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REVIEW

Advances in understanding of air—sea exchange and cycling of greenhouse gases in the upper ocean

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The air–sea exchange and oceanic cycling of greenhouse gases (GHG), including carbon dioxide (CO_2), nitrous oxide (N_2O), methane (CH_4), carbon monoxide (CO), and nitrogen oxides ($NO_x = NO + NO_2$), are fundamental in controlling the evolution of the Earth's atmospheric chemistry and climate. Significant advances have been made over the last 10 years in understanding, instrumentation and methods, as well as deciphering the production and consumption pathways of GHG in the upper ocean (including the surface and subsurface ocean down to approximately 1000 m). The global ocean under current conditions is now well established as a major sink for CO_2, a major source for N_2O and a minor source for both CH_4 and CO. The importance of the ocean as a sink or source of NO_x is largely unknown so far. There are still considerable uncertainties about the processes and their major drivers controlling the distributions of N_2O, CH_4, CO, and NO_x in the upper ocean. Without having a fundamental understanding of oceanic GHG production and consumption pathways, our knowledge about the effects of ongoing major oceanic changes—warming, acidification, deoxygenation, and eutrophication—on the oceanic cycling and air–sea exchange of GHG remains rudimentary at best. We suggest that only through a comprehensive, coordinated, and interdisciplinary approach that includes data collection by global observation networks as well as joint process studies can the necessary data be generated to (1) identify the relevant microbial and phytoplankton communities, (2) quantify the rates of ocean GHG production and consumption pathways, (3) comprehend their major drivers, and (4) decipher economic and cultural implications of mitigation solutions.

Keywords: Carbon dioxide, Nitrous oxide, Methane, Carbon monoxide, Nitrogen oxides, Surface ocean

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1. Introduction

The interactions between the atmosphere and the ocean are driven by the exchange of momentum, energy, and material (i.e., gases and particles) across the surface ocean/lower atmosphere interface (e.g., Liss and Johnson, 2014). Atmospheric trace gases such as carbon dioxide (CO_2), nitrous oxide (N_2O), methane (CH_4), carbon monoxide (CO), and nitrogen oxides ($NO_x = NO + NO_2$) play significant roles in developing Earth's climate, and the chemistry of the Earth's atmosphere and oceans, with continuous exchanges between the lower atmosphere and the surface ocean that vary in time and space (**Table 1**).

CO_2 and N_2O are classified as long-lived atmospheric trace gases (Gulev et al., 2021). In contrast, CH_4, CO, and NO_x, which have comparably shorter lifetimes, are classified as short-lived atmospheric trace gases (Szopa et al., 2021). On the global scale, all gases except for CO show increasing tropospheric mole fractions since the industrial revolution (**Table 1**). They all act as greenhouse gases (GHG) and contribute to the ongoing warming of the Earth, that is, the greenhouse effect. CO_2 and N_2O are direct GHG. CO and NO_x are indirect GHG because they have an indirect impact on the radiative balance of the Earth through their chemical reactivity in the atmosphere leading to the formation of GHG such as CH_4, CO_2, ozone (O_3) as well as nitrate aerosols (Szopa et al., 2021). CH_4 is both a direct and an indirect GHG. The effective radiative forcing of CO_2, N_2O, CH_4, and CO is positive (a warming effect), whereas NO_x has an overall negative effective radiative forcing (a cooling effect; **Table 1**).

A comprehensive overview on trace gas cycling in the ocean is given in Liss and Johnson (2014). Regionally focused overviews on GHG cycling in polar regions, coastal upwelling systems and the Indian Ocean are presented in Willis et al. (2023), Lachkar et al. (n.d.), and Kumar et al. (n.d.), respectively. In this synthesis, we focus on reviewing the advances over the last decade in the knowledge and understanding of air–sea gas exchange and cycling of GHG in the upper ocean (0–1000 m water depth) and their related impacts and feedback processes within the upper oceanic and lower atmospheric system.

2. Greenhouse gases

2.1. Carbon dioxide (CO_2)

The exchange of CO_2 between the atmosphere and the biosphere (on land and at sea) has several unique physical and socio-economic attributes. CO_2 is the dominant

Table 1. Overview of the climate relevance of CO_2, N_2O, CH_4, CO, and NO_x (Gulev et al., 2021; Szopa et al., 2021)

Greenhouse Gas	Atmospheric Lifetime	Mean Atmospheric Mole Fraction	Atmospheric Growth Rate	Climate Effects	Effective Radiative Forcing[a] ($W\ m^{-2}$)	Atmospheric Chemistry Effects
CO_2	Multiple[b]	409.9 ppm (in 2019)	1.56 ppm $year^{-1}$ (1959–2019)	Direct	2.1	Weathering of rocks
N_2O	109 years	332.1 ppb (in 2019)	0.9 ppb $year^{-1}$ (1985–2019)	Direct	0.24	Stratospheric O_3 depletion
CH_4	9 years	1866 ppb (in 2019)	7 ppb $year^{-1}$ (2007–2016)	Direct, indirect	1.2	Stratospheric O_3 depletion, oxidative capacity
CO	1–4 months	40/120 ppb (SH/NH)[c]	–0.32% $year^{-1}$ (2000–2017)	Indirect	0.44[d]	Oxidative capacity
NO_x	Hours–days	30–40 ppt/up to several ppb[e]	Increasing (1850–2015)	Indirect	−0.27	Tropospheric O_3 formation, acid rain, aerosol formation

[a] Emission-based estimates for the period 1750–2019 (Szopa et al., 2021).

[b] The lifetime of CO_2 cannot be represented with a single value because the gas is not destroyed over time; instead it moves among different parts of the ocean/atmosphere/land system. Some of the excess CO_2 is absorbed quickly (e.g., by the ocean surface), but some will remain in the atmosphere for thousands of years, due in part to the very slow process by which carbon is transferred from ocean surface to ocean sediments (e.g., Tans, 1997).

[c] Southern hemisphere/northern hemisphere.

[d] Includes non-methane volatile organic compounds.

[e] Background over the ocean (Warneck, 2000)/in populated, urbanized, and industrialized regions.

human-produced GHG in the atmosphere and quantifying the fluxes between the reservoirs is critical for the global stocktake, which reviews global progress towards the Paris Agreement goals (https://unfccc.int/process-and-meetings/the-paris-agreement) every 5 years (Roelfsema et al., 2020). The air–sea exchange is the main conduit between ocean and atmosphere and determines the role of the ocean in modulating atmospheric levels of GHG. The ocean plays a major role in sequestering CO_2 on a global scale and annually captures approximately 26% of anthropogenic CO_2 emissions, C_{anth} (carbon emitted as CO_2 by burning fossil fuels, land use change, and cement production; Gruber et al., 2023). In response to increasing atmospheric CO_2 mole fractions, the ocean CO_2 sink increased from 1.1 ± 0.4 Pg C yr^{-1} in the 1960s to 2.9 ± 0.4 Pg C yr^{-1} for the 2012–2021 period (Friedlingstein et al., 2022). Coastal oceans take up about 0.4 Pg C yr^{-1} (Chen et al., 2013). In contrast to coastal and open oceans, estuaries are emitting CO_2 to the atmosphere. A recent study revealed estuarine emissions of about median (Q1–Q3) 0.03 (0.02–0.05) Pg C yr^{-1} (Rosentreter et al., 2023), which is lower than most of the recent mean estimates (Chen et al., 2013).

Increases in CO_2 concentrations in the ocean deplete carbonate ions while increasing hydrogen ions, which decreases the ocean's CO_2 buffering capacity and increases acidification of the ocean, respectively. These changes have been shown to pose a measurable impact on marine habitats and their resident organisms (Doney et al., 2009; Hauck, 2018; Kwiatkowski and Orr, 2018). CO_2 differs from other GHG in that ocean inventories of inorganic carbon in the form of bicarbonate and carbonate are about 40 times greater than the atmospheric CO_2 content, and thus cause the ocean to be a buffer with respect to CO_2. The rate of uptake of atmospheric CO_2 is controlled by air–sea exchange, and its transport into the ocean's interior. On millennial timescales 90% of the C_{anth} will be captured by the ocean (Archer et al., 2009).

On global scales the ocean–atmosphere CO_2 fluxes are estimated by different empirical modelling and mass balance approaches (e.g., Broecker et al., 1979; Landschützer et al., 2016). The identified increase in ocean absorption is closely tied to the increasing atmospheric CO_2 mole fractions from C_{anth}, with the ratio of uptake to C_{anth} release of 0.25 ± 0.02 from 1959 to 2021 (McKinley et al., 2020; Friedlingstein et al., 2022). This close connection implies that surface ocean pCO_2 (partial pressure of CO_2 in water) or fCO_2 (fugacity of CO_2 in water, which includes a correction of pCO_2 for non-ideal behavior; e.g., Wanninkhof et al., 2022) has kept up with the increases in atmospheric CO_2 mole fractions over the past 60 years. This net global CO_2 uptake is a composite of regional and seasonal fluxes that vary greatly in magnitude and direction, and assessments are based on modelled and observed datasets. These assessments rely upon surface ocean pCO_2 data, which are commonly measured using automated systems with commercially available units capable of determining water pCO_2 within 2 μatm and air pCO_2 within 0.2 μatm (Steinhoff et al., 2019). However, sparsity of these measurements requires interpolation in time and space that is done by a variety of methods, including machine learning techniques, or empirical approaches that use predictor variables (e.g., location, time, sea surface temperature, and mixed layer depth) that can be obtained at higher time and space resolutions (see the examples given in Rödenbeck et al., 2015). Along with these surface ocean pCO_2 datasets, the net CO_2 uptake assessments and approaches are also critically dependent on remotely sensed variables and re-analysis data products for providing the predictor inputs (for interpolating the sparse surface ocean pCO_2 data) and for providing the inputs for calculating the gas exchange velocity and the atmospheric and oceanic CO_2 concentrations when using bulk gas flux methods (Shutler et al., 2020).

The gas transfer velocity (k_w) is now considered a dominant source of uncertainty in global ocean CO_2 flux assessments (Woolf et al., 2019). The term k_w is often expressed as a quadratic dependence, with wind speed tuned to global constraints, or based on dedicated deliberate passive tracer studies. Over the last decade, consistent results have been obtained between these methods (Ho et al., 2011). This formulation overlaps within the respective uncertainty bounds with frequently used parameterizations, as in Nightingale et al. (2000) and Wanninkhof (2014), over intermediate wind speeds (5–12 m s^{-1}). However, few observational constraints are available at higher and lower winds; therefore, work is still needed to further constrain exchange within these regimes where other factors affect gas transfer, in addition to wind-forced surface turbulence. Surfactants (diurnal), heating and cooling, and stratification influence transfer at low wind conditions (e.g., Yang et al., 2021), whereas bubbles and changes in surface drag become important at high wind conditions (e.g., Blomquist et al., 2017). These issues apply to the air–sea exchange of all gases, including CH_4, N_2O, and CO, as well as CO_2. A detailed discussion of the uncertainties associated with k_w is being prepared in a companion paper for this special feature focused on air–sea interfacial and exchange processes.

These local and often short duration phenomena are now probed with micrometeorological (direct) flux measurement approaches (Businger and Delany, 1990), including eddy covariance. These direct methods serve as an independent method to verify fluxes obtained by indirect bulk formulations, and for evaluating or refining their underlying assumptions (Dong et al., 2021). In particular, independent enhancements of wind speed and the suppression of k_w can be fully investigated. However, addressing these phenomena within some regions remains challenging for either, or both, of these methods (e.g., polar regions; Willis et al., 2023).

The last decade has seen a substantial community effort to be consistent in collating and performing quality control on surface ocean pCO_2 (fCO_2) data (led by those within the Surface Ocean CO_2 Atlas, SOCAT, community; e.g., Bakker et al., 2016), thereby helping to elevate the importance of observation-based assessments of surface ocean CO_2 absorption (e.g., Friedlingstein et al., 2022), enabling the harmonization of methods across disciplines, and identifying the importance for community-consistent methods (Fay et al., 2021). Increased awareness of the importance of more accurate and precise calculations has been identified (Woolf et al., 2016). This increased awareness, in combination with more consistent use of well characterized satellite remote sensing data (e.g., climate data records and their uncertainty information), which global and regional assessments increasingly rely upon, is likely to be important to enable new knowledge of CO_2 uptake by the global ocean.

Assuring high data quality comes at some costs. The SOCAT database relies on under-resourced efforts to maintain the database and voluntarily perform quality control on the increasing number of data sets. One approach to support these efforts would be the standardization of instrumentation and clear community guidance on uncertainty methodologies. Here the Surface Ocean CO_2 Reference Network (SOCONET) and the Integrated Carbon Observation System (ICOS; Steinhoff et al., 2019), together with other networks, can play a guiding role in the future.

The variety of instrumentation for measuring surface ocean pCO_2 has grown over the last decade, ranging from classical air-water equilibration systems (Pierrot et al., 2009) to membrane-based systems (e.g., Arruda et al., 2020; Macovei et al., 2021). This growth has opened up new opportunities for measuring pCO_2 in the ocean: while installations with air-water equilibration are still the backbone of SOCAT with well-established operating and calibration procedures, the smaller instrumentation can be installed on autonomous surface vehicles or smaller vessels like sailing yachts. Such installations are increasing the observation capacity in so far under-sampled regions (e.g., Indian Ocean, Southern Ocean). However, the increase in availability of instrumentation also comes with an increased number of different, and sometimes unknown, qualities of these instruments. In 2021 the Ocean Thematic Centre of the European research infrastructure ICOS organized a pCO_2 instrument intercomparison with more than 25 ocean pCO_2 instruments. Outcomes so far suggest that: (1) air-water equilibration systems using at least one non-zero reference gas are most suitable to provide high-quality data; and (2) the community needs to increase emphasis on reporting complete metadata and rigorous uncertainty estimates, all of which follow Findability, Accessibility, Interoperability, Reuse (FAIR) principles and will support users in identifying the quality of each data set. A data set with known uncertainties, even if the uncertainties are considered large, is more useful than data with vaguely reported or missing uncertainty information and meta-data.

Community recommendations as presented in the Integrated Ocean Carbon Research report (2021) rely heavily on improved knowledge and measurements of GHG fluxes. In particular, addressing two of the main questions—"Will the oceanic uptake of anthropogenic CO_2 continue as primarily an abiotic process?" and "How are humans altering the ocean carbon cycle and resulting feedback?"—will depend critically on better measurement and understanding of air–sea fluxes. These efforts will likely benefit from machine learning and increased use of remote sensing approaches (Shutler et al., 2020; Cronin et al., 2023).

The increasing importance of observation-based assessments of the ocean CO_2 uptake has helped to identify the increasing strength of the ocean absorption, but has also identified a clear discrepancy between the observations and modelling assessments, which are steadily diverging with time (as evident in Friedlingstein et al., 2022). Sensitivity evaluation of model parameterizations and their data, alongside observation-based data (in situ, re-analyses and remote sensing) with the full inclusion of uncertainty information, could help to begin elucidating the differences between the model and observation assessments. Some efforts are already providing advances, but more analyses and approaches are needed, which is likely to be major focus for the community over the next 2–5 years and beyond. For example, recent modelling work (Bellenger et al., 2023) has identified one reason for differences between many models and one observation-based dataset presented in Friedlingstein et al. (2022), resulting in recommendations to the modelling community, whereas another recent model and observation comparison study (Hauck et al., 2023) has identified the impact of sparse sampling (which varies with time and space) which is more apparent within some observation-based data products. More advanced model and data inter-comparison approaches, where data from each source (model data versus observation-based data products versus in situ data) are treated on a more equal or standardized basis (e.g., as developed by Land et al., 2023), are likely to lead to an improved ability to identify the sources of any model versus observational differences. Future advancements in understanding of gas transfer, which is now considered a dominant source of uncertainty within global air–sea flux assessments (Woolf et al., 2019), and knowledge gained from indirect and direct flux comparisons will also help to strengthen confidence in the observation-based approaches and to further constrain exchange within all biogeochemical regimes of the open and coastal oceans.

2.2. Nitrous oxide (N_2O)

2.2.1. Global surface distribution and emissions of N_2O

The most recent assessment of oceanic N_2O emissions reported a mean annual flux of 4.2 ± 1.0 Tg N yr^{-1} (Yang et al., 2020). This estimate is not significantly different from the often-cited estimate of 4 (1.2–6.8) Tg N yr^{-1} by Nevison et al. (1995), but the associated uncertainty markedly decreased from ± 70% to ± 20%. Coastal oceans and estuaries emit about 0.7 and 0.04 Tg N yr^{-1}, respectively, to the atmosphere (Seitzinger and Kroeze, 1998; Rosentreter et al., 2023).

The global large-scale distribution of N_2O in surface waters was established in the late 1990s (Nevison et al., 1995; Suntharalingam and Sarmiento, 2000). The determination of the small-scale distribution of N_2O was made possible only during the last decade due to the development of the cavity-enhanced absorption spectroscopy technique, which enabled high-resolution surveys of dissolved N_2O in ocean surface waters (Arévalo-Martínez et al., 2013; Arévalo-Martínez et al., 2015). The increasing

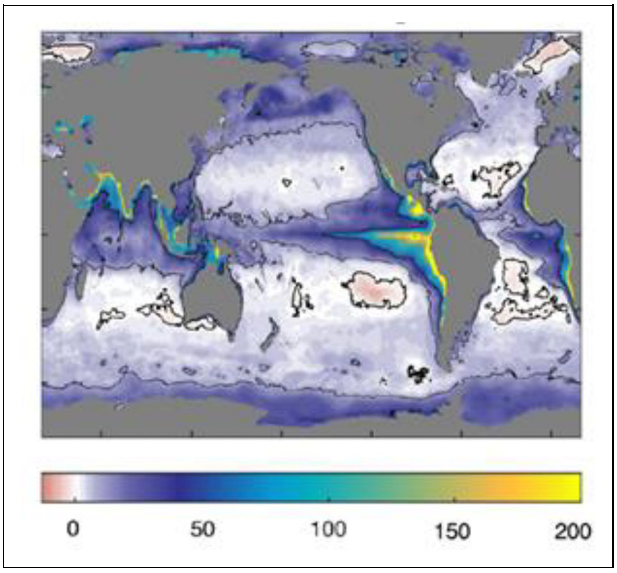

Figure 1. Global mean distribution of surface N_2O from Yang et al. (2020). The figure is taken from Wilson et al. (2020). Color-coded values are expressed as the difference of the partial pressures of N_2O (pN_2O) in the ocean surface layer and the atmosphere ($\Delta pN_2O = pN_2O_w - pN_2O_a$ in natm). Positive values of ΔpN_2O suggest N_2O outgassing, whereas negative values indicate uptake by surface waters.

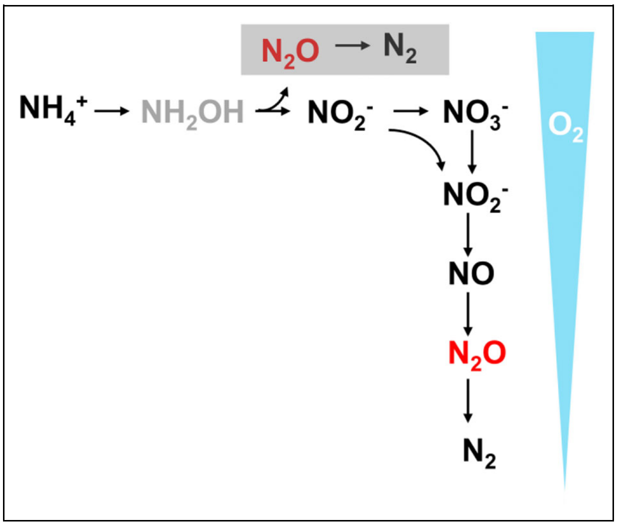

Figure 2. Production pathways of N_2O with nitrification taking place aerobically and denitrification under oxygen depletion. A new way of N_2O consumption via reduction has been identified as having the potential to take place in oxic surface waters (grey box).

number of observations (Bange et al., 2019a) in combination with the application of machine learning algorithms (Yang et al., 2020) led to an improved understanding of the spatial variability of oceanic N_2O (**Figure 1**). Despite the progress made, the majority of open and coastal ocean waters remain under-sampled, and therefore our knowledge about the seasonal and interannual variability of oceanic N_2O concentrations and emissions is still rudimentary at best. For example, the variability of N_2O in subpolar and polar oceans remains largely unresolved. Whereas some studies suggest these regions are weak-to-moderate sources of N_2O to the atmosphere, other studies have shown slight N_2O sinks (Rees et al., 2022, and references therein). Studies of N_2O within sea ice are scarce. However, they show severe source/sink shifts in response to ice formation cycles (Randall et al., 2012; Geilfus et al., 2021).

In order to further reduce uncertainties in current global N_2O oceanic emission estimates and better constrain and understand temporal and spatial variability of those emissions, more measurements are needed. Importantly, the oceanographic community is taking steps to standardize these measurements and improve the interoperability of the resulting datasets (Wilson et al., 2020).

2.2.2. N_2O production and consumption pathways

Classically, N_2O has been described as produced by two microbially driven pathways, that is, nitrification and denitrification (**Figure 2**; Bakker et al., 2014). In oxygen minimum zones (OMZ), denitrification dominates N_2O production (Ji et al., 2015; Ji et al., 2018). A recent study emphasized the importance of denitrification for N_2O production also in waters adjacent to OMZ (Frey et al., 2020), a role previously overlooked due to the lack of direct rate measurements. Apart from denitrification, nitrification by ammonia-oxidizing archaea also plays an important role for N_2O production in waters adjacent to OMZ (e.g., Löscher et al., 2012). See also the discussion of N_2O production in OMZ connected to upwelling regions in Lachkar et al. (n.d.). N_2O can be produced by anammox as well (Wan et al., 2023a). However, its importance for N_2O cycling in the ocean remains to be demonstrated. Only recently a study showed that N_2O production in the subtropical oceans covaries with remineralization of organic matter and thus is counteracting the radiative benefit of CO_2 removal by the marine biological pump (Wan et al., 2023b). N_2O photoproduction was demonstrated to take place in reservoirs and might take place in oceanic surface waters as well (León Palmero, 2021). Moreover, the role of N_2O production by phytoplankton is still largely unknown (McLeod et al., 2021). Decreasing pH (i.e., ocean acidification) has also been shown to affect N_2O production: the results from studies point to both increasing and decreasing N_2O production (Rees et al., 2016; Breider et al., 2019; Zhou et al., 2023).

N_2O consumption has long been assumed to take place only during the last step of denitrification under suboxic/anoxic conditions. However, this traditional view has been challenged by recent findings from the oxic surface waters of various ocean regions, including the eastern subtropical South Pacific Ocean, the Red Sea and the Arctic Ocean (e.g., Cornejo et al., 2015; Zhan et al., 2016; Bange et al., 2019b) that exhibited significant undersaturation in N_2O compared to the atmosphere. Undersaturation implies a pronounced in-situ consumption of N_2O, turning the surface ocean from a N_2O source into a N_2O sink. One explanation for this "oceanic nitrous oxide paradox" might be biological N_2 fixation, where N_2 fixers (diazotrophs) consume N_2O instead of N_2, as was suggested for the eastern South Pacific Ocean. N_2O consumption by diazotrophs is caused by a low specificity of the key enzyme nitrogenase, which is able to bind to different small linear molecules (Farías et al., 2013). Moreover, denitrifiers possessing a *nos*Z gene (the key functional marker of N_2O reduction to N_2), but not the other genes of canonical denitrification, were found in the ocean surface layer which indicates a potential consumption of N_2O under oxic conditions. This pathway might provide an alternative explanation for N_2O consumption in oxic waters (**Figure 2**; Rees et al., 2021; Sun et al., 2021) and might indeed help to explain the apparent N_2O consumption by diazotrophs (Farías et al., 2013): in a first step, N_2O could be denitrified to N_2 and, in a second step, N_2 could be fixed into biomass by diazotrophs. N_2O consumption by a coupled denitrification/N_2 fixation mechanism in the oxic surface ocean, however, awaits further investigation. Our present view of oceanic N_2O cycling might be incomplete and further process studies are needed to decipher the prevailing N_2O production/consumption pathways and their major drivers.

2.2.3. N_2O in coastal ecosystems and estuaries

Coastal ecosystems (estuaries, coastal upwellings, mangrove waters) are typified by large, positive N_2O anomalies (i.e., supersaturations with respect to atmospheric equilibrium) fueled by high nitrogen inputs that in turn drive large sea-to-air emissions. Consequently, they are significant contributors to marine-derived atmospheric N_2O (Bakker et al., 2014). Indeed, a recent reconstruction of marine N_2O emissions proposed that coastal upwelling regions, occupying less than 3% of the global ocean area, are responsible for around 20% of total marine N_2O emissions (Yang et al., 2020). Nevertheless, with respect to terrestrially influenced systems in particular (e.g., estuaries, mangroves), high variability of inputs and process rates with questions surrounding data processing and interpretation, such as the robustness of the Intergovernmental Panel on Climate Change (IPCC) N_2O emission factors (Maavara et al., 2019), continues to bring high uncertainty (Smith, 2017). For a recent summary review of N_2O formation and consumption processes in coastal ecosystems see Bakker et al. (2014). One means to reduce this uncertainty is to gain additional data in key regions. Until recently most data derived from Europe and North America, but data for other regions, notably tropical and subtropical locations, are increasingly available (Yang et al., 2020). Even so, in addition to improving spatial-temporal data resolution, new approaches are required. New insight has been derived in recent years, for example, from mixed-effect regression models that predict N_2O concentrations from water quality parameters (Lequy et al., 2022). There is also growing evidence for a more widespread occurrence of coastal N_2O sinks than was previously thought (Foster and Fulweiler, 2016; Maher et al., 2016; Rees et al., 2021; Lequy et al., 2022). As these studies are few and biased towards temperate locations, further work is required, particularly in tropical and sub-tropical regions, to assess the nature and wider implications of local-to-regional N_2O sinks for the global N_2O budget.

Other recent work in estuarine waters and sediments has established a widespread enhancement of N_2O production at low O_2 concentrations, with maximal rates at suboxic levels (Foster andFulweiler, 2016; Barnes and Upstill-Goddard, 2018; Tang et al., 2022). An important implication is that increased coastal N_2O emissions should be expected from the expansion of estuarine and coastal hypoxia/anoxia in response to global change (Breitburg et al., 2018). Importantly, associated positive warming feedback has been proposed (Ni et al., 2019). Moreover, Wells et al. (2018) showed how changes in land use intensity affect estuarine N_2O fluxes.

Combined investigative approaches drawing on cross-disciplinary expertise show substantial future promise. For example, microplastics are now known to modify the marine nitrogen cycle (e.g., Seeley et al., 2020), especially in coastal waters; more specifically, plastic-associated biofilm can enhance anoxic conditions, potentially favoring denitrifiers and stimulating N_2O production (Su et al., 2022). A recent combination of biogeochemical (dual-isotope, ^{15}N-^{18}O, labelling) and genetic (DNA fingerprinting) approaches revealed increased N_2O production in an estuarine sediment via the microbial colonization of plastic debris (Chen et al., 2022b). The isotope work showed that petroleum-based and biodegradable microplastics stimulated N_2O production by providing additional carbon and nitrogen sources, while the DNA techniques identified a micro-niche supporting elevated nitrification, heterotrophic denitrification, and nitrifier-denitrification. Biodegradable microplastics increased N_2O production by 50% (Chen et al., 2022b), which is important given their increasing contribution to environmental plastic waste (Seeley et al., 2020; Li et al., 2022). Another study combining these techniques found sediment antibiotic residues to enhance N_2O production via nitrification and heterotrophic denitrification, by 17% and 64%, respectively (Chen et al., 2022a).

The role of anthropogenic pollutants in coastal waters is clearly an important emerging aspect of N_2O biogeochemistry. Currently, the contribution from these emerging sources relative to those from more widely studied inputs, such as rivers and the nearshore deposition of reactive atmospheric nitrogen (Bakker et al., 2014), is not possible to quantify. Further targeted research is therefore required.

2.3. Methane (CH_4)

2.3.1. CH_4 emissions from the open and coastal oceans

In the surface waters of the open ocean, away from benthic or coastal sources, CH_4 is frequently supersaturated with respect to atmospheric equilibrium. Global oceanic coastal and open ocean emissions including geological (offshore) and biogenic open and coastal CH_4 emissions were estimated at 9–22 (mean of 13) Tg CH_4 yr^{-1} in a recent global synthesis (Saunois et al., 2020), with a slightly narrower uncertainty of 6–12 Tg CH_4 yr^{-1} provided by Weber et al. (2019). The phenomenon of CH_4 in the surface ocean was described originally as the "oceanic methane paradox" because it was not evident how CH_4 production could be so ubiquitous in oxygenated waters. The advances in our scientific understanding of the mechanisms that produce CH_4 in oxic surface waters (Section 2.3.2) have been supported by technological advances in CH_4 analyzers, such as cavity-enhanced absorption spectroscopy, which have increased the number of surface measurements (e.g., Gülzow et al., 2013).

Compared to the open ocean, the coastal zone is a hotspot of CH_4 production and emissions, particularly the shallow vegetated coastline fringed by mangroves, salt marshes, and seagrass meadows (Weber et al., 2019; Al-Haj and Fulweiler, 2020; Rosentreter et al., 2021b; Rosentreter et al., 2023). A recent regional and global data-driven meta-analysis showed that methane emissions from mangroves, salt marshes, and seagrasses combined (median (Q1–Q3) 0.8 (0.5–1.4) Tg CH_4 yr^{-1}) are more than double of those from estuaries (median (Q1–Q3) 0.3 (0.1–0.5) Tg CH_4 yr^{-1}; Rosentreter et al., 2023). Coastal aquaculture ponds alone emit a median (Q1–Q3) value of 0.6 (0.01–1.0) Tg CH_4 yr^{-1} (Rosentreter et al., 2021b). However, because of the high spatial-temporal variability within and between coastal systems and the overall global paucity of data, global estimates generally show a large range for both coastal vegetation (0.02–6.2 Tg CH_4 yr^{-1}; e.g., Garcias-Bonet and Duarte, 2017; Al-Haj and Fulweiler, 2020; Rosentreter et al., 2023) and estuaries (0.02–6.6 Tg CH_4 yr^{-1}; e.g., Bange et al., 1994; Upstill-Goddard et al., 2000; Borges and Abril, 2011; Rosentreter et al., 2023). Combining the median (Q1–Q3) coastal methane flux of 1.8 (0.6–5.6) Tg CH_4 yr^{-1} from estuaries, coastal wetlands, coastal aquaculture, and tidal flats (Rosentreter et al., 2021b; Rosentreter et al., 2023) with the diffusive CH_4 flux for near-shore (1.5 Tg CH_4 yr^{-1}), outer shelf (0.6 Tg CH_4 yr^{-1}), slope (0.3 Tg CH_4 yr^{-1}), and ebullitive flux for continental shelf (6–10 Tg CH_4 yr^{-1}) based on machine learning (Weber et al., 2019) gives a total of approximately 12 Tg CH_4 yr^{-1} for diffusive and ebullitive coastal ocean CH_4 emissions. In contrast, the global open ocean has substantially lower diffusive CH_4 emissions (approximately 1 Tg CH_4 yr^{-1}) because of the much lower areal CH_4 flux rates compared to coastal ecosystems (Weber et al., 2019; Rosentreter et al., 2023).

Increasing the temporal and spatial resolution of CH_4 observations will help to constrain the ocean inventory, emissions, and biogeochemical processes that influence its surface concentrations. Importantly, the oceanographic community is taking steps to standardize these measurements and improve the interoperability of the resulting datasets (Wilson et al., 2020).

2.3.2. CH_4 production and consumption pathways

The scientific discoveries that CH_4 could be produced aerobically in both aquatic and terrestrial environments raised the question of how widespread is this metabolic capability (Ernst et al., 2022). Marine microorganisms are now known to synthesize CH_4 via the metabolism of methylated compounds (Repeta et al., 2016; Klintzsch et al., 2019) and other pathways (Stawiarski et al., 2019; Bižić et al., 2020). Some of the CH_4 present in the surface ocean may also plausibly result from non-biological processes. For example, abiotic CH_4 photoproduction from chromophoric dissolved organic matter (CDOM) has been shown to occur across a wide range of marine environments including the open ocean (Li et al., 2020).

CH_4 emissions from coastal ecosystems are determined largely by the balance of sedimentary sources and sinks of CH_4 in both sediments and the water column (e.g., Mao et al., 2022). Recent work has focused on the environmental variables that regulate the efficiency of CH_4 production and consumption pathways, like temperature (Bussmann et al., 2021), CH_4 and oxygen concentrations (Steinle et al., 2017), trace metal concentration (Crespo-Medina et al., 2014), light irradiance (Thottathil et al., 2018), and transport and development of the methanotrophic microbial community in CH_4 plumes (Jordan et al., 2021). Apart from this focus, the role of alternative electron acceptors, such as nitrite and nitrate in the oxygen-depleted waters, remains elusive and a subject of ongoing research (Guerrero-Cruz et al., 2021).

Once produced, sedimentary CH_4 can reach the atmosphere via diffusion, ebullition (via gas bubbles), or through the aerenchyma of vascular plants (Jeffrey et al., 2019; Schorn et al., 2022). Other factors that can control CH_4 production and consumption processes, and thus its net release to the atmosphere, include hydrological (e.g., groundwater discharge), physicochemical (e.g., temperature), climatic (e.g., floods, droughts), biotic (e.g., bioturbation), and geomorphological (e.g., soil porosity) factors (Al-Haj and Fulweiler, 2020; Rosentreter et al., 2021a). Anthropogenic impacts such as wastewater pollution and land-use change are expected to elevate coastal CH_4 emissions in the future. Recently, plastics were shown to represent an unrecognized source of CH_4 (Royer et al., 2018) that might increase, as more plastic is accumulating in the coastal and open oceans. At present, the different production, consumption, and transportation pathways of CH_4 in coastal ecosystems are not well understood (Rosentreter et al., 2021a). The presence of plants further complicates coastal CH_4 budgets. More research is required to better quantify coastal CH_4 fluxes and to advance our fundamental understanding of CH_4 cycling in the coastal zone.

A central challenge in surface ocean CH_4 biogeochemistry is to assess a solid mass balance for multiple systems. This assessment requires an in-depth understanding on how environmental variables shape the pathways of CH_4 production and consumption, which is also a prerequisite for robust predictions on how oceanic methane emission will react to expected environmental changes.

2.4. Carbon monoxide (CO)

2.4.1. CO emissions from open and coastal oceans

Global oceanic CO emission estimates from recent studies fall in the range of 1–10 Tg C yr^{-1}, with the most likely values being 4–6 Tg C yr^{-1} (Stubbins et al., 2006a; Park and Rhee, 2016; Conte et al., 2019). According to Park and Rhee (2016) emissions from the coastal ocean account for about 0.9–1.5 Tg C yr^{-1}. The open and coastal oceans are a minor source of atmospheric CO, contributing up to 1%

to its natural and anthropogenic sources (Zheng et al., 2019). However, CO has a comparably short atmospheric lifetime of 1–4 months (Szopa et al., 2021); thus, its oceanic emissions can contribute significantly to the CO budget in the atmospheric marine boundary layer of remote areas such as the Arctic and Southern oceans where the contribution of other CO sources is low. In view of declining anthropogenic CO sources (Szopa et al., 2021), the importance of the oceanic source might increase in the future.

CO emissions from estuaries, coastal seas, and upwelling regions are often greater than those in oligotrophic open oceans but are highly variable (Day and Faloona, 2009; Kitidis et al., 2011; Zhang and Xie, 2012; Schmidt and Heikes, 2014). Coastal areas, however, are severely under-sampled, with the exception of the marginal seas off the coasts of northern China where a relatively large CO dataset has been collected during the last decade (e.g., Zhang et al., 2019). A limited number of surveys in the Arctic Ocean suggest that net CO emissions are probably enhanced compared to warmer waters due to slower microbial uptake and cyclic sea ice dynamics in cold waters (Xie et al., 2009a; Song et al., 2011; Tran et al., 2013; Gros et al., 2023).

2.4.2. CO production and consumption processes

Comparable to CH_4, CO emissions to the atmosphere are determined by the balance of CO sources and sinks. Oceanic CO is primarily produced from photodegradation of CDOM in sunlit surface waters (e.g., Zafiriou et al., 2003; Ossola et al., 2022). Estimates of the strength of this photochemical source on global scales spanned an enormous range in early studies (130–820 Tg C yr^{-1}; reviewed by Zafiriou et al., 2003) but became more consistent among later studies employing coupled optical-photochemical modelling: 50 (30–70) Tg C yr^{-1} (Zafiriou et al., 2003), 47 (38–84) Tg C yr^{-1} (Stubbins et al., 2006b) and 41 Tg C yr^{-1} (Fichot and Miller, 2010). Photodegradation of particulate organic matter (POM) has also been suggested to be a significant source of CO in marine environments on regional and global scales, being 10%–50% of the CDOM-driven photochemical source (Xie and Zafiriou, 2009; Song et al., 2013; McLeod et al., 2021). Under very high particulate loads, the POM source could even exceed the CDOM counterpart (Song et al., 2015; Song and Xie, 2017). In addition to the photochemical production pathway, dark (thermal) production of CO from organic matter has been measured directly in sterilized water samples (Zhang et al., 2008; Kitidis et al., 2011) or inferred from modelling temporal evolution of CO concentrations (Day and Faloona, 2009; Schmidt and Heikes, 2014). Dark CO production in the global ocean has been estimated to be 10%–30% of the global CDOM photochemical production (Zhang et al., 2008). The mechanisms of both the light and dark CO production are not well understood so far. In organic-rich estuarine waters, the dark source could be comparable in magnitude to the CDOM photochemical source (Zhang and Xie, 2012). A biogenic source of CO has also been observed in laboratory incubations of diverse marine phytoplankton (Gros et al., 2009) and inferred from vertical profiles of CO in the Arctic Ocean (Tran et al., 2013).

Microbial consumption represents the dominant loss for oceanic CO, consuming about 80%–90% of the produced CO (Zafiriou et al., 2003; Zafiriou et al., 2008; Stubbins et al., 2006a). Microbial CO consumption rates nevertheless are poorly constrained due partly to the ^{14}CO-based method yielding much lower rates than those obtained from non-^{14}CO methods (Tolli and Taylor, 2005; Xie et al., 2005). The majority of reported first-order microbial consumption rate coefficients (k_{bio}) vary by two orders of magnitude, with high values observed in warm estuarine and coastal waters and low values usually in cold offshore waters. To minimize this high variability, Kwon et al. (2020) recently proposed a second-order bacterial biomass-specific consumption rate coefficient, k_{bio}*, which is posited to be spatially and temporally constant. The k_{bio} and bacterial abundance data simultaneously collected from the Estuary and Gulf of St. Lawrence (Canada), however, showed substantial spatial-temporal variability of k_{bio}* (Xie et al., 2009b).

Recently published results from incubation experiments conducted in the Fram Strait (Arctic Ocean) under different environmental conditions showed that lower pH did not affect CO production and consumption rates, whereas they correlated positively with CDOM and dissolved nitrate concentrations, respectively (Campen et al., 2023). For future studies, further mapping of CO distributions (particularly in coastal waters) and k_{bio} are needed to improve the CO emission and consumption estimates. The methodological discrepancy between the ^{14}CO method and the non-^{14}CO methods for determining k_{bio} remains to be resolved.

2.5. Nitric oxide (NO) and nitrogen dioxide (NO_2)

The first studies about NO cycling in the ocean were published in the 1980s (e.g., Zafiriou et al., 1980) but only recently have the number of studies on oceanic NO increased. The overwhelming majority of the dissolved NO concentration measurements published during the last years are from the open and coastal areas of the East China and Yellow seas and from the western tropical North Pacific Ocean (Liu et al., 2017; Tian et al., 2019; Tian et al., 2020; Tian et al., 2021; Gong et al., 2023). Some studies from the Seto Inland Sea (Japan) have also been published (Olasehinde et al., 2010; Anifowose and Sakugawa, 2017; Adesina et al., 2018; Adesina and Sakugawa, 2021). Dissolved NO concentrations can be enhanced in coastal waters (e.g., Liu et al., 2017; Gong et al., 2023). Surface waters of the open and coastal oceans are now established as the usual source of atmospheric NO (Tian et al., 2020; Tian et al., 2021). However, atmospheric pollution events with high atmospheric mole fractions of NO over coastal waters can turn the surface ocean into a NO sink, resulting in an uptake of atmospheric NO (Gong et al., 2023). Oceanic NO emissions are small compared to other sources of atmospheric NO (Tian et al., 2019; Yao et al., 2020).

The early finding of photochemical NO production from dissolved nitrite in the surface ocean has been confirmed by actual field studies (e.g., Tian et al., 2020). NO is also a short-lived intermediate of various microbial processes of the marine nitrogen cycle, such as nitrification, denitrification, and anammox (e.g., Schreiber et al., 2012; Zhang et al., 2020). As a signaling molecule, NO is also involved in algal-bacterial interactions (Abada et al., 2023). However, the role of (micro)biological pathways for oceanic NO cycling is still largely unknown (Ward and Zafiriou, 1988; Lutterbeck et al., 2018; Gong et al., 2023). The loss processes for NO are under debate: on the one hand, its release to the atmosphere has been argued as the major sink for oceanic NO (Adesina and Sakugawa, 2021); on the other hand, a comparison of air–sea flux densities with photochemical production rates indicated a missing NO consumption process beside oxidation in ocean surface waters (Tian et al., 2019; Tian et al., 2020).

Considerably less is known about the cycling of NO_2 in the ocean. It can be produced from the photolysis of dissolved nitrite and nitrate but its hydrolysis is rapid (Tian et al., 2021). (Micro)biological production and/or consumption pathways of NO_2 in seawater are unknown. At this time, there is only one study of the distribution of dissolved NO_2 concentrations in the ocean: this study of NO_2 in the surface layer of the East China Sea by Tian et al. (2021) revealed an undersaturation of dissolved NO_2, indicating that the East China Sea was taking up NO_2 from the atmosphere at the time of the measurements.

Global estimates of the oceanic NO release as well as the NO_2 uptake are lacking, mainly because of the limited number of available measurements. To this end, further mapping of NO distributions (particularly in coastal waters) and production/consumption rates are needed to improve the NO emission and consumption estimates.

3. Climate feedback and sensitivity

3.1. CO_2

Variability in air–sea CO_2 fluxes on the order of 0.3–0.7 Pg C yr^{-1} are observed in response to large-scale climate reorganizations. The changes in upwelling of carbon-rich waters in the Equatorial Pacific caused by the El Niño-Southern Oscillation is the largest contributor of interannual variability in fluxes (Feely et al., 2006). The Southern Ocean had a decadal decrease in its role as CO_2 sink from 1995 to 2005 (Le Quéré et al., 2007) with a reinvigoration thereafter (Landschützer et al., 2015). This pattern is attributed to changes in intermediate water formation and ventilation but exact climate drivers are not fully clear. These variations do not manifest themselves in atmospheric CO_2 mole fractions, in part because of their relatively small magnitude in the atmosphere and also because in some instances the terrestrial uptake of CO_2 acts in the opposite sense to climate variability (Chatterjee et al., 2017). Year-to-year variability in terrestrial CO_2 uptake dominates the atmospheric variability.

The ocean CO_2 uptake will likely decline in the future but with uncertain magnitude. Ocean uptake could drop with decreasing CO_2 emissions (McKinley et al., 2020). The buffering of the surface ocean will decrease due to increases in the ocean CO_2 content, resulting in less uptake capacity. Climate change will lead to a warmer ocean and a more stratified ocean, with both factors decreasing CO_2 uptake as well (Chikamoto et al., 2023). Monitoring and understanding these changes is essential.

3.2. N_2O

The climate sensitivity of oceanic N_2O outgassing and the resulting feedbacks remain poorly understood. Changes in the marine N_2O cycle reflect variations in the balance between net production rates, transport and accumulation in the water column, and air–sea fluxes (Wilson et al., 2020; Yang et al., 2020). Ultimately, oceanic N_2O production is tied (via nitrification and denitrification; Section 2.2.1) to remineralization of organic matter produced in surface layers. Thus, future oceanic N_2O production will likely respond directly to changes in surface primary production and organic matter export. Declines in primary production and export in the low- to mid-latitude ocean, projected as a result of surface warming and increased stratification (Bopp et al., 2013; Kwiatkowski et al., 2020), are likely to reduce N_2O sources from remineralization pathways. However, future projections of primary production are still uncertain, with recent Earth system model simulations suggesting that expansion of nitrogen fixation in the low-latitude ocean may help to stabilize productivity across the 21st century (Bopp et al., 2022). Furthermore, changes in the nitrogen and carbon stoichiometry of newly produced organic matter (Bopp et al., 2022) may have cascading effects on N_2O sources that are difficult to assess without dedicated studies.

In contrast, ongoing oxygen loss (Bopp et al., 2013; Schmidtko et al., 2017) is likely to increase N_2O production from both nitrification and denitrification, given their high yield at low oxygen concentrations (Nevison et al., 2003; Babbin et al., 2015; Ji et al., 2015). Meanwhile, shrinking volumes of anoxic waters in the tropical ocean (Deutsch et al., 2014; Cabré et al., 2015; Busecke et al., 2022) would reduce N_2O consumption by denitrification. Thus, projected changes in oxygen concentrations could partially offset the effects—albeit uncertain—of declining remineralization on N_2O production (Martinez-Rey et al., 2015; Landolfi et al., 2017; Battaglia and Joos, 2018).

Strengthening of oceanic stratification following ocean warming is expected to curtail mixing and ventilation pathways (Gnanadesikan et al., 2007; Bopp et al., 2013) leading to N_2O accumulation and increasing inventories in the ocean interior (Martinez-Rey et al., 2015). Superposed to these changes, increasing atmospheric N_2O mole fractions from human emissions (Tian et al., 2021) are expected to lead to a global reduction in N_2O fluxes to the atmosphere. However, because of the relatively slower atmospheric N_2O increase, this perturbation of the ocean N_2O content is presumably minor compared to the case of CO_2. In parallel, warming-driven decrease in N_2O solubility and shrinking sea ice in polar regions would strengthen N_2O outgassing, with regional modulation by changes in wind speed and gas-transfer velocity.

Taken together, reduction in net production rates, accumulation in the ocean interior, and the increase in N_2O mole fractions are likely to reduce oceanic N_2O outgassing, counteracting the effects of oxygen loss, and providing a negative (albeit weak) feedback on climate (Martinez-Rey et al., 2015). However, because N_2O emissions reflect interaction of tightly coupled biogeochemical and physical processes (Berthet et al., 2023), these projections and the resulting climate feedbacks remain uncertain. Better constraining them will require model improvements, expanded observational syntheses, and coordinated assessments similar to those carried out for CO_2 (Gruber et al., 2023).

3.3. Other gases

The climate sensitivity of oceanic cycling and outgassing of CH_4, CO, and NO as well as the resulting feedbacks are largely unknown because our knowledge about the major production and consumption processes and their major drivers is rudimentary. The complex interactions of physical processes (e.g., air–sea gas exchange and mixing), on the one side, and the various (micro)biological processes (e.g., methanogenesis, microbial oxidation, production by phytoplankton), and chemical reactions (e.g., photochemical production) on the other side hamper a simple model parameterization of the gas production/consumption pathways. Therefore, model studies for CH_4 and NO are largely missing, and only a few model studies on CO have been published (Conte et al., 2019; Kwon et al., 2020).

A key component seems to be dissolved and particulate organic matter, which is directly involved in CH_4 and CO production and indirectly involved in NO production (Sections 2.3–2.5). The occurrence and fluxes of organic matter, in turn, are directly related to biological productivity in the ocean or riverine input to coastal regions. CH_4 production in the ocean was modelled as a flux from sinking organic particles (Nihous and Masutani, 2006). Comparable with N_2O (Section 3.2), the projected declines in organic matter export and remineralization are thus likely to reduce CH_4 production from particle-associated methanogenesis. Ongoing environmental changes, such as ocean warming, eutrophication and deoxygenation, very likely will affect future oceanic cycling of CH_4, CO, and NO. For the time being, however, how and to what extent these environmental changes will be affected by climate change in the future remains unclear.

4. Social and global relevance and perspectives

Despite recent results from modelling studies suggesting that climate change has reduced the ocean CO_2 sink globally by approximately 4% in the last decade, this small change cannot be discerned by surface observations, with oceans continuing to be responsible for the absorption of 25% of anthropogenic emissions from 1960 to 2021 (Friedlingstein et al., 2022). This ocean CO_2 sink serves human society by buying time to reduce emissions and limit global climate change. Without the oceanic CO_2 uptake, the atmospheric CO_2 mole fraction would have already exceeded 500 ppm. The downside of the anthropogenic oceanic CO_2 sink is that it alters the ocean chemical composition. These changes lead to ocean acidification, aragonite undersaturation, and the depletion of the buffering capacity of CO_2, which pose a threat to marine ecosystem habitability and the role of the ocean as a sink for CO_2 (McNeil and Matear, 2008; Kwiatkowski and Orr, 2018; Gruber et al., 2021; Heinze et al., 2021; Cooley et al., 2022) and could potentially result in crossing tipping points (Lenton et al., 2008) caused by the cumulative impact of warming, acidification, and deoxygenation. These effects may be fragmented both regionally and in time but add up to global dimensions, particularly in combination with other threats such as exploitation, mining, pollution, habitat destruction, overfishing and farming, invasive species, and shipping, with the potential to cause ecosystem-wide regime shifts (Heinze et al., 2021). Impacts are already observed on biodiversity, ecosystem functioning, and services (FAO, 2020; Cooley et al., 2022; Santana-Falcón and Séférian, 2022). Past and future changes in biogeochemical variables are leading large marine areas towards environmental conditions not experienced in the present or recent climate (Williams and Jackson, 2007; Blenckner et al., 2021), as illustrated by Santana-Falcón and Séférian (2022), who concluded that by 2100 concomitant changes in both lower and upper boundaries can expose pelagic ecosystems to thermal environments never experienced before.

Over this century, ocean heat and carbon uptake will continue, but the ocean may take up a smaller fraction of CO_2 emissions as atmospheric CO_2 concentrations rise (Armstrong et al., 2019; Gulev et al., 2021; Müller et al., 2023). At the same time as carbon cycling and CO_2 storage are being, and continue to be, affected by climate change (Müller et al., 2023), interventions to enhance ocean carbon storage are being explored for mitigation (Shukla et al., 2022; Bach et al., n.d.; Johnson et al., n.d.). The global community has set forth ambitions for a net-zero emission society by 2050. The global average air–sea CO_2 fluxes have a smaller variability than CO_2 fluxes associated with the land biosphere (**Figure 3**). Nevertheless, this smaller variability hides the fact that the net ocean CO_2 sink exhibits large regional variability and non-negligible interannual-to-decadal variability, and even a relatively small but persistent change in the efficiency will affect the net-zero target (**Figure 3**). The processes driving these variabilities are still not fully understood, which is highlighted by the large (approximately 1 Gt C yr^{-1}) discrepancy between estimates based on global circulation models and estimates based on gap-filled observational data products (Friedlingstein et al., 2022; Gruber et al., 2023). Quantifying air–sea exchange of CO_2 to the degree necessary will require large resources. If we are to rely on the ocean to help us reach net-zero emissions, then we need to put effort into a truly global estimate of net GHG exchange. Data from different measuring networks need to be connected together more efficiently to maximize benefits from all the data collected. We need all the smaller parts in order to see the big picture.

Human adaptation to minimize harm and exploit opportunities from climate-driven ocean changes is already taking place. Adaptation through nature-based solutions, including marine protected areas and habitat restoration, and carbon dioxide removal (CDR) initiatives can bring social, economic, and cultural co-benefits, but these are constrained by the degree of mitigation action

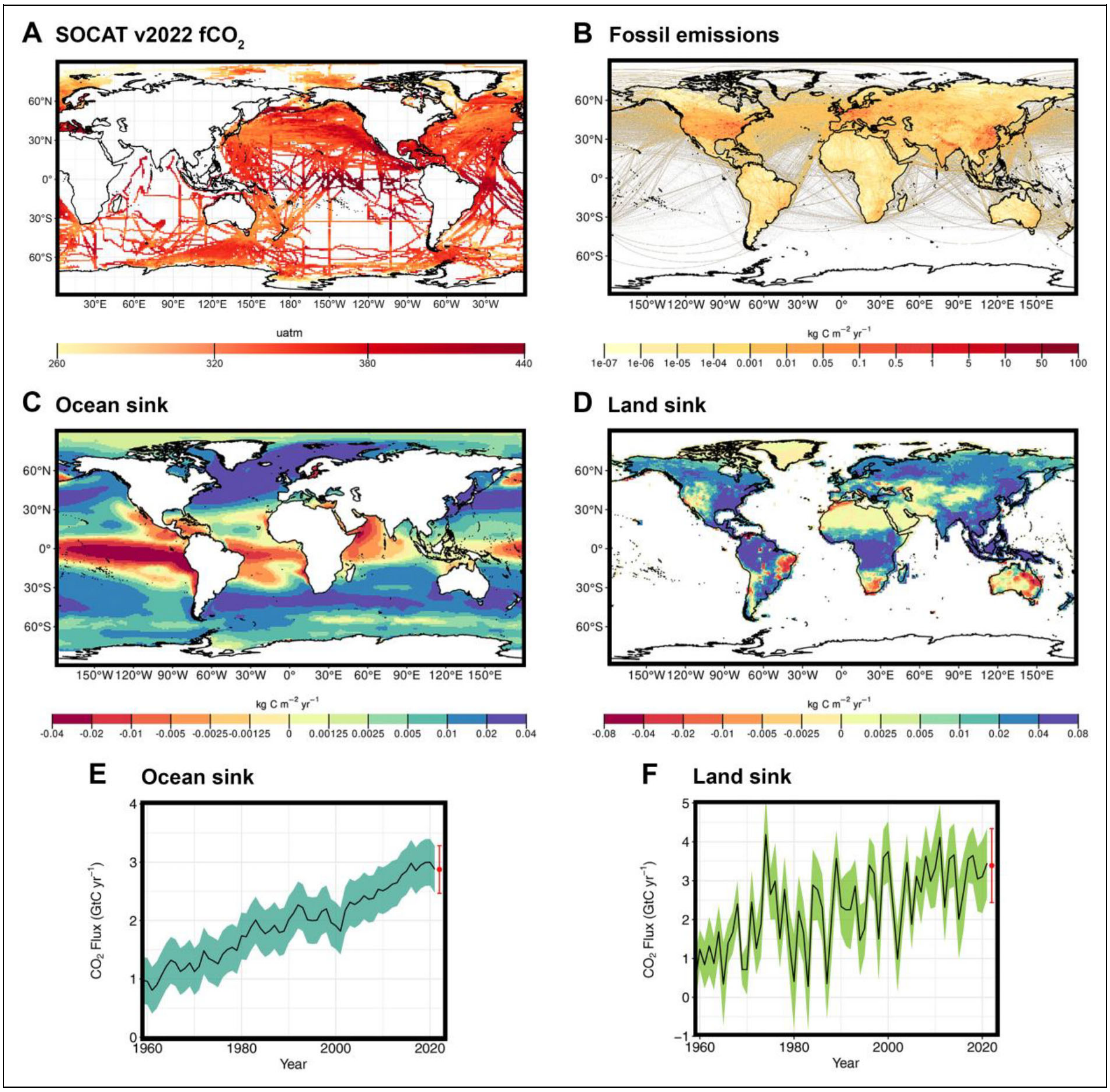

Figure 3. Spatial distributions and temporal evolutions of the ocean and land CO_2 sinks for 2012–2021. (A) fCO_2 measurements (µatm) from the Surface Ocean CO_2 Atlas (SOCAT) v2022; (B) fossil CO_2 emissions (kg C m^{-2} yr^{-1}); (C) ocean CO_2 sink (kg C m^{-2} yr^{-1}); (D) land CO_2 sink (kg C m^{-2} yr^{-1}); (E) temporal evolution of the ocean CO_2 sink (Gt C yr^{-1}); and (F) temporal evolution of the land CO_2 sink (Gt C yr^{-1}). In panels C and D negative values (red and yellow colors) represent a release to the atmosphere, whereas positive values (green and blue colors) represent an uptake from the atmosphere. In panels E and F the uncertainty bounds representing ±1 standard deviation are given in shaded color. Note the different scales in each panel. Panels are from Friedlingstein et al. (2022), with permission.

and investment in the necessary technological innovation (Shukla et al., 2022; Bach et al., n.d.; Johnson et al., n.d.). A recent review by Williamson and Gattuso (2022) concluded that CDR using coastal blue carbon restoration has questionable cost-effectiveness for climate mitigation due to different issues, among which are lateral carbon transport, fluxes of CH_4 and N_2O, and carbonate formation and dissolution. For example, coastal wetlands have been discussed as a natural climate solution due to their capacity to sequester large amounts of blue carbon in their sediments (Gattuso et al., 2021; Macreadie et al., 2021). However, CH_4 emissions can offset some of the carbon sink capacity in coastal wetlands (Rosentreter et al., 2018; Al-Haj and Fulweiler, 2020), thereby complicating blue carbon assessments (Rosentreter et al., 2021a; Williamson and Gattuso, 2022). Such offsets, however, are currently highly uncertain and also need to incorporate N_2O fluxes. Only recently, near-shore coastal CH_4 and N_2O emissions have been estimated to offset the coastal CO_2 sink by 9%–20% globally (Rosentreter et al., 2023).

Looking forward, there is enormous potential benefit to society in understanding the environmental consequences of climate change in general and ocean-atmosphere interactions with GHG in particular (Garçon et al., 2014). The main environmental metrics employed to date in relation to climate change mitigation are atmospheric GHG mole fractions and global mean surface temperatures. However, more refined mitigation metrics need to be developed that consider potential changes in the physical climate system and the biogeochemical properties of the ocean, such as ocean oxygen content, marine net primary productivity or upper ocean metabolic index (Heinze et al., 2021; Tran et al., 2021). Societal decision-making must consider surface ocean and lower atmosphere interactions (e.g., through GHG uptake/emissions) and how these are linked to human societies and social processes of change (Weaver et al., 2014). This knowledge needs to be integrated into societal decision-making and communicated effectively to the public through adequate tools and processes. Essential in this process is the role of social sciences in elucidating the processes that turn knowledge into action, that is, knowledge that is practically relevant, usable, legitimate, and actionable.

Humans living on land need the atmosphere. The vast open ocean is and has always been far removed from human lives; nevertheless, it is the largest part of our planetary biosphere and, while having great inertia, it is not immutable. Great strides have been made in the past several decades to properly observe how the ocean and the air–sea interactions of GHG are changing. However, the ocean remains severely undersampled and lacks, for example, observational data from the Southern Hemisphere (**Figure 3**), from wintertime, and from the subsurface. Process level understanding and quantification of oceanic GHG fluxes is critical in order to fulfil the Paris Agreement mitigation targets and secure adaptation solutions that support the social, economic, and environmental United Nations' Sustainable Development Goals (SDGs: https://sdgs.un.org/goals; van Doorn et al., n.d.). Integrated planning, financing, and implementation can help to overcome challenges and ensure that marine adaptations do not compromise overall human equity or specific SDGs but are fully synergistic with these goals (Nash et al., 2021).

5. Summary and outlook

This study set out to review advances in surface ocean air–sea fluxes and GHG cycling in the upper ocean (0–1000 m) during the past 10 years, focusing on CO_2, N_2O, CH_4, CO, and NO_x. Overall, we find that significant advances have been made in understanding mechanistic pathways, and reducing uncertainties in measurements and flux estimates. The global ocean is a large and increasing sink for CO_2, a major source of N_2O, and a minor source of CH_4 and CO (**Table 2**). Estimates of the oceanic sources and sinks of NO_x are missing. The amount of CO_2 taken up by the open and coastal oceans is well known, and the estimates are associated with a relatively small uncertainty compared to the emission estimates of the other GHG (**Table 2**). This is mainly resulting from the fact the number of available surface ocean pCO_2 measurements is much larger than the number of available surface concentration measurements of the other GHG: The latest version of SOCAT (2022; https://socat.info) has 35.6 million observations from 1957 to 2022 for the open and coastal oceans (**Figure 3**), whereas the latest version of the MarinE MethanE and NiTrous Oxide database (MEMENTO version 2021; https://memento.geomar.de) has archived about 150,000 N_2O and CH_4 concentration data.

Emissions from coastal areas contribute a large fraction (N_2O, CO) to the overall annual oceanic emissions or even dominate (CH_4) the global emissions (**Table 2**). Except for CO_2, the oceanic cycling of GHG is mainly driven by microbial production and consumption pathways (**Table 2**). These microbial pathways and photochemical production, in turn, depend directly (e.g., denitrification, CO photoproduction) or indirectly (e.g., methanogenesis, nitrification)

Table 2. Summary of emissions (positive values) and uptake (negative values) of greenhouse gases with their major production and consumption processes in the open and coastal oceans and estuaries (see Section 2 for details)

Location or Process	CO_2 (Pg C yr^{-1})	N_2O (Tg N yr^{-1})	CH_4 (Tg C yr^{-1})	CO (Tg C yr^{-1})	NO_x[a] (Tg N yr^{-1})
Open ocean	-2.5[b] (***)[c]	3.5[b] (***)	0.8 (***)	3.8 (**)[c]	? (*)[c]
Coastal ocean	-0.4 (***)	0.7 (**)	7.8[d] (**)	1.2 (**)	? (*)
Estuaries	0.03 (***)	0.04 (**)	0.2 (**)	? (*)	? (*)
Production	Microbial remineralization of organic matter	Microbial nitrification, denitrification	Microbial production	Photochemistry	NO: photochemistry, microbial N cycling; NO_2: unknown
Consumption	Photosynthesis	Microbial denitrification	Microbial oxidation	Microbial uptake	Chemical reactions

[a] NO + NO_2.

[b] Calculated as the total oceanic emissions minus emissions from coastal areas (as given in Sections 2.2.1 and 2.3.1).

[c] Level of confidence: unknown or very low (*), low (**) or high (***).

[d] Includes diffusive and ebullitive emissions from continental shelves (as given in Section 2.3.1).

Table 3. Summary of expected changes[a] and level of understanding[b] of the changes in CO_2 uptake and N_2O, CH_4, CO, and NO emissions from open and coastal oceans in view of ongoing environmental changes

Environmental Changes	Consequences or Potential Linkages	CO_2	N_2O	CH_4	CO	NO
Warming	Decrease of solubility	− (***)	+ (***)	+ (***)	+ (***)	+ (***)
	Changes in biological production, community composition	± (**)	? (*)	? (*)	? (*)	? (*)
	Changes in microbial production/consumption pathways	− (**)	± (**)	± (**)	− (**)	? (*)
	Enhanced stratification, leading to reduced mixing, deoxygenation of subsurface waters	− (**)	± (**)	+ (**)	? (*)	? (*)
Acidification	Changes in biological production, community composition	± (**)	± (**)	? (*)	± (**)	? (*)
Eutrophication[c]	Enhanced biological production leading to enhanced sinking of organic material, enhanced organic matter decomposition, deoxygenation of subsurface/bottom waters	± (***)	+ (**)	+ (**)	? (*)	? (*)
	Enhanced biological production leading to enhanced CDOM concentrations, enhanced photochemistry	− (**)	? (*)	+ (**)	+ (***)	na
Deoxygenation	Shift from aerobic to anaerobic organic matter decomposition in subsurface/bottom waters	− (**)	± (***)	+ (**)	? (*)	? (*)
Pollution	Increasing amount of plastics	? (*)	+ (**)	+ (**)	? (*)	na

[a] Increase (+), decrease (−), unknown (?), or not applicable (na).

[b] Level of understanding: unknown or very low (*), low (**), or high (***).

[c] Including nutrient inputs by rivers, atmospheric deposition and submarine groundwater discharge.

on biological productivity by photosynthesis in the sunlit surface layer of the open and coastal oceans.

The ocean's continuing ability to absorb CO_2 means that observation-based estimates of this absorption, based upon existing and new understanding (and resulting from activities of the Surface Ocean-Lower Atmosphere Study, SOLAS) will continue to be critically important for global and regional carbon assessments. Furthermore, machine learning–based gaps filling products using ship- and buoy-collected ocean pCO_2, satellite observations and reanalysis outputs are playing an increasingly crucial role in improving ocean CO_2 estimates. The synthesis of CO_2 measurements by SOCAT has been and will be of significant help in ensuring that the global community maintains centralized standards in quality control and the sharing of CO_2 measurements. The role of weather and climate, as well as cooling associated with volcanic eruptions, are postulated as drivers of interannual variability for global air–sea CO_2 fluxes. The most extensive air–sea CO_2 flux interannual variability occurs in the Southern Ocean and the equatorial regions of the tropical Pacific Ocean, where CO_2 measurements and mechanistic insights of drivers remain the least understood. To this end, the introduction of biogeochemical Argo floats, especially through the Southern Ocean Carbon and Climate Observations and Modelling project, the use of autonomous surface vehicles and the continuing of ship-based measurements play crucial roles in improving CO_2 measurements in the Southern Ocean.

Improvements in measurement techniques and increased availability of datasets have reduced the uncertainty of N_2O, CH_4, and CO emission estimates during the last years. Traditionally, their oceanic emissions were thought to be determined largely by the balance of microbial (N_2O, CH_4) or photochemical (CO) production and consumption processes. However, recently published findings have questioned this conventional thinking; for example, there is increasing evidence that N_2O can be consumed in the oxic ocean surface layer. The "oceanic methane paradox," despite a bundle of potential explanations, remains enigmatic. The role of phytoplankton as a source of N_2O, CH_4, and CO and the molecular mechanisms associated with the photochemical production of CH_4, CO, and NO remain unclear and require further evaluation. Additional sources such as N_2O and CH_4 production associated with marine microplastics (recently reviewed by Goddijn-Murphy et al., 2023) and submerged aquatic vegetation remain poorly characterized as well. We can thus conclude that our knowledge of the production, consumption, and emissions of N_2O, CH_4, CO, and NO from the ocean is still far from complete. In addition to these persistent gaps in knowledge, ongoing environmental changes, such as ocean warming (and associated changes in stratification and ice coverage), ocean acidification, deoxygenation, and eutrophication due to increasing anthropogenic inputs of nutrients via rivers and atmospheric deposition of aerosols, might significantly alter the production and consumption of N_2O, CH_4, CO, and NO, their distribution patterns and, ultimately, their release to the atmosphere (**Table 3**). As a consequence, ocean biogeochemical models fail to provide an adequate representation of reality, which in turn affects projections of the distributions and emissions of these gases under future scenarios of ocean warming, acidification, deoxygenation, and eutrophication (**Table 3**).

In order to improve data coverage and standardize measurements, MEMENTO was launched and, on the basis of a first method inter-comparison (Wilson et al., 2018), standard measurement protocols for N_2O and CH_4 have been published. Unfortunately, MEMENTO does not yet include data or protocols for CO and NO_x. The importance of additional, routine oceanic N_2O measurements has been recognized by the Global Ocean Observing System program, which recently added N_2O to its list of Essential Ocean Variables (www.goosocean.org). Building on these efforts the Global N_2O Ocean Observation Network was proposed (Bange et al., 2019a). Although this network was originally designed for N_2O only, adding measurements of CH_4, CO, and NO_x will be facilitated by deploying instruments on the basis of the same technique used for N_2O measurements (i.e., cavity-enhanced absorption spectroscopy). Those advances will enable us to improve estimates of oceanic N_2O, CH_4, CO, and NO emissions. However, they only tackle part of the problem. In order to address existing knowledge gaps and fully decipher the production and consumption pathways of these gases in a changing ocean (**Tables 2** and **3**), complementary approaches to a global observation network are needed, as suggested in Bange (2022). Joint process studies should be established in open ocean sites in all major ocean basins and coastal ocean regions with high biological productivity and/or anthropogenic influence, conducted across multiple seasons and years, and designed holistically, combining microbial, molecular biological and biogeochemical and atmospheric methods. Such a comprehensive, coordinated, and interdisciplinary approach will result in the data necessary to identify the relevant phytoplankton and microbial communities, quantify the rates of oceanic N_2O, CH_4, CO, and NO production/consumption pathways, and identify their major drivers. In combination with the continuous collection of data from routinely operated observation networks, these process data will help to establish or improve the parameterizations of GHG cycling and emissions in models (Séférian et al., 2020).

Finally, studies to detect early significant changes of ocean acidification, deoxygenation, eutrophication, and aragonite saturation in marine ecosystems will be necessary to optimize conservation and mitigation strategies. A holistic approach that considers scientific collaboration and measurement collection as well as consideration of the cultural and economic implications of mitigation solutions for GHG emissions will be necessary for navigating the Anthropocene on a global scale.

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Competing interests

There are no competing interests.

Author contributions

HWB, PM and JDS coordinated the writing of the article. All authors contributed to the writing of the article and approved its submission.

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