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CARBON CYCLE

Strong Southern Ocean carbon uptake evident in airborne observations

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The Southern Ocean plays an important role in determining atmospheric carbon dioxide (CO2), yet estimates of air-sea CO2 flux for the region diverge widely. In this study, we constrained Southern Ocean air-sea CO2 exchange by relating fluxes to horizontal and vertical CO2 gradients in atmospheric transport models and applying atmospheric observations of these gradients to estimate fluxes. Aircraft-based measurements of the vertical atmospheric CO2 gradient provide robust flux constraints. We found an annual mean flux of $-0.53 \pm 0.23$ petagrams of carbon per year (net uptake) south of 45°S during the period 2009–2018. This is consistent with the mean of atmospheric inversion estimates and surface-ocean partial pressure of CO2 (Pco2)-based products, but our data indicate stronger annual mean uptake than suggested by recent interpretations of profiling float observations.

Oceano column carbon inventories suggest that the Southern Ocean accounts for more than 40% of the cumulative global ocean uptake of anthropogenic CO2 (J, 2). However, estimates of contemporary net Southern Ocean air-sea CO2 exchanges based on surface-ocean partial pressure of CO2 (Pco2) observations or atmospheric inversions remain highly uncertain (3–8). Recent interpretations of profiling float observations have introduced further complications, proposing a profound revision of the Southern Ocean carbon budget, with reduced summertime uptake and larger wintertime outgassing (9, 10). Given the Southern Ocean’s critical role as a sink for anthropogenic CO2, as well as indications that regional fluxes vary substantially on decadal time scales (7, 11, 12), it is essential to develop more robust constraints on Southern Ocean air-sea CO2 exchange. Observations of atmospheric CO2 provide an opportunity for doing so, as the atmosphere effectively integrates signal fluxes over large surface regions. Atmospheric inversion models provide a formal statistical method to estimate fluxes that optimally satisfy atmospheric observational constraints, given circulation simulated by data-constrained atmospheric transport models (4, 13, 14). However, global-scale atmospheric inversion models have not converged on consistent Southern Ocean fluxes, as they suffer from inaccuracies in the simulated transport, reliance on uncertain “prior” flux estimates, and requirements to meet tighter constraints elsewhere in the world, where signals are stronger and measurements less sparse (4, 13–17).

In this study, we derived “emergent constraints” on regional air-sea CO2 fluxes by relating fluxes in a collection of models to observable gradients in CO2 in the atmosphere directly overlying the Southern Ocean. We used observations from nine deployments of three recent aircraft projects: the HIAPER Pole-to-Pole Observations (HIPPO) project (18), the O3/N2O Ratio and CO2 Airborne Southern Ocean (ORCAS) study (19), and the Atmospheric Tomography (Atm) mission (20) (see supplementary materials, hereafter SM). We also examined 44 atmospheric CO2 records from surface monitoring stations in the high-latitude Southern Hemisphere, selecting and filtering the highest-quality data (SM). Collectively, these observations show a distinct pattern in the seasonal variability of atmospheric CO2 overlying the Southern Ocean, most notably characterized by lower-troposphere CO2 depletion in austral summer and neutral to weakly positive enhancement in austral winter (Figs. 1 and 2, A to C). To isolate CO2 gradients driven by Southern Ocean fluxes, we examined CO2 anomalies relative to a local reference, using potential temperature (θ) to delineate boundaries in the vertical dimension (SM). We defined a metric quantifying the vertical CO2 gradient, $\Delta q_{CO2}$, as the difference between the median value of CO2 observed south of 45°S, where $\theta < 280$ K, and that in the mid- to upper-troposphere, where $295 < \theta < 305$ K. The aircraft observations suggest that the amplitude of seasonal variation in CO2 is minimized within this upper θ range relative to the rest of the column (fig. S7); it is also above the vertical extent of wintertime, near-surface homogeneity (Fig. 2A) and below altitudes substantially influenced by the stratosphere, making it a good reference for detecting regional air-sea flux signals (see SM). Similarly, we defined a metric of the horizontal surface gradient, $\Delta q_{CO2}$, as the difference between CO2 averaged across stations in the core latitudes of summertime CO2 drawdown (Fig. 1, C and D, shaded region) and that at the South Pole Observatory (SPO). $\Delta q_{CO2}$ is strongly negative in the austral summer, followed by near-neutral conditions in the austral winter through spring (Fig. 2B). Correspondingly, $\Delta q_{CO2}$ also indicates summertime drawdown at the surface and weakly positive to near-neutral conditions in winter (Fig. 2C), although the amplitude of seasonal variation in $\Delta q_{CO2}$ is more than three times larger than that in $\Delta q_{CO2}$. Variation in drawdown intensity across stations contributing to $\Delta q_{CO2}$ likely reflects differential sampling of air exposed to strong ocean productivity signals (fig. S4).

We developed inferences about austral summer CO2 fluxes from these gradient metrics by examining a collection of atmospheric inverse models that simulate time-varying, three-dimensional CO2 fields sampled to replicate observations (SM). The inverse models demonstrate that seasonality in $\Delta q_{CO2}$ and $\Delta q_{CO2}$ is dominated by Southern Ocean air-sea fluxes. Although land and fossil fuel fluxes are small south of 45°S, extraregional contributions do influence local gradients via transport from the north. The models explicitly simulate CO2 tracers responsive to ocean (CO2,20) and land (CO2,20), and fossil fuel (CO2,20) fluxes and subject to identical transport fields. The simulations of these tracers indicate that the influence of land fluxes generally opposes the effect of fossil fuel emissions for both gradient metrics, and the seasonality in the land and fossil fuel tracers is much weaker than the ocean-derived signal (Fig. 2, D and E, and fig. S6). The negative vertical (positive horizontal) gradient in fossil fuel CO2 is consistent with elevated CO2 concentrations in the equatorward portion of the
domain, particularly at high altitude (Fig. 1, A and B, and figs. S2 to S4). Ancillary measurements of methane-mixing ratios confirm that this feature reflects long-range transport of emission signals from land, but that it has little influence on $\Delta_q CO_2$ (figs. S6 and S8). Additional evidence that fossil fuel emissions make only small contributions to the annual mean and seasonality in $\Delta_q CO_2$ comes from ancillary observations of sulfur hexafluoride—which provides an analog for fossil fuel CO2 (2f) and shows very little spatial or temporal structure over the Southern Ocean (fig. S9).

To develop quantitative flux estimates, we related simulated $\Delta_q CO_2$ and $\Delta_q CO_2$ to regionally integrated, temporally averaged air-sea flux in each modeling system (Fig. 3). In addition to inverse models, we included forward atmospheric transport integrations forced with spatially explicit surface-ocean $PCO_2$-based flux datasets (SM). Ultimately, each model realization was a forward simulation producing three-dimensional CO2 fields from which we computed gradient metrics consistent with the model’s surface fluxes and atmospheric transport. The relationship between the fluxes and simulated gradient metrics across the collection of models enabled using the observed gradients to constrain Southern Ocean fluxes. We assumed that the relevant surface-influence region can be approximated as the area south of a particular latitude and focused on fluxes integrated over the region south of 45°S, noting that the flux products indicate strong meridional gradients and seasonality in the zonal mean fluxes south of 30°S (fig. S18, A and B). We averaged the regional fluxes over individual seasons to regress against the surface mole fraction observations and over 90 days before each aircraft campaign (see SM for sensitivity tests, including an assessment of different region boundaries and a similar analysis based on gradients in total CO2). There is a robust relationship between the CO2 flux south of 45°S and $\Delta_q CO_2$ across the models (Fig. 3, A and B). The sensitivity of $\Delta_q CO_2$ to fluxes varies seasonally, as indicated by a change in slope between seasons. December to February (DJF) is distinct in having a smaller slope (higher sensitivity); the other seasons individually have larger slopes that are similar to each other, thus we grouped data from campaigns flown in March to November together (Fig. 3B). For the surface data, we find a significant positive relationship between the regional air-sea flux and $\Delta_q CO_2$ in DJF across the models (Fig. 3C); the flux-$\Delta_q CO_2$ relationship dwindles in strength during nonsummer months, however, and there is no significant relationship in austral winter [June to August (JJA)] (Fig. 3D). The spread enabling these relationships results from the diversity of flux estimates, while the scatter about the fits represents both different realizations of atmospheric transport and spatiotemporal mismatch between the true surface influence function and our coarse spatiotemporal approximation. The smaller slope for the aircraft data in DJF is consistent with greater atmospheric stability (reduced vertical mixing) over the cold ocean during austral winter, intensifying the flux signal in the lower troposphere; more-energetic vertical mixing in other seasons, as well as stronger horizontal flow, results in diminished sensitivity in $\Delta_q CO_2$ and no clear relationship...
between fluxes and the surface station–based $\Delta CO_2$ metric in winter.

Vertical lines in Fig. 3 show representative observations of each gradient metric corrected for land and fossil fuel contributions; the intersection of these lines with the flux-gradient fit provides a quantitative flux estimate. Applying this emergent constraint for each aircraft campaign yields 10 flux estimates spread over 7 months of the year; these data suggest that the Southern Ocean is a strong sink for CO2 in austral summer, with fluxes that are near-neutral during winter (Fig. 4A).

Applying a two-harmonic fit to the data, we estimated an annual mean flux spread over the aircraft observing period (2009–2018) of −0.53 ± 0.23 petagrams of carbon (Pg C) year$^{-1}$ (Fig. 4B). The seasonal cycle of fluxes estimated from aircraft campaigns largely agrees with flux estimates derived from the Surface Ocean CO2 Atlas (SOCAT) $P_{CO_2}$ data product, using either neural network interpolation (22) or the Jena mixed-layer scheme (23) (Fig. 4A). Similarly, the aircraft-based fluxes agree with the multimodel mean of inverse estimates, although inversions tend to underestimate summer uptake (Fig. S12D), and show greater than 100% disagreement on the annual mean flux. We have not explicitly accounted for interannual variability or trends in the fluxes over the period of aircraft data collection (Fig. S12, C and D), although we expect this to be a relatively small effect, as seasonal coverage between HIPPO and ATom is relatively uniform (Fig. 4A). The flux estimates obtained from the surface atmosphere CO2 gradient in summer are consistent with the aircraft-based estimates (fig. S12C) but have larger uncertainty—indeed, the magnitude of the $\Delta CO_2$ signal is small relative to analytical uncertainty (SM), a particular challenge in this region, where sites are remote, conditions are harsh, and intercomparison between the multiple laboratories maintaining CO2 records is limited (24, 25). Despite the large uncertainty, however, trends in $\Delta CO_2$ are consistent with increasing Southern Ocean uptake since 2005 (7, 26) (see SM); for instance, $\Delta CO_2$ declined by about 0.16 ± 0.03 parts per million (ppm) decade$^{-1}$ over the period 2005–2019 for both DJF and JJA, and while there was no significant flux-gradient relationship in JJA (Fig. 3D), the associated $\Delta CO_2$-based flux estimates suggest the DJF flux was $-0.5 ± 0.7$ Pg C year$^{-1}$ from 2005 to 2009, $-1.1 ± 0.9$ Pg C year$^{-1}$ from 2010 to 2014, and $-1.3 ± 1.1$ Pg C year$^{-1}$ from 2015 to 2019 (fig. S12C).

Notably, the aircraft-based flux estimates indicate stronger annual mean uptake than fluxes incorporating $P_{CO_2}$ estimates from the Southern Ocean Carbon and Climate Observations and Modeling (SOCCOM) profiling float pH measurements (10, 27). The primary SOCCOM flux product we examined (SOCAT+SOCCOM) is derived from neural network interpolation including both ship-based surface-ocean $P_{CO_2}$ observations as well as float-derived $P_{CO_2}$ estimates [see SM and (27)]; this product yields weaker annual mean uptake but tracks the individual aircraft campaign flux estimates within uncertainty (Fig. 4A).

The other two SOCCOM-based products presented here are sensitivity runs (10, 27) that selectively exclude ship-based $P_{CO_2}$ observations in the Southern Ocean (see SM). While these products have a seasonal phase and amplitude similar to those of the aircraft flux estimates, they indicate greater outgassing in winter and less ingassing during summer than the aircraft-based fluxes (Fig. 4). Such large fluxes should have clear atmospheric signatures...
but no such signatures are evident in any of the Southern Ocean aircraft campaigns (Fig. 2, A and B, and figs. S2, S10, and S11).

Our analysis demonstrates that Southern Ocean air-sea fluxes impart a coherent pattern in atmospheric CO2 as measured by aircraft. The surface station network is only detectably sensitive to variations in fluxes during austral summer and hampered by measurement noise commensurate with flux signals. Our results highlight the difficulty global atmospheric in-

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**Fig. 3. Emergent constraints on air-sea fluxes south of 45°S.** (A and B) 90 day–mean air-sea fluxes south of 45°S versus $\Delta$CO$_2$$_{ocn}$ from model simulations (see SM) replicating aircraft observations collected during (A) December to February and (B) March to November. Colored vertical lines show an observed value of $\Delta$CO$_2$$_{ocn}$ [ORCAS during January in (A) and ATom-1 in (B)] corrected for land and fossil fuel influence, with shading indicating both analytical uncertainty and model spread in the correction (see SM); colored points highlight the model samples from these particular campaigns, while gray points show data from other campaigns in the (A) December to February or (B) March to November timeframe. Figures S10 and S11 show similar plots for each individual aircraft campaign. (C and D) Seasonal-mean surface fluxes versus $\Delta$CO$_2$$_{ocn}$ computed from models for (C) summer (DJF) and (D) winter (JJA) over the period 1999–2019. Points correspond to individual models; whiskers denote the standard deviation of interannual variability. Light blue vertical lines show the observed $\Delta$CO$_2$$_{ocn}$ corrected for land and fossil fuel influence; shading shows analytical uncertainty and model spread in the correction (see SM; fig. S12, A and B, shows $\Delta$CO$_2$$_{ocn}$ time series). The sign convention for fluxes is positive upward. Diagonal lines, where significant, show the best-fit line to all data points shown; inset text shows an estimate of the slope with standard error (SM), and goodness-of-fit statistics are also shown. Table S3 provides detailed information on the model products, defining the acronyms used in the legend. Note that the axis bounds differ by panel. See fig. S16 for a version of this plot based on total CO$_2$.

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or unidentified transport biases common across models. The collection of models we included use four different underlying meteorological reanalysis datasets, four different transport models, and differ in spatial resolution and treatment of vertical transport (table S3); moreover, they make up a substantial proportion of the models commonly used for long-term global CO₂ inversions. However, inclusion of additional model solutions would improve confidence in our result by increasing the number of independent realizations of transport. Despite this potential limitation, aircraft observations leverage the broad integrative power of the atmosphere, which provides an advantage over estimating fluxes from surface ocean \( P_{CO_2} \) observations: The ocean surface is heterogeneous, making representative sampling difficult; air-sea fluxes computed from \( P_{CO_2} \) estimates depend on an uncertain gas exchange parameterization (28); and float-based estimates have additional uncertainty associated with estimating \( P_{CO_2} \) itself (29). However, we resolved fluxes only over a broadly defined Southern Ocean region; finer-scale spatial features present in surface-ocean \( P_{CO_2} \) data can provide important mechanistic insight, reinforcing the need for more high-quality, widely distributed ocean observations to advance process understanding. Uncertainty regarding Southern Ocean carbon uptake is a critical limitation in current understanding of the global carbon cycle (30). Our results can be used to validate Earth system models and inversion-based assessments of the Southern Hemisphere carbon budget. Critical challenge, though, is that the atmospheric \( CO_2 \) budget require balanced fluxes; therefore, our result of strong Southern Ocean uptake alleviates the need to identify missing Southern Hemisphere land or subtropical ocean sinks, as suggested by the float observations. Finally, our analysis has important implications for effective monitoring of the Southern Ocean carbon sink. A regular program of aircraft observations could provide a cost-effective approach to drastically improve estimates of the carbon budget for the Southern Ocean and globally, helping to fulfill a societal requirement for clear understanding of mechanisms driving variation in atmospheric \( CO_2 \).

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
Figs. S1 to S19
Tables S1 to S4
References (40–83)

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Strong Southern Ocean carbon uptake evident in airborne observations

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Up in the air
Understanding ocean-atmospheric carbon dioxide (CO₂) fluxes in the Southern Ocean is necessary for quantifying the global CO₂ budget, but measurements in the harsh conditions there make collecting good data difficult, so a quantitative picture still is out of reach. Long et al. present measurements of atmospheric CO₂ concentrations made by aircraft and show that the annual net flux of carbon into the ocean south of 45°S is large, with stronger summertime uptake and less wintertime outgassing than other recent observations have indicated. —HJS

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