Enhancing air–sea CO_2 exchange and modulating seawater carbonate–pH dynamics: the role of wave effect mechanisms in the POP2–waves coupled model

Yung-Yao Lan[1,2], Huang-Hsiung Hsu[1], Wei-Liang Lee[*1] and Simon Chou[1]

[1]Research Center for Environmental Changes, Academia Sinica, Taipei 11529, Taiwan

[2]National Land Resources Conservation Center, National Chung Hsing University, Taichung 40227, Taiwan

Correspondence to: Wei-Liang Lee (leelupin@gate.sinica.edu.tw)

Abstract

In this study, a wave module is online-coupled into the Parallel Ocean Program version 2 (POP2) within the CESM1.2.2 framework (hereafter POP2–waves). Unlike empirical data analyses of observation-based products and offline-forced ocean models that treat wave-induced gas transfer velocity and the air–sea difference in partial pressure of CO_2 ($dpCO_2$) as decoupled variables, the online-coupled POP2–waves model captures their interactive, $dpCO_2$-driven negative feedbacks. This setup enables wave properties to dynamically modulate gas transfer velocity and physical mixing through sequential processes. POP2–waves is evaluated alongside a control simulation (B–CTL) against the National Oceanic and Atmospheric Administration (NOAA) CarbonTracker (CT2022) inversion product. The spatial air–sea CO_2 flux in POP2–waves shows closer structural agreement with the NOAA CT2022 than the control B–CTL. Specifically, bubble-mediated transfer accounts for up to 41.3% of the total flux, consistent with the ~40% contribution reported in recent research. POP2–waves enhances regional oceanic CO_2 uptake and outgassing by 11.8% and 41.6%, respectively, yielding a 1.8% increase in the global net ocean CO_2 sink compared to the B–CTL simulation. Globally, air–sea $dpCO_2$ and pH exhibit the strongest positive and negative regression coefficients with the air–sea CO_2 flux. Regionally, the gas transfer velocity shows a positive (negative) regression coefficient within oceanic CO_2 outgassing (uptake) regions, whereas SST displays the opposite trend.

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已删除: Wave and bubble-mediated processes impact air–sea CO_2 flux by enhancing the gas transfer velocity—normalized to a Schmidt number of 660 ($kK_{w,660}$)—primarily through their dependence on significant wave height (H_s). Neglecting wave and bubble processes may lead to an underestimation of air–sea CO_2 flux under high 10-m wind speeds (U_{10}) in most state-of-the-art climate models. In this study, a waves module from the Princeton Ocean Model (POM) is integrated into the Parallel Ocean Program version 2 (POP2) within the CESM1.2.2 framework. This coupled model, termed POP2–waves, is evaluated alongside a control simulation (B–CTL) against National Oceanic and Atmospheric Administration (NOAA) CarbonTracker (CT2022) air–sea CO_2 flux data. Overall, bubbles contribute up to 41.3% to the total air–sea CO_2 flux, a finding consistent with recent literature. Furthermore, the spatial distribution of air–sea CO_2 flux in POP2–waves broadly agrees with NOAA CT2022 product, although regional discrepancies remain. Under the $dpCO_2$-driven negative feedback, the POP2–waves experiment yields increases of 11.8% and 41.6% in negative and positive air–sea CO_2 flux regions, with a 1.8% increase in the global ocean carbon sink compared to the B–CTL control. The air–sea $dpCO_2$ (pH) exhibits the strongest positive (negative) regression coefficient with air–sea CO_2 flux across the global ocean. Additionally, the $K_{w,660}$ gas transfer velocity shows a positive (negative) regression coefficient within regions of the positive (negative) air–sea CO_2 flux regions, whereas SST displays the opposite pattern

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1. Introduction

The exchange of CO_2 between the air and sea is a crucial component of the global carbon cycle, carrying significant implications for Earth's climate (Bange et al., 2024; Friedlingstein et al., 2022; McKinley et al., 2020; Müller et al., 2023; Shutler et al., 2019). Traditionally, this air–sea exchange is characterized using the bulk formula (e.g., Wanninkhof, 1992, 2014; McKinley et al., 2020; Dong et al., 2022; Fay et al., 2021; Zhou et al., 2023; Heimdal et al., 2024):

$$F_{CO_2} = k_{w,660} \cdot k_0 \cdot (pCO_2^w - pCO_2^a) \cdot (1 - ice) \qquad (1)$$

where the air–sea CO_2 flux (F_{CO_2}, mol m^{-2} yr^{-1}) is determined by the gas transfer velocity expressed relative to a Schmidt number of 660 ($k_{w,660}$, cm hr^{-1}), the difference in partial pressure of CO_2 ($dpCO_2$, µatm) between the seawater and atmosphere (indicated by superscripts " w " and " a ", respectively), and the solubility constant (k_0; mol m^{-3} atm^{-1}), which varies with salinity and temperature (Weiss, 1974). Note that an appropriate scaling factor (8.76×10^{-5}) is implicitly required to convert the dimensions of $k_{w,660}$ and $dpCO_2$ into the final flux unit (mol m^{-2} yr^{-1}). The sign of air–sea CO_2 flux is determined by $dpCO_2$, where a positive air–sea CO_2 flux indicating ocean outgassing to the atmosphere. Additionally, the calculated fluxes are weighted by the ice-free fraction ($1 - ice$), where 'ice' represents the sea ice fraction. The estimation of bulk air–sea CO_2 fluxes using the gas transfer velocity is typically based on the Wanninkhof (1992) formulation:

$$k_{w,660} = 0.31 \langle U_{10}^2 \rangle (Sc/660)^{-0.5} \qquad (2)$$

where $\langle U_{10}^2 \rangle$ represents the squared neutral mean wind speed at 10 m above the surface (m s^{-1}), and Sc denotes the Schmidt number. The flux estimate error can reach as high as 34% due to model limitations and uncertainties surrounding bulk parameters. (Signorini and McClain, 2009). Furthermore, substantial uncertainties persist in air–sea

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CO_2 flux estimates, primarily stemming from an incomplete understanding of the spatiotemporal variability in the governing mechanisms (Shutler et al., 2019).

Some studies have highlighted that estimating air–sea CO_2 fluxes involves complexities well beyond neutral wind speed and solubility factors. Monahan and Spillane (1984) previously regarded whitecaps as "low impedance vents", that effectively "shorten" the water-side transfer resistance. Without wave breaking, air–sea CO_2 exchange occurs via mass transport, which under non-turbulent conditions is governed by molecular diffusion, wave breaking then acts as a transitional process from laminar to turbulent flow (Deike, 2022). Furthermore, bubbles provide additional surface area for gas transfer; as they rise through the aqueous mass boundary layer and burst at the sea surface, they significantly enhance near-surface turbulence (Soloviev and Lukas, 2010; Bell et al., 2017; Deike and Melville, 2018; Krall et al., 2019; Czerski et al., 2022). Gutiérrez-Loza et al. (2022) pointed out that during high and intermediate wind speeds (>6–8 m s^{-1}), enhanced air–sea CO_2 exchange is primarily driven by wave-breaking dynamics and bubble mediation, which occur alongside the generation of sea spray droplets that contribute to atmospheric cloud condensation nucleation. Conversely, under low wind conditions (<6 m s^{-1}), water-side convection becomes the dominant control mechanism. Beyond these calm periods, the critical role of breaking waves and bubble injection mechanisms in facilitating CO_2 transfer has been widely corroborated (e.g., Andreas et al., 2016; Brumer et al., 2017; Blomquist et al., 2017; Reichl and Deike, 2020; Li et al., 2023; Zhou et al., 2023; Rustogi et al., 2025). Specifically, Deike and Melville (2018) demonstrated that the bubble-mediated contribution to CO_2 transfer exceeds 40% at a neutral 10-meter wind speed (U_{10}) ≥ 10 m s^{-1}, becomes dominant at $U_{10} \geq 15$ m s^{-1} and reaches 60% at 20 m s^{-1}.

Monitoring sea surface CO_2 is crucial for understanding Earth system dynamics, as climate change has begun to impact the ocean's carbon uptake capacity (Behncke et

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al., 2024). However, air–sea CO_2 fluxes estimates derived from products based on the partial pressure of CO_2 (pCO_2) often suffer from significant uncertainties, primarily because they rely on empirical parameterizations of the gas transfer velocity (e.g., Williams et al., 2017; Gray et al., 2018; Coggins et al., 2023; Behncke et al., 2024; Fay et al., 2024; Yang et al., 2024). To resolve this issue and independently constrain $k_{w,660}$, direct flux measurements using the eddy covariance (EC) method are widely utilized as a benchmark reference (Dong et al., 2021). By directly acquiring the turbulent flux F_{CO_2} via the EC method and pairing it with simultaneous measurements of $dpCO_2$, researchers can inversely calculate the gas transfer velocity ($k_{w,660} = F_{CO_2} / (k_0 \times dpCO_2)$). This approach provides a direct observational constraint to evaluate and calibrate these observation-based products under complex marine conditions. However, obtaining reliable direct fluxes from shipborne EC setups presents its own instrumental challenges. Marine EC systems are generally conducted using either open- or closed-path infrared gas analyzers. A closed-path analyzer (e.g., LI-7000, LI-COR) operates with an enclosed measurement chamber (Sutton et al., 2014, 2021; Bakker et al., 2016; Sabine et al., 2020; Akhand et al., 2021; Wu and Qi, 2023), whereas open-path analyzers (e.g., LI-7500, LI-COR) measure infrared absorption directly in ambient air (Edson et al., 2011; Tokoro et al., 2014; Bell et al., 2017; Dong et al., 2021; Van Dam et al., 2021). While both configurations are inherently susceptible to H_2O cross-sensitivity, each system possesses distinct advantages and systematic limitations (Honkanen et al., 2018). Evaluating and constraining global Earth system models (ESMs) with direct, long-term observations of air–sea CO_2 flux remains challenging due to the severe scarcity of continuous field measurements. Consequently, alternative validation datasets are typically derived by upscaling sparse shipbased pCO_2 observations combined using statistical interpolation, machine learning, atmospheric inversion, and climatological averaging to construct long-term, gridded flux products.

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Although several recent studies have estimated the air–sea CO_2 flux using Eq. (1), current parameterizations in oceanic and atmospheric models still rely exclusively on neutral wind speed (Long et al., 2013; Moore et al., 2013; Couldrey et al., 2016; Jin et al., 2017; Lovenduski et al., 2019; Ziehn et al., 2020; Chikamoto and DiNezio, 2021). Similarly, surface ocean observation-based products typically calculate those flux using standard parameterizations (Wanninkhof et al., 1992, 2014; Fay et al., 2021). Crucially, the persistent uptake of anthropogenic CO_2 is lowering seawater pH and altering carbonate system in nonlinear ways, which further reducing the oceans' capacity to absorb additional CO_2 (Moore et al., 2013). The mechanisms of breaking waves and bubble injection in facilitating the total gas transfer velocity have been investigated primarily through empirical data analyses of observation-based products (e.g., Andreas et al., 2016; Blomquist et al., 2017; Reichl and Deike, 2020; Zhou et al., 2023). Rustogi et al. (2025) employed a fully coupled ocean–biogeochemistry modeling system based on the MOM6 ocean circulation model and the COBALTv2 biogeochemical module. Utilizing this framework, they simulated ocean dynamics, temperature, and carbon cycling processes—including dissolved inorganic carbon (DIC) and pCO_2—while explicitly accounting for wave effects on air–sea CO_2 exchange. Furthermore, they identified a nonlinear pCO_2 feedback mechanism within the coupled ocean–carbon system that regulates air–sea CO_2 flux. However, their configuration relies on ocean and biogeochemical modules that are strictly forced by offline atmospheric reanalysis and wave model outputs. This decoupled or single-directional forcing approach faces limitations in capturing high-frequency, transient air–sea interactions (e.g., rapid wind shifts or wave breaking dynamics) that significantly modulate gas exchange. In contrast, our study introduces an online coupling framework that integrates wave effects directly into the flux simulation interface at each model time step. This online approach provides a key value-add by enabling real-time, interactive feedback between wave dynamics

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and the seawater carbonate system, thereby resolving the non-linear high-frequency physical controls on the air–sea CO_2 flux that are often overlooked in offline-forced systems.

In this study, we investigate how breaking waves and bubble injection mechanisms influence air–sea CO_2 flux and ocean's buffering capacity. Specifically, we examine the biogeochemical responses driven by these physical mechanisms within the seawater carbonate–pH system. To achieve this, we utilize the Parallel Ocean Program version 2 (POP2; Smith et al., 2010) of the Community Earth System Model version 1.2.2 (CESM1.2.2; Hurrell et al., 2013) online coupled with waves module (Mellor et al., 2008) of the Princeton Ocean Model (POM; Blumberg and Mellor, 1987). The coupled model is referred to as the POP2–waves model.

The structure of this paper is organized as follows. Section 2 introduces the model, data, methodology, and experiments employed in this study, and defines twelve key regions of high air–sea CO_2 flux variability identified by NOAA CT2022 to validate the model simulation. Section 3 evaluates the performance of the POP2–waves coupled model in simulating air–sea CO_2 flux and its interaction with the carbonate–pH system over the Western Pacific (WP; 160–180°E, 35–40°N) and the Equatorial Pacific (EP; 230–250°E, 0–5°S) regions. Section 4 compares the carbonate–pH dynamics of POP2–waves and B–CTL, examine the primary drivers of air–sea CO_2 exchange, and evaluate the uncertainties arising from the absence of $dpCO_2$-driven negative feedback. Finally, Section 5 presents the conclusions.

2. Data, model experiments, and methodology

2.1 Description of the model framework and experiments

In this study, we investigated the role of waves and bubble mechanisms in modulating ocean biogeochemistry response by comparing the control simulation (B–

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CTL) with the coupled POP2–waves model within the CESM1.2.2 framework (Fig. 1). The CESM simulation was conducted using a fully coupled component set over a 30-year period, with well-mixed greenhouse gases (CO_2, CH_4, N_2O, etc.), ozone, and aerosols fixed to year-2000 values (the B_2000_CAM5 compset). The framework features a horizontal resolution of 1.9° × 2.5° in the Community Atmosphere Model 5.3 (CAM5.3) and a nominal 1° horizontal resolution with 60 vertical layers in POP2, the Community Land Model version 4 (CLM4), the prognostic Los Alamos Sea Ice Model (CICE), and the coupler (CPL) (Hurrell et al., 2013). POP2 is forced by CAM5.3 through CPL via four primary fields: (1) momentum fluxes (zonal and meridional wind stress and friction velocity), (2) heat fluxes (net shortwave radiation, sensible heat flux, longwave radiation, and heat flux from snow/ice melt), (3) freshwater fluxes (precipitation, evaporation, river runoff, and ice melt), and (4) surface winds (10 m zonal and meridional wind speeds). Furthermore, POP2 interacts with the Marine Biogeochemistry (MARBL; Long et al., 2021) module to regulate carbonate chemistry and air–sea CO_2 fluxes. This interaction is mediated via ocean dynamics (currents and sea surface height), K-profile vertical mixing, and tracer transport, with wave-induced processes providing additional physical constraints. Within MARBL, variations in DIC and nutrients (NO_3, PO_4, and Fe) are primarily controlled by biological processes: photosynthesis consumes DIC and nutrients, whereas respiration and remineralization return organic carbon to the DIC pool. Together with the wave module, these biogeochemical and physical processes jointly regulate the carbon cycle.

The control simulation (B–CTL), which served as the baseline for air–sea CO_2 flux and seawater acidification, utilized Eq. (1) with the gas transfer velocity according to Wanninkhof (1992) (Eq. 2). Both the B–CTL and POP2–waves experiments were integrated under a present-day scenario (starting from the year 2000) to estimate air–sea CO_2 flux pH variations. This experimental design allowed us to isolate the impact

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of wave effects on carbonate chemistry and directly compare our results with the NOAA CarbonTracker CT2022 product (Jacobson et al., 2023). For the POP2–waves experiment, the waves module (Mellor et al., 2008) from the POM model was online coupled into POP2. This configuration incorporated bubble-mediated gas transfer—computed from a mechanistic model for air bubble entrainment at the breaking-wave scale (Deike and Melville, 2018)—integrated with the POP2 marine biogeochemistry module.

Specifically, POP2–waves experiment adopts the advanced parameterization framework expanded by Reichl and Deike (2020), and Deike (2022). In this framework, both the non-bubble and bubble-mediated gas transfer velocity are explicitly resolved as a function of the ocean surface friction velocity, (u^*, m s^{-1}) and significant wave height, (H_s, m). This parameterization effectively captures the main wave effect, as well as solubility and diffusivity. To standardize the formulation for CO_2, the total gas transfer velocity is expressed relative to a Schmidt number of 660. Following Deike and Melville (2018), it is calculated as the sum of the non-bubble ($k_{wNB,660}$) and bubble ($k_{wB,660}$) components:

$$K_{w,660} = K_{wNB,660} + K_{wB,660} = A_{NB} u^* \left(\frac{Sc}{660}\right)^{-1/2} + \frac{A_B}{K_0\, R\, T_0} (u^*)^{5/3} (g\, H_s)^{2/3} \left(\frac{Sc}{660}\right)^{-1/2} \quad (3)$$

where A_{NB} is an empirical, nondimensional coefficient given as 1.55 × 10^{-4}, A_B is a dimensional fitting coefficient (1.2 × 10^{-5} s^2 m^{-2}), R is the ideal gas constant (0.082 L atm mol^{-1} K^{-1}), T_0 is the sea surface temperature, SST (K), and g is gravitational acceleration (9.806 m s^{-1}). Section 3.3 examines the contribution ratios of the non-bubble and bubble components in simulating the air–sea CO_2 flux.

The marine carbonic acid system is a nonlinear, coupled system governed by CO_2 dissolution equilibrium, ($CO_{2(aq)} + H_2O \overset{K_H}{\leftrightarrow} H_2CO_{3(aq)}$), where the Henry's constant (K_H) for CO_2 in water at 25°C is approximately 3.4 x 10^{-2} (m atm^{-1}), two-step

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dissociation reactions, and the conservation of the total dissolved inorganic carbon (DIC) and total alkalinity (TA), ultimately determining pH and the distribution of carbonate species. The H_2CO_3 has two hydrogen ions and dissociates in two steps:

$$H_2CO_3 \overset{K_{a1}}{\longleftrightarrow} H^+ + HCO_3^- \qquad K_{a1} = 4.4 \times 10^{-7} \qquad (4)$$

$$HCO_3^- \overset{K_{a2}}{\longleftrightarrow} H^+ + CO_3^{2-} \qquad K_{a2} = 4.7 \times 10^{-11} \qquad (5)$$

The DIC can be expressed as

$$\mathrm{DIC} = [H_2CO_3] + [HCO_3^-] + [CO_3^{2-}]$$

$$= K_H pCO_2^w + \frac{K_{a1} K_H pCO_2^w}{[H^+]} + \frac{K_{a1} K_{a2} K_H pCO_2^w}{[H^+]^2}$$

$$= K_H pCO_2^w \left(1 + \frac{K_{a1}}{[H^+]} + \frac{K_{a1} K_{a2}}{[H^+]^2}\right)$$

$$= K_H pCO_2^w ([H^+]^2 + K_{a1} [H^+] + K_{a1} K_{a2})/[H^+]^2 \qquad (6)$$

The marine carbonate system describes the distribution of DIC among its chemical species in seawater and is jointly governed by TA and pH. From Eq. (6), if the oceanic pCO_2^w and pH are known, the DIC can be determined, conversely, pCO_2^w can be inferred from DIC. TA is defined as the sum of the proton-neutralizing capacities of all weak acid anions in seawater (Dickson, 1981) and can be expressed as: $TA = [HCO_3^-] + 2[CO_3^{2-}] + [B(OH)_4^-] + [OH^-] + [HPO_4^{2-}] + 2[PO_4^{3-}] + [SiO(OH)_3] + [NH_3] + [HS^-] - [H^+] - [H_3PO_4]$. In the POP2–waves framework, TA, pH and biogeochemistry are also used to calculate pCO_2^w.

When TA increases, seawater becomes more alkaline, leading to a higher CO_3^{2-} concentration and enhanced buffering of hydrogen ions. Consequently, $[H^+]$ decreases, causing pH to rise and boosting the ocean's capacity to absorb CO_2. Conversely, when TA decreases, the buffering capacity weakens, $[H^+]$ increases, pH drops, rendering the ocean more acidic with a reduced ability to uptake CO_2.

2.2 Coupling POP2–waves Methodology

Except for the ocean component, all other components adhere strictly to the CESM 1.2.2 framework. The wave module is forced by variables exchanged through the coupler, the POP2, and a dedicated input initialization file. Specifically, in addition to the baseline component interactions in CESM1.2.2, the framework passes the zonal and meridional 10-m winds, along with surface friction velocity (u^*, accounting for both open-ocean and t sea-ice fractions) from the CPL to evaluate the wave properties and the associated gas transfer velocity. To drive the wave module calculations, key physical variables from POP2; this involves interpolating the depth-variable currents from the POP2 z-coordinates (60 levels of zonal and meridional horizontal velocities) onto the POM (waves) sigma-coordinates (21 vertical levels). This vertical mapping is r equired to compute both the depth-dependent wave radiation equation and the specified spectrum. Additionally, spatial grid configurations across the the x, y, and z domains and timestep configurations are exchanged. The wave module further integrates external variables (F1–F4), which represent the spectrally averaged components of wave radiation stress incorporated into the momentum equation.

Following Mellor et al. (2008), the significant wave height (H_s) utilized in the wave radiation equation represents the total wave energy integrated over the full spectrum, which inherently encompasses both locally wind-generated seas and remotely generated swells. During the simulation of wave energy and propagation, improper handling of parallel boundary formulations within the domain decomposition can induce artificial wave energy accumulation at the subdomain interfaces. To resolve this, this study mirrors the parallel computing infrastructure of the POP2 Message Passing Interface (MPI) protocol. Key wave-module prognostic variables—including the zonal and meridional 10-m winds, wave radiation stress, subsurface momentum, and wave radiation and energy densities—are explicitly integrated across processor

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subdomains to establish a robust, scalable online coupling framework.

2.3 Model Validation

We evaluate the simulated spatial patterns of the air–sea CO_2 flux from both the uncoupled control simulation (B–CTL) and the coupled POP2–waves model against estimates from the NOAA CarbonTracker data assimilation system (CT2022; Jacobson et al., 2023). Unlike our forward, ocean-centric modeling frameworks, CT2022 constraints the system from an atmospheric perspective, top-down inferring net surface CO_2 fluxes by assimilating observed atmospheric CO_2 mole fractions. Consequently, this product captures the integrated signature of both anthropogenic emissions and natural carbon cycle variability to providing continuous global flux estimates spanning January 2000 through December 2020.

Following the inversion framework established by Jacobson et al. (2007), NOAA CT2022 partitions the global ocean into 30 distinct basins to capture large-scale circulation and biogeochemical dynamics. This basin-based approach groups regions characterized coherence in physical processes—such as major currents systems, upwelling zones, and boundary mixing layers—as well as shared biogeochemical behaviors, including air–sea CO_2 exchange rates and biological productivity. Moreover, because numerous observational datasets and ocean carbon products are resolved at regional scales, a basin-scale aggregation ensures greater methodological consistency. We evaluate the seasonal variability and mean state of global air–sea CO_2 fluxes, specifically excluding regions characterized by seasonal reversals in flux sign—a feature that can introduce substantial artifacts during model–data comparisons. Instead, our analysis focuses primarily on regions exhibiting robust, clear oceanic CO_2 flux signals. Recognizing that inherent uncertainties in atmospheric inversions stem from sub-grid scale anthropogenic emissions and localized sea-ice melt,; we follow the

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protocols of Fay et al. (2024) and exclude both the coastal ocean and high-latitude marginal ice zones from the model-data evaluation.

Based on the spatial patterns of oceanic CO_2 outgassing, uptake, and low-flux regimes characterized in the Pacific (Fig. 2c), this study delineates 12 key regions exhibiting high air–sea CO_2 flux variability as identified by NOAA CT2022 (Fig. 2d). These are selected from the 30 ocean basins defined in the CT2022 model documentation (Jacobson et al., 2023) for optimizing flux inversions. These oceanic domains are categorized into three major basins: (1) the Pacific Ocean, (2) the Indian Ocean, and (3) the Atlantic Ocean (Table 1). Within the Pacific basin, underlined labels denote the regions exhibiting the most pronounced oceanic CO_2 outgassing (EEP) and uptake (NEP).

3. Results

3.1 The climatological mean and seasonal variations of air–sea CO_2 flux

We utilize the traditional air–sea CO_2 flux bulk formula (Eq. 1) in CESM1.2.2 while also incorporating wave and bubble-mediated effects via a gas transfer velocity parameterized by u_*, SST, and H_s (Eq. 3). A comparison of the climatological means and monthly standard deviations highlights that the spatial distribution of the model-simulated air–sea CO_2 fluxes is broadly consistent with NOAA CT2022, with regional differences generally remaining below 0.5 mol m^{-2} yr^{-1} (Figs. 2a–f). Notably, the Pacific CO_2 outgassing and uptake regions simulated by the POP2–waves experiment show smaller range-normalized relative magnitudes of model–data biases (Table 3) relative to NOAA CT2022 than the B–CTL baseline, despite a higher monthly SD in POP2–waves over both the Pacific and Indian Oceans.

To clarify our shortlisting criteria, the 12 regions were categorized based on two quantitative features:

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1. Seasonal phasing consistency: Whether the simulated seasonal cycles (temporal trends) align with the NOAA CT2022 atmospheric inversion framework.
2. Relative magnitudes of model–data biases: The monthly averaged deviation as a percentage of the overall range (95% minus 5%) relative to the NOAACT2022 baseline.

The nine regions in Fig. 3 exhibit both highly coherent seasonal phasing and relatively small monthly model–data biases. In contrast, the three regions in Fig. 4 display prominent structural mismatches, such as inverted seasonal trends. Importantly, these temporal phase misalignments directly amplify the calculated monthly model–data biases, as the model and data fluctuate in opposing directions throughout the annual cycle. This phase mismatch ultimately leads to substantially larger relative monthly mean biases; specifically, the model–data biases averaged across the three regions in Fig. 4 (32.9 %) are significantly larger than those in the nine regions of Fig. 3 (27.5% in B–CTL and 23.0% in POP2–waves, respectively).

In terms of the regional mean values of air–sea CO_2 flux, the POP2–waves demonstrate closer structural agreement with NOAA CT2022 compared to the uncoupled B–CTL baseline across eight key regions (excluding the Northwest Pacific (NWP) in Fig. 3). Nevertheless, despite this larger mean offset in specific areas like the NWP, the POP2–waves successfully captured the autumn transition of oceanic CO_2 from a sink to a source as depicted in NOAA CT2022 — a feature that remains unsimulated in B–CTL. Table 3. presents the range-normalized relative magnitudes of model–data biases across nine selected regions, providing a robust metric to evaluate the simulated seawater carbonate-pH and flux field performance against the NOAA CT2022 atmospheric inversion baseline. Overall, the wave-coupled framework (POP2–waves) demonstrates a pervasive and substantial reduction in systematic biases compared to the uncoupled control simulation (B–CTL). Specifically, the regional mean absolute deviation dropped noticeably in major ocean basins, with the most

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pronounced improvement captured in the Equatorial Eastern Pacific (EEP), where the relative bias was drastically attenuated by 25.0 % (from 71.5 % in B–CTL down to 46.5 % in POP2–waves. Notable bias reductions are also observed in the Northwest Pacific (NWP, down 4.9 %), South Indian Ocean Region 1 (SIO1, down 4.1%), and South Atlantic Ocean (SAO, down 4.4 %, confirming that incorporating wave-effect mechanisms successfully rectifies over- or under-estimations in air–sea carbon exchange dynamics. Conversely, the uncoupled baseline (B–CTL) slightly outperforms the coupled model only in the Equatorial Atlantic Ocean (EAO) (11.9 % vs. 15.0 %), suggesting minor localized over-compensations in wave parameterizations within this sub-basin. Nonetheless, the consistent narrowing of relative biases across the vast majority of regions firmly underscores the superior performance and statistical robustness of the online-coupled framework.

The POP2–waves simulation generally exhibits higher a SD than B–CTL, reflecting an amplified seasonal variability (Figs. 3–4). Although these SD values typically remain below 3 mol m^{-2} yr^{-1}, they peak during boreal winter, a phenomenon driven by intense oceanic CO_2 uptake in high-latitude regions of the Northern Hemisphere, such as the Northwestern and Northeastern Pacific, and North Atlantic Ocean 1. In contrast, the Southern Hemisphere exhibits pronounced monthly SD alongside robust oceanic CO_2 uptake during the austral winter (boreal summer), particularly across regions such as the South Pacific Ocean 1 (SPO1), South Indian Ocean 1 (SIO1), and South Atlantic Ocean (SAO). The monthly SD derived from NOAA CT2022 exhibits substantial regional heterogeneity. Notably, variability within the Northwest Pacific (NWP), Equatorial Eastern Pacific (EEP), and North Atlantic Ocean 1 (NAO1) exceeds that simulated by both the coupled POP2–waves model and the uncoupled control B–CTL framework.

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Three specific sub-basins—the South Pacific Ocean Region 2 (SPO2), South Indian Ocean Region 2 (SIO2), and North Atlantic Ocean Region 2 (NAO2)—exhibit prominent model-data discrepancies when evaluated against NOAA CT2022 (Fig. 4). Notably, these systematic deviations are not attributable to wave-mediated processes or the parameterization of the CO_2 flux bulk formula. Within the NAO2, both B–CTL and POP2–waves models simulate a strong seasonal variations that mirrors the NAO1, characterized by robust oceanic CO_2 uptake during the boreal winter; conversely, NOAA CT2022 displays the opposite seasonal trajectory in this sub-basin. A structurally similar mismatch is observed in the mid-latitude South Pacific (SPO2), where the simulated seasonal cycles of both models exhibit strong synchronization with those in SPO1 but contrast sharply with the inverse trend captured by the NOAA CT2022 atmospheric inversion framework.

3.2 The relationship between U_{10} and $k_{w,660}$ in two regions with contrasting wind speed regimes

To demonstrate that the sea state–dependent gas transfer velocity (Deike and Melville, 2018) provides a more suitable estimate of air–sea CO_2 flux than the wind-only formulation (Wanninkhof, 1992) under high-wind conditions ($U_{10} > 10$ m s^{-1}), we selected the Western Pacific (WP) and the Equatorial Pacific (EP) regions due to their contrasting wind regimes. The WP region experiences a high frequency of high-wind conditions (exceeding 30%), whereas wind speeds in the EP region consistently remain below this threshold. Because both regions exhibit stable and significant oceanic CO_2 outgassing or uptake, they enable a clear comparative analysis of the correlation between $k_{w,660}$ and U_{10} (Figs. 5a, b).

Generally, the B–CTL data shows higher R^2 but lower $k_{w,660}$ values compared to POP2–waves $k_{w,660}$ across both the WP and EP. The POP2–waves experiment exhibits

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a slightly lower R^2 than B–CTL due to greater deviations between $k_{w,660}$ and U_{10}, particularly at higher surface wind speeds. This pattern is consistent with the findings of Gutiérrez-Loza et al. (2022) under conditions where H_s > 1.5 m. The relationship between $k_{w,660}$ and U_{10} was evaluated under two scenarios: baseline monthly climatological means (H_s < 1.5m) and high-resolution daily data filtered for H_s > 1.5m (enhanced conditions) aggregated into 30-year monthly averages. Notably, both approaches substantially diminish the differences in $k_{w,660}$ at identical U_{10} values.

However, short-term (30 min) observations indicate that gas transfer velocity can be up to twice as high under similar wind speeds when H_s > 1.5 m (Gutiérrez-Loza et al., 2022. Divergence between the models becomes significant when U_{10} exceed 12 m s^{-1} over the WP region (Fig. 5a); whereas over the EP region, the convergence of $k_{w,660}$ estimates is most notable around U_{10} = 9 m s^{-1} (Fig. 5b), suggesting that mid-range wind speeds dominate the EP flux signal.

As shown in Fig. 5c, the enhanced WP cases (H_s > 1.5 m) have data available for all months except August and October, during which their $k_{w,660}$ values consistently exceed both the all-data POP2–waves and B–CTL averages. In contrast, the EP region exhibits H_s > 1.5 m data only during the summer months (July to October), where the corresponding $k_{w,660}$ values also exceed both the all-data POP2–waves and B–CTLaverage. The monthly SD indicates no significant differences between the enhanced (H_s > 1.5 m) data and the standard POP2–waves simulations over the WP, with the $k_{w,660}$ discrepancies between the two datasets remaining minimal from January to May because the average H_s during this period consistently remains below 1.5 m. However, over the EP, a pronounced discrepancy is observed between the H_s > 1.5 m condition and the all-data POP2–waves average from July to September.

3.3 Regression of CO_2 flux against U_{10} under high-wind conditions

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The wind-only parameterization of gas transfer velocity tends to underestimate values under high-wind conditions (U_{10} > 10 m s^{-1}) or high significant wave heights (H_s > 1.5 m) (e.g., Gutiérrez-Loza et al., 2022; Zhou et al., 2023). Under these high-wind conditions, the monthly mean air–sea CO_2 fluxes derived from the B–CTL and POP2–waves simulations diverge notably within the Western Pacific region (WP) (Fig. 6), highlighting key discrepancies between the wind-only and wave-modified $k_{w,660}$ formulations. The data were filtered to include only grid points that met this high-wind threshold before being regionally averaged, Because not all months contained grid points satisfying the threshold, a final subset of 154 data points was utilized for the linear regression analysis. These high-wind conditions over the WP region occur primarily in winter (accounting for 42% of the valid data), when the concurrent low sea surface temperatures further enhance CO_2 solubility. The slopes of the regression equations (–0.77 mol m^{-2} yr^{-1} per m s^{-1} for B–CTL and –1.40 mol m^{-2} yr^{-1} per m s^{-1} for POP2–waves) alongside the mean CO_2 fluxes (Y_{avg}: –6.35 mol m^{-2} yr^{-1} for B–CTL and –6.97 mol m^{-2} yr^{-1} for POP2–waves) both indicate that POP2–waves a higher sensitivity to wind speed, leading to stronger oceanic CO_2 uptake across the WP domain under elevated wind speeds.

The correlation coefficient (R) of the POP2–waves simulation is higher than that of B–CTL, indicating that the sea state–dependent $k_{w,660}$ formulation provides a more robust estimate of the CO_2 flux than the wind-only $k_{w,660}$ scheme under high-wind conditions. This finding aligns with the observations of Gutiérrez-Loza et al. (2022) and Zhou et al. (2023). The average air–sea CO_2 flux in B–CTL over the WP region during DJF and JJA is −0.021 and −0.004 mol m^{-2} yr^{-1}, respectively. Meanwhile, POP2–waves exhibits only slight discrepancies relative to B–CTL during these respective seasons, with differences of more than 0.001 mol m^{-2} yr^{-1} and less than 0.002 mol m^{-2} yr^{-1} in CO_2 uptake (data not shown). This contrast

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3.4 Analysis of surface fields and $k_{w,660}$ component in POP2–waves and B–CTL

The spatial co-variations of sea level pressure (PSL), U_{10}, and H_s underscore their combined role as key meteorological and wave drivers steering the regional $k_{w,660}$ patterns (Figs. 7a, b). The region with $H_s > 1.5$ m is generally situated where $U_{10} > 8$ m s^{-1} and lies south of the high PSL region. The spatial patterns of climatological U_{10} and H_s align closely with the findings of Reichl and Deike (2020) (Figs. 7a, b). Furthermore, a dominance of bubble-mediated transfer in the POP2–waves simulation, where the $k_{wB,660}/k_{w,660}$ ratio exceeds 0.5, primarily occurs in regions characterized by $H_s > 1.5$ m (Fig. 7b). However, an elevated H_s does not always correspond to a higher $k_{wB,660}/k_{w,660}$ ratio in the POP2–waves simulation; this decoupling is explicitly observed in the North Pacific (Fig. 7b).

Deike and Melville (2018) demonstrated that the bubble contribution to CO_2 transfer exceeds 40% when the $U_{10} \geq 10$ m s^{-1}. In our POP2–waves simulation, the bubble fraction ($k_{wB,660}/k_{w,660}$) accounts for approximately 38% of total gas transfer velocity, which is slightly higher than ~30% reported by Reichl and Deike (2020). This discrepancy may be attributed to differences in the A_B scaling coefficients (Eq. 3), which is derived from field data, where gas transfer velocity are estimated from eddy covariance flux measurements (Reichl and Deike, 2020; Brumer et al., 2017).

The structural differences in $k_{w,660}$ (POP2–waves − B–CTL) reveal that the most pronounced discrepancies are primarily distributed in regions characterized by elevated H_s, particularly in the Southern and South Indian Oceans (Fig. 7c). Although U_{10} in the Indian Ocean and tropical Pacific is relatively low (<5 m s^{-1}; Fig. 7a), the $k_{w,660}$ values simulated by POP2–waves remain approximately 9 cm hr^{-1} higher than

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those in B–CTL. In these regions, this wave-driven enhancement of the total gas transfer velocity is predominantly governed by the non-bubble-mediated component ($k_{wNB,660}$; Fig. 7d). Although H_s is also elevated in the Southern Ocean (Fig. 7b), it does not contribute substantially to the $k_{w,660}$ difference between POP2–waves and B–CTL; this is because the high U_{10} in this region (Fig. 7a) already yields high gas transfer velocities through the empirical formulation of Wanninkhof (1992). The spatial patterns of both the B–CTL $k_{w,660}$ and the wave-induced discrepancies $k_{w,660}$ (POP2–waves − B–CTL) closely align with the findings of Reichl and Deike (2020) (Fig. 7c). Overall, the two simulations exhibit consistent geographic distributions, yielding 30-year domain-wide mean gas transfer velocities of 17.0 cm hr^{-1} for B–CTL and 22.7 cm hr^{-1} for POP2–waves, despite their reliance on distinct input parameters (U_{10} vs. u_* and H_s). Aside from the polar regions, the total gas transfer velocity in the POP2–waves coupled model exceeds that in B–CTL, with the most pronounced differences localized in the tropics (Fig. 7c). To elucidate the driving mechanisms behind this global enhancement, it is necessary to quantify whether the non-bubble ($k_{wNB,660}$) or bubble-mediated ($k_{wB,660}$) component dominates the total gas transfer velocity across the different ocean basins. These discrepancies reveal a dominance of bubble-mediated transfer ($k_{wB,660} > k_{wNB,660}$) under elevated H_s regimes, contrasting with a shift toward non-bubble dominance ($k_{wB,660} < k_{wNB,660}$) in regions where 10-m wind speeds remain below 7 m s^{-1} (Fig. 7).

3.5 Mean State and model differences in pCO_2^w and surface pH

Integrating wave dynamics induces a pronounced spatial reorganization of both pCO_2^w and ocean surface pH over the 30-year climatological period (Fig. 8). Specifically, the baseline B–CTL simulation shows that high pCO_2^w is concentrated in the equatorial eastern Pacific (EEP), contributing to a larger oceanic CO_2 source (Fig.

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8a). In contrast, low pCO_2^w is found in high-latitude regions of the Pacific and Atlantic (e.g., NWP, NEP, NAO1, and NAO2), driving strong oceanic CO_2 uptake (Fig. 8a). Over the study period, the climatological standard deviation of pCO_2^w in the B–CTL simulation fluctuates between 10 and 30 ppm (Fig. 8a).

In the CESM1 framework of the B_2000_CAM5 compsets, atmospheric pCO_2^a is held constant (367 ppm). The pCO_2^w in the Eastern Pacific exceeds 440 ppm, indicating an air–sea $dpCO_2$ greater than 73 ppm in B–CTL (109.8 and 92.7 ppm during DJF and JJA, respectively), whereas the pCO_2^w in the Western Pacific is below 320 ppm, indicating an air–sea $dpCO_2$ lower than −47 ppm (−53.4 and −31.4 ppm during DJF and JJA, respectively). Notably, POP2–waves reduces the high pCO_2^w by more than 10 ppm over the Eastern Equatorial Pacific and increases the low pCO_2^w in other regions (Fig. 8b). Consequently, in the oceanic CO_2 outgassing (uptake) regions, the POP2–waves coupled model decreases (increases) pCO_2^w, thereby reducing the magnitude of the air–sea $dpCO_2$. The SD of the pCO_2^w difference between POP2–waves and B–CTL varies significantly in Eastern Equatorial Pacific, while SD changes in other regions are less pronounced.

The biogeochemical module of POP2 (MARBL) updates TA in each grid cell by solving a coupled transport–reaction equation (Long et al., 2021). The average pH of the ocean is currently around 8.1 in oceanic CO_2 uptake regions (e.g., the Western Pacific) and 8.0 in outgassing regions (e.g., the Equatorial Pacific, Fig. 8c), reflecting slightly alkaline conditions. Within the DIC system, bicarbonate ions (HCO_3^-) are the dominant species because the prevailing seawater pH lies between pK_{a1} and pK_{a2}. However, continued uptake of atmospheric CO_2 drives a decline in pH—a process termed ocean acidification. Our study highlights that wave-induced $k_{w,660}$ significantly influences the marine CO_2 system, altering alkalinity, pH, and carbonate speciation. These chemical shifts, in turn, provide a feedback mechanism that modulates the overall

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dynamics of the air–sea CO_2 flux. In other words, perturbations in $k_{w,660}$ influence both $dpCO_2$ and air–sea CO_2 flux within the Earth System Model.

Ocean pH and surface ocean pCO_2^w are strongly negatively correlated (Macovei et al. 2021). Consequently, low pH values are observed in the Equatorial Eastern Pacific (Fig. 8c), as a direct result of the elevated DIC and pCO_2^w concentrations characteristic of that region (Fig. 8a). Conversely, higher pH values are sustained in regions exhibiting lower pCO_2^w levels, a feature predominantly found across the high-latitude domains of both the Pacific and Atlantic Oceans. Rustogi et al. (2025) indicate that accelerated equilibration of oceanic pCO_2 in the wind–wave–bubble simulation, driven by enhanced gas exchange, ultimately reduces the magnitude of the air–sea $dpCO_2$. This pCO_2 feedback is strongest in the extratropics, where it suppresses CO_2 uptake. Our POP2–waves experiment reveals a consistent response, characterized by increasing in both oceanic pCO_2 (Fig. 8b) and hydrogen ion concentration, which are accompanied by a corresponding decline in pH (Fig. 8d) across the extratropics. Accounting for wave effects results in simulated pH shifts of approximately + (-) 0.01 in oceanic CO_2 outgassing (uptake) regions (Fig. 8d). This suggests that incorporating this mechanism alleviates acidification in the equatorial Pacific, whereas it intensifies ocean acidification in the high-latitude domains of both the Pacific and Atlantic Oceans (Fig. 8d). Rustogi et al. (2025) also showed that the effects of Δk_w (the difference between wave-induced and based on U_{10}) and $\Delta dpCO_2$ partially offset each other. However, because the Δk_w effect is generally 20%–30% stronger, it dominates the counteracting $\Delta dpCO_2$ effect, explaining the modest increase observed in both oceanic outgassing and uptake CO_2 regions within the wave-effect simulation.

4. Discussion

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4.1 Impact of wave-dependent k_w on global air–sea CO_2 flux: a comparison between POP2–waves and standard Earth System Models

Several investigations have parameterized the air–sea CO_2 flux using Eq. (1), yet current $k_{w,660}$ parameterizations within prominent global frameworks still rely exclusively on the neutral 10-m wind speed (U_{10}) as seen in models such as MPI-ESM1.2 (Mauritsen et al., 2019), CESM2 (Danabasoglu et al., 2020), NorESM2 (Seland et al., 2020), UKESM1 (Ziehn et al., 2020), MIROC6 (Chikamoto and DiNezio, 2021), CMCC-ESM2 (Lovato et al., 2022), and CanESM5 (Sigmond et al., 2023) (Table 2). Concurrently, pioneering efforts have begun quantifying wave-induced effects on both air–sea CO_2 flux and long-term ocean carbon storage by incorporating coupled wind–wave–bubble gas transfer formulations into the Ocean general circulation models (OGCMs), such as MOM6–COBALTv2 (Rustogi et al., 2025) and NEMO–PISCES (Wu et al., 2025). Notably, Wu et al. (2025) underscore that integrating these wave-mediated boundary layer dynamics into comprehensive Earth System Models is essential for reducing systemic uncertainties in global carbon cycle projections.

Several studies have incorporated wave effects following the parameterization of Deike and Melville (2018), utilizing the Surface Ocean CO_2 Atlas (SOCAT) database (Bakker et al., 2016) to calculate diagnostic air–sea CO_2 fluxes. In these offline configurations, however, the computed air–sea CO_2 flux is treated independently of pCO_2^w dynamics and surface pH evolution, lacking any interactive feedback mechanisms between the atmosphere and the upper-ocean carbonate system.

Rustogi et al. (2025) utilized an ocean-biogeochemistry system forced by offline atmospheric reanalysis and wave model outputs. While comprehensive, such an offline-forced setup is limited in its ability to capture high-frequency, episodic, and synchronous interactions between physical wave states and surface water chemistry

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during rapid weather transitions. In contrast, our study implements an online coupling framework. The key value-add of our setup is its capacity to simulate step-by-step, interactive feedbacks where wave properties dynamically alter the gas transfer velocity ($k_{w,660}$) and physical mixing in real-time. The coupled behavior of POP2–waves includes $dpCO_2$-driven negative feedbacks, which fundamentally differs from traditional obs-based products, that treat wave-induced $k_{w,660}$ and $dpCO_2$ as independent variables when estimating air–sea CO_2 flux (e.g., Reichl and Deike, 2020). This online coupling mechanism also contrasts with global ocean model forced offline by atmospheric reanalysis and wave model outputs (e.g., Rustogi et al., 2025), where such interactive feedbacks are often omitted. This allows our model to capture non-linear, high-resolution impacts on the air–sea CO_2 flux during highly dynamic conditions, which are often smoothed out in offline-forced simulations.

4.2 Impact of air–sea $dpCO_2$, $k_{w,660}$, SST, and pH on air–sea CO_2 flux

To evaluate which driver exerts the most direct control on air–sea CO_2 flux, Fig. 9 displays the spatial distributions of the linear regression coefficients (LRCs) between the flux and various standardized variables: air–sea $dpCO_2$ (Fig. 9a), $k_{w,660}$ (Fig. 9b), SST (Fig. 9c), and pH (Fig. 9d). To remove the confounding artifacts of differing physical units and enable a direct comparison on a common scale, both the air–sea CO_2 flux and the independent variables were standardized into anomalies (with a mean of 0 and a standard deviation of 1) prior to the regression analysis. The statistical significance of these regressed relationships was subsequently evaluated using a Student's t-test at the 95% confidence level. This standardization constraints the LRCs to range strictly between −1 and +1. Here, we do not delve into the intricate chemical and biochemical feedback mechanisms governing carbonate species. Nevertheless, it is worth noting that the collective influence of carbonate

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composition—specifically TA, and dissolved inorganic carbon (DIC)—on pCO_2^w variability has been shown to outweigh that of solubility changes driven by sea surface salinity and SST (e.g., Koseki et al., 2023).

Among these drivers, air–sea $dpCO_2$ and air–sea CO_2 flux exhibit the strongest positive LRCs, with coefficients ranging from 0.6 to 0.8 across the global ocean; this underscores the role of the ocean acting as net CO_2 source or sink depending on the sign of the air–sea $dpCO_2$ gradient. Additionally, wave effects increase the LRCs by approximately 0.1 to 0.2, particularly within regions of intense oceanic CO_2 outgassing or uptake. A clear positive (negative) LRCs exists between $k_{w,660}$ and the air–sea CO_2 flux in oceanic CO_2 outgassing (uptake) regions. Furthermore, wave effects reduce the absolute LRCs between $k_{w,660}$ and air–sea CO_2 flux in both oceanic CO_2 outgassing (uptake) regions (Fig. 9b). This indicates that once interactions between the modeled CO_2 flux and the ocean carbonate-pH system are considered, the wave-influenced CO_2 flux becomes less correlated with $k_{w,660}$ and more closely linked to air–sea $dpCO_2$. This shift can be attributed to the limitations of wind-only parameterizations; as indicated by Zhou et al. (2023), wind-only formulas tend to underestimate gas transfer velocities when U_{10} exceeds 10 m s^{-1}, whereas enhanced gas transfer velocity frequently occurs under conditions of strong wave breaking and high significant wave height. Johansson et al. (2022) reported that H_s generally increases with wind speed, but the relationship is strongly modulated by wind direction and regional conditions, leading to spatially variable and nonlinear wind–wave coupling.

In contrast to the former, there is a clear positive (negative) LRCs between SST and CO_2 flux in oceanic CO_2 uptake (outgassing) regions (Fig. 9c). A negative correlation exists between $k_{w,660}$ and SST, reflecting the latitudinal gradient where colder, high-latitude waters are characterized by more intense sea states and elevated

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$k_{w,660}$ values. This pattern occurs because lower temperatures at high latitudes increase CO_2 solubility (Séférian et al., 2020), thereby enhancing oceanic uptake. As SST increases, the kinetic energy of CO_2 molecules in seawater rises, enhancing their diffusion and consequently reducing the Schmidt number (Sc = ν/D, defined as the ratio of kinematic viscosity to the molecular diffusion coefficient). Because $k_{w,660}$ is parameterized as a function of $Sc^{-0.5}$, a decrease in Sc leads to an increase in both $k_{w,660}$ and net air–sea CO_2 flux. Fay et al. (2024) attributed the discrepancies and variations in air–sea CO_2 fluxes to a combination of factors, including choices in wind speed products, SST datasets, biological carbon uptake, and the parameterization of gas transfer velocity. Wave effects only slightly reduce the LRCs between SST and the air–sea CO_2 flux—by approximately 0.1—within the tropics, with negligible impacts observed in other regions. The strong negative correlation between ocean pH and pCO_2^w is dictated by the acid dissociation equilibria of the marine carbonate system (Figs. 8a, c), which govern the fundamental relationship between dissolved CO_2 and hydrogen ion concentration (Williams et al., 2017). As seawater pH decreases, the resulting increase in hydrogen ion ([H^+]) concentration drives a reaction with carbonate ions (CO_3^{2-}). This consumption of carbonate ions ultimately reduces the ocean's buffering capacity for CO_2 uptake. Consequently, pCO_2^w increases and $dpCO_2$ decreases, suppressing the air–sea CO_2 flux into the ocean. Reflecting this mechanism, pH and air–sea CO_2 flux exhibit strong negative LRCs (less than –0.9) across most of the global ocean, with exceptions limited to the Eastern Equatorial Pacific and along the Arctic margins (Fig. 9d). Wave effects marginally attenuate this relationship in the Eastern Equatorial Pacific (EEP), reducing the LRCs by approximately 0.1.

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4.3 Uncertainty arising from the absence of interactions between air–sea CO_2 flux and the ocean carbonate–pH system

Rustogi et al. (2025) indicate that as surface seawater pCO_2^w responds to changes in DIC and alkalinity, the air–sea $dpCO_2$ gradient is partially attenuated. This induces a negative feedback loop whereby enhanced CO_2 uptake (or outgassing) is systematically damped by chemical re-equilibration within the marine carbonate system, a mechanism that is highly consistent with the findings of this study. To assess the uncertainty in CO_2 flux resulting from the absence of interactions between the flux and the ocean carbonate–pH system (i.e., the decoupling of the $dpCO_2$-driven negative feedback). The air–sea CO_2 flux was reconstructed using the uncoupled $dpCO_2$ fields from the control simulation (B–CTL) and alongside the wave-influenced $k_{w,660}$ parameterization from the coupled wave simulation (POP2–waves). This experimental setup explicitly isolates the statistical mean and standard deviation of the flux under a scenario defined by the "lack of $dpCO_2$-driven negative feedback". Only data significant at a 95% confidence level of air–sea CO_2 flux difference between "lack of $dpCO_2$-driven negative feedback" and POP2–waves are shown in Fig. 10b. Compared to the baseline (Fig. 2b), the CO_2 flux in the "lack of $dpCO_2$-driven negative feedback" scenario is stronger than that in the POP2–waves simulation across both oceanic CO_2 outgassing and uptake regions, accompanied by a larger SD. The most pronounced discrepancies occur in the Pacific basin across both outgassing and uptake zones. However, certain mid-to-high latitude subregions—specifically the Northeastern Pacific (NEP), South Indian Ocean 2 (SIO2), and South Pacific Ocean 2 (SPO2)—do not reach the 95% confidence threshold.

Rustogi et al. (2025) demonstrate that while wave-enhanced $k_{w,660}$ accelerates local air–sea CO_2 exchange, it simultaneously attenuates the air–sea $dpCO_2$ gradient. In this study, we conceptualize this process as a manifestation of the ocean's buffering

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capacity, which encompasses the dynamic coupling between CO_2 flux (estimated via $k_{w,660}$) and the marine carbonate–pH system. Within the coupled POP2–waves framework, incorporating wave effects alleviates surface acidification and reduces air–sea $dpCO_2$ over oceanic CO_2 outgassing regions; conversely, the resulting relatively higher pH levels systematically enhance the ocean's capacity to absorb atmospheric CO_2 within regions characterized by oceanic CO_2 uptake region in the coupled POP2–waves model. In contrast, the simulation lacking the $dpCO_2$-driven negative feedback—artificially omits realistic marine buffering effects—overestimates the air–sea exchange, yielding excess CO_2 outgassing into the atmosphere (>0.4 mol m^{-2} yr^{-1}) over source regions. Concurrently, it amplifies excess CO_2 uptake (<−0.4 mol m^{-2} yr^{-1}) within regions characterized by oceanic CO_2 uptake regions (Fig. 10b) compared to the coupled POP2–waves model.

5 Conclusions

The study advances Earth System Model by developing an online coupling framework (POP2–waves) that incorporates wave effects directly into air–sea CO_2 flux simulations, effectively capturing the $dpCO_2$-driven negative feedback in the marine carbonate-pH system. Neglecting this coupling by treating $dpCO_2$ and wave-induced $k_{w,660}$ independently leads to a substantial overestimation of global fluxes. Fully integrating these wave effects into ocean biogeochemical models addresses three grand challenges in climate modeling:

1. Technical implementation: Resolving coordinate interpolation, boundary discontinuities in wave energy, and MPI parallelization.
2. Interdisciplinary analysis: Harmonizing kinematic, thermodynamic, and biogeochemical frameworks across distinct Earth system components.

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3. Validation constraints: Overcoming sparse observational data by synthesizing indirect flux estimates from atmospheric inversions (e.g., NOAA CT2022), ships, and buoys.

Within the coupled POP2–waves framework, the gas transfer velocity is resolved using the parameterization of Deike and Melville (2018), which attributes bubble-mediated transfer to the combined effects of friction velocity and significant wave height. This setup extends the framework of Wu et al. (2025) by utilizing a dynamically coupled ocean–wave system. This advanced coupling explicitly captures how flux-driven alterations in surface $dpCO_2$ feedback nonlinearly via the interactive interface to modulate the global air–sea CO_2 flux itself. Our regional evaluation reveals a mixed and basin-dependent performance when introducing wave effects. Across the majority of the 12 key regions of high variability defined by NOAA CT2022, the POP2–waves simulation shows a closer structural agreement with the inversion data compared to the uncoupled B–CTL baseline. However, this improvement is not universal; notable discrepancies persist within specific sectors, including the South Pacific Ocean 2, South Indian Ocean 2, and North Atlantic Ocean 2, where the wave-coupled model does not clearly outperform the baseline.

Significant deviations between POP2–waves and B–CTL simulations emerge when significant wave height exceeds 1.5 m. Consistent with Gutiérrez-Loza et al. (2022), this wave-induced divergence results in a systematically air–sea CO_2 flux under high 10-m wind speed and significant wave height. Across the entire spatiotemporal domain, the bubble-mediated contribution to the total gas transfer velocity ($k_{wB,660}/k_{w,660}$) in POP2–waves averages approximately 38%. This finding slightly exceed the ~30% baseline reported by Reichl and Deike (2020). These high $k_{wB,660}/k_{w,660}$ ratios align with regions of elevate 10-m wind speed and significant wave height. Concurrently, our results indicate that bubbles-mediated processes account for up to 41.3% of the total

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air–sea CO_2 flux, which is consistent with the ~40% contribution reported by both Reichl and Deike (2020) and Zhou et al. (2023).

Within oceanic CO_2 outgassing regions, POP2–waves attenuates the high air–sea $dpCO_2$ gradient, whereas it enhances low gradients elsewhere. This feedback drives surface pH shifts of approximately ±0.01 that spatially align with the sign of regional flux. On a global scale, the air–sea $dpCO_2$ (pH) displays the strongest positive (negative) regression coefficients with the flux. Additionally, while gas transfer velocity scales positively with the absolute air–sea CO_2 flux magnitude, whereas SST demonstrates an inverse relationship to this gas transfer velocity sensitivity.

Compared to the B–CTL baseline, POP2–waves simulations show that oceanic CO_2 uptake and outgassing regions expand by 11.8% and 41.6%, respectively, resulting in a 1.8% increase in the global ocean CO_2 sink. Consequently, while wave-induced enhancements in $k_{w,660}$ boost instantaneous air–sea CO_2 flux, the coupled carbonate–pH system limits the net long-term impact via this this $dpCO_2$-driven feedback (Rustogi et al., 2025). This feedback decouples local increases in CO_2 flux from proportional proportional changes in the global mean ocean carbon sink.

The impacts of the $k_{w,660}$ bulk formula (Wanninkhof et al., 1992) and wave dynamics (Deike and Melville, 2018) on air–sea CO_2 flux, surface pH, and air–sea $dpCO_2$ are schematically summarized in Fig. 11. The B–CTL baseline captures a pronounced seasonal contrast in Western Pacific air–sea CO_2 fluxes, characterized by strong uptake during DJF that is heavily suppressed in JJA due to SST-driven reductions in solubility. This seasonal variability in air–sea $dpCO_2$ activates a negative feedback loop with the POP2–waves framework , which ultimately modulate the overall air–sea CO_2 flux. Conversely, the Equatorial Pacific region exhibits lower pH than the Western Pacific and acts as a persistent CO_2 source across both seasons, showing no significant, with no obvious seasonal variation, a finding consistent with Fay et al. (2024).

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While the current POP2–waves framework demonstrates the critical importance of online coupling and $dpCO_2$-driven feedbacks, several avenues remain for future model refinement. A key priority is the continuous improvement of gas transfer velocity formulations. Future efforts will focus on transitioning to the new generalized formulation proposed by Deike et al. (2025), which incorporates updated coefficients and accounts for an asymmetric bubble flux contribution. While this advancement is expected to have a minor impact on oceanic CO_2 flux estimations, it holds potential significance for constraining global marine O_2 fluxes. Furthermore, moving toward a fully coupled Earth System Model (ESM)—which integrates dynamic atmospheric feedback alongside ocean and wave components—will be essential to fully unravelling the long-term impacts of wave-induced processes on global marine biogeochemical cycles.

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Code and data availability. The model code of POP2–waves coupled model is available at https://doi.org/10.5281/zenodo.15795234 (Lan, 2025). Input data of POP2–waves using the climatological Hadley Centre Sea Ice and Sea Surface Temperature dataset, including 30-year numerical experiments, are available at https://doi.org/10.5281/zenodo.5510795 (Lan et al., 2021). CarbonTracker CT2022 data are provided by the National Oceanic and Atmospheric Administration (NOAA) and available from Global Monitoring Laboratory https://gml.noaa.gov/aftp/products/carbontracker/co2/CT2022/ (Jacobson et al., 2023).

Author contributions. YYL is the sole developer of the POP2–waves coupled model and writes the majority part of the paper. HHH provides computational support and analysis suggestions. WLL supports reorganization and offers analytical recommendations and SC offers a non-parallel wave module as part of the POM source code.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Characteristics of the 12 key regions exhibiting prominent air–sea CO_2 flux variability across the three major ocean basins.

Basins	Key regions
the Pacific Ocean	the Northwestern Pacific (NWP), Eastern Equatorial Pacific (EEP), Northeastern Pacific (NEP), South Pacific Ocean 1 (SPO1), and South Pacific Ocean 2 (SPO2)
the Indian Ocean	the North Indian Ocean (NIO), South Indian Ocean 1 (SIO1), and South Indian Ocean 2 (SIO2)
the Atlantic Ocean	the North Atlantic Ocean 1 (NAO1), North Atlantic Ocean 2 (NAO2), Equatorial Atlantic Ocean (EAO), and South Atlantic Ocean (SAO)

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Table 2. Intercomparison of Earth System Model for estimating air–sea CO_2 flux

Earth System Model	Ocean components	Wave-informed air–sea CO_2 flux	$k_{w,660}$ parameterization	Reference
CESM1[1]	POP2[2]	no	U_{10} (Wannikhof 1992)	Chikamoto and DiNezio, 2021; Chikamoto et al., 2023
CESM2[3]	POP2[2]	no	U_{10} (Wannikhof 2014)	Danabasoglu et al., 2020
NorESM2[4]	BLOM[5]	no	U_{10} (Wannikhof 2014)	Seland et al., 2020; Tjiputra et al., 2020
MPI-ESM1.2[6]	MPIOM 1.6[7]	no	U_{10} (Wannikhof 2014)	Mauritsen et al., 2019; Liu et al., 2021
ACCESS-ESM1.5[8]	MOM5[9]	no	U_{10} (Wannikhof 1992)	Ziehn et al., 2020
CMCC-ESM2[10]	NEMO v3.6[11]	no	U_{10} (Wannikhof 1992)	Lovato et al., 2022
CanESM5[12]	CanNEMO[13]	no	U_{10} (Wannikhof 1992)	Sigmond et al., 2023
GFDL-ESM4.1[14]	MOM6[15]	no	U_{10} (Wannikhof 2014)	Stock et al., 2020; Roobaert et al., 2024
IPSL-CM6A-LR[16]	NEMO-PISCES[17]	no	U_{10} (Wannikhof 1992)	Boucher et al., 2020
MIROC-ES2L[18]	COCO 4.0[19]	no	U_{10} (Wannikhof 1992)	Hajima et al., 2020
UKESM1[20]	NEMO v3.6[11]	no	U_{10} (Wannikhof 1992)	Yool et al., 2021
CNRM-ESM2-1[21]	NEMO-PISCES[17]	no	U_{10} (Wannikhof 1992)	Séférian et al., 2019
-[22]	NEMO-PISCES[17]	Yes	H_s, u^* (Deike and Melville, 2018)	Wu et al. 2025
-[22]	MOM6-COBALTv2[23]	Yes	H_s, u^* (Deike and Melville, 2018)	Rustogi et al. 2025
CESM1-POP2–waves[24]	POP2–waves coupled model	Yes	H_s, u^* (Deike and Melville, 2018)	This study

Note:

[1]CESM1: the Community Earth System Model version 1 of the National Center for Atmospheric Research (NCAR);

[2]POP2: the Parallel Ocean Program version 2 of NCAR;

[3]CESM2: CESM version 2;

[4]NorESM2: The Norwegian Earth System Model version 2;

[5]BLOM: Bergen Layered Ocean Model;

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[6]MPI-ESM1.2: the Max Planck Institute for Meteorology Earth System Model version 1.2;

[7]MPIOM 1.6: the Max-Planck Institute Ocean Model version 1.6;

[8]ACCESS-ESM1.5: the Australian Community Climate and Earth System Simulator form an Earth System Model version 1.5;

[9]MOM5: the GFDL Modular Ocean Model version 5;

[10]CMCC-ESM2: the Euro-Mediterranean Centre on Climate Change (CMCC) Earth System Model version 2;

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[11]NEMO v3.6: Nucleus for European Modelling of the Ocean version 3.6;

[12]CanESM5: the Canadian Earth System Model version 5;

[13]CanNEMO: NEMO version 3.4 modified for CanESM;

[14]GFDL-ESM4.1: the Geophysical Fluid Dynamics Laboratory's Earth System Model 4.1;

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[15]MOM6: the GFDL Modular Ocean Model version 6;

[16]IPSL-CM6A-LR : version 6 of the Institut Pierre-Simon Laplace (IPSL) climate model;

[17]NEMO-PISCES: Nucleus for European Modelling of the Ocean, Pelagic Interaction Scheme for Carbon and Ecosystem Studies ocean general circulation and biogeochemistry model;

[18]MIROC-ES2L: the Model for Interdisciplinary Research on Climate, Earth System version 2;

[19]COCO 4.0: CCSR (Center for Climate System Research) Ocean Component Model version 4.0:

[20]UKESM1: the U.K. Earth System Model;

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[21]CNRM-ESM2-1: the Earth system (ES) model of second generation developed by the Centre National de Recherches Météorologiques (CNRM);

[22]The simulations were conducted using global ocean-only models rather than full Earth System Models.

[23]MOM6-COBALTv2: Geophysical Fluid Dynamics Laboratory global ocean model (Modular Ocean Model, MOM6) coupled with sea ice and biogeochemistry (Carbon, Ocean Biogeochemistry and Lower Trophics, COBALTv2);

[24]CESM1-POP2–waves: POP2–waves coupled model based on CESM1.

Table 3. Relative magnitudes of model–data biases (%), defined as the monthly mean absolute deviation normalized by the model's corresponding monthly variability range (95th minus 5th percentile)

Model	NWP	EEP	NEP	SPO1	NIO	SIO1	NAO1	EAO	SA[illegible]
POP2–waves	21.9	46.5	24.7	21.1	14.5	17.8	14.3	15.0	31.[illegible]
B–CTL (baseline)	26.8	71.5	25.3	22.1	16.2	21.9	15.4	11.9	36.[illegible]

Note:

Abbreviations: NWP: Northwestern Pacific; EEP: Eastern Equatorial Pacific; NEP: Northeastern Pacific; SPO1: South Pacific Ocean 1; NIO: North Indian Ocean; SIO1: South Indian Ocean 1; NAO1: North Atlantic Ocean 1; EAO: Equatorial Atlantic Ocean; SAO: South Atlantic Ocean.

The relative magnitude of model–data bias (%) across the 360-month period is defined and calculated as follows:

$$\text{Model} - \text{data biases } (\%) = \frac{1}{12}\sum_{i=1}^{12}\frac{|Mi - Oi|}{P_{95}(M) - P_5(M)}$$

where *Mi* and *Oi* represent the monthly mean values simulated by (POP2–waves or B–CTL) and reference (NOAA CT2022 inversion) air–sea CO_2 fluxes for the month mean of month i (i = 1, 2, …, 12), respectively. P_{95} (*M*) and P_5 (*M*) denote the 95th and 5th percentiles, respectively, of each calendar month's values over the 30-year period, representing the corresponding range of monthly variability in the model.

已刪除: Table 3. Integrated monthly mean air–sea CO₂ flux biases (mol m⁻² yr⁻¹) between the model simulations and NOAA CT2022, based on the Fig. 3. Model

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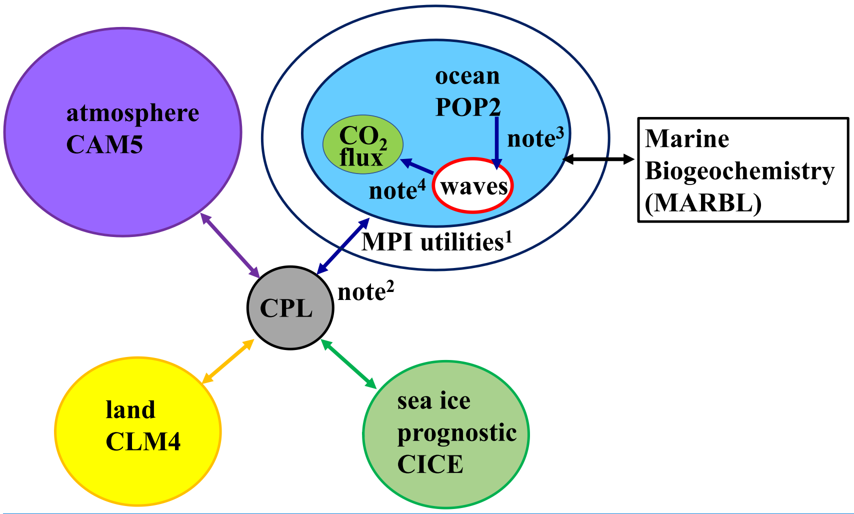


Figure 1. Architecture diagram for POP2–waves experiment in CESM1.2.2 framework, all components adhere to the CESM 1.2.2 framework, except for certain parts of the ocean component. The model components including components for the atmosphere [Community Atmosphere Model version 5 (CAM5)], land [Community Land Model version 4 (CLM4)], ocean [Parallel Ocean Program, version 2 (POP2)], along with its associated modules—the waves module (waves) and the Marine Biogeochemistry (MARBL) module—sea ice [prognostic Los Alamos Sea Ice Model (CICE)], and the coupler (CPL).

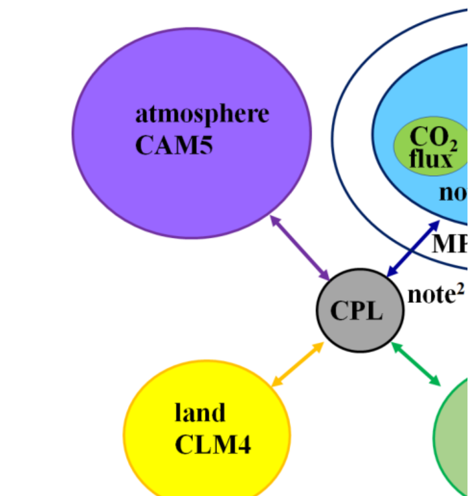
Note:

[1]The wave-module variables are communicated between neighboring blocks via MPI, including zonal and meridional 10-m winds, wave radiation stress, subsurface momentum, and wave radiation and energy densities.

[2]To transfer variables such as zonal and meridional 10-m winds and friction velocity (including sea surface and ice fraction) from the CPL to POP2.

[3]The wave module receives variables from POP2, including the interpolation of depth-varying currents from z-coordinates (60 levels) to the wave module's vertical sigma coordinates (21 levels), along with ocean grid information in the x, y, and z directions.

[4]The wave module outputs significant wave height and friction velocity from the CPL for calculating the bubble-mediated gas transfer velocity in Eq. (3).



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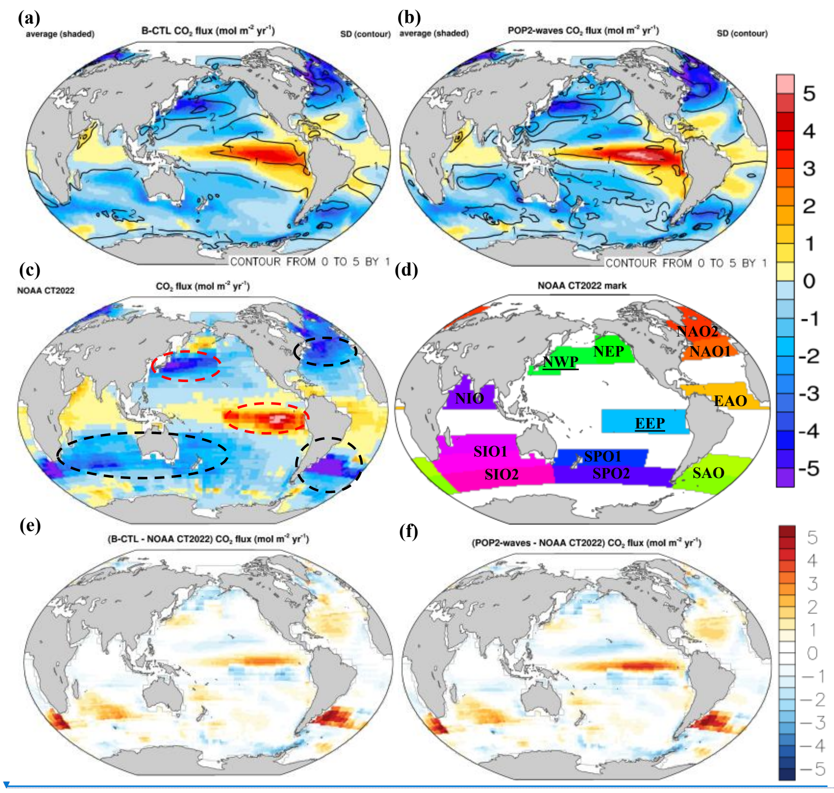

Figure 2. Climatological mean air–sea CO_2 flux (mol m^{-2} yr^{-1}; shaded) and monthly standard deviation (SD; contour): (a) B–CTL; (b) POP2–waves; (c) NOAA CT2022: red dashed lines highlight the primary oceanic CO_2 outgassing and uptake regions in the Pacific Ocean, while black dashed lines mark regions with an air–sea CO_2 flux below -2 mol m^{-2} yr^{-1}; (d) Based on NOAA CT2022, 12 key regions exhibiting significant air–sea CO_2 flux variability have been identified among the 30 ocean basins defined in the CT2022 model documentation (Jacobson et al., 2023) for optimizing flux inversions. These include: (1) Pacific Ocean: Northwestern Pacific (NWP), Eastern Equatorial Pacific (EEP) (underlined labels highlighted with red dashed lines in Fig. 2c), as well as Northeastern Pacific (NEP), South Pacific Ocean 1 (SPO1), and South Pacific Ocean 2 (SPO2); (2) Indian Ocean: North Indian Ocean (NIO), South Indian Ocean 1 (SIO1), and South Indian Ocean 2 (SIO2); (3) Atlantic Ocean: North Atlantic Ocean 1 (NAO1), North Atlantic Ocean 2 (NAO2), Equatorial Atlantic Ocean (EAO), and South Atlantic Ocean (SAO), (e) the air–sea CO_2 flux difference between B–CTL and NOAA CT2022, and (f) same as (e), but between POP2–waves and NOAA CT2022.

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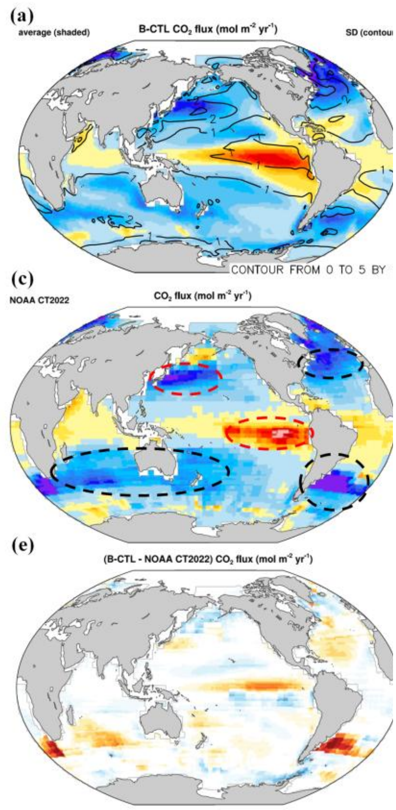

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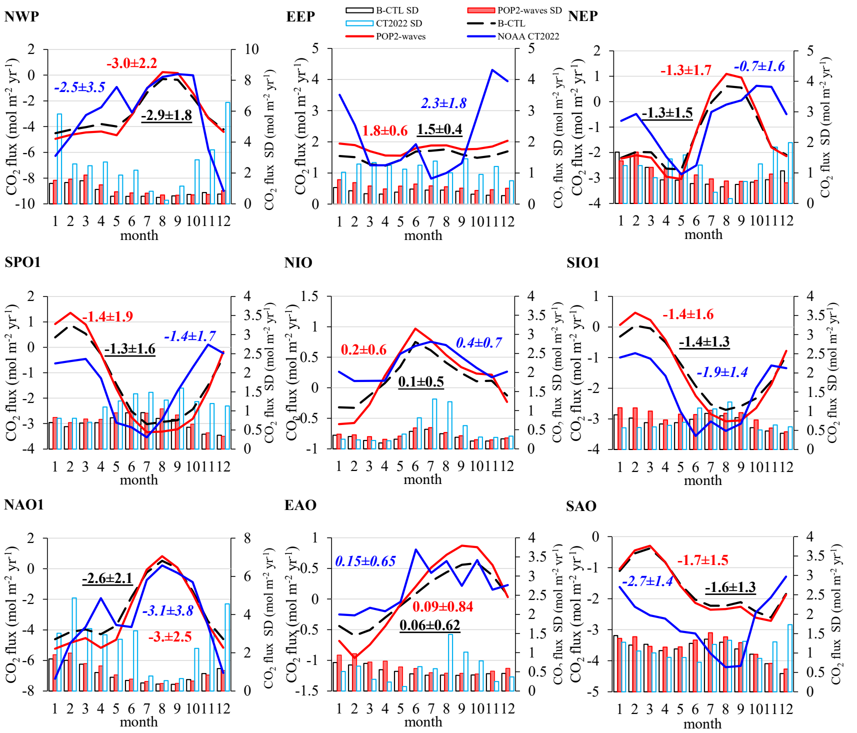

Figure 3. Climatological monthly mean CO_2 flux for nine highly correlated regions defined in Fig. 2d (NWP, EEP, NEP, SPO1, NIO, SIO1, NAO1, EAO, and SAO). Lines indicate values for B–CTL (black dashed), POP2–waves (red solid), and NOAA CT2022 (blue solid). Regional-average values are distinguished by a black underline for B–CTL, red font for POP2–waves, and blue italics for NOAA CT2022. Monthly standard deviation (SD) is shown as bars: black hollow (B–CTL), red solid (POP2–waves), and light blue hollow (NOAA CT2022).

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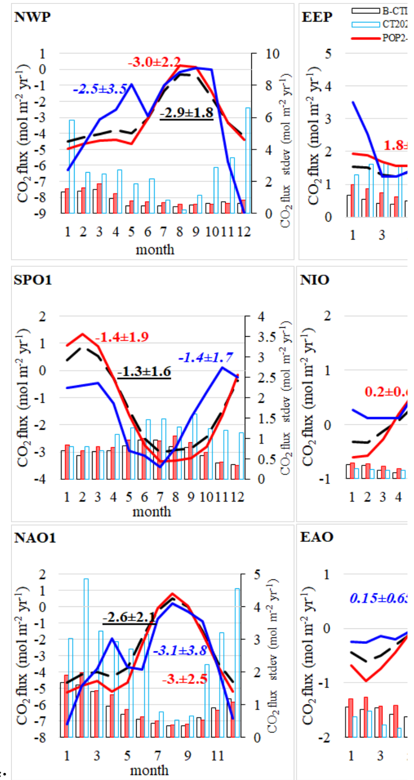

已刪除: **Figure 3.** The monthly mean values of the high-correlation climatological average of CO_2 flux with NOAA CT2022 are based on nine characteristic regions (NWP, EEP, NEP, SPO1, NIO, SIO1, NAO1, EAO, and SAO) shown in Fig. 2d. The black dashed line represents B–CTL, the red solid line represents POP2–waves, and the blue solid line represents NOAA CT2022. The bar graph illustrates the standard deviation (stdevSD) for each month. Black hollow bars correspond to B–CTL, red solid bars to POP2–waves, and light blue hollow bars to NOAA CT2022.

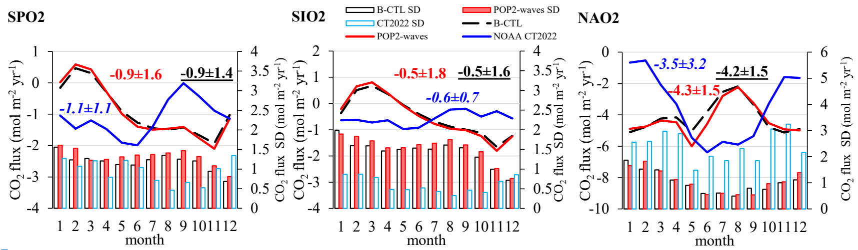

Figure 4. Same as Fig. 3, but for the low-correlation climatological average of the simulated CO_2 flux compared with NOAA CT2022, based on the three characteristic regions (SPO2, SIO2, and NAO2).

*Note: SPO2: South Pacific Ocean Region 2; SIO2: South Indian Ocean Region 2; NAO2: North Atlantic Ocean Region 2.

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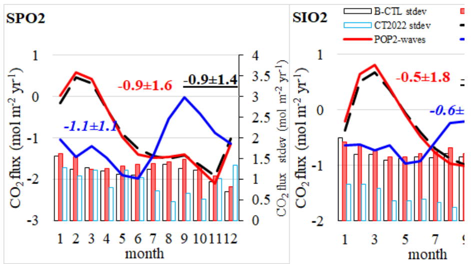

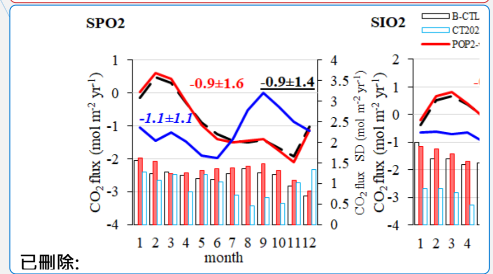

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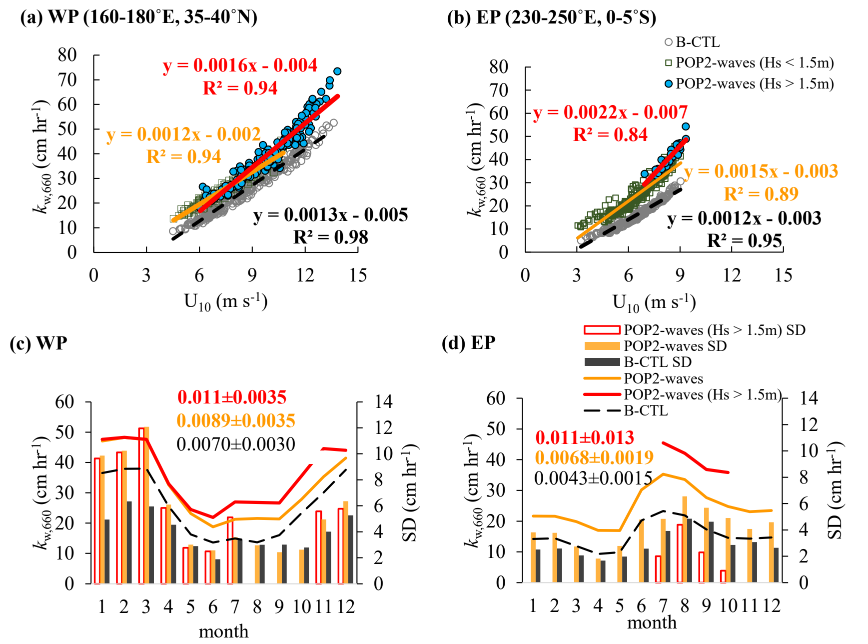

Figure 5. Scatter plots of monthly averaged gas transfer velocity ($k_{w,660}$) versus U_{10} for (a) the Western Pacific (WP; 160–180°E, 35–40°N) and (b) the Eastern Pacific (EP; 230–250°E, 0–5°S). Gray open circles, green open squares, and blue dots with black edges represent B–CTL, POP2–waves data for H_s < 1.5 m, and H_s > 1.5 m, respectively. Corresponding simple linear regressions (SLRs) and squared correlation coefficients (R^2) are shown with black dotted lines/text (B–CTL), orange solid lines/text (POP2–waves, H_s < 1.5 m), and red solid lines/text (POP2–waves, H_s > 1.5 m). Panels (c) and (d) display the monthly mean values of $k_{w,600}$ for WP and EP, respectively. Red and orange solid lines/text indicate POP2–waves (H_s > 1.5 m) and the average of all POP2–waves data, while the black dotted line/text represents B–CTL. The bar graph illustrates the standard deviation (SD) for each month over the WP and EP, red hollow bars for POP2–waves (H_s > 1.5 m), orange for average POP2–waves, and black for B–CTL.

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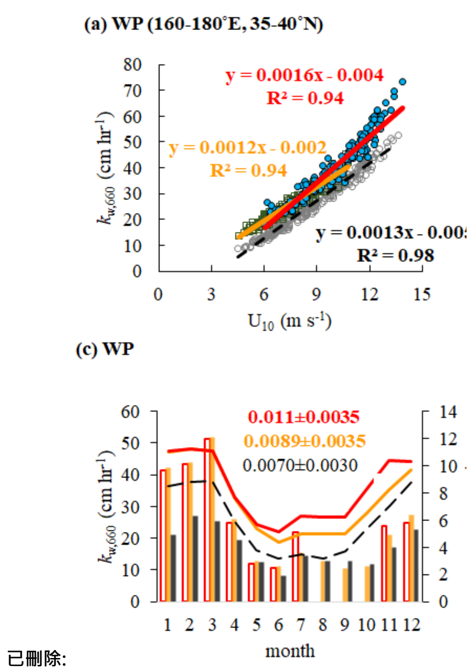

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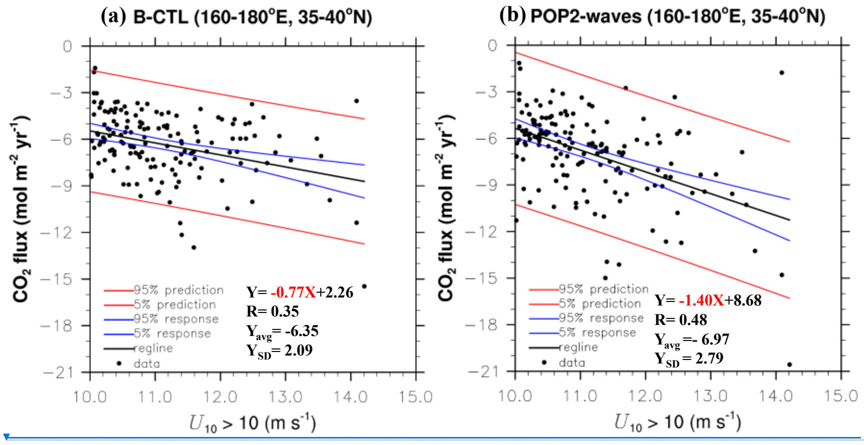

Figure 6. Scatter plots of air–sea CO_2 flux versus U_{10} with regression lines over the Western Pacific (160–180°E, 35–40°N) under high-wind conditions (U_{10} > 10 m s^{-1}) for (a) B–CTL and (b) POP2–waves experiments. Black dots indicate monthly mean values from each experiment. Blue lines denote the 95% and 5% confidence limits of the mean response, while red lines denote the 95% and 5% prediction intervals. The regression equation (Y = mx + b), correlation coefficient (R), mean CO_2 flux (Y_{avg}), and standard deviation (Y_{SD}) are shown to the right of the legend.

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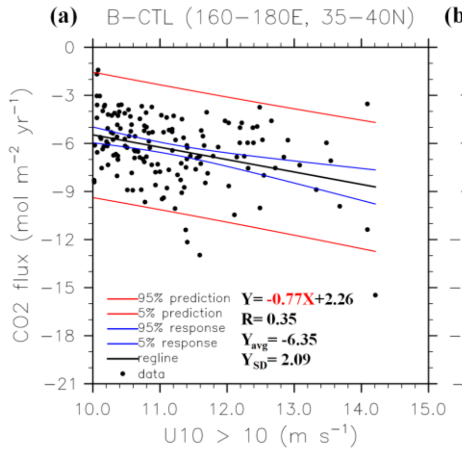

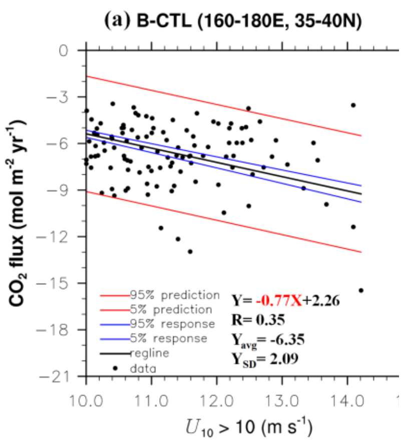

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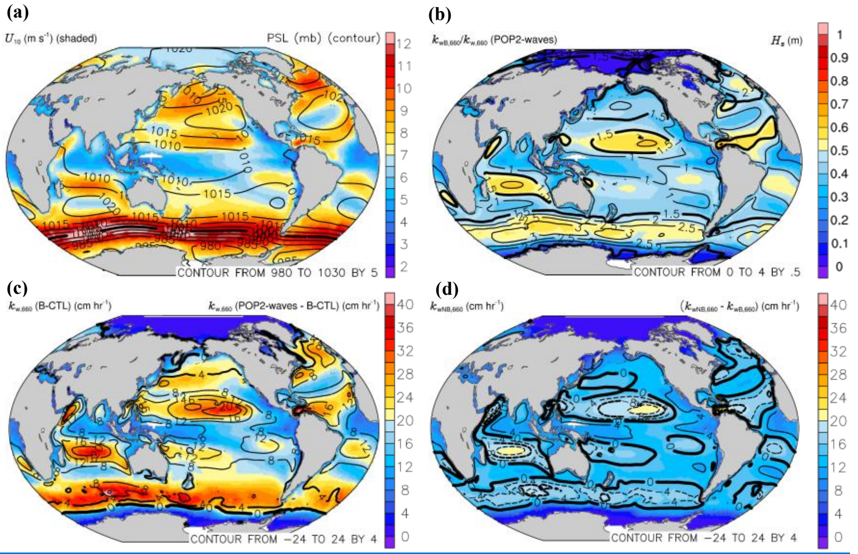
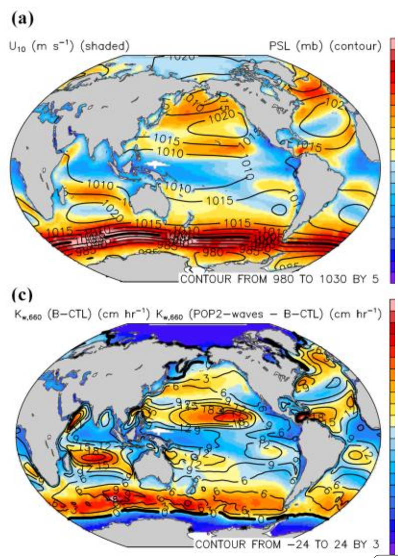


Figure 7. Effects of atmospheric factors and wave components on average gas transfer velocity: (a) Shaded areas represent 10-m wind speed (U_{10}, m s^{-1}) and contours show sea level pressure (PSL, mb); (b) color denotes the ratio of bubble-mediated ($k_{wB,660}$) to POP2–waves $k_{w,660}$ components, with contours representing significant wave height H_s (m) and thick lines marking H_s = 1.5m; (c) shading represents $k_{w,660}$ (B–CTL) and contours represent the difference $k_{w,660}$ (POP2–waves − B–CTL) in cm hr^{-1}; (d) shading shows the non-bubble-mediated component ($k_{wNB,660}$) and contours show the difference ($k_{wNB,660}$ − $k_{wB,660}$) in cm hr^{-1}, with the thick contour lines at 0.



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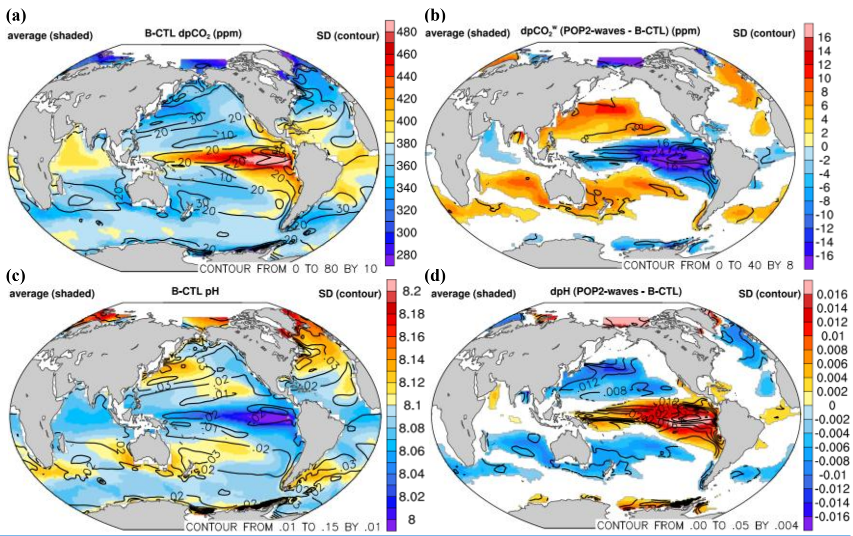
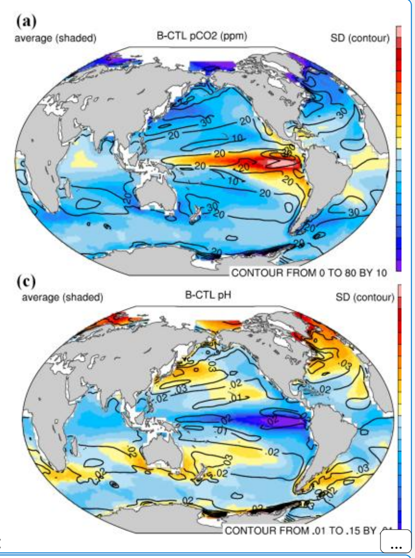


Figure 8. Spatial distributions of the 30-year climatological averages and model differences for $dpCO_2^w$ (ppm; panels a, b) and ocean surface pH (panels c, d). Climatological means are represented by shading, and their corresponding standard deviation (SD) is superimposed as contours. Panels (a) and (c) present baseline results from the B–CTL simulation. Panels (b) and (d) depict the structural differences between the POP2–waves and B–CTL experiments (POP2–waves minus B–CTL), where stippling indicates statistical significance at the 95% confidence level based on a Student's t-test.



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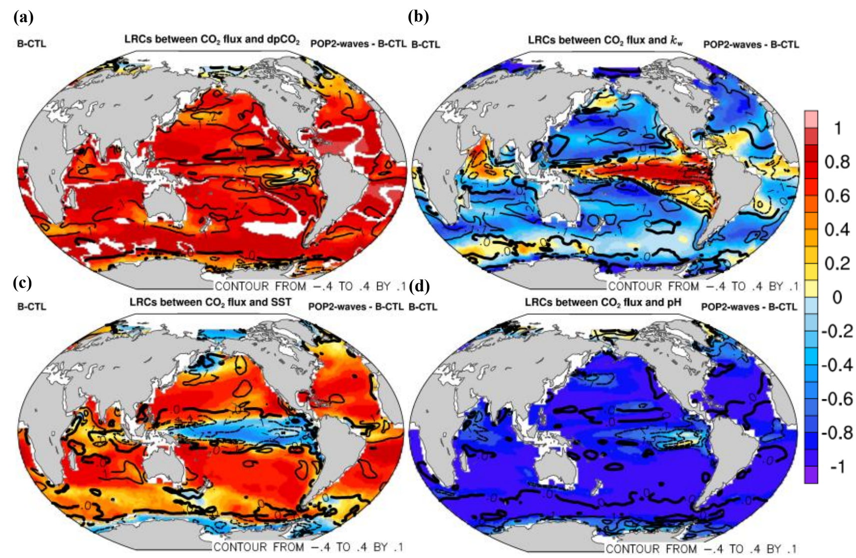
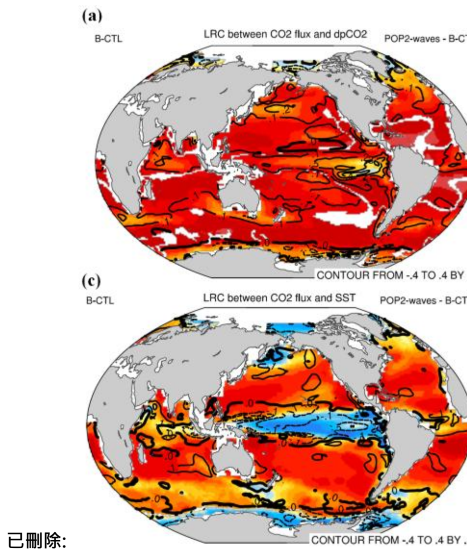


Figure 9. Spatial distributions of the 30-year averages of the linear regression coefficients (LRCs) between the CO_2 flux and (a) $dpCO_2$, ((b) $k_{w,660}$, (c) SST, and (d) pH. Shading represents the baseline LRCs from the B–CTL simulation, while overlaid contours depict the structural differences between the POP2–waves and B–CTL experiments (POP2–waves minus B–CTL). White areas denote regions where the LRCs are statistically insignificant at the 95% confidence level (based on a Student's t-test). All variables were standardized prior to the regression analysis.



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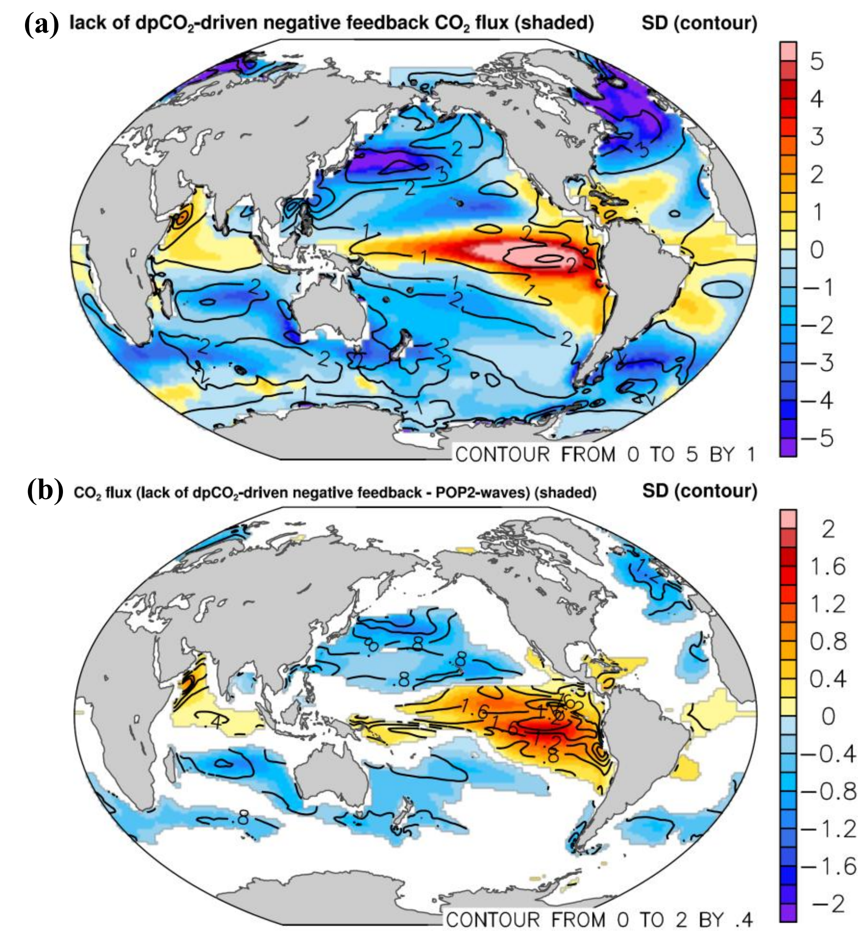
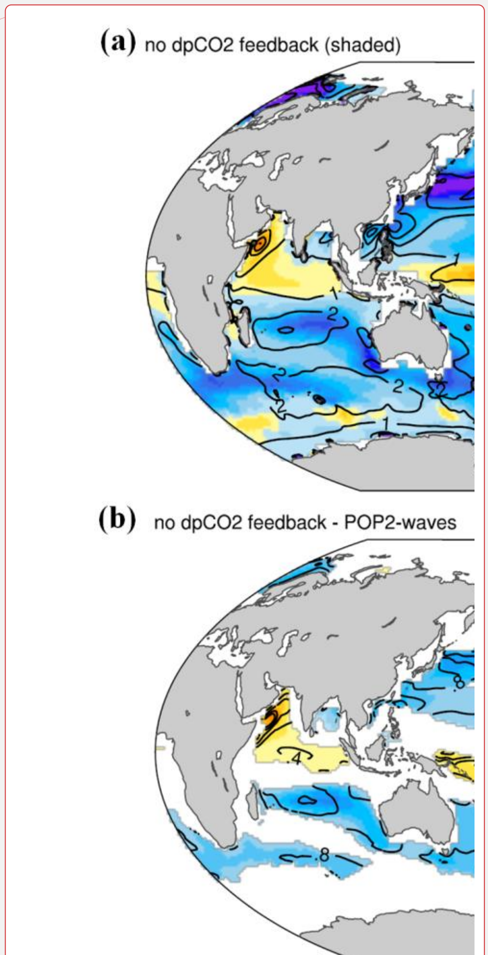


Figure 10. Estimated air–sea CO_2 flux assuming the absence of the $dpCO_2$-driven negative feedback mechanism. (a) Air–sea CO_2 flux calculated using uncoupled, independent $dpCO_2$ values from the B–CTL baseline and wave-dependent $k_{w,660}$ formulations (labeled as the "lack of $dpCO_2$-driven negative feedback" case). Shading represents the climatological mean, and contours indicate the corresponding standard deviation (SD). (b) Spatial difference in CO_2 flux between the decoupled feedback case and the fully coupled POP2–waves simulation (decoupled minus POP2–waves). Shading denotes the mean difference, and overlaid contours indicate regions where the difference is statistically significant at the 95% confidence level based on a Student's t-test.



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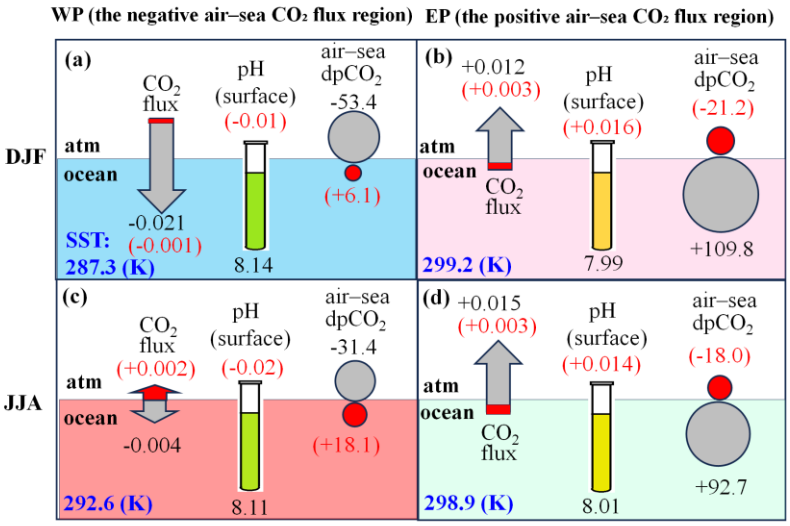

Figure 11. Schematic diagrams illustrate B–CTL (gray colors) and the differences between POP2–waves and B–CTL (red colors) in air–sea CO_2 flux (arrows), surface pH (tube colors indicator), and air–sea dpCO₂ (circle markers) over the oceanic CO_2 uptake region (WP; 160–180°E, 35–40°N) and the oceanic CO_2 outgassing region (EP; 230–250°E, 0–5°S). Each panel includes an ocean-atmosphere interface, with ocean color shading representing relative SST levels across different seasons. The lower-left corner displays the SST value, (a) WP in DJF seasonal mean, (b) EP in DJF seasonal mean, (c) WP in JJA seasonal mean, and (d) EP in JJA seasonal mean.

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