

Abrupt changes in biomass burning during the last glacial period

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Ben Riddell-Young^{1,2✉}, James Edward Lee³, Edward J. Brook¹, Jochen Schmitt⁴, Hubertus Fischer⁴, Thomas K. Bauska⁵, James A. Menking⁶, René Iseli⁷ & Justin Reid Clark⁸

Understanding the causes of past atmospheric methane (CH_4) variability is important for characterizing the relationship between CH_4 , global climate and terrestrial biogeochemical cycling. Ice core records of atmospheric CH_4 contain rapid variations linked to abrupt climate changes of the last glacial period known as Dansgaard–Oeschger (DO) events and Heinrich events (HE)^{1,2}. The drivers of these CH_4 variations remain unknown but can be constrained with ice core measurements of the stable isotopic composition of atmospheric CH_4 , which is sensitive to the strength of different isotopically distinguishable emission categories (microbial, pyrogenic and geologic)^{3–5}. Here we present multi-decadal-scale measurements of $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$ from the WAIS Divide and Talos Dome ice cores and identify abrupt 1‰ enrichments in $\delta^{13}\text{C}-\text{CH}_4$ synchronous with HE CH_4 pulses and 0.5‰ $\delta^{13}\text{C}-\text{CH}_4$ enrichments synchronous with DO CH_4 increases. $\delta\text{D}-\text{CH}_4$ varied little across the abrupt CH_4 changes. Using box models to interpret these isotopic shifts⁶ and assuming a constant $\delta^{13}\text{C}-\text{CH}_4$ of microbial emissions, we propose that abrupt shifts in tropical rainfall associated with HEs and DO events enhanced ^{13}C -enriched pyrogenic CH_4 emissions, and by extension global wildfire extent, by 90–150%. Carbon cycle box modelling experiments⁷ suggest that the resulting released terrestrial carbon could have caused from one-third to all of the abrupt CO_2 increases associated with HEs. These findings suggest that fire regimes and the terrestrial carbon cycle varied contemporaneously and substantially with past abrupt climate changes of the last glacial period.

Ice core measurements of atmospheric methane (CH_4) reveal its past variability. Identifying the causes of this variability is important for understanding global CH_4 biogeochemistry and its relation to climate. Because most CH_4 emissions originate from the terrestrial biosphere, drivers of past atmospheric variability can reveal the terrestrial biogeochemical and hydrologic components of past abrupt climate change at temporal scales not recorded by other proxies^{1,2,8–10}.

Rapid CH_4 changes of 50–150 ppb during the last glacial period (115–11.5 kyr) were contemporaneous with Dansgaard–Oeschger (DO) cycles^{2,11}, which are defined by abrupt, millennial-scale Northern Hemisphere (NH) temperature variations likely driven by rapid changes in Atlantic Meridional Overturning Circulation (AMOC) strength¹². The warm phases of these cycles (DO interstadials) were associated with abrupt northward shifts of tropical rain belts and NH monsoon intensification^{13,14}, which likely enhanced microbial wetland CH_4 emissions^{8,11,15}.

Heinrich events (HEs) were extensive, multi-decadal-scale iceberg discharges into the North Atlantic Ocean that occasionally occurred within the stadial phase of DO cycles and considerably weakened AMOC¹⁶, causing cascading impacts on the global climate system^{7,17}. High-resolution

speleothem records indicate both rapid drying of Asian Monsoon (ASM) regions¹⁴ and Southern Hemisphere (SH) monsoon intensification¹⁸ approximately coincident with HEs¹⁴. Multi-decadal-scale CH_4 pulses (about 50 ppb) have been linked to HEs, with proposed abrupt southward shifts of tropical rain belts enhancing southern tropical wetland CH_4 emissions^{1,19}. However, quantitative constraints on the CH_4 budget across HEs and DO events are currently lacking.

Causes of past CH_4 variability can be investigated using measurements of the carbon and hydrogen stable isotopic composition of CH_4 ($\delta\text{D}-\text{CH}_4$ and $\delta^{13}\text{C}-\text{CH}_4$) (refs. 3–5). The three main emission categories—microbial (wetlands, ruminants and termites), pyrogenic (biomass burning, or BB) and geologic (marine clathrates, cold seepage and mud volcanoes)—have distinguishable isotopic compositions^{4,5,20} (Extended Data Table 1). Changes in the proportion of these sources are reflected in the atmospheric isotopic composition. Specifically, $\delta\text{D}-\text{CH}_4$ and $\delta^{13}\text{C}-\text{CH}_4$ are both primarily influenced by the dominant, isotopically depleted microbial source, although $\delta^{13}\text{C}-\text{CH}_4$ is particularly sensitive to changes in ^{13}C -enriched pyrogenic emissions, whereas $\delta\text{D}-\text{CH}_4$ is strongly influenced by deuterium-enriched geologic emissions^{4,5,20}.

¹College of Earth, Ocean, and Atmospheric Sciences (CEOAS), Oregon State University, Corvallis, OR, USA. ²Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO, USA. ³Los Alamos National Laboratory, Los Alamos, NM, USA. ⁴Climate and Environmental Physics, Physics Institute, and Oeschger Centre for Climate Change Research, University of Bern, Bern, Switzerland. ⁵British Antarctic Survey, Cambridge, UK. ⁶Commonwealth Scientific and Industrial Research Organization (CSIRO), Aspendale, Victoria, Australia.

⁷Adolphe Merkle Institute, University of Fribourg, Fribourg, Switzerland. ⁸Institute of Arctic and Alpine Research, University of Colorado, Boulder, CO, USA. ✉e-mail: Benjamin.riddell-young@noaa.gov

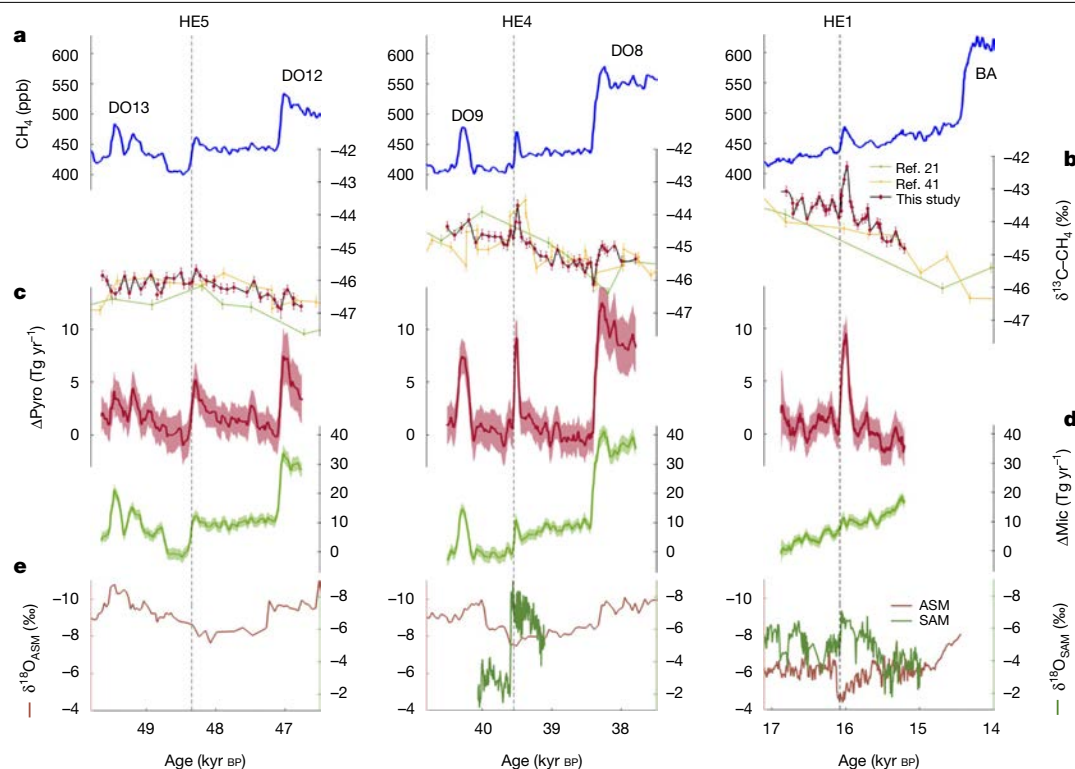


Fig. 1 | $\delta^{13}\text{C}-\text{CH}_4$ for HE5 and DO12, HE4 and DO8 and HE1. **a**, WDC CFA CH_4 data (blue)^{39,53}. **b**, $\delta^{13}\text{C}-\text{CH}_4$ data from ref. 21 (green), ref. 41 (yellow) and this study (red diamonds, black line) each plotted with 1σ measurement uncertainties. **c,d**, Relative changes in pyrogenic (red) and microbial (green) emissions from the one-box model used to interpret the $\delta^{13}\text{C}-\text{CH}_4$ data. Shaded bands show Monte-Carlo-derived 95% confidence intervals related to analytical, firn diffusive, geologic emission and source signature uncertainties

(Methods). **e**, $\delta^{18}\text{O}-\text{CaCO}_3$ records from speleothems in Asian Monsoon (ASM) (brown; HE4 and HE5, ref. 54; HE1, ref. 55) and South American Monsoon (SAM) regions (green; HE4, ref. 18; HE1, ref. 44). DO events and the Bölling Alleröd (BA) warming are labelled and HEs are indicated by vertical dashed lines. Note that the approximately 50 ppb CH_4 pulses discussed here are assumed synchronous with HEs¹.

The temporal resolution of existing Pleistocene $\delta^{13}\text{C}-\text{CH}_4$ records limits interpretations to multi-millennial timescales and existing $\delta\text{D}-\text{CH}_4$ data are limited²¹. Several records that used Greenland ice cores to increase data resolution across abrupt CH_4 changes^{4,22} were likely contaminated by non-atmospheric CH_4 released during the analysis of dusty Greenland ice^{23,24}.

Abrupt CH_4 isotopic variations

We present multi-decadal-scale measurements across several prominent abrupt CH_4 changes associated with HEs and DO events using samples from Antarctic ice cores, which are not affected by non-atmospheric CH_4 contamination^{23,24}. Specifically, we measured $\delta^{13}\text{C}-\text{CH}_4$ across the abrupt around 50 ppb CH_4 pulses associated with HE5, 4 and 1, and the abrupt 100–150 ppb CH_4 increases associated with DO12 and DO8 using ice from the WAIS Divide Ice Core (WDC) and WDC Replicate Core. WDC is ideal for high-resolution measurements because the high accumulation rates result in minimal smoothing of the gas record by firn processes (mean sample gas age distribution of 24–58 years) (ref. 9). Samples were also measured for $\delta\text{D}-\text{CH}_4$ using the WDC Replicate Core across the HE4 CH_4 pulse and the Talos Dome Ice Core (TALDICE) across DO8 (see Methods for detailed descriptions of sample measurement and data processing).

We identify hitherto undetected $+1\text{‰}$ $\delta^{13}\text{C}-\text{CH}_4$ anomalies synchronous with the abrupt CH_4 increases associated with HE4 and HE1 (ref. 1) and a smaller $+0.5\text{‰}$ anomaly across HE5 (Fig. 1b). During the larger CH_4 increases at the onset of DO12 and DO8, we observe $+0.5\text{‰}$ $\delta^{13}\text{C}-\text{CH}_4$ anomalies (Fig. 1b). Across the DO8 and HE4 CH_4 changes, $\delta\text{D}-\text{CH}_4$ varies little (Fig. 2e). To interpret these data, we used a one-box CH_4 stable

isotope model^{6,25} (equations (1–5)) and ran forward simulations using high-resolution continuous flow analysis (CFA) CH_4 data¹⁹ and our interpolated $\delta^{13}\text{C}-\text{CH}_4$ data (Methods). The magnitude and $\delta^{13}\text{C}-\text{CH}_4$ of the global CH_4 source calculated from the model were used to determine the balance of pyrogenic and microbial emissions (equations (6) and (7) and Fig. 1c,d) using established source signatures (Extended Data Table 1) and assuming geologic emissions of 2.5 Tg yr^{-1} ($1\sigma = 1.3 \text{ Tg yr}^{-1}$), as suggested by radiocarbon-derived constraints across deglacial abrupt CH_4 changes^{10,26}.

We tested our assumption of stable geologic emissions by comparing our DO8 and HE4 $\delta\text{D}-\text{CH}_4$ datasets with theoretical $\delta\text{D}-\text{CH}_4$ records generated using $\delta^{13}\text{C}-\text{CH}_4$ -derived emissions that either assumed stable geologic emissions or stable pyrogenic emissions (Fig. 2e and Methods). The scenario in which deuterium-enriched geologic emissions²⁰ were allowed to vary while holding pyrogenic emissions constant caused substantial $+15\text{--}20\text{‰}$ $\delta\text{D}-\text{CH}_4$ anomalies at the HE4 and DO8 CH_4 increases (Fig. 2e). No such anomalies are observed in the WDC and TALDICE $\delta\text{D}-\text{CH}_4$ datasets. The theoretical $\delta\text{D}-\text{CH}_4$ record that assumed constant geologic emissions more closely matches the measured data (Fig. 2e). This supports radiocarbon-based evidence^{10,26} that previously hypothesized marine CH_4 clathrate destabilization during abrupt DO interstadial warming²⁷ or HE iceberg discharge²⁸ did not occur at sufficient scale to impact atmospheric CH_4 .

With geologic emissions assumed stable, $\delta^{13}\text{C}-\text{CH}_4$ -derived pyrogenic emissions increased by $9\text{--}15 \text{ Tg yr}^{-1}$ ($+90\text{--}150\%$ of annual pyrogenic CH_4 emissions) at the onset of each HE (Methods and Extended Data Table 2) and explain most of the CH_4 pulses across HEs (Fig. 1c). $\delta^{13}\text{C}-\text{CH}_4$ -derived microbial emissions increase by about 10 Tg yr^{-1}

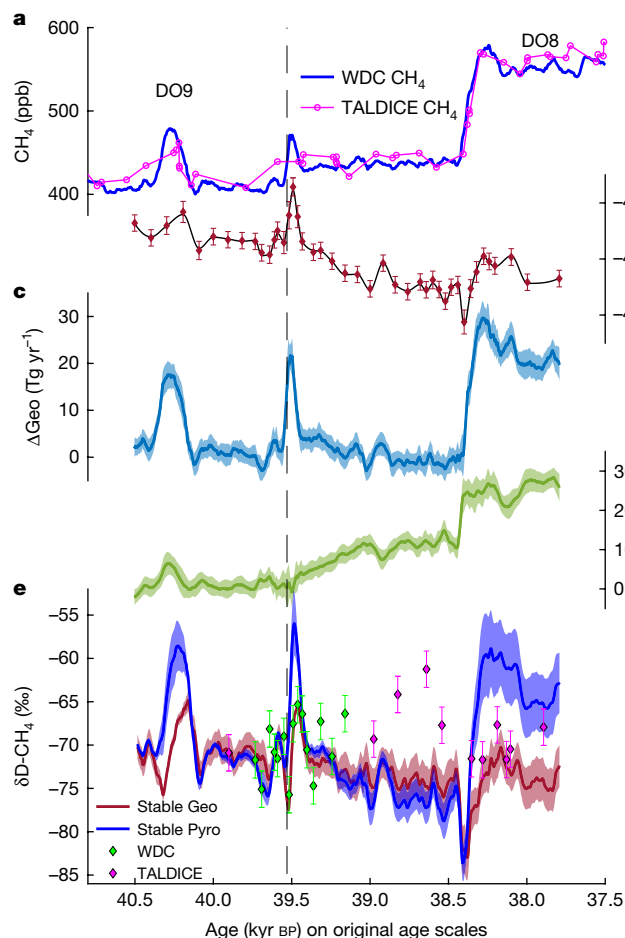


Fig. 2 | $\delta D-CH_4$ across HE4 and DO8. **a**, WDC CFA CH_4 data in blue¹⁹, TALDICE CH_4 data in magenta⁵³ using the WDC-synchronized gas age scale from refs. 56, 57. **b**, $\delta^{13}C-CH_4$ data from this study (red diamonds, black line) and 1σ measurement uncertainties. **c, d**, Relative changes in modelled geologic (light blue) and microbial (green) emissions assuming constant pyrogenic emissions. Shaded bands show Monte-Carlo-derived 95% confidence intervals related to analytical, firn diffusive and source signature uncertainties (Methods). **e**, Calculated theoretical $\delta D-CH_4$ using one-box model output for assumptions of constant geologic (red) and constant pyrogenic (dark blue) emissions. Measured $\delta D-CH_4$ and 1σ measurement uncertainties from WDC ice (this study, light green) and TALDICE (this study and ref. 58, magenta) plotted on the WDC-synchronized gas age scale from refs. 21, 57. Shaded bands in **c–e** show Monte-Carlo-derived 95% confidence intervals related to analytical, firn diffusive and geologic emission and source signature uncertainties (Methods). DO interstadials are labelled and the onset of HE4 is indicated by a vertical dashed line.

to higher baselines across the HE5 and HE4 CH_4 pulses, suggesting a microbial driver of the observed elevated CH_4 after the pulses. Increasing background CH_4 surrounding HE1 is driven by a steady increase in modelled microbial emissions. The model primarily attributes the much larger abrupt CH_4 increases at the onsets of DO8 and DO12 to 20 $Tg\ yr^{-1}$ and 30 $Tg\ yr^{-1}$ increases in microbial emissions, respectively, but also suggests enhanced pyrogenic emissions comparable in magnitude to those modelled during HEs (Fig. 1c,d).

It is important to consider how variables other than changes to the proportion of emission categories might have contributed to observed atmospheric $\delta^{13}C-CH_4$ and $\delta D-CH_4$ variability. First, the relative contributions of different atmospheric CH_4 sinks, and thus sink fractionation, likely changed little across past abrupt CH_4 changes (Methods). Second, the Antarctic $\delta^{13}C-CH_4$ response to atmospheric CH_4 disequilibrium during times of rapid change (Extended Data Fig. 3) and the impact of latitudinal shifts in CH_4 emissions on the mean CH_4 age and thus isotopic

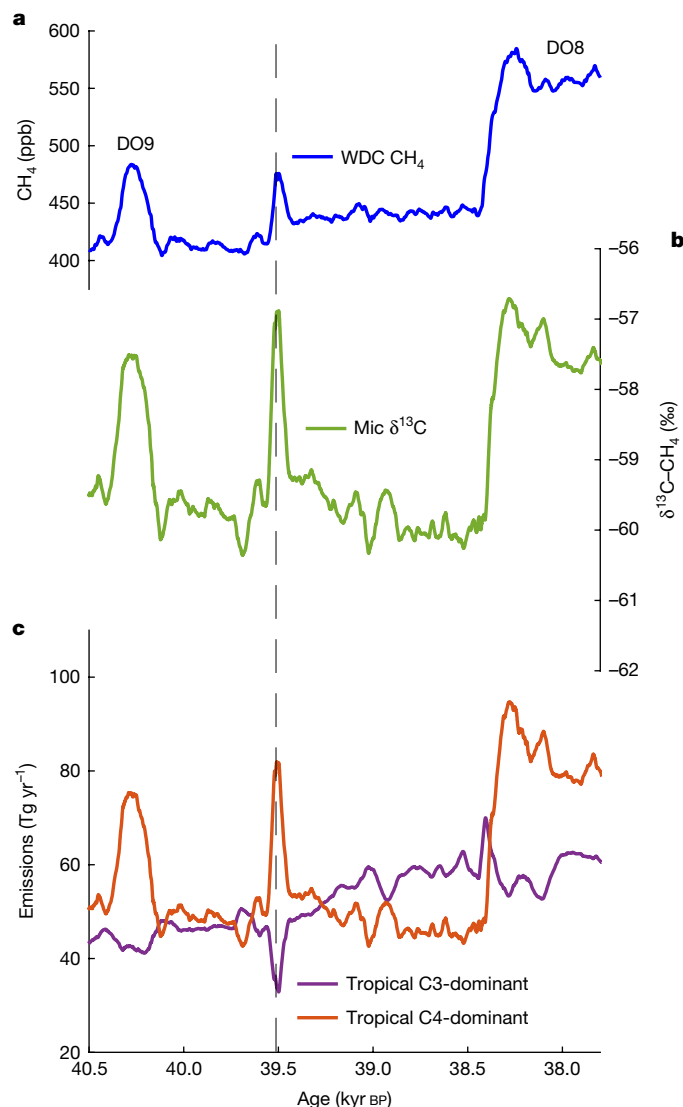


Fig. 3 | Sensitivity of analyses to variability in microbial $\delta^{13}C-CH_4$ across the HE4, DO8 interval. **a**, CFA CH_4 data^{9,58} (blue). **b**, One-box-modelled microbial $\delta^{13}C-CH_4$ assuming constant pyrogenic and geologic $\delta^{13}C-CH_4$ and emissions. **c**, Changes in tropical microbial CH_4 emissions derived from C3-dominant (purple) or C4-dominant (orange) precursor biomass that are required to explain the observed atmospheric CH_4 and $\delta^{13}C-CH_4$ variability, assuming constant pyrogenic and geologic $\delta^{13}C-CH_4$ and emissions (**b**). DO interstadials are labelled and the onset of HE4 is indicated by a vertical dashed line.

composition of CH_4 reaching Antarctica (Extended Data Fig. 4) are both minimal relative to the observed $\delta^{13}C-CH_4$ variations (Methods). These effects are more pronounced for Antarctic $\delta D-CH_4$ because of stronger isotopic sink fractionation. Uncertainties associated with $\delta D-CH_4$ measurement precision²², source signatures and sink fractionation^{20,21} are also high. We therefore limit our interpretation of the $\delta D-CH_4$ datasets to exploring our assumption of stable geologic emissions, rather than use them to make quantitative constraints on the CH_4 budget.

More importantly, atmospheric $\delta^{13}C-CH_4$ is sensitive to changes in the $\delta^{13}C-CH_4$ of microbial emissions. For example, the $\delta^{13}C-CH_4$ of microbial and pyrogenic CH_4 emissions is partially determined by whether the precursor biomass is derived from C3 or C4 photosynthetic pathways²⁰, with C4-derived biomass 12‰ more enriched²⁹. Precipitation changes related to HEs and DO events may have shifted the locations of tropical wetland CH_4 emissions to regions of differing vegetation type^{13,30}, affecting the magnitudes of C3- and C4-derived

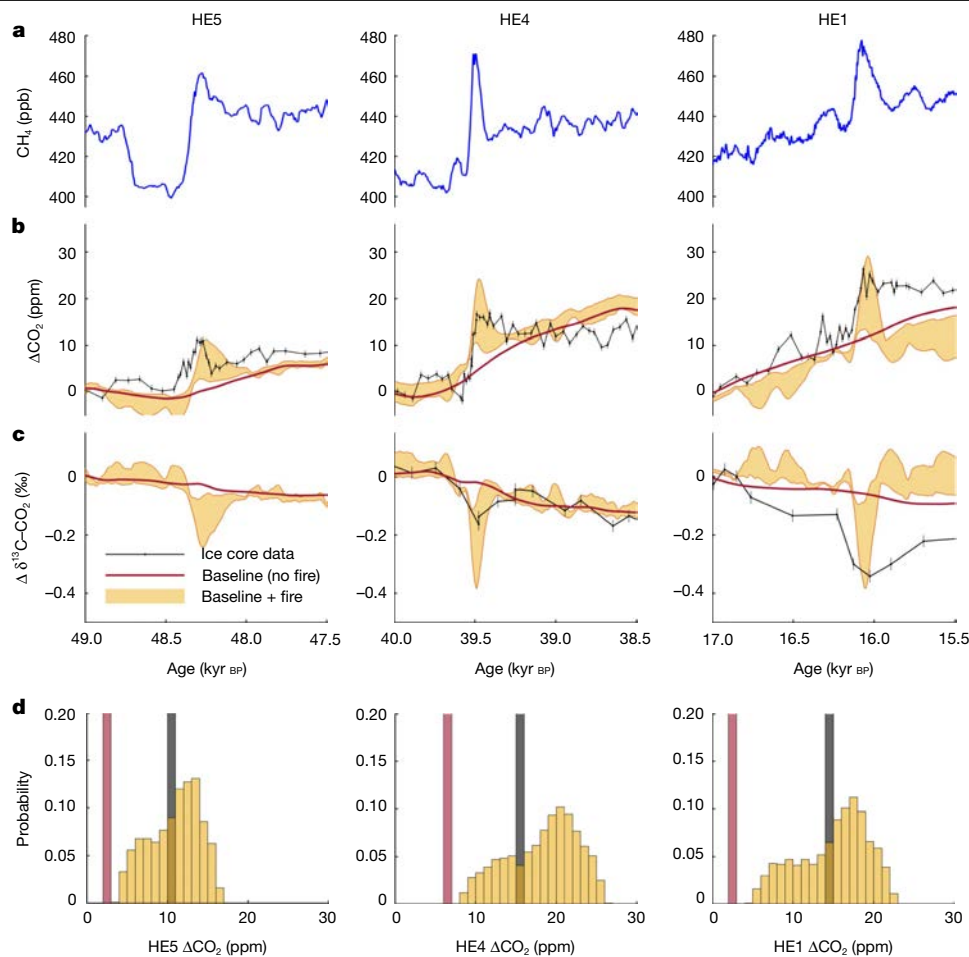


Fig. 4 | Modelled CO₂ across HE5, HE4 and HE1. **a**, WDC CFA CH₄ data (blue)^{9,53}. **b**, Atmospheric CO₂ data and 1σ measurement uncertainties (WDC, black)^{7,49,53,59}, baseline carbon cycle box model output with stable terrestrial-atmosphere carbon flux (red) (model output from ref. 47), and the error envelope of the model output (95% confidence) including CO₂ fluxes related to pyrogenic emissions calculated using our CH₄ pyrogenic emissions (yellow shading). The error envelope was determined using a Monte Carlo error propagation related to uncertainties in pyrogenic CO₂ emissions and post-fire regrowth

times (Methods). Data are plotted relative to CO₂ at the start of each interval. **c**, Same as **b** but for relative changes in δ¹³C–CO₂. Ice core δ¹³C–CO₂ data (plotted with 1σ measurement uncertainties) were measured using ice from Taylor Glacier (TG) and were shifted 150 years younger to match the WDC gas age scale⁷. **d**, Histogram showing the magnitude of CO₂ increase across each HE. Baseline model change (red)⁴⁹, WDC measured change (black) and Monte-Carlo-derived probability density function of the change of the baseline + pyrogenic results, which incorporates all uncertainties described in **b** (yellow).

microbial CH₄ emissions. Specifically, intensified methanogenesis in freshly inundated, C4-dominant regions might have caused the overshoots characteristic of the HE and DO event CH₄ increases¹. Using our one-box model and equations (9) and (10) (Methods), we estimate that C3- and C4-derived microbial CH₄ emissions would need to have decreased by 35% and increased by 75% during HE4, respectively, to explain the observed CH₄ and δ¹³C–CH₄ change (Fig. 3). Similarly, a doubling of C4-derived microbial emissions would be required across the DO8 CH₄ increase.

These shifts in the C3–C4 balance of precursor biomass would have been quite substantial, but the palaeoecological evidence that allows examination of this scenario is limited and equivocal. Proxies of terrestrial precipitation³¹ and paleo-vegetation models³² suggest that the regions that experienced increased precipitation during HEs were both C3-dominant (Amazonia) and C4-dominant (Southern Africa and the Sunda/Sahul Shelves). Similarly, aridification occurred in regions composed of both C4 grasslands (Central Africa) and C3 forests (Eastern Asia)^{31,32}, highlighting the complexity of quantifying this effect. Although we cannot rule this mechanism, a change in pyrogenic emissions can plausibly explain our observations. Therefore, we hereafter explore the implications of this scenario, assuming constant microbial δ¹³C–CH₄ across HEs and DO events.

The terrestrial carbon cycle

The observation of comparable abrupt increases in pyrogenic CH₄ emissions across several HEs and DO events suggests that enhanced BB was a characteristic component of the carbon cycle during the abrupt climate variability of the last glacial period. We propose that during HEs, the southward displacement of tropical rain belts markedly dried regions of the NH tropics^{14,33,34}. The resulting aridification of high biomass ecosystems in previously monsoon-influenced regions provided considerable fuel load for enhanced BB. The establishment of low fuel load ecosystems in the newly arid NH tropical environments^{32,35} supports the modelled reduction of pyrogenic CH₄ emissions immediately following HEs (Fig. 1c). A microbial driver of elevated baseline CH₄ after HE5 and HE4 (Fig. 1d) is consistent with the evidence of enhanced precipitation and wetland expansion in SH tropical regions^{1,13,34}. The microbial driver of increasing CH₄ surrounding HE1 was likely caused by rising global temperatures and the fertilization effects of increasing CO₂ during the last deglaciation³⁶.

Enhanced NH tropical BB during HEs is supported by proxy evidence of an abrupt onset of widespread Afro-Asian megadroughts during HE1 (refs. 33,37). Furthermore, a composite of charcoal records from Chinese Loess Plateau deposits indicates peak regional fire activity

contemporaneous with HE1 (ref. 38). The short duration of HEs limits terrestrial-based evidence of BB variability before HE1. Aerosol-based paleo-fire proxies in polar ice cores (for example, ammonia, formate and black carbon) do not record BB activity beyond polar and mid-latitude regions because of their short atmospheric residence times³⁹. Ice core measurements of longer-lived atmospheric ethane and carbon monoxide offer more hemispheric integrations of BB⁴⁰ but records at sufficient precision and resolution have not yet been measured. Our $\delta^{13}\text{C}-\text{CH}_4$ data thus provide the first direct evidence of a global increase in BB during HEs.

Hypotheses of a dominant microbial driver of the CH_4 increases at the onset of DO12 and DO8 by enhanced NH tropical wetland emissions are consistent with our $\delta^{13}\text{C}-\text{CH}_4$ record and box model analyses^{11,14,30,41}. The microbial CH_4 response to shifting precipitation was greater for DO events than for HEs because NH tropical regions contain greater land area conducive to wetland development^{1,30}. This is in line with the proposed relationship between atmospheric CH_4 and global terrestrial ecosystem productivity^{2,11,13,25}. These ecosystem and regional microbial CH_4 emission shifts are supported by records of the CH_4 inter-polar difference³⁶, earth system modelling studies^{15,42} and ASM speleothem records^{14,34}.

We link the increases in $\delta^{13}\text{C}-\text{CH}_4$ -derived pyrogenic emissions at the onset of DO interstadials (Fig. 1c) to enhanced BB from both rapid drying in SH tropical regions^{43,44} and increased NH vegetation cover that was susceptible to combustion during the dry season^{39,45,46}. Evidence of enhanced BB during DO events is found in both global⁴⁶ and SH tropical⁴³ charcoal compilations. Similar to HEs, Chinese Loess Plateau charcoal records show enhanced BB contemporaneous with several late-glacial DO events³⁸. Furthermore, more frequent ammonium deposition events in Greenland during DO events have been linked to increased North American BB^{39,46}.

The significant increases in pyrogenic CH_4 emissions during both HEs and DO events suggest major and abrupt changes to global ecosystem structure, terrestrial biogeochemistry and carbon cycling. Rapid atmospheric CO_2 increases of up to 15 ppm have been linked to HEs and were contemporaneous with the abrupt CH_4 pulses^{7,47,48}. Ice core records of atmospheric $\delta^{13}\text{C}-\text{CO}_2$ show abrupt isotopic depletion synchronous with the HE1 and HE4 CO_2 increases, suggesting organic carbon of terrestrial or marine origin^{7,49}. An oceanic source has received the most attention, with compelling, although resolution-limited, evidence of rapid Southern Ocean overturning of CO_2 -rich deep water that abruptly outgassed and increased atmospheric CO_2 (refs. 48,50). Our $\delta^{13}\text{C}-\text{CH}_4$ record and calculated pyrogenic emissions provide evidence of an important terrestrial CO_2 source.

We explore this possibility using a 14-box, isotope-enabled ocean, atmosphere and terrestrial biosphere carbon cycle model that is forced by proxies of ocean circulation and climate derived from ocean sediments and ice cores⁴⁹ (Methods) to quantify the impact of enhanced pyrogenic emissions on atmospheric CO_2 . Previously, with terrestrial carbon exchange held constant, the model captured glacial-interglacial and millennial-scale CO_2 and $\delta^{13}\text{C}-\text{CO}_2$ variability by Southern Ocean mechanisms with comparable accuracy to state-of-the-art carbon cycle models but failed to reproduce the abrupt changes related to HEs⁷. Here, we varied terrestrial carbon fluxes using our $\delta^{13}\text{C}-\text{CH}_4$ -derived pyrogenic CH_4 emissions and modern CO_2 -to- CH_4 emission ratios to calculate global pyrogenic CO_2 emissions and eventual post-fire regrowth (Methods). The resulting time series was then input into the model as a terrestrial carbon flux to the atmosphere.

Including our estimates of pyrogenic CO_2 emissions markedly improves the agreement of the model with CO_2 and $\delta^{13}\text{C}-\text{CO}_2$ across HEs (Fig. 4). We estimate that enhanced BB contributed to 5–15 ppm (33–100%) of the abrupt CO_2 increases associated with HE5, 4, and 1 and that these BB events combusted an additional 285–637 million hectares per year at their peak; equivalent to 1/2 to roughly all of modern annual global burned area (Methods). Shortcomings of the model

are exemplified by overestimates of the smaller abrupt CO_2 increases at DO interstadial onsets⁴⁷. This disagreement could stem from the assumption of the model of stable terrestrial carbon reservoirs outside of changes associated with BB. Increased NH tropical vegetation cover during DO interstadials^{30,32,45} is not captured in the model but would offset the carbon released from enhanced BB (Methods and Extended Data Fig. 5). We propose that this offsetting effect was minimal during stadial intervals when terrestrial carbon accumulation was probably minor and limited to the SH tropics, as suggested by several paleo-ecological modelling studies^{32,51}.

The agreement between modelled and observed CO_2 and $\delta^{13}\text{C}-\text{CO}_2$ across HEs suggests that our $\delta^{13}\text{C}-\text{CH}_4$ data and CH_4 model output provide plausible estimates of changes to the CH_4 budget and global BB. Furthermore, our data offer empirical evidence that the terrestrial biosphere played a key part in multi-decadal-scale CO_2 increases associated with HEs, although we cannot rule out an important oceanic contribution⁴⁸. Vegetation-coupled earth system models have been run to equilibrium in climate states with both active and collapsed AMOC to simulate the response of terrestrial carbon to HEs^{30,32,51,52}. The resulting net terrestrial carbon loss (28–91 Pg) (ref. 32) is comparable to the loss implied by our modelled transient excess pyrogenic carbon emissions (43–146 Pg), suggesting that our estimates are reasonable. Our time series may provide a valuable comparison for future paleo-ecological studies that simulate the transient response of the terrestrial biosphere to HEs. The findings of this study show that global fire regimes varied contemporaneously and substantially with past climate changes over very short time periods and reveal a new mechanistic relationship between CH_4 and CO_2 , the terrestrial carbon cycle and abrupt climate change.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41586-024-08363-3>.

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Methods

Ice core measurements

The ice core CH₄ stable isotope data presented in this study were measured using analytical systems at the University of Bern^{22,60,61} and Oregon State University (OSU). The OSU system is modelled after the Bern system and consists of an extraction line for separating CH₄ from other air components and measuring the total amount of air in the sample, a combustion system to produce CO₂ from CH₄ (Thermo GC-IsoLink) and a Thermo Delta V mass spectrometer configured to measure $\delta^{13}\text{C}-\text{CH}_4$ (for further details, see Extended Data Fig. 6 and Supplementary Information section 1).

The pooled standard deviation of replicate $\delta^{13}\text{C}-\text{CH}_4$ measurements is comparable between the OSU system ($1\sigma = 0.14\text{‰}$ for 250 g samples) and the Bern system ($1\sigma = 0.15\text{‰}$ for 150 g samples)⁶⁰. For $\delta\text{D}-\text{CH}_4$, Bern reports a 1σ precision of 2.1‰ for 300 g samples^{22,61}. Both systems are calibrated to the VPDB-CO₂ scale (Supplementary Information section 2).

On the OSU system, WDC samples were measured for $\delta^{13}\text{C}-\text{CH}_4$ across HE5 and DO12 from 49.6 kyr to 46.8 kyr (43 samples at 67-year mean resolution) and HE1 from 17.0 kyr to 15.3 kyr (41 samples at 40-year mean resolution). The Bern system was used to measure ice samples from the WAIS Divide Replicate Core^{62,63}, covering HE4 and DO8 for both $\delta^{13}\text{C}-\text{CH}_4$ (40.5–37.8 kyr; 43 samples at 63-year mean resolution) and $\delta\text{D}-\text{CH}_4$ (39.7–39.2 kyr; 15 samples at 38-year mean resolution). The WAIS Divide Replicate Core is offset in depth from WDC by 0.7375 m for the depth range we analysed. Finally, 11 samples from TALDICE were measured for $\delta\text{D}-\text{CH}_4$ across the DO8 transition on the Bern system, with a mean resolution of 110 years. The TALDICE gas age scale from refs. 57,58 was used to synchronize the TALDICE record to the WDC gas age scale (Fig. 2a,e). Although these HEs and DO events were prioritized because of their prominence and the notable labour involved in measurements, future studies should investigate other abrupt CH₄ changes to better define the recurrence of our observed isotopic anomalies.

The measured $\delta^{13}\text{C}-\text{CH}_4$ in ice core samples must be corrected for fractionation in the porous firn column. First, gravitational fractionation enriches the bottom of the firn column with $^{13}\text{CH}_4$ (ref. 64). This effect is quantified using $\delta^{15}\text{N}-\text{N}_2$ measurements^{64,65}, which have been made at high resolution in WDC samples⁵³. The gravitational fractionation correction based on $\delta^{15}\text{N}-\text{N}_2$ is about 0.3‰ in WDC. Second, $^{13}\text{CH}_4$ and $^{12}\text{CH}_4$ diffuse at different rates, causing a diffusive fractionation when concentration gradients are present in the firn, for example, at times of abrupt atmospheric CH₄ change. This effect was corrected for by using the methods described in ref. 66. The high-resolution of WDC CFA CH₄ data¹ provides robust quantifications of this effect across the abrupt CH₄ change investigated in this study. The CFA data were first smoothed using a Gaussian smoothing window of 20 years to dampen analytical noise that caused unrealistic estimates of instantaneous rates of CH₄ change. The diffusive column height and gas age at lock-in, two variables needed to calculate diffusive fractionation, were both determined using $\delta^{15}\text{N}-\text{N}_2$ (refs. 66,67). We find that, during the intervals of the most rapid CH₄ change, diffusive fractionation imparts a maximum −0.8‰ anomaly on the $\delta^{13}\text{C}-\text{CH}_4$ signal trapped in the ice, highlighting the importance of these corrections. However, the measured positive anomalies in $\delta^{13}\text{C}-\text{CH}_4$ are observable in data both corrected and not corrected for diffusive fractionation and are thus not dependent on the accuracy of this correction (Extended Data Fig. 1). The WDC and TALDICE $\delta\text{D}-\text{CH}_4$ data were also corrected for gravitational and diffusive fractionation using $\delta^{15}\text{N}-\text{N}_2$ (refs. 56,66,67), although these effects are small relative to δD measurement precision and signal amplitude.

The 1σ total uncertainties for each measurement were calculated by propagating uncertainties associated with gravitational fractionation (0.04‰), diffusive fractionation (30% of the calculated value⁶⁶) and analytical error (0.14‰). The largest uncertainties (0.24‰) occur

during intervals of abrupt CH₄ change when diffusive fractionation is the greatest but do not limit conclusive interpretations of the data.

Box model analyses

To interpret our CH₄ stable isotope data, we used a one-box model⁶. These models are advantageous because they offer a simplified yet well-constrained framework for understanding complex CH₄ biogeochemistry^{68,69}. We first averaged replicate samples and interpolated our $\delta^{13}\text{C}-\text{CH}_4$ record to match the resolution of the 20-year Gaussian-smoothed WDC CH₄ CFA data. Latitudinal gradients in CH₄ (the Interpolar Difference, or IPD) and $\delta^{13}\text{C}-\text{CH}_4$ may have existed during the investigated time intervals^{11,36,70,71}. However, owing to limited IPD measurements and centennial-scale uncertainties³⁶, we assume a stable IPD at 2% of total atmospheric CH₄ (refs. 11,36,70) and multiply the WDC CH₄ record by a factor of 1.01 to estimate the global mean burden. This assumption is not impactful to our interpretations of the CH₄ budget. Two-box model sensitivity tests (see below) indicate that Antarctic $\delta^{13}\text{C}-\text{CH}_4$ was minimally affected by possible past IPD variability³⁶ (Extended Data Fig. 4). The smoothed CH₄ record was then used to calculate the total source ($^{12}\text{CH}_4 + ^{13}\text{CH}_4$) by solving for S in equation (1):

$$\frac{dX_n}{dt} = S - \frac{X_n}{L} \quad (1)$$

where S is the annual global CH₄ source (Tg yr^{−1}), L is the atmospheric lifetime (9 years) (ref. 72) and X is the atmospheric CH₄ burden (Tg of all isotopologues) at timestep n , calculated using the following mass balance:

$$X = \frac{M_A}{MM_A} \times \frac{\text{CFA} \times 1.01}{10^9} \times \frac{MM_{\text{CH}_4}}{10^{12}} \quad (2)$$

where M_A is the mass of the atmosphere (5.15×10^{21} g), MM_A is the molar mass of the atmosphere (28.97 g mol^{-1}), CFA is the smoothed Antarctic CH₄ CFA record and MM_{CH_4} is the molar mass of CH₄ (16.04 g mol^{-1}). Equation (1) can be modified to calculate the $^{13}\text{CH}_4$ source ($S^{13\text{C}}$) at each timestep:

$$\frac{dX_n^{13\text{C}}}{dt} = S^{13\text{C}} - \frac{X_n^{13\text{C}}}{L} \times \alpha \quad (3)$$

where $X^{13\text{C}}$ refers to the atmospheric $^{13}\text{CH}_4$ burden and α is the kinetic fractionation factor of the combined effect of all CH₄ sinks (reaction with OH, chlorine and soil oxidation; 0.994) (refs. 73,74). Changes in the relative contributions of different sinks could affect $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$. For example, increased mineral dust interaction with sea salt aerosols during stadial intervals could have increased the chlorine sink⁷⁵, causing stronger sink fractionation and $\delta^{13}\text{C}-\text{CH}_4$ enrichment. This may explain some of the multi-millennial-scale $\delta^{13}\text{C}-\text{CH}_4$ variability identified in refs. 21,41. Although low-latitude dust archives lack multi-decadal-scale resolution, the abrupt CH₄ pulses and $\delta^{13}\text{C}-\text{CH}_4$ anomalies are not accompanied by pulses of mineral dust and sea salt aerosol in Greenland and Antarctic ice cores^{19,76,77}. We, therefore, assume that the chlorine sink, lifetime and sink fractionation were stable over the short intervals investigated in this study⁷².

The $^{13}\text{CH}_4$ atmospheric burden, or $X^{13\text{C}}$ in equation (3), is calculated from our measured, interpolated atmospheric $\delta^{13}\text{C}-\text{CH}_4$ record ($\delta^{13}\text{C}$) using equation (4):

$$X^{13\text{C}} = \left(\frac{\delta^{13\text{C}}}{1,000} + 1 \right) \times R_{\text{std}} \times X \quad (4)$$

where R_{std} is the VPDB standard for $\frac{^{13}\text{C}}{^{13}\text{C} + ^{12}\text{C}}$. We then calculate the $\delta^{13}\text{C}-\text{CH}_4$ of the global CH₄ source ($\delta^{13}\text{C}_s$) using equation (5):

$$\delta^{13}\text{C}_S = \left(\frac{R_S^{13}\text{C}}{R_{\text{std}}^{13}\text{C}} - 1 \right) \times 1,000 \quad (5)$$

where $R_S^{13}\text{C}$ is the $\frac{^{13}\text{C}}{^{12}\text{C}}$, or $\frac{S^{13}\text{C}}{S}$, of the global CH_4 source. With the strength (S) and $\delta^{13}\text{C}-\text{CH}_4$ ($\delta^{13}\text{C}$) of the global CH_4 source known, the following isotope mass balance system of equations can be used to estimate microbial (Mic), geologic (Geo) and pyrogenic (Pyro) emissions:

$$S = S_{\text{Mic}} + S_{\text{Pyro}} + S_{\text{Geo}} \quad (6)$$

$$S \times \delta^{13}\text{C}_S = S_{\text{Mic}} \times \delta^{13}\text{C}_{\text{Mic}} + S_{\text{Pyro}} \times \delta^{13}\text{C}_{\text{Pyro}} + S_{\text{Geo}} \times \delta^{13}\text{C}_{\text{Geo}} \quad (7)$$

where the subscripts denote each emission type (Extended Data Table 1). Solving the system of equations, we obtain estimates of pyrogenic and microbial emissions in Tg yr^{-1} (Fig. 1c,d) assuming geologic emissions of 2.5 Tg yr^{-1} ($1\sigma = 1.3 \text{ Tg yr}^{-1}$).

The total uncertainty of each measurement is incorporated into our box model using a 10,000-iteration Monte Carlo approach. Because our interpretations primarily focus on relative changes to the CH_4 budget over time, and because we assume stable $\delta^{13}\text{C}-\text{CH}_4$ of major sources over multi-decadal timescales, we show emission estimates as relative change over time (Figs. 1 and 2). This enables us to isolate uncertainties in the global CH_4 budget from uncertainties in how the budget changes over time.

To explore our assumption of stable geologic emissions, the magnitudes of emission types were calculated for two scenarios using equations (6) and (7) by assuming both stable geologic and pyrogenic emissions. Equation (8) (see Extended Data Table 1 for source signatures)²¹ was then used to estimate the $\delta\text{D}-\text{CH}_4$ of the total source (δD_S):

$$S \times \delta\text{D}_S = S_{\text{Mic}} \times \delta\text{D}_{\text{Mic}} + S_{\text{Pyro}} \times \delta\text{D}_{\text{Pyro}} + S_{\text{Geo}} \times \delta\text{D}_{\text{Geo}} \quad (8)$$

The resulting time series of S and δD_S were then used with δD -enabled versions of equations (1–5) ($\alpha = 0.7634$) (refs. 73,78) to calculate atmospheric $\delta\text{D}-\text{CH}_4$ for the HE4–DO8 interval for both emission scenarios (Fig. 2). Under stable pyrogenic emissions, $\delta\text{D}-\text{CH}_4$ would have experienced +15–20‰ anomalies across HE4 and DO8 that were driven by enhanced geologic emissions. These anomalies could have been larger if previously proposed marine clathrate destabilization^{27,28} drove the CH_4 increases due to isotopic enrichments during water column oxidation before atmospheric release^{79,80}.

Sensitivity tests

Changes in source CH_4 isotopic signature. To quantify how much microbial $\delta^{13}\text{C}-\text{CH}_4$ would need to have changed to cause the observed CH_4 and $\delta^{13}\text{C}-\text{CH}_4$ variability across HE4 and the onset of DO8, we assumed constant pyrogenic and geologic emissions and solved equations (6) and (7) for S_{Mic} and $\delta^{13}\text{C}_{\text{Mic}}$. We estimate that abrupt roughly 4‰ enrichments in $\delta^{13}\text{C}_{\text{Mic}}$ would be required across both abrupt CH_4 increases (Fig. 3b). This shift could only occur through a marked change in the C3:C4 ratio of methanogenesis precursor biomass. We established a new isotope mass balance to determine C3- and C4-dominant components of microbial emissions (see Extended Data Table 1 for source signatures):

$$S_{\text{Mic}} = S_{\text{HL}} + S_{\text{C3}} + S_{\text{C4}} \quad (9)$$

$$S_{\text{Mic}} \times \delta^{13}\text{C}_{\text{Mic}} = S_{\text{HL}} \times \delta^{13}\text{C}_{\text{HL}} + S_{\text{C3}} \times \delta^{13}\text{C}_{\text{C3}} + S_{\text{C4}} \times \delta^{13}\text{C}_{\text{C4}} \quad (10)$$

where S_{HL} is the strength of high-latitude NH microbial emissions (assumed stable at 10 Tg yr^{-1}) (ref. 10). The results (Fig. 3c and Extended Data Fig. 2) are minimally sensitive to the assumed strengths of high-latitude NH microbial, geologic and pyrogenic emissions.

Using similar methods, we find that the scenario in which only S_{Mic} and $\delta^{13}\text{C}_{\text{Pyro}}$ are allowed to vary, which might arise because of changes in the C3:C4 ratio of pyrogenic precursor material⁸¹, results in unrealistic changes to $\delta^{13}\text{C}_{\text{Pyro}}$.

Firn smoothing and isotopic disequilibrium. When CH_4 increases, atmospheric disequilibrium causes a depletion of $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$ because sinks remove the lighter isotopologue faster^{6,82,83}. Although this effect is incorporated into our one-box model analyses, we visualized its impact by running our model forward across the HE4–DO8 interval using S from equation (1) and constant isotopic source signatures of –50‰ and –290‰ for $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$, respectively. For $\delta^{13}\text{C}-\text{CH}_4$, this effect is small (at most 0.2‰) relative to observed variability (Extended Data Fig. 3b). Owing to stronger sink fractionation, the effect is more pronounced for $\delta\text{D}-\text{CH}_4$ (up to 8‰) and is comparable in magnitude to observed variability across HE4 and DO8 (Extended Data Fig. 3c). However, the true rate of CH_4 change may have been 50% higher than what is captured by WDC because of the smoothing effects of the firn column^{9,64}, causing our box model to underestimate the atmospheric disequilibrium effect by up to 50%. The largest disequilibrium effect for $\delta^{13}\text{C}-\text{CH}_4$ in our box model is 0.2‰, so it may be underestimated by at most 0.1‰. This is below our reported analytical precision and does not notably affect our analyses.

Because the amplitude of the atmospheric CH_4 pulses observed in WDC during HEs may be smoothed, it is likely that the pyrogenic emission pulses were larger in amplitude and shorter in duration than what is captured in our box model. Within each HE interval, box-modelled annual pyrogenic emissions increase by a maximum of 6–10 Tg yr^{-1} (Fig. 1c). Assuming the amplitude of the HE CH_4 pulses were dampened by 33% in WDC⁹, we estimate that the true maximum increase in annual pyrogenic emissions during HEs was 9–15 Tg yr^{-1} (Extended Data Table 2). Because the CH_4 changes during DO events were stepwise, firn smoothing minimally affects modelled pyrogenic emission variability. To calculate the per cent change in global pyrogenic emissions (main text), we assume that baseline glacial CH_4 pyrogenic emissions were 10 Tg yr^{-1} , in line with the estimates from modelling studies^{15,42,84,85}. Note that the absolute value of our per cent change estimates is highly sensitive to this baseline estimate and uncertainties related to firn smoothing⁹. Although firn processes dampen the amplitude of HE CH_4 pulses, the integrated magnitude of the CH_4 pulse is fully captured in WDC. We, therefore, estimate that pyrogenic pulses across each HE released 438–739 Tg of CH_4 over the course of 100–250 years, depending on the HE (Extended Data Table 2).

Latitudinal shifts in sources. Latitudinal shifts in CH_4 emissions affect the mean age of atmospheric CH_4 at ice core sites and thus measured $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$ because of sink fractionation^{4,6,21}. These shifts are tracked by the past relative inter-pole difference (rIPD):

$$\text{rIPD} = \left(\frac{\text{CH}_{4,\text{N}}}{\text{CH}_{4,\text{S}}} - 1 \right) \times 100\% \quad (11)$$

where $\text{CH}_{4,\text{N}}$ is NH CH_4 and $\text{CH}_{4,\text{S}}$ is SH CH_4 (ref. 86). For example, northward shifts of CH_4 emissions increase the age of atmospheric CH_4 in the SH, resulting in positive isotopic shifts over Antarctica that are not reflective of changes in source mixture. This effect was explored using a two-box model framework⁶ with an interhemispheric exchange rate of 1 year (ref. 87). For both $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$, the model was run forward with different ratios of NH to SH emissions but equal source signatures and sink fractionation factors from zero atmospheric burden to equilibrium. The results are plotted with NH and SH CH_4 isotopic signatures as a function of the resulting rIPD (Extended Data Fig. 4).

Based on constraints from existing ice-core-derived CH_4 rIPD data^{36,71}, we find that this mechanism would have affected Antarctic $\delta^{13}\text{C}-\text{CH}_4$ by at most 0.2‰. Specifically, ice core records suggest that the rIPD was about

3 percentage points higher during DO interstadials than stadials^{11,24,70}, which would have minimally impacted Antarctic $\delta^{13}\text{C}-\text{CH}_4$ (<0.1‰ at DO onsets) and our box model interpretations. HE-related CH_4 changes are too short in duration to accurately measure the rIPD (ref. 36), although we suspect that rIPD-induced $\delta^{13}\text{C}-\text{CH}_4$ changes were also small. The effect is more pronounced for $\delta\text{D}-\text{CH}_4$ (up to 8‰), which is comparable to the magnitude of observed $\delta\text{D}-\text{CH}_4$ variability (Extended Data Fig. 4).

CO₂ modelling

The 14-box ocean, atmosphere, and terrestrial biosphere carbon cycle model described in refs. 7,49 contains physical and biogeochemical constraints that realistically simulate interactions and lags in the carbon cycle. The model is ideal for testing how multiple components of the carbon cycle can combine to modulate atmospheric CO₂, without the added complexity and assumptions of more computationally expensive earth system models. The baseline scenario presented here (Fig. 4) was previously derived by forcing the model with climate records from Greenland and Antarctica, greenhouse gas forcing calculated from ice core gas records and Pa/Th records from the Bermuda Rise as proxies for AMOC strength^{7,49}. The strength of each forcing was varied in a Monte Carlo approach to best match ice core CO₂ and $\delta^{13}\text{C}-\text{CO}_2$ data over the past 65,000 years. We use this best-fit scenario as a baseline to perform our experiments, which test how the addition of BB fluxes may have contributed to some of the abrupt, enigmatic features of the CO₂ record that were not previously captured in the model.

Estimates of CO₂:CH₄ pyrogenic emission ratios were used to calculate CO₂ emissions from our box-modelled CH₄ emissions⁸⁸. Because we report pyrogenic CH₄ emissions as relative change, we estimated total pyrogenic emissions by uniformly adjusting the HE5, HE4 and HE1 time series to initialize at 10 Tg yr⁻¹. CO₂:CH₄ emission ratios are well-characterized by modern observations but differ among ecosystems and fire regimes^{88–90}. Uncertainties in tropical and mid-latitude vegetation and where burning occurred across HEs and DO events³² limit our ability to pinpoint past CO₂:CH₄ emission ratios. We thus conservatively use a range derived from the mean and standard deviation of annual global emission ratios (125–170 gC gCH₄⁻¹ from 1997 to 2022)^{89,90}.

The resulting time series of pyrogenic CO₂ emissions were incorporated into the carbon cycle model as a terrestrial flux to the atmosphere with a $\delta^{13}\text{C}-\text{CO}_2$ of -24‰. Following the pulses of increased BB, carbon stocks return to pre-pulse levels by imposing increases in ecosystem productivity with a 10–100-year e-folding timescale to represent the regrowth of ecosystems ranging from shrubland to mature forest. Uncertainties in pyrogenic CO₂ emissions and regrowth timescales were incorporated into the model assuming a 1,000-iteration Monte Carlo approach. The model was run from 65 kyr to 10 kyr with pyrogenic CO₂ emissions held constant between intervals for which estimates were available to ensure model equilibration (Fig. 4).

Several uncertainties are not incorporated into our model that could influence estimates of the impact of BB on atmospheric CO₂. First, although we incorporate absolute source signature and atmospheric $\delta^{13}\text{C}-\text{CH}_4$ uncertainties into our one-box model, uncertainties related to temporal variability in source signature (Fig. 3 and Extended Data Fig. 2) are not included (see sensitivity tests). Specifically, if the observed $\delta^{13}\text{C}-\text{CH}_4$ enrichments were partially driven by increases in microbial CH₄ emissions derived from ¹³C-enriched C4 precursor biomass, modelled pyrogenic CH₄ and thus CO₂ emissions may be overestimated during HEs and DO events. Second, we assume that all terrestrial carbon combusted and emitted to the atmosphere is eventually regrown. However, the regional aridification induced by abrupt shifts in precipitation may have resulted in lower carbon biomass in newly established, post-fire ecosystems during HEs and DO events³². Carbon may have accumulated in regions in which precipitation increased, although terrestrial carbon loss in the drying, land-dominated NH tropics probably outweighed gains in the moister SH tropics during HEs^{30,32,51,52}. The opposite was probably true during DO interstadials, with carbon uptake in the NH

tropics from increased precipitation exceeding carbon loss in the land-limited SH tropics and leading to net terrestrial carbon uptake not captured by our model^{30,32,91} (Extended Data Fig. 5). To prevent the over-parameterization of the model with poorly constrained variables, attempts to quantify terrestrial carbon fluxes other than those from BB emissions were not included outside of Extended Data Fig. 5, but their discussion is important for model interpretation.

At the peak of HE4 and HE1, our model indicates a 0.75–1.70 Pg yr⁻¹ increase in pyrogenic carbon emissions. Assuming the amplitude of the approximately 150-year HE pyrogenic pulses were dampened by 33% in WDC⁹ (see above), the true maximum pyrogenic carbon emission increase more likely ranged from 1.12 Pg yr⁻¹ to 2.55 Pg yr⁻¹. Using the global mean modern ratio of 4 tonnes of carbon per hectare of burned area⁹⁰, we estimate that 285–637 million more hectares were burnt each year globally during HEs. This rough approximation is sensitive to the type of vegetation burned⁹² but helps to conceptualize the geographic extent of the enhanced BB implied by our $\delta^{13}\text{C}-\text{CH}_4$ record.

Data availability

The WDC $\delta^{13}\text{C}-\text{CH}_4$ data measured on the OSU analytical system across HE5, DO12 and HE1 are publicly available and archived at the USAP DC (<https://doi.org/10.15784/601683>) and the NOAA National Climatic Data Center (<https://www.ncdc.noaa.gov/access/paleo-search/study/39699>). The WDC $\delta^{13}\text{C}-\text{CH}_4$ data and $\delta\text{D}-\text{CH}_4$ data and the TALDICE $\delta\text{D}-\text{CH}_4$ measured at the University of Bern across HE4 and DO8 are publicly available on the USAP DC (TALDICE $\delta\text{D}-\text{CH}_4$: <https://www.usap-dc.org/view/dataset/601814>; WDC $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$: <https://www.usap-dc.org/view/dataset/601813>) and the NOAA National Climatic Data Center (TALDICE $\delta\text{D}-\text{CH}_4$ and WDC $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$: <https://www.ncdc.noaa.gov/access/paleo-search/study/39699>). Box model and carbon-cycle model outputs are publicly available at Zenodo (<https://doi.org/10.5281/zenodo.13974600>; ref. 93) and can be found in the source data figure files associated with the online version of this paper. Source data are provided with this paper.

Code availability

The code used to run the methane isotope box model and plot the results in main text Figs. 1–4 is archived and publicly available at GitHub (https://github.com/benryoung23/Riddell-Young_etal_2024v2). The entire suite of Monte Carlo simulations and box model code used for the carbon cycle model simulations is publicly available at Zenodo (<https://doi.org/10.5281/zenodo.13861216>; ref. 94).

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Competing interests The authors declare no competing interests.

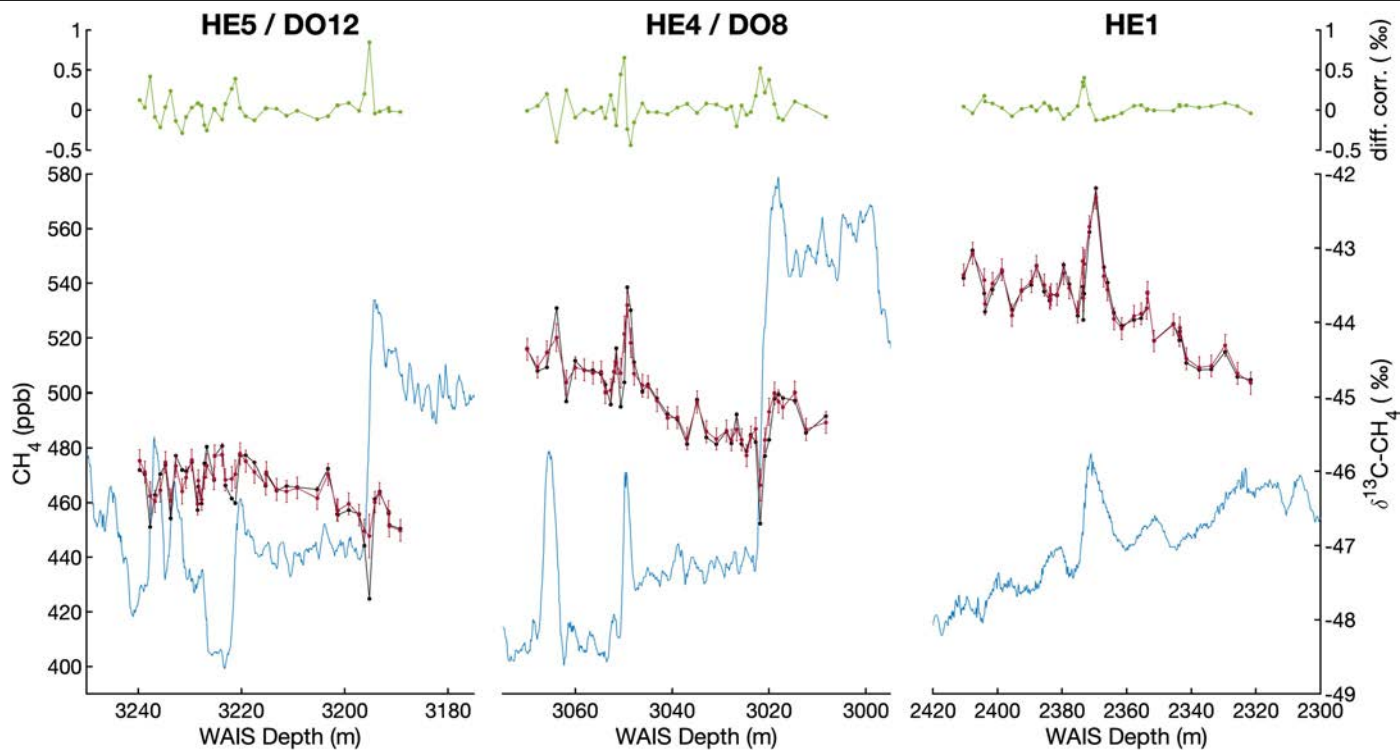
Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41586-024-08363-3>.

Correspondence and requests for materials should be addressed to Ben Riddell-Young.

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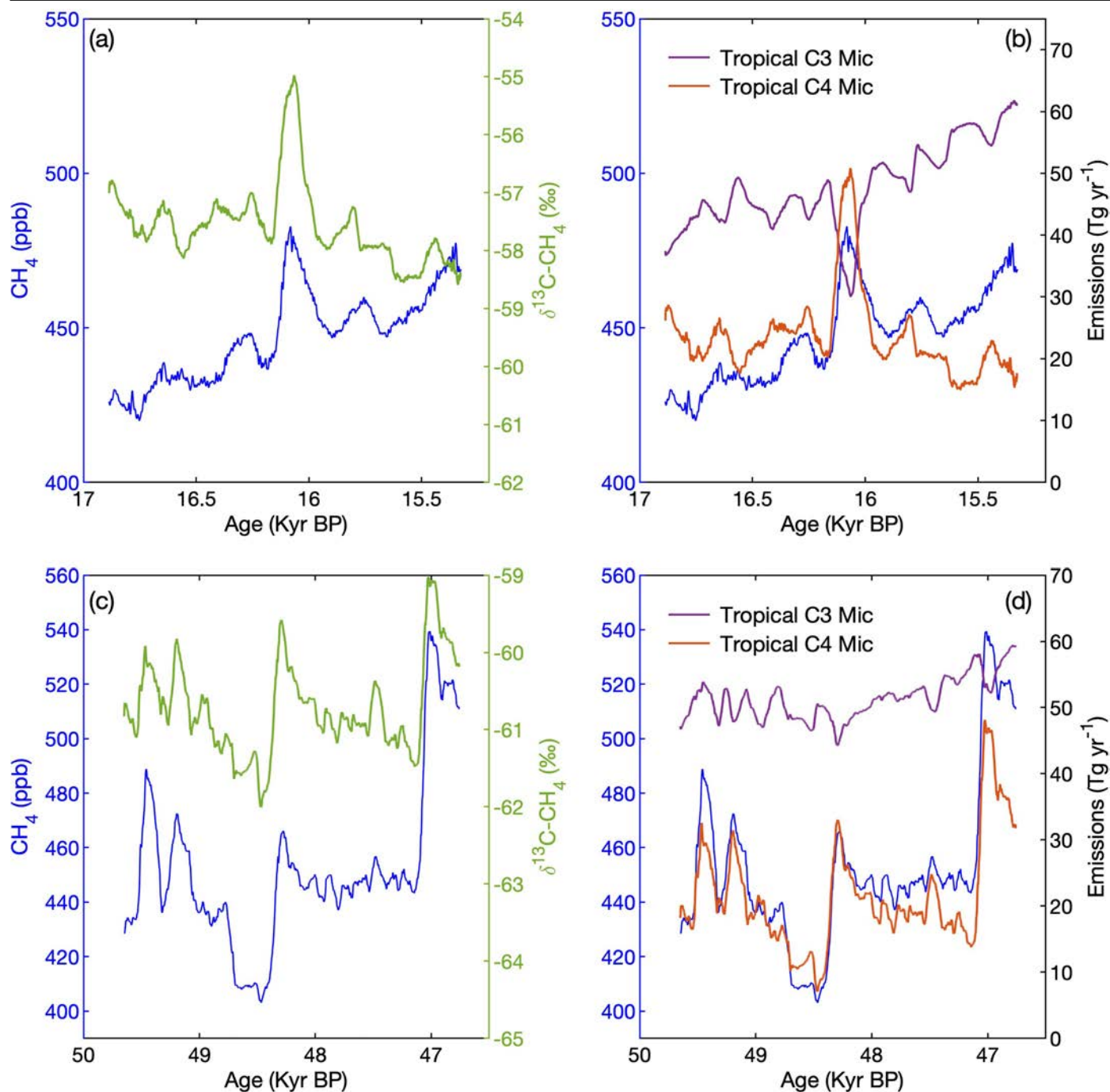
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Extended Data Fig. 1 | Raw and fractionation-corrected $\delta^{13}\text{C}-\text{CH}_4$ data.

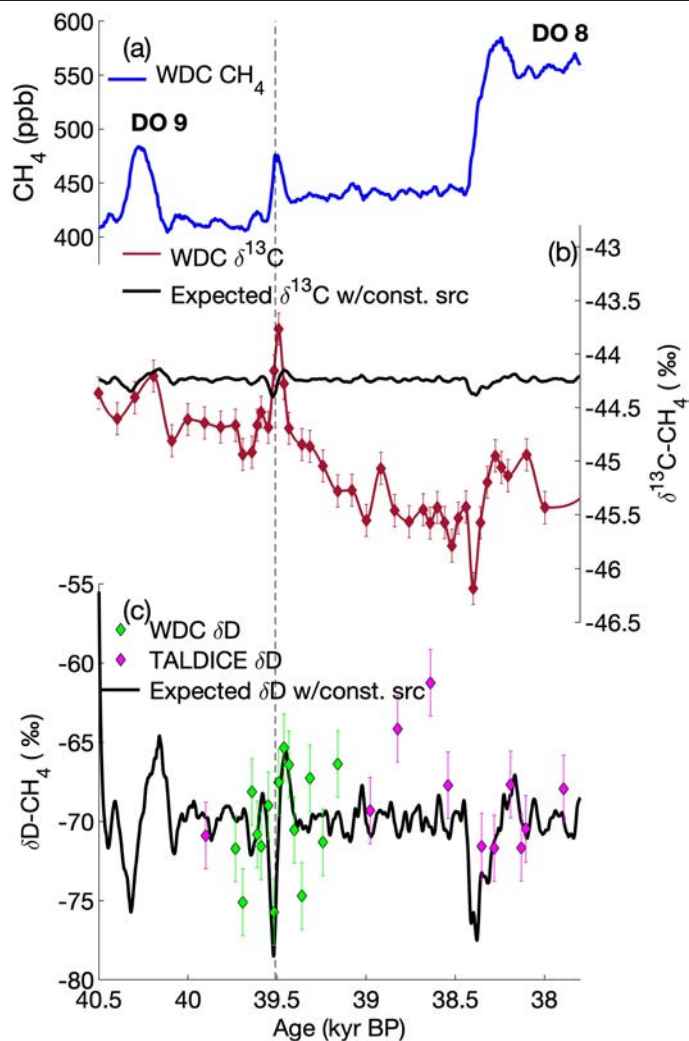
Gravitational fractionation-corrected $\delta^{13}\text{C}-\text{CH}_4$ data (black) and gravitational and diffusional corrected $\delta^{13}\text{C}-\text{CH}_4$ data (maroon) for HE5/DO12 (left), HE4/DO8 (middle) and HE1 (right) plotted on the WAIS Divide depth scale. One-sigma analytical uncertainties, calculated using measurements precision and

uncertainties related to the fractionation corrections, are plotted as maroon error bars for each data point. The WDC CH₄ CFA record is plotted in light blue in each subplot¹. The diffusional correction is plotted above in green for each data point.

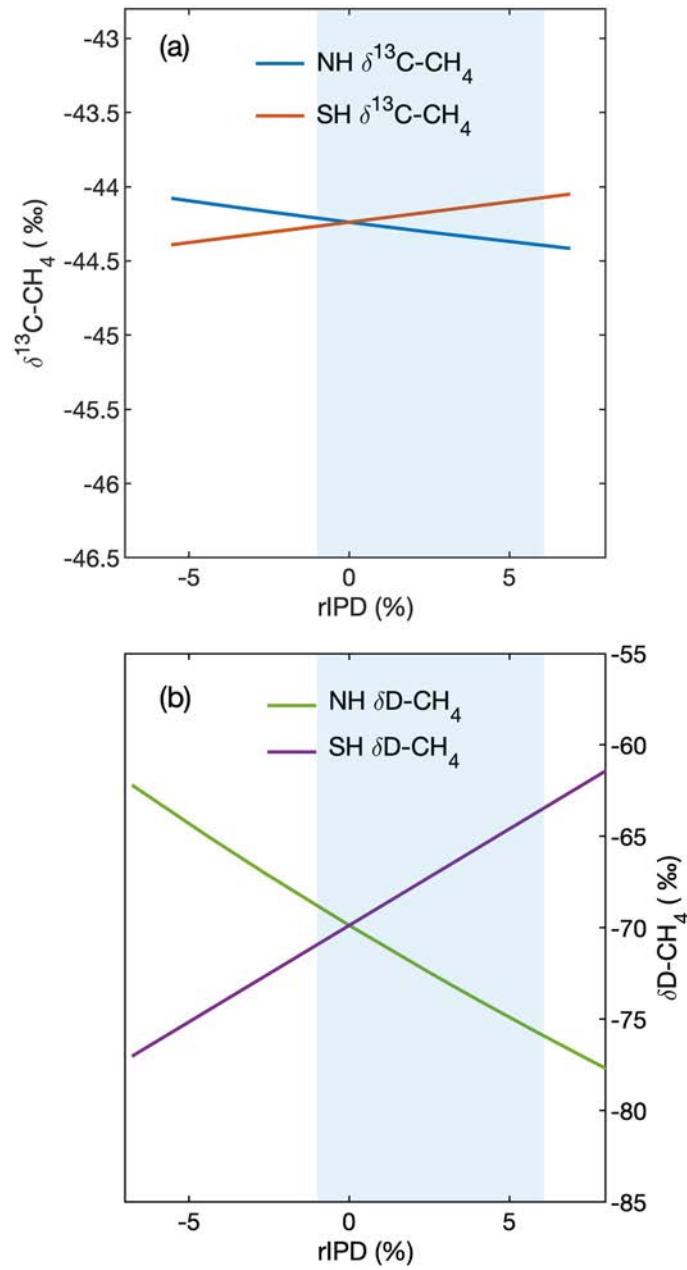


Extended Data Fig. 2 | Sensitivity tests for HES/DO12 and HE1 datasets. Modelled microbial $\delta^{13}\text{C}-\text{CH}_4$ assuming constant pyrogenic and geologic $\delta^{13}\text{C}-\text{CH}_4$ and emissions for (a) HE1 and (c) HES/DO12. Calculated emission strength of microbial CH_4 emissions derived from C3-dominant (green) or

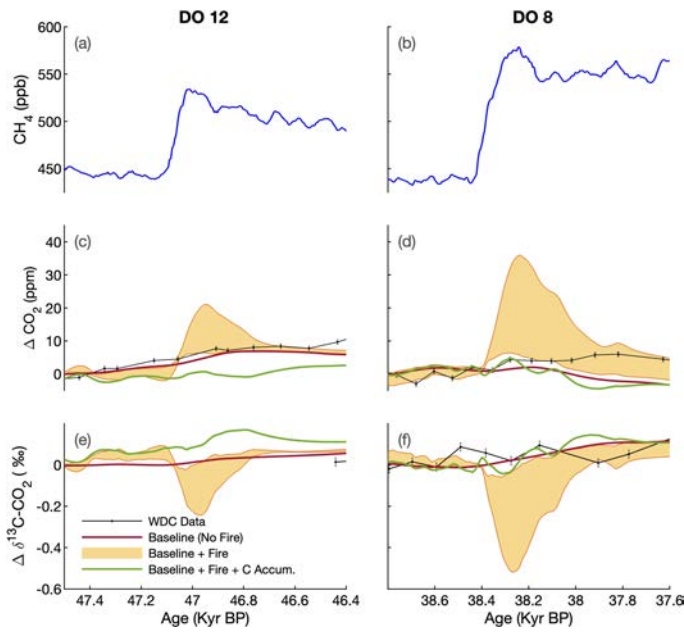
C4-dominant (orange) precursor biomass from the tropical regions using the results of the lefthand panels for (b) HE1 and (d) HES/DO12. The WDC CFA record is shown in blue in each subplot¹.



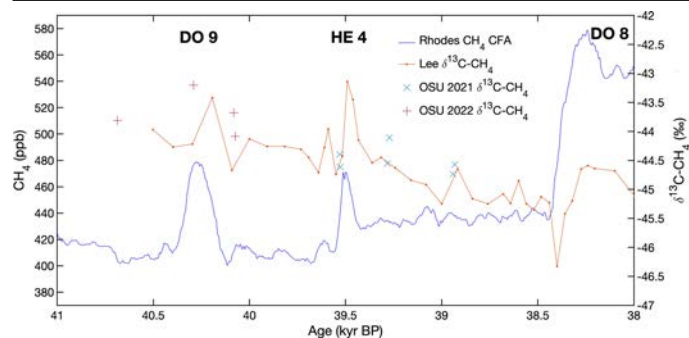
Extended Data Fig. 3 | Atmospheric disequilibrium effect for $\delta\text{D}-\text{CH}_4$ and $\delta^{13}\text{C}-\text{CH}_4$. (a) WDC CH_4 CFA data (blue)^{1,53}. (b) Measured $\delta^{13}\text{C}-\text{CH}_4$ data (red diamonds) plotted with 1-sigma measurement uncertainties and spline-interpolated to the resolution of the CFA CH_4 data (red). Modelled atmospheric $\delta^{13}\text{C}-\text{CH}_4$ assuming constant $\delta^{13}\text{C}-\text{CH}_4$ of the global source (black) to visualize the impact of atmospheric disequilibrium on $\delta^{13}\text{C}-\text{CH}_4$. (c) Measured $\delta\text{D}-\text{CH}_4$ data plotted with 1-sigma uncertainties from WDC (green diamonds) and TALDICE (magenta diamonds) plotted on the WDC-synchronized gas age scale from 57,58 . Modelled atmospheric $\delta\text{D}-\text{CH}_4$ assuming constant $\delta\text{D}-\text{CH}_4$ of the global source (black) to visualize the impact of atmospheric disequilibrium on $\delta\text{D}-\text{CH}_4$. The onset of HE4 is indicated by a vertical dashed line.



Extended Data Fig. 4 | Two-box model sensitivity tests. Two-box model equilibrium for (a) $\delta^{13}\text{C-CH}_4$ and (b) $\delta\text{D-CH}_4$ for scenarios with varying latitudinal source distribution (indicated by the x-axis: rIPD), but the same isotopic source signature. Varying latitudinal source distribution is displayed on the x-axis as the rIPD, which is the percent difference in NH and SH atmospheric CH₄ mixing ratio^{11,25,70,95}. The range of observed rIPD values indicated by ice core studies are shaded in blue. (a) $\delta^{13}\text{C-CH}_4$ for the NH (green) and SH (purple) are shown as a function of rIPD. (b) $\delta\text{D-CH}_4$ for the NH (green) and SH (purple) are also shown as a function of rIPD. The equilibrium isotopic composition was calculated using a two-box model with prescribed hemispheric source signatures (methods)⁶.



Extended Data Fig. 5 | Modelled CO₂ change during DO Events. Same as main text Fig. 4 but showing carbon cycle box-model results for DO12 (left) and DO8 (right) and including model results with changes to NPP associated with abrupt climate events (green). Changes to terrestrial carbon uptake due to changes in primary productivity were calculated using our one-box modelled microbial CH₄ emissions as a proxy for terrestrial carbon storage with a theoretical conversion factor of 1 Tg of microbial CH₄ emissions per 8 Pg of carbon stocks. Our modelled increases in terrestrial carbon stock at the onset DO Interstadials cancel carbon losses implied by the modelled pyrogenic CO₂ emissions and are consistent with earth system modelling studies^{15,32,52,91}. These studies similarly attribute the increase in terrestrial carbon to enhanced productivity associated with greater moisture availability over the land-dominated NH tropics.



Extended Data Fig. 6 | Comparison of OSU and Bern $\delta^{13}\text{C}-\text{CH}_4$ data across HE4, DO8 and DO9 from the WDC ice core. WDC CFA CH₄ data (left axis, blue line)^{1,53}, $\delta^{13}\text{C}-\text{CH}_4$ measured by James Lee at the Bern (orange), OSU $\delta^{13}\text{C}-\text{CH}_4$ measurements made in 2021 prior to the H1 WDC measurement campaign (light blue Xs), and OSU $\delta^{13}\text{C}-\text{CH}_4$ measurements made in 2022 prior to the HE5/DO12 measurement campaign (maroon +s). Data are not corrected for gravitational or diffusive firn fractionation, which allows for direct comparison between the $\delta^{13}\text{C}-\text{CH}_4$ that is trapped in the ice sample.

Extended Data Table 1 | Stable isotope signatures used in the 1-box model output isotope mass balance^{21,96}

CH ₄ Source	δ ¹³ C-CH ₄ (‰)	δD-CH ₄ (‰)
Global Microbial (Mic)	-60 +/- 1.5	-318 +/- 13
Pyrogenic (Pyro)	-22 +/- 1.8	-211 +/- 15
Geologic (Geo)	-44 +/- 0.4	-197 +/- 10
C3-dominant Microbial (C3)	-60 +/- 1.5	-
C4-dominant Microbial (C4)	-50 +/- 1.5	-
High-Lat NH Microbial (HI)	-68 +/- 3.1	-

One-sigma uncertainties for δ¹³C and δD source signatures from²¹ were incorporated into our box model output uncertainty. The δ¹³C-CH₄ emitted from C3- and C4-dominant biomass, the high-latitude NH microbial source δ¹³C-CH₄, and one-sigma uncertainties were obtained from⁹⁷. Parentheses denote the subscripts used in Eqs. 1–10 (methods).

Extended Data Table 2 | Summary of the magnitude of the pyrogenic pulses associated with HE5, 4 and 1

Heinrich Event	Box-model max annual increase (Tg yr ⁻¹)	Est. max annual increase (Tg yr ⁻¹)	Pyrogenic Pulse Duration (yr)	Total pyrogenic pulse emissions (Tg)	1-sigma Unc. (Tg)	Avg. emission rate (Tg yr ⁻¹)
HE5	6	9	250	501	196	2.0
HE4	9	14	110	438	104	4.0
HE1	10	15	180	739	154	4.1

From left to right: (1) The HE; (2) The box-modelled maximum annual increase in pyrogenic emissions from baseline values during each HE (Tg yr⁻¹); (3) The estimated true maximum annual increase based on the assumption of a 33% damping of the signal amplitude due to firn smoothing in WDC⁹ (Tg yr⁻¹); (4) The duration of the CH₄ pulses associated with each HE (yr); (5) The integrated total anomalous pyrogenic CH₄ emissions across each HE (Tg); (6) The 1-sigma uncertainty of (5) (Tg); (7) The average annual increase in pyrogenic emissions (Tg yr⁻¹), calculated by dividing (5) by (4).