

Monsoon-driven biogeochemical dynamics in an equatorial shelf sea: time-series  
observations in the Singapore Strait

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## Abstract

Coastal tropical waters are experiencing rapid increases in anthropogenic pressures, yet coastal biogeochemical dynamics in the tropics are poorly studied. We present a multi-year biogeochemical time series from the Singapore Strait in Southeast Asia's Sunda Shelf Sea. Despite being highly urbanised and a major shipping port, the strait harbours numerous biologically diverse habitats, and is a valuable system for understanding how tropical marine ecosystems respond to anthropogenic pressures. Our results show strong seasonality driven by the semi-annual reversal of ocean currents: dissolved inorganic nitrogen (DIN) and phosphorus varied from  $\leq 0.05 \mu\text{mol l}^{-1}$  during the intermonsoons to  $\geq 4 \mu\text{mol l}^{-1}$  and  $\geq 0.25 \mu\text{mol l}^{-1}$ , respectively, during the southwest monsoon.  $\text{Si(OH)}_4$  exceeded DIN year-round. Based on nutrient concentrations, their relationships to salinity and coloured dissolved organic matter, and the isotopic composition of  $\text{NO}_x^-$ , we infer that terrestrial input from peatlands is the main nutrient source. This input delivered dissolved organic carbon (DOC) and nitrogen, but was notably depleted in dissolved organic phosphorus. In contrast, particulate organic matter showed little seasonality, and the  $\delta^{13}\text{C}$  of particulate organic carbon ( $-21.0 \pm 1.5\%$ ) is consistent with a primarily autochthonous origin. Diel changes in dissolved  $\text{O}_2$  varied seasonally with a pattern that suggests that light availability controls primary productivity more than nutrient concentrations. However, diel changes in pH were greater during the southwest monsoon, when remineralisation of terrestrial DOC lowers the seawater buffer capacity. We conclude that terrestrial input results in mesotrophic conditions, and that the strait might be vulnerable to further eutrophication if nutrient inputs increase during seasons when light availability is high. Moreover, the seasonality of diel pH variation suggests that coral reefs exposed to terrestrial organic matter in the Sunda Shelf may be at significant risk from future ocean acidification.

## 1. Introduction

Tropical shelf seas are ecologically and economically important, but are under increasing anthropogenic pressure from coastal development, land-use change, resource extraction, and terrestrial inputs (Jennerjahn, 2012). At the same time, our understanding of the biogeochemistry of tropical shelf seas is more limited than for higher-latitude environments, which makes it harder to predict how anthropogenic pressures will affect tropical seas (Lønborg et al., 2021b; Vieillard et al., 2020).

Tropical coastal waters are usually nutrient-poor because shelf seas exchange large volumes of water with adjacent nutrient-poor open oceans (Brunskill, 2010). However, tropical coastal waters also receive more than half of the global river discharge and nutrient input (Jennerjahn, 2012; Mayorga et al., 2010). Nutrient concentrations and ratios delivered by tropical rivers can be different from temperate regions: for example, tropical rivers typically have higher dissolved silicon concentrations (Jennerjahn et al., 2006). While consistently high temperature and sunlight in the tropics allow for fast rates of uptake and biogeochemical transformation of nutrients in shelf waters, high input of sediments and dissolved organic matter in the tropics can also attenuate sunlight and potentially slow the rates of nutrient utilisation (Nittrouer et al., 1995). As anthropogenic nutrient input is projected to increase substantially within the next decades, particularly in tropical Asia (Sinha et al., 2019), it is pressing to better understand the biogeochemistry of tropical coastal waters.

Southeast Asia's Sunda Shelf Sea is one of the largest and most biodiverse shelf seas globally; however, it also has dense human populations along the coast, placing a high degree of pressure on its marine habitats (Jennerjahn, 2012; Veron et al., 2009). This is especially the case in urbanised marine ecosystems that experience high anthropogenic stress (Heery et al., 2018; Todd et al., 2019). The Sunda Shelf Sea receives a large amount of precipitation and river input that deliver freshwater (Lee et al., 2019), terrestrial carbon (Baum et al., 2007;

Huang et al., 2017; Zhou et al., 2019), and dissolved nutrients (Jennerjahn et al., 2004; Jiang et al., 2019). The climate is driven by the Asian monsoon system, which results in seasonally varying patterns of precipitation across the region and causes a semi-annual reversal in physical ocean circulation across the shelf (Lee et al., 2019; Susanto et al., 2016). While these environmental conditions might be expected to result in dynamic coastal biogeochemistry, the biogeochemistry of Southeast Asia's coastal seas has received comparatively little research attention to date (Partelow et al., 2018). Across the open waters of the Sunda Shelf, chlorophyll-*a* concentrations are typically  $\leq 0.2 \mu\text{g l}^{-1}$  (Kartadikaria et al., 2015; Ke et al., 2014), indicative of oligotrophic waters. Closer to shore and within estuaries, significant levels of eutrophication due to nutrient input have been reported (Chai et al., 2021; Damar et al., 2019; Lim et al., 2021; Lim et al., 2018; Tomascik et al., 1994). However, very few studies have collected multi-year biogeochemical time series in Southeast Asia, and where such time series do exist (Lim et al., 2018), often only a limited number of parameters are available. This limits our understanding of the drivers of coastal biogeochemistry in Southeast Asia.

Here, we present time-series data collected over multiple years in the Singapore Strait, an urbanised coastal area in the central Sunda Shelf, to examine the dynamics and drivers of nutrient biogeochemistry, and their potential impacts on planktonic productivity. This study complements our previous analyses of seawater carbonate system and bio-optical variability in the Singapore Strait, which revealed a strong seasonal input of terrigenous dissolved organic matter from regional peatlands (Martin et al., 2021; Zhou et al., 2021).

## 2. Materials and Methods

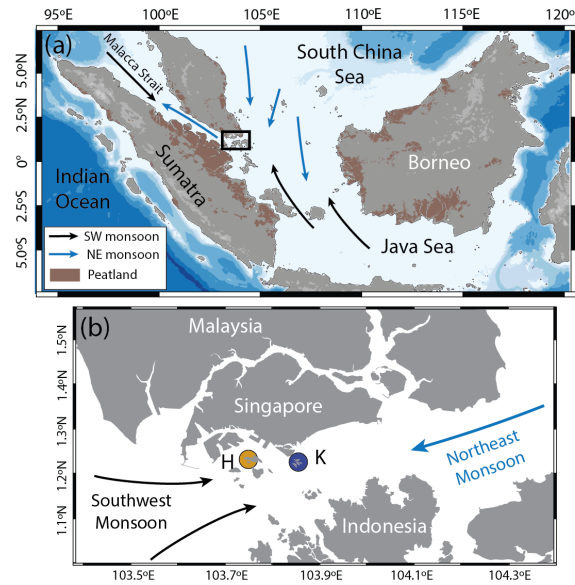


Fig. 1 Map of the study region, modified from Martin et al. (2021), indicating the seasonal current reversal between southwest (SW) and northeast (NE) monsoons (arrows). Yellow marker in (a) indicates the Singapore Strait. The two sampling sites, Hantu (“H”) and Kusu (“K”) are indicated in (b) by the two crossed yellow circles.

## 2.1 Site description and study design

The Singapore Strait is situated in the central part of the Sunda Shelf Sea at 1.2°N 103.8°E (Fig. 1) and is subject to two monsoon seasons (Mayer et al.; Susanto et al.; van Maren & Gerritsen). During the northeast (NE) monsoon (mid-November to March), the prevailing mean circulation carries water from the open South China Sea across the Sunda Shelf, west- and northwards through the Singapore Strait and the Malacca Strait, and southwards into the Java Sea. During the southwest (SW) monsoon (mid-May to mid-September), the circulation reverses and water flows northwards from the Java Sea towards the South China Sea, while the flow in the Malacca Strait ceases or flows weakly southwards. Flow through the Singapore Strait is then eastwards and out towards the open South China Sea. The annual mean flow follows the direction of the NE monsoon, forming the South China Sea limb of the

Indonesian Throughflow. During the two intermonsoon periods (April to mid-May and mid-September to mid-November) the mean residual flow through the Singapore Strait stagnates. Singapore experiences rainfall year-round, but rainfall is highest during the early NE monsoon (mid-November to January) and is lowest during the late NE monsoon and early intermonsoon (February and March). Because the seawater chemistry changes considerably from early to late NE monsoon, but the late NE monsoon and first intermonsoon are very similar, for the purposes of our analysis we classify March as part of intermonsoon 1, consistent with our previous work (Martin et al., 2021; Zhou et al., 2021). We thus define the seasons as: NE monsoon = 15 Nov to end Feb; intermonsoon 1 = 01 Mar to 15 May; SW monsoon = 15 May to 15 Sept; intermonsoon 2 = 15 Sept to 15 Nov. Because of the reversing ocean circulation, the Singapore Strait is influenced by considerable river input from Sumatra and the western Malay Peninsula during the SW monsoon, with a notable seasonal decrease in salinity (Tanzil et al., 2019; Zhou et al., 2021) and input of terrestrial dissolved organic matter (DOM) from regional peatlands (Martin et al., 2021; Zhou et al., 2021). During the early NE monsoon, there is freshwater input from local rainfall and river flow, with a shorter period of reduced salinity and a small input of terrestrial DOM (Martin et al., 2021; Zhou et al., 2021). Here, we present time-series data collected by regular *in-situ* water sampling at two sites in the Singapore Strait, Kusu Island (1.226°N 103.860°E) and Hantu Island (1.227°N 103.746°E) (Fig. 1). Both sites are small islands with narrow fringing reefs, with diverse communities of hard corals down to approximately 6–8 m depth (Guest et al., 2016; Huang et al., 2009). Below this depth, the seafloor consists largely of soft sediment and slopes down to 15–20 m. Sampling started in mid-2015 on an opportunistic basis for inorganic nutrients, and was carried out at monthly to bi-weekly frequency from mid-2017 for an increasing number of parameters.

## 2.2 Sensor installation

In mid-2015, a Seabird SeaCat 19plusV2 conductivity-temperature-depth sensor (CTD) and a SeaFET pH sensor (previously Satlantic, now Seabird) were installed at approximately 5 m depth on the reef slope of Kusu Island, using iron stakes such that the sensors were approximately half a metre above the seafloor. In mid-2017, a PME miniDOT dissolved oxygen sensor was added at the same depth. The SeaFET and the miniDOT were both equipped with copper biofouling guards; the SeaCat had no antifouling protection but was operated with the pump running during sampling to prevent sediment accumulation in the conductivity cell. All sensors were installed horizontally (except the miniDOT, which initially faced upwards but was then installed horizontally to prevent sediment accumulation between the biofouling guard and the sensor membrane) and measured every 10 min. Sensors were recovered for data download and cleaning every 1–4 months. Gaps in the sensor records occurred as a result of occasional technical problems, the need for maintenance back in the laboratory and factory recalibration, and periods of bad data (see Section 2.5). All salinity data are expressed on the practical salinity scale.

## 2.3 Water sampling

Water samples were collected with a Niskin bottle at 5 m depth, and a CTD profile was measured using a Valeport FastCTD with chlorophyll-*a* fluorometer. Prior to October 2017, samples were only collected for dissolved inorganic nutrient analysis, by syringe-filtering (0.2 µm Acrodisc polyethersulfone [PES] filters, 25 mm diameter) water into acid-washed, 15 ml polypropylene centrifuge tubes. From October 2017, a larger number of parameters was measured, and sample water was filtered directly from the Niskin bottle with a peristaltic pump and an in-line polycarbonate filter housing (Pall, product 1119) using 0.2 µm PES membranes (47 mm diameter, Supor, Millipore). The filter, tubing, and filter housing were

170 rinsed with 300 ml ultrapure water ( $18.2 \text{ M}\Omega \text{ cm}^{-1}$ , hereafter Elga water) and about 100 ml  
171 sample water immediately before samples were collected.

172 Water for  $\delta^{13}\text{C}$  of dissolved inorganic carbon ( $\delta^{13}\text{C}$ -DIC, 1 ml) was filtered into a syringe  
173 without allowing air contact and injected into a He-flushed 12 ml Exetainer (Labco, UK) with  
174 butyl rubber septum containing 1 ml of 80%  $\text{H}_3\text{PO}_4$ . Water for DIC concentration was filled  
175 to overflowing into 12 ml Exetainers and capped with minimal headspace with a butyl rubber  
176 septum. Water for total alkalinity (TA) was filled into 125 ml acid-washed high-density  
177 polyethylene (HDPE) bottles. Water for dissolved organic carbon (DOC) and coloured  
178 dissolved organic matter (CDOM) was filled into pre-ashed ( $450^\circ\text{C}$ , 4 hours) 40 ml amber  
179 borosilicate vials with PTFE-lined septa. Water for  $\delta^{13}\text{C}$  of DOC ( $\delta^{13}\text{C}$ -DOC) was filled into  
180 50 ml acid-washed polypropylene centrifuge tubes. Water for dissolved organic and  
181 inorganic nutrients was filled into acid-washed 15 ml polypropylene centrifuge tubes. Water  
182 for nitrate isotopes ( $\delta^{15}\text{N}$ - and  $\delta^{18}\text{O}$ - $\text{NO}_x^-$ ) was filled into acid-washed 125 ml HDPE bottles.  
183 Samples for dissolved nutrients were immediately frozen in cryogenic dry shippers in the  
184 field. All other samples were stored at ambient temperature in the dark until return to the lab  
185 (within 2–5 hours). All samples were then stored at  $+4^\circ\text{C}$  except for dissolved nutrients,  
186  $\delta^{15}\text{N}$ - and  $\delta^{18}\text{O}$ - $\text{NO}_x^-$  ( $-20^\circ\text{C}$ ),  $\delta^{13}\text{C}$ -DOC ( $-20^\circ\text{C}$ ), and  $\delta^{13}\text{C}$ -DIC (room temperature).

187 Samples for DOC concentration were acidified on the day of collection with  $100 \mu\text{l}$  50%  
188  $\text{H}_2\text{SO}_4$  per 30 ml sample.

189 In addition, unfiltered water was collected in the field into 1 l acid-washed HDPE bottles to  
190 measure particulate organic carbon (POC) and particulate nitrogen (PN), particulate  
191 phosphorus (PP),  $\delta^{13}\text{C}$  of POC ( $\delta^{13}\text{C}$ -POC),  $\delta^{15}\text{N}$  of PN ( $\delta^{15}\text{N}$ -PN), and chlorophyll-*a*. These  
192 samples were stored at ambient temperature in the dark until return to the laboratory, and  
193 then vacuum-filtered (0.5–1.5 l, depending on date and parameter) onto 25 mm Whatman  
194 GF/F filters (nominal pore size  $0.7 \mu\text{m}$ ). Except for chlorophyll-*a*, all GF/F filters were pre-



ashed (450°C 4 hours), and samples were briefly rinsed with Elga water upon filtration. All samples were then wrapped in aluminium foil and frozen at -20°C, except chlorophyll samples, which were flash-frozen in liquid nitrogen and stored at -80°C.

## 2.4 Analyses

### 2.4.1 Dissolved nutrients and organic matter

Dissolved inorganic nutrients, i.e. nitrate ( $\text{NO}_3^-$ ), nitrite ( $\text{NO}_2^-$ ), ammonia ( $\text{NH}_4^+$ ), phosphate ( $\text{PO}_4^{3-}$ ), and silicate ( $\text{Si}(\text{OH})_4$ ), were analysed on a SEAL AA3 segmented-flow autoanalyser. The methods for  $\text{NO}_3^- + \text{NO}_2^-$ ,  $\text{NO}_2^-$ ,  $\text{PO}_4^{3-}$ , and  $\text{Si}(\text{OH})_4$  followed the colorimetric techniques outlined in Hansen and Koroleff (1999) according to SEAL methods G172, G173, G297, and G177.  $\text{NH}_4^+$  was analysed fluorometrically after reaction with o-phthaldialdehyde according to SEAL method G327, based on K  rouel and Aminot (1997). Detection limits were 0.05  $\mu\text{mol l}^{-1}$  ( $\text{NO}_3^- + \text{NO}_2^-$ ), 0.01  $\mu\text{mol l}^{-1}$  ( $\text{NO}_2^-$ ), 0.016  $\mu\text{mol l}^{-1}$  ( $\text{PO}_4^{3-}$ ), 0.1  $\mu\text{mol l}^{-1}$  ( $\text{Si}(\text{OH})_4$ ), and 0.25  $\mu\text{mol l}^{-1}$  ( $\text{NH}_4^+$ ).

Dissolved organic nitrogen and phosphorus (DON and DOP) were analysed as for inorganic  $\text{NO}_3^-$  and  $\text{PO}_4^{3-}$  following wet oxidation. 4.5 ml of sample water were mixed in glass centrifuge tubes with 0.5 ml of a solution consisting of 6 g  $\text{K}_2\text{S}_2\text{O}_8$  with 3 g  $\text{B}(\text{OH})_3$  in 100 ml of 0.42 M NaOH, tightly capped and autoclaved at 120°C for 30 min, and diluted 1:1 with artificial seawater prior to analysis. For DOP analysis, we introduced an additional ascorbic acid solution with the first reagent line (instead of just surfactant as for inorganic  $\text{PO}_4^{3-}$  analysis), such that the solution stream on the AA3 had a concentration of 40 mmol  $\text{l}^{-1}$  ascorbic acid before the colour reagents were introduced. This reduces interference from chlorine species generated during wet oxidation (Ma et al., 2017). Oxidation efficiency was monitored with ethylenediamine tetraacetic acid (DON) and glucose-6-phosphate (DOP), and was always  $\geq 92\%$ .

Dissolved organic carbon (DOC) was analysed on a Shimadzu TOC-L instrument equipped with the manufacturer's high-salt kit, calibrated with potassium hydrogen phthalate standards, and validated using the University of Miami deep-sea certified reference material. Coloured dissolved organic matter (CDOM) was measured on a Thermo Evolution300 dual-beam spectrophotometer against Elga water as the reference using 10-cm pathlength quartz cuvettes. Data were baseline corrected (Green and Blough, 1994), smoothed using a loess function, and converted to Napierian absorption coefficients. We use the CDOM absorption coefficient at 350 nm,  $a_{350}$ , as a measure of CDOM concentration.

#### 2.4.2 Particulate C, N, and P

Particulate organic carbon (POC) and particulate nitrogen (PN) were analysed together after acid-fuming the filters overnight in a desiccator with concentrated HCl, then drying them overnight at 40°C, and pelleting the samples in tin foil discs (Sercon, SC1032). They were then analysed at the University of Hong Kong on a Eurovector elemental analyser and Nu Instruments Perspective isotope ratio mass spectrometer (IRMS) calibrated with acetanilide standards. Particulate phosphorus (PP) was measured by submerging filters in 3 ml of oxidative solution (made as for DOP analysis) in tightly-capped glass centrifuge tubes and autoclaving them for 30 min at 120°C. Samples were then diluted between 1:40 and 1:100 with artificial seawater and analysed as for inorganic  $\text{PO}_4^{3-}$ ; the high dilution in this case eliminates the need for additional ascorbic acid. Oxidation efficiency was monitored with glucose-6-phosphate and was always >88%.

#### 2.4.3 Isotope analyses

Stable carbon and nitrogen isotopes of POC and PN ( $\delta^{13}\text{C}$ -POC and  $\delta^{15}\text{N}$ -PN) were measured with a separate set of filters from POC and PN concentration measurements, for which a

larger volume of water was filtered to yield higher instrument signals. These filters were not acid-fumed, because calcifying plankton species are essentially absent from these waters. Samples were otherwise prepared and analysed as for POC/PN analysis, but without using acetanilide concentration standards. Instead, the analysis was calibrated with isotope standards USGS40 and 41a. Analytical precision was assessed with acetanilide standard 1 from University of Indiana, and was  $\pm 0.2\%$  or better for both  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ .

$\delta^{15}\text{N}$ - and  $\delta^{18}\text{O}$ - $\text{NO}_x^-$  was analysed on a subset of samples from Kusu in 2019–2020 for which the  $\text{NO}_3^-$  concentration was sufficiently high. Analysis was performed using an IRMS after converting all  $\text{NO}_x^-$  into  $\text{N}_2\text{O}$  with the denitrifying bacterium *Pseudomonas chlororaphis* strain ATCC 13985 (Sigman et al., 2001). During the analysis, the standard reference materials IAEA- $\text{NO}_3^-$  ( $-1.8\%$  vs. air) and USGS 34 ( $4.7\%$  vs. air) were prepared every 8-10 samples to constrain the performance of the bacterial conversion and the stability of the mass spectrometer. The stability of the mass spectrometer was further monitored by a series of prepared  $\text{N}_2\text{O}$  gas vials at the beginning and the end of each run and every 10 samples. The average standard deviation of the  $\delta^{15}\text{N}$  measurements on sample replicates was less than  $\pm 0.1\%$ .

$\delta^{13}\text{C}$ -DIC was analysed partly at the UC Davis Stable Isotope Laboratory and partly at Nanyang Technological University using a Gas Bench connected to a Thermo Delta V IRMS, as described by Zhou et al. (2021). Analytical precision was  $\pm 0.1\%$  at UC Davis and  $\pm 0.2\%$  for our in-house analysis.

#### 2.4.4 Chlorophyll-*a*

Chlorophyll-*a* was measured on a Horiba Fluoromax4 spectrofluorometer at excitation 436 nm and emission 680 nm (both with bandpass of 5 nm) according to Welschmeyer (1994), after extracting the samples in 90% acetone at  $+4^\circ\text{C}$  overnight in the dark. Samples were

thoroughly mixed and centrifuged at 500 rcf for 10 min prior to analysis. Calibration was performed using spinach chlorophyll-*a* (Sigma-Aldrich, C5753).

## *2.5 Sensor data quality control and analysis*

The Valeport CTD profiles were used to calculate the mean salinity, temperature, and chlorophyll-*a* between 4.5 and 5.5 m depth to accompany our water sample data. The chlorophyll fluorometer malfunctioned and returned values consistently below 0.1  $\mu\text{g l}^{-1}$  between October 2018 and July 2019 until the fault was repaired; these measurements have been omitted.

All data from the moored sensors were checked manually to remove spikes and periods with suspect data, e.g. when heavy biofouling was noted on the sensor parts. For dates when both SeaCAT and Valeport data were available, the Valeport data were compared to the SeaCAT measurement closest in time; except for two outliers in salinity, the data agreed with root mean squared error (RMSE) of  $\pm 0.07$  (salinity) and  $\pm 0.08^\circ\text{C}$  (Fig. S1).

The SeaFET data were compared to pH calculated from TA and DIC, which were measured during each sampling date starting in August 2018. pH was calculated using the R package *seacarb* (Gattuso et al., 2016) based on the Valeport salinity and temperature and the measured dissolved inorganic nutrient concentrations. We used the dissociation constants from Lueker et al. (2000) for carbonic acid, from Dickson (1990) for  $\text{HSO}_4^-$ , and from Perez and Fraga (1987) for fluoride; total boron concentration followed Uppström (1974). The SeaFET pH data were first reprocessed based on the salinity record from the SeaCAT, and were then compared to the calculated pH based on TA and DIC measurements. For dates when no SeaCAT data were available, a salinity of 30 was assumed for the SeaFET data processing (note that the salinity correction is too small to impact our results significantly). A direct comparison of our TA+DIC-based calculated pH with the closest matching individual

SeaFET pH measurement for all dates when good-quality SeaFET measurements were available within 3 hours of the water sampling showed that the data clustered around the 1:1 line with root mean squared error of 0.028 pH units across a pH range of 7.85–8.01 (Figure S1).

Lab-based measurements of dissolved oxygen concentration are not available for comparison to the miniDOT. Hence, the data were only examined qualitatively to remove periods where either the diel amplitude increased considerably or we observed a clear decrease in daily average concentration together with a large increase in diel amplitude. These cases were suspected to be caused by biofouling and/or accumulation of excessive amounts of suspended particulate matter on the sensor interface, as the variability decreased again after sensor cleaning.

## *2.6 Data analysis and previous data use*

All data analysis and plotting were carried out in R (R Core Team, 2020). To test for seasonal variation, we used non-parametric Kruskal-Wallis tests with post-hoc Dunn tests. All averages are quoted as mean  $\pm$  standard deviation unless specified otherwise.

The majority of the data for salinity, temperature, DOC, CDOM, chlorophyll-*a*,  $\delta^{13}\text{C}$ -DOC,  $\delta^{13}\text{C}$ -DIC, and the daily averages of seawater pH were previously used in our analyses of carbonate system variability (Zhou et al., 2021) and bio-optical variability (Martin et al., 2021). The data for dissolved inorganic nitrogen,  $\text{PO}_4^{3-}$ , and chlorophyll-*a* are also used in an analysis of alkaline phosphatase activity in the Singapore Strait (Nichols et al., submitted manuscript).

## **3. Results**

### 3.1 Hydrographic variability

Salinity and temperature ranged typically between 29–33 and 28–31°C and showed pronounced seasonal variation, with highest values during the late NE monsoon and intermonsoon 1 seasons (February to early May), and also during intermonsoon 2 (mid-September to mid-November) (Fig. 2). Salinity and temperature decreased during both monsoon seasons, but with distinct patterns: salinity was lowest throughout the SW monsoon (mid-May to mid-September), but only decreased for a shorter period during the early NE monsoon (mid-November to January) and not always by as much as during the SW monsoon. Temperature decreased more strongly during the NE monsoon and less during the SW monsoon. While temperature was essentially identical between both sites, the salinity during the SW monsoon was consistently lower at Hantu than at Kusu by an average of  $0.49 \pm 0.22$ . Both sites typically showed little vertical density variation without a clear pycnocline (Fig. S2), confirming that the water column is generally well mixed.

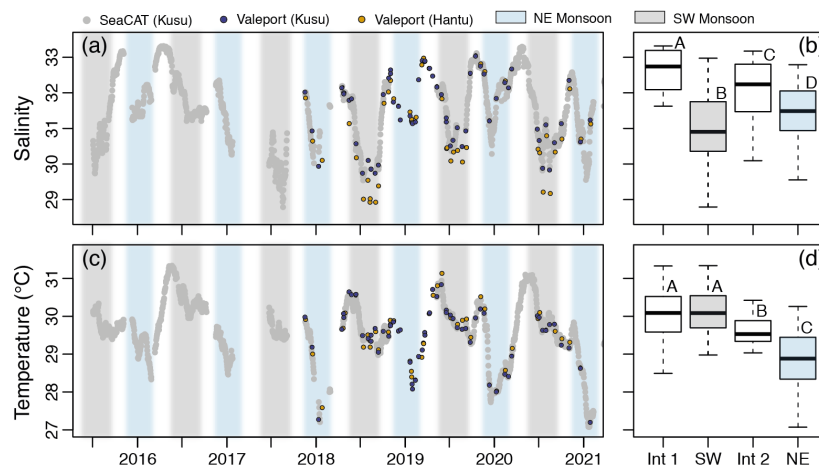


Fig. 2. Time series and seasonal boxplots of (a,b) salinity and (c,d) temperature in the Singapore Strait. Grey shading indicates the southwest (SW) monsoon, blue shading indicates the northeast (NE) monsoon. In the boxplot x-axis, Int 1 = intermonsoon 1; SW = southwest monsoon; Int 2 = intermonsoon 2; NE = northeast monsoon. Data from the Seabird SeaCAT moored CTD (at Kusu) are daily mean values; data from the Valeport FastCTD are from profiling measurements averaged at 5 m. Letters in the boxplots indicate significant differences between seasons (Kruskal-Wallis with post-doc Dunn test,  $p < 0.05$ ).

### 3.2 Dissolved inorganic nutrients

Except for  $\text{NH}_4^+$ , the dissolved inorganic nutrient concentrations also showed very strong and statistically significant seasonal variation, with elevated concentrations during the SW monsoon, a smaller and shorter increase during the early NE monsoon, and mostly very low concentrations (often close to or below detection limits) during both intermonsoon periods (Fig. 3; Table 1). Concentrations of  $\text{NO}_3^- + \text{NO}_2^-$  reached 4–5  $\mu\text{mol l}^{-1}$  during the SW monsoon, with  $\text{NO}_2^-$  typically contributing 0.5–1.0  $\mu\text{mol l}^{-1}$  during this time, but  $\text{NH}_4^+$  was only sporadically detectable (usually during the NE monsoon), with highest concentrations during 2018 (Fig. 3a-f).  $\text{PO}_4^{3-}$  showed very similar seasonal patterns to  $\text{NO}_3^- + \text{NO}_2^-$ , with values ranging from below detection to around 0.35  $\mu\text{mol l}^{-1}$  (Fig. 3g,h).  $\text{Si(OH)}_4$  differed slightly from the other nutrients in never dropping below detection limits, and was  $>2 \mu\text{mol l}^{-1}$  in all but seven samples, reaching up to 13.8  $\mu\text{mol l}^{-1}$  (Fig. 3i,j). Moreover,  $\text{Si(OH)}_4$  showed clearer peaks during the NE monsoon than the other nutrients.

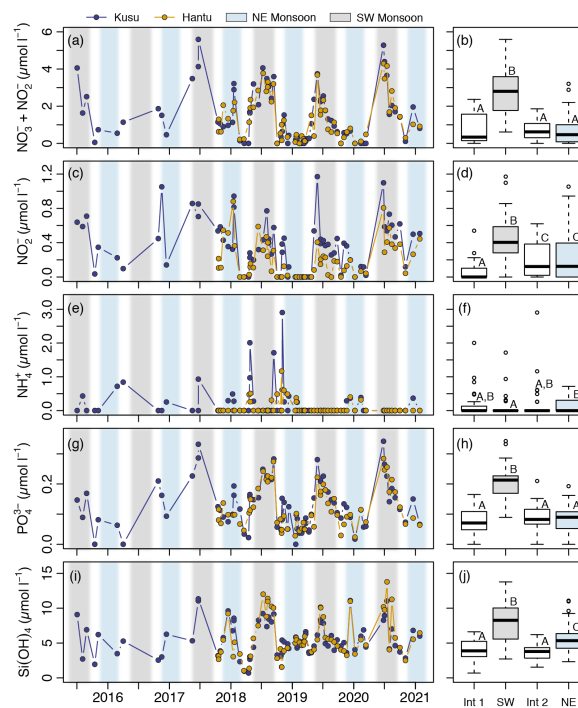


Fig. 3. Time series and seasonal boxplots of dissolved inorganic nutrient concentrations in the Singapore Strait.

Colour shading and boxplot labelling are as in Fig. 2.

Parameter		Intermonsoon 1	SW monsoon	Intermonsoon 2	NE monsoon	$\chi^2$	p
Salinity (daily mean)	Min, Max	31.63, 33.32	28.79, 32.97	30.09, 33.17	29.55, 32.79	547	<0.0001
	Mean (SD)	32.63 (0.53)	31.01 (0.85)	32.09 (0.79)	31.44 (0.70)		
	Median	32.74	30.90	32.24	31.49		
Temperature (daily mean)	Min, Max	28.49, 31.33	28.98, 31.34	29.03, 30.42	27.07, 30.26	662	<0.0001
	Mean (SD)	30.06 (0.66)	30.14 (0.52)	29.64 (0.37)	28.81 (0.75)		
	Median	30.09	30.08	29.53	28.88		
pH (daily mean)	Min, Max	7.942, 7.999	7.821, 7.99	7.859, 8.033	7.865, 8.035	440	<0.0001
	Mean (SD)	7.968 (0.013)	7.896 (0.037)	7.937 (0.048)	7.980 (0.034)		
	Median	7.964	7.888	7.937	7.987		
Dissolved O <sub>2</sub> (daily mean)	Min, Max	155, 193	164, 195	169, 207	155, 198	43.6	<0.0001
	Mean (SD)	181 (9.8)	178 (6.1)	184 (5.8)	180 (11.7)		
	Median	183	179	183	180		
Diel change in pH	Min, Max	0.014, 0.103	0.019, 0.1589	0.017, 0.1444	0.010, 0.162	262	<0.0001
	Mean (SD)	0.056 (0.020)	0.068 (0.023)	0.059 (0.022)	0.041 (0.019)		
	Median	0.0563	0.0662	0.0556	0.0366		
Diel change in dissolved O <sub>2</sub>	Min, Max	15.3, 78.3	13.5, 80.5	11.8, 69.5	10.0, 63.4	31.1	<0.0001
	Mean (SD)	39.5 (13.2)	35.3 (13.8)	32.8 (12.6)	28.3 (10.2)		
	Median	40.1	32.2	31.5	27.0		
NO <sub>3</sub> <sup>-</sup> + NO <sub>2</sub> <sup>-</sup>	Min, Max	<0.05, 2.37	0.613, 5.60	<0.05, 1.86	<0.05, 3.21	71.6	<0.0001
	Mean (SD)	0.803 (0.817)	2.74 (1.16)	0.704 (0.523)	0.733 (0.816)		
	Median	0.332	2.80	0.621	0.467		
NO <sub>2</sub> <sup>-</sup>	Min, Max	<0.01, 0.538	<0.01, 1.17	<0.01, 0.620	<0.01, 1.05	42.9	<0.0001
	Mean (SD)	0.071 (0.126)	0.451 (0.251)	0.208 (0.201)	0.241 (0.292)		
	Median	<0.01	0.404	0.122	0.125		
NH <sub>4</sub> <sup>+</sup>	Min, Max	<0.25, 2.01	<0.25, 1.72	<0.25, 2.90	<0.25, 0.719	8.78	0.032
	Mean (SD)	0.211 (0.474)	0.075 (0.285)	0.210 (0.575)	0.149 (0.200)		
	Median	<0.25	<0.25	<0.25	<0.25		
PO <sub>4</sub> <sup>3-</sup>	Min, Max	<0.016, 0.165	0.089, 0.342	<0.016, 0.210	<0.016, 0.193	82.3	<0.0001
	Mean (SD)	0.080 (0.045)	0.206 (0.054)	0.089 (0.041)	0.085 (0.042)		
	Median	0.071	0.213	0.083	0.090		
Si(OH) <sub>4</sub>	Min, Max	0.688, 6.62	2.71, 13.77	1.54, 6.20	2.32, 11.08	61.6	<0.0001
	Mean (SD)	3.89 (1.69)	7.88 (2.57)	3.70 (1.14)	5.60 (2.09)		
	Median	3.89	8.26	3.79	5.35		
POC	Min, Max	8.74, 19.9	6.78, 29.8	8.33, 22.3	7.84, 22.3	11.9	0.008
	Mean (SD)	13.5 (3.48)	11.7 (4.21)	14.0 (3.28)	14.2 (3.81)		
	Median	13.6	11.2	13.6	13.2		
PN	Min, Max	1.41, 3.63	0.97, 5.35	1.49, 3.32	1.23, 4.19	7.89	0.048
	Mean (SD)	2.39 (0.75)	2.08 (0.80)	2.46 (0.51)	2.50 (0.80)		
	Median	2.18	2.06	2.58	2.16		
PP	Min, Max	0.02, 0.19	0.01, 0.16	0.05, 0.20	0.05, 0.22	22.6	<0.0001
	Mean (SD)	0.08 (0.04)	0.07 (0.03)	0.10 (0.04)	0.11 (0.04)		
	Median	0.08	0.07	0.08	0.11		
$\delta^{13}\text{C}$ -POC	Min, Max	-24.8, -17.1	-24.2, -18.0	-23.2, -19.3	-22.3, -18.0	21	0.0001
	Mean (SD)	-20.9 (1.6)	-21.8 (1.4)	-21.0 (1.0)	-20.1 (1.2)		
	Median	-20.7	-21.9	-20.9	-20.3		
$\delta^{15}\text{N}$ -PN	Min, Max	1.8, 7.5	1.9, 6.3	3.8, 7.0	1.0, 6.5	14.7	0.002
	Mean (SD)	4.2 (1.3)	4.0 (1.3)	5.1 (0.7)	4.8 (1.2)		
	Median	4.2	3.8	5.1	5.2		
DOC	Min, Max	66.0, 85.6	65.9, 108	62.6, 77.2	55.4, 91.1	55.1	<0.0001
	Mean (SD)	74.8 (4.9)	84.6 (9.7)	69.3 (3.4)	77.9 (7.1)		
	Median	73.6	84.3	69.1	77.9		
DON	Min, Max	4.6, 11.7	4, 9.7	2.9, 8.9	5.9, 16.6	12.6	0.0057
	Mean (SD)	7.1 (1.7)	7.1 (1.4)	6.5 (1.5)	8.7 (2.6)		
	Median	7.1	7.2	6.6	7.8		
DOP	Min, Max	<0.01, 0.39	<0.01, 0.13	0.03, 0.42	<0.01, 0.5	49.8	<0.0001
	Mean (SD)	0.12 (0.08)	0.03 (0.03)	0.13 (0.10)	0.15 (0.09)		
	Median	0.12	0.02	0.11	0.14		
DOC:DON ratio	Min, Max	6.9, 17.3	7.7, 19.4	7.9, 23.5	3.8, 13	16.0	0.0011
	Mean (SD)	11.1 (2.47)	12.4 (2.70)	11.6 (4.05)	9.7 (1.98)		
	Median	10.8	12.0	10.1	10.3		
DON:DOP ratio	Min, Max	18.6, 681	38.9, 1800	18.1, 146	49.4, 98.9	26.7	<0.0001
	Mean (SD)	111 (150)	309 (366)	63.8 (42.9)	63.9 (15.2)		
	Median	59.6	155	46.3	62.1		
Chl-a (CTD- and filter data)	Min, Max	0.43, 1.72	0.38, 2.55	0.56, 2.60	0.27, 2.39	9.22	0.0266
	Mean (SD)	0.89 (0.31)	0.97 (0.46)	1.42 (0.65)	1.17 (0.58)		
	Median	0.84	0.88	1.32	1.01		

Table 1. Summary values of physical and biogeochemical properties by season.  $\chi^2$ - and p-values show results of

Kruskal-Wallis tests for differences between seasons (all d.f. = 3).



### 3.3 Dissolved and particulate organic matter and stable isotope ratios

DOC showed a clear seasonal cycle with a typical peak of 80–100  $\mu\text{mol l}^{-1}$  during both the SW and the NE monsoon, and lower values (usually 65–70  $\mu\text{mol l}^{-1}$ ) during the intermonsoon seasons (Fig. 4a,b). DON showed less clear seasonality, ranging mostly between 4–12  $\mu\text{mol l}^{-1}$ , but was slightly and significantly higher during the NE monsoon (Fig. 4c,d). In contrast, DOP had clear seasonal variation, with very low concentrations during the SW monsoon (when DOP was sometimes not detected) and a peak of ~0.1–0.2  $\mu\text{mol l}^{-1}$  during the intermonsoon and NE monsoon periods (Fig. 4e,f).

Unlike the dissolved nutrients, particulate nutrient concentrations had limited seasonal variation, with concentrations ranging mostly between 7–20  $\mu\text{mol l}^{-1}$  for POC, 1–4  $\mu\text{mol l}^{-1}$  for PN, and PP between 0.025–0.2  $\mu\text{mol l}^{-1}$  (Fig. 4g–l). The values of all particulate concentrations were very similar between both sites. However, the concentrations of all three were significantly lower during the SW monsoon and higher during either the intermonsoon or the NE monsoon periods (Fig. 4h,j,l; Table 1). The chlorophyll-*a* concentration ranged mostly between 0.5–2.5  $\mu\text{g l}^{-1}$  with an overall mean of  $1.1 \pm 0.52 \mu\text{g l}^{-1}$  (Fig. 4m). Filter-based chlorophyll-*a* concentrations agreed with the Valeport FastCTD fluorometer with root mean squared error = 0.51  $\mu\text{g l}^{-1}$  (Fig. S3). Combining the Valeport data and the filter-based data showed that chlorophyll-*a* was on average slightly but significantly elevated during intermonsoon 2 (Fig. 4n; Table 1).

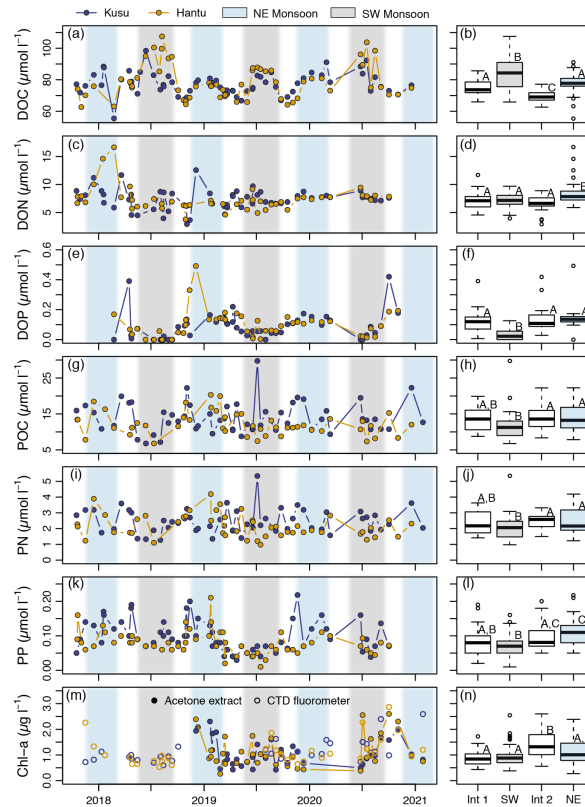


Fig. 4. Time series and seasonal boxplots of dissolved and particulate organic matter and chlorophyll-*a* concentrations. Colour shading and boxplot labelling are as for previous figures. Panel (m) shows data both from filter samples measured after acetone extraction and *in-vivo* measurements from the Valeport FastCTD fluorometer. The boxplot in panel (n) includes the fluorometer data for dates when no filter-based measurements are available.

$\delta^{13}\text{C}$ -POC ranged between -24.8‰ to -17.1‰, while  $\delta^{15}\text{N}$ -PN ranged between +1.0‰ to +7.5‰; both showed moderate but significant differences between seasons, with lower values during the SW monsoon (Fig. 5a–d; Table 1).  $\delta^{13}\text{C}$ -DIC showed an extremely clear seasonal cycle, ranging mostly from -1.5‰ to +0.5‰, with low values during the SW monsoon and high values during all other seasons (Fig. 5e,f).  $\delta^{15}\text{N}$ - $\text{NO}_x^-$  varied between +2.6‰ to +8.5‰ while  $\delta^{18}\text{O}$ - $\text{NO}_x^-$  ranged from -0.3‰ to +9.0‰ (Fig. 5g).  $\text{NO}_3^-$  concentrations were only sufficiently high during the SW and early NE monsoon to allow the isotopic composition to be measured; hence we did not test for seasonality. However, the three measurements from

the NE monsoon had the lowest  $\delta^{15}\text{N}$  values. There was no significant relationship between  $\delta^{18}\text{O}-\text{NO}_x^-$  and  $\delta^{15}\text{N}-\text{NO}_x^-$ , and the values could be consistent with either marine or terrestrial  $\text{NO}_3^-$  sources (Fig. 6).

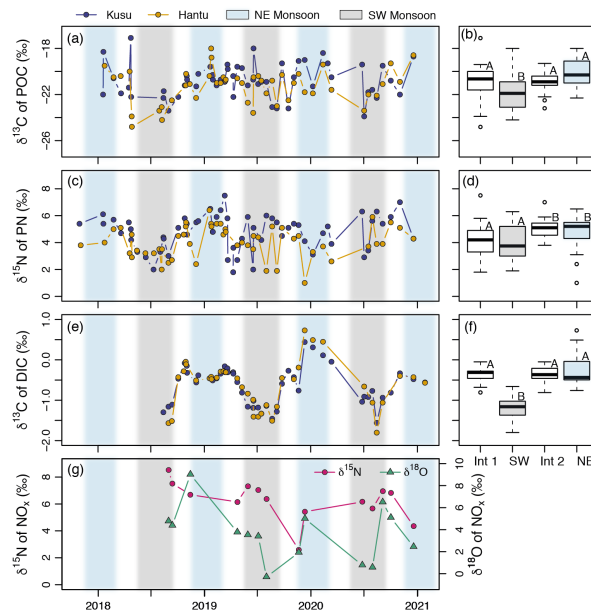


Fig. 5. Time series and seasonal boxplots of the isotopic composition ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) of (a–d) particulate organic matter, and (e,f) dissolved inorganic carbon. Time series of  $\delta^{15}\text{N}$  and  $\delta^{18}\text{O}$  of  $\text{NO}_x^-$  (measured only at Kusu) is shown in (g). Since  $\text{NO}_x^-$  isotopes were only measured for dates with sufficiently high  $\text{NO}_3^-$  concentration, the seasonality was not analysed. Colour shading and boxplot labelling are as for previous figures.

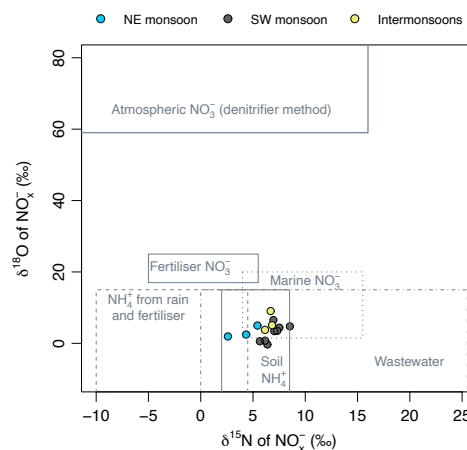


Fig. 6. Isotopic composition of dissolved  $\text{NO}_x^-$  at Kusu Island, together with approximate ranges of the main potential nitrogen sources drawn according to Kendall et al. (2007).

### 3.4 Nutrient stoichiometry and relationships between parameters

Regression statistics between parameters are given in Table 2. Dissolved inorganic nitrogen (DIN) showed a strong relationship to dissolved  $\text{PO}_4^{3-}$  that was close to the canonical Redfield ratio of 16:1, but with a tendency towards excess  $\text{PO}_4^{3-}$  (Fig. 7a). DIN was also closely related to  $\text{Si(OH)}_4$ , but with a considerable excess of silicon: the DIN: $\text{Si(OH)}_4$  ratio was invariably lower than 1, and the regression slope was 0.30 (Fig. 7b). POC and PN also showed a strong relationship that was close to the 6.6:1 Redfield stoichiometry (Fig. 7c),

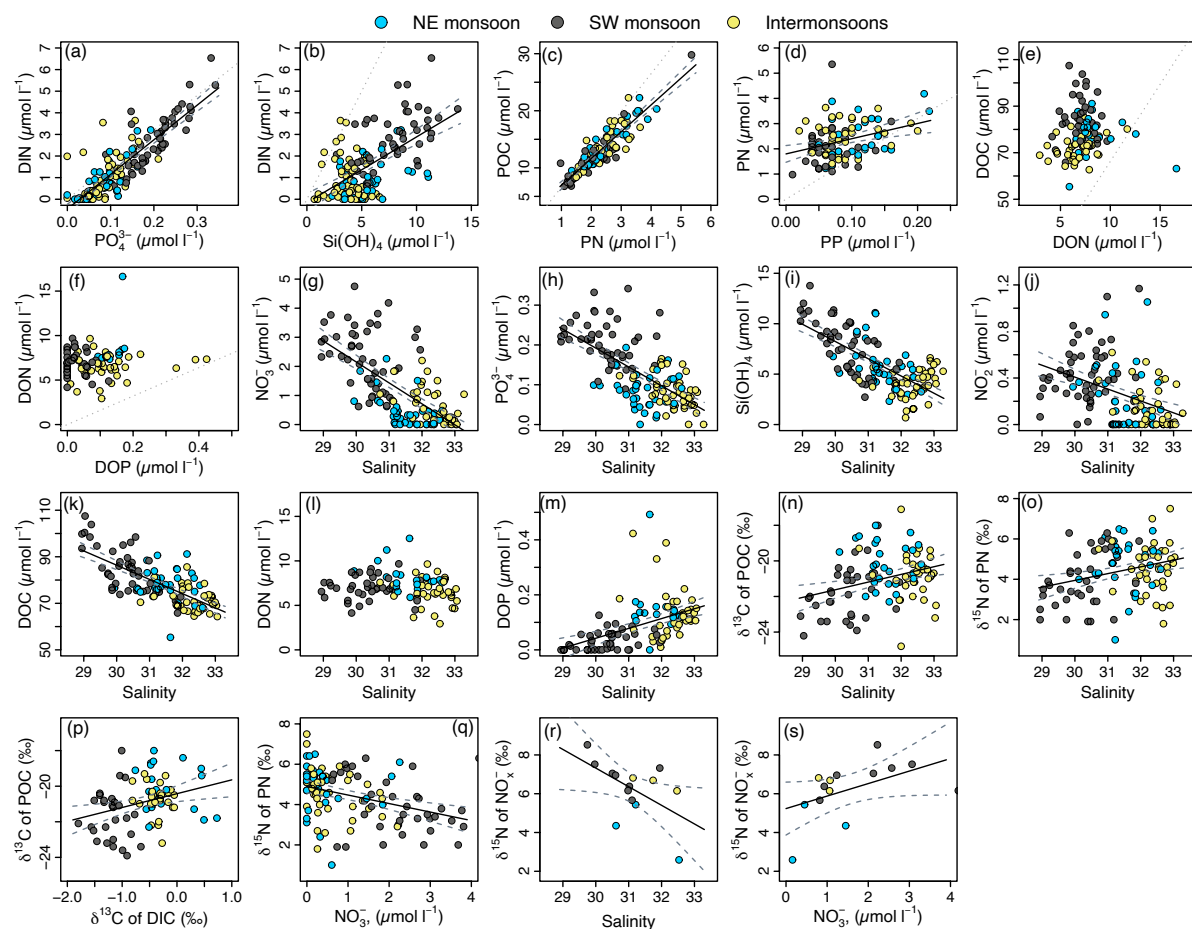


Fig. 7. Scatter plots between parameters to test for relationships and stoichiometric ratios. Solid black lines with dark-grey dashed lines indicate linear regression slopes with 95% confidence intervals; for regression parameters, see Table 2. Dotted grey lines in panels a–f indicate the canonical Redfield ratios (C:N:P = 106:16:1, N:Si = 1:1).

while PN and PP were less strongly related, with an N:P ratio above Redfield (mean of  $33 \pm 20 \text{ mol mol}^{-1}$ ; Fig. 7d). There was also no strong relationship between DOC and DON (Fig. 7e) or DON and DOP (Fig. 7f); the DOC:DON ratio was enriched in carbon relative to the Redfield value of 6.6, while the DON:DOP ratio was enriched in nitrogen relative to the Redfield value of 16 (Fig. 7e,f). Neither the POC:PN nor the PN:PP ratio showed significant seasonal variation, but both the DOC:DON and the DON:DOP ratio were significantly higher during the SW monsoon than at other times (Table 1). There was no significant relationship between POC and chlorophyll-*a* concentration, and the POC:chlorophyll-*a* ratio was on average relatively high (mean of  $182 \pm 113 \text{ g g}^{-1}$ ).

Parameters (y versus x)	Slope estimate (SE)	Intercept estimate (SE)	Statistics
DIN versus $\text{PO}_4^{3-}$	16.0 (0.77)	-0.45 (0.11)	$r^2=0.753$ , $p<0.0001$
DIN versus $\text{Si(OH)}_4$	0.304 (0.035)	-0.16 (0.22), NS	$r^2=0.355$ , $p<0.0001$
POC versus PN	4.69 (0.22)	2.26 (0.53)	$r^2=0.819$ , $p<0.0001$
PN versus PP	6.03 (1.74)	1.80 (0.17)	$r^2=0.111$ , $p<0.001$
$\text{NO}_3^-$ versus salinity	-0.720 (0.068)	23.7 (2.16)	$r^2=0.461$ , $p<0.0001$
$\text{PO}_4^{3-}$ versus salinity	-0.047 (0.004)	1.61 (0.139)	$r^2=0.467$ , $p<0.0001$
$\text{Si(OH)}_4$ versus salinity	-1.71 (0.15)	59.5 (4.80)	$r^2=0.494$ , $p<0.0001$
$\text{NO}_2^-$ versus salinity	-0.102 (0.020)	3.48 (0.61)	$r^2=0.176$ , $p<0.0001$
DOC versus salinity	-6.31 (0.581)	276 (18.2)	$r^2=0.504$ , $p<0.0001$
DOP versus salinity	0.0358 (0.007)	-1.034 (0.221)	$r^2=0.214$ , $p<0.0001$
$\delta^{13}\text{C}$ -POC versus salinity	0.432 (0.132)	-34.6 (4.15)	$r^2=0.102$ , $p<0.01$
$\delta^{15}\text{N}$ -PN versus salinity	0.336 (0.112)	-6.12 (3.53), NS	$r^2=0.081$ , $p<0.01$
$\delta^{15}\text{N}$ - $\text{NO}_x^-$ versus salinity	-0.961 (0.422)	36.1 (13.1)	$r^2=0.302$ , $p<0.05$
pH anomaly versus dissolved $\text{O}_2$ anomaly (10-min frequency)	$1.705 \times 10^{-3}$ ( $7.95 \times 10^{-6}$ )	$-1.883 \times 10^{-5}$ ( $5.606 \times 10^{-5}$ ), NS	$r^2=0.653$ , $p<0.0001$
Diel pH change versus diel dissolved $\text{O}_2$ change	$1.079 \times 10^{-3}$ ( $1.25 \times 10^{-4}$ )	0.0288 (0.0045)	$r^2=0.299$ , $p<0.0001$

Table 2. Results of regression analyses for relationships between key biogeochemical and physical parameters. Intercepts that are not statistically significant are marked NS. p-values refer to the significance of the regression slopes.

NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, and Si(OH)<sub>4</sub>, and to a lesser degree NO<sub>2</sub><sup>-</sup>, all showed strong inverse relationships with salinity (Fig. 7g–j). This was also clearly seen for DOC, but not for DON (which seemed to show a unimodal relationship), while DOP was positively related to salinity, but with high variability (Fig. 7k–m). POC, PN, and PP did not show significant relationships to salinity (not shown), but both  $\delta^{13}\text{C}$ -POC and  $\delta^{15}\text{N}$ -PN were weakly related to salinity (Fig. 7n,o). Consequently,  $\delta^{13}\text{C}$ -POC and  $\delta^{15}\text{N}$ -PN were also weakly related to  $\delta^{13}\text{C}$ -DIC and NO<sub>3</sub><sup>-</sup> concentrations, respectively (Fig. 7p,q).  $\delta^{15}\text{N}$ -NO<sub>x</sub><sup>-</sup> showed a clear inverse relationship to salinity and consequently a positive relationship with NO<sub>3</sub><sup>-</sup> concentrations (Fig. 7r,s). Parameters that were significantly related to salinity were also significantly related to CDOM (as  $a_{350}$ ), mostly with equal or slightly higher  $r^2$  values (Fig. S4; Table S1).

### 3.5 Dissolved O<sub>2</sub> and pH dynamics

Seawater pH varied mostly between 7.80–8.05, showing appreciable high-frequency variability. Moreover, there was a strong seasonal difference in daily mean pH, with lowest values during the SW monsoon (Fig. 8a,b, Table 1). In contrast, dissolved O<sub>2</sub> chiefly showed high-frequency variability between typically 150–210  $\mu\text{mol l}^{-1}$  (Fig. 8c). Although seasonal differences in daily mean dissolved O<sub>2</sub> were technically significant, their magnitude was too small to be environmentally relevant (<5  $\mu\text{mol l}^{-1}$  difference in medians; Fig. 8d, Table 1). Plotting the daily anomaly of pH and dissolved O<sub>2</sub> against time revealed a clear diel cycle with a minimum at 05:30–06:30 local time and a maximum at 14:00–15:00; the average diel change was 0.022 pH units and 14.2  $\mu\text{mol O}_2 \text{ l}^{-1}$  (Fig. 8e,f). There was a strong relationship between the pH and dissolved O<sub>2</sub> anomalies at 10-min frequency (Fig. 8g), and a relationship between the diel change in pH and dissolved O<sub>2</sub> (Fig. 8h). The diel variation in both pH and of dissolved O<sub>2</sub> showed some evidence of seasonal variability, with diel pH change greatest

during the SW and lowest during the NE monsoon, and diel dissolved O<sub>2</sub> change greatest during intermonsoon 1 and lowest during the NE monsoon (Fig. 8i–l, Table 1).

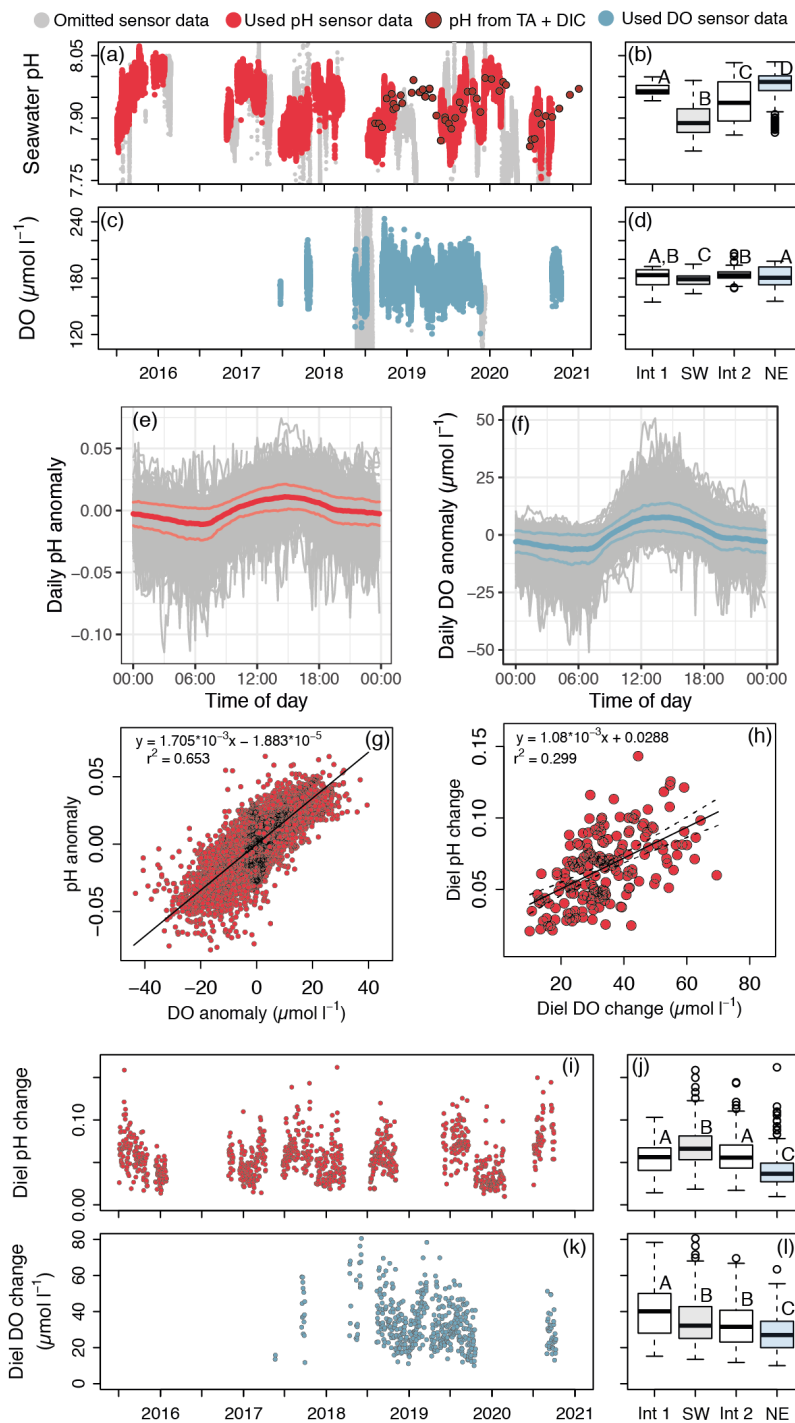


Fig. 8. Time series and seasonal boxplots of (a,b) seawater pH and (c,d) dissolved O<sub>2</sub> at Kusu. Diel anomalies of (e) seawater pH and (f) dissolved O<sub>2</sub>; grey lines show individual daily data, solid blue and red lines show overall mean ± standard deviation. Relationships between (g) individual diel anomalies in pH and dissolved O<sub>2</sub> at 10-min frequency, and (h) between the maximum diel change in pH and dissolved O<sub>2</sub> (on point for each day). Solid black and dashed grey lines indicate linear regression with 95% confidence interval. Time series and seasonal boxplots of maximum diel change in (i,j) seawater pH and (k,l) dissolved oxygen. Monsoon shading in the time series has been omitted for

clarity; boxplot labelling is as for previous figures.

## 4. Discussion

### 4.1 Seasonality and range in dissolved nutrient concentrations

Dissolved inorganic nutrient concentrations have rarely been reported from the Singapore Strait, and only for shorter time periods and/or with lower measurement frequency. The values we measured are broadly consistent with previous studies (Browne et al., 2015; Chénard et al., 2019; Deignan and McDougald, 2021; Gin et al., 2000), which also noted that dissolved nutrient concentrations are typically elevated during the SW monsoon. Our data clearly show that the Singapore Strait undergoes strong and consistent seasonal variation from low concentrations typical of oligotrophic waters (especially during and after the late NE monsoon) to significantly higher values during the SW monsoon (Fig. 3). Our nutrient concentrations during the late NE monsoon and intermonsoon 1 are similar to reports from the open Sunda Shelf and the South China Sea (Kartadikaria et al., 2015; Liu et al., 2020; Ning et al., 2004). In contrast, nutrient concentrations during the SW monsoon are similar to values reported from the Malacca Strait (Lim et al., 2021; Lim et al., 2015). However, chlorophyll-*a* concentrations were above  $0.4 \mu\text{g l}^{-1}$  in all but two samples, with an average of about  $1 \mu\text{g l}^{-1}$  (Fig. 4). This is greater than reported from the open Sunda Shelf Sea (Kartadikaria et al., 2015; Ke et al., 2014; Liu et al., 2020), and indicates that the Singapore Strait does not experience genuinely oligotrophic conditions, even when dissolved inorganic nutrients are low. For comparison, eutrophication thresholds for coral reefs are typically considered to be  $\sim 1 \mu\text{mol l}^{-1}$  DIN,  $0.1\text{--}0.2 \mu\text{mol l}^{-1}$   $\text{PO}_4^{3-}$ , and  $0.2\text{--}0.4 \mu\text{g l}^{-1}$  chlorophyll-*a* (Bell, 1992; Bell et al., 2014).

The dissolved organic matter pool showed a somewhat different seasonal pattern. While DOC showed similar seasonality to the inorganic nutrients, as described previously (Zhou et al., 2021), the DON concentration showed very limited seasonal changes, and the DOP



concentration showed an opposite seasonal pattern, with lowest concentrations during the SW monsoon and highest during the NE and intermonsoon (Fig. 4). The DOC and DON concentrations are similar to those in other coastal locations in the tropical Indo-Pacific (Kuwahara et al., 2010; Lønborg et al., 2021a; Martin et al., 2018). The DOP concentrations in Singapore are lower than reported from the Great Barrier Reef by Lønborg et al. (2021a) (average of  $0.21 \pm 0.16 \mu\text{mol l}^{-1}$ ), but similar to recent reports from coastal locations around the Malay Peninsula (Lim et al., 2018) (average of  $0.07 \pm 0.03 \mu\text{mol l}^{-1}$ ). The fact that DOP was sometimes undetectable during the SW monsoon (i.e. the total dissolved P concentration was equal to the dissolved  $\text{PO}_4^{3-}$  measurement) might indicate that a refractory DOP fraction was incompletely oxidised, or alternatively that a labile DOP fraction was hydrolysed during dissolved  $\text{PO}_4^{3-}$  measurements. However, the close agreement between our two sites and consistent seasonal patterns between years clearly suggest that at least the relative variation in DOP is accurate.

Overall, and taken together with the strong seasonality in salinity (Fig. 2), our results confirm that dissolved biogeochemical properties in the Singapore Strait are largely controlled by the regional advection of distinct water masses according to the monsoon-driven changes in ocean currents, as previously described for DOC and for coloured dissolved organic matter (Martin et al., 2021; Zhou et al., 2021). Our data also provide further support for classifying the Singapore Strait as a mesotrophic environment (Lim et al., 2021) despite the seasonally low concentrations of inorganic nutrients.

#### *4.2 Sources of nutrients*

The strong inverse relationships of  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$ , and  $\text{Si}(\text{OH})_4$  to salinity (Fig. 7) clearly indicate a terrestrial source from regional river input. Using stable carbon isotopes, Zhou et al. (2021) demonstrated that a large fraction of DOC in the Singapore Strait during the SW

monsoon originates from regional peatlands. The fact that all three nutrients were more strongly correlated with CDOM than with salinity (as seen by the higher  $r^2$  values; Fig. S4) provides further support for a predominantly terrestrial source of nutrients in the Singapore Strait, as CDOM is a good tracer of terrestrial DOC in the Sunda Shelf Sea (Martin et al., 2018; Martin et al., 2021; Mizubayashi et al., 2013; Siegel et al., 2019).

The intercepts of our nutrient–salinity regressions can be taken as estimated riverine end-member concentrations. This implies average river concentrations of  $23.7 \pm 2.2 \mu\text{mol l}^{-1}$  for  $\text{NO}_3^-$ ,  $1.61 \pm 0.14 \mu\text{mol l}^{-1}$  for  $\text{PO}_4^{3-}$ , and  $59.5 \pm 4.8 \mu\text{mol l}^{-1}$  for  $\text{Si}(\text{OH})_4$  (Fig. 7, Table 2). This is at the low end of  $\text{Si}(\text{OH})_4$  concentrations in small tropical rivers in Asia (Jennerjahn et al., 2006), and our inferred riverine end-member values of all three nutrients are several-fold lower than reported from human-impacted river systems on Java (Damar et al., 2019; Jennerjahn et al., 2004). Tropical peatland-draining rivers carry low nutrient concentrations, often  $<5 \mu\text{mol l}^{-1}$  DIN, unless they are affected by anthropogenic nutrient inputs (Alkhatib et al., 2007; Baum, 2008), consistent with efficient nutrient retention within the peatland ecosystem (Mishra et al., 2021). However, our inferred riverine end-member concentrations are well within the range reported for peatland-draining rivers with varying degrees of anthropogenic impact from Sumatra and Borneo (Alkhatib et al., 2007; Bange et al., 2019; Baum, 2008; Gandois et al., 2020; Jiang et al., 2019).

The fact that  $\text{NO}_2^-$  was only weakly related to salinity and CDOM (Fig. 7), while  $\text{NH}_4^+$  was unrelated to salinity and was consistently low during the SW monsoon (Fig. 3), suggests that these two nitrogen species originate primarily from internal recycling, rather than being derived directly from land.  $\text{NO}_2^-$  and  $\text{NH}_4^+$  concentrations in peatland rivers have only been reported from the Siak and Rajang rivers, where  $\text{NO}_2^-$  is typically  $<0.5 \mu\text{mol l}^{-1}$  but  $\text{NH}_4^+$  is usually  $2\text{--}15 \mu\text{mol l}^{-1}$  (Baum, 2008; Jiang et al., 2019). Our data therefore suggest that riverine  $\text{NH}_4^+$  is rapidly transformed in the estuaries and coastal waters, likely through a

combination of phytoplankton uptake and nitrification, as also noted along the salinity gradient of the Rajang delta on Borneo (Jiang et al., 2019). Recycling of organic nitrogen (either terrestrial or aquatic in origin) by ammonification followed by nitrification could also act as a source of  $\text{NO}_2^-$  in coastal waters. In subtropical coastal waters of North America, temperature-dependent  $\text{NO}_2^-$  accumulation can occur when  $\text{NH}_4^+$  oxidation and  $\text{NO}_2^-$  oxidation become uncoupled between 20–30°C, owing to the different temperature sensitivities of the microbial taxa responsible for each of these nitrification steps (Schaefer and Hollibaugh, 2017). Whether the permanently high temperatures in tropical coastal waters might promote  $\text{NO}_2^-$  accumulation over complete nitrification may be worth testing in future research, especially in the context of increasing tropical surface temperatures. Since the water column at our site was always oxygenated (Fig. 8), it is unlikely that hypoxia was mediating nitrification rates (Schaefer and Hollibaugh, 2017).

The  $\delta^{15}\text{N}-\text{NO}_x^-$  and  $\delta^{18}\text{O}-\text{NO}_x^-$  values are slightly higher compared to values measured in the Rajang River in northwest Borneo (Jiang et al., 2019), but do not provide an unambiguous source fingerprint (Fig 6; (Kendall et al., 2007)). Although the isotopic composition could be consistent with marine  $\text{NO}_3^-$ , this is only plausible for the NE monsoon period, given the strong inverse relationship of  $\text{NO}_3^-$  concentration with salinity. For the SW monsoon, the isotopic data would be consistent with soil N, wastewater, or a combination of fertiliser and wastewater (Fig. 6). Most of the peatlands on Sumatra are used for agriculture and have synthetic fertilisers applied regularly; Baum (2008) reported that fertilisers and wastewater are the main nutrient sources to the Siak, which is one of the main river systems on Sumatra. Although atmospheric deposition was suggested previously to be a significant source of  $\text{NO}_3^-$  to the Singapore Strait (Sundarambal et al., 2010), our isotope data are not consistent with a significant contribution from atmospheric  $\text{NO}_3^-$  deposition during any season. The monthly mean  $\delta^{15}\text{N}-$  and  $\delta^{18}\text{O}-\text{NO}_3^-$  values in rainfall in Singapore are nearly all  $<2\text{‰}$  and  $>50\text{‰}$ ,

respectively (Li et al., 2020). The fact that  $\delta^{15}\text{N-NO}_x^-$  during the NE monsoon had the lowest values measured, even below typical marine  $\text{NO}_3^-$  values (Fig. 6) may point to a significant contribution from N fixation: the shallow subsurface  $\text{NO}_3^-$  pool in the South China Sea is isotopically depleted due to diazotrophy (Ren et al., 2017; Zhang et al., 2020), and additional N fixation is likely to take place across the Sunda Shelf Sea. For example, N fixation was reported from the Singapore Strait even during the DIN-rich SW monsoon, both in the water column and associated with corals, albeit at relatively low rates (Moynihan et al., 2021). In contrast to the main inorganic nutrients and to DOC, neither DON nor DOP appeared to have a predominant terrestrial source, given their different seasonal patterns (Fig. 4). While very low DOP concentrations during the SW monsoon might seem surprising, this provides further support for the view that peatlands are the main source of river run-off influencing the Singapore Strait during the SW monsoon (Zhou et al., 2021). Tropical peatlands are ombrotrophic, nutrient-poor systems that retain nutrients efficiently and can become particularly phosphorus-poor, while nitrogen-fixing vegetation continues to provide a nitrogen source even in old peatlands (Mishra et al., 2021; Troxler, 2007). Hence, the most likely explanation for the DON and DOP dynamics is that the DOP is predominantly of marine origin (and is hence high during the NE monsoon and intermonsoon months), while the DON has both significant marine and terrestrial sources, and hence shows little seasonality. Measurements of both DON and DOP in peatland-draining rivers would be valuable to clarify this further. While disturbed peatlands converted to agricultural use can clearly be a source of inorganic nutrients through fertiliser input, as reflected in our inorganic nutrient data, the dissolved organic matter exported from disturbed peatlands consists of increasingly old soil organic matter (Evans et al., 2014), which would explain the different dynamics of the inorganic and the organic nutrients at our site.

At present, the bioavailability of DON and DOP in the Sunda Shelf Sea is unknown. In the Great Barrier Reef, DON and DOP contributed more bioavailable N and P than either dissolved inorganic or particulate nutrients (Lønborg et al., 2018), but the DOM pool in that study was predominantly of marine origin. In the open South China Sea, isotopic evidence points to active DON cycling with DON production and remineralization rates approximately in balance (Zhang et al., 2020). However, peatland-derived DOC in Southeast Asia appears to be relatively refractory to direct remineralisation by heterotrophic microbes, even when inorganic N and P are added (Nichols and Martin, 2021), suggesting that the terrestrial organic nutrient pools might also be relatively refractory. Nevertheless, the majority of this peatland DOC is ultimately remineralised before leaving the Sunda Shelf (Wit et al., 2018; Zhou et al., 2021), although the biogeochemical processes driving this remineralisation remain unclear. If peatland DON is also remineralised to a similar degree as DOC in the Sunda Shelf, then given the relative lack of DOP, this might lead to an imbalanced N:P ratio in areas receiving peatland run-off. Interestingly, the N:P stoichiometry of the particulate pool is typically higher than the canonical Redfield ratio of 16:1 (Fig. 7), although this might also reflect a contribution from detrital organic matter to the particulate pool (see Section 4.3). The Singapore Strait also has measurable alkaline phosphatase activity year-round (Nichols et al., submitted manuscript), suggesting that least a fraction of DOP is actively cycled. Resolving the bioavailability of both the terrestrial and marine DON and DOP pools will be important both for understanding the patterns of primary productivity in this region, and to reach a more complete understanding of how land use-driven changes in terrestrial DOM fluxes (Moore et al., 2013) affect coastal seas.

#### *4.3 Composition and dynamics of particulate organic matter*

The fact that the particulate organic matter pool and the chlorophyll-*a* concentration showed only relatively limited seasonality and did not directly follow the seasonal cycle in inorganic nutrients indicates that phytoplankton biomass is not primarily controlled by nutrient availability. Even during the low-nutrient intermonsoon periods, chlorophyll-*a* was usually  $>0.5 \mu\text{g l}^{-1}$ , which suggests that nutrients are efficiently recycled during these periods to maintain phytoplankton productivity and standing stock. This is consistent with the observation of year-round measurable levels of alkaline phosphatase activity in the water column, averaging  $9 \text{ nmol l}^{-1} \text{ h}^{-1}$ , most likely from heterotrophic microbial sources (Nichols et al., submitted manuscript). However, chlorophyll-*a* was on average elevated during intermonsoon 2 and partly also during the NE monsoon (Fig. 4). This does suggest that the SW monsoon nutrient input stimulates a delayed phytoplankton growth response. The delay might be linked to greater light limitation during the SW monsoon due to the presence of peatland CDOM, which leads to shoaling of the euphotic zone and changes in the spectral quality of light underwater (Martin et al., 2021). Phytoplankton production in tropical and subtropical estuaries experiencing episodic nutrient delivery is similarly thought to be modulated by light limitation from high turbidity during flood events (Burford et al., 2012; Eyre, 2000), and depth-integrated phytoplankton productivity along the Great Barrier Reef is consistently greater at offshore stations with greater light availability (Furnas et al., 2005). However, this apparent seasonal increase in chlorophyll-*a* concentration was small and not consistent between years, e.g. no increase was seen in 2019 (Fig. 4). It is probable that grazing, especially by microzooplankton, is also a key factor controlling phytoplankton biomass, as noted, for example, in Darwin Harbour and the Great Barrier Reef (Burford et al., 2008; Furnas et al., 2005).

Although the POC:PN ratio was very close to the Redfield ratio, the PN:PP ratio was higher (Fig. 7), and the POC:chlorophyll-*a* ratio was greater than might be expected for

phytoplankton biomass in the relatively low-light environment of the Singapore Strait (Arteaga et al., 2016; Geider et al., 1997). This suggests that a significant part of the particulate organic matter pool was likely detrital. Whether the detrital POM fraction is primarily from marine production or originates from terrestrial input is unclear: the overall mean  $\delta^{13}\text{C}$ -POC ( $-21.0 \pm 1.5\text{‰}$ ) is compatible with a purely marine origin (Fry and Sherr, 1989; Verwege et al., 2021), but the small decrease in  $\delta^{13}\text{C}$ -POC during the SW monsoon (Fig. 5) might indicate a contribution of terrestrial POC during this period. However, the strong decrease in  $\delta^{13}\text{C}$ -DIC during the SW monsoon, caused by peatland DOC remineralisation (Zhou et al., 2021), means that this might also reflect phytoplankton carbon assimilation from a more isotopically depleted DIC pool (Chanton and Lewis, 1999). The  $\delta^{15}\text{N}$ -PN data don't help to resolve this question, as  $\delta^{15}\text{N}$ -PN in Sumatran peatland rivers appears to be mostly in the range of  $+2\text{‰}$  to  $+6\text{‰}$  (Alkhatib et al., 2007; Baum, 2008), essentially the same as our data.

#### 4.4. Net community metabolism

Net community metabolism causes correlated diel changes in pH and dissolved  $\text{O}_2$ , as seen in Fig. 8. On corals reefs, such variation is driven by a combination of photosynthesis–respiration and calcification–carbonate dissolution (Albright et al., 2015; DeCarlo et al., 2017; Page et al., 2019). These previous studies were typically conducted on extensive reef flats with limited water exchange, and they consequently observed larger changes, with diel variation in pH mostly  $\geq 0.1$  unit and in dissolved  $\text{O}_2 \geq 50 \mu\text{mol l}^{-1}$ . In our case, the sensors were installed along a reef slope exposed to the open sea, and the coral reef only extends about 20 m from the shoreline. Therefore, the variation in our dataset is most likely dominated by planktonic photosynthesis and respiration rather than by benthic metabolism and calcification.

The diel change in dissolved O<sub>2</sub> varied by a factor of 3–4 within each season, but was on average highest during intermonsoon 1 and lowest during the NE monsoon (Fig. 8). This pattern suggests that light availability is an important control over primary productivity: Singapore experiences lower rainfall and more solar radiation during the late NE and intermonsoon 1 than in other seasons (Fong, 2012), while CDOM input during the SW monsoon (and to a lesser degree during the early NE monsoon) increases the absorption of sunlight underwater, especially at blue wavelengths (Martin et al., 2021). At the same time, suspended matter concentrations can vary substantially over short (<1 day) periods on reefs in Singapore (Morgan et al., 2020), further contributing to variation in light attenuation and therefore productivity rates within seasons.

Our data therefore indicate that phytoplankton productivity in the Singapore Strait is likely more limited by light than by nutrient availability. While this would imply that increases in nutrient input might not trigger phytoplankton blooms, the tendency for chlorophyll-*a* to increase after the SW monsoon nutrient input (Fig 6) suggests that phytoplankton biomass might still respond to increases in nutrients. This could be the case especially during the low-nutrient intermonsoon periods, when CDOM concentrations are lower. Increases in nutrient concentrations, such as from future aquaculture expansion (Tan, 2020) or from increased riverine supply, might therefore still cause further eutrophication in the strait. Continued monitoring is hence warranted. Our inorganic nutrient data point towards potential N- rather than P-limitation, but also reveal an excess of Si(OH)<sub>4</sub> over N (Fig. 7). This is consistent with observed chlorophyll-*a* increases in experimental incubations of Singapore Strait water upon N addition (Gin et al., 2006), and the fact that diatoms are consistently a large fraction of the phytoplankton community (Chénard et al., 2019; Gin et al., 2006).

The fact that the diel change in pH does not follow the same seasonal pattern as dissolved O<sub>2</sub>, but is on average highest during the SW monsoon (Fig. 8), reflects the seasonal change in the



mean state of the carbonate system: the low salinity during the SW monsoon means that total alkalinity is lower than during other seasons, while peatland DOC remineralisation leads to a relative excess of DIC (Zhou et al., 2021). Consequently, the buffering capacity is lowest during the SW monsoon, and so the diel change in pH for a given change in dissolved CO<sub>2</sub> concentration due to metabolic processes is necessarily larger (Pacella et al., 2018). In contrast, the low diel pH variation during the NE monsoon results from both the higher buffering capacity and the lower net community metabolism at this time. Marine ecosystems in this part of the Sunda Shelf might therefore be more vulnerable to future ocean acidification, because the seawater buffering capacity will be depleted both by uptake of atmospheric CO<sub>2</sub> across the South China Sea and further by terrigenous DOC remineralisation on the shelf (Zhou et al., 2021). We therefore expect that the diel variation in the carbonate system will increase further, especially during the SW monsoon. Such diel carbonate system variation due to community respiration can lead to periods of net CaCO<sub>3</sub> dissolution on reefs, especially from the reef sediment (Stoltenberg et al., 2021). Coral reefs in the Singapore Strait already have relatively low net CaCO<sub>3</sub> accretion rates and low vertical accretion potential even without considering possible sedimentary dissolution (Januchowski-Hartley et al., 2020). Future ocean acidification of the incoming open-ocean water is therefore an important potential threat to these reefs.

## 5. Conclusions

Our results show that terrestrial input is a major source of dissolved inorganic nutrients to the central Sunda Shelf, with coastal waters characterised by DIN:PO<sub>4</sub><sup>3-</sup> ratios close to or slightly below the Redfield ratio, and an excess of dissolved Si over DIN. The source of nutrients is

likely a combination of natural input from soils together with fertiliser and wastewater input, but atmospheric deposition appears to be at most minor. Rivers additionally provide a major input of peatland-derived dissolved organic matter to coastal waters, although this input is notably poor in DOP. The Singapore Strait experiences strong seasonal variation because the semi-annual current reversal advects either terrestrially influenced water masses or water from the open South China Sea through the Strait. The particulate organic matter pool shows far less seasonal variation, suggesting a greater control from local production within coastal waters and possibly resuspension from the sediment. While there is likely a significant detrital contribution to the particulate organic matter pool, phytoplankton biomass is higher than would be expected for genuinely oligotrophic tropical waters. Phytoplankton productivity is probably limited more by light than by nutrients, but phytoplankton biomass does appear to increase in response to seasonal nutrient inputs, at least in some years. Net community metabolism causes stronger diel variation in pH during the SW monsoon than in other seasons, when terrigenous DOC remineralisation depletes the buffering capacity. This metabolic feedback, in combination with the impact of terrigenous DOC remineralisation, may render marine ecosystems in this region of Southeast Asia more vulnerable to the future effects of ocean acidification.

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Data availability

All data and analysis codes are archived in the Nanyang Technological University data repository and can be accessed at <https://doi.org/10.21979/N9/2FQEGW>.

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## Supplementary Material

P. Martin *et al.* Monsoon-driven biogeochemical dynamics in an equatorial shelf sea: time-series observations in the Singapore Strait

Table S1. Relationships between biogeochemical parameters and CDOM  $a_{350}$ . Slope and intercept estimates are given together with their respective standard errors; non-significant intercepts are marked “NS”. P-values refer to the regression slopes.

Parameter	Slope estimate (SE)	Intercept estimate (SE)	Statistics
$\text{NO}_3^-$	1.79 (0.13)	-0.05 (0.10), NS	$r^2=0.612$ , $p<0.0001$
$\text{PO}_4^{3-}$	0.118 (0.008)	0.048 (0.007)	$r^2=0.611$ , $p<0.0001$
$\text{Si(OH)}_4$	4.20 (0.33)	2.98 (0.26)	$r^2=0.563$ , $p<0.0001$
$\text{NO}_2^-$	0.234 (0.044)	0.010 (0.035)	$r^2=0.186$ , $p<0.0001$
DOC	14.3 (1.18)	68.6 (0.95)	$r^2=0.540$ , $p<0.0001$
DOP	-0.097 (0.015)	0.160 (0.013)	$r^2=0.311$ , $p<0.0001$
$\delta^{13}\text{C-POC}$	-1.24 (0.31)	-20.2 (0.23)	$r^2=0.143$ , $p<0.0001$
$\delta^{15}\text{N-PN}$	-0.907 (0.241)	5.01 (0.19)	$r^2=0.116$ , $p<0.001$
$\delta^{15}\text{N-NO}_x^-$	2.59 (0.96)	4.42 (0.75)	$r^2=0.376$ , $p<0.05$

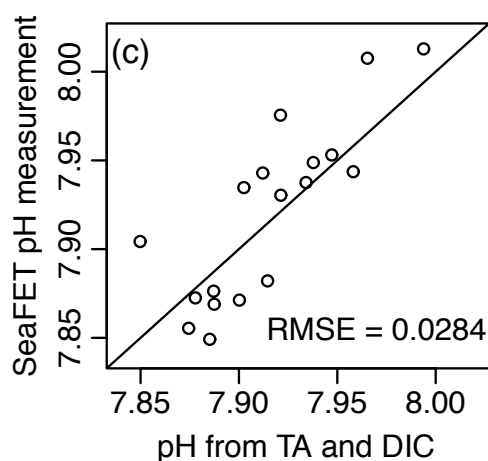
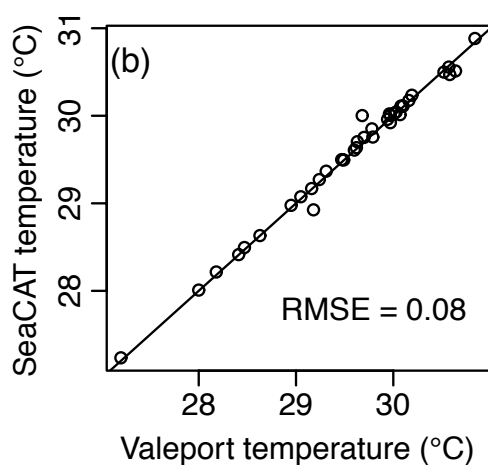
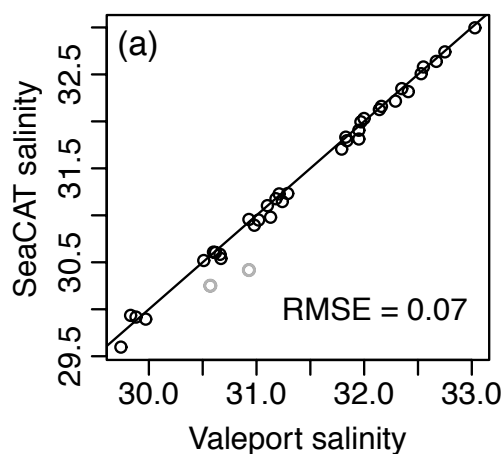


Fig. S1. Comparison of (a) salinity and (b) temperature measured by Seabird SeaCAT moored CTD at Kusu and by Valeport FastCTD profiler at Kusu. The RMSE in (a) excludes the two outliers indicated in grey. (c) Comparison of seawater pH measured by SeaFET pH sensor at Kusu and pH calculated from laboratory measurements of total alkalinity and dissolved inorganic carbon concentration. Solid lines show 1:1 relationships.

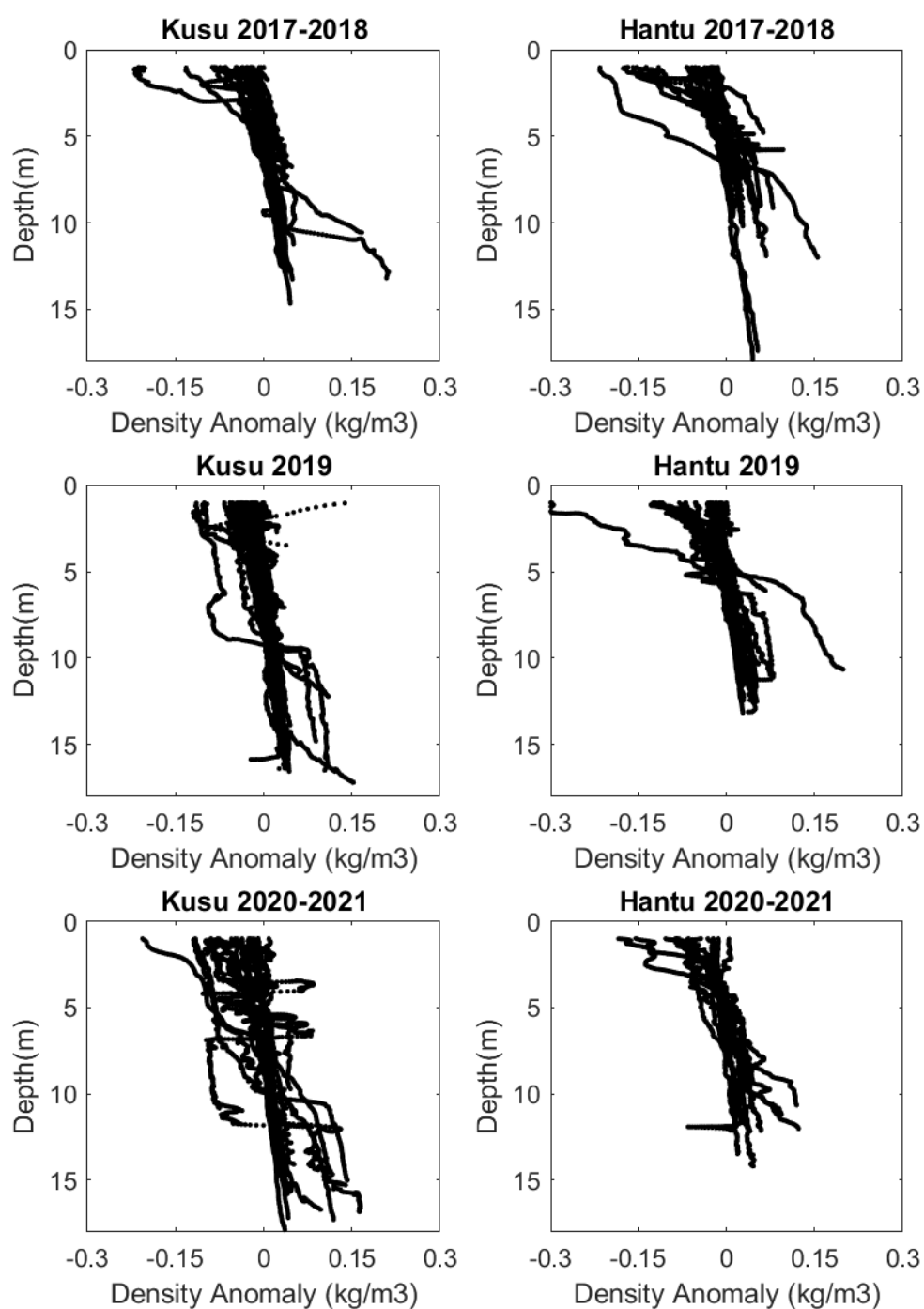


Fig. S2. Vertical profiles of potential density anomaly calculated from Valeport FastCTD profile data (calculated by subtracting the mean potential density of each profile). Note that there is typically no clear pycnocline, indicating little consistent water column stratification.

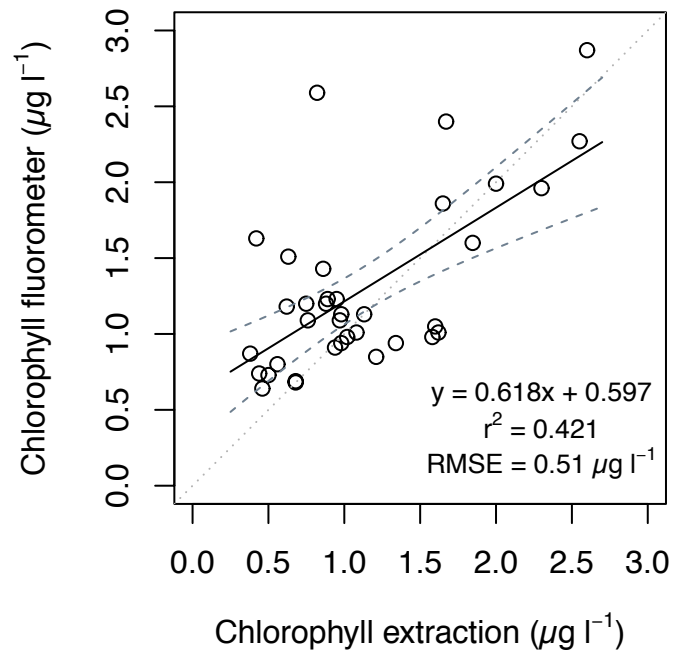


Fig. S3. Comparison of chlorophyll-*a* as measured *in-vivo* by the fluorometer on the Valeport FastCTD *versus* measurements from filtered water samples that were extracted in acetone and measured in the laboratory. Solid black and dashed dark grey lines indicate linear regression and 95% confidence interval. Dotted grey line in the 1:1 relationship.

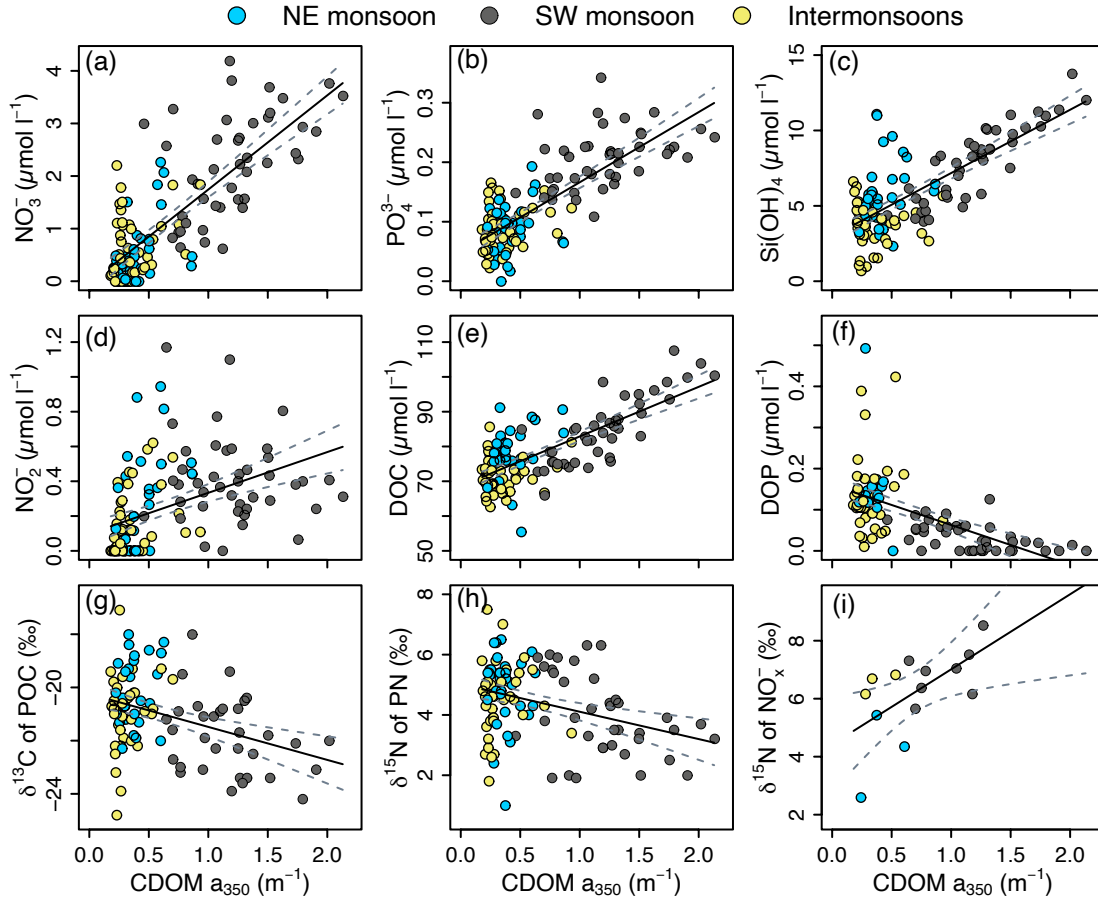


Fig. S4. Relationships between parameters that showed significant relationships to salinity and coloured dissolved organic matter (measured as absorption coefficient at 350 nm;  $a_{350}$ ). Solid black and dashed dark grey lines indicate linear regressions and 95% confidence intervals.