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1 Holocene dynamics of the Southern Hemisphere westerly winds and links to CO_2 outgassing

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15 The Southern Hemisphere westerly winds (SHW) play a significant role in regulating the 16 capacity of the Southern Ocean carbon sink. They modulate upwelling of carbon-rich deep 17 water and, with sea ice, determine the ocean surface area available for air-sea gas exchange. 18 Some models suggest the current strengthening and poleward shift of the SHW will weaken 19 the carbon sink. If correct, centennial- to millennial-scale reconstructions of SHW intensity 20 should be linked with past changes in atmospheric CO₂, temperature, and sea ice. Here, we 21 present a 12,300-year reconstruction of wind strength based on three independent proxies 22 that track inputs of sea salt aerosols and minerogenic particles accumulating in lake

sediments on sub-Antarctic Macquarie Island. Between c. 12.1–11.2 ka BP and since c. 7 ka BP, wind intensities were above their long-term mean, and corresponded with increasing atmospheric CO₂. Conversely, from c. 11.2–7.2 ka BP, wind intensities were below their long-term mean and corresponded with decreasing atmospheric CO₂. These observations are consistent with model inferences of enhanced SHW contributing to the long-term outgassing of CO₂ from the Southern Ocean.

The Southern Ocean currently accounts for $43 \pm 3\%$ of the global oceanic anthropogenic CO_2 uptake 1 mitigating (perhaps temporarily) the climatic effects of enhanced greenhouse gases in the atmosphere. The capacity of the Southern Ocean to absorb CO_2 at the surface is determined by the balance between processes sequestering carbon (e.g., diffusion and the biological carbon pump) versus processes releasing old carbon from the deep ocean to the atmosphere (e.g., upwelling and outgassing) 2,3 . Attempts to model these processes have yielded conflicting results. One model based on instrumental data collected between 1981 and 2004 suggested that the Southern Ocean carbon sink has weakened 4 . Conversely, analyses of the same data extended to 2011 suggest a reinvigoration of the carbon sink since 2002^{5} . This latter trend is also seen in measurements of the difference between the partial pressure of CO_2 in ocean surface water and the overlying atmosphere (ΔpCO_2) of the Southern Ocean in Drake Passage 6 . Spatial extrapolation of the relatively few pCO_2 measurements from the Southern Ocean as a whole suggests a trend towards a weakening sink in the 1990s, but a strengthening one in the 2000s 7 .

One of the main drivers of the Southern Ocean CO_2 sink are the Southern Hemisphere westerly winds (SHW), which are strongest from 50-55°S over the Southern Ocean ⁸ (Fig. 1a). Changes in the SHW are mainly determined by atmospheric temperature gradients, sea surface temperature and regional sea ice ⁹. In turn they influence ocean circulation ^{10,11}, regulate sea ice extent ¹² and control the upwelling of dissolved inorganic carbon-rich deep water to Antarctic surface waters ². All are processes that modulate the net uptake of CO_2 by the ocean from the atmosphere ¹³. Studies that propose a weakening of the Southern Ocean CO_2 sink attribute it to the recent strengthening and poleward shift of the SHW resulting from changes in surface temperature gradients due to human activities, including ozone depletion ⁴. This has brought carbon-rich waters to the surface ocean and reduced ΔpCO_2 ¹.

Despite the potential importance of the SHW in modulating net uptake of CO_2 by the Southern Ocean, recent extrapolations of the future behaviour of the CO_2 sink are solely based on short-term instrumental records and low spatial and temporal measurements of $\Delta p CO_2$ ^{4,5}. If current theories regarding a potential weakening of the CO_2 sink are correct, past reconstructions of changes in the intensity of the SHW over the Southern Ocean should show clear links between atmospheric CO_2 , temperature and sea ice over longer (centennial-millennial) time scales-

To date, palaeoclimatic reconstructions of the SHW derive mostly from southernmost South America, the only continental landmass intersecting the mid to northern core of the SHW. However, they 'do not provide a consistent picture of the SHW during the Holocene' (14, p.14). This can be partly attributed to a reliance on proxies of past changes in effective precipitation

and/or temperature to infer changes in wind strength. Examples include sub-fossil pollen assemblages in peat and lake sediment cores (e.g., ^{15,16}), geochemical proxies of the precipitation-evaporation balance (e.g., ¹⁷), and rainfall changes influencing runoff into fjord ¹⁸ and ocean ¹⁹ sediments. The application of proxies that measure windblown transport (e.g., exotic pollen ^{20,21}, dust ²²) are rare, and their relationship to wind strength not always straightforward ¹⁴.

Avoiding continental landmasses with complex orographic effects, and applying multi-proxy (and independent) methods to track past changes in wind intensity circumvents these issues.

The latter tests whether different wind proxies record congruent patterns in the direction and relative magnitude of change, providing a validation that the proxies are responding to the external forcing of the SHW rather than local or internal dynamics at the study site.

We carried out a detailed reconstruction of Holocene changes in SHW intensity at sub-Antarctic Macquarie Island. The first aim was to reconstruct changes in the dynamics and relative strength of the SHW over the core jets of the Antarctic Circumpolar Current; the region most relevant to Southern Ocean air-sea gas exchange ²³. The second was to test which of the competing models of a weakening ⁴ or reinvigoration ⁵ of the carbon sink in recent decades is supported by the long-term palaeo-record. We did not consider changes in the position of the SHW core belt (50-55°S) in detail. More well-dated, multi proxy records are needed from broader latitudinal and longitudinal ranges to reliably separate changes in intensity from changes in latitudinal position.

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Sub-Antarctic Macquarie Island

Macquarie Island is a small, 130 km², sub-Antarctic island located a few hundred kilometres north of the Polar Front (54°S, 158°E; Fig. 1 and Supplementary Note 1). The sub-Antarctic islands are the only landmasses, other than southernmost South America, that lie in the core of the SHWs. Macquarie Island experiences mean annual wind speeds of ~35 km hr⁻¹, a mean daily wind run of 751 km, gusts of up to 185 km hr^{-1 24} and its climate is representative of the core belt of the SHW (Fig. 1 c, d). Wind almost exclusively comes from the west-northwest (Supplementary Figs. 2 and 3). This results in strong west to east gradients in sea-spray ²⁵ and minerogenic wind-blown aerosols being deposited across the island. These aerosols accumulate over time in lake and peat sediments, preserving a record of relative changes in SHW intensity, with periods of more (or less) sea spray or deposited minerogenic material reflecting phases of stronger (or weaker) winds. We applied a combination of three methods to reconstruct past changes in SHW intensity using a sedimentary sequence from Emerald Lake, a small lake perched on the western edge of the Macquarie Island plateau, and directly exposed to westerly air flow (Supplementary Note 2). The sediment sequence was radiometrically dated with 44 ¹⁴C and 11 ²¹⁰Pb dates (Supplementary Note 3). Freshwater diatom-based inference models provided a measure of past sea spray aerosol inputs through their effect on the conductivity and diatom species assemblages in lakes ²⁵. These were compared with two independent measures of minerogenic aerosol inputs based on micro X-ray fluorescence (μ-XRF) core scanning and hyperspectral imaging. These proxies, and the sedimentology of the core, are described in Supplementary Notes 4, 5.

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Holocene dynamics of the SHW

The three proxies used in the present study show similar patterns in wind strength over Macquarie Island over the last c. 12.3 ka BP (Fig. 2, Supplementary Note 5). The strongest correlation is between the two methods measuring minerogenic inputs (r=0.85, p <0.0001, Supplementary Table 4). These are also significantly correlated with the independent diatominferred conductivity / sea spray proxy (r=0.56 and r=0.65, p <0.0001). The slightly weaker correlations with the diatom-inferred conductivity / sea spray proxy can be attributed to lake water nutrients and pH which explain smaller yet independent portions of the variance in the diatom data ²⁵. The correlations between the minerogenic wind proxies and diatom-inferred conductivity excludes changes in dust supply from source regions as a major factor modifying the reconstructions. The period from 7.5–7.2 ka BP shows a negative correlation between the minerogenic proxies and diatom-inferred conductivity. Based on the presence of sub-aerial diatoms, this was attributed to a brief period of low water levels (see Supplementary Note 4). In general, the proxies show high relative wind intensity (defined here as periods when at least two of the three wind proxies have values greater than the 95% upper bound of their mean, Supplementary Table 6) between 12.1–11.2, 9.2–8.5, 7.9–7.7, 7.0–5.6, 5.3–0.2 and 0.1–0 ka BP. Low relative wind intensities were recorded from 11.2-9.2, 8.4-7.9, 7.7-7.0 and 0.2-0.1 ka BP. The key features are high relative winds spanning the end of the Last Glacial-Interglacial Transition (LGIT) to the early Holocene (12.1–11.2 ka BP), low relative winds during the early Holocene Thermal Maximum (11.2–9.2 ka BP), increased winds and higher amplitude changes

between 9.2–5.3 ka BP, and a period of sustained relatively intense winds from 5.3 to 0.2 ka BP (although the resolution of the record is lower during this period) (Fig. 2). After 0.2 ka BP the wind intensity drops and then increases in the most recent decades.

SHW dynamics, atmospheric CO₂, sea ice and temperature

Significant correlations between the three independent wind proxies support the hypothesis that, together, they provide a reliable record of changes in the dynamics of the SHW in their core belt, and more specifically at Macquarie Island. Therefore, to address our wider objective of evaluating the role of the SHW as a driver of natural CO_2 variations, we compared past changes in the SHW at Macquarie Island with other hemispheric changes in the ocean and atmosphere. These include far-field proxy records in the EPICA Dome C ice core of sea ice (sea salt Na+ aerosol flux)²⁶, temperature and CO_2^2 , and marine core microfossil-based measurements of winter sea ice concentration (WSI %) in selected (and not necessarily representative) marine cores from the South Atlantic (PS2090²⁷) and South Pacific sector near Macquarie Island (E27-23²⁸, Fig. 3).

12.1–11.2 ka BP: The end of the LGIT and early Holocene are characterised by intense SHW. Many records at similar latitudes in southernmost South America (Supplementary Fig. 15c-e), and elsewhere in the Southern Hemisphere (Supplementary Note 7) agree. This period of intense SHW is accompanied by marked increases in atmospheric CO₂ and temperature (Fig 3e, f), and declines in sea ice (Fig. 3c, d). There are both proxy and model-based studies that support the magnitude and direction of these changes across the LGIT ^{10,29,30}. Specifically, the

persistent winds identified here are consistent with enhanced Southern Ocean upwelling 10 accounting for at least part of the deglacial rise in atmospheric CO_2 , and accompanied by increases in atmospheric 30 and sea surface temperatures 29 . Temporal patterns in our records are also consistent with the retreat of sea ice having a positive feedback on the ocean surface area available for the outgassing of CO_2 29 (Fig. 3).

11.2–9.2 ka BP: An early Holocene Thermal Maximum between 11.2–9.2 ka BP is clearly resolved in Antarctica ³¹ and other records across the Southern Hemisphere, including the western side of the Andes from 49–55°S ¹⁴. This reduced the thermal gradient between the mid and high latitudes and resulted in persistent low intensity winds at Macquarie Island (Fig. 3a, b). This is consistent with selected records from South America ^{32,33}, Tasmania and New Zealand ³² (Supplementary Note 7), and a reduced thermal gradient in the Southern Ocean east and south of New Zealand ³⁴. If replicated in further studies, this would suggest a widespread decrease in wind intensity across the Southern Hemisphere from 41°S to 54°S (Macquarie Island, see Supplementary Note 7). The sustained warm conditions and low wind intensity during this period corresponded with reduced sea ice in marine ^{27,29} and ice core records ³⁵, and was followed by the only long-term decline in atmospheric CO₂ (c. 10.5–7.2 ka BP) since the Antarctic Cold Reversal ³⁶.

9.2–5.3 ka BP: All three wind proxies at Emerald Lake show a period of increased and higher amplitude changes in wind intensity (relative to the early Holocene Thermal Maximum), punctuated by relatively shorter lived (multi-centennial) periods when wind strength is

significantly lower. This corresponds to some terrestrial records of enhanced precipitation in South America (e.g., Tamar Lake ³² and Lago Cipreses ³³, but not others ⁸; see Supplementary Note 7). These increased winds coincide with a downturn in temperature and increases in sea ice (Fig. 3c, d). During the first part of this period the positive relationship between enhanced winds and sea ice appears similar to the beginning of the record (12.1–11.2 ka BP), but differs, at least until 7 ka BP, because the winds did not correspond with an increase in atmospheric CO₂ (Fig. 3f). We attribute this to the concurrent development of alternative carbon sinks in the terrestrial biosphere, including mineral soils, peat and permafrost carbon ^{37,38}. However, after 7 ka BP, increased wind strength and sea ice correspond to the onset of the steady increase in atmospheric CO₂ that characterised the middle and late Holocene.

5.3–0 ka BP: This period of relatively constant and intense winds at Macquarie Island and elevated rainfall inferred from several studies on the western side of the Andes (see Supplementary Note 7, Supplementary Fig 15) corresponds with: relatively high sea ice in the Atlantic sector (Fig. 3c); increases in the ice core ssNa after 4 ka (Fig. 3d); a sustained increase in atmospheric CO₂ (Fig. 3f). Combined, this evidence implies that persistent strong winds led to net Southern Ocean outgassing during the latter part of the Holocene. Over the last 0.2 ka BP, our record of Ti aerosol inputs into Emerald Lake show an initial decline, followed by an increase in the last c. 100 years. The latter is consistent with instrumental records from Macquarie Island, and other areas of the Southern Ocean, which have shown an intensification and southward shift in the main wind belt ⁵ (Supplementary Fig. 2). However, during this period,

all proxies are potentially compromised by erosional inputs associated with the activities of introduced species ³⁹ ('rabbit-influenced zone'; Fig. 2).

SHW, CO₂, temperature and sea ice

To develop our understanding of Southern Hemisphere climate dynamics, we now focus on the centennial to millennial relationships between the SHW, atmospheric CO₂, temperature and sea ice, and the extent to which they are replicated in selected Global Climate Models.

First, the Macquarie record shows significant positive relationships between SHW and sea ice particularly through the late LGIT and early Holocene, and possibly during the period of sustained relatively intense winds from 5.3 to 0.2 ka BP (Supplementary Table 4, Supplementary Note 5). This is consistent with CMIP5 models that show significant relationships between SHW jet strength and sea ice area ⁴⁰. It has been suggested that a strengthened SHW jet leads to increased Ekman upwelling bringing cooler subsurface water to the surface and strengthened equatorward transport, which is conducive to increased sea ice ¹². Alternatively, at least part of the correspondence between the SHW reconstruction and ice core ssNA could be attributed to direct wind-driven transport of ssNa to the Dome C ice core site, independent of changes in sea ice extent, although this has previously been questioned based on chemical signatures in the ice ⁴¹. Thus, except for the 9.2–7 ka BP period, where the relationship is less clear, the Macquarie Island record suggests that, at centennial to millennial timescales during the Holocene, sea ice and the intensity of the SHW were broadly in phase.

Second, in terms of the relationship between the SHW and atmospheric temperature and CO₂, the Macquarie Island proxy records show that windier periods (i.e., greater than the 95% upper bound of their long-term mean) correspond with periods of increasing atmospheric CO₂ (orange boxes in Fig. 3b, f) from 12.1–11.2 ka BP, and in the last 7 ka. Conversely, the period of lower than mean winds from 11.2 ka BP corresponds with decreasing atmospheric CO₂ (blue boxes in Fig. 3b, f), and a downturn in temperature (Fig. 3e), which persisted until c. 8 ka BP. This relationship is reproduced in short term CMIP5 model experiments, which consistently show a poleward shift and strengthening of the SHW jet in response to increasing greenhouse gases and stratospheric ozone 42. These observations are consistent with enhanced wind driven upwelling of CO₂ rich deep ocean waters, accounting for at least part of the increase in atmospheric CO₂ across the LGIT and into the early Holocene ³ and the last 7ka, whilst periods of reduced wind strength enhance the Southern Ocean CO₂ sink. The larger magnitude of the increase in CO₂ during the late LGIT and early Holocene (Fig. 3) can be attributed to the higher CO₂ available for release from the glacial deep ocean ⁴³. In contrast, the moderate increases in CO₂ since 7 ka BP can be attributed to less CO₂ available for release from the ocean and the development of terrestrial carbon sinks ³⁷.

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SHW and the future of the Southern Ocean carbon sink

Our new records of the SHW, which are aligned with the core jets of the present-day Antarctic Circumpolar Current, suggest large changes in wind intensity over the last 12.1 ka BP. This is in marked contrast to model simulations of the SHW across the Last Glacial termination and Holocene which simulate only relatively small wind speed anomalies (\pm 1 m s⁻¹) 9,23,44,45 . The

correspondence between strong and persistent SHW and the rise in CO_2 during the latter part of the LGIT into the early Holocene, and from 7 ka BP suggests that the winds have contributed to the long-term outgassing of CO_2 from the ocean during these periods. This provides a longer-term perspective on the 30-year instrumental record of ΔpCO_2 and SHW strength, which has been used to evaluate short-term changes in the behaviour of the Southern Ocean carbon sink been used to evaluate short-term changes in the behaviour of the Southern Ocean carbon sink 4.5. Specifically, the palaeo-data support the Le Quéré model 4, which suggests a wind-driven weakening of the Southern Ocean carbon sink (enhanced outgassing) prior to 2007 CE as the dominant process. It does not support the alternative Landschützer model 5, which proposes that a stabilisation of the surface waters has counteracted the wind induced upwelling and reinvigorated the carbon sink during the last decade. Therefore, over multi-decadal to millennial timescales, further increases in wind strength will lead to faster accumulation of CO_2 in the atmosphere.

Methods

A series of lake sediment cores dating back to c. 12.1 ka BP were extracted from Emerald Lake on the western edge of the Macquarie Island plateau. Perched within a small catchment, Emerald Lake is ideally situated to record changes in sea salt and mineral aerosol inputs (Supplementary Notes 2, 3). We applied three independent methods to reconstruct past changes in SHW intensity (Supplementary Note 5). First, to reconstruct past sea salt aerosol inputs, we developed a method to track past changes in lake water conductivity (a function of sea salt aerosol input) using diatom-based inference models ^{25,46}. These utilise the strong statistical relationship between modern diatom species assemblages and lake water

conductivity (weighted averaging partial least squares, two components; $r^2 = 0.92$, $r^2_{jack} = 0.72$, RMSEP = 230 μS cm⁻¹ (Supplementary Note 5), which is applied as a transfer function to reconstruct past changes in conductivity from subfossil species assemblages preserved in lake sediments. Second, ITRAXTM micro X-ray fluorescence (μ-XRF) core scanner data were used to track inputs of minerogenic aerosols into the lake, focusing on titanium (Ti), one of the most widely used indicator elements for increased allochthonous inputs ⁴⁷ from distal and proximal sources ⁴⁸. Third, cores were scanned using a Specim hyperspectral camera, which measures reflected optical properties between wavelengths from 400–1000 nm ⁴⁹. The ratio of reflectance between 850 and 900 nm (R₈₅₀/R₉₀₀) was used as an additional indicator of minerogenic inputs. Statistical analyses of the proxies were undertaken using R 2.15.2 (R Foundation for Statistical Computing), MATLAB® (Supplementary Note 3 for details) and in Sigmaplot v13, using original and 100-year interval 2nd order polynomial LOESS (Local tricube weighting and polynomial regression) smoothing. The calibration and performance of each of these methods together with details of core sedimentology and chronology are described in Supplementary Notes 3, 4 and 5. Potential influences on the proxies (e.g., lake ice cover and relative sea level) are described in Supplementary Note 6.

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Data Availability

All data are archived at the Australian Antarctic Data Centre (https://data.aad.gov.au/) and Natural Environment Research Council Polar Data Centre (https://www.bas.ac.uk/data/uk-pdc/).

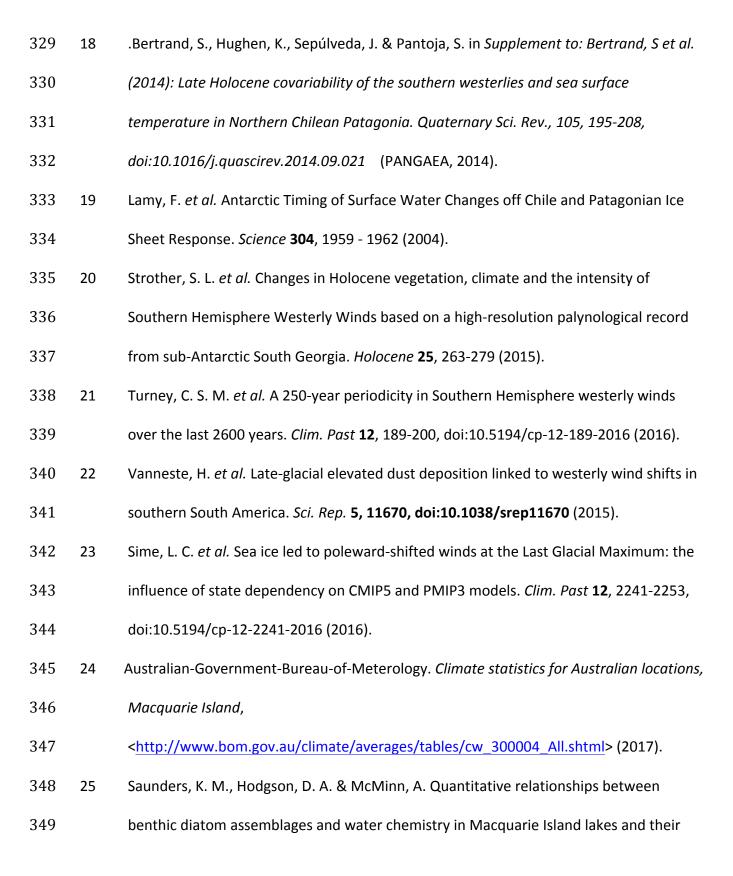
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Author contributions

DH, KS and SR contributed equally to this work. Fieldwork was carried out by KS, DH, SR, WVN.

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WVN (fieldwork, diatom analysis), KS, CB and MG (hyperspectral imaging) and LS (modelling).

DH, KS, SR and BP wrote the manuscript and supplementary information with input from all authors.

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Figure captions

Figure 1. Southern Hemisphere westerly winds and wind anomalies in relation to the location of Macquarie Island (white circle) a. Location of Macquarie Island in the Southern Ocean within the core of the Southern Hemisphere Westerly wind (SHW) belt. White square = EPICA Dome C ice core (EDC). Grey triangle = marine core PS2090, white triangle = marine core E27-23; SHW = Southern Hemisphere Westerlies, PF = Polar Front, SACCF = Southern Antarctic Circumpolar Current Front. b. Location of Macquarie Island and Emerald Lake (pansharpened LANDSAT8 satellite image, bands 4,3,2). c. Annual mean and d. Decadal wind speed anomalies associated with the top 20% of highest wind speed events at Macquarie Island (see Supplementary Notes 1 and 2 for details).

Figure 2. Changes in relative strength of the Southern Hemisphere westerly winds based on proxies in a sediment core from Emerald Lake. a. Diatom-inferred (D-I) conductivity. b. Titanium (Ti) micro-XRF analysis. c. Micro-XRF Principal Components Analysis Axis 1. d. Hyperspectral ratios (R_{850}/R_{900}). Red lines = 100-year interval 2nd order LOESS-smoothing.

Periods of increased wind strength defined as > than the 95% upper bound of the mean (blue horizontal lines; see Supplementary Note 5 and Supplementary Table 6). Green shading indicates when proxies compromised by erosional inputs from non-indigenous species ⁴². LGIT = Last Glacial-Interglacial Transition, EH = early Holocene, MH = mid Holocene, LH = late Holocene. Horizontal bar = formal EH. Lithology described in Supplementary Table 3 and Note 4. Figure 3. Comparison of Macquarie Island wind proxies with sea ice, temperature and CO₂ a. Diatom-inferred (D-I) conductivity **b.** Titanium (Ti) micro-XRF data. **c.** Southern Ocean winter sea ice concentration (WSIC) from marine cores PS2090 ²⁹ and E27-23 ³⁰. **d.** EPICA Dome C sea salt Na⁺ aerosol flux (representing new sea ice surfaces) ³⁸. **e.** EPICA Dome C temperature anomaly ³². **f.** EPICA Dome C CO₂ ³⁹. Grey dotted lines are lithological zones defined in Figure 2. Orange highlights periods when above-mean winds correspond to increasing CO₂, blue when below-mean winds correspond to decreasing CO₂. LGIT = Last Glacial-Interglacial Transition, EH = early Holocene, MH = mid Holocene, LH = late Holocene. Horizontal bar = formal EH.

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