#### **OCEANOGRAPHY**

# Southern Ocean anthropogenic carbon sink constrained by sea surface salinity

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The ocean attenuates global warming by taking up about one quarter of global anthropogenic carbon emissions. Around 40% of this carbon sink is located in the Southern Ocean. However, Earth system models struggle to reproduce the Southern Ocean circulation and carbon fluxes. We identify a tight relationship across two multimodel ensembles between present-day sea surface salinity in the subtropical-polar frontal zone and the anthropogenic carbon sink in the Southern Ocean. Observations and model results constrain the cumulative Southern Ocean sink over 1850-2100 to  $158 \pm 6$  petagrams of carbon under the low-emissions scenario Shared Socioeconomic Pathway 1-2.6 (SSP1-2.6) and to  $279 \pm 14$  petagrams of carbon under the high-emissions scenario SSP5-8.5. The constrained anthropogenic carbon sink is 14 to 18% larger and 46 to 54% less uncertain than estimated by the unconstrained estimates. The identified constraint demonstrates the importance of the freshwater cycle for the Southern Ocean circulation and carbon cycle.

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#### **INTRODUCTION**

Anthropogenic carbon ( $C_{\rm ant}$ ) uptake by the ocean is playing a crucial role in slowing global warming. Since 1850, the global ocean has taken up between 20 and 30% (160  $\pm$  20 Pg of C) of  $C_{\rm ant}$  emissions from fossil fuel combustion, cement production, and land-use change (1, 2). Around 40% of this global ocean  $C_{\rm ant}$  uptake has taken place in the Southern Ocean south of 30°S (3–9), making this particular region the largest oceanic sink of  $C_{\rm ant}$ .

Observation-based (5–8) and model (9–13) estimates of the Southern Ocean C<sub>ant</sub> uptake come with large uncertainties (Fig. 1). The uncertainty in observation-based estimates is mainly caused by data scarcity, especially during wintertime (14-16). The uncertainty among models can be attributed to shortcomings in simulating the complex circulation of the Southern Ocean (11, 17-19). Observation-based cumulative Southern Ocean Cant fluxes scaled to the period from 1850 to 2005 (see Materials and Methods) range from 40 to 71 Pg of C (5, 7, 8), while simulated C<sub>ant</sub> uptake over the same period ranges from 44 to 63 Pg of C (intermodel range) when using the Earth system models (ESMs) from phase 5 of the Coupled Model Intercomparison Project (CMIP5; fig. S1A) (9) and from 43 to 62 Pg of C when using the new CMIP6 model generation (Fig. 1A and Table 1). By 2100, the projected range grows to 194 to 279 Pg of C under the high emissions scenario for CMIP5 [Representative Concentration Pathway 8.5 (RCP8.5) (20)] and to 204 to 309 Pg of C under the high emissions scenario for CMIP6 [Shared Socioeconomic Pathway 5-8.5 (SSP5-8.5) (21)]. Thus, the new generation of CMIP6 ESMs has, despite further model development and overall increased horizontal and vertical ocean model resolution (22), not reduced the uncertainties in Southern Ocean Cant uptake.

In cases of large projection uncertainties, such as for Southern Ocean  $C_{ant}$  uptake, the use of emergent constraints offers an opportunity to reduce these uncertainties (23–25). Emergent constraints first relate model projections of a particular variable to observable historical trends, sensitivities, or base states of the same or different

variable across an ESM ensemble and then exploit this relationship with observations of the historical trend, sensitivity, or base state. The fidelity of an emergent constraint, thus, depends on the correlation of the relationship and the uncertainty of the observations. Emergent constraints have been applied to constrain many climate-related variables, such as transient (26, 27) and equilibrium climate sensitivity (28, 29), Arctic snow albedo feedback (24), carbon cycle feedbacks (30, 31), marine primary production (32), and Arctic Ocean acidification (33). However, as emergent constraints may conflict with one another (28, 34) and can even be derived from data-mined pseudo-correlations (35), it is essential to understand and demonstrate the mechanisms that underpin them and to test the identified emergent constraint in an independent model ensemble (25, 36).

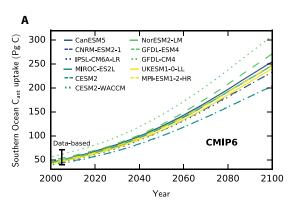
In this study, we develop and apply an emergent constraint to reduce uncertainties in the Southern Ocean sink of  $C_{ant}$ . The constraint relies on observed sea surface salinity. The constraint is supported by physical and biogeochemical process understanding, presented for the CMIP6 ensemble, and confirmed using the CMIP5 ensemble.

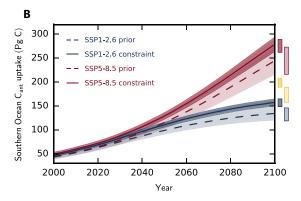
## Southern Ocean circulation and Cant sink

The important role of the Southern Ocean in the global C<sub>ant</sub> uptake is due to its unique circulation (12, 37). Westerly winds drive a strongly divergent surface flow that allows old Circumpolar Deep Water carrying little C<sub>ant</sub> to come back to the surface ocean at the polar front (PF), the southern limit of the Antarctic Circumpolar Current (12, 37, 38). Only a small fraction of upwelled and surface waters moves southward and is converted into Antarctic Bottom Water (12, 38). The largest fraction flows through Ekman transport northward while taking up large amounts of Cant via air-sea gas exchange. Eventually, these northward flowing water masses are transformed to Antarctic Intermediate Water (AAIW) and Subantarctic Mode Water (SAMW) (38) via surface heat uptake and mixing with southward flowing water from the mid-latitudes (15). AAIW and SAMW, now enriched in Cant, are then subducted in the vicinity of the subtropical front (STF) below the light subtropical waters into the deeper ocean (38). The amount of Cant taken up in the Southern Ocean is, thus, primarily dictated by the rate of SAMW and AAIW

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**Fig. 1. Projections of cumulative Southern Ocean C**<sub>ant</sub> **uptake in CMIP6.** ESM projections of the 21st century Southern Ocean (>30°S) cumulative C<sub>ant</sub> uptake since 1850 from (**A**) 11 CMIP6 models following the SSP5-8.5 scenario (21) with the data-based estimate for cumulative Southern Ocean uptake from 1850 to 2005 (black vertical line) (5, 7, 8). (**B**) Time series of the multimodel mean C<sub>ant</sub> uptake under the SSP1-2.6 (blue) and SSP5-8.5 (red) scenarios with ±1 SD for the CMIP6 model ensembles before (transparent) and after the emergent constraint is applied (opaque). The bars indicate the range of ±1 SD of the cumulative C<sub>ant</sub> uptake in 2100 under SSP1-2.6 (blue), SSP2-4.5 (yellow), and SSP5-8.5 (red) before (transparent) and after (opaque) the constraint is applied.

Table 1. Unconstrained (prior) and constrained (after constraint) cumulative Southern Ocean  $C_{ant}$  uptake (petagrams of C) in CMIP5 and CMIP6\*.

Simulations	Prior	After constraint
CMIP6		
Historical		
1850–2005	49 ± 5	55 ± 3
1850–2014	58 ± 6	65 ± 4
SSP1-2.6		
1850–2100	134 ± 13	158±6
2015–2100	78 ± 10	96 ± 4
SSP2-4.5	••••	
1850–2100	173 ± 16	200 ± 8
2015–2100	117 ± 12	138±6
SSP5-8.5		
1850–2100	245 ± 28	278 ± 13
2015–2100	187 ± 22	214±11
CMIP5 <sup>†</sup>	••••	
Historical	•••••	••••
1850-2005	51 ± 5	53 ± 4
RCP2.6	•	•
1850–2100	138 ± 10	142±6
2006–2100	86 ± 8	88 ± 7
RCP4.5		
1850–2100	174 ± 15	183±7
2006–2100	122 ± 12	129±7
RCP8.5		
1850–2100	241 ± 24	253 ± 9
2006–2100	190 ± 21	200 ± 10

\*Not all models were available for all future scenarios so that historical and future estimates do not always add up exactly to the estimates of the full period. TGFDL-ESM2M and GFDL-ESM2G simulations started in 1861.

formation with other factors, such as the buffer capacity of surface waters and the air-sea equilibration time of  $CO_2$ , being less important (3, 4, 9, 19, 39, 40).

The total subduction rate of SAMW and AAIW depends on the density of the northward flowing surface waters (41, 42) and on the density structure of the upper ocean (12, 17). Heavier surface waters, corresponding to larger outcrop areas of SAMW and AAIW (fig. S1C) (41), have the potential to penetrate deeper and to occupy a larger volume below the surface than lighter waters (12, 42). Similarly, surface waters penetrate more easily when density increases weakly with depth than in a strongly stratified ocean. Across the CMIP6 and CMIP5 model ensembles the volume of ocean interior water ventilated by surface waters that lies between the PF and the STF, namely, SAMW and AAIW, increases with increasing sea surface density ( $r^2 = 0.74$ ; figs. S2 to S5). Sea surface density is, thus, a physically supported indicator of the formation rate of SAMW and AAIW (12, 41, 43, 44) and, in turn, of  $C_{ant}$  uptake by the Southern Ocean (12, 41).

The sea surface density variations in the cold Southern Ocean depends strongly on variations in surface salinity ( $r^2 = 0.84$ ; fig. S2A) (13, 42–45) and less on variations in surface temperature ( $r^2 = 0.01$ ; fig. S2B). The salinity at the surface is influenced by the hydrological balance of evaporation minus precipitation, sea and land ice melt, and net salinity transport into the surface layers via circulation. This balance of complex and intricate processes and, in turn, surface salinity and density is difficult to represent correctly in models (10, 17, 18, 46). In CMIP5 models, for example, the Southern Ocean surface waters were found to have on average a fresh bias (fig. S2A) (10, 17, 18), but the model spread is substantial. A negative salinity bias is possibly caused by too much precipitation (47), too little subsurface inflow of Circumpolar Deep Water into the Southern Ocean (17), too weak and equatorward displaced winds that therefore do not create enough upwelling (48), and a too large decline of the sea ice extent over the past decades (46). Nevertheless, the outlined mechanistic explanation supported by the above presented model results suggests that sea surface salinity in the Southern Ocean is a strong and well observable indicator of SAMW and AAIW formation and, hence, Cant uptake.

#### **RESULTS**

# Emergent relationship between Southern Ocean sea surface salinity and C<sub>ant</sub> uptake

A tight relationship between Southern Ocean sea surface salinity and C<sub>ant</sub> uptake is found for the CMIP5 and CMIP6 models. ESMs, such as the Community Earth System Model version 2 (CESM2), that simulate low mean surface salinities (and densities) between the PF and the STF also simulate a small Cant uptake in the Southern Ocean south of 30°S, while ESMs that simulate high salinities, such as the Geophysical Fluid Dynamics Laboratory's CM4.0 physical climate model (GFDL-CM4), also simulate a high Cant uptake (Fig. 2 and figs. S6 and S7). We, therefore, expect and find a strong correlation between the cumulative Cant uptake and the mean of present-day sea surface salinity between the PF and STF averaged over 1986-2005. This correlation holds across the two latest model generations (CMIP6 and CMIP5) and for every year of the 21st century (Fig. 3 and figs. S8 and S9, A and B). This relation yields, when combined with observations of sea surface salinity, a so-called emergent constraint (11, 36) on the cumulative  $C_{ant}$  uptake in the Southern Ocean across 11 ESMs from CMIP6 (Fig. 3) and across 13 ESMs from CMIP5 (fig. S8).

Southern Ocean cumulative  $C_{ant}$  uptake since 1850 was calculated for each ESM as the difference between the cumulative air-sea  $CO_2$  flux south of 30°S between the historical and future simulation and the corresponding preindustrial control (pi-Control) simulation. This definition of  $C_{ant}$  uptake includes the flux driven by increasing atmospheric  $CO_2$  concentrations and the changes in the natural air-sea  $CO_2$  flux due to climate change (e.g., warming and changes in circulation and biology). The PF and STF were identified for each

month and each longitude by the maximum temperature gradients based on the sea surface temperature over 1986–2005 for each model (49–51) (see Materials and Methods and fig. S10) to compute mean surface salinity from monthly data for this frontal region. One ensemble member was used per ESM as internal model variability is small compared to the differences between models (see Materials and Methods). The relationship between the cumulative  $C_{\rm ant}$  uptake and the mean surface salinity between the PF and STF was calculated using a linear regression with equal weight for each model. The constrained estimate of the cumulative  $C_{\rm ant}$  uptake is then calculated as the normalized product of the conditional probability density functions (PDFs) of the emergent relationship and the observations assuming Gaussian distributions.

With this emergent constraint, the Southern Ocean cumulative  $C_{\rm ant}$  uptake from 1850 to 2005 of the CMIP6 model ensemble changes from 49  $\pm$  5 to 55  $\pm$  3 Pg of C ( $r^2$  = 0.74; Table 1 and Fig. 3) (the uncertainty refers to  $\pm$ 1 SD, and  $r^2$  is the coefficient of determination between simulated sea surface salinity and  $C_{\rm ant}$  uptake). The unconstrained and constrained mean estimates are significantly different (Student's t test, 5% significance level). The constrained mean estimate is also closer to the central value of the observation-based estimates of 58  $\pm$  13 Pg of C (5, 7) and 54  $\pm$  14 Pg of C (8) over the same period.

# Constrained cumulative C<sub>ant</sub> uptake from 1850 to 2100 in CMIP6

After applying the emergent constraint to the cumulative Southern Ocean  $C_{ant}$  uptake from 1850 to 2100, the uptake changes from

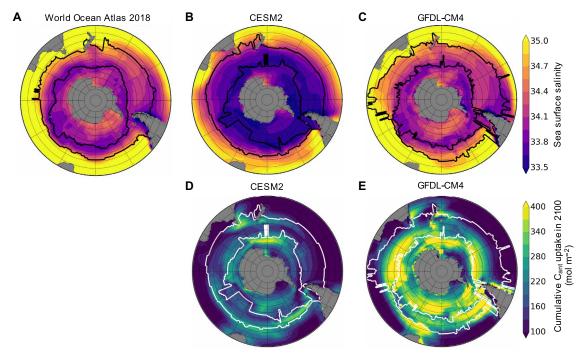
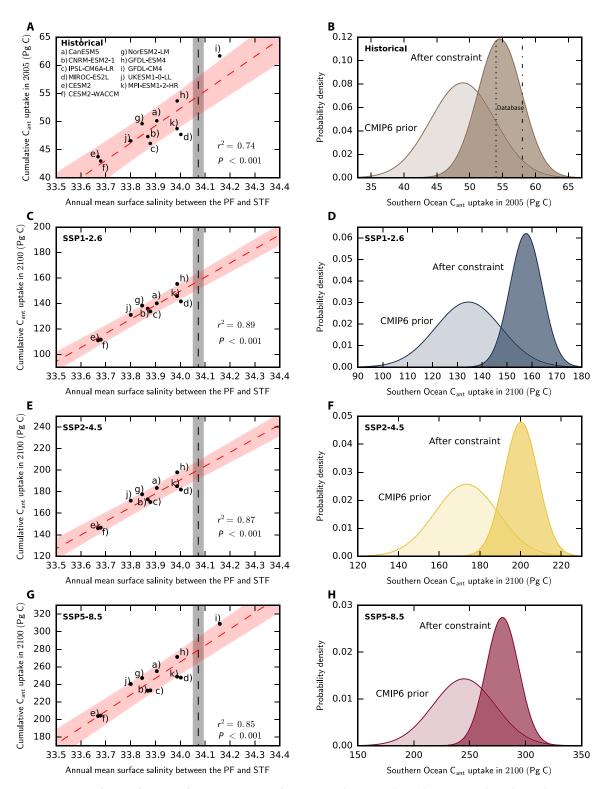


Fig. 2. Sea surface salinity and cumulative C<sub>ant</sub> uptake in the Southern Ocean. (A) Present-day sea surface salinity in August from World Ocean Atlas 2018 (80) and the sea surface salinity in August averaged over 1986–2005 simulated by the (B) CESM2 and (C) GFDL-CM4 models. Both models are part of CMIP6. Black and white contour lines delineate the PF and the STF in August. Simulated cumulative Southern Ocean (>30°S) C<sub>ant</sub> uptake over 1850–2100 for the (D) CESM2 and (E) GFDL-CM4 models. CESM2 is the minimum of the CMIP6 ensemble for both present-day (1986–2005) mean sea surface salinity between the PF and STF (33.67) and projected cumulative C<sub>ant</sub> uptake in 2100 (204 Pg of C), while GFDL-CM4 is the ensemble maximum (34.16 and 309 Pg of C). The observation-based mean sea surface salinity between the PF and STF is 34.07 [World Ocean Atlas 2018 (80)].



**Fig. 3. Emergent constraints on the cumulative Southern Ocean C**<sub>ant</sub> **uptake in CMIP6.** The projected cumulative C<sub>ant</sub> uptake in the Southern Ocean south of 30°S across the CMIP6 model ensemble for (**A**) the historical period from 1850 to 2005, and the historical and future period from 1850 to 2100 under (**C**) SSP1-2.6, (**E**) SSP2-4.5, and (**G**) SSP5-8.5 against the present-day (1986–2005) mean sea surface salinity between the PF and STF. Linear regression fits (red dashed lines) and the associated 68% prediction intervals are shown in (A), (C), (E), and (G), as are observation-based estimates of present-day annual sea surface density between the PF and STF (black dashed lines) with the associated uncertainty (black shaded area). Probability density functions for the cumulative Southern Ocean C<sub>ant</sub> uptake from (**B**) 1850 to 2005 and from 1850 to 2100 under (**D**) SSP1-2.6, (**F**) SSP2-4.5, and (**H**) SSP5-8.5, before ("CMIP6 prior," transparent) and after ("after constraint," opaque) the emergent constraint is applied. Data-based estimates are shown in (B) as dash-dotted (*5*, *7*) and dotted (*8*) lines and are scaled to the same definition of C<sub>ant</sub> as the models (see Materials and Methods).

 $134 \pm 13$  to  $158 \pm 6$  Pg of C ( $r^2 = 0.89$ ; Table 1 and Fig. 3) under the low emissions SSP1-2.6 scenario. For the SSP2-4.5 scenario with moderate CO<sub>2</sub> emissions, which peak in the middle of the 21st century and decrease afterward, the Southern Ocean cumulative Cant uptake changes from 173  $\pm$  16 to 200  $\pm$  8 Pg of C ( $r^2 = 0.87$ ). Under the high emissions scenario SSP5-8.5, the Southern Ocean cumulative  $C_{ant}$  uptake changes from  $244 \pm 28$  to  $278 \pm 13$  Pg of  $C(r^2 = 0.85)$ . Thus, the uncertainty of the estimated cumulative C<sub>ant</sub> uptake in 2100 is reduced by 46 to 54% across these three CMIP6 scenarios, while the estimated uptake itself is increased by 14 to 18%. Across the CMIP6 ensemble, 10 of 11 ESMs underestimate the annual sea surface salinity between the PF and STF and, therefore, also the uptake of C<sub>ant</sub>. By correcting for this systematic sea surface salinity bias, the constrained Southern Ocean cumulative  $C_{ant}$  uptake in 2100 is significantly (Student's t test, 5% significance level) larger than the unconstrained uptake in each scenario.

#### **Comparison to CMIP5**

To test the robustness of the emergent constraint, we further applied the constraint to the CMIP5 model ensemble. In this ensemble, the cumulative Southern Ocean  $C_{ant}$  uptake from 1850 to 2100 changes from 138  $\pm$  10 to 142  $\pm$  6 Pg of C under the low emissions RCP2.6 scenario ( $r^2 = 0.77$ ), from 173  $\pm$  16 to 183  $\pm$  8 Pg of C under the moderate emissions RCP4.5 scenario ( $r^2 = 0.88$ ), and from 241  $\pm$  24 to 253  $\pm$  9 Pg of C under the high emissions RCP8.5 scenario ( $r^2 = 0.90$ ) (fig. S8).

As for the CMIP6 model ensemble, the uncertainty of the constrained estimates is approximately half (40 to 63%) of the uncertainty of the unconstrained estimate. The increase in the best estimate in the constrained results of the CMIP5 ensemble, however, is with 3 to 6% not significant (Student's *t* test, 5% significance level) and smaller than the relative increase in the CMIP6 model ensemble (14 to 18%). This smaller change in the best estimate is due to a smaller model-averaged mean salinity bias in CMIP5 (0.11) than in CMIP6 (0.18).

The Southern Ocean C<sub>ant</sub> uptake is generally smaller for RCPs than for their SSP counterparts. This difference is to a large extent caused by higher prescribed atmospheric CO<sub>2</sub> trajectories over the 21st century in the SSP than in the RCP scenarios, due to different energy and land use assumptions (20, 21). In the high emissions scenarios, for example, prescribed anthropogenic CO<sub>2</sub> in the atmosphere is, on average over the 21st century, around 15% larger in SSP5-8.5 than in RCP8.5 (52, 53). This explains to a large extent why the constrained cumulative Southern Ocean C<sub>ant</sub> uptake from 2005 to 2100 is 12% larger for SSP5-8.5 than for RCP8.5 (Table 1).

#### Extending the constraint to Southern Ocean acidification?

Our emergent constraint suggests that the Southern Ocean may take up more  $C_{ant}$  than estimated by the CMIP6 model mean. However, enhanced  $C_{ant}$  uptake may also cause stronger ocean acidification (54), i.e., a decrease in pH and in the saturation state of seawater relative to the calcium carbonate mineral aragonite ( $\Omega_{arag}$ ) and calcite ( $\Omega_{calc}$ ). These changes in seawater chemistry have been shown to negatively affect keystone aragonite and calcite shell-forming species and other marine organisms (55–57).

By extending the emergent constraint approach from  $C_{ant}$  uptake to ocean acidification at different depth levels (33) in the Southern Ocean, we project slightly greater ocean acidification across the CMIP6 model ensemble north of the STF between 30°S and 40°S, where most of the subducted AAIW and SAMW are located (fig. S11). In

waters between 300 and 1500 m, end-of-century  $\Omega_{arag}$  under SSP5-8.5 projected by the CMIP6 model ensemble is reduced by the constraint from 0.78  $\pm$  0.06 to 0.74  $\pm$  0.06, and  $\Omega_{calc}$  is reduced by the constraint from 1.22  $\pm$  0.09 to 1.16  $\pm$  0.10. Constrained end-of-century  $\Omega_{arag}$  and  $\Omega_{calc}$  projections are only significantly different (Student's t test, 5%) from the unconstrained projections between 300 and 600 m. Below 1500 m where SAMW and AAIW do not occur (17), there is no relationship between sea surface salinity between the PF and STF and end-of-century  $\Omega_{arag}$  and  $\Omega_{calc}$ .

Compared to the emergent constraint on cumulative Southern Ocean  $C_{ant}$ , constraining  $\Omega_{arag}$  and  $\Omega_{calc}$  with sea surface salinity does not reduce the uncertainties in  $\Omega_{arag}$  and  $\Omega_{calc}$  and only very slightly adjusts the best estimate. While sea surface salinity was shown to constrain the  $C_{ant}$  uptake from the atmosphere, it does not constrain other processes that influence  $\Omega_{arag}$  and  $\Omega_{calc}$ , such as the interior ocean  $C_{ant}$  transport from the Southern Ocean to the subtropics, which varies strongly across models (9), and changes in alkalinity, nutrients, temperature, and salinity.

#### DISCUSSION

#### **Potential limitations**

Use of emergent constraints has limits when important processes for the identified relationship are not included or poorly represented in ESMs (25). In this section, the possible role of mesoscale eddies, freshwater input from Antarctic ice melt, acceleration of the Southern Ocean meridional overturning circulation, and biological processes are discussed.

Mesoscale eddies in the Southern Ocean influence the transport of tracers, such as heat, salinity, carbon, and nutrients (58-61). However, the explicit simulation of these mesoscale eddies requires high horizontal and vertical ocean model resolutions, especially in high latitudes such as the Southern Ocean (62). Most of the CMIP5 and CMIP6 models use ocean models with horizontal resolution of about 1° (22, 63). To date, conducting transient simulations with fully coupled ESMs in higher resolution is computationally too expensive, especially because these simulations also need a sufficiently long spin-up to reach a stable equilibrium (64, 65). Therefore, the effect of eddies on the mean ocean circulation and the transport of ocean tracers, such as salinity and carbon, are parametrized within the CMIP models. While the eddy parametrization has an effect on the simulated sea surface salinity and Cant uptake (58-61), this effect cannot be quantified by the state-of-the art CMIP6 ESMs due to their relatively coarse resolution and merits further investigation when eddy-resolving ocean models incorporated in global coupled ESMs will become more widely available.

Furthermore, changes in freshwater input from Antarctic ice melt are not included in any of these models. This freshwater input has the potential to further reduce sea surface salinity and, thus, the uptake of  $C_{\rm ant}$  in the Southern Ocean. Although the effect of land ice melt on the sea surface salinity between the PF and STF in recent decades (estimated to be <0.05 over 1995–2015) (66) is small compared to the present day (1986–2005) intermodel variability (0.8; Fig. 3 and fig. S2A), more research is needed to quantify the effect of land ice melt over the 21st century.

Another observed process that is not well represented by the models is the acceleration of the upper cell of the Southern Ocean meridional overturning circulation in the past decades (67). This acceleration and potential further acceleration in the future might

reduce the residence time of waters at the surface below the around 10 months that are necessary for surface ocean partial pressure of  $\rm CO_2$  to equilibrate with the atmosphere and, thus, potentially reduce the uptake of  $\rm C_{ant}$  from the atmosphere (15).

In addition, our emergent constraint is purely physical and does not account for the representation of biological processes and their potential changes over the 21st century, which varies largely among these models (68, 69). However, recent studies (70, 71) have suggested that biological processes, in comparison to ocean physics, currently play a minor role for the oceanic C<sub>ant</sub> uptake despite their importance for natural CO<sub>2</sub> air-sea fluxes (72).

# "Confirmed" emergent constraint

The development and increasing use of emergent constraints in recent years not only led to major advances in the understanding of the Earth system but also led to conflicting results, i.e., emergent constraints for the same projected variable, but different observable variables led to different results (28, 34). Moreover, it is shown that in a large dataset such as the CMIP dataset, emergent constraints can be derived even from data-mined pseudo-correlations (35). To avoid spurious constraints, Hall *et al.* (25) propose three criteria for emergent constraints to be confirmed. These criteria are (i) a plausible mechanism behind the emergent relationship, (ii) verification of the proposed mechanism, (iii) and out-of-sample testing. All three criteria are met in our analysis.

First, we proposed a plausible underlying mechanism, namely, that the Southern Ocean sea surface salinity determines the amount of SAMW and AAIW that is subducted below the surface ocean and thereby the transport of  $C_{\rm ant}$  from the surface to the subsurface ocean. The more  $C_{\rm ant}$  is removed from the surface ocean, the more  $C_{\rm ant}$  can be taken up from the atmosphere via air-sea  $CO_2$  flux. Second, we verified this mechanism by demonstrating that the salinity in the frontal region is related to the outcrop area of SAMW and AAIW and to the volume of ventilated waters in the subsurface Southern Ocean and to  $C_{\rm ant}$  uptake (figs. S2 to S5). Third, we applied out of sampling testing across a different ESM ensemble (CMIP5). Although models in new CMIP generations are not strictly independent from their predecessors from which they were developed, this out-of-sample testing in a previous model generation is regarded as useful evidence of an underlying emergent constraint (25).

While the relationship between the volume of subducted SAMW and AAIW and the Southern Ocean C<sub>ant</sub> uptake might be more direct, we chose the sea surface salinity as the observable quantity because its observations are less uncertain. Sea surface salinity provides the best compromise between a good linear correlation and low observational uncertainties.

Our results do not directly constrain the  $C_{ant}$  uptake of the global ocean. The Southern Ocean  $C_{ant}$  uptake over 1850–2100 south of 30°S is not correlated to the ocean  $C_{ant}$  sink north of 30°S ( $r^2$  = 0.07), i.e., a weak or strong Southern Ocean  $C_{ant}$  sink is not systematically compensated by a strong or weak  $C_{ant}$  sink north of 30°S. Thus, intermodel differences in the  $C_{ant}$  uptake north of 30°S remain unchanged.

#### The importance of the Southern Ocean freshwater cycle

Our results suggest that the simulation of sea surface salinity and of the cumulative Southern Ocean  $C_{\rm ant}$  uptake are in better agreement with observations in the CMIP5 model ensemble than in the new CMIP6 model ensemble. This is unexpected as the ocean's vertical and horizontal resolution is increased from CMIP5 to CMIP6 in

almost all ESMs (22) and the representation of the wind forcing and sea surface temperature in the Southern Ocean is improved (73). However, large biases with respect to the freshwater cycle and the sea surface salinity remain or even increased from CMIP5 to CMIP6. We show that it is indeed sea surface salinity and the freshwater cycle in the Southern Ocean that is crucial for simulating the Southern Ocean circulation (74, 75) and the associated ocean  $C_{\rm ant}$  uptake (45, 76). An improved ability to properly simulate the Southern Ocean freshwater cycle is urgently needed to pin down one of the largest uncertainties in projections of the fate of  $C_{\rm ant}$  and the climate.

#### **MATERIALS AND METHODS**

#### Earth system models

We used 24 ESMs in this study, 11 from the CMIP6 (table S1), and 13 from the CMIP5. All models include coupled ocean biogeochemistry schemes and have been applied within the context of both climate and ocean biogeochemical projections (68, 69). A single ensemble member of the concentration-driven simulations was used for each ESM (ensemble member 1 was used when available, otherwise ensemble member 2 was used). All model simulations cover the period 1850 to 2100 (1861-2100 for the GFDL-ESM2G and GFDL-ESM2M, respectively) following historical greenhouse gas and aerosol and natural forcing changes over the period 1850-2005 (CMIP5) or 1850-2014 (CMIP6). For CMIP5 models, the simulations follow RCP2.6, RCP4.5, and RCP8.5 over 2006-2100, and for CMIP6, they follow SSP1-2.6, SSP2-4.5, and SSP5-8.5 over 2015-2100. GFDL-CM4 output was only available for SSP5-8.5, Centro Euro-Mediterraneo sui Cambiamenti Climatici - Community Earth System Model (CMCC-CESM) output only for RCP8.5, and Hadley Centre Global Environment Model version 2 - Carbon Cycle (HadGEM2-CC) and Community Earth System Model version 1-Biogeochemistry (CESM1-BGC) output only for RCP4.5 and RCP8.5.

Simulated annual air-sea  $CO_2$  flux and monthly temperature and salinity output fields south of 30°S were used. All these output fields were analyzed on the native model grid. The anthropogenic air-sea  $CO_2$  flux was calculated as the difference between the air-sea  $CO_2$  flux in historical simulations merged with the future simulations and the concurrent pi-Control simulations. Hence, any model drift was directly accounted for. Note that this definition of the anthropogenic air-sea  $CO_2$  flux includes both the flux driven by increasing atmospheric  $CO_2$  concentrations and any flux from changes in the natural air-sea  $CO_2$  flux. Changes in the natural air-sea  $CO_2$  flux may arise from climate change driven by anthropogenic and natural forcing and internal climate variability.

The mean Southern Ocean sea surface salinity between the PF and STF was calculated for each ESM based on a monthly climatology of sea surface temperature and salinity outputs from 1986 to 2005. Throughout the manuscript, salinity is reported on the Practical Salinity Scale. The fronts were identified for each month, longitude, and model by the maximum latitudinal temperature gradients over a temperature range from 1° to 6°C in the case of the PF and a range from 9° to 18°C in the case of the STF across the entire model ensemble (fig. S10) (49–51). The monthly area-averaged salinities between the fronts were then averaged to obtain an annual mean. Our definition of the fronts leads sometimes to large shifts in the latitudinal position (fig. S10). However, our results are insensitive to the definition of the fronts. Even if the fronts are chosen over a wider range of isotherms, the emergent constraint remains robust (table S2).

 $\Omega_{arag}$  and  $\Omega_{calc}$  were calculated offline from simulated dissolved inorganic carbon, total alkalinity, temperature, salinity, and, where available, dissolved inorganic phosphorus and silicon using mocsy2.0 (77) and the equilibrium constants recommended for best practices (78). To account for carbonate chemistry biases in the present-day mean state of the ESMs, model anomalies of all input variables relative to 2002 were added to the observation-based Global Ocean Analysis Project version 2 (GLODAPv2) observational product (79), which is normalized to the year 2002. For models without dissolved inorganic phosphorus and silicon output, anomalies were assumed to be zero. Model anomalies were corrected for potential model drift using concurrent pi-Control simulations. All grid cells with GLODAPv2 observational coverage (~96% of Southern Ocean volume) were used. Basin-wide averages between 30°S and 40°S of  $\Omega_{arag}$  and  $\Omega_{calc}$  were weighted on the basis of grid cell volumes.

#### The role of internal model variability

The influence of the model internal variability on the detected emergent constraint was assessed using the first four ensemble members of the Institut Pierre-Simon Laplace - Climate Model 6A - Low Resolution (IPSL-CM6A-LR). The internal variabilities of present-day annual sea surface density between the PF and STF (33.85 to 33.88) and the projected cumulative  $C_{\rm ant}$  uptake in the Southern Ocean (233.2 to 234.6 Pg of C) were found to be negligible in comparison with the range across the CMIP5 and CMIP6 model ensembles (33.56 to 34.34; 194 to 309 Pg of C).

#### **Observational constraints**

The sea surface salinity between the PF and STF was calculated identically as for the ESM ensemble but using observation-based sea surface salinities and temperatures from the World Ocean Atlas 2018 climatologies (80). The uncertainty associated with the sea surface salinity was estimated using the area-averaged SE of the sea surface salinity of the World Ocean Atlas 2018 in every surface grid cell within the region.

# Observation-based Cant fluxes

The observation-based cumulative  $C_{ant}$  fluxes from 1850 to 2005 were derived from observation-based  $C_{ant}$  fluxes for the year 1995 [1.12  $\pm$  0.26 Pg of C year  $^{-1}$  (5, 7) and 1.04  $\pm$  0.27 Pg of C year  $^{-1}$  (8)]. Following standard practice (5, 7, 8, 81), the  $C_{ant}$  flux for a year t was derived from the observation-based  $C_{ant}$  fluxes in 1995 by scaling the flux proportional to the anthropogenic  $CO_2$  concentration in the atmosphere

$$F_{\text{ant}}^{1765}(t) = F_{\text{ant}}^{1765}(1995) \frac{\Delta \text{CO}_2^{1765}(t)}{\Delta \text{CO}_2^{1765}(1995)}$$
(1)

with t being the respective year,  $F_{\rm ant}^{1765}$  being the  $C_{\rm ant}$  flux into the ocean relative to year 1765, and  $\Delta CO_2^{1765}(t)$  being the perturbation in the  $CO_2$  mixing ratio in the atmosphere relative to 1765 [e.g., for year 1995, it is 82 parts per million (ppm) = 360 ppm – 278 ppm].

CMIP5 and CMIP6 models start in the year 1850 and therefore define all  $C_{ant}$  emitted after 1850 as  $C_{ant}$ , while observation-based estimates commonly define  $C_{ant}$  as all  $C_{ant}$  emitted after 1765. This different definition of  $C_{ant}$  leads to an underestimation of  $C_{ant}$  by ESMs when compared to observation-based estimates for two reasons: The time during which the ocean takes up  $C_{ant}$  is reduced by

85 years, and the difference in the natural background  $CO_2$  concentration is around 7 ppm (82). When combined, this has been found to yield a difference in the global ocean cumulative  $C_{ant}$  uptake of 29 Pg of C by 1995 (29%) (82).

To be able to compare model results to observation-based estimates, we scaled the observation-based estimate of the  $C_{ant}$  fluxes to the  $C_{ant}$  definition that is used in the ESMs using the following equation

$$F_{\text{ant}}^{1850}(t) = F_{\text{ant}}^{1765}(1995) \frac{\Delta \text{CO}_2^{1850}(t)}{\Delta \text{CO}_2^{1765}(1995)}$$
(2)

with  $F_{\rm ant}^{1850}$  and  $F_{\rm ant}^{1765}$  being the  $C_{\rm ant}$  flux into the ocean estimated from measurements relative to the reference year 1850 and 1765, respectively,  $\Delta {\rm CO_2}^{1850}$  being the perturbation in the  ${\rm CO_2}$  mixing ratio in the atmosphere relative to 1850 as used by the ESMs, and  $\Delta {\rm CO_2}^{1765}$  being the perturbation in the  ${\rm CO_2}$  mixing ratio in the atmosphere relative to 1765 as used by the observation-based estimates. By doing so, the observation-based estimates of cumulative Southern Ocean  $C_{\rm ant}$  uptake from 1765 to 2005 are reduced by 25% for the period 1850 to 2005, e.g., from 77 to 58 Pg of C (5, 7) and from 72 to 54 Pg of C (8).

#### **Emergent constraints**

For the emergent constraints, a linear regression was calculated between the projected variable ( $C_{ant}$  uptake or  $\Omega_{arag}$  and  $\Omega_{calc}$ ) and the sea surface salinity between the PF and STF. All models were weighted equally. PDFs of the projected variable were calculated for the unconstrained (prior) ensemble and the emergent constraints. The prior PDF was derived assuming all models were equally likely and sampled from a Gaussian distribution. The constrained PDFs were calculated as the normalized product of the conditional PDF of the emergent relationship and the PDF of the observational constraint following previously established methodologies (26, 27, 29–33).

#### **SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/18/eabd5964/DC1

View/request a protocol for this paper from Bio-protocol.

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