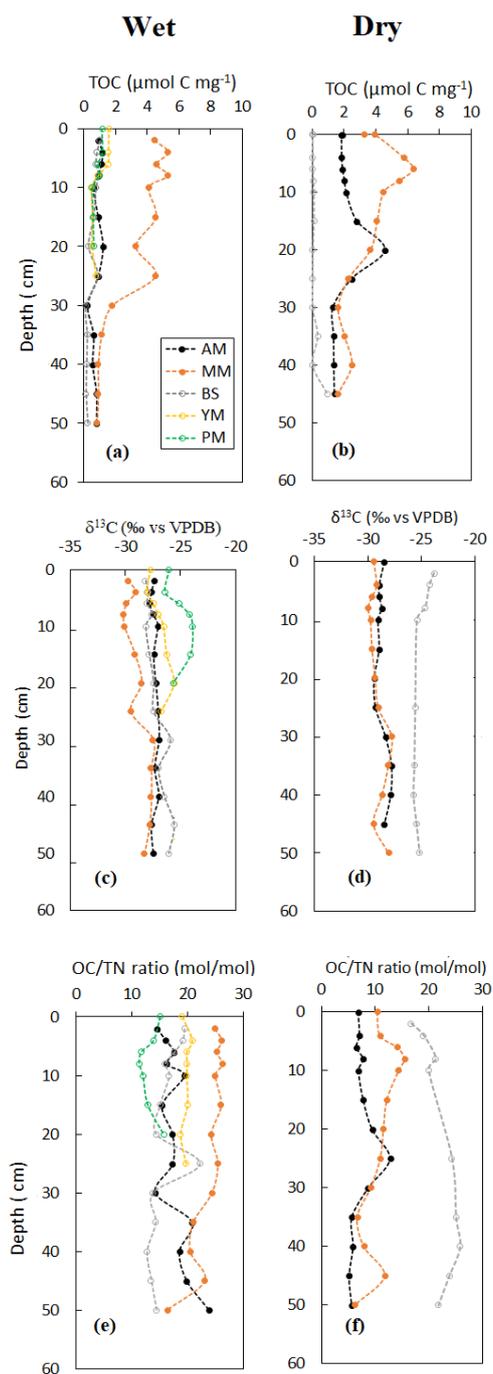


Fig. 3.

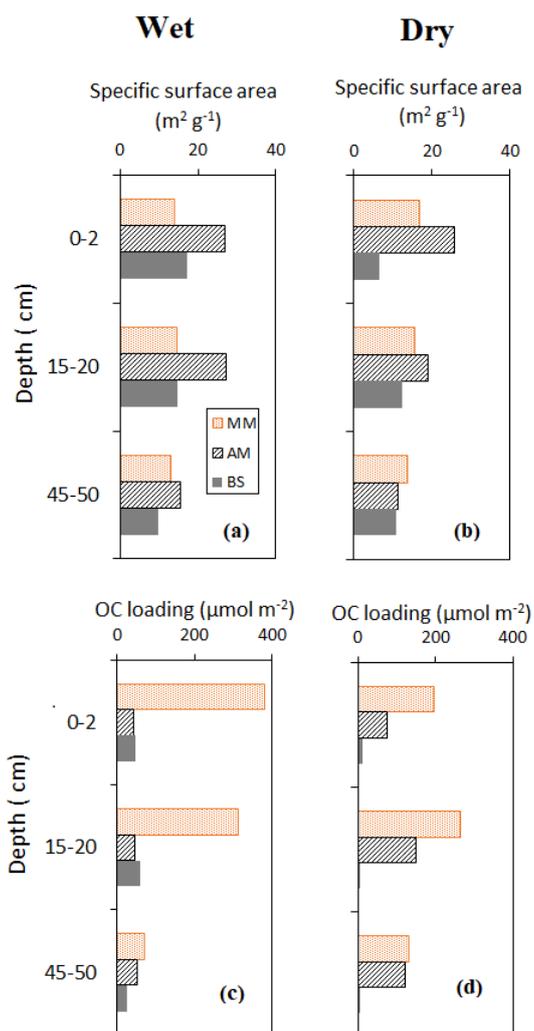


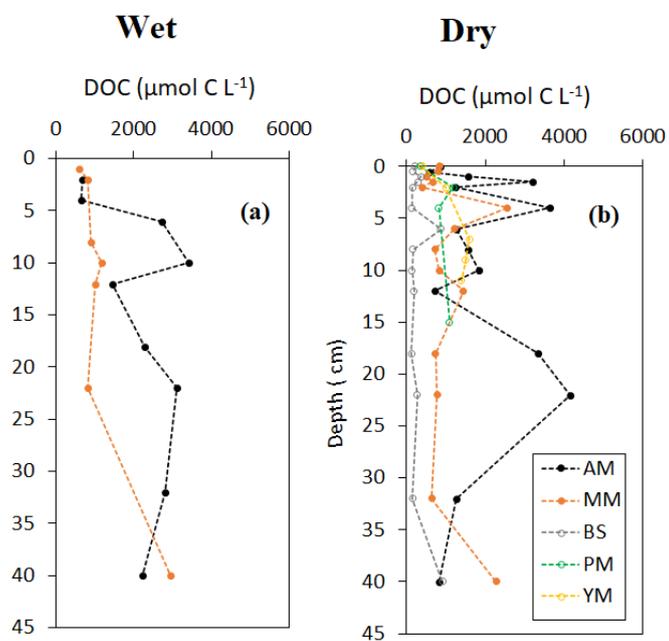
Fig. 4.

790

795



800



**Fig. 5.**

805

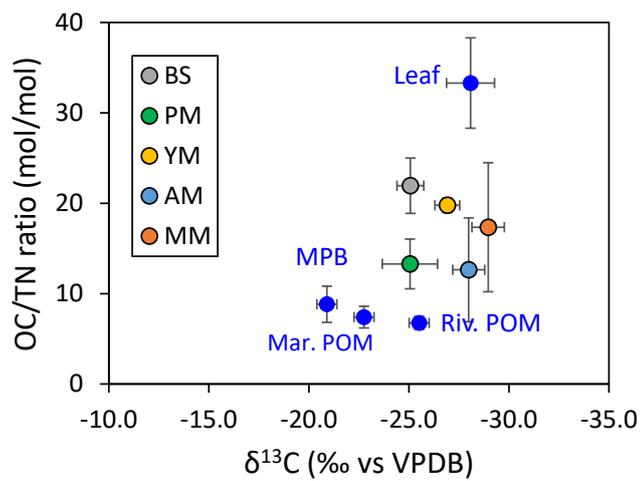
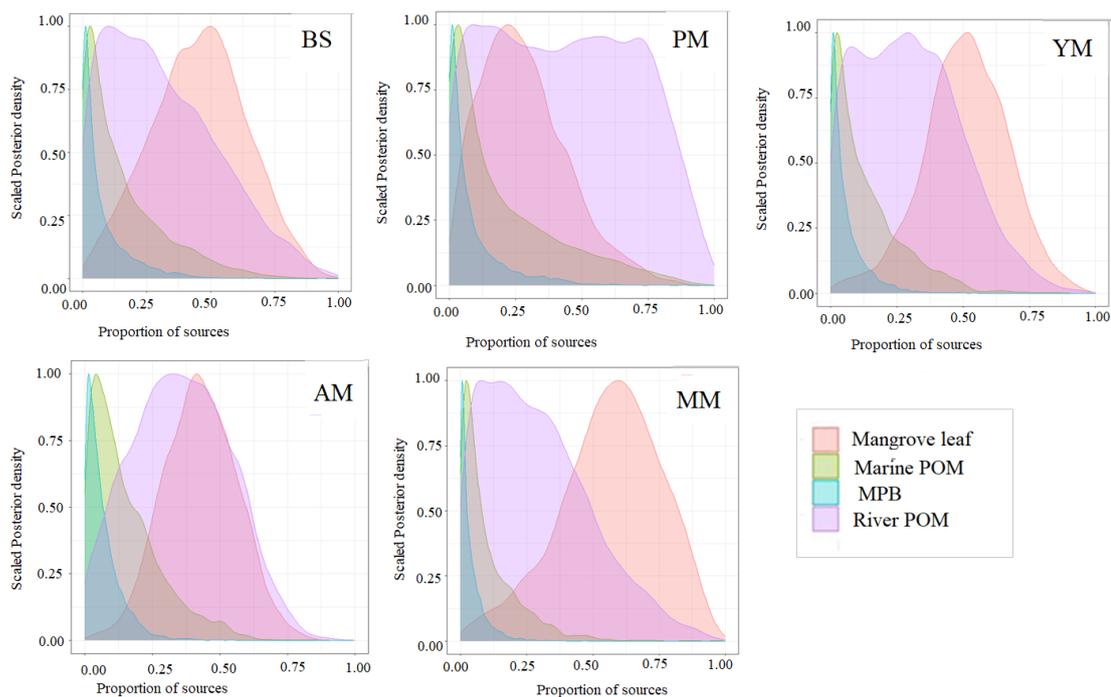
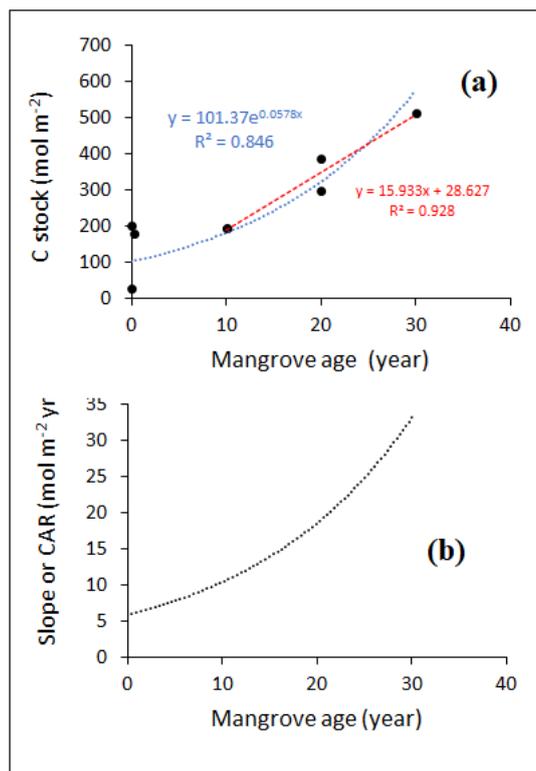


Fig. 6.



**Fig. 7.**



820 **Fig. 8.**

825

830