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**Carbon Fluxes in the Coastal  
 Ocean: Synthesis, Boundary  
 Processes and Future Trends**

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### Keywords

carbon cycle, coastal ocean, air-sea CO<sub>2</sub> flux, continental margin, river-dominated ocean margin, RiOMar, ocean-dominated margin, OceMar, carbon neutrality

### Abstract

This review examines the current understanding of the global coastal ocean carbon cycle and provides a new quantitative synthesis of air-sea CO<sub>2</sub> exchange. This reanalysis yields an estimate for the globally integrated coastal ocean CO<sub>2</sub> flux of  $-0.25 \pm 0.05$  Pg C year<sup>-1</sup>, with polar and subpolar regions accounting for most of the CO<sub>2</sub> removal (>90%). A framework that classifies river-dominated ocean margin (RiOMar) and ocean-dominated margin (OceMar) systems is used to conceptualize

coastal carbon cycle processes. The carbon dynamics in three contrasting case study regions, the Baltic Sea, the Mid-Atlantic Bight, and the South China Sea, are compared in terms of the spatio-temporal variability of surface  $p\text{CO}_2$ . Ocean carbon models that range from box models to three-dimensional coupled circulation-biogeochemical models are reviewed in terms of the ability to simulate key processes and project future changes in different continental shelf regions. Common unresolved challenges remain for implementation of these models across RiOMar and OceMar systems. The long-term trends in coastal ocean carbon fluxes for different coastal systems under anthropogenic stress that are emerging in observations and numerical simulations are highlighted. Knowledge gaps in projecting future perturbations associated with before and after net-zero  $\text{CO}_2$  emissions in the context of concurrent changes in the land-ocean-atmosphere coupled system pose a key challenge.

- A new synthesis yields an estimate for globally integrated coastal ocean carbon sink of  $-0.25 \text{ Pg C year}^{-1}$ , with greater than 90% of atmospheric  $\text{CO}_2$  removal occurring in polar and subpolar regions.
- The sustained coastal and open ocean carbon sink is vital in mitigating climate change and meeting the target set by the Paris Agreement.
- Uncertainties in the future coastal ocean carbon cycle are associated with concurrent trends and changes in the land-ocean-atmosphere coupled system.
- The major gaps and challenges identified for current coastal ocean carbon research have important implications for climate and sustainability policies.

## 1. INTRODUCTION

Ocean sequestration of atmospheric  $\text{CO}_2$  is increasingly recognized as an important climate mitigation pathway to realize the target of a  $1.5\text{--}2.0^\circ\text{C}$  temperature increase set by the Paris Agreement (Aricò et al. 2021, Pörtner et al. 2019) (see Sidebar titled Key Terms). The sustained ocean carbon sink is vital in meeting this target because it accounts for an overall uptake of  $\sim 37\%$  of the fossil fuel  $\text{CO}_2$  emissions, or  $\sim 25\%$  of the combined fossil fuel burning and emissions due to land use changes between 1850 and 2019 (Friedlingstein et al. 2020). Hoegh-Guldberg et al. (2019) further projected that ocean-based mitigation options could reduce the emissions gap by up to  $\sim 21\%$  for a  $1.5^\circ\text{C}$  pathway and by  $\sim 25\%$  for a  $2.0^\circ\text{C}$  pathway, making the ocean a primary carbon sink.

Though relatively small in surface area, the coastal ocean (defined in this review as ocean margin or continental margin as detailed in **Supplemental Material Section 1**), with its rich spatial, economic, and biological resources and diverse ecosystems, provides important ecosystem services in support of human society. The absorption of anthropogenic  $\text{CO}_2$  is one of the most important ecosystem services provided by the coastal ocean and its associated nearshore ecosystems (e.g., Cooley et al. 2009).

The coastal ocean is characterized by elevated primary production and is the site for long-term burial of organic matter and calcium carbonate ( $\text{CaCO}_3$ ), both important contributors to the global carbon budget (Gattuso et al. 1998, Mackenzie et al. 2005). The coastal ocean provides a highly dynamic land-ocean interface where biogeochemical processes modulate and transform carbon inputs from both land and the open ocean, thereby controlling subsequent air-sea  $\text{CO}_2$  exchanges and exports of carbon to the open ocean (Bauer et al. 2013, Cai 2011, Chen & Borges 2009, Hofmann et al. 2011, Lacroix et al. 2021a). This role of the coastal ocean makes its inclusion as a component of the global carbon cycle critical for development of carbon and climate policies. The complexity of processes in the coastal ocean, however, presents significant challenges for

## KEY TERMS

### Acronyms, Abbreviations, and Symbols

**CO<sub>2</sub>**: carbon dioxide  
**DIC**: dissolved inorganic carbon  
**TA**: total alkalinity  
**pCO<sub>2</sub>**: partial pressure of CO<sub>2</sub>  
**HCO<sub>3</sub><sup>-</sup>**: bicarbonate  
**CaCO<sub>3</sub>**: calcium carbonate  
**NO<sub>3</sub>**: nitrate  
**NO<sub>2</sub>**: nitrite  
**PO<sub>4</sub>**: phosphate  
**SST**: sea surface temperature  
**MARCATS**: MARGins and CATchment Segmentation  
**SD**: Standard Deviation  
**NEP**: net ecosystem production  
**NEC**: net ecosystem calcification  
**CDR**: carbon dioxide removal  
**ESMs**: Earth system models  
**GOBMs**: global ocean biogeochemical models  
**OceMar**: ocean-dominated margin  
**RiOMar**: river-dominated ocean margin  
**EBC**: eastern boundary current  
**WBC**: western boundary current  
**SCS**: South China Sea  
**WNP**: western North Pacific  
**BS**: Baltic Sea  
**MAB**: Mid-Atlantic Bight  
**ECS**: East China Sea  
**SEATS**: Southeastern Asia Time-series Study  
**RCP**: representative concentration pathway  
**SOCAT**: Surface Ocean CO<sub>2</sub> Atlas

### Variables

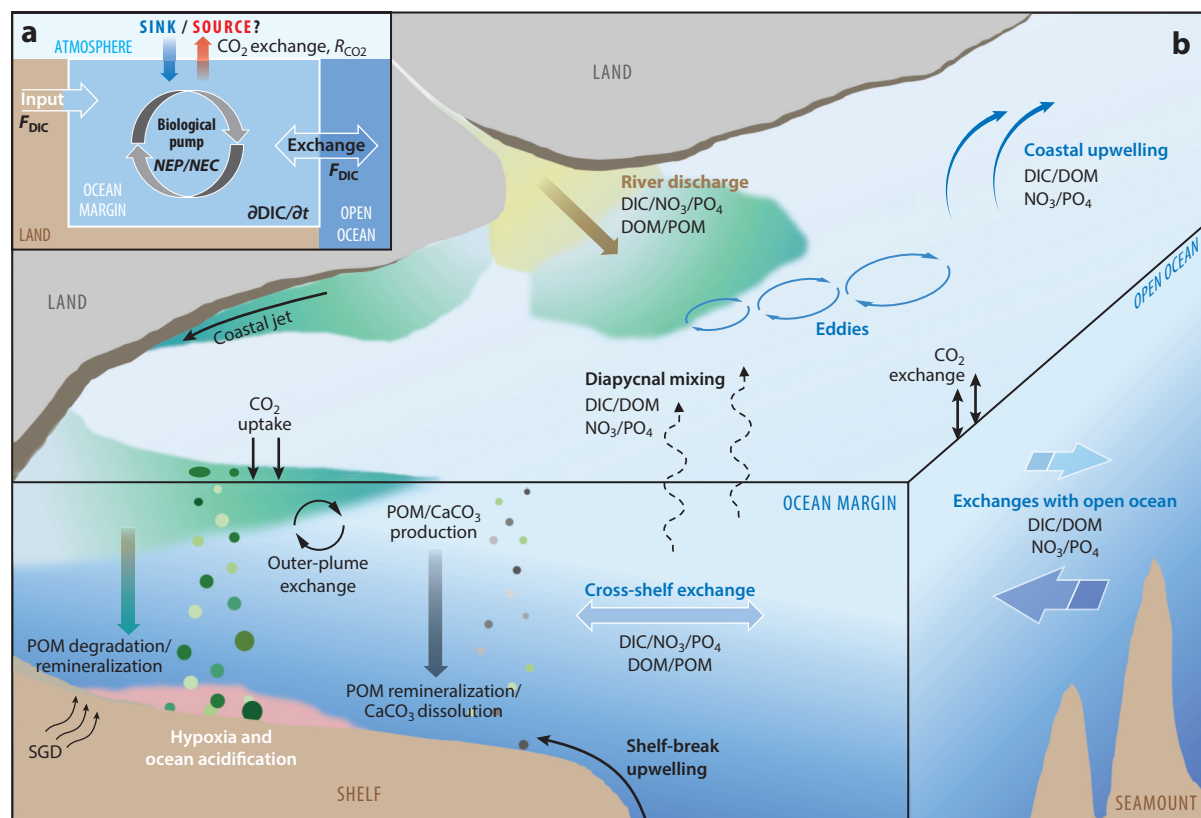
**R<sub>CO<sub>2</sub></sub>**: the air-sea CO<sub>2</sub> flux  
**F<sub>DIC</sub>**: the input or output flux of DIC across the boundary  
**∂DIC/∂t**: the change in the amount of DIC over time  
**NO<sub>3</sub> + NO<sub>2</sub>**: the sum of concentrations of nitrate and nitrite  
**PO<sub>4</sub>**: the phosphate concentration  
**∂DIC\***: changes in the DIC concentration solely induced by air-sea CO<sub>2</sub> exchange  
**ΔpCO<sub>2</sub>**: the difference in the partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>) between seawater and the atmosphere

its inclusion in Earth system models, one of the essential tools for developing climate and sustainability policies.

The removal (uptake) or release of CO<sub>2</sub> from or to the atmosphere is referred as a sink or source of atmospheric CO<sub>2</sub>, respectively (**Figure 1a**). By convention, the ocean's uptake of CO<sub>2</sub> from the

**Sink of atmospheric CO<sub>2</sub>**: uptake of CO<sub>2</sub> from the atmosphere by the ocean





**Figure 1**

Conceptual schematics of air-sea  $\text{CO}_2$  exchanges (a) and major physical and biogeochemical processes in the coastal ocean (b), highlighting the transport of matter between land, ocean margin, and open ocean. (a) The sea-air  $\text{CO}_2$  flux ( $R_{\text{CO}_2}$ ) is balanced by the sum of DIC inputs and outputs ( $F_{\text{DIC}}$ ) across the boundaries, the NEP and NEC, and the change in the amount of DIC over time ( $\partial \text{DIC} / \partial t$ ) within the coastal system (Equation 1 in the text). (b) The ocean margin is bordered by the coastline on the land side and by the open ocean on the outer side. Rivers discharge DIC,  $\text{NO}_3$ ,  $\text{PO}_4$ , DOM, and POM onto the continental shelf via a buoyant plume, and SGD also provides important fluxes of carbon and nutrients to coastal waters. The riverine nutrients stimulate enhanced phytoplankton growth and POM production in the mid- and far fields of the plume, which exchanges with oceanic waters at plume fronts and can extend beyond the shelf break when entrained by sub-meso- and mesoscale eddies. The POM that is produced sinks and is degraded or remineralized in bottom waters, leading to severe consequences such as hypoxia and OA. Wind-induced upwelling at the shelf break and in nearshore regions, and upward diapycnal mixing also supply nutrients as well as DIC and DOM to the surface, facilitating primary production (POM/ $\text{CaCO}_3$  production). The POM is remineralized and  $\text{CaCO}_3$  is dissolved when sinking to deep waters. The biogeochemistry of upwelled or upward mixed waters is controlled by the exchange of DIC, nutrients, DOM, and/or POM at the ocean margin-open ocean interface. The sea-air  $\text{CO}_2$  exchange ultimately depends on the relative supply or consumption of DIC and nutrients due to water mass mixing and the intrinsic biological metabolism. Panel b is adapted from Liu et al. (2010). Abbreviations:  $\text{CaCO}_3$ , calcium carbonate; DIC, dissolved inorganic carbon; DOM, dissolved organic matter; NEC, net ecosystem calcification; NEP, net ecosystem production;  $\text{NO}_3$ , nitrate; OA, ocean acidification;  $\text{PO}_4$ , phosphate; POM, particulate organic matter; SGD, submarine ground discharge.

**Source of atmospheric  $\text{CO}_2$ :** transfer of  $\text{CO}_2$  out of the ocean to the atmosphere

atmosphere (sink) is indicated by a negative sign, and release of  $\text{CO}_2$  into the atmosphere (source) is indicated by a positive sign. The role of the coastal ocean as a source or sink of atmospheric  $\text{CO}_2$  in the global carbon budget has received considerable attention (e.g., Borges et al. 2005, Cai & Dai 2004, Cai et al. 2006, Chen & Borges 2009, Dai et al. 2013, Laruelle et al. 2014, Roobaert et al. 2019, Thomas et al. 2004). The current role of the global ocean margin as a carbon sink is

relatively well constrained, ranging from  $-0.19$  to  $-0.45$  Pg C year $^{-1}$  with a flux density between  $-0.5$  and  $-1.0$  mol C m $^{-2}$  year $^{-1}$  (Borges et al. 2005, Cai et al. 2006, Chen et al. 2013, Dai et al. 2013, Laruelle et al. 2014), with more recent estimates pointing toward the lower bound of these ranges (Roobaert et al. 2019). However, these global flux estimates are only beginning to resolve the seasonality and heterogeneity of regional ocean margin systems (Roobaert et al. 2019), many of which are known to alternate between carbon sinks and sources at different spatial and temporal scales (e.g., Cai et al. 2020). The basic spatial patterns of CO $_2$  fluxes provided by regional observational programs show that individual coastal ocean regions have extensive spatial variability, making it difficult to categorize coastal ocean systems and quantify these fluxes (e.g., Cai et al. 2020, Guo et al. 2015, Li et al. 2020b). Temporal variability of the coastal ocean CO $_2$  flux is even less well known, although understanding of its seasonality is emerging (e.g., Guo et al. 2015). Finally, interannual variability or long-term temporal trends of coastal ocean CO $_2$  fluxes remain elusive and controversial (e.g., Laruelle et al. 2018).

A simplified mass balance equation (Cao et al. 2020, Mackenzie & Lerman 2006) provides a framework to examine the processes and mechanisms that control the air-sea CO $_2$  flux ( $R_{\text{CO}_2}$ , **Figure 1a**) as:

$$R_{\text{CO}_2} = \sum F_{\text{DIC}} + \text{NEP} + \text{NEC} + \partial \text{DIC} / \partial t, \quad 1.$$

where  $\sum F_{\text{DIC}}$  is the sum of the dissolved inorganic carbon (DIC) inputs and outputs,  $\text{NEP}$  and  $\text{NEC}$  are the net ecosystem production and calcification, respectively, and  $\partial \text{DIC} / \partial t$  is the change in the amount of DIC over time, which is assumed to be zero at steady state ( $\partial \text{DIC} / \partial t \sim 0$ ). In most coastal regions,  $\text{NEC}$  is relatively small except for some tropical continental shelves with coral reefs and carbonate banks where  $\text{NEC}$  can be greater than  $\text{NEP}$  (Mackenzie et al. 2005). Thus, for many ocean margins,  $R_{\text{CO}_2}$  is primarily determined by the difference between the net DIC transport flux (inputs – outputs) and  $\text{NEP}$ .

Exchanges across the air-sea interface act by opposing or compensating the net change in DIC and force the system toward equilibrium with the atmosphere (Bauer et al. 2013, Cai 2011, Cao et al. 2020, Dai et al. 2013). Land-derived inputs originate from upland soils (Regnier et al. 2013b) and also from adjacent coastal marsh-estuarine systems (Cai 2011). At the ocean margin-open ocean interface, cross-shelf/slope transport provides a primary exchange pathway, quantification of which, however, remains difficult (e.g., Bauer et al. 2013). Conceptualizations that include the myriad processes and mechanisms contributing to the coastal carbon cycle have been developed (**Figure 1b**), but quantification of processes is often based on individual case studies (e.g., Bauer et al. 2013, Cao et al. 2014, Dai et al. 2013).

The coastal ocean carbon cycle is further complicated by anthropogenic perturbations that act across the land-ocean continuum and evolve over time (e.g., Mackenzie et al. 2005, Regnier et al. 2013b). During preindustrial times, the coastal ocean may have acted as a net source of atmospheric CO $_2$  (e.g., Aricò et al. 2021, Bauer et al. 2013, Mackenzie et al. 2004). However, analysis of recent global coastal ocean simulations suggested that the coastal ocean was a small net preindustrial sink of atmospheric CO $_2$  (Bourgeois et al. 2016, Lacroix et al. 2021a). Regnier et al. (2013b) developed an intermediate view of a near-neutral coastal ocean carbon sink prior to significant human perturbations by assuming that the coastal ocean perturbation scaled to that of the open ocean. These authors also proposed that anthropogenic perturbations originate from increased land-to-ocean carbon inputs, rather than only from atmospheric uptake. However, with increasing atmospheric CO $_2$  levels, it is anticipated that the future coastal ocean will take up more anthropogenic CO $_2$  from the atmosphere and export more DIC to the open ocean, at least before carbon emissions reach their peak (Bauer et al. 2013, Bourgeois et al. 2016).



**Net-zero carbon emission:** achieving an overall balance between CO<sub>2</sub> emissions produced and removed from the atmosphere

**Carbon dioxide removal:** anthropogenic activities removing CO<sub>2</sub> from the atmosphere and durably storing it in geological, terrestrial, or ocean reservoirs, or in products

Advances have been made in development and implementation of models to investigate coastal carbon cycling processes. Yet, modeling the coastal ocean at global scales remains difficult because of the paucity of observations for model evaluation and the heterogeneity of coastal systems, both of which affect the ability to adequately represent global-scale coastal processes (Hofmann et al. 2011, Holt et al. 2009). So far, only regional models allow inclusion of region-specific biogeochemical processes representative of the spatial and temporal variability. Reconciling and combining results from global and regional models is an important step toward developing the predictive capability that is needed to inform policies aimed at net-zero carbon emission (i.e., achieving an overall balance between CO<sub>2</sub> emissions and that removed from the atmosphere) by the mid-century, which likely will include the implementation of some types of land- and/or ocean-based carbon dioxide removal (CDR) techniques. The implementation of CDR techniques will potentially lead to further and rapid alterations of the global carbon cycle, including the coastal carbon cycle, that will be superimposed on the changes that have already occurred since the start of the industrial period and are expected to continue in the future. Understanding the response of the coastal ocean carbon to such changes poses a grand challenge, which requires careful observational and modeling assessment at both regional and global scales.

Critical knowledge gaps remain in understanding and quantifying coastal carbon fluxes and the underlying controls. These include internal process interactions, responses to external and boundary forcings, and responses to human-induced local and global perturbations. This review provides an analysis of the current qualitative and quantitative understanding of the coastal ocean carbon cycle, with a focus on the air-sea CO<sub>2</sub> exchange, at regional to global scales. It includes novel findings obtained using the full breadth of methodological approaches, from observation-based studies and advanced statistical methods to conceptual and theoretical frameworks, and numerical modeling. Most importantly, it provides a critical review of the current status of coastal ocean carbon research, pointing to the major gaps and challenges that give rise to opportunities for future research.

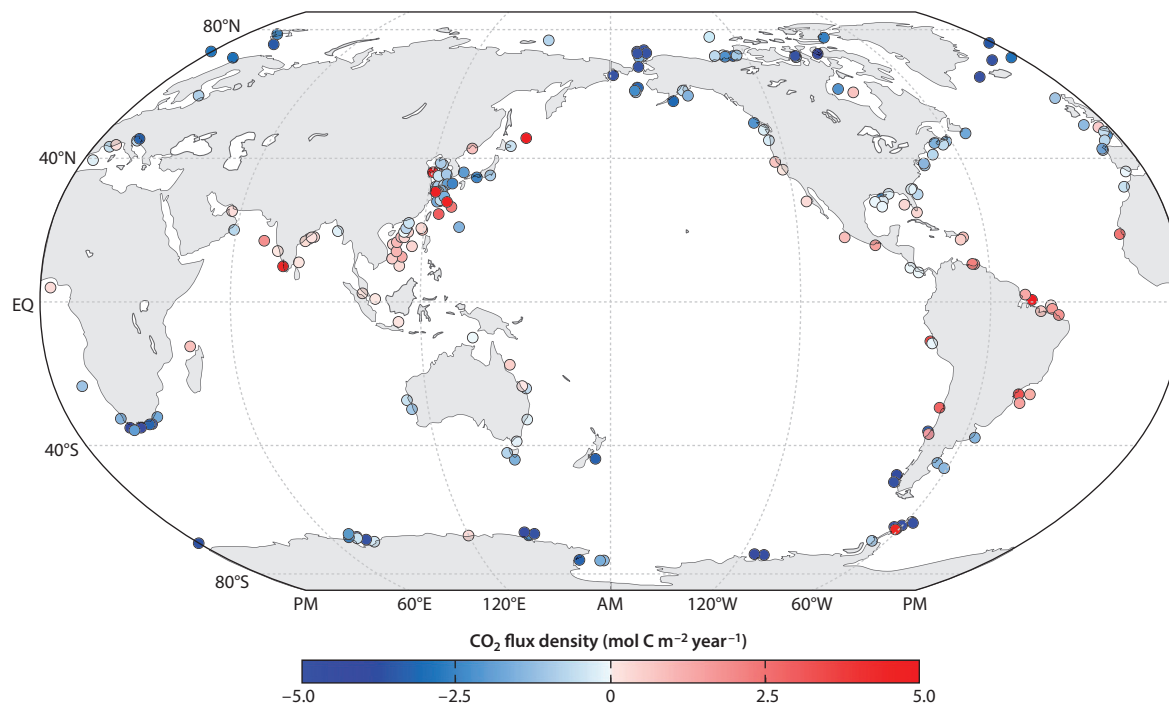
## 2. SYNTHESIS OF REGIONAL AIR-SEA CO<sub>2</sub> FLUXES

Assessment of the global coastal ocean air-sea CO<sub>2</sub> flux has advanced significantly over the past four decades. Tsunogai et al. (1999) extrapolated a case study from the East China Sea (ECS) to derive a carbon sink of  $-1 \text{ Pg C year}^{-1}$  for the global continental margin. A subsequent North Sea case study was also extrapolated to the global scale, resulting in a smaller carbon sink of  $-0.4 \text{ Pg C year}^{-1}$  (Thomas et al. 2004). These studies highlighted the importance of the coastal ocean but showed that individual coastal systems vary considerably (Cai & Dai 2004). This motivated more advanced global synthesis efforts based on compilations of local and regional CO<sub>2</sub> flux measurements and province-based extrapolations (e.g., Borges 2005, Cai et al. 2006, Chen & Borges 2009, Chen et al. 2013, Dai et al. 2013, Laruelle et al. 2010).

### 2.1. New Data Synthesis

A new synthesis of the global coastal ocean CO<sub>2</sub> flux, based on reported region-based integrated CO<sub>2</sub> flux estimates from 1998 until the present, is provided here with detailed methodological explanations given in **Supplemental Material Section 2**. This new synthesis includes 214 regional CO<sub>2</sub> flux estimates in coastal systems (**Figure 2**; **Supplemental Figure 1.1**; **Supplemental Table 1**) with uncertainty and caveats considered (**Supplemental Material Section 2.2**) and is based on regional studies that provided sufficient information to allow recalculation of the gas transfer velocity using the relationships given in Wanninkhof et al. (2013) or Wanninkhof (2014). The calculated CO<sub>2</sub> flux estimates were allocated into 46 MARgins and CATchment Segmentation





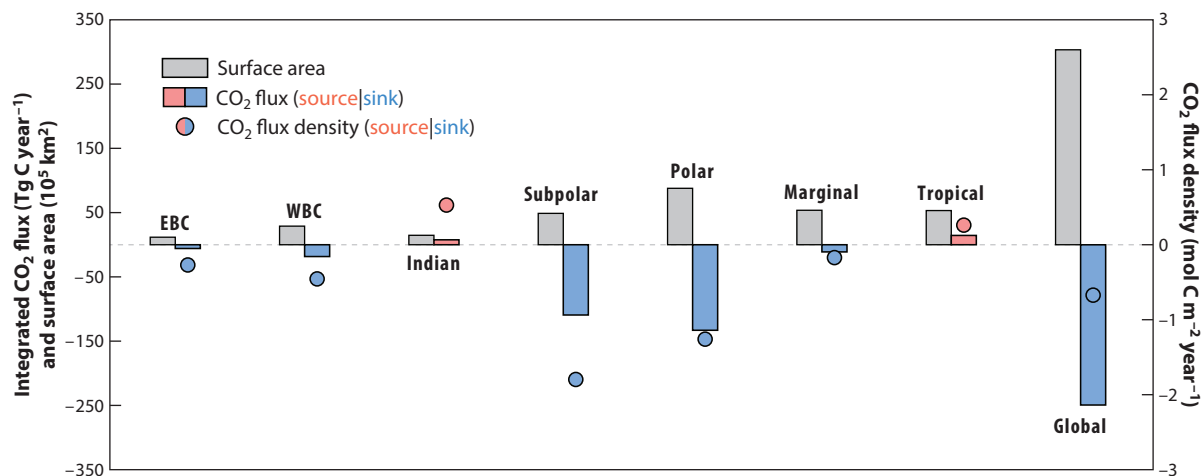
**Figure 2**

Updated sea-air CO<sub>2</sub> flux density (mol C m<sup>-2</sup> year<sup>-1</sup>) in the global coastal oceans. Individual sites and associated calculations are detailed in **Supplemental Table 1**.

units, a widely accepted segmentation of the global coastal ocean, the geographical boundaries of which were obtained from Laruelle et al. (2013) with slight modifications, and further categorized into seven major shelf classes (**Supplemental Figure 1.1**) based on the characteristics given in the **Supplemental Material Section 1**.

The new synthesis of air-sea CO<sub>2</sub> fluxes shows an integrated air-sea CO<sub>2</sub> flux (a sink) in the global coastal ocean of  $-0.25 \pm 0.05$  Pg C year<sup>-1</sup>, which is within the range of  $-0.45$  to  $-0.19$  Pg C year<sup>-1</sup> reported in previous studies (Borges et al. 2005, Chen & Borges 2009, Chen et al. 2013, Laruelle et al. 2014, Roobaert et al. 2019). The polar ( $-134$  Tg C year<sup>-1</sup>) and subpolar ( $-108$  Tg C year<sup>-1</sup>) regions contribute the majority to the global sink (**Figure 3**), followed by the western boundary current (WBC) systems ( $-17$  Tg C year<sup>-1</sup>), marginal seas ( $-11$  Tg C year<sup>-1</sup>), and eastern boundary current (EBC) systems ( $-4$  Tg C year<sup>-1</sup>). The tropical ( $17$  Tg C year<sup>-1</sup>) and Indian Ocean ( $9$  Tg C year<sup>-1</sup>) margins are weak sources (**Figure 3**).

The bulk of the CO<sub>2</sub> uptake translates into a globally averaged CO<sub>2</sub> flux density of  $-0.68 \pm 0.14$  mol C m<sup>-2</sup> year<sup>-1</sup>, which suggests that the coastal ocean is a more efficient CO<sub>2</sub> sink relative to the value of  $-0.5$  mol C m<sup>-2</sup> year<sup>-1</sup> estimated for the open ocean (Wanninkhof et al. 2013). This flux density represents the lower bound of previously reported ranges [ $-0.71$  mol C m<sup>-2</sup> year<sup>-1</sup> (Cai et al. 2006);  $-1.09$  mol C m<sup>-2</sup> year<sup>-1</sup> (Chen et al. 2013);  $-0.73$  mol C m<sup>-2</sup> year<sup>-1</sup> (Laruelle et al. 2014)], but is only slightly higher than the value of  $-0.58$  mol C m<sup>-2</sup> year<sup>-1</sup> reported in the recent assessment by Roobaert et al. (2019), considering the associated uncertainty in both estimates and distinct methodological approaches. Here, we assess fluxes from a literature review of regional studies while Roobaert et al. (2019) applied a neural-network interpolation technique



**Figure 3**

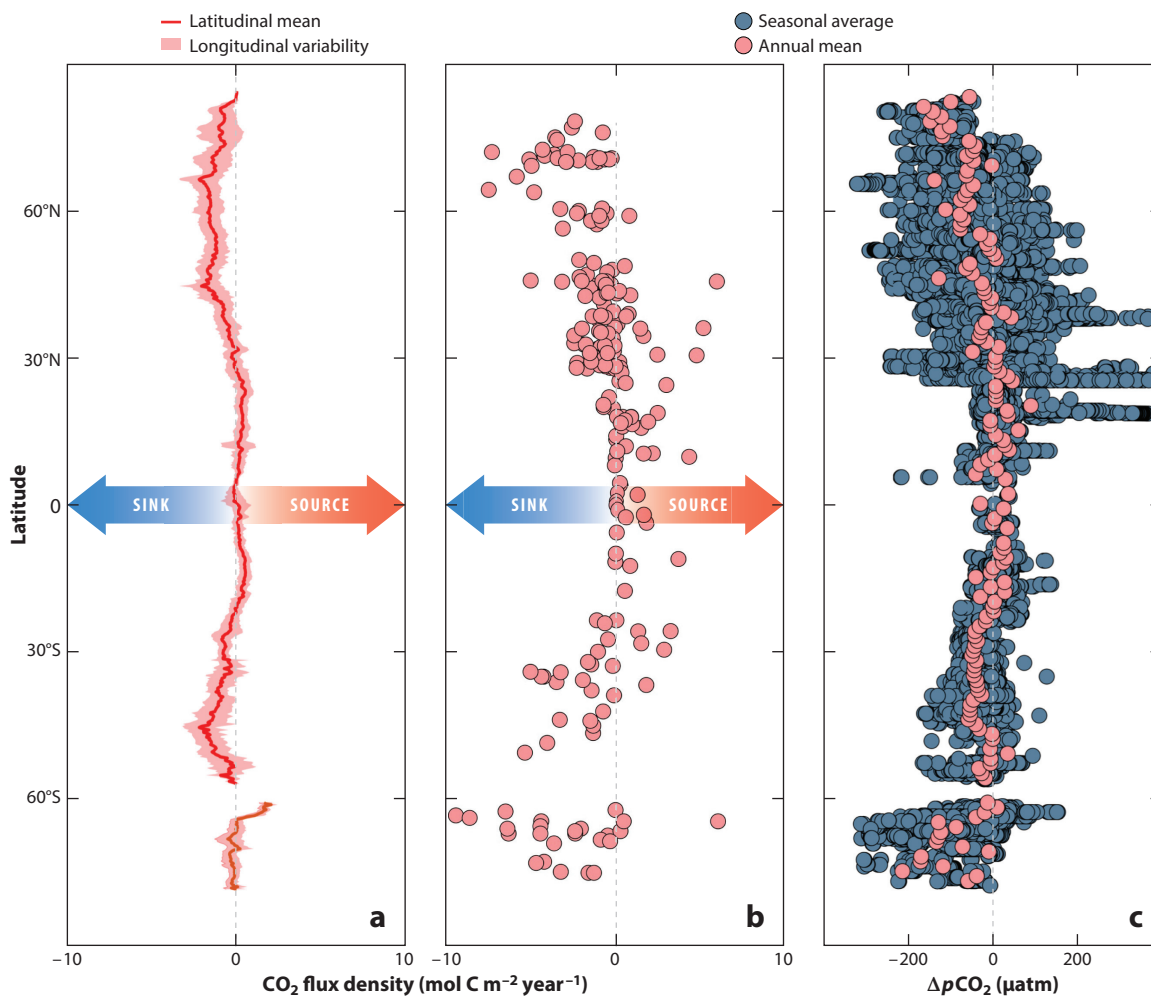
The shelf surface area (gray bars), CO<sub>2</sub> flux density (colored circles), and spatially integrated CO<sub>2</sub> flux (red and blue bars) estimated for the seven shelf classes and global shelves. Abbreviations: EBC, eastern boundary current; Indian, Indian Ocean margins; marginal, marginal sea; WBC, western boundary current.

to the CO<sub>2</sub> data gathered in the Surface Ocean CO<sub>2</sub> Atlas (SOCAT) version 4 product (Bakker et al. 2016). Both assessments show a consistent pattern in terms of defining sinks and sources of the seven shelf classes. The larger CO<sub>2</sub> flux obtained from the present synthesis relative to the results of Roobaert et al. (2019) can be attributed to the higher CO<sub>2</sub> flux densities obtained for polar ( $-1.27$  versus  $-0.75$  mol C m<sup>-2</sup> year<sup>-1</sup>) and subpolar ( $-1.80$  versus  $-1.23$  mol C m<sup>-2</sup> year<sup>-1</sup>) regions and the larger shelf surface areas of  $88 \times 10^5$  versus  $81 \times 10^5$  km<sup>2</sup> in polar and  $50 \times 10^5$  versus  $47 \times 10^5$  km<sup>2</sup> in subpolar regions (surface area estimates are detailed in the **Supplemental Material Section 2.1** and **Supplemental Table 1**). The total differences ( $-100$  Tg C year<sup>-1</sup>) of the CO<sub>2</sub> sinks in polar ( $-134$  versus  $-74$  Tg C year<sup>-1</sup>) and subpolar ( $-108$  versus  $-69$  Tg C year<sup>-1</sup>) regions account for 40% and 51% of the global coastal CO<sub>2</sub> flux derived in this study ( $-0.25$  Pg C year<sup>-1</sup>) and that of Roobaert et al. (2019) ( $-0.20$  Pg C year<sup>-1</sup>), respectively.

## 2.2. Seasonality and Latitudinal Pattern

The latitudinal pattern in the coastal CO<sub>2</sub> flux described by Laruelle et al. (2014) also showed the seasonality of the flux. In this study, the highest CO<sub>2</sub> uptakes were found at high latitudes, followed by moderate uptake at mid-latitudes (Laruelle et al. 2014). At low latitudes, seasonally alternating uptake and outgassing of CO<sub>2</sub> resulted in low net annual air-sea exchanges. This study recognized the large contribution of Arctic and sub-Arctic shelves in the global coastal ocean carbon budget. Seasonal dynamics of the CO<sub>2</sub> fluxes are evident in the mid- and high latitudes of the Atlantic Ocean, showing an atmospheric CO<sub>2</sub> sink in spring, a strong source in summer, and a gradual decrease over fall and into winter (Signorini et al. 2013). This seasonality is mostly driven by changes in sea surface temperature (SST) and NEP. Roobaert et al. (2019) similarly showed CO<sub>2</sub> flux amplitudes that were large at high latitudes, moderate at mid-latitudes, and low at low latitudes (**Figure 4a**).

The large and consistent uptake of atmospheric CO<sub>2</sub> in polar and subpolar regions was also found in this synthesis (**Figure 3**). The largest variability occurs in the mid-latitudes. At low latitudes, the CO<sub>2</sub> fluxes vary from near neutral to positive (sources of atmospheric CO<sub>2</sub>). A striking



**Figure 4**

Latitudinal distributions of (a) annual  $\text{CO}_2$  flux density reported by Roobaert et al. (2019), (b) annual  $\text{CO}_2$  flux density obtained from compilation of data presented in the literature, and (c) seasonal and annual averages  $\Delta p\text{CO}_2$  (difference of  $p\text{CO}_2$  between seawater and the atmosphere) at a distance of 50 km from the major land masses. Abbreviation:  $p\text{CO}_2$ , partial pressure of  $\text{CO}_2$ .

contrast is that the literature-based estimates have a more pronounced dynamic range than that shown in **Figure 4a**.

The latitudinal distribution of SST, sea surface partial pressure of  $\text{CO}_2$  ( $p\text{CO}_2$ ), and  $\Delta p\text{CO}_2$  (defined as the difference in  $p\text{CO}_2$  between the sea and the air) based on SOCAT product (version 2021) (**Figure 4c**; **Supplemental Figure 2.3**) at three different distances—50, 100, and 200 km (**Supplemental Figures 2.4–2.6**)—away from the global shoreline provide further characterization of the global pattern, similar to the analysis in Cao et al. (2020). The  $\Delta p\text{CO}_2$ , indicative of the air-sea  $\text{CO}_2$  fluxes, is similar to the global pattern based on our literature synthesis (**Figures 4a,b**), but with a larger dynamic range. This larger dynamic range is even more evident for the seasonal data (blue dots in **Figure 4c**), relative to the annual mean, suggesting strong seasonal heterogeneity in  $\text{CO}_2$  fluxes at the same latitudes and along latitudes.

**River-dominated ocean margin:**

the coastal system of which CO<sub>2</sub> fluxes are dominated by riverine inputs of carbon and nutrients at the land-ocean margin interface

**Ocean-dominated margin:**

the coastal system of which CO<sub>2</sub> fluxes are dominated by oceanic inputs of carbon and nutrients at the ocean margin-open ocean interface

The  $p\text{CO}_2$  and  $\Delta p\text{CO}_2$  have a less defined latitudinal pattern during each season than that of SST (**Supplemental Figures 2.4–2.6**). The  $p\text{CO}_2$  is low at mid- and high latitudes and slightly higher in tropical zones. The spatial variance of  $p\text{CO}_2$  at 50 km is large at latitudes that are influenced by upwelling and terrestrial input, and shows considerable seasonal variability. Also, summer  $p\text{CO}_2$  drawdown is pronounced at high latitudes due to biological CO<sub>2</sub> consumption. In autumn, the latitudinal pattern of  $p\text{CO}_2$  is not as clear. Seasonal changes in  $p\text{CO}_2$  are smaller for data collected at greater distance offshore (i.e., at 200 km), with the majority of values around 400  $\mu\text{atm}$  (**Supplemental Figures 2.4–2.6**). The 200-km spatial and temporal distribution patterns are rather similar to those expected for the open ocean, and the  $\Delta p\text{CO}_2$  latitudinal pattern is similar to that of sea surface  $p\text{CO}_2$ .

The preceding description suggests that both thermal and nonthermal drivers control the latitudinal  $p\text{CO}_2$  and the associated fluxes, as previously reported (Cao et al. 2020). The effect of temperature increases toward the equator, where increasing SST elevates the surface seawater  $p\text{CO}_2$ . Nonthermal factors, such as water mass mixing with open ocean waters (e.g., upwelling), and local biological alterations (e.g., higher biological productivity at mid- and high latitudes, and lower productivity but higher riverine organic matter input at low latitudes) are superimposed on the thermal control pattern (Cao et al. 2020, Chen et al. 2013).

The relative importance of thermal and nonthermal factors on coastal CO<sub>2</sub> dynamics and fluxes was further assessed for interseasonal  $p\text{CO}_2$  changes using the approach given in Cao et al. (2020). With this approach, the observed interseasonal  $p\text{CO}_2$  change is decomposed into a temperature-controlled change and a non-temperature-controlled residual. The main controls of interseasonal  $p\text{CO}_2$  changes in different latitudinal bands during seasonal transitions differ. However, the interseasonal changes of global coastal ocean  $p\text{CO}_2$  are generally more determined by nonthermal factors rather than temperature, which is supported by a statistically significant positive relationship between the total change and nonthermal factors (**Supplemental Figure 2.7**).

### 3. OCEAN-DOMINATED MARGINS VERSUS RIVER-DOMINATED OCEAN MARGINS

The previously developed framework that categorizes river-dominated ocean margin (RiOMar) and ocean-dominated margin (OceMar) systems allows conceptualization of coastal ocean carbon cycle processes (Cao et al. 2020, Dai et al. 2013). Both systems are characterized by interactive exchanges and transport at the boundaries along the river-estuary-shelf-ocean continuum and intrinsic biogeochemical reactions that determine the air-sea CO<sub>2</sub> flux. However, the distinction recognizes differences in controls on CO<sub>2</sub> fluxes and carbon cycling in coastal systems that are dominated by riverine inputs at the land-ocean margin interface versus those that are dominated by oceanic inputs at the ocean margin-open ocean interface (Cao et al. 2020, Dai et al. 2013).

A theoretical framework was developed to resolve the CO<sub>2</sub> source/sink nature of OceMar systems (**Supplemental Material Section 3.1**), such as systems affected by boundary currents and coastal upwelling, and RiOMar systems, such as river plume-shelf systems receiving large terrestrial loads. In OceMar systems, DIC and nutrients, supplied by the open ocean and distributed by the three-dimensional circulation, are transported to the ocean margin and subsequently upwelled or mixed into the euphotic zone, where the nutrients support enhanced primary production. Changes in DIC relative to nutrients, both transformed by physical transport and biological processes, determine whether there is an excess or deficit in DIC. Excess DIC in seawater is eventually released to the atmosphere, making the ocean margin a source of atmospheric CO<sub>2</sub>, whereas a DIC deficit is compensated by input of atmospheric CO<sub>2</sub> making the



region a CO<sub>2</sub> sink (Cao et al. 2020, Dai et al. 2013). In RiOMar systems, the DIC and nutrients originating from land or rivers are delivered to nearshore waters via estuarine/bay circulation and subsequently exported to the continental shelf via wind-driven circulation of the low saline river plume (Gan et al. 2009). The balance between the externally sourced DIC and nutrients and internal biogeochemical interactions thus controls the CO<sub>2</sub> fluxes (Cao et al. 2020).

The RiOMar and OceMar designations are spatially and temporally variable and can be interchangeable. Thus, a RiOMar can shift to an OceMar depending on the relative inputs of water and biogeochemical compounds across the land-ocean margin and ocean margin-open ocean interfaces and the intensity of the physical forcing that regulates these inputs. A decrease in river discharge during dry seasons constrains the spatial scale of river plumes, potentially converting a RiOMar to an OceMar, particularly if coastal upwelling or vertical mixing is strengthened by the interaction between wind-driven shelf currents and topography.

#### Diatom-diazotroph assemblages:

a widespread marine planktonic symbiosis between diatoms and N<sub>2</sub>-fixing bacteria or prokaryotes

### 3.1. River-Dominated Ocean Margin Systems: Characteristics and Diagnosis

Buoyant surface plumes that extend from large rivers and estuaries into the coastal ocean are characteristic of RiOMar systems. Estimates of the plume area associated with the world's 19 largest rivers showed a total annual mean plume area of  $3.7 \times 10^6$  km<sup>2</sup> (Kang et al. 2013), accounting for ~14% of the total continental shelf area worldwide. Extrapolation of the CO<sub>2</sub> flux density in river plumes of the Amazon, Changjiang, Mississippi, and Loire Rivers indicates a global CO<sub>2</sub> sink for large river plumes of about  $-0.066$  Pg C year<sup>-1</sup>, contributing to ~25% of the global continental shelf CO<sub>2</sub> sink (Cai et al. 2014). The hydrodynamics of individual plumes are complex and heterogeneous. The Amazon River plume flows northwestward and can be partly carried eastward by retroflexion of the North Brazil Current to feed into the North Equatorial Countercurrent (Ibanez et al. 2016). The Mississippi-Atchafalaya River plume propagates offshore toward the remote southern Gulf of Mexico (Schiller & Kourafalou 2014). The Mekong River plume discharged into the South China Sea (SCS) can also veer into the deep SCS basin via coastal jet separation (Gan & Qu 2008).

Mixing of seawater with riverine freshwater dominates *p*CO<sub>2</sub> dynamics in river plumes by altering the carbonate chemistry. High bicarbonate (HCO<sub>3</sub><sup>-</sup>) concentrations in subtropical to mid-latitude river waters (e.g., the Changjiang, Mississippi, and Pearl Rivers) exhibit a stronger buffering capacity to changes in *p*CO<sub>2</sub> than that of tropical river waters (e.g., the Amazon and Orinoco Rivers) with lower HCO<sub>3</sub><sup>-</sup> concentrations (Cai et al. 2014). The *p*CO<sub>2</sub> in HCO<sub>3</sub><sup>-</sup>-rich river waters therefore decreases gradually along the plume, while in HCO<sub>3</sub><sup>-</sup>-poor river waters it decreases rapidly in the near field of the plume and approaches the seawater *p*CO<sub>2</sub> at the plume front.

River plume *p*CO<sub>2</sub> dynamics are further affected by biological processes that depend on the trade-off between degradation of terrestrial organic matter and riverine nutrient-enhanced primary production. Along a river plume, the sea surface often shifts from a source of atmospheric CO<sub>2</sub> in the near field, due to organic matter degradation and excess CO<sub>2</sub> inherited from the river, to a sink as a result of enhanced biological removal of CO<sub>2</sub> (Huang et al. 2015). The biologically mediated low *p*CO<sub>2</sub> surface water is transported away from estuaries because the air-sea *p*CO<sub>2</sub> equilibrium time is often much longer than the residence time of the plume or surface waters (Zhao et al. 2021). In the high-salinity far field, the sea surface tends to be in a near-neutral state due to weakened phytoplankton production resulting from depleted nutrients, and temperature becomes the dominant control on the surface *p*CO<sub>2</sub> and air-sea CO<sub>2</sub> flux. However, for some systems, such as the Amazon and Mekong River plumes, nitrogen fixation by diatom-diazotroph assemblages can supply nutrients that sustain the biological pump in the outer plume.



In addition to the analysis for the Pearl River plume (Cao et al. 2020), the carbon dynamics of the RiOMar systems represented by the Mekong River plume and the Mississippi-Atchafalaya River plume were assessed using the theoretical diagnostic framework (**Supplemental Material Section 3.1; Supplemental Figure 3.1a,b**). The Mekong River discharge has relatively high nutrient concentrations, greater than  $40 \mu\text{mol L}^{-1}$  of nitrate + nitrite ( $\text{NO}_3 + \text{NO}_2$ ) and  $\sim 1 \mu\text{mol L}^{-1}$  of phosphate ( $\text{PO}_4$ ), that support new production in the SCS (Grosse et al. 2010). Similarly to the Amazon River plume, the Mekong River plume is influenced by  $\text{N}_2$  fixation potentially by diatom-diazotroph assemblages with rates up to  $0.55 \mu\text{mol N L}^{-1} \text{ day}^{-1}$ , which accounts for 1–47% of the primary production nitrogen demand (Grosse et al. 2010).

The diagnostic approach estimate for DIC consumption fueled by riverine nutrients in the Mekong River plume is  $7.9 \pm 2.4 \mu\text{mol kg}^{-1}$  ( $\text{NO}_3 + \text{NO}_2$  based) or  $2.7 \pm 0.6 \mu\text{mol kg}^{-1}$  ( $\text{PO}_4$  based), resulting in high  $\delta\text{DIC}^*$  (changes in the DIC concentration solely induced by air-sea  $\text{CO}_2$  exchange as explained in **Supplemental Material Section 3**) values of  $31 \pm 7 \mu\text{mol kg}^{-1}$  based on  $\text{NO}_3 + \text{NO}_2$  and  $37 \pm 10 \mu\text{mol kg}^{-1}$  based on  $\text{PO}_4$  in the surface plume (salinity  $< 33.5$ ) in late summer of 2007 (**Supplemental Figure 3.2**). The DIC consumption fueled by  $\text{N}_2$  fixation can offset at most 47% of the  $\delta\text{DIC}^*$ . The Mekong River plume is therefore a source of atmospheric  $\text{CO}_2$ , having already released  $\text{CO}_2$  to the atmosphere over the timescale considered by the framework with sea surface  $p\text{CO}_2$  decreasing from  $\sim 500 \mu\text{atm}$  before air-sea exchange to  $\sim 379 \pm 15 \mu\text{atm}$  at measurements, almost equilibrating with the atmospheric  $p\text{CO}_2$  of  $\sim 381 \mu\text{atm}$ .

The Louisiana shelf in the northern Gulf of Mexico annually receives  $\sim 0.7\text{--}1.0 \text{ Tg N}$  of nitrate (Lehrter et al. 2013) and  $\sim 17 \text{ Tg C}$  of  $\text{HCO}_3^-$  from the Mississippi-Atchafalaya River system (Lohrenz et al. 2013). Although highly supersaturated  $p\text{CO}_2$  values were observed in narrow regions near river mouths, the inner shelf influenced by the riverine inputs remained largely undersaturated with respect to the atmospheric  $\text{CO}_2$  (Huang et al. 2015). The diagnostic result for the Mississippi-Atchafalaya River plumes (salinity  $< 35$ ) in early summer of 2006 (**Supplemental Figure 3.3**) shows that primary production consumes DIC by  $232 \pm 110 \mu\text{mol kg}^{-1}$  based on  $\text{NO}_3 + \text{NO}_2$ , lowering the seawater  $p\text{CO}_2$  to less than  $100 \mu\text{atm}$ . The spatially averaged  $\delta\text{DIC}^*$  is  $-67 \pm 56 \mu\text{mol kg}^{-1}$ , indicating a strong deficit of DIC, which can explain its behavior as a sink of  $\text{CO}_2$  over the inner shelf with a  $\Delta p\text{CO}_2$  of  $-125 \pm 75 \mu\text{atm}$ .

### 3.2. Ocean-Dominated Margin Systems: Characteristics and Diagnosis

For OcoMar systems, the material exchange across the ocean margin-open ocean interface is poorly known and is dependent on the diverse geomorphology and ecosystems that characterize these systems. Quantifying this exchange at regional or global scales is challenging because of the lack of clearly defined boundaries, insufficient understanding of associated processes such as boundary current intrusions, cross-slope dynamics, and exchanges induced by meso- and sub-mesoscale oceanic processes (Dai 2021, and references therein).

**3.2.1. Eastern boundary current systems.** The EBC systems and other coastal upwelling systems are dependent on inputs of oceanic-derived nutrients to sustain their high biological productivity (Wollast 1991). The California Current System, an example of an EBC upwelling system (**Supplemental Material Section 1; Supplemental Figure 3.4a**), transports DIC- and nutrient-enriched subsurface waters, derived from offshore deep oceanic waters, across the shelf to the nearshore surface waters (Cai et al. 2020). The upwelling brings water from 150–200 m to the surface, resulting in  $p\text{CO}_2$  levels that can reach  $1,000 \mu\text{atm}$ . This water is then transported seaward and southward while the  $p\text{CO}_2$  is drawn down by biological productivity to  $\sim 200 \mu\text{atm}$ , far below the atmospheric  $p\text{CO}_2$  value (Feely et al. 2008). Such a pronounced decrease in seawater

$p\text{CO}_2$  occurs because complete utilization of preformed nutrients in upwelled waters exceeds the corresponding net DIC consumption, which has been diagnosed semiquantitatively (Cao et al. 2014). Controls of the  $\text{CO}_2$  flux in other EBC systems in which the majority of DIC and nutrients in the upper layer originate from locally wind-induced cross-shelf transport of the nonlocal deep waters in the subtropical gyre of the open ocean are likely similar. Depending on the DIC/nutrient ratios in source waters,  $\text{CO}_2$  flux density can vary, but the DIC deficit relative to that of nutrients makes the system a sink of atmospheric  $\text{CO}_2$  (Section 2).

**3.2.2. Western boundary current systems.** The WBC systems are typically found along coastal margins with wide shelves. The outer and mid-shelf dynamics are dominated by inputs from the open ocean. The ECS (**Supplemental Figure 3.4e**), a OcéMar system, is characterized by intrusions of warm, high-salinity, and high-nutrient waters onto the outer shelf that are derived from the Kuroshio, a WBC in the western North Pacific (WNP) (Guo et al. 2015, Liu & Gan 2012). Zhang et al. (2019) estimated that the Kuroshio contributes 72% and 84% of external inputs of dissolved inorganic nitrogen (DIN) and phosphorus (DIP), respectively, to the entire ECS shelf, supporting 50% of DIN-based primary production and 61% of DIP-based primary production. Budget analysis showed that the total influx of DIC ( $75 \text{ Tmol C year}^{-1}$ ) from the intrusion of Kuroshio waters into the ECS exceeded the DIC consumption fueled by the Kuroshio-sourced DIN ( $0.15 \text{ Tmol N year}^{-1}$ ) and DIP ( $0.011 \text{ Tmol P year}^{-1}$ ) (Chen & Wang 1999). However, the biological consumption of open ocean-sourced DIC produces a deficit relative to nutrient use, causing the outer ECS to behave as a sink for the atmospheric  $\text{CO}_2$  (Guo et al. 2015).

**3.2.3. Marginal seas.** Semi-enclosed marginal seas are characterized by complex transport pathways and lateral and vertical fluxes/exchanges with water masses of relatively longer residence times that allow for biogeochemical transformations. Using a simple one-dimensional advection-diffusion model, Dai et al. (2013) showed that the DIC and nutrient inputs from upwelling of the deep Pacific water provided via the SCS overflow result in excess DIC. The DIC excess relative to open ocean-sourced nutrients, as diagnosed previously (Cao et al. 2020, Dai et al. 2013), is eventually released to the atmosphere. Chou et al. (2017) adopted this approach to explain the occurrence of atmospheric  $\text{CO}_2$  source areas within the Japan/East Sea in summer that are supported by  $\text{CO}_2$  sources from the adjacent open ocean.

The Caribbean Sea is the largest marginal sea of the Atlantic Ocean (**Supplemental Figure 3.4b**) and experiences overflow from the Atlantic similar to that from the WNP into the SCS, but with a more complex circulation pattern (MacCready et al. 1999). Mid-depth water, which is sourced from Upper North Atlantic Deep Water, flows from the Atlantic into the Virgin Islands basin, and then over the Jungfern-Grappler complex into the eastern Caribbean (MacCready et al. 1999). This sole source of water for the deep Caribbean is presumably balanced by diffusively driven upwelling elsewhere in the basin (Ribbat et al. 1976). Upward water displacement exists in the deep layer (Gallegos 1996), induced by renewal flows across the deepest passages, that supplies both DIC and nutrients to the surface mixed layer, where the relative  $\text{CO}_2$  consumption/release determines whether this OcéMar system acts as a  $\text{CO}_2$  source or sink (Dai et al. 2013). Samples collected in April 2021 showed  $\delta\text{DIC}^*$  values of  $1.4 \pm 2.5 \mu\text{mol kg}^{-1}$  and  $\Delta p\text{CO}_2$  of  $3 \pm 5 \mu\text{atm}$  (**Supplemental Figures 3.1c, 3.5**), further demonstrating that the Caribbean Sea is nearly in equilibrium with the atmospheric  $\text{CO}_2$ .

**3.2.4. Indian Ocean margins.** The Arabian Sea on the western Indian Ocean margin is an OcéMar system with intrusion of deep waters from the Indian Ocean onto the continental margin via horizontal advection, which are then transported upward via vertical mixing and upwelling (Valsala & Murtugudde 2015) (**Supplemental Figure 3.4f**). During the southwest monsoon



season when wind-driven coastal upwelling frequently occurs, high primary production is often observed (Brand & Griffiths 2009). Cao et al. (2020) estimated positive  $\text{PO}_4$ -based  $\delta\text{DIC}^*$  values during all seasons ( $\sim 23 \mu\text{mol kg}^{-1}$  in winter, summer, and autumn and  $\sim 4 \mu\text{mol kg}^{-1}$  in spring), suggesting that the Arabian Sea acts as a weak source of  $\text{CO}_2$  to the atmosphere.

### 3.3. Arctic Margins

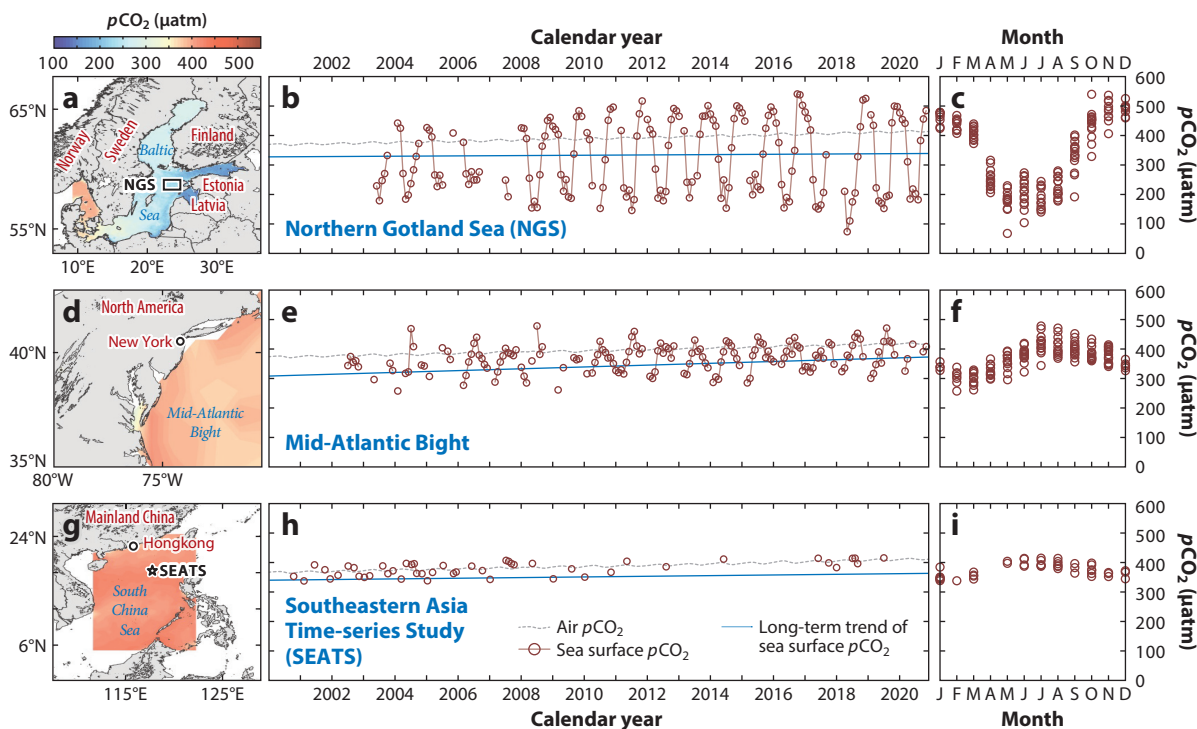
Arctic continental shelves are influenced by both river inputs and exchanges with the Pacific and Atlantic Oceans (Roeske et al. 2012, Terhaar et al. 2021). The large influx of freshwater from riverine inputs makes the Arctic Ocean well stratified with a distinct surface layer of reduced salinity (Abrahamsen et al. 2009). The Arctic Ocean also receives inflows from the Atlantic and Pacific Oceans via the Greenland-Scotland Ridge (Hansen et al. 2008) and Fram Strait and the Barents Seas Opening, respectively (e.g., Beszczynska-Möller et al. 2012, Ingvaldsen et al. 2002, Schauer et al. 2004).

The Bering Sea (**Supplemental Figure 3.4c**) provides an example system for analyzing possible controls on the air-sea exchanges featuring both riverine input and exchanges with the open ocean (Aiello & Ravelo 2012). As such, the Bering Sea is intermediate between OcéMar and RiOMar systems, with surface  $p\text{CO}_2$  and  $\text{CO}_2$  fluxes characterized by large spatial-temporal variability and diverse controlling processes. In spring and summer, the Bering Sea shelf is undersaturated with respect to atmospheric  $p\text{CO}_2$  mainly because of biological production (Bates et al. 2011, Chen et al. 2004). However, Sun et al. (2020) observed that in summer the central Bering Strait is supersaturated with respect to the atmospheric  $p\text{CO}_2$  as a result of upwelling. In contrast, the Bering Sea basin and eastern nearshore regions are a neutral or weak  $\text{CO}_2$  sink, as a result of the high nutrients/low chlorophyll status and riverine input, respectively. Chen (1985) suggested that in winter the Bering Sea shelf was a potential sink for atmospheric  $\text{CO}_2$  due to cooling, while Kelley & Hood (1971) and Cross et al. (2014) found supersaturation of  $p\text{CO}_2$  in winter in the Bering Sea shelf area.

The Alaskan Stream flows into the Bering Sea and transports relatively warm water in the surface layer. This water mixes with the dichothermal water from  $\sim 150$  m that forms during the winter and warm subsurface Alaskan Stream (depth of  $\sim 300$  m) via eddy-induced upwelling and along tilted-up isopycnals (Miura et al. 2002, Mizobata et al. 2002). Using data collected in October 2007 in the eastern Bering Sea basin, the diagnostic approach estimated that biological production consumed  $2.0 \pm 0.6 \mu\text{mol kg}^{-1} \text{NO}_3 + \text{NO}_2$ , equivalent to a DIC consumption of  $13.1 \pm 4.0 \mu\text{mol kg}^{-1}$  (**Supplemental Figures 3.1d, 3.6**). The resultant  $\delta\text{DIC}^*$  is  $1.5 \pm 1.8 \mu\text{mol kg}^{-1}$ , indicating a neutral or weak  $\text{CO}_2$  sink for the Bering Sea basin in autumn. However, the dominant controls on the  $\text{CO}_2$  source/sink nature for the entire Bering Sea (including shelf and basin regions) in all seasons warrant further examination.

## 4. THREE REGIONAL CASE STUDIES ON SPATIAL AND SEASONAL VARIABILITY AND LONG-TERM TRENDS OF SEA SURFACE $p\text{CO}_2$ : THE BALTIC SEA, THE MID-ATLANTIC BIGHT, AND THE SOUTH CHINA SEA

The Baltic Sea (BS), the SCS, and the Mid-Atlantic Bight (MAB) (**Supplemental Figure 4.1**) are regions with long-term sea surface  $p\text{CO}_2$  measurements at high spatio-temporal resolution that allow estimates of air-sea  $\text{CO}_2$  fluxes and characterization of biogeochemical transformations (**Figure 5**). In addition, coupled circulation-biogeochemical models have been implemented to investigate carbon cycling dynamics in these three regions (for additional information, see **Supplemental Material Section 4**).



**Figure 5**

Spatial, long-term, and seasonal variability of sea surface  $p\text{CO}_2$  in (a–c) the BS, (d–f) the MAB, and (g–i) the SCS. The spatial distribution of sea surface  $p\text{CO}_2$  (a,d,g) represents Northern Hemisphere summer conditions and is based on remote sensing–supported  $p\text{CO}_2$  estimates from the climatological monthly average of August (2002–2011) with a horizontal resolution of  $0.0417^\circ \times 0.0417^\circ$  for the BS (Zhang et al. 2021), a spatial interpolation of the gridded summer data ( $1^\circ \times 1^\circ$ , SOCAT version 2021) for the MAB, and reconstructed data ( $0.5^\circ \times 0.5^\circ$ ) for the SCS with relatively high reliability (Supplemental Figure 2.2) (Wang et al. 2021). The air  $p\text{CO}_2$  data (b,e,h) were obtained from the Mauna Loa Observatory (Keeling et al. 2005). The monthly averaged sea surface  $p\text{CO}_2$  data for the BS (b,e) were calculated from cruise averages of each crossing of the NGS as shown in Schneider & Müller (2018). The MAB data (e,f) are based on monthly gridded data from SOCAT version 2021 ( $1^\circ \times 1^\circ$ ) as shown in Xu et al. (2020). The SCS data (h,i) are based on time-series measurements at the SEATS station ( $116^\circ\text{E}$ ,  $18^\circ\text{N}$ ) in the northern basin of the SCS according to Sheu et al. (2010) and Dai (unpublished data). The long-term trends of sea surface  $p\text{CO}_2$  are based on a linear regression fit to the seasonally detrended monthly mean values (b,e,h). Abbreviations: BS, Baltic Sea; MAB, Mid-Atlantic Bight; NGS, Northern Gotland Sea;  $p\text{CO}_2$ , partial pressure of  $\text{CO}_2$ ; SCS, South China Sea; SEATS, Southeastern Asia Time-series Study; SOCAT, Surface Ocean  $\text{CO}_2$  Atlas.

The BS, a semi-enclosed marginal sea, can be considered one of the world’s largest estuaries subjected to perturbation by anthropogenic activities, notably by eutrophication. Among the three case study sites, the BS has the largest spatial variability [standard deviation (SD)  $\sim 55.1 \mu\text{atm}$  in summer (Figure 5a)] and the strongest seasonality of sea surface  $p\text{CO}_2$  [(mean amplitude defined as the difference between the highest and the lowest monthly mean  $p\text{CO}_2$  across all years  $\sim 319.3 \mu\text{atm}$  (Figure 5c)]. Primary production and mineralization processes have been identified as the main drivers for the seasonality of sea surface  $p\text{CO}_2$  (Schneider & Müller 2018). The MAB, located on the North American East Coast, is characterized by a broad, shallow shelf with moderate riverine inputs discharged through large estuaries. The MAB shows intermediate spatial [SD  $\sim 32.3 \mu\text{atm}$  in summer (Figure 5d)] and seasonal [mean amplitude  $\sim 116.6 \mu\text{atm}$  (Figure 5f)] variability in sea surface  $p\text{CO}_2$ . The seasonal cycle of  $p\text{CO}_2$  in this system is primarily driven by the SST (DeGrandpre et al. 2002, Signorini et al. 2013, Xu et al. 2020). Local biological

production partially counteracts the thermally induced  $p\text{CO}_2$  increase during spring and summer, but the overall seasonality is not significantly altered because stratification inhibits the nutrient supply to the surface water. Finally, the SCS, one of the largest marginal seas, is influenced by the Asian monsoons and spans a wide range of biogeographic regimes with characteristic oligotrophy in its basin area. Here, sea surface  $p\text{CO}_2$  has the lowest spatial [SD  $\sim 13.0 \mu\text{atm}$  in summer (**Figure 5g**)] and seasonal heterogeneity [mean amplitude  $\sim 64.3 \mu\text{atm}$  (**Figure 5i**)] relative to that observed for the BS and MAB. The heterogeneous nature of  $p\text{CO}_2$  and uneven observations in the SCS affect the uncertainty of gridded  $p\text{CO}_2$  used in the flux estimate (**Supplemental Figure 2.1**). Controls on the  $p\text{CO}_2$  seasonality in the SCS differ among its subregions, but overall it is modulated by SST and mixed layer depth that govern the seasonal nutrient supply from depth (Li et al. 2020b, Yang et al. 2021).

The long-term trends of sea surface  $p\text{CO}_2$  differ substantially between the three regions. Synthesis of all available data collected at the Southeastern Asia Time-series Study site (**Figure 5g**) in the SCS basin shows a long-term increasing trend of  $2.5 \pm 1.0 \mu\text{atm year}^{-1}$  ( $R^2 = 0.33$ ), based on a linear regression, that is synchronous with the atmospheric  $p\text{CO}_2$  level since 2000 (**Figure 5b**). However, when seasonal variability is removed, this long-term trend is reduced to  $1.4 \pm 0.5 \mu\text{atm year}^{-1}$  ( $R^2 = 0.45$ ), which is less than the rate of atmospheric  $p\text{CO}_2$  increase. In the MAB, the long-term increase in sea surface  $p\text{CO}_2$  is  $2.4 \pm 0.6 \mu\text{atm year}^{-1}$  after seasonal detrending, which is comparable to or slightly more than that of  $\sim 1.8 \mu\text{atm year}^{-1}$  for atmospheric  $\text{CO}_2$  (**Figure 5e**). In the BS, the seasonally detrended  $p\text{CO}_2$  data show a smaller rate of increase of  $0.06 \mu\text{atm year}^{-1}$  during the period 2003–2020, but associated with a large uncertainty of  $\pm 1.02 \mu\text{atm year}^{-1}$  (**Figure 5b**). In contrast to the large uncertainty associated with  $p\text{CO}_2$  trend analysis, the BS  $\text{CO}_2$  system shows a long-term and steady increase in alkalinity that emerged in the mid-1990s (Müller et al. 2016). The amplitude of seasonal variability in sea surface  $p\text{CO}_2$  (**Figure 5c,f**) for the BS and MAB is larger than the secular trend, making it difficult to determine a meaningful rate of change on decadal timescales and thus reliably predict the temporal evolution of the respective carbon sinks (for additional information, see **Supplemental Material Section 4**).

## 5. MODELING COASTAL OCEAN CARBON

Numerical models are capable of examining complex processes in a holistic way and provide an approach for reconstructing historical changes in coastal carbon fluxes, as well as projecting changes into the future (e.g., Bourgeois et al. 2016, Lacroix et al. 2021a, Legge et al. 2020, Siedlecki et al. 2021). Furthermore, numerical models are applied to quantify critical processes that are difficult to constrain through in situ measurements, such as NEP (Gan et al. 2010, Herrmann et al. 2015, Lacroix et al. 2021a) and the cross-shelf and/or cross-interface transfer of carbon (Druon et al. 2010, Yuan et al. 2018). In addition to considering the impacts of climate change and atmospheric  $\text{CO}_2$  increase on the coastal carbon cycle, current biogeochemical models also provide assessments of other environmental threats posed by human activities that contribute to eutrophication, acidification, and hypoxia (Fennel & Testa 2019, Laruelle et al. 2017, Li et al. 2020a, Takeshita et al. 2015, Yu et al. 2021) that have direct and indirect consequences for coastal carbon cycling.

Coastal ocean carbon models range from conceptual box (budget) models to three-dimensional coupled circulation-biogeochemical models, some of which include data assimilative capability (e.g., Xiao & Friedrichs 2014). Modeling studies have been successfully conducted in different coastal regions and encompass a range of space and timescales (**Supplemental Table 2**). A common requirement of these models is the need to identify and constrain the processes that control carbon cycling and the flow of organic and inorganic carbon into and out of the coastal system.

Thus, quantification of coastal carbon cycling requires accurate representation of exchanges and fluxes across interfaces with the open ocean, land, atmosphere, and sediment. Equally important are representations of the internal transport and transformations of carbon via circulation and biogeochemical processes (e.g., NEP, heterotrophic respiration, NEC). Current approaches in representing these fluxes and processes, imposed limitations, and the implications for carbon cycling at global and regional scales are highlighted by reviewing representative global and regional models.

### 5.1. Budgets and Box Models

Carbon budgets constructed for continental margins are based on aggregate approaches that average processes, fluxes, and exchanges in space and time. These mass balance approaches have provided useful insights regarding the magnitude of carbon fluxes and inventories at global and regional scales, with a specific focus on the trophic status of the global coastal ocean and the air-sea CO<sub>2</sub> exchange (e.g., Gattuso et al. 1998, Mackenzie et al. 2005, Smith & Hollibaugh 1993, Wollast 1993). Advances in global coastal carbon budgets are facilitated by observations that constrain key fluxes such as the air-sea CO<sub>2</sub> exchange, the inclusion of overlooked fluxes attributed to coastal vegetated ecosystems (Bauer et al. 2013), and the explicit accounting of the anthropogenic perturbation of land-to-open ocean carbon fluxes (Regnier et al. 2013b). Nevertheless, the outstanding question of whether continental shelves exhibit net autotrophy or heterotrophy remains to be resolved (Bauer et al. 2013).

In parallel with implementation of carbon budget models, conceptual global box models have provided complementary, mechanistic insights into how changes in physical and biogeochemical processes resulting from the combined effects of changing climate, atmospheric composition, and land use may have affected past global carbon fluxes and may indicate impacts on projected future fluxes (Mackenzie et al. 1998, 2000, 2002, 2004). These models are useful in isolating the effects of different stressors on carbon fluxes in the global coastal ocean and allow for a wide range of sensitivity analyses, but considerations of the spatio-temporal heterogeneity arising from complex circulation patterns or widely varying biological activity are not allowed by the model structure.

### 5.2. Global and Regional Ocean Biogeochemical Models

Coupled three-dimensional physical-biogeochemical models, at either global or regional scales, extend the understanding of coastal carbon processes and fluxes over spatial and temporal scales that go beyond the constraints of box model approaches and limited measurements.

**5.2.1. Global ocean biogeochemical models.** Global ocean biogeochemical models (GOBMs) (Dunne et al. 2020, Tjiputra et al. 2020), such as those used in the United Nations Intergovernmental Panel on Climate Change (Ciais et al. 2013) or the Global Carbon Project (Friedlingstein et al. 2020), have resolutions that are typically too coarse to provide a reliable basis for the analysis of coastal ocean carbon fluxes (Holt et al. 2009, Mathis et al. 2017). An equally important limitation is that these models do not include or only provide simplistic representations of processes relevant to the coastal ocean interface, such as time-varying riverine fluxes (Lacroix et al. 2020), their modulation by the estuary-coastal vegetation continuum (Bauer et al. 2013), and benthic carbon processing (Bianchi et al. 2021, Krumins et al. 2013). However, improved model resolution, afforded by advances in computational capabilities, allows GOBMs to represent important features of the coastal ocean, such as three-dimensional material and substance transports, residence times (Lacroix et al. 2021a, Liu & Gan 2017), air-sea CO<sub>2</sub> exchange (Bourgeois et al. 2016), and riverine fluxes (Dunne et al. 2020; Lacroix et al. 2020, 2021a,b). Additionally, the biogeochemical



representation of specific coastal processes (e.g., heterotrophic respiration and burial of organic carbon, benthic calcification) greatly lags behind improvements in model resolution.

Downscaled and nested models combine the advantages of the coarse resolution of large-scale global models with high-resolution regional models. Downscaling allows exchange of information between coastal and oceanic domains, thereby maintaining connectivity between oceanic and coastal systems (Mathis et al. 2017, Mayer et al. 2018). The higher-resolution regional model allows detailed representations of processes that are unresolved (e.g., mesoscale and sub-mesoscale circulation) or are system specific (e.g., lower trophic-level structure) in the global model. Two-way nested high-resolution regional models are also capable of providing feedbacks to the global model of coastal information to better predict global carbon cycle. An added benefit is that global models typically provide longer simulations due to their coarse resolution without the need of boundary conditions. Inherent difficulties associated with resolving the fine-scale features of the coastal ocean (e.g., estuarine plumes, coastal fronts) within state-of-the-art global models make regional models essential in assessment of coastal carbon cycling and fluxes and related environmental consequences, such as hypoxia (Fennel & Testa 2019).

**5.2.2. Regional ocean biogeochemical models.** Regional coastal carbon models represent a diversity of approaches that reflect system understanding and characteristics, and data availability. Regional models have been applied to a wide range of systems (**Supplemental Table 2**) to investigate controls on carbon cycling (e.g., Druon et al. 2010), cross-shelf export (e.g., Fennel & Wilkin 2009, Frischknecht et al. 2018), seasonal and spatial variability in carbon chemistry (e.g., Fennel et al. 2008, Gomez et al. 2020, Turi et al. 2014), and historical trends (e.g., Mathis et al. 2019). Regional models also provide the ability to examine subregional variability that is not easily accessible with global models. Although implemented for a diversity of systems, these models yield common metrics, such as export flux estimates (**Supplemental Table 2**), that have the potential to perform across-system comparisons of the processes controlling coastal carbon cycling.

Estuaries, which function as a filter and/or conduit across the land-ocean interface, have also been modeled at the regional scale using coupled physical-biogeochemical models (Regnier et al. 2013a). To date, only a few modeling studies have partly addressed the catchment-river-estuary-shelf continuum in a single framework (St-Laurent et al. 2020). The inclusion of such linkages across the land-estuarine-ocean continuum is critical for assessing past and future trends in carbon cycling.

Regional carbon cycling models developed for the SCS, the MAB region, and the BS provide a comparison of three contrasting systems and discussions of limitations and implications for model results (**Supplemental Material Section 4**). Simulation of  $p\text{CO}_2$  fluxes in these coastal oceans has proven to be difficult and depends on regionally based physical and biological dynamics. In the BS, the lack of simulation of additional nitrogen source (e.g.,  $\text{N}_2$  fixation or vertical mixing) inhibited reproducing the regional  $\text{CO}_2$  flux (Gustafsson et al. 2015) during the period between spring blooms and the accumulation of cyanobacteria in July when nitrogen is depleted (Fransner et al. 2018, Schneider et al. 2014). In the SCS, simulation of the  $p\text{CO}_2$  dynamics has proven difficult because of the spatio-temporal patterns of salinity and surface total alkalinity (TA) that partly drive the carbonate system but are not fully represented by the model dynamics. These difficulties stem from the poor representation of the dilution or concentration effects driven by precipitation and evaporation, the accuracy of current atmospheric models, and/or the fact that remote sensing data are insufficient to adequately portray these processes. For the MAB, the simulated air-sea  $\text{CO}_2$  flux was controlled by the cross-shelf transport of organic matter (Fennel & Wilkin 2009), the effective TA flux from shelf denitrification (Fennel et al. 2006), and near-surface wind speed (Previdi et al.

2009), all with different space and time variability. More detailed challenges in simulating these complex regimes are provided in **Supplemental Material Section 4**.

In spite of these limitations and challenges, regional and nested coupled physical-biogeochemical models have advanced sufficiently in process representation and skill to allow scenario testing for responses of carbon cycling in coastal waters to past conditions of global and local forcing, as well as projected future conditions. Mathis et al. (2019) used a nested model forced by representative concentration pathway (RCP) climate change scenarios to investigate future reductions in open inflows to the North Sea and showed that this may not necessarily lead to lower biological carbon uptake in the region. Xiu et al. (2018) modeled a complex biological carbon uptake pattern in the California Current System in response to future global warming.

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**Representative concentration pathway:** a trajectory of atmospheric greenhouse gas concentration (not emissions) that describes climate futures

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### 5.3. Challenges for Coupled Circulation-Biogeochemical Models

Simulation of coastal ocean carbon dynamics is dependent on realistic representation of the physical circulation that is governed by both geostrophic (e.g., topographically driven flow) and ageostrophic dynamics (e.g., mixing and nonlinearity), input of terrestrial buoyancy, and water mass and momentum exchanges between the coastal and open oceans (Gan et al. 2010). Accurate representation of the coastal circulation dynamics is challenging because of the temporally variable and spatially heterogeneous processes that link the land and the open ocean. The spatio-temporal resolution required to resolve local topographic controls and high-frequency forcing (such as tides) also introduces challenges for numerical approaches. Another considerable challenge to biogeochemical modeling is the quantification of the parameters that control carbon cycling rates, which are limited by understanding of ecosystem dynamics and observational capabilities.

Simulation of the coastal ocean carbon dynamics also requires three-dimensional boundary fluxes and forcing, some of which can be provided by downscaling of fields from large-scale or global models, which are not always reliable. In addition, numerical open boundary conditions for downscaled fields that allow computational stability and retain the continuum between coastal and open oceans remain an important challenge, although the issue is often neglected (Liu & Gan 2017).

Specification of other boundary fluxes and forcing, such as riverine inputs and ocean nutrient inputs, requires different approaches that depend on climatology and observations (e.g., Fennel et al. 2008), reanalysis products, or other models (Fennel & Wilkin 2009). However, these approaches resolve different spatio-temporal scales and potentially different dynamics (i.e., biogeochemical processes) that may not match those resolved in a regional model. Thus, the current approaches used to specify boundary fluxes and forcing impose constraints and limitations on model skill and simulation results. An important challenge for coastal carbon models is thus to resolve these potential mismatches and not introduce additional uncertainties into simulations.

### 5.4. State of Current Understanding

Global models offer a large-scale perspective that is useful in informing global assessments of the carbon cycle (e.g., Friedlingstein et al. 2020). However, model constraints imposed by representation of coastal zone circulation dynamics, as well as specific biogeochemical processes relevant to individual regions, have limited the use of GOBMs in studies of coastal carbon cycling. Nevertheless, models with increased resolution have been used for the analysis of long-term trends in coastal carbon without addressing the shorter-term seasonal or interannual temporal variability (Bourgeois et al. 2016, Lacroix et al. 2021a). Analyses of biogeochemical model



structure, complexity, and uncertainty show that increasing model skill is a trade-off between including additional components, accurate representation of rates and processes, and data to constrain processes and provide verification (Ward et al. 2010), all of which have implications for data requirements and process understanding for model parameterization, calibration, and verification.

A limitation of regional and global biogeochemical models is the lack of representation of benthic formation and dissolution of  $\text{CaCO}_3$ , which is an essential component of the carbon counter pump in the coastal ocean (e.g., Smith & Mackenzie 2016). The global role of benthic carbonate cycling has recently been addressed in a conceptual model framework (O'Mara & Dunne 2019), which provides insights into how these processes can be incorporated into three-dimensional coupled physical-biogeochemical models. Representation of organic matter decomposition in the water column and sediments, which generally relies on a limited number of pools or even a single pool of organic matter of constant stoichiometry and reactivity, is another important limitation of global and regional carbon models (Hülse et al. 2018). Also, understanding of how organic matter degrades in marine sediments has implications for development of regional and global carbon models, as well as for elucidation of the role of sediments in carbon budgets (LaRowe et al. 2020). Promising avenues nevertheless exist, such as the representation of terrestrial dissolved organic matter as an independent carbon pool (Lacroix et al. 2021a), or the representation of organic matter degradability across a reactive continuum (Aumont et al. 2017).

Assessment of model skill via comparisons with observations, which often have limitations, or via model intercomparisons is a critical step in refining model structure, determining model-specific responses of biogeochemical cycles to short- (change in land cover) versus long-term (climate) forcing, and developing scenarios to explore the consequences of natural and anthropogenic changes. An important limitation of current models is that model-based estimates are typically not constrained by uncertainty estimates, although efforts have begun to emerge (Meier et al. 2019). In contrast to the open ocean, large-scale carbon models, identifying common, overarching research questions remains very often elusive for coastal carbon models. Therefore, assessing model uncertainties through ensemble runs that address key metrics of the carbon cycle is so far uncommon (Meier et al. 2018). Thus, developing scenarios and overarching questions focused on fundamental controls of the coastal carbon cycle provides an important challenge to the modeling community.

Projecting the consequences of natural and human-induced changes will place more emphasis on models (Jacox et al. 2020). Moreover, coastal ecosystem goods and services will continue to expand (Martínez et al. 2007) as human development leads to more demands (Pendleton et al. 2012), thus further stressing coastal carbon resources. The ability to predict the impacts of multiple stressors and respond to management requirements for coastal resources will require coastal carbon modeling frameworks that can inform economic, social, and governance structures, which remain to be developed.

## 6. TEMPORAL EVOLUTION OF COASTAL OCEAN CARBON SINK AND ITS POSSIBLE FUTURE TRENDS UNDER POSITIVE AND NEGATIVE EMISSION SCENARIOS

The injection of anthropogenic  $\text{CO}_2$  into the atmosphere and its subsequent distribution across the ocean, land, and atmosphere components of the Earth system have led to an unprecedented perturbation of the global carbon cycle that has been ongoing since the Industrial Revolution (Gruber et al. 2019). The ocean, a sustained sink for anthropogenic  $\text{CO}_2$  released into the atmosphere by fossil fuel burning over the past  $\sim 150$  years (**Supplemental Figure 5.1**), has been altered by this  $\text{CO}_2$  uptake, and the effects of this perturbation are evident in the coastal ocean.



The complexities of the land-ocean-atmosphere coupled system introduce large uncertainties into projections of ocean carbon fluxes, in particular into defining the role of the coastal carbon sink and its evolution, both of which are of fundamental importance to climate science and climate policies developed before and after achievement of net-zero CO<sub>2</sub> emissions (i.e., carbon neutrality).

The Paris Agreement defined climate warming targets of 2°C, 1.5°C, and 1°C, associated with end-of-century atmospheric CO<sub>2</sub> concentrations of ca. 450, 400, and 350 ppm, respectively. These scenarios approach carbon neutrality by about 2070, 2055, and 2040, respectively, and remain negative thereafter (Hansen et al. 2017). More than 130 countries have proposed commitments on the reduction of individual carbon neutrality-related emission.

Adoption and implementation of the actions proposed by many countries will introduce another set of perturbations to the Earth system on decadal timescales. Continued emission growth, followed by the transition from peak to net-zero emissions (2040–2060), will be superimposed on changes that have already occurred throughout the industrial era. Confounding this adjustment to emissions are intended CDR (e.g., artificial ocean alkalization) and ecosystem restoration (e.g., mangrove forest restoration) activities that are planned or already underway. The combination of these activities will again affect the global carbon cycle by producing abrupt CO<sub>2</sub> redistribution within the Earth system, including at the land-ocean interface. Understanding the response of the coastal ocean carbon sink to these perturbations, individually and collectively, is a major challenge that has implications for the overall global carbon budget and carbon-climate coupled system. The temporal evolution of the air-sea CO<sub>2</sub> flux in the coastal ocean from preindustrial to present time provides useful guidance for suggesting scenarios for possible future trends (**Figure 6**).

### 6.1. From Preindustrial to Present Time

Analysis of coastal ocean carbon cycling based on conceptual box models suggested that the global continental margin was a preindustrial source of atmospheric CO<sub>2</sub>, providing 0.1–0.3 Pg C year<sup>-1</sup> (intermediate value of ~0.2 Pg C year<sup>-1</sup> in **Figure 6a**) as a result of respiring terrestrially derived organic matter (e.g., Mackenzie & Lerman 2006; Mackenzie et al. 2004, 2002, 2000). Bauer et al. (2013) further suggested that over the past century these ocean margins have progressively shifted to a CO<sub>2</sub> sink of –0.1 to –0.4 Pg C year<sup>-1</sup> as a result of rising atmospheric CO<sub>2</sub> levels, circulation changes, and increasing riverine nutrient inputs. This source-to-sink shift suggests a current anthropogenic CO<sub>2</sub> sink of –0.2 to –0.7 Pg C year<sup>-1</sup> (intermediate value of ~–0.45 Pg C year<sup>-1</sup> in **Figure 6b**), with a larger flux density than the open ocean, which is driven by enhanced biological production in the coastal ocean (Mackenzie et al. 2000, 2004).

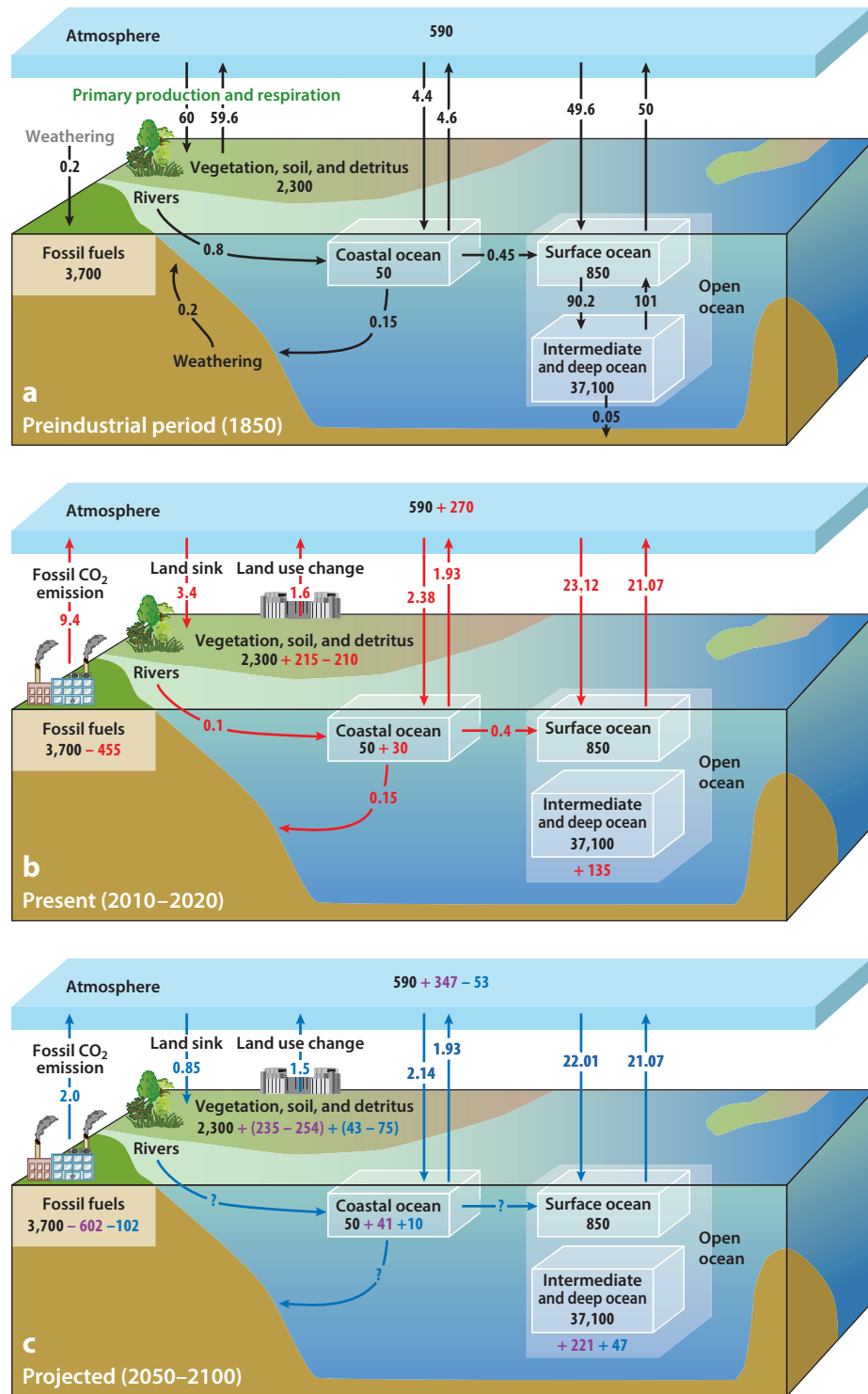
In contrast, simulations using a GOBM suggested that the preindustrial coastal ocean may have been a CO<sub>2</sub> sink of –0.04 Pg C year<sup>-1</sup> (Lacroix et al. 2021a), mostly driven by low temperate and biological production on outer shelves and efficient offshore export of terrestrial dissolved organic matter. The same simulations indicated that this sink increased to –0.15 Pg C year<sup>-1</sup> in present time, suggesting a current anthropogenic CO<sub>2</sub> uptake of only –0.11 Pg C year<sup>-1</sup> (Lacroix et al. 2020). In another GOBM-based analysis, Bourgeois et al. (2016) found a similar anthropogenic sink of –0.12 Pg C year<sup>-1</sup>. The lower anthropogenic coastal CO<sub>2</sub> sink suggested by the GOBMs compared to that estimated using box models and mass balance analysis may be a result of different definitions and representations of the coastal ocean. Global box models do not consider the circulation heterogeneity of different coastal regions, which is explicitly resolved in GOBMs, thereby producing shorter water residence time on the shelves (Lacroix et al. 2021a), although the resolution of GOBMs remains too coarse to fully resolve the physical processes on continental shelves. In contrast, box models typically include estuaries, salt marshes, and

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**Carbon neutrality:**  
a state in which anthropogenic CO<sub>2</sub> emissions are balanced globally by anthropogenic CO<sub>2</sub> removals over a specified period

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(Caption appears on following page)

**Figure 6** (Figure appears on preceding page)

Schematic of (a) preindustrial (1850), (b) present (2010–2020), and (c) future (2050–2100) global carbon budgets. Fluxes of total carbon (organic plus inorganic, in Petagrams of carbon per year) between different reservoirs (numbers in the boxes represent the reservoirs in Petagrams of carbon) for preindustrial (*black arrows and numbers*; natural background), present (*red arrows and numbers*; anthropogenically induced changes), and after carbon neutrality (*blue arrows and numbers*; anthropogenically induced changes) are estimated based on Sarmiento & Gruber (2002), Aricò et al. (2021), Bauer et al. (2013), Friedlingstein et al. (2020), Regnier et al. (2013b), and Jones et al. (2016). The fluxes in panels *b* and *c* are on top of the natural fluxes in panel *a*. In panel *a*, the land source of  $59.6 \text{ Pg C year}^{-1}$  includes the degassing of  $\sim 1 \text{ Pg C year}^{-1}$  from inland waters during the preindustrial era. The cumulative  $\text{CO}_2$  in the atmosphere is expected to peak (Jones et al. 2016) is shown in panel *c* (*purple numbers*). For the land sink, the positive number is an inferred terrestrial land sink and the negative value is the decrease due to deforestation. In panels *a* and *b*, the gross oceanic carbon source to the atmosphere is partitioned between the coastal ocean and the open ocean based on surface area proportions of 8.4% for the global coastal ocean and 91.6% for the open ocean. In panels *b* and *c*, the increase in the coastal ocean carbon inventory is estimated based on the ratio of contemporary anthropogenic  $\text{CO}_2$  uptake by the coastal ocean to global ocean. Details of the estimates are provided in **Supplemental Material Section 6**.

mangroves, the water components of which generally serve as a source of carbon to the atmosphere and are not resolved in three-dimensional GOBMs.

Nevertheless, observational studies have suggested a tendency for enhanced ocean margin uptake of atmospheric  $\text{CO}_2$  over the past three decades (Laruelle et al. 2018). Using a conceptual model, Bauer et al. (2013) and Cai (2011) also suggested an increasing global shelf  $\text{CO}_2$  sink in response to increased atmospheric  $p\text{CO}_2$ . These observation- and model-based studies agree that temperate and subpolar shelves may be stronger sinks of anthropogenic  $\text{CO}_2$  than the open ocean. In contrast, modeling studies suggest weak anthropogenic sinks for most tropical and Arctic shelves, regions that are poorly represented in the study of Laruelle et al. (2018), and may explain the differing conclusions at the global scale.

Significant differences in trends of air-sea  $\text{CO}_2$  fluxes, however, occur between many coastal regions and the adjacent open-ocean waters, particularly for coastal upwelling systems (e.g., Peruvian upwelling current, OceMar) and those dominated by large river plumes (e.g., Amazon River plume, RiOMar) (Bourgeois et al. 2016). The three case studies presented in Section 4 also demonstrated clear differences in long-term trends of  $\text{CO}_2$  fluxes. These observed differences indicate that physical and/or biogeochemical processes other than the abiotic atmospheric  $\text{CO}_2$  can be important controls on carbon flux evolution in specific coastal regions.

The different models are consistent in identifying terrestrial inputs via river discharges and ocean margin-open ocean exchanges as primary controls of coastal ocean carbon cycling. Regnier et al. (2013b) suggested an anthropogenic perturbation of an enhanced riverine discharge flux of  $0.10 \text{ Pg C year}^{-1}$  (**Figure 6b**), 70% of which can be exported to the open ocean (Bourgeois et al. 2016). Increased riverine nutrients and TA loads affect coastal carbon cycling by enhanced biological uptake and changes in seawater buffering capacity, which are rarely represented in current GOBMs that assume constant present-day riverine loads of both carbon and nutrients. Lacroix et al. (2020) estimated that present-day global riverine nutrients loads have increased relative to preindustrial times by 8–197%, 37–144%, and 115–141% for phosphorus, nitrogen, and silicate, respectively (Beusen et al. 2009, 2016; Frings et al. 2016; Mayorga et al. 2010). Changes in TA are less well constrained, although Goll et al. (2014) suggested an increase of ca. 11% in the weathering supply of alkalinity over the historical time period. A schematic for the evolution of the land-derived forcing is shown in **Supplemental Figure 5.2**.

With increased terrestrial inputs into the coastal ocean, the anthropogenic-induced increase in sedimentary burial of organic carbon and the lateral export to the open ocean are estimated to



**RCP 2.6:** one pathway where radiative forcing peaks at approximately  $3 \text{ W m}^{-2}$  and then declines to be limited at  $2.6 \text{ W m}^{-2}$  in 2100

be  $0.15$  (Regnier et al. 2013b) and  $0.4 \text{ Pg C year}^{-1}$  (Bauer et al. 2013), respectively, comparable to the natural fluxes at preindustrial times (**Figure 6a,b**). The enhanced biological production and carbon sink increased the coastal ocean carbon inventory by  $30 \text{ Pg C}$  from 1850 to 2010–2020, nearly 60% of the coastal ocean carbon inventory estimated for preindustrial times.

## 6.2. From Present to Future

The temporal evolution of atmospheric  $p\text{CO}_2$  at the end of this century has been assessed for various RCP scenarios. Under the low carbon or neutrality scenario (RCP 2.6), atmospheric  $\text{CO}_2$  will peak in two to three decades followed by a gradual decrease until the end of this century (**Supplemental Figure 5.2i**). In response to these projected atmospheric  $\text{CO}_2$  changes, the strength of the global ocean carbon sink may slowly increase for another 10 years but rapidly shift to a continuous decrease (Jones et al. 2016). The eventual decline in atmospheric  $p\text{CO}_2$  under the RCP 2.6 scenario may be offset by  $\text{CO}_2$  fluxes from the ocean, an expected ocean feedback to reductions in emissions (Keller et al. 2018, Koch et al. 2021). Implementation of marine CDR would be needed to sustain the ocean carbon sink, which may further change ocean carbon cycling.

With progressive emission reductions and CDR, the atmospheric  $p\text{CO}_2$  will decline and eventually approximate that of sea surface water, resulting in a less efficient natural uptake of atmospheric  $\text{CO}_2$  in the coastal ocean (**Supplemental Figure 5.2k**). Changes in coastal-ocean physics and biology as well as in riverine inputs and sediment interactions will modify the distribution of carbon and alkalinity (Hu & Cai 2011, Mackenzie et al. 2004), also affecting the potential of the coastal ocean to absorb anthropogenic carbon. A potential future outcome is that enhanced anthropogenic activities (Regnier et al. 2013b) and precipitation (Battin et al. 2009) will increase carbon export from land to the coastal ocean. Riverine organic carbon inputs tend to produce excess DIC in the coastal ocean because of efficient remineralization of the higher carbon to nitrogen ratio in terrestrial organic matter relative to use by aquatic primary production (Bauer et al. 2013, Lacroix et al. 2020). Another possibility is a decline in the terrestrial input of nutrients after the peak of fertilizer use (**Supplemental Figure 5.2c,d**), producing reduced DIC deficits in RiOMar systems and leading to a decline in coastal ocean carbon sinks and carbon burial (**Supplemental Figure 5.2**).

The responses of OceMar systems are complex because of the influence of boundary currents that are part of the large-scale ocean basin circulation (**Supplemental Figure 5.2g–i**). Taking the ocean margins adjacent to the WBC in the WNP as an example, warming in the Kuroshio region would lead to a northward shift of the subtropical circulation system (Hu et al. 2015) and strengthening of the Kuroshio (Chen et al. 2015), thereby weakening the intrusion of open ocean water masses into the adjacent ocean margins (e.g., Wu et al. 2017). As a result, reduced nutrient and DIC influxes from the open ocean would be expected. Projections for EBC systems suggest that the strength of these upwelling systems will be enhanced under warming scenarios (Sydean et al. 2014), thereby increasing the supply of carbon and nutrients from depth to the surface waters. However, how the  $\text{CO}_2$  source/sink role of OceMar systems will change is dependent on changes in the ratio between DIC and nutrients of source waters and the physical settings that affect water residence times and hence biological controls on the carbon cycle.

Interventions that move toward carbon neutrality will significantly influence the global carbon cycle and the carbon fluxes among different reservoirs. Anthropogenic carbon emissions are projected to decrease by more than two-thirds between 2050 and 2100 under the RCP 2.6 scenario, reducing the atmospheric carbon inventory by  $53 \text{ Pg C}$  (**Figure 6c**). Correspondingly, the carbon sink of the ocean, either the open or coastal ocean, will also decrease by more than half relative to the present-day scenario, from  $-2.05$  to  $-0.94 \text{ Pg C year}^{-1}$  for the open ocean



and from  $-0.45$  to  $-0.21$  Pg C year<sup>-1</sup> for the coastal ocean (**Figure 6b,c**). The carbon inventory of the coastal ocean and open ocean is estimated to increase by 10 and 47 Pg C, respectively, relative to the present-day scenario (**Figure 6c**). The coastal ocean carbon budget thus shows the greatest response to progressive future reductions in emission reductions and implementation of CDR techniques. However, carbon transport fluxes along the land-coastal ocean-open ocean continuum under low carbon emission scenarios are essentially unknown (**Figure 6c**). Thus, the challenges associated with identifying and quantifying current and projected responses of the coastal ocean and its source/sink role in the global carbon budget are daunting. Addressing these challenges requires coordinated observational and modeling efforts focused at both regional and global scales. The development of these programs is a priority for the ocean carbon research and management community.

### SUMMARY POINTS

1. Rapid increases in ocean carbon observations have allowed further improvement in estimating the global coastal ocean CO<sub>2</sub> sink and its spatial distribution. An updated compilation of air-sea CO<sub>2</sub> fluxes based on observations reported in the literature shows that the global coastal ocean represents an integrated CO<sub>2</sub> sink of  $-0.25 \pm 0.05$  Pg C year<sup>-1</sup> with a flux density of  $-0.68 \pm 0.14$  mol C m<sup>-2</sup> year<sup>-1</sup>, confirming that the coastal ocean is an efficient CO<sub>2</sub> sink. The strongest CO<sub>2</sub> uptake occurs at high latitudes with the combined contributions from polar ( $-134$  Tg C year<sup>-1</sup>) and subpolar ( $-108$  Tg C year<sup>-1</sup>) regions accounting for the majority of the global coastal carbon sink. Moderate CO<sub>2</sub> uptake with highest seasonal dynamic ranges occurs at mid-latitudes. Alternating uptake and outgassing of CO<sub>2</sub> occurs at low latitudes. The latitudinal pattern in CO<sub>2</sub> flux and its seasonality are controlled by both temperature and nonthermal factors, such as water mass mixing and net primary production, characteristics of coastal ocean carbon cycling that differ from those of the open ocean.
2. A framework based on river-dominated ocean margin (RiOMar) and ocean-dominated margin (OceMar) systems is used to conceptualize coastal ocean carbon cycle processes and provide a semiquantitative diagnosis of individual coastal ocean systems. Application of this framework highlights differences in exchanges and boundary transport along the river-estuary-shelf-ocean continuum and the intrinsic biogeochemical reactions that determine air-sea CO<sub>2</sub> fluxes.
3. Coastal ocean carbon models, extending from conceptual box (budget) models to three-dimensional coupled circulation-biogeochemical models, have been successfully implemented in a range of coastal ocean regions. These models simulate key processes, provide projections of future trends, and highlight uncertainties and challenges in our understanding of coastal carbon cycling. Global ocean biogeochemical models (GOBMs) with improved spatial resolution allow simulation of important features and global trends in the global coastal ocean, such as coastal water residence times, air-sea CO<sub>2</sub> exchange, and riverine fluxes. GOBMs are also instrumental in simulating global trends in coastal carbon cycle processes and fluxes.
4. Observations and numerical model simulations are revealing long-term trends in ocean carbon fluxes, which have been perturbed since preindustrial times. The anthropogenic alterations are anticipated to continue before and after net-zero CO<sub>2</sub> emissions (i.e.,



carbon neutrality) are reached, but considerable uncertainty is associated with impacts arising from concurrent changes in the land-ocean-atmosphere coupled system. The observed differences in carbon fluxes and their long-term trends among different coastal systems indicate that physical and/or biogeochemical processes other than the abiotic atmospheric CO<sub>2</sub> can be important controls on the evolution of air-sea CO<sub>2</sub> exchanges in specific coastal regions.

## FUTURE ISSUES

1. The ability to conceptualize and simulate future trends in the coastal ocean carbon cycle has been greatly advanced by improved understanding of spatio-temporal variability of carbon fluxes. This highlights the importance of continuing and enhancing ocean carbon observation programs that are coordinated with measurements of physical, chemical, and biological parameters that can be used to provide information that is responsive to regional interests, policies, and management. The continued development of global data products, such as the Surface Ocean CO<sub>2</sub> Atlas, with improved coordination across the land-ocean continuum is critical to further advance understanding of ocean carbon dynamics.
2. Regional models that fully couple the land-ocean continuum and global coastal ocean models that meaningfully resolve and reconcile coastal physical processes are equally important for the development of predictive capabilities at both regional and global scales. Predictive capability is increasingly needed to project the role of the coastal ocean in the global carbon cycle and inform policies developed to achieve net-zero carbon emissions by the mid-twenty-first century. Here, improvements in data collection and advances in model structure and computational resources that allow integration of data and models are essential priorities. Improved representation of coastal processes through use of conceptual frameworks such as the RiOMar and OceMar systems provides an avenue to improve model skill and identify needed observations.
3. Identification of a baseline conditions representative of preindustrial times is critical for assessing past effects of human activities and for projecting future trends in coastal ocean carbon cycling and air-sea CO<sub>2</sub> fluxes. Estimating this baseline condition and its associated uncertainty has important implications for advancing the ability to detect and quantify changes in the coastal ocean carbon cycle.

## DISCLOSURE STATEMENT

The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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