

# *Southern Ocean drives multi-decadal atmospheric CO<sub>2</sub> rise during Heinrich Stadials*

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# Southern Ocean drives multidecadal atmospheric CO<sub>2</sub> rise during Heinrich Stadials

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The last glacial period was punctuated by cold intervals in the North Atlantic region that culminated in extensive iceberg discharge events. These cold intervals, known as Heinrich Stadials, are associated with abrupt climate shifts worldwide. Here, we present CO<sub>2</sub> measurements from the West Antarctic Ice Sheet Divide ice core across Heinrich Stadials 2 to 5 at decadal-scale resolution. Our results reveal multi-decadal-scale jumps in atmospheric CO<sub>2</sub> concentrations within each Heinrich Stadal. The largest magnitude of change ( $14.0 \pm 0.8$  ppm within  $55 \pm 10$  y) occurred during Heinrich Stadal 4. Abrupt rises in atmospheric CO<sub>2</sub> are concurrent with jumps in atmospheric CH<sub>4</sub> and abrupt changes in the water isotopologs in multiple Antarctic ice cores, the latter of which suggest rapid warming of both Antarctica and Southern Ocean vapor source regions. The synchronicity of these rapid shifts points to wind-driven upwelling of relatively warm, carbon-rich waters in the Southern Ocean, likely linked to a poleward intensification of the Southern Hemisphere westerly winds. Using an isotope-enabled atmospheric circulation model, we show that observed changes in Antarctic water isotopologs can be explained by abrupt and widespread Southern Ocean warming. Our work presents evidence for a multi-decadal- to century-scale response of the Southern Ocean to changes in atmospheric circulation, demonstrating the potential for dynamic changes in Southern Ocean biogeochemistry and circulation on human timescales. Furthermore, it suggests that anthropogenic CO<sub>2</sub> uptake in the Southern Ocean may weaken with poleward strengthening westerlies today and into the future.

ice core | paleoclimate | carbon cycle | Heinrich Stadials | carbon dioxide

The last glacial period was punctuated by millennial-scale cold intervals in the northern hemisphere (NH), referred to as stadials. A Heinrich Stadal (HS) is defined as a stadal that culminated in a rapid and extensive ice rafting event in the North Atlantic, known as a Heinrich Event (HE) (1–3). The unique impacts of these century-scale ice-rafting events and the HS they occurred in have been identified in paleoclimate records worldwide (4). A key mechanism that facilitated the global propagation of these impacts was a weakening of the Atlantic Meridional Overturning Circulation (AMOC) during NH stadials (5, 6), which amplified the interhemispheric temperature contrast through an oceanic teleconnection known as the thermal bipolar seesaw (7, 8). During all stadials (Heinrich and non-Heinrich alike), synchronous decreases in global methane (CH<sub>4</sub>) concentrations (9), rainfall decrease in the NH subtropics (10, 11), and rainfall increase in the Southern Hemisphere (SH) subtropics (12) have been linked to southward shifts in the Intertropical Convergence Zone (ITCZ) in response to northern high-latitude temperature perturbations (13, 14). Unlike non-Heinrich stadials, HSs are characterized by an abrupt rise in CH<sub>4</sub> of the order of ~10s ppb that occurred within the millennial-scale cold interval of low CH<sub>4</sub> (15). These short-lived CH<sub>4</sub> peaks within HSs coincide with maximum wet conditions in the SH subtropics (16) and are attributed to an expansion of SH subtropical wetland emissions due to a maximum southerly displacement of the ITCZ in response to a North Atlantic HE (15).

Abrupt rises in atmospheric carbon dioxide (CO<sub>2</sub>), also referred to as carbon dioxide jumps or CDJs (17), have been found during HS-1 (18), HS-4, and HS-5 (19–21). The timing of each jump is within dating uncertainty of the HE that occurred during each HS, as previously observed in high-resolution CH<sub>4</sub> records (15). For example, the beginning of HE 1 (16.2 ka B.P., where “present” is the year 1950) (22) shows a CO<sub>2</sub> jump of  $12 \pm 1$  ppm within the span of a century, coinciding with a ~40 ppb spike in CH<sub>4</sub> measured in the same ice core (15, 18). Evidence for comparable CO<sub>2</sub> jumps of ~10 ppm per century has been recently identified in older glacial periods, specifically during Marine Isotope

## Significance

Earth's climate system and carbon cycle interact in myriad ways that can add or remove CO<sub>2</sub> from the atmosphere. We use Antarctic ice cores to resolve four multi-decadal-scale CO<sub>2</sub> rises of up to 14 ppm that occurred during the most recent glacial period. These abrupt rises coincide with cold periods and iceberg discharge in the North Atlantic. Ice cores show synchronous abrupt warming in Antarctica and vapor source regions, which is consistent with increasing Southern Ocean ventilation due to shifting Southern Hemisphere westerly winds. Our results highlight past periods of dynamic changes in Southern Ocean biogeochemistry and circulation that occurred on human timescales and suggest that Southern Ocean CO<sub>2</sub> uptake may weaken as Southern Hemisphere westerlies strengthen in the future.

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The authors declare no competing interest.

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Stages 10 and 12 (17). However, current records of CO<sub>2</sub> during the last glacial period are limited by >100-y resolution (21), and therefore lack the necessary detail needed to examine the exact timing, rate, and magnitude of these rapid carbon-cycle events associated with all HSs of the last glacial period.

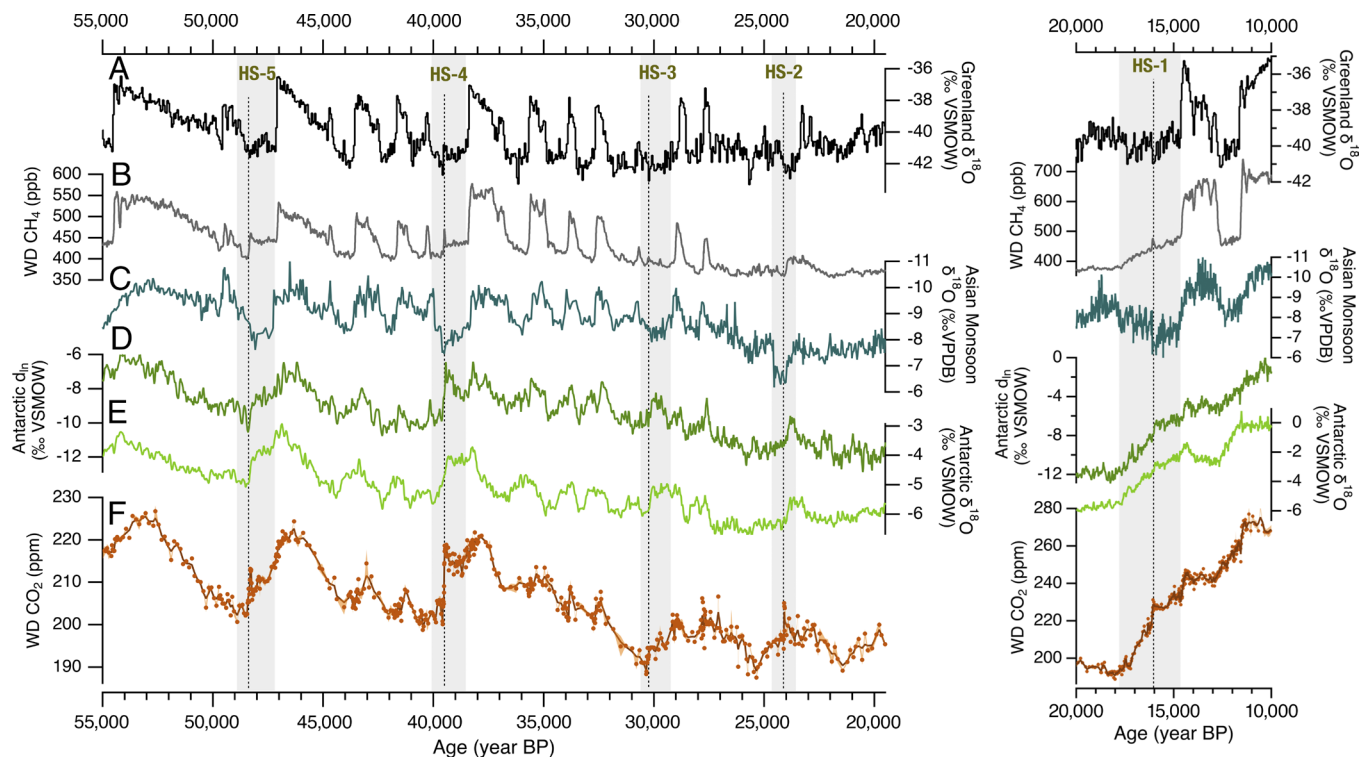
The drivers and source of these centennial-scale CO<sub>2</sub> jumps are unknown. The proposed mechanisms include enhanced ocean-atmosphere CO<sub>2</sub> exchange in the Southern Ocean (via wind-driven upwelling of carbon-rich deep waters or invigorated convection), surface ocean temperature changes (impacting CO<sub>2</sub> solubility), biomass burning (via NH subtropical wildfires), or a combination of mechanisms (18, 23–29). The ongoing debate on CO<sub>2</sub> sourcing is exemplified by a lack of consensus in Earth system and ocean models, which disagree on the sign of change from terrestrial sources and the dominant oceanic source region when forced with idealized stadial conditions (30). Improving the characterization of CO<sub>2</sub> jumps during HSs is important for identifying the mechanisms by which abrupt shifts in atmospheric and oceanic circulation may alter carbon sources and sinks, with relevance to climate change in both the past and future.

## Results

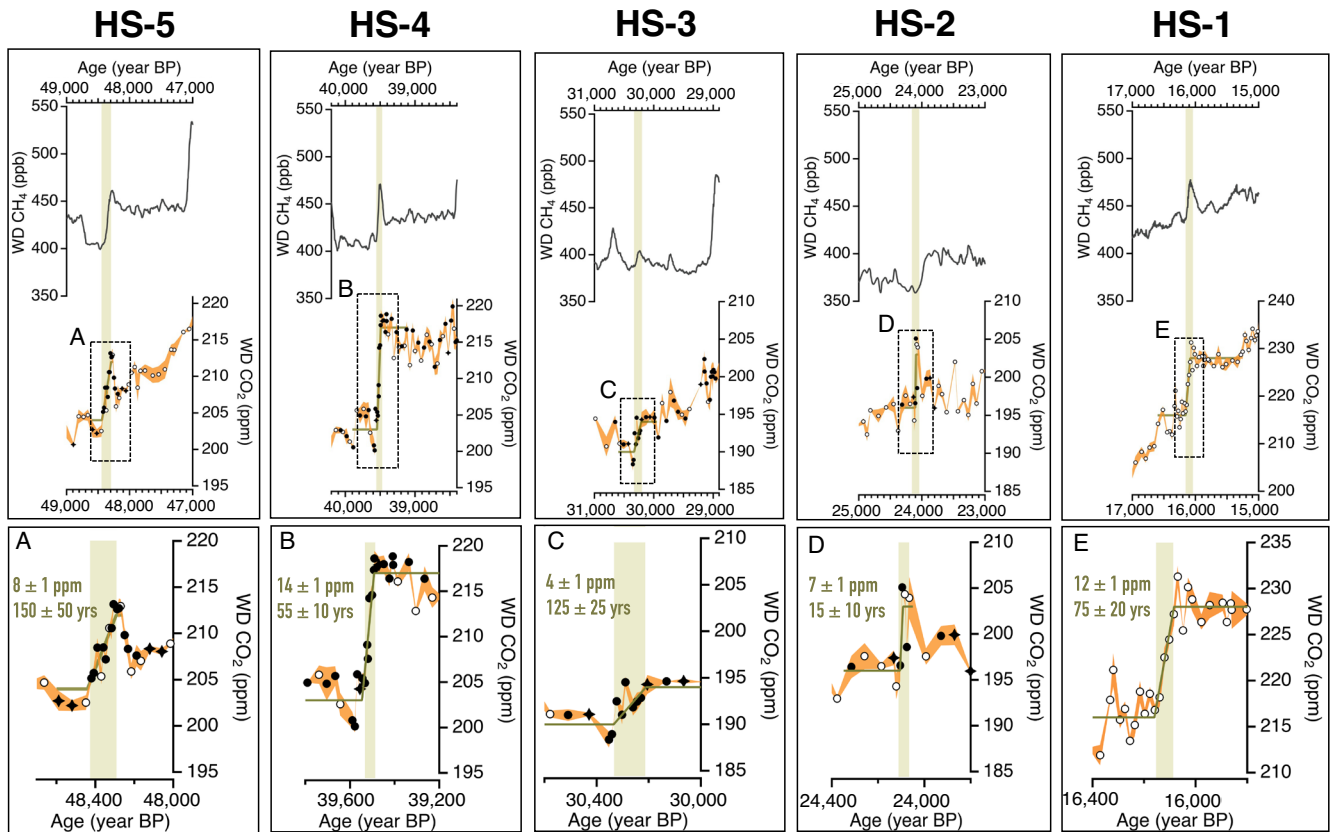
Our high-resolution WD CO<sub>2</sub> record reveals variations in atmospheric CO<sub>2</sub> levels that can be examined at comparable temporal resolution to atmospheric CH<sub>4</sub> variations and the oxygen and hydrogen isotopes of Antarctic ice (henceforth referred to as water isotopologs) across HSs 2 to 5 (Fig. 1). Abrupt rises in atmospheric CO<sub>2</sub> levels are clearly identifiable during HSs 1, 2, 4, and 5, with a smaller-magnitude jump during HS-3 that does not exceed background variability, yet has the same phasing relative to CH<sub>4</sub> and Antarctic water isotopologs as seen during other HSs (Fig. 2).

Each increase in CO<sub>2</sub> is resolved by multiple data points that were replicated by 2 to 3 measurements at each depth. No relationship between the timing of WD CO<sub>2</sub> variability and the carbonate dust in the ice is observed that would suggest in situ production of CO<sub>2</sub> (SI Appendix, Fig. S1). To test reproducibility, a WD replicate core spanning HS-4 was measured across 42 depths at the University of Bern using a friction-free dry-extraction technique (17, 31) (Materials and Methods). Absolute CO<sub>2</sub> values produced in the separate laboratories were reproducible to within an average of 1.6 ppm (SI Appendix, Fig. S2). This small offset is likely due to small differences in methods and/or standard materials used in the separate laboratories.

To determine the timing, duration, and magnitude of the rapid CO<sub>2</sub> jumps associated with HSs 2 to 5, we employed the function Rampfit (37). The largest Rampfit-determined feature is a  $14.0 \pm 0.8$  ppm CO<sub>2</sub> increase within  $55 \pm 10$  y starting at  $39.5 \pm 0.4$  ka ( $2\sigma$ ) during HS-4 (Fig. 2), with a full amplitude (peak-to-peak) of 18 ppm. The CO<sub>2</sub> rise observed during HS-4 is comparable in magnitude and duration to the CO<sub>2</sub> jump associated with HS-1, during which a Rampfit-determined jump of  $12 \pm 0.7$  ppm over  $75 \pm 20$  y is observed (18). The Rampfit-estimated rate of rise is 0.25 ppm/y during HS-4 and 0.16 ppm/y during HS-1. Lower magnitude jumps of  $8 \pm 1.4$  ppm during HS-5 ( $48.4 \pm 0.4$  ka) and  $4 \pm 0.9$  ppm during HS-3 ( $30.3 \pm 0.3$  ka) occurred within  $150 \pm 50$  y and  $125 \pm 10$  y, respectively. A rapid jump of  $7 \pm 1.2$  ppm occurred within  $15 \pm 10$  y during HS-2 ( $24.1 \pm 0.3$  ka). For HS-1, HS-3, and HS-4, higher CO<sub>2</sub> concentrations are maintained after the abrupt event, while for HS-2 and HS-5, the CO<sub>2</sub> jump is part of a short-lived excursion. These rates of CO<sub>2</sub> increase are among the most rapid seen in the ice core record, yet still about an order of magnitude slower than the current rate of anthropogenic CO<sub>2</sub> rise (38).



**Fig. 1.** Paleoclimate records of polar climate, greenhouse gas variability, and East Asian monsoon intensity reflecting meridional displacement of the Intertropical Convergence Zone. (A) Averaged  $\delta^{18}\text{O}$  from Greenland Ice Core Project (GRIP) and Greenland Ice Sheet 2 (GISP2) (black) (32, 33) on the GICC05 timescale (34) multiplied by 1.0063 after 31 ka (35), (B) WAIS Divide (WD) CH<sub>4</sub> (gray) (15), (C) Chinese speleothem  $\delta^{18}\text{O}$  (blue) (11), (D) Antarctic six-core average  $d_{1n}$  (dark green) (36), (E) Antarctic six-core average  $\delta^{18}\text{O}_{\text{ice}}$  (light green) (36), (F) WD CO<sub>2</sub> (orange dots) with binomial smoothed average (dark orange line), and pooled  $1\sigma$  standard uncertainty (orange shading) (this study; 18, 21). Gray bars show the timing of Heinrich Stadials (HS) 1 to 5 (SI Appendix, Table S1). Dashed lines represent timing of decadal- to centennial-scale rises and decreases in WD CO<sub>2</sub> and CH<sub>4</sub>, respectively.



**Fig. 2.** Centennial-scale  $\text{CO}_2$  and  $\text{CH}_4$  variability during Heinrich Stadials 1 to 5. *Upper panel:* WD  $\text{CH}_4$  (15), WD  $\text{CO}_2$  from this study (closed circles), and previous publications (open circles) (18, 21) and replicated from previous publications (closed black diamonds). Light orange shading represents pooled  $1\sigma$  standard uncertainty of individual  $\text{CO}_2$  measurements. Shaded vertical bars highlight centennial-scale jumps in WD  $\text{CO}_2$  and  $\text{CH}_4$ . Green lines indicate breakout determinations (see text). *Lower panel:* WD  $\text{CO}_2$  as in *Upper panel* sections A–E, with the magnitude and duration of abrupt  $\text{CO}_2$  transitions shown in green.

## Discussion

As with HS-1, the multi-decadal- to centennial-scale jumps in  $\text{CO}_2$  associated with HSs 2 to 5 are synchronous with an abrupt, short-lived rise in atmospheric  $\text{CH}_4$  concentrations recorded in the same ice core (15, 18) (Fig. 2).  $\text{CH}_4$  changes during an HS are interpreted as follows: cooler NH temperatures prompted a southward shift of the ITCZ (13), resulting in drying of the NH tropics (10, 11) and a subsequent decrease in  $\text{CH}_4$  emissions via a desiccation of NH wetlands (9, 15) and reduction in their carbon stock (39). High-resolution sampling of ice cores reveals a smaller-magnitude (10s ppb) multi-decadal-scale increase in  $\text{CH}_4$  that occurs approximately midway through each HS (15, 18, 40). The timing of the mid-HS jump in atmospheric  $\text{CH}_4$  levels associated with HS-1 (the HS with best age control) coincides with the onset of ice-rafted debris deposition in the North Atlantic, within age uncertainties (22). Rhodes et al. (15) argue that NH cold intervals associated with ice rafting events triggered a maximum southerly displacement of the ITCZ, as corroborated by subtropical rainfall proxy evidence (10, 11, 16), which led to an expansion of SH subtropical wetlands and a subsequent increase in  $\text{CH}_4$  emissions. This rapid reorganization of global atmospheric circulation likely occurred within a decade of the initiation of ice rafting events (14); thus, mid-HS jumps in atmospheric  $\text{CH}_4$  are interpreted as an indirect proxy for the timing of HEs (15).

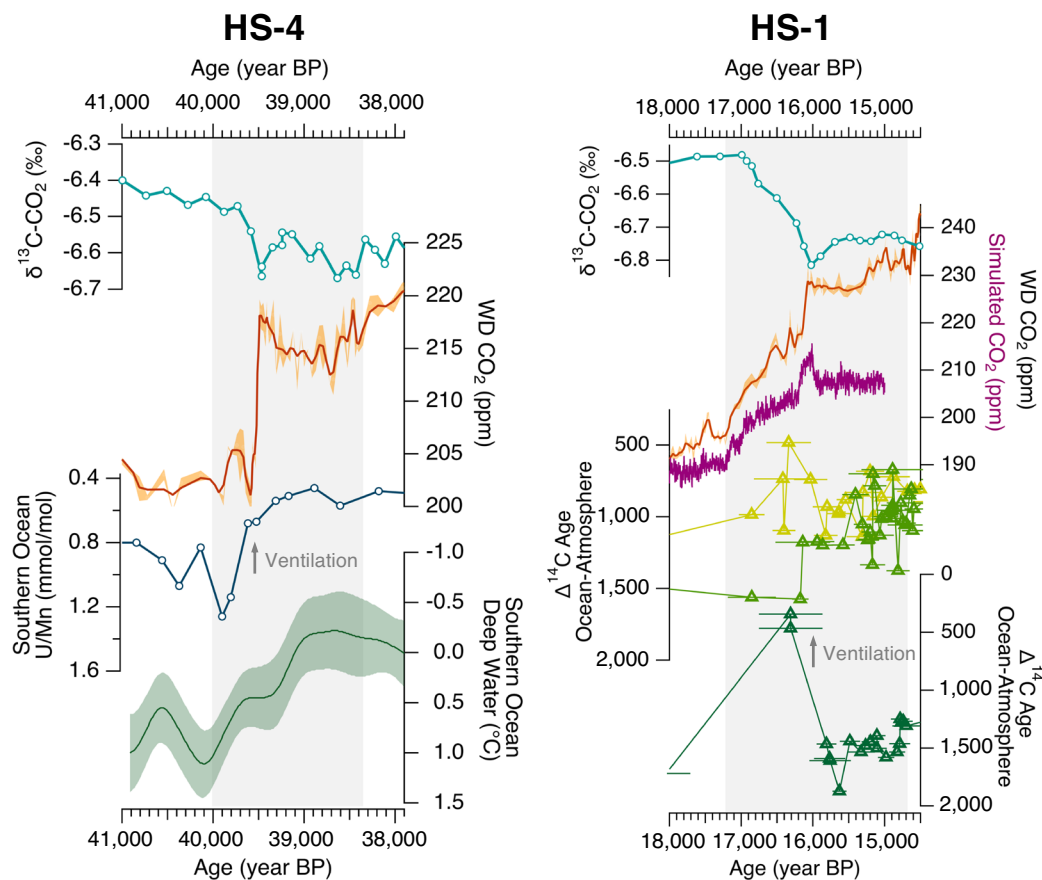
Ice cores also reveal a 0.2 to 0.25‰ depletion in the carbon isotopic composition of  $\text{CO}_2$  ( $\delta^{13}\text{C}-\text{CO}_2$ ) that coincides with  $\text{CO}_2$  and  $\text{CH}_4$  jumps during HS-1 and HS-4 (20, 29) (Fig. 3). Similar high-resolution  $\delta^{13}\text{C}-\text{CO}_2$  data are unavailable for the other events.  $\delta^{13}\text{C}-\text{CO}_2$  reflects combined changes in the terrestrial

carbon pool, marine export production, oceanic circulation, and air–sea gas exchange linked to temperatures (29, 41, 42). Explaining the multi-decadal- to centennial-scale jump in  $\text{CO}_2$  during HSs requires a mechanism that is directly or indirectly linked to concurrent jumps in  $\text{CH}_4$  and depletion of  $\delta^{13}\text{C}-\text{CO}_2$ .

Here, we examine the hypothesis that abrupt shifts in global atmospheric circulation facilitated concomitant rises in  $\text{CH}_4$  and isotopically light  $\text{CO}_2$  during HSs. General ocean–atmosphere circulation model simulations suggest that the SH westerlies respond in parallel to latitudinal shifts in the ITCZ (8, 46, 47), with a southward shift in the ITCZ prompting a poleward shift and strengthening of the SH westerlies. A poleward intensification of the SH westerlies during HSs 1 to 6 and HS-11 is supported by increased salt leakage in the Agulhas Corridor (48–50), increased rainfall in western Patagonia during HS-1 (51), maximum opal productivity in the Southern Ocean during HSs 1 to 5 (24), and a poleward shift in the Southern Ocean SST front latitude (i.e., the latitude at which meridional SST gradient is steepest) during HS-1 (28). Numerical models suggest that poleward intensification of the SH westerlies enhances Southern Ocean ventilation and rapidly releases  $\text{CO}_2$  from the ocean interior (26, 52–54). Even when taking into account the mesoscale response to the wind-driven circulation, increased SH westerly wind stress and parallel changes in surface buoyancy fluxes are thought to enhance the upwelling of deep and mid-depth, carbon-rich waters in the Antarctic Zone of the Southern Ocean (26, 53–56).

Enhanced Southern Ocean ventilation and Southern Ocean–atmosphere  $\text{CO}_2$  exchange during HSs is supported by southern high-latitude sediment records (Fig. 3), which suggest enhanced Antarctic Bottom water formation that delivers cold water to depth





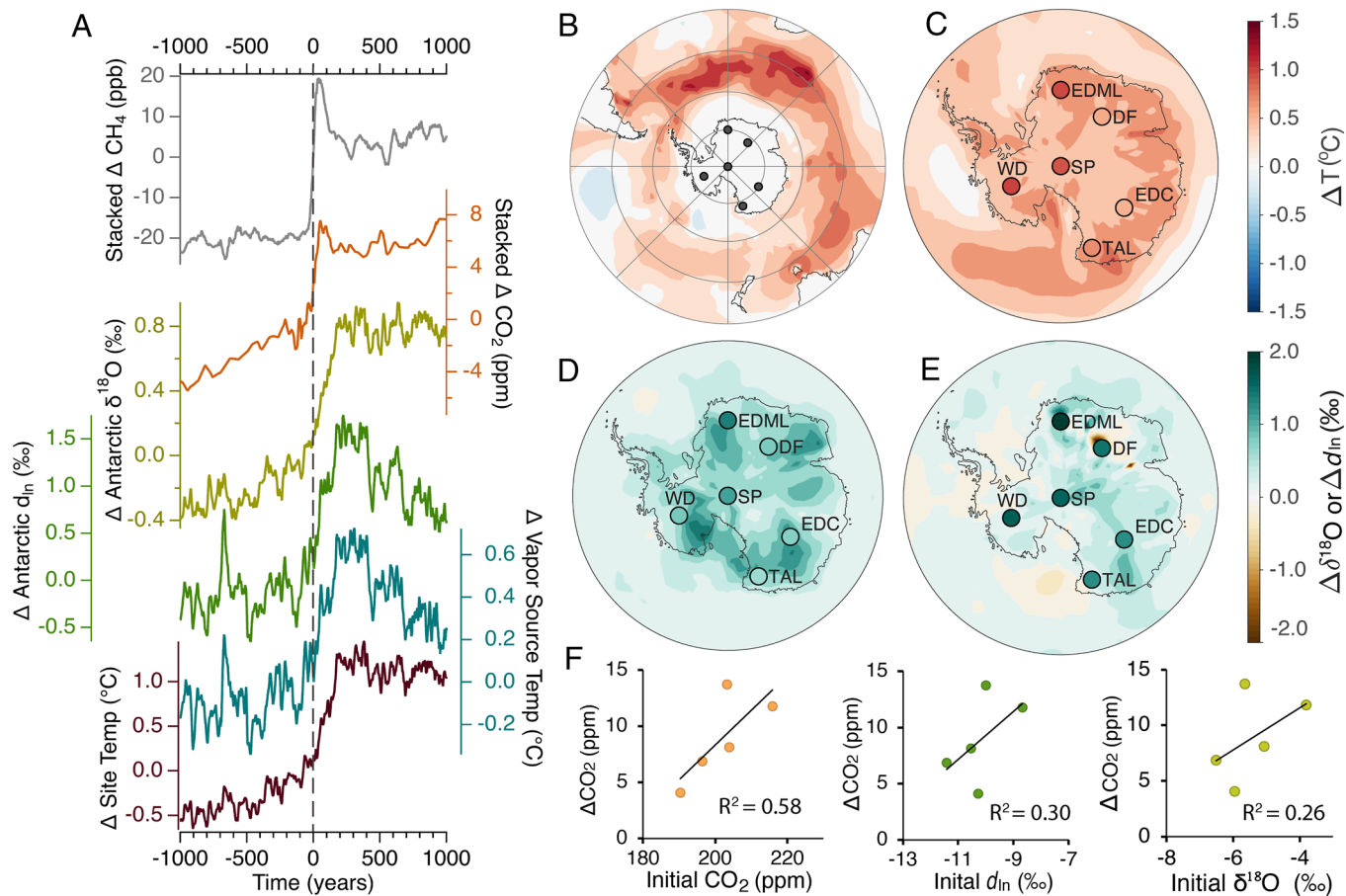
**Fig. 3.** Atmospheric  $\delta^{13}\text{C-CO}_2$ ,  $\text{CO}_2$ , and Southern Ocean marine sediment proxies during HS-4 and HS-1. *Left panel:* Taylor Glacier  $\delta^{13}\text{C-CO}_2$  (light blue) (20), WD  $\text{CO}_2$  (orange) (this study; 21), South Atlantic sediment proxies for deep-water oxygenation (blue) (43), and deep-water temperature (44). *Right panel:*  $\delta^{13}\text{C-CO}_2$  (light blue) (29), WD  $\text{CO}_2$  (orange) (this study; 21), simulated atmospheric  $\text{CO}_2$  (pink) (26; LH1-SO-SHW simulation), and difference in Southern Ocean and atmosphere  $^{14}\text{C}$  ages from shallow (yellow), intermediate (green), and deep (dark green) corals (27, 45) including  $2\sigma$  U-Th dating uncertainties. Respectively, 160 and 150 y were subtracted from the Taylor Glacier chronology to align Taylor Glacier and WD  $\text{CO}_2$  peaks at HS4 and HS1.

(44), greater deep water oxygenation (43, 44), lower Southern Ocean  $^{14}\text{C}$  ventilation ages (43, 45, 57–59), and increased upwelling (24). A depletion of atmospheric  $\delta^{13}\text{C-CO}_2$  (20, 29, 42) (Fig. 3) and enrichment of benthic foraminiferal  $\delta^{15}\text{C}$  south of  $30^\circ\text{S}$  (58–61) during HSs are consistent with a wind-driven release of isotopically light carbon from ventilation of the ocean interior.  $^{14}\text{C}$ -based ventilation reconstructions from precisely dated deep-sea corals also suggest deep vertical mixing in the Southern Ocean during HS-1 (27, 45, 62) (Fig. 3) with a distinct centennial-scale pulse of deep ocean ventilation around  $16.3 \pm 0.4$  ( $2\sigma$ ) ka (27) which is within the dating uncertainty of the rapid  $\text{CO}_2$  rise in HS-1 recorded in the WD ice core (Fig. 3).

Previous studies show that enhanced vertical transport in the Southern Ocean during HSs would likely result in increased sea surface temperatures (SST) due to enhanced upwelling of relatively warm Circumpolar Deep Water (26, 44, 63, 64) (Fig. 3). Temperature reconstructions from precisely dated corals support this hypothesis, with an abrupt  $\sim 5^\circ\text{C}$  warming centered on the interval of rapid  $\text{CO}_2$  rise within HS-1 in corals bathed by waters sourced from the Southern Ocean surface (65). However, resolving centennial-scale Southern Ocean SST anomalies beyond the coral record remains a challenge, given sediment mixing by benthic organisms, insufficient marine sedimentation rates, and poor calcite preservation. To circumnavigate these issues, we examine the relative phasing of high-resolution Antarctic water isotopolog records and abrupt atmospheric  $\text{CO}_2$  jumps during HSs at multidecadal precision. Variations in oxygen isotopes ( $\delta^{18}\text{O}$ ) in the ice phase of ice cores largely reflect changes in site air temperatures. Deuterium excess ( $d_{\text{in}}$ ) is a second-order isotope

parameter that reflects temperature conditions during source evaporation. Previous work shows that  $d_{\text{in}}$  is positively correlated with SSTs at the vapor origin site (66), and it has been used to reconstruct source water temperatures (66–69). To examine similarities across all four HSs, we construct composite (or stacked) records of i) WD  $\text{CO}_2$ , ii) WD  $\text{CH}_4$ , iii)  $\delta^{18}\text{O}$ , and iv)  $d_{\text{in}}$  averaged from six Antarctic ice cores [EPICA Dome C (EDC), EPICA Dronning Maud Land (EDML), South Pole (SP), Talos Dome (TAL), Dome Fuji (DF), and WAIS DIVIDE (WD); Fig. 4]. We also compare the (v) site and (vi) vapor source temperatures calculated by Markle and Steig (67) using  $\delta^{18}\text{O}$  and  $d_{\text{in}}$  records from each ice core (Fig. 4). We interpret the stacked records as a faithful representation of an Antarctic-wide signal of continental surface temperatures and Southern Ocean-wide SSTs for all 5 HSs (Fig. 4).

Results show a jump in atmospheric  $\text{CO}_2$  concentrations associated with HSs 1 to 5 that averages +9 ppm over a 100-y envelope surrounding the atmospheric  $\text{CH}_4$  mid-point ( $\text{CO}_2$  jumps an average of +12 ppm when stacking only HS-1, -4, -5). An abrupt  $0.4\text{‰}$  rise in  $\delta^{18}\text{O}$  and  $0.7\text{‰}$  rise in  $d_{\text{in}}$  occurred within the first century following the mid-point of each  $\sim 40$  ppb jump in atmospheric  $\text{CH}_4$  levels (Fig. 4), with both proxies reaching maximum values within two centuries after the onset of atmospheric  $\text{CO}_2$  rise. On average, HSs show an abrupt  $0.6^\circ\text{C}$  rise in site temperatures and  $0.4^\circ\text{C}$  rise in vapor-source temperatures within the first century following the mid-point of each  $\text{CH}_4$  jump, reaching maximum values of approximately  $1.3^\circ\text{C}$  and  $0.6^\circ\text{C}$ , respectively, within the second century (67). Overall, the data suggest an abrupt centennial-scale increase in Southern Ocean SSTs associated with



**Fig. 4.** Centennial-scale Antarctic temperature response during Heinrich Stadials. (A) Stacked average change in WD  $\text{CH}_4$  (gray) (15), WD  $\text{CO}_2$  (orange) (this study), Antarctic  $\delta^{18}\text{O}$  (yellow) (36), Antarctic  $d_{\text{in}}$  (green) (36), vapor source temperature (blue) (67), and site temperature (purple) (67) during HSs 1 to 5. Time “0” represents the mid-point of the associated WD  $\text{CH}_4$  jump in HSs 1 to 5 (see text). (B) SST anomaly forcing based on Ferreira et al. (64) in response to a poleward displacement of the SH westerlies. The SST response from Ferreira et al. (64) was multiplied by two to better match Antarctic water isotopologs. Spatial pattern of the differences in (C) modeled surface temperature, (D)  $\delta^{18}\text{O}$  (D), and (E)  $d_{\text{in}}$  simulated in iCAM5 before and after imposing SST temperature forcing as shown in (B). Location, abbreviation, and HSs proxy value of each ice core are indicated on each map by the colored dots. (F) Scatter plots showing the magnitude of change in WD  $\text{CO}_2$  concentrations and HSs proxies associated with HSs 1 to 5 versus background values for  $\text{CO}_2$ ,  $d_{\text{in}}$ , and  $\delta^{18}\text{O}$ .

a mid-HS increase in atmospheric  $\text{CH}_4$  and  $\text{CO}_2$  concentrations. Alternative interpretations of the  $d_{\text{in}}$  signal include a northward shift of the vapor source and thus an apparent warming in water isotopologs, however, this would fail to account for the observed site warming. The increase in greenhouse radiative forcing from each  $\text{CO}_2$  jump (e.g.,  $0.35 \text{ W/m}^2$  at HS-4) accounts for  $\leq 20\%$  of the  $\sim 1^\circ\text{C}$  change in SSTs as indicated by our stacked Antarctic  $\delta^{18}\text{O}$  record and is therefore insufficient to explain the full magnitude of SST increase (*Materials and Methods*). Instead, the upwelling of warm,  $\text{CO}_2$ -rich water masses accounts for both a release of heat and  $\delta^{13}\text{C}$ -depleted  $\text{CO}_2$  from the Southern Ocean to the atmosphere (42, 44) (Fig. 3).

To assess whether a multi-decadal-scale increase in Southern Ocean SST due to wind-driven upwelling could influence Antarctic ice core  $\delta^{18}\text{O}$  and  $d_{\text{in}}$ , we perform a series of experiments using the isotope-enabled Community Atmosphere Model 5 (iCAM5) (70). To apply an SST anomaly between  $45^\circ\text{S}$  and  $60^\circ\text{S}$  (approximate latitudinal band of SH westerlies) that broadly matches the  $0.4^\circ\text{C}$  warming implied by ice core  $d_{\text{in}}$ , we multiplied the SST anomaly from Ferreira et al. (64) by a factor of 2 (*Materials and Methods*) and applied this SST anomaly to the iCAM5 model under LGM background climate conditions (Fig. 4B and *SI Appendix, Table S3*). The simulated anomalies in surface air temperature, precipitation  $\delta^{18}\text{O}$ , and precipitation  $d_{\text{in}}$  (Fig. 4C–E) are compared to the abrupt HS signals reconstructed from the Antarctic ice cores (Fig. 4C–E). We find that the model correctly captures the sign of all three signals, yet

underestimates the magnitude of the  $d_{\text{in}}$  shift (*SI Appendix, Table S3*). Similar results are obtained using the simulated SST anomaly from a transient experiment of HS-1 with stronger SH westerlies (LH1-SHW in ref. 26; *SI Appendix, Fig. S4*) as well as an idealized  $1^\circ\text{C}$  SST warming anomaly (*SI Appendix, Fig. S5*), suggesting that the spatial details of the warming pattern are of secondary importance in explaining the magnitude of the observed  $d_{\text{in}}$  shift. In addition, model results predict continent-wide increases in Antarctic snow accumulation rates in parallel with the isotope changes to SST anomalies (*SI Appendix, Fig. S7*). Overall, the iCAM5 simulations support the hypothesis that the observed increase in Antarctic  $\delta^{18}\text{O}$  and  $d_{\text{in}}$  can be explained by a rise in Southern Ocean SST, which is anticipated to occur during intervals of increased wind-driven upwelling due to a poleward intensification of the SH westerlies.

The magnitude of change in Antarctic water isotopologs is consistent between HSs 1 to 5 (Fig. 4F and *SI Appendix, Fig. S8*), even for those HSs that show little to no changes in AMOC such as HS-2 and HS-3 (71). By contrast, the magnitude of the abrupt jumps in atmospheric  $\text{CO}_2$  levels that occur mid-HS is variable among HSs 1 to 5 and ranges from  $4 \pm 1$  ppm during HS-3 to  $14 \pm 1$  ppm during HS4. We find that the magnitude of rapid atmospheric  $\text{CO}_2$  jumps is correlated with background climate conditions, defined here as the temperature proxy values ( $\delta^{18}\text{O}$  and  $d_{\text{in}}$ ) prior to each event, such that colder climate states (lower atmospheric  $\text{CO}_2$ ,  $\delta^{18}\text{O}$  and  $d_{\text{in}}$ ) tend to have a smaller mid-HS  $\text{CO}_2$  jump (Fig. 4F and *SI Appendix, Fig. S8*). We speculate that

background climate may control the extent of Antarctic sea ice, which in turn may control the extent of ocean–atmosphere CO<sub>2</sub> exchange (72, 73).

High-resolution isotopic data from Antarctic ice support the hypothesis that SH high-latitude changes are different for Heinrich and non-Heinrich stadial phases of the Dansgaard–Oeschger (DO) cycle (74) and that the former is not simply an amplified version of the latter. While gradual Antarctic warming via the thermal bipolar seesaw occurs during all DO stadials (Heinrich and non-Heinrich alike), our results suggest that abrupt Southern Ocean SST warming occurs during HSs only, whereas no such abrupt Southern Ocean SST warming is seen during non-HS DO stadials (36). Similarly, no abrupt rises in atmospheric CO<sub>2</sub> or CH<sub>4</sub> are observed during non-HS DO stadials (15, 21). This qualitative difference may reflect a nonlinear (threshold) response in the strength or zonal pattern of the SH westerlies during North Atlantic stadial periods, or perhaps the strong involvement of the AMOC and/or Pacific Ocean in HE dynamics (6, 75, 76).

Our decadal-resolution atmospheric CO<sub>2</sub> record from the WD ice core reveals multi-decadal- to centennial-scale jumps in atmospheric CO<sub>2</sub> levels that are synchronous with jumps in atmospheric CH<sub>4</sub> concentrations and Antarctic water isotopologs during HSs (15, 36). The synchronicity of these jumps can be explained by a southward displacement of the ITCZ in response to North Atlantic ice rafting events, which ultimately led to a poleward enhancement of the SH westerlies. Enhanced wind stress drove increased vertical transport and ventilation of deep waters in the Southern Ocean, resulting in rapid CO<sub>2</sub> outgassing that drove a pulse of atmospheric CO<sub>2</sub> rise of up to 14 ppm within half a century. Other potential mechanisms, including a multidecadal scale release of CO<sub>2</sub> from a terrestrial carbon pool, cannot be excluded and may have occurred in parallel to Southern Ocean changes. The rate and magnitude of atmospheric CO<sub>2</sub> rises resolved in this study provide critical constraints on carbon-cycle variability during abrupt climate shifts and urge caution that the modern-day Southern Ocean carbon sink has the potential to weaken in response to continued poleward enhancement of the SH westerlies (28, 77).

## Materials and Methods

A total of 453 measurements were made from 249 individual depth levels on the WAIS Divide Ice core (79.48°S, 112.11°W). Measurements spanning HSs 2 to 5 have a median sampling resolution of 25 y (*SI Appendix, Table S1*). The timing of HSs 2 to 5 is defined in Martin et al. (74). The mean pooled 1 $\sigma$  SD of all measurements is 0.7 ppm.

CO<sub>2</sub> amount fractions were determined using dry extraction methods at Oregon State University, Corvallis, USA (78). Subsamples were cut at 0.5 to 0.15 m intervals along selected depths of the WD ice core using a Laguna bandsaw in a –25 °C walk-in freezer. Bandsaw blade and tabletop surface were decontaminated with ultrapure ethanol. On the morning of extraction, subsamples were trimmed into ~2.5 cm wide cubes with an average mass of 12  $\pm$  1 (1 $\sigma$ ) g.

Subsamples were inserted into a custom-designed stainless-steel vacuum chamber cooled to –35 °C then crushed 50 times using a stainless-steel plate containing 91 steel pins. The liberated air was expanded to a stainless-steel coil water trap in an ethanol dewar flask cooled to –85 °C, and then condensed in a 6 cm<sup>3</sup> stainless-steel sample tube housed in a cryogenic system at 10 K. Total extraction time per sample was 7 min. Following extraction, sample tubes were warmed to room temperature using a water bath. The air (~1 cm<sup>3</sup> at standard temperature and pressure) was then expanded into an Agilent 6890 N Gas Chromatograph using a vacuum inlet line connected to a sample loop (5 cm<sup>3</sup>) installed on a six-port two-position VICI Valco valve. Pressures in the sample loop were measured with a high-precision MKS 622 Baratron capacitance manometer (0.15% accuracy). The GC system is fitted with a Ni catalyst and flame-ionization detector. N<sub>2</sub> was used as the carrier gas to maximize the detector stability over a

greater range of air-flow rate and temperature conditions. Signals were acquired and integrated using Agilent ChemStation software.

The extracted ice core air was calibrated to dry standard air from the National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory with a known amount fraction of 268.80  $\pm$  0.01 ppm on the WMOX2019 scale for reference gases. In-house verification of standard air values using a lower concentration standard (197.54  $\pm$  0.03 ppm on WMOX2019 scale) yielded no difference between the assigned and measured values within uncertainties. CO<sub>2</sub> amount fractions measured prior to 2019 that were originally calibrated to WMOX2007 are presented here on the updated to the WMOX2019 scale (difference in calibrated results is <0.02 ppm). The amount fractions of standards used here represent the range expected for atmospheric CO<sub>2</sub> concentrations during the last glacial (21). Daily calibrations were performed by introducing standard air to the sample loop at a pressure range comparable to the sample air aliquots. Typical precision for standard air is ~0.2 ppm (1 $\sigma$ ) for n = 6 measurements. A daily calibration curve was constructed using a second-order polynomial fit, as outlined in ref. 78.

Two additional variables were monitored during sample extraction and analysis: i) instrumental drift and ii) adsorption or desorption of CO<sub>2</sub> on the surface of the vacuum chamber during extraction. Instrumental drift includes both variations in baseline (i.e., GC sensitivity) and slope of the calibration curve. The former is automatically detected and subtracted by Agilent ChemStation software, while the latter is monitored daily by bracketing sample measurements with standard air analyses. Measured standard air values, as determined by respective calibration slopes before and after sample measurements, showed no difference within uncertainties. Potential adsorption or desorption of CO<sub>2</sub> was monitored by introducing standard air into the system following crushing. The expanded standard air was then collected in a separate sample tube following normal sampling procedures. Each measurement for a crushed sample is corrected by adding or subtracting the detected net offset from the calibrated standard value. Mean offsets were 4  $\pm$  3.8 (1 $\sigma$ ) ppm throughout sampling.

To determine background values, standard air was expanded over air-free ice and measured using standard methods (78). Gas-free ice was made by boiling deionized water in a vacuum chamber, and then slowly freezing it by placing the chamber in an ethanol bath. One standard air-over-gas-free-blank-ice measurement was performed every 10th sample measurement in order to quantify the influence of the extraction procedure on the raw measurement results. Measurement of gas-free ice resulted in a 0.96  $\pm$  1.02 (1 $\sigma$ ) ppm CO<sub>2</sub> enrichment of standard air (n = 61 measurements over 2 y). The detected ratio of CO<sub>2</sub> enrichment per mass of gas-free ice was used to determine the total contamination per sample mass, which was subtracted from results. Averaged biweekly background values averaged 0.3 ppm/g and ranged from 0.6 to 0.1 ppm/g ice. Raw CO<sub>2</sub> amount fractions were additionally corrected for gravitational enrichment using  $\delta^{15}\text{N-N}_2$  from ref. 79, with gravitational effects ranging from 1.0 to 1.4 ppm. Each sampling depth was replicated 2 to 3 times.

Gas ages were interpolated from the WD2014 chronology (35, 80), which is synchronized to the radiometrically dated Hulu Cave record prior to ~30 thousand years BP (ka). The difference in age between the gas and ice phases ( $\Delta$ age) is relatively well constrained in WD (e.g.,  $\Delta$ age uncertainties of  $\pm$ 73 y at 40 ka BP), such that the time of changes in CO<sub>2</sub> relative to the chemical composition of ice can be precisely determined.

**Breakpoint Determination.** To provide an objective means for determining the timing and duration of the CO<sub>2</sub> transitions across HSs, we employed the function Rampfit (37). Both the timing and duration of abrupt jumps in CO<sub>2</sub> were determined using 25 to 10-y time spacing. Calculated uncertainties are the result of 1,000 Monte Carlo analyses (*SI Appendix, Table S2*).

**Record Stacking.** We constructed a composite (or stacked) records for HSs recorded in WD CO<sub>2</sub>, WD CH<sub>4</sub>, and water isotopologs from six ice cores across Antarctica [EPICA Dome C (EDC), EPICA Dronning Maud Land (EDML), South Pole (SP), Talos (TAL), Dome Fuji (DF), and WAIS DIVIDE (WD)]. Individual HSs were aligned and then averaged to identify their common climatic signal. The methods described here are identical to stacking methods used in ref. 35. We define the midpoint ( $t = 0$ ) of the abrupt CH<sub>4</sub> transitions associated with HSs 1, 2, 4, and 5. We excluded HS-3 due to its relatively low signal. We define a time vector  $\tau$  that runs from time  $t = -1,200$  to  $t = 1,200$  in 1-y increments. The WD06 CO<sub>2</sub>



and CH<sub>4</sub> records were resampled at annual time steps onto time vector  $\tau$ . The uniformly sampled events were averaged to obtain the stacked record. The same methods were used to stack site and source temperature reconstructions (67) from the same six ice cores, as well as  $\delta^{18}\text{O}$  and  $d_{\text{in}}$  in the logarithmic definition from each respective ice core.

**External Reproducibility.** The WD replicate core (WDC-R1) was measured at Climate and Environmental Physics (CEP), Physics Institute, University of Bern, Switzerland, using the Centrifugal Ice Microtome (CIM) in 2017. Using this friction-free dry-extraction method, we performed a total of 132 single measurements on 42 depth levels with three true replicate measurements for each depth level (SI Appendix, Fig. S2). In four cases, four to five single measurements of two directly vertically adjacent samples (2 cm apart) were averaged and are reported as a single value for their average depth. At least 3 to 5 mm were trimmed from all outer surfaces that were in contact to the atmosphere prior to sampling. The average sample weight was  $7.74 \pm 0.30$  g (1 $\sigma$ ). The pooled SD of 0.54 ppm for all depth levels is surprisingly low, probably owing to the high quality of the ice samples (little to no cracks or clathrate-relaxation features) and the number of true replicate measurements. One standard-air-over-gas-free-blank-ice measurement was performed for every four WD-R1 sample measurements to quantify the influence of the extraction procedure on the raw measurement results. The average correction for the extraction offset amounts to  $1.85 \pm 0.67$  ppm (1 $\sigma$ ). All measured raw CO<sub>2</sub> amount fractions have been corrected for gravitational enrichment using  $\delta^{15}\text{N-N}_2$  from ref. 79. The average correction for this gravitational effect amounts to  $1.24 \pm 0.04$  ppm (1 $\sigma$ ). For a detailed description of the CIM method please refer to refs. 31 and 17. All Bern CO<sub>2</sub> measurements made using the CIM were originally referenced to the WMOX2007 amount fraction scale using four primary dry air standards calibrated at the NOAA Earth System Research Laboratory, Boulder, USA ranging between  $192.44 \pm 0.11$  and  $363.08 \pm 0.06$  ppm (17) and later transferred the WMOX2019 amount fraction scale (Dataset S1). Replicate core depths were corrected to align with main core (WD) depths using electrical conductivity data (81). At these depths, the average depth offset between WD-R1 and WD cores is 0.735 m (81).

**Radiative Forcing Calculation.** Changes in radiative forcing due to a rise of CO<sub>2</sub> concentrations were calculated using established equations (82). Assuming all other forcings are held constant, we can approximate the associated change in temperature using a simplified climate sensitivity equation:

$$\Delta T = F/\lambda,$$

where  $\Delta T$  is the temperature change in response to the radiative CO<sub>2</sub> forcing ( $F$ ), and  $\lambda$  is the climate feedback parameter ( $\text{Wm}^{-2}\text{K}^{-1}$ ).  $\lambda$  can be approximated using the mean climate sensitivity per doubling of CO<sub>2</sub> ( $\sim 3.25$  °C) from the range of climate sensitivity estimates cited in recent literature (e.g., ref. 83).

**Isotope-Enabled Modeling.** In this study, we use an atmosphere-only modeling setup that is part of the Community Earth System Model 1.2 (CESM 1.2) (84). Specifically, we use the isotope-enabled Community Atmosphere Model (iCAM 5) (70). We use a nominal 2-degree finite volume resolution, with  $96 \times 144$  latitude by longitude grid size, and 30 vertical levels. Precipitation isotope results from iCESM are well-verified (85).

For all model results presented here, the atmosphere-only iCAM5 model was run for 200 y. The first 50 y are treated as the model spin-up and removed, and the last 150 y were averaged to obtain the equilibrium monthly climatology. Rather than attempting to simulate the warming events dynamically, we instead run one equilibrium snapshot before and one after the abrupt event and interpret the difference between the two as the event magnitude. For the control (CTRL), we run iCAM5 using LGM background conditions (orbital configuration, atmospheric greenhouse gas concentrations, sea level, ice sheet topography), with land and sea surface conditions [including SST and sea ice coverage (SIC)] taken from fully coupled LGM atmosphere-ocean GCM simulations using the Community Climate System Model 4. **iCAM5 deuterium excess in Antarctica.** The traditional linear definition of deuterium excess ( $d = \delta^2\text{H} - 8 \times \delta^{18}\text{O}$ ) is not suitable for situations of strong isotopic depletion (e.g., interior Antarctica or the glacial period) where the  $\delta^2\text{H}-\delta^{18}\text{O}$  slope for equilibrium fractionation begins to deviate from the value of 8. Using the linear definition in such cases results in spurious signals in  $d$  that reflect equilibrium fractionation processes en route, rather than the kinetic fractionation at the

vapor source regions that deuterium excess seeks to capture. To resolve this issue, Uemura et al. (86) introduced an empirical logarithmic definition of deuterium excess that more closely follows water-stable isotope fractionation systematics:

$$d_{\text{in}} = \ln[1 + \delta^2\text{H}] - \left( -2.85 \times 10^{-2} \times (\ln[1 + \delta^{18}\text{O}])^2 + 8.47 \times \ln[1 + \delta^{18}\text{O}] \right). \quad [1]$$

The Uemura  $d_{\text{in}}$  definition is derived by performing a second-order polynomial fit between  $\ln(1 + \delta^{18}\text{O})$  and  $\ln(1 + \delta^2\text{H})$  in isotopic data from global meteoric waters and Antarctic snow, thereby ensuring that there is no residual trend between  $d_{\text{in}}$  and  $\delta^{18}\text{O}$  (86). Unlike the linear definition, the logarithmic definition of deuterium excess shows great coherence across Antarctica on orbital and millennial timescales making it more suitable for climatic interpretation (86–88).

The iCAM5 relationship between  $\ln(1 + \delta^{18}\text{O})$  and  $\ln(1 + \delta^2\text{H})$  in Antarctica for the LGM control run deviates slightly from the observational relationship (SI Appendix, Fig. S3). Therefore, Eq. 1 is not adequate when evaluating  $d_{\text{in}}$  in these simulations. Following the approach by Uemura et al. (86), we perform a second order polynomial fit between the simulated  $\ln(1 + \delta^{18}\text{O})$  and  $\ln(1 + \delta^2\text{H})$  south of 45°S in the iCAM5 LGM control run (SI Appendix, Fig. S3) and derive the following operational definition of  $d_{\text{in}}$  for iCAM5 LGM simulations:

$$d_{\text{in}} = \ln[1 + \delta^2\text{H}] - \left( -1.497 \times 10^{-2} \times (\ln[1 + \delta^{18}\text{O}])^2 + 9.058 \times \ln[1 + \delta^{18}\text{O}] \right). \quad [2]$$

We use Eq. 2 in our analysis of Antarctic  $d_{\text{in}}$  in this paper. However, we urge caution in interpreting the  $d_{\text{in}}$  results due to the observed model-data offset in the  $\delta^2\text{H}-\delta^{18}\text{O}$  relationship over Antarctica (SI Appendix, Fig. S3).

**Southern Ocean SST anomalies.** From studies that prescribe a forced and persistent shift in the SH westerlies, we expect a Southern Ocean SST warming on longer (decadal to centennial) timescales (26, 89), which is also the signal implied by the ice core isotopic signals (Fig. 4). To investigate the impact of such SST anomalies on water isotope ratios in Antarctic precipitation, we run a series of iCAM5 scenarios. We test three different Southern Ocean SST anomalies. First, we use the long-term (years 11 to 20) SST response to a southward shift in the SH westerlies (positive SAM anomaly) from the CCSM3.5 model as found in Ferreira et al. (64). The Ferreira et al. (64) SST anomaly was added to the monthly LGM CTRL climatology and used to force the atmospheric model. A better  $\delta^{18}\text{O}$  model-data agreement was obtained after doubling the Ferreira et al. SST (64) anomaly, and that run is reported here. Second, we use the SST response from Menviel et al. (26) to a simulated southward shift around 16.3 ka BP in the SH westerlies in the LOVECLIM Earth system model run LH1-SHW (SI Appendix, Fig. S4). SST changes from Menviel et al. (26) are much stronger than those from Ferreira et al. (64)—even after multiplying the latter by a factor of 2. Third, we use an idealized SST anomaly that consists of a 1 °C SST warming in a 15 degrees wide latitudinal band (SI Appendix, Fig. S5), the southern edge of which is placed just north of the winter sea ice edge (to avoid applying any warming underneath the sea ice). This SST anomaly is added to the monthly LGM CTRL climatology. The three SST anomalies are shown in Fig. 4 and SI Appendix, Figs. S4 and S5, together with the simulated changes in surface air temperature (SAT),  $\delta^{18}\text{O}$  of precipitation, and  $d_{\text{in}}$  of precipitation. Root mean squared deviation of model results from proxy observations presented in SI Appendix, Table S3. See SI Appendix for additional discussion of modeling results.

**Data, Materials, and Software Availability.** All study data are included in the supporting information.

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