

Atmospheric methane: Comparison between methane's record in 2006-2022 and during glacial terminations.

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The air is getting hotter. There's a rumbling in the skies.

I've been wading through the high muddy water, With the heat rising in my eyes.

Bob Dylan: Trying to get to Heaven before they close the door.

Key points

The rapid growth in the atmospheric methane burden that began in late 2006 is very different from methane's past observational record. Recent studies point to strongly increased emissions from wetlands, especially in the tropics. This increase is comparable in scale and speed to glacial/interglacial terminations when the global climate system suddenly reorganised.

Abstract

Atmospheric methane's rapid growth from late 2006 is unprecedented in the observational record. Analyses of atmospheric methane data attribute a large fraction of this atmospheric growth to increased natural emissions over the tropics, which appear to be responding to changes in anthropogenic climate forcing. Isotopically lighter measurements of $\delta^{13}\text{C}_{\text{CH}_4}$ are consistent with the recent atmospheric methane growth being mainly driven by an increase in emissions from microbial sources, particularly wetlands. The global methane budget is currently in disequilibrium and new inputs are as yet poorly quantified. Although microbial emissions from agriculture and waste sources have increased between 2006-2022 by perhaps 35-40 Tg/yr, approximately another 35-45 Tg/yr of the recent net growth in methane emissions may have been driven by natural biogenic processes, especially wetland feedbacks to climate change. A model comparison shows that recent changes may be comparable or greater in scale and speed than methane's growth and isotopic shift during past glacial/interglacial termination events. It remains possible that methane's current growth is within the range of Holocene variability, but it is also possible that methane's recent growth and isotopic shift may indicate a large-scale reorganisation of the natural climate and biosphere is under way.

Plain language summary

Atmospheric methane's unprecedented current growth, which in part may be driven by surging wetland emissions, has strong similarities to ice core methane records during glacial-interglacial 'termination' events during global reorganisations of the planetary climate system. Here we compare current and termination-event methane records to test the hypothesis that a termination-scale change may currently be in progress.

Keywords

Atmospheric methane growth driven by biogenic sources. Sudden global climate change. Glacial/interglacial comparison.

Main Text

1. Introduction

In the past, rapid changes in atmospheric methane have signalled rapid global climate shifts. Does atmospheric methane's recent growth, sustained from the end of 2006 through 2022 and continuing, similarly signal a major global climate shift?

In the modern air, the global methane burden – the total amount of methane in the air – includes both emissions from human activities such as methane from fossil fuel use, agriculture and waste, as well as natural biogenic inputs. Both agricultural and natural emissions are subject to feedbacks from the sensitivity of the biosphere to changes in climate. Summing global biospheric change from measurement data across the planet is a complex task with large uncertainties, but provided direct anthropogenic forcing can be deducted (a difficult and complex task), the remaining non-anthropogenic growth in biogenic methane emissions provides a direct measure of change.

Unlike CO_2 , whose annual cycle is strongly influenced by the spring growth and autumn leaf fall of deciduous plants in the temperate northern hemisphere, the natural methane cycle

reflects change across a much wider latitudinal spectrum, tropical, temperate and Arctic. Natural biogenic sources include tropical and boreal wetlands, ruminants, termites, Arctic bogs and permafrost, as well as forest and grassland fires, while anthropogenic biogenic methane comes from agriculture and waste, especially food waste disposal. Aquatic ecosystems, both natural and anthropogenic, contribute up to half the total emission (Rosentreter et al. 2021).

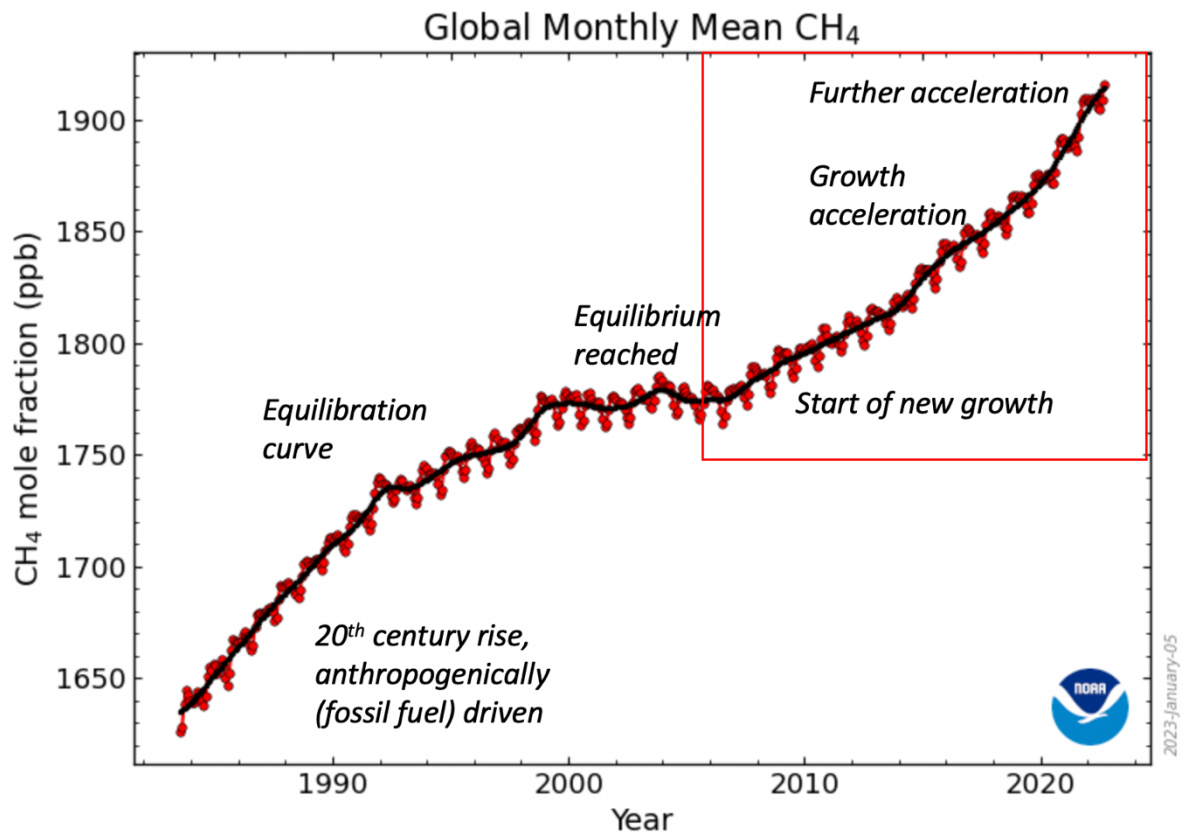
Starting in 2006 the atmospheric methane burden has grown strongly (Dlugokencky et al. 2011, Nisbet et al., 2014, Schaefer et al., 2016, Mikaloff-Fletcher and Schaefer 2019), accompanied by a sustained negative shift in the carbon isotope delta value ($\delta^{13}\text{C}_{\text{CH}_4}$). This shift indicates present-day atmospheric growth is primarily driven by biogenic emissions (Nisbet et al. 2016, 2019; Lan et al. 2021a,b, 2022; Basu et al 2022) and not primarily from increased fossil fuel emissions, nor by greatly stronger sink processes. Note that we distinguish between the atmospheric '*burden*' – the total mass of methane in the air expressed in Tg; '*emissions*' or fluxes, in Tg/yr; and '*emissions growth*' in Tg/yr per year. 1 ppb of methane, as an average mass fraction of the global atmosphere, has a mass of about 2.77Tg.

Methane's dominant sink is the atmospheric hydroxyl radical [OH], especially in the tropical mid-troposphere, with minor sinks including reaction with atomic chlorine in the marine boundary layer and bacterial methanotrophy in upland and forest soils. The formation and distribution of [OH] is changing as the climatological tropics widen with expansion of the Hadley cells (Nicely et al. 2018). While it remains unclear how much of the observed tropical expansion is within the range of natural variability (Grise et al. 2019) the present rate of increase is exceptional in the long-term record.

The following sections review recent work on the complex and often contradictory puzzle of methane's post-2006 growth. That synthesis is then compared with past events in the glacial paleoclimatological record, where is strong evidence (Möller et al. 2013) that fast methane rises during rapid glacial to interglacial shifts in the global climate are driven by feedbacks involving tropical wetlands that respond rapidly to warming.

Figure 1. Global monthly mean of atmospheric CH₄, NOAA network. Note convex equilibration curve to 2006; concave acceleration curve from 2007. The convex curve from the 1980s to early 2000s is consistent with relaxation to a source-sink steady state (Dlugokencky et al., 2003), but then the start of a renewed growth became apparent in 2007, and the slope of the new concave curve steepened in 2013 (acceleration of growth), followed by further acceleration in 2020. Modified from Lan et al. (2022).

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122

123 1.1 Feedbacks

124 Anthropogenic methane emissions have dramatically added to the primary forcing of the
 125 climate (Saunio et al., 2020), but the extent of the feedback response of methane
 126 emissions as climate has become warmer and wetter remains uncertain (Dean et al, 2018).
 127 For methane in particular, it is difficult to separate primary emissions (e.g. gas industry and
 128 coal mine leaks), natural feedbacks (e.g. increased wetland emissions) and changing
 129 anthropogenic biogenic emissions from farming, sewage, and landfills. As fertiliser
 130 production, especially fixed nitrogen, has grown, the productivity of neighbouring wetlands
 131 may have risen, driven by nitrogen runoff. Rising CO₂ also acts as a fertiliser, globally
 132 promoting C3 plant growth (trees and bushes) (Haverd et al. 2020, Graven et al. 2013,
 133 Matthews, 2007, Möller et al. 2013), though this impact is eventually limited by supply of
 134 other nutrients such as fixed nitrogen and phosphorous (Wang et al, 2020).

135 Considering the interlinkages between climate and methane productivity, Dean et al. (2018)
 136 concluded that wetlands will form the majority of the methane climate feedback up to the
 137 year 2100. Higher primary productivity, in response to warmth, precipitation and
 138 inundation, supplies and enhances microbial activity in anaerobic environments.
 139 Methanogenic processes may respond more quickly to change than methanotrophic
 140 processes (Wen et al, 2018). We may have entered a new era of increased natural methane
 141 emissions from wetlands (Rosentreter et al. 2021), driven by multiple feedback factors,
 142 including increased rainfall and temperatures, higher rainfall in key areas (Lunt et al 2021),
 143 and higher ecosystem production, aided by agricultural impacts. If present-day feedback
 144 responses are indeed strong, then the biogenic component of methane's very strong current

growth may be an integrative signal responding to, indicative of, and feeding global-scale change.

Any modern global-scale climate change, by cause and starting point, must be fundamentally different from past changes in the palaeoclimate record. Nevertheless, there are strong analogies between methane's current growth and ice core methane records during glacial-interglacial 'terminations'. In these events, the methane record tracked planetary-scale meteorological reorganisations of the climate system (Broecker & Denton, 1990; Cheng et al. 2009; Denton et al. 2010). Thus, the question arises (Nisbet 2022, 2023): Is methane's modern growth (Fig. 2) also a signal that a planetary-scale climate reorganisation is underway?

Figure 2. Annual mean growth (blue bars; ppb/yr) and acceleration in half-year intervals (red, black bars; ppb/yr²) of atmospheric methane inferred from NOAA data, 1984-2022 (NOAA 2023a). Red bars denote the gradient of the growth rate calculated using second-order accurate central differences of the interior points and first-order accurate one-side differences of the end points. Black bars denote the difference in atmospheric growth between successive years.

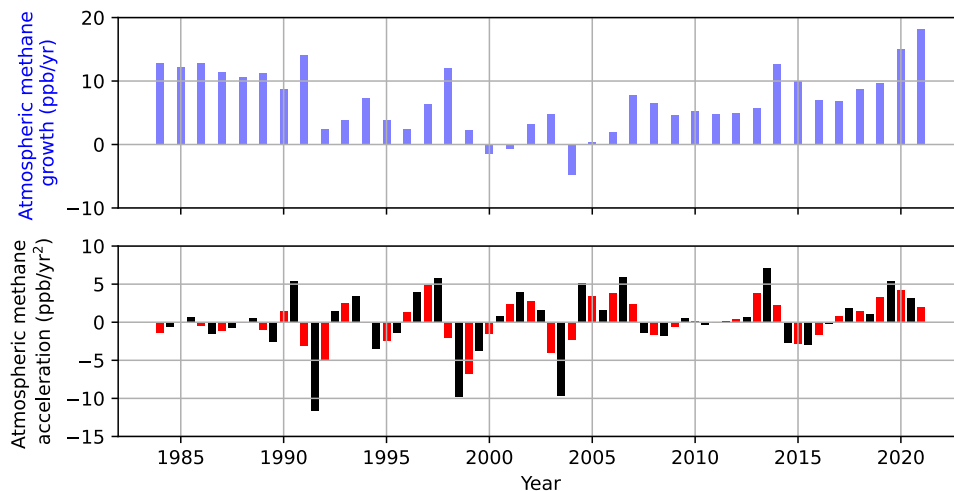
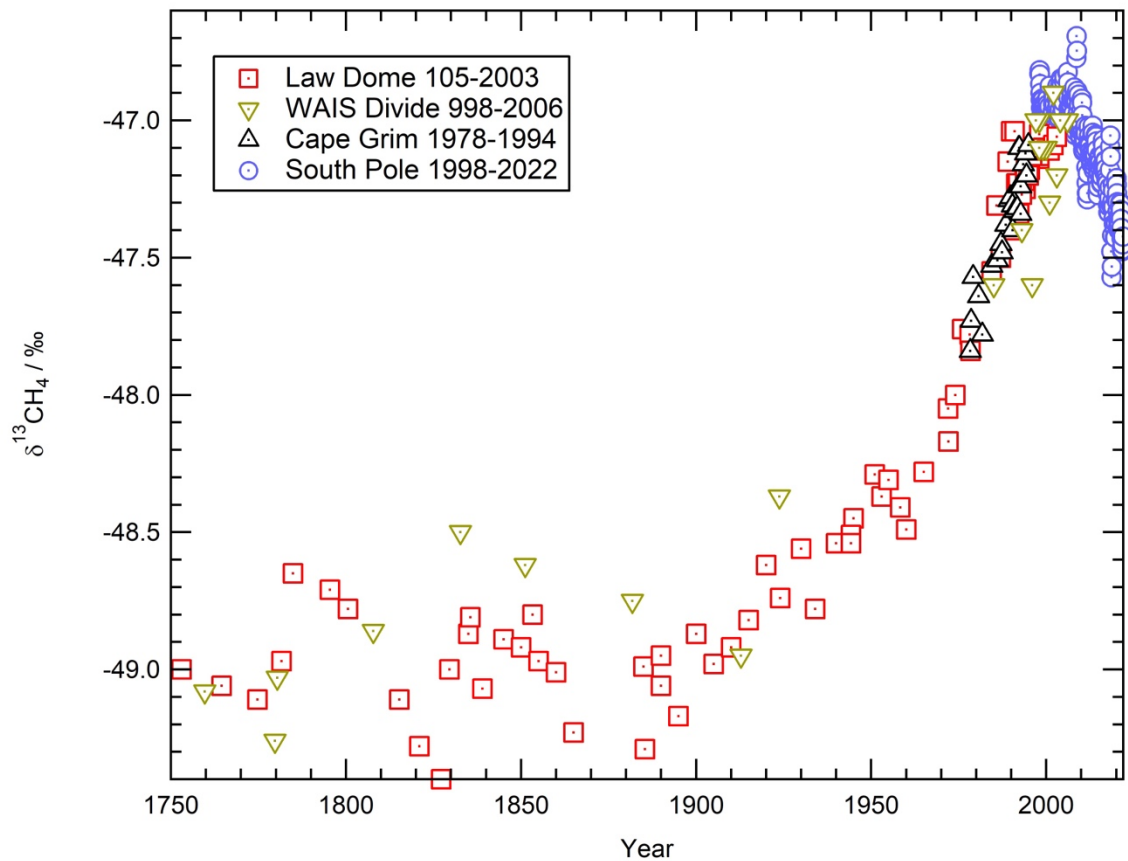
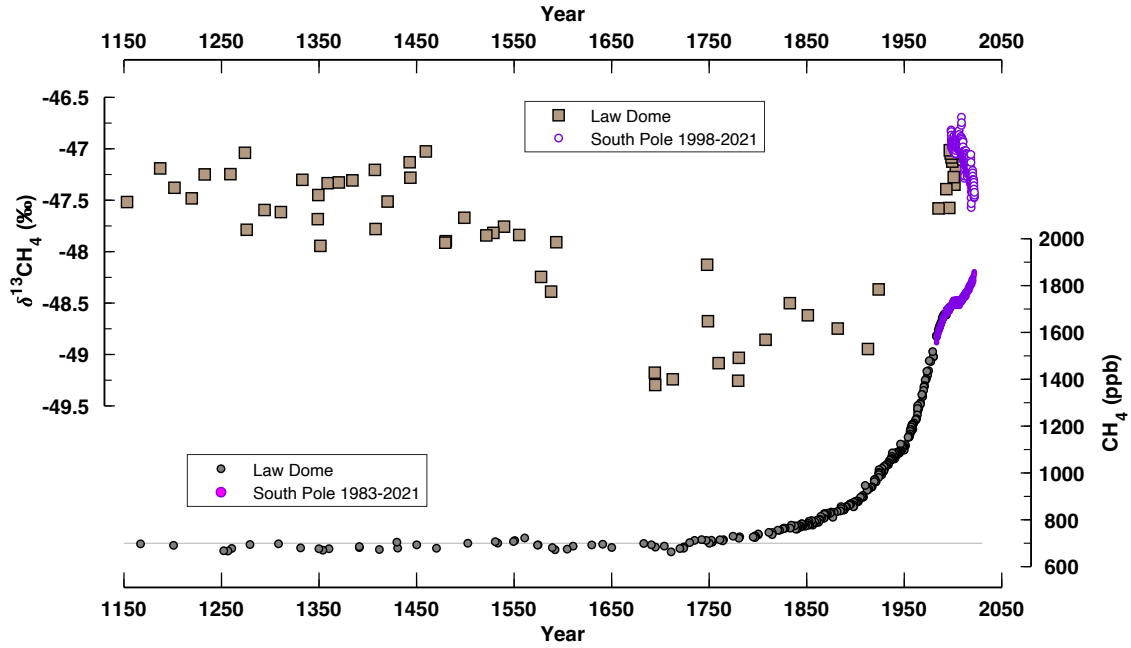


Figure 3. Past records of $\delta^{13}\text{C}_{\text{CH}_4}$.

3a. Methane and $\delta^{13}\text{C}_{\text{CH}_4}$ record back to the year 1150 CE, showing the background evolution of both CH_4 and $\delta^{13}\text{C}_{\text{CH}_4}$ before the budget was disturbed by human interferences (i.e. before ca. 1500 -1600 AD), to indicate historic variability due to changes in climate and wetland emissions on multidecadal or longer time scales. Note the CH_4 background is essentially flat over pre-industrial time, but there is a notable negative trend of about 2‰ between 1500-1650 that is gradually reversed as coal-fired industrialisation begins. For longer record see Sapart et al. (2012).

3b. Post-1750 $\delta^{13}\text{C}_{\text{CH}_4}$ record from South Pole and ice core measurements, showing recent record in more detail. Note long +ve trend in $\delta^{13}\text{C}_{\text{CH}_4}$, driven by fossil fuel emissions, and sustained reversal of trend since 2007.

Data from Law Dome: Ferretti et al. 2005; see also Rubino et al. 2019; WAIS Divide: Mischeler et al 2009; Sapart et al. 2012 and from NOAA https://gml.noaa.gov/aftp/data/trace_gases/ch4c13/flask/surface/.



2. Methane growth, pre-industrial period to present

There has been strong centennial-scale variation in methane's recent historic record (Fig. 3). From Antarctic ice core measurements, Ferretti et al. (2005), Sapart et al. (2012) and Mischler et al (2009) found that $\delta^{13}\text{C}_{\text{CH}_4}$ was about -47‰ around 800 CE, but then showed a

strong depletion trend from about 900 CE to 1700 CE, until $\delta^{13}\text{C}_{\text{CH}_4}$ was about -49‰. Prior to the onset of industrialisation, in the medieval years from the 12th to the 19th centuries CE, methane rose by around 100 ppb, accompanied by a negative trend in $\delta^{13}\text{C}_{\text{CH}_4}$ from -47.5‰ to -49‰ (Fig. 3). Ferretti et al. (2005) interpreted this medieval 2‰ depletion as evidence for a major drop in anthropogenic biomass burning emissions between 800-1700 CE, for example a reduction in deliberate and intense North American grassland burning. CH_4 was rising during this period, implying the reduction in biomass burning emissions must have been more than balanced by other sources. This interpretation is supported by CO evidence from ice cores. Agricultural methane emissions may have helped strengthen the negative trend (Mischler et al. 2009). But that historic depletion rate was about 0.0025‰ per year, totalling less than 0.04‰ over 15 years, very different from what has taken place in the 15 years 2007-2022.

Starting around the years 1750 to 1800 (Fig. 3b), with the onset of coal-fired and then oil and gas-powered industrialisation, the $\delta^{13}\text{C}_{\text{CH}_4}$ trend rapidly shifted positive (Etheridge et al. 1998, Ferretti et al. 2005, Mischler et al. 2009, Sapart et al. 2012). That exceptionally rapid positive shift accompanying fossil-fuel industrialisation from the years 1800-2007 was +2‰ in 250 years, or about 0.01‰ per year, interpreted as evidence that the rise was primarily driven by fossil fuel emissions, first mainly from coal, then with methane emissions as a by-product of oil extraction and use, and more recently from the gas industry too.

In contrast, the ongoing 2007-2022 reverse negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$ has been -0.55‰ in 15 years: a rate of negative shift four times faster than the positive rise during industrialisation, and more than tenfold the rate of the medieval depletion trend. However, it should be noted that many biogenic sources, especially in extra-tropical wetlands, have greater leverage on atmospheric $\delta^{13}\text{C}_{\text{CH}_4}$ than fossil fuel emissions which are closer in source signature to the bulk atmospheric value.

Sustained accurate methane measurement began in the late 1970s and by 1983 NOAA led a co-operative global network collecting flask samples of air from remote sites. Between 1984 and 1990 the growth rate was over 10 ppb/yr, accompanied by a continuing positive shift in $\delta^{13}\text{C}_{\text{CH}_4}$ (Quay et al. 1988, Etheridge et al. 1998, Ferretti et al. 2005), but in the early 1990s Steele et al. (1992) first recognised a slowing-down of growth and predicted stabilisation around 2006. This was confirmed by Dlugokencky et al. (1994, 1998, 2003) and Lowe et al. (1994) as the budget equilibrated and the growth rate in fossil fuel emissions slowed. After some years of equilibrium in the early 2000s, the return of growth in the atmospheric methane burden became apparent from late 2006 and into 2007 (Figure 3).

Methane's recent growth is wholly different from the late 20th century pattern. Unlike the convex, equilibrating trend of the late 20th century, growth since the end of 2006 has been on a concave, accelerating trend with the highest growth rates on record in 2020 and 2021 (Dlugokencky et al. 2011, Nisbet et al., 2014, 2016, 2019, 2021, Lan et al. 2021a,b, 2022, 2023; Fig. 1). Moreover, methane's post-2006 isotopic record is isotopically quite unlike the record of the 'fossil-fuel' industrial 19th and 20th centuries. The long trend towards more positive $\delta^{13}\text{C}_{\text{CH}_4}$, indicative of fossil fuel emissions, has unexpectedly reversed, replaced by sustained strong trend to more negative $\delta^{13}\text{C}_{\text{CH}_4}$. The trend is not a subtle statistical drift but rather it is a strong planet-wide shift far outside any plausible statistical variability.

In 2006, the global mean of CH_4 in the marine background air was 1775 ppb, rising to 1912 ppb in 2022, a total rise of 137 ppb (about 379 Tg) in the methane burden. In 2021 alone,

growth was around 18 ppb or about 50 Tg. Rapid growth continues with an increase in 2022 of 14 ppb.

Synchronous with the onset of sustained sharp growth in CH₄ has been the isotopic reversal, initiating a sustained strong negative trend in $\delta^{13}\text{C}_{\text{CH}_4}$ (Fig. 3; also Nisbet et al. 2019, Lan et al. 2021a,b). $\delta^{13}\text{C}_{\text{CH}_4}$ values in Arctic air declined from -47.3‰ in 2006 towards -48‰ in mid-2022. At the South Pole (Fig. 4), $\delta^{13}\text{C}_{\text{CH}_4}$ values were about -46.9‰ in 2006, sinking towards -47.4‰ in 2022 (Michel et al. 2022). These trends were constructed from detailed individual flask records (e.g. see Figure SI 1 showing the flask-by-flask record from Ascension).

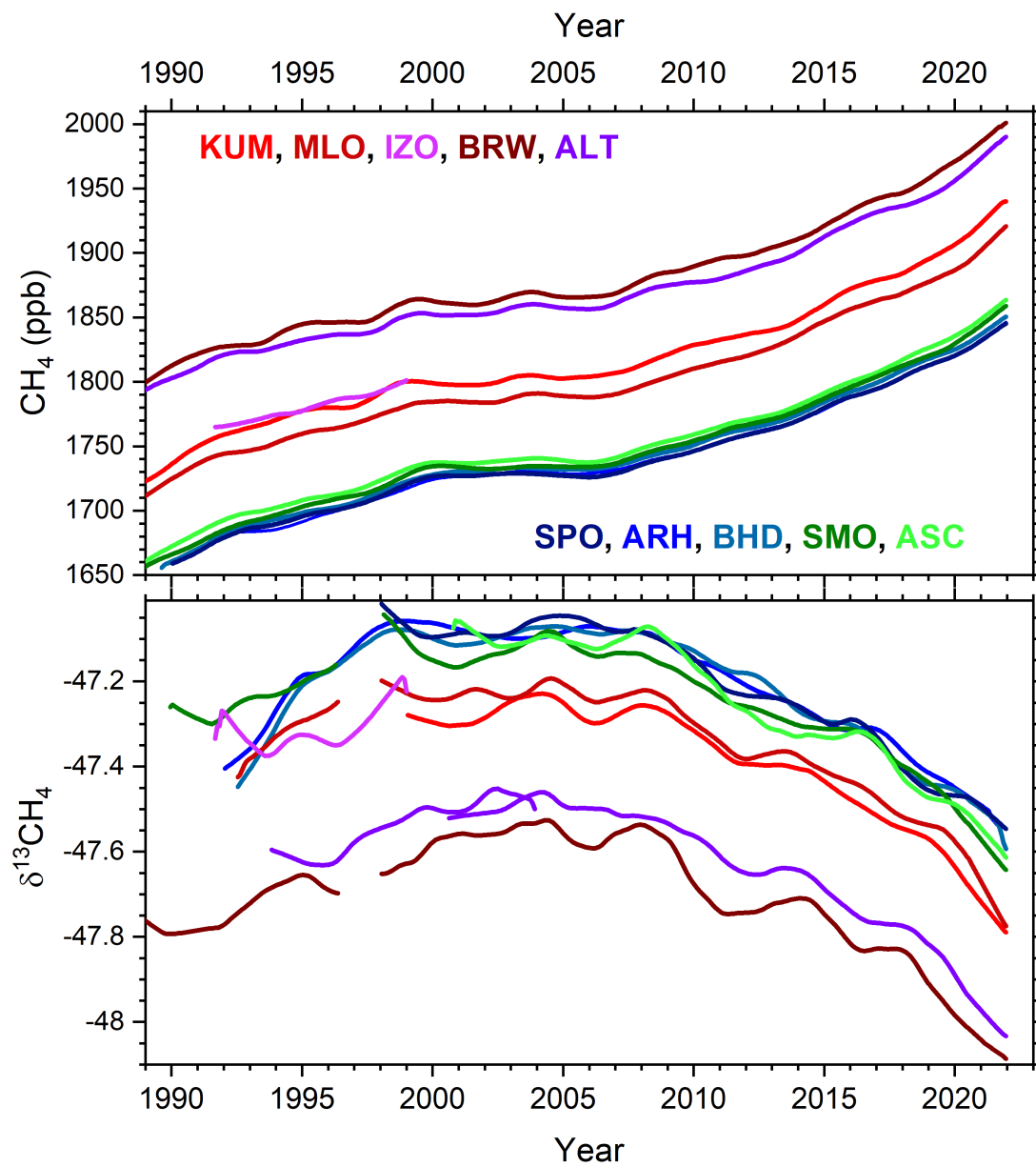
As seen in Figure 4, the last 30 years have had two phases for an increase in CH₄ mole fraction but with these having strong opposite and global trends in $\delta^{13}\text{C}_{\text{CH}_4}$. Longer term records from ice-core and firn air show that this 'turnover' in $\delta^{13}\text{C}_{\text{CH}_4}$, has reversed a centuries-long trend towards 'heavier' (more ¹³C-rich) methane that ended around 1998, then a few years of stability, and onset of the current trend towards 'lighter' (less ¹³C-rich) values that began in late 2006 and has been sustained ever since. Globally, the negative $\delta^{13}\text{C}_{\text{CH}_4}$ shift over 2006-2022 has been about -0.55‰, from the maximum (most positive) record in March 2008 to the more recent 2022 (Lan et al 2021a,b and update 2023).

Figure 4. Trends in methane mixing ratio (top panels) and $\delta^{13}\text{C}_{\text{CH}_4}$ (lower panels)

Figure 4a) Trends in methane mixing ratio (top panel) and $\delta^{13}\text{C}_{\text{CH}_4}$ (lower panel) for five long term monitoring sites in each hemisphere. In the Southern Hemisphere the measurement stations are: NOAA's SPO – South Pole (90°S), SMO – Samoa (14°S), and ASC – Ascension (8°S); and NIWA's ARH – Arrival Heights, McMurdo, Antarctica (78°S) and BHD – Baring Head, NZ (41°S). In the Northern Hemisphere they are: NOAA's KUM – Cape Kumukahi (20°N), MLO – Mauna Loa (20°N), BRW – Barrow, Alaska (71°N), ALT – Alert, Canada (83°N), and University of Heidelberg's IZO – Izana (28°N). Early $\delta^{13}\text{C}_{\text{CH}_4}$ data for SMO, MLO and BRW are from Quay et al (1991) and for IZO from Levin et al., (2012).

Figure 4b) Detailed patterns showing seasonality for four selected sites, BRW and MLO in the Northern Hemisphere, and ASC and SPO in the Southern Hemisphere. NOAA data.

NOAA data from https://gml.noaa.gov/aftp/data/trace_gases/ch4c13/flask/surface/. NIWA data in the World Data Center for Greenhouse Gases <https://gaw.kishou.go.jp/>. For further details see Supplementary Information. Intercomparison follows Umezawa et al. (2018). SPO and MLO are at 2810 and 3397 m elevation respectively but the other points are close to sea level. Details of the data from these sites are shown in supplementary material while this figure summarises the similar patterns seen globally.



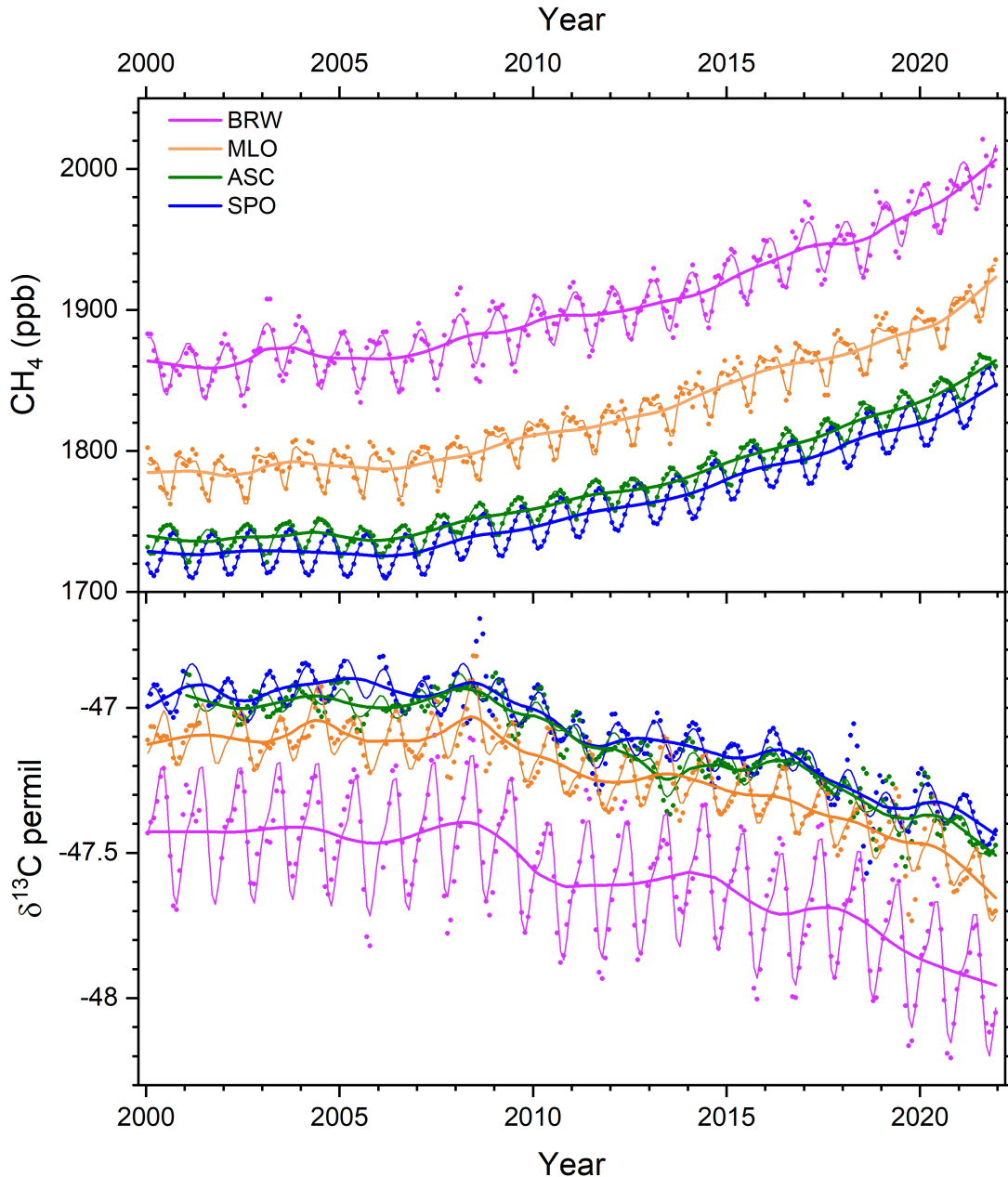
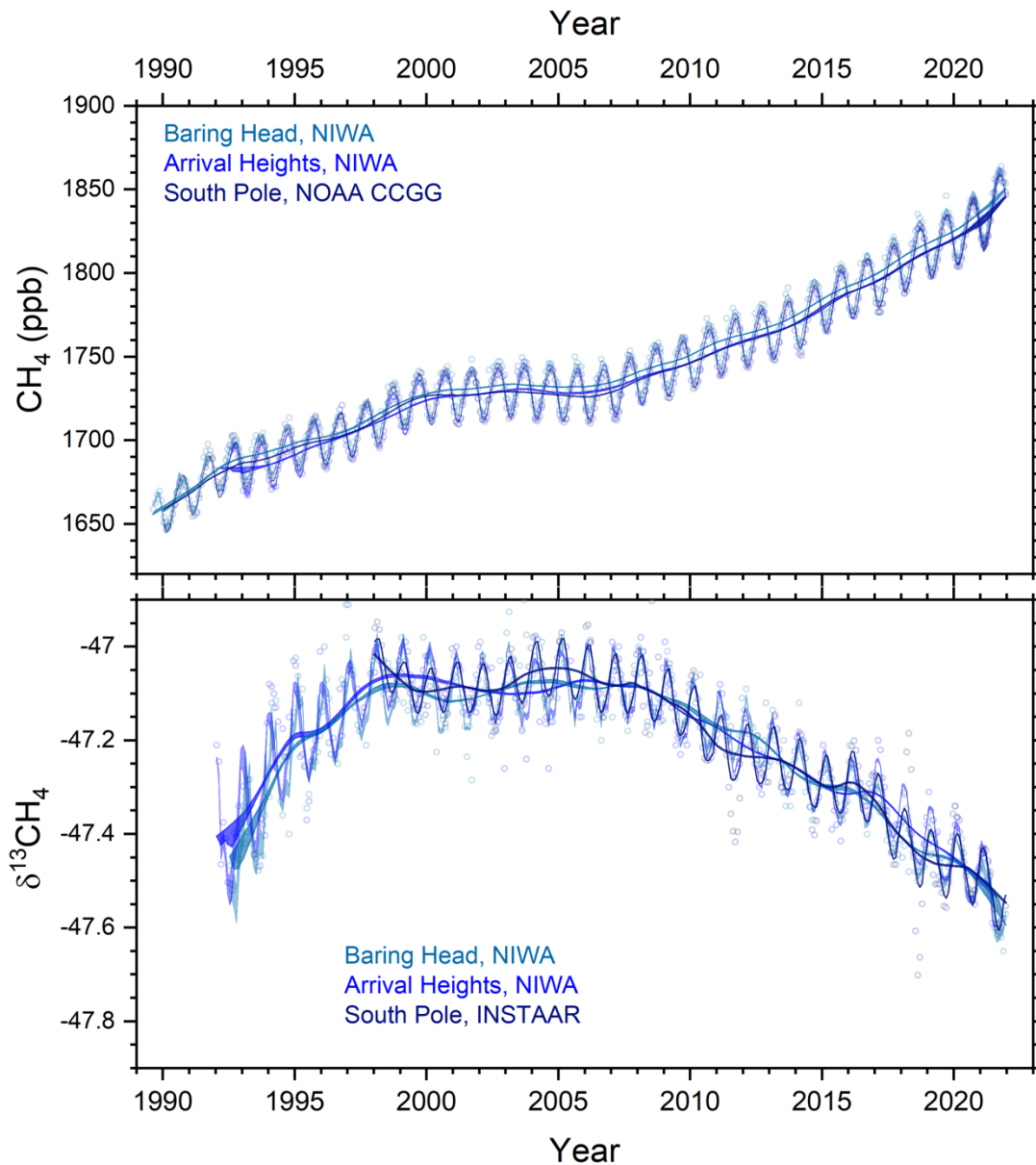


Figure 5 shows in detail the 'turnover' and seasonality in the remote (i.e. global) $\delta^{13}\text{C}_{\text{CH}_4}$ trend. The centuries-long trend towards 'heavier' (more ^{13}C -rich) methane ended around 1998 (e.g. see Dlugokencky et al. 1994). Then came a few years of stability. The onset of the trend towards 'lighter' (less ^{13}C -rich) values began in late 2006 and has been sustained ever since. Globally, the negative $\delta^{13}\text{C}_{\text{CH}_4}$ shift over 2006-2022 has been about -0.55‰, from the maximum (most positive) record in March 2008 to 2022 (Lan et al 2021a,b and update 2023).

Fig. 5 Remote Southern Hemisphere Isotopic Trend since the 1980s. Data from NIWA New Zealand (Baring Head (41°S), Arrival Heights (78°S) and NOAA/INSTAAR measurements (SPO 90°S). See Fig. 4 for data sources; intercomparison after Umezawa et al. (2018). https://gml.noaa.gov/aftp/data/trace_gases/ch4c13/flask/surface/).



3. Methane sources and sinks; isotopic impacts.

The bulk source of global methane emissions has $\delta^{13}\text{C}_{\text{CH}_4} \cong -53\text{‰}$ for all sources (Dlugokencky et al. 2011, Nisbet et al. 2019, Sherwood et al. 2017). Oh et al. (2022) estimated a mean global wetland $\delta^{13}\text{C}_{\text{CH}_4}$ signature of $-61.3 \pm 0.7\text{‰}$. This reflects a mix between northern high-latitude C3 plant wetlands (mean -67.8‰) and tropical C4-rich wetlands (mean -56.7‰) (Quay et al, 1988, Dlugokencky et al. 2011; Sherwood et al. 2017, 2021, Ganesan et al. 2018, MOYA/ZWAMPS Team (2021), Menoud et al., 2022). Note the means and precisions cited here are from partial and perhaps unrepresentative sampling: real world averages may be several ‰ different. In addition to the global C3/C4 plant distribution, the distribution of hydrogenotrophic / acetoclastic methanogens and vegetation-specific CH_4 transport all contribute to the $\delta^{13}\text{C}_{\text{CH}_4}$ variability of wetland emissions (Ganesan et al. 2018). France et al. (2021) found methane from South American

and African southern outer tropical (seasonal) wetlands had $\delta^{13}\text{C}_{\text{CH}_4}$ $-60 \pm 5\text{‰}$ while equatorial wetlands emitted methane with $\delta^{13}\text{C}_{\text{CH}_4}$ $-52 \pm 2\text{‰}$. Similarly, Oh et al. (2022) found tropical wetland emissions are enriched by about 11‰ relative to boreal wetlands.

Agricultural emissions, like wetland emissions, vary with latitude and C3/C4 proportions in ruminant diets (Menoud et al. 2022, and tropical measurements are scarce. For rice fields, France et al. (2021) found $\delta^{13}\text{C}_{\text{CH}_4}$ $-61 \pm 4\text{‰}$. Farmed ruminant emissions vary with diet, with a global average $\delta^{13}\text{C}_{\text{CH}_4}$ perhaps around -65‰ (e.g. Chang et al. 2019), but data are scarce from tropical ruminants where C4 fodder can dominate; for example, for Kenyan pastured cattle, $\delta^{13}\text{C}_{\text{CH}_4}$ is around -57‰ (MOYA/ZWAMPS Team 2021). Fossil fuel emissions have $\delta^{13}\text{C}_{\text{CH}_4}$ globally roughly between -43‰ and -45‰ (Lan et al. 2021a). However, $\delta^{13}\text{C}_{\text{CH}_4}$ values range widely from highly positive values (e.g. -25‰ for some gasfields) to very negative (as light as -70‰ in some cases). Howarth (2019) suggested that about a third of growth in methane emissions from all sources globally over the past decade came from shale gas production in North America. Nevertheless, Milkov et al. (2020), Basu et al. (2022) and Zhang et al. (2023) showed this to be unlikely.

The steady-state lifetime for atmospheric methane is between 9 and 10 years (Ehhalt 1974; Dlugokencky et al. 2011; Nisbet et al. 2019; Lan et al. 2021a). Methane's perturbation lifetime, which accounts for atmospheric chemistry relaxation times and is more relevant for climate impacts of emission reductions, is approximately 12 years (Prather et al. 2012, Myhre et al. 2013). Methane's main sinks, atmospheric hydroxyl [OH], Cl, and soil methanotrophy, are all subject to interannual variability as well as to the growth in the methane burden. Nicely et al. (2018) found the expected decline in [OH] caused by methane growth was largely offset by positive trends in tropospheric [OH] responding to changes in water vapour, NO_x , and O_3 . Methane's sinks all respond to climate change. Yin et al. (2021) showed that the longer-term impact of [OH] changes was modest, a conclusion updated by Basu et al (2022). Similarly, Thompson et al. (2018) and Drinkwater et al. (2023) also found the impact of changes in the atmospheric sink did not appear to be a key driver of methane's strong and sustained growth. However, in some years specific events can have large impacts on methane growth. In particular, [OH] fluctuations can make a major contribution to methane growth in hot El Niño years (Zhao et al. 2020), especially when there are enhanced CO emissions from biomass burning. Such effects smooth out during a full El Niño /Southern Oscillation (ENSO) cycle, as CO emissions from biomass burning decrease again in subsequent cool, regionally-wet La Niña years.

Sources and sinks show interannual variability and may have been affected by covid lockdowns in 2020 (Laughner et al. 2021), though that transient impact is unlikely to have been sustained significantly into 2021 and 2022. For the unusual year 2020, when anthropogenic emissions of air pollution were much affected by COVID lockdowns (e.g. Cooper et al. 2022), the growth in methane was driven both by a decline in [OH], and by increased wetland emissions (Peng et al. 2022), especially in the tropical and sub-tropical Northern Hemisphere and in western Siberia and central Canada. Using analysis of GOSAT retrievals, Qu et al (2022) concluded 14% of growth was from [OH] changes. Likewise, Feng et al (2023) also found an [OH] reduction comparable to Peng et al.'s results, but concluded that most of the observed increase in atmospheric methane during 2020 and 2021 was caused by increased emissions, especially in Eastern Africa. The full causes of methane's remarkable growth in 2021 and 2022, when covid lockdowns were less severe, are still being debated but it is

notable that over the period 2020-2022 the methane growth histogram matches the very unusual 'triple-dip' 3-year La Niña event (NOAA 2023b).

Methane's minor sinks are destruction by atmospheric chlorine (Hossaini et al. 2016), oxidation by soil methanotrophs, and upward transport into the stratosphere where methane is destroyed (and isotopically fractionated) by photooxidation. Methane destruction by chlorine is particularly interesting: although it only accounts for ~2.5% of the total methane sink (but with larger regional-scale effects (Hossaini et al. 2016)), it is highly fractionating for C and H isotopes (Strode et al. 2020). All these sinks will have interannual variability, especially in the ENSO cycle, and will also be subject to changing feedbacks to long-term climate warming.

All biological methane sources, both purely 'natural' and anthropogenic, respond to climate change and the rapid strong positive feedbacks make methane an excellent integrating indicator of large-scale wetland ecosystem change. Increased warmth and moisture in the 'wet tropics' are likely to increase plant growth and thus ruminant productivity, and also microbial activity in manure and wetlands. In the boreal and Arctic realms warmth is also likely to increase wetland emissions. Moreover, as mentioned above (section 1.1), CO₂ acts as a potent fertiliser for C3 plants, increasing plant productivity (Lobell and Field, 2008; Uddin et al. 2018; Haverd et al. 2020) and thus supply of organic matter to eventual anaerobic decay, though it is possible this impact is eventually limited by supply of other nutrients, primarily fixed nitrogen and phosphorus (Wang et al. 2020).

Table 1: Simplified comparison of some recent assessments of wetland emissions and their growth.

a) Examples of some estimated outer tropical wetland emissions

<i>Location</i>	<i>Latitude, area</i>	<i>Emissions Tg/yr</i>	<i>Reference</i>
Bangweulu, Upper Congo basin, Zambia	11°S, >10000 km ²	1 to 3	Shaw et al, 2021
Pantanal, Brazil	Around 17°S, around 150000 km ²	2 to 3.3	Gloor et al., 2021
Llanos de Moxos, Bolivia	Around 12-15°S, Around 70000 km ²	3.6	France et al, 2022
<i>Freshwater wetlands</i>	<i>Global</i>	138-165	Rosentreter et al 2021
<i>East Africa</i>	<i>Regional</i>	Mar-May 2018 6Tg Oct-Dec 2019 9Tg	Lunt et al. 2021
<i>South Sudan</i>	<i>Regional</i>	7.4 ± 3.2 Tg	Pandey et al. 2021

b) Global emissions growth (all sources) in the single years 2020 and 2021

Period	Emissions growth in year (Tg)	Reference
2020	27	Feng et al. 2023
2021	21	Feng et al. 2023
2020	25-37 (48% from Africa)	Qu et al.

c) Global wetland-only emissions growth in the single years 2020 and 2021

Period	Emissions growth in year (Tg)	Reference
2020	14-26	Zhang et al. 2023
2021	13-23	Zhang et al. 2023
2020	6 - 7	Peng et al. 2022
2019-2020	22-32 (of which 15 in Africa)	Qu et al. 2022

d) Global emissions in the years from the onset of growth prior to 2020

Reference	Period	Growth in biogenic emissions Tg/yr	Estimated emissions growth acceleration Tg/yr/yr
Basu et al. 2021	2007-2012	26	4
Basu et al. 2021	2013-2017	32	
Zhang et al. 2023	2007-2021	8 - 10	1.3 -1.4
Oh et al. 2022	2006-2016	28 assumed scenario	

Table 1 Note: numbers given here are for comparison purposes only. See cited sources for original details. Numbers here highly simplified, rounded off from the disparate results in the many cited studies, and are very approximate with large uncertainties.

4. The wetland hypothesis

The wetland hypothesis is that biogenic emissions, particularly from wetlands, are growing rapidly and are the dominant cause of recent growth (Lunt et al. 2019, Nisbet et al. 2016, 2019, Lan et al. 2021a,b, Basu et al. 2022, Oh et al. 2022, Zhang et al. 2023). Aquatic ecosystems are enormously important in the global methane budget (Rosentreter et al. 2021). Dean et al. (2018) found wetland emission growth can be primarily ascribed to climate change feedbacks. Table 1 provides a highly simplified comparison of results from recent studies in a very active and complex field.

The 'wetland hypothesis' has been supported by powerful evidence that increased wetland emissions, especially in the tropics, drove methane's exceptional growth in 2020 (Lunt et al. 2019, 2021, Qu et al. 2022, Feng et al. 2022, 2023; Peng et al. 2022). The isotopic evidence is strong. Using a process-based biogeochemistry model, Oh et al. (2022) concluded the spatial distribution of wetland $\delta^{13}\text{C}_{\text{CH}_4}$ emissions is consistent with the observed latitudinal gradient in $\delta^{13}\text{C}_{\text{CH}_4}$, implying the growth in microbial emissions has been the dominant driver for methane's post-2006 rise and concurrent negative trend in $\delta^{13}\text{C}_{\text{CH}_4}$. The causes of increased wetland emissions likely include increased rainfall and inundation in drainage basins (Pandey et al. 2017, Lunt et al. 2019, 2021, Feng et al. 2022), and also overall warm temperatures in the post-2006 period, as plant and microbial productivities rise with temperature, provided water and nutrients are available.

Methane's remarkable growth in 2020-2022 has been synchronous with an unusual La Niña event (Hasan et al. 2022) with a 'triple-dip' 3-year period (McPhaden et al. 2023) that has brought sustained seasons of exceptional rainfall to the important regions in the moist tropics. However, emissions from aquatic systems are highly variable (Rosentreter et al. 2021), and inherently difficult to parameterize (Ringeval et al., 2014).

Because cloud cover is typically deep and persistent during wet seasons over key emitting regions, it is difficult to study tropical wetland emissions in rainy season using satellite remote sensing directly overhead, but the long atmospheric lifetime of methane allows for these emissions to be quantified via observations collected downwind. Feng et al. (2022) used satellite observations to infer that over 80% of the observed growth in the global methane burden between 2010-2019 came from tropical terrestrial emissions. Separately, Feng et al. (2023) estimated that 66 % of methane's global increase in 2020 was due to increased emissions, particularly from the tropics. They found evidence for growing microbial sources over the tropics with a very large increase (14 ± 3 Tg) in recent (2020)

emissions from northern hemisphere Eastern Africa (e.g. the Nile Sudd – see Lunt et al. 2019), while emissions decreased in China and India.

These results are supported by the findings of Qu et al. (2022) from GOSAT retrievals, that 82% of methane growth from 2019 to 2020 was driven by increased emissions, with a large contribution from African wetlands. Similarly, Drinkwater et al. (preprint submitted 2023) found methane recent growth (2004-2020) has been driven by a progressive increase in emissions, with emissions from tropical and sub-tropical sources (30°N to 30°S, i.e. including the monsoonal climate belt) accelerating by 3.8 Tg/yr/yr. They concluded methane's growth was primarily driven by biogenic emissions, especially from regions with large wetlands, with increases in annual emissions over the period of about 17 Tg from Northern Hemisphere Africa, and 21 Tg from Tropical South America, though Chinese annual emissions also increased very strongly (27 Tg).

Anthropogenic impacts can also increase wetland emissions. Although widespread drainage of wetlands is also taking place, tropical wetlands in Africa can host large cattle populations (own observations). Indirect anthropogenic forcing factors include CO₂ fertilisation (Holden et al. 2013, Haverd et al 2020), run-off to wetlands from nitrogen fertiliser application (Ackerman et al 2018), sulphate deposition (Gauci et al. 2005) in regions like southern Africa and China with large coal-industry emissions, and perhaps also the complex impacts of human predation on wetland fauna. The fertilising impact of run-off from arable croplands into nearby wetlands by nitrogen fertilisers (Fowler et al. 2013) may be important. However, most boreal wetlands are far from farming drainage, while in many parts of the tropics, although fertiliser application in arable lands around wetlands is increasing rapidly (e.g. Uganda), the use of fertilisers is still very low compared to farming in northern temperate regions (Dangal et al, 2019; FAO 2017, IFAsat 2022).

In particular, CO₂ fertilisation of C3 plants may be important. There is evidence (Möller et al. 2013) for CO₂ fertilisation impact in past transitions: the strong correlation between $\delta^{13}\text{C}_{\text{CH}_4}$ and CO₂ in the ice core record of the past 160 ka is much better than the correlation between $\delta^{13}\text{C}_{\text{CH}_4}$ and CH₄. This suggests CO₂ fertilisation was an active feedback in past changes by increasing plant productivity and hence microbial methane emissions during the initiation of interglacial episodes, with isotopic impacts favouring C3 plants (more negative $\delta^{13}\text{C}_{\text{CH}_4}$) over C4 grasses. Thus the glacial/interglacial $\delta^{13}\text{C}_{\text{CH}_4}$ record may reflect *in situ* shifts in plant populations and productivity as well as shifts in the geographic loci of emission. However, on the modern planet this impact is difficult to assess. It is also likely that nitrogen cycle constraints and phosphorus availability may be progressively limiting the effect of CO₂ fertilisation (Wang, S. et al. 2020, Huntingford et al, 2022).

Wetland models have large uncertainties (Chang et al. 2023) and tropical wetland emissions may be much larger than hitherto inferred in land surface models of wetland methane sources (Shaw et al. 2022; Zhang et al. 2023; Nisbet 2023). Recent tropical wetland field campaigns that included both low-flying aircraft and on-ground sampling have shown that emissions from some wetlands are very substantial indeed. For example, overflying the Llanos de Moxos wetlands in Bolivian Amazonia in March 2019 (late wet season), France et al. (2022) found extremely large methane enhancements (500 ppb over background). These were inferred to have come from very high surface fluxes of methane. Annual emissions were estimated around 3.6 Tg/yr from the Llanos de Moxos wetlands. Further south, in the Pantanal wetlands of Brazil and Bolivia in the South American seasonal outer tropics, Gloor et al. (2021) used a planetary boundary layer budgeting technique to estimate the

Pantanal's total regional emissions of 2.0 to 2.8 Tg/yr, while their Bayesian inversion using an atmospheric transport model found even higher emissions of ~3.3 Tg/yr.

At about the same southern outer tropical (seasonal) latitude in Africa, from an aircraft campaign in February 2019 (mid-wet season) in the Upper Congo Bangweulu wetlands of Zambia, Shaw et al. (2022) derived very large emissions of 1.2 to 3.0 Tg/yr. Bangweulu is only part of the Upper Congo's extensive suite of wetlands. Emissions from Zambezi basin wetlands were also significant (Shaw et al. 2022). This work suggests total emissions from wetlands in the Congo and Zambezi basins may be very large. Interestingly the regional $\delta^{13}\text{C}_{\text{CH}_4}$ isotopic signatures of emissions of outer (single rainy season) tropical C4/C3 wetlands in Zambia and Bolivian Amazonia were very similar, around -59‰ to -60‰ (France et al. 2021, MOYA team et al. 2021), presumably reflecting the similarity of the seasonal outer tropical C4/C3 vegetation mix.

Unfortunately, on-ground or low-level aircraft/UAV *in situ* monitoring of major tropical wetlands is inadequate (Ringeval et al. 2014, Nisbet 2023), so it is not possible to use *in-situ* monitoring to confirm year-on-year changes. From satellite observations, Lunt et al. (2021) found strong XCH₄ enhancements in East Africa consistent with large positive seasonal precipitation anomalies. Zonal observations from remote stations of the NOAA network (Nisbet et al. 2016, 2019) broadly support the findings of increased wetland emissions. Interannual rainfall and temperature variability can be very large (Lunt et al. 2021, Peng et al., 2022, Palmer et al. 2023), depending on shifts in the position and intensity of the Inter-Tropical Convergence Zone (ITCZ). Thus expansion and intensification of the tropical Hadley cell circulation can have dramatic impacts on methane emissions.

To turn to bottom-up assessments, Zhang et al. (2023), applying climate data from meteorological stations and reanalysis in a land surface wetland model, found that climate change impacts increased annual wetland emissions by 8-10 Tg/yr in 2007-2021 compared to the 2000-2006 level. Specifically, they also found huge positive anomalies of 14-26 Tg in 2020 and 13-23 Tg in 2021, and that tropical methane emissions contributed a large part of methane's growth in those two years. For the single year between 2019 and 2020, the bottom-up estimate of Peng et al. (2022) found an increase in wetland emissions of 6.0 ± 2.3 Tg. Their top-down 3D atmospheric inversion gave a global increase in surface emissions of 6.9 ± 2.1 Tg/yr between 2019 and 2020, which they also mostly attributed to increased wetland emissions. These very large increments are consistent with the hypothesis that growth has been supercharged by precipitation in the unusual 2020-2022 La Niña "triple dip" event (NOAA 2023b, McPhaden et al. 2023), though it should be noted that earlier intense La Niña events did not coincide with years of especially extreme methane growth.

Table 2: Anthropogenic methane emissions in the years 2000, 2010 and 2020 and decadal growth rate in the last two decades. Data from EDGARv7.0 (EDGAR, 2023, Crippa et al., 2021)

IPCC sector	Emission Tg yr ⁻¹ CH ₄			Growth rate (Tg CH ₄ yr ⁻²)	
Year	2000	2010	2020	2000_2010	2010_2020
1 Energy	103.51	123.98	137.42	2.05	1.34
2 Industrial processes and product use	0.19	0.33	0.49	0.01	0.02
3 Agriculture, forestry and other land use	137.41	151.93	162.47	1.45	1.05
3A1 Enteric Fermentation	91.70	101.69	111.03	1.00	0.93
3A2 Manure Management	10.87	11.69	12.54	0.08	0.08

3C1 Biomass Burning	1.35	1.71	1.96	0.04	0.03
3C7 Rice Cultivations	33.49	36.84	36.94	0.33	0.01
4 Waste	62.72	70.60	83.58	0.79	1.30
4A Solid Waste Disposal	29.64	30.65	35.65	0.10	0.50
4B Biological Treatment of Solid Waste	0.17	0.23	0.28	0.01	0.01
4C Incineration and Open Burning of Waste	1.14	1.32	1.55	0.02	0.02
4D Wastewater Treatment and Discharge	31.76	38.40	46.10	0.66	0.77
5 Other	0.15	0.15	0.15	0.00	0.00
Total anthropogenic	303.98	346.99	384.11	4.30	3.71
Total 'microbial anthropogenic' (excluding FF etc)*	197.98	219.98	243.18	2.2	2.3

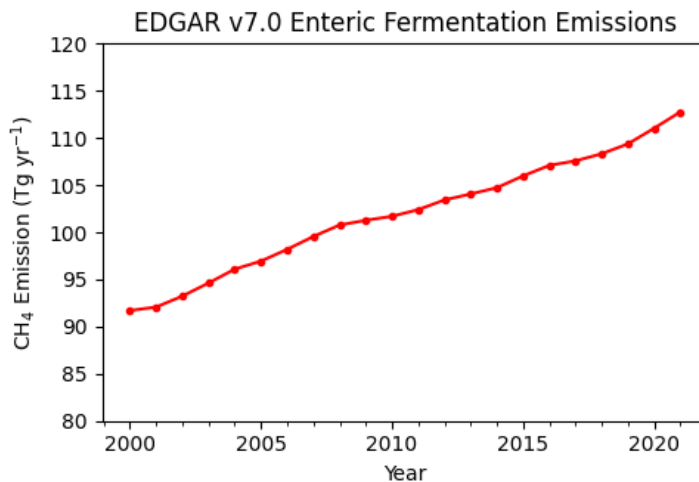
* excludes Energy (1), Biomass burning (3C1), and Incineration and open burning of waste (4C).

5. Agricultural, biomass burning, and waste emissions

Agriculture is a major source of biogenic methane emissions, from cattle breath ($\delta^{13}\text{C}_{\text{CH}_4}$ slightly more negative than the bulk methane burden), wetland rice cultivation, and biomass burning ($\delta^{13}\text{C}_{\text{CH}_4}$ much more positive than the burden)(for isotopic signatures of sources see Dlugokencky et al., 2011, Nisbet et al. 2016, 2019, MOYA/ZWAMPS Team 2021).

Livestock emissions are growing but the rate of growth is difficult to quantify. Zhang et al. (2022) estimated, with uncertainty around 20%, that methane emissions from global livestock populations were around 105 Tg in 2000, 121 Tg in 2011, and 132 Tg in 2019, growing by 27 Tg from 2000-2019, or 1.4 Tg/yr/yr. Using an inversion framework to verify methane emissions from satellite-based observations, Worden et al. (2023) reported a growth rate of livestock emissions of 0.25 to 1.3 Tg/yr/yr. The growth rate they estimated was similar to EDGAR results, although Worden et al. (2023) considered the EDGAR assessment had underestimated livestock emissions. From 2000-2020, the EDGAR (2023) assessment finds an increase in emissions from enteric fermentation of 20 to 21 Tg, (Fig. 6), with another 2 Tg increase from animal manure (EDGAR 2023) but no significant acceleration in the rate of increase in the past decade. Our assessment (Fig. 6) of growth in ruminant emissions from enteric fermentation, estimated from the EDGAR (2023) database, is nearly 1 Tg/yr/yr for 2000-2020 (Table 2), somewhat less than the Zhang et al. (2022) estimate but within the very wide uncertainties.

Figure 6. Estimated growth in methane emissions from enteric fermentation (ruminants), from EDGAR (2023). See Table 2. Recent (2010-2021) growth rate about 1 Tg/yr/yr.



The densest cattle populations are in South Asia, tropical Africa and tropical South America. Livestock population growth will be facilitated by increased grazing and browsing productivity in a warmer wetter climate. However, it is not clear how much agricultural expansion in the tropics has increased total land surface methane emission. Africa and more recently South America are undergoing a widespread transition from a complex 'natural' ecology of diverse ruminants and pseudo-ruminants to a now human-dominated pastoral system of cattle, goats and sheep (e.g. see Zhang et al. 2022).

In the 'natural' African ecology, ruminants from browsing giraffe to grazing buffalo exploit all parts of the floral variety. In contrast, in cattle-dominated monocultures, where most or all natural ruminants are eliminated, food production specifically for the very rapidly growing human populations will have increased, but the total body mass per unit area of the ruminants that are producing methane may not be larger (e.g. see data in Taylor and Walker (1978). Moreover, much tropical cattle manure falls on dry pastures in aerobic conditions not supportive of methanogens, although it should be noted that in some parts of Africa cattle are kept overnight in 'bomas' (corrals) where manure collects under urine and methane emissions can be significant (Leitner et al. 2023). Thus the overall net change in methane emissions from replacing an original productive and diverse natural ecology with anthropogenic cattle or maize monocultures is unclear.

Biomass burning emissions come from incomplete combustion. The amount of burning varies from year to year, which has significant impact on emissions (Worden et al. 2017). In the tropics, biomass fires are closely linked to changes in the Inter-Tropical Convergence Zone and the ENSO cycle. Grassland fires (mainly C4 plants in the tropics) are widespread in dry winters after good rainy seasons, often covering very large areas. Crop waste fires also occur mainly in dry season, varying with precipitation in the previous wet season. These fires, particularly of ¹³C-rich tropical C4 crop plants like maize, sugarcane, and millet, emit isotopically heavy methane.

Burning of tropical C3 woodland and forest also occurs, especially in clearing land. Tracking biomass burning is difficult: satellite observation quantifies burnt area but it is more difficult to assess fuel load and intensity of fire (especially important in crop waste fires). Barker et al. (2020) found high variability in emissions measured in flights over active fires, with methane emissions of 1.8 ± 0.19 g/kg of fuel over outer-tropical Senegal, but 3.1 ± 0.35 g/kg over equatorial Uganda. Boreal biomass burning, in contrast, is mainly wildfire in northern C3 forests, highly dependent on summer temperatures, and more easily assessed by remote sensing.

'Biogenic' CO is mainly produced by biomass burning, but the CO record in the 2000-2010 decade is affected by Europe's decade of 'dieselisation' favouring diesel-fuel passenger cars, which are low emitters of CO. Contrasting the period 2001–2007 to the 2008–2014, Worden et al (2017) concluded biomass burning emissions of methane decreased by $3.7 (\pm 1.4)$ Tg CH₄ per year, and this drop may have contributed to the standstill in methane growth in the early years of the 21st century. For the periods 2007-2012 and 2013-2017, inversions of both mixing ratio and isotopic data suggest overall global pyrogenic emissions dropped slightly or showed little change, (Zhang et al. 2021, comparison in Basu et al. 2022).

Much agricultural growth comes from expansion of farmed land area, displacing natural ruminants from grasslands or cutting forest and draining wetlands and burning peatlands. Thus, for tropical agricultural emissions it is difficult to allocate proportions of 'feedback-

driven' and 'technology-driven' emissions growth. The impact on crop yields and ruminant fodder supply of indirect factors such as CO₂ fertilisation and sulphate deposition, is not easily quantified.

Bottom-up (EDGAR 2023) inventories suggest anthropogenic emissions from wastewater, solid waste, and landfills are increasing worldwide. Despite successful efforts to reduce waste emissions in Europe and other regions, the rapid growth of tropical cities is driving increased urban waste disposal and these emissions are very poorly quantified (Nisbet et al. 2020). Waste disposal in tropical cities is increasing in large poorly-managed landfills (Nisbet et al. 2020), but food waste comes from local agriculture, so waste emissions are linked to local agricultural productivity. In the EDGAR v7.0 inventory (EDGAR 2023) the estimated growth of annual solid waste and landfill emissions from 2006 to 2021 is about 5.7 Tg, while methane from wastewater has increased by about 11 Tg/yr over the same period.

The 'climate feedback' proportion of growth in emissions of methane from tropical agricultural and waste disposal is uncertain. Growth in ruminant and landfill biogenic emissions depends both on global change feedbacks (more productivity in a warmer, wetter, CO₂-rich environment) and also on 'direct-action' factors increasing agricultural productivity such as nitrogen fertiliser application and genetic modification of seeds.

Overall, including solid and liquid waste, manure handling and enteric fermentation in agricultural ruminants, EDGAR inventory data (Table 2) suggest microbial methane emissions from anthropogenic activities rose each year by about 2.3 Tg/yr, or an increase of about 35 Tg from the end of 2006 to 2020. Although there is wide uncertainty on these bottom-up estimates, they appear broadly consistent with top-down estimates. Importantly, there is no evidence in the data for a sudden acceleration in the growth of agricultural and waste emissions post-2006.

6. Inference: increased natural emissions are a major cause of recent methane growth.

Quantifying emissions from 'natural' sources as they respond to climate change is difficult. Ongoing experimental measurement of methane fluxes from wetlands is minimal, especially in Africa. An alternative is to use isotopic information to assess global total (i.e. natural and anthropogenic) biogenic emissions and then subtract bottom-up estimates of emissions from biogenic sources under anthropogenic control, with the caveat that these too respond to climate warming.

Studying the period 1999-2016, Basu et al. (2022) estimated methane's 2008-2014 rise was driven by global emissions growth of $27.1 \pm 0.6 \text{ Tg yr}^{-1}$ or, nearly 4 Tg/yr/yr. They found the negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$ can be balanced by assuming about 15%, or 50 Tg, of the recent rise is from growth in fossil fuel emissions (i.e. with $\delta^{13}\text{C}_{\text{CH}_4}$ more positive than the methane already in the air). Comparing the period 2007-2012 with the period 2013-2017, Basu et al. (2022) found an increase in biogenic emissions of $32.3 \pm 3.2 \text{ Tg/yr}$. After 2017 and especially in 2020-2023, the burden grew more rapidly.

Similarly, looking at the surge in growth in 2019-2020, using a global inverse analysis of GOSAT satellite observations, Qu et al. (2022) found the imbalance between sources and sinks of methane increased by 31 Tg in the year 2019-20. They found that half the growth in methane emissions in 2019-2020 took place in Africa, especially East Africa, most likely from wetlands, with strong emissions growth in Canada also. For the year 2020, Qu et al. (2022)

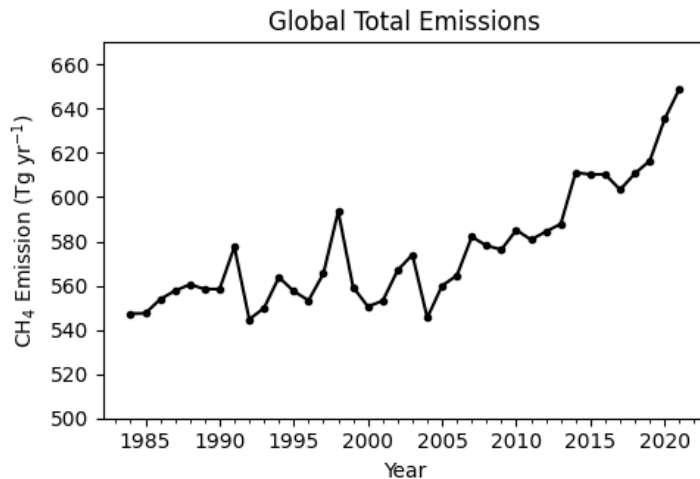
attributed $86 \pm 18\%$ of the growth surge to increased emissions, especially from wetlands in Africa (15 Tg/yr) and Canada and Alaska (4.8 Tg/yr), although there was also evidence for 14% of the growth coming from a decrease in methane's [OH] sink, to which also Stevenson et al. (2022) attribute part of methane's growth in 2020. Overall, Qu et al. (2022) found total emissions grew by 25–37 Tg/yr in 2019–2020. If their estimate that about 86% of growth was biogenic is accepted, then the growth in biogenic emissions in 2019–2020 was as large as 22–32 Tg.

Drinkwater et al. (2023), over the period 2004–2020, used *in situ* data to estimate a global annual emission increase of 93 Tg/yr over the period (620 Tg/yr minus 527 Tg/yr), with 83% of that increase occurring since 2011 when the global growth first began to accelerate. Assuming 2.3 Tg/yr/yr of growth is from anthropogenic microbial sources, increased wetland emissions will be responsible for the balance of emissions growth, of 2 to 2.5 Tg/yr/yr (with very wide uncertainty). Note also that the acceleration of methane's rise from 2020 implies the growth rate of emissions was much higher in 2020–2022.

More broadly, accepting the estimates of Basu et al. (2021) and Qu et al. (2022), and extrapolating to end-2022, a very rough guess is that about 70–80 Tg/yr of the global emission increase between 2006 and end-2022 came from specifically microbial sources, both natural and anthropogenic. As detailed above, microbial emissions from anthropogenic agricultural and waste sources have probably increased between 2006–2020 by about 35 Tg/yr (assuming linear growth, Fig. 6 and Table 2). If this is correct, the remaining recent net growth in methane emissions is not likely to be primarily attributable to agricultural expansion but has been driven by 'natural' biogenic processes, especially wetland feedbacks to climate change.

Fig. 7 updates estimated emissions into the 2020s, using the methodology of Lan et al (2021a) and Basu et al. (2022). Subtracting (say) 35 Tg for growth over this period in anthropogenic emissions from agriculture and waste, the growth in 'natural' biogenic emissions may have been 35–45 Tg. More generally, if total biogenic emissions continue to accelerate at the same rate as in the period 2006–2022 (a very large assumption), then by the year 2037, 30 years after the start of growth, specifically biogenic emissions might have grown by perhaps 150 Tg/yr.

Figure 7. Global methane emissions, estimated and updated using the top-down methodology of Lan et al. (2021a) and Basu et al. (2022). For further details, see supplementary information in Lan et al. 2021a, Fig S2b, red curve, updated.



7. Glacial Terminations

During the past half-million years, several "termination" events have marked rapid changes of global climate from cold glacial to warmer inter-glacial conditions. Terminations were rapid major reorganisations of the Earth's climate systems, with degassing of upwelling oceans and revival of a warmer biosphere (Broecker and van Donk, 1970; Raymo 1997; Cheng et al. 2009; Denton et al. 2010; Lang and Wolff 2011, Schmitt et al. 2012). They are characterised by very large-scale shifts of global and regional weather circulation patterns, for example from glacial climates toward a more La Niña-like state (Cheng et al 2020).

Broecker and Denton (1990) interpreted 'terminations' in the broadest terms, as global-scale climate reorganisations, which were synchronous with sharp changes in the methane burden. likely driven by wetland emissions (Nisbet 1990, Nisbet and Chappellaz 2009). The modern palaeoclimate understanding of a recognised 'Termination', named in Roman numerals from I (youngest) to IX (nearly 800 ka ago) refers to specific transitions from glacial to interglacial periods that took place about every 100 ka. In the finer detail of the glacial record are Dansgaard-Oeschger (D-O) events, which were abrupt warming events, with sharp temperature rises over decadal timescales, followed by slow relaxation over millennia. These D-O events were very rapid, with decadal progression (Erhardt et al. 2019). While Dansgaard-Oeschger events occurred within glacials and were likely linked to rapid changes in sea ice cover (Boers et al. 2018), the numbered Terminations marked global-scale climate changes.

Although full terminations took several millennia before the new warmer interglacial climate was established, the transitions typically included brief abrupt phases when methane growth and climate warming was very rapid indeed. It is likely that the new warmer weather patterns became established within a few centuries or less (Severinghaus et al. 1998), although it took millennia for the great ice domes to melt and, by their shrinking, to change boreal albedo and replace ice with wetlands and forest, thus completing the full climate transition of the termination.

Sudden methane growth is the bellwether of these changes (Möller et al. 2013, Bock et al. 2017). Methane emissions are very responsive to climate (Nisbet 1990, Petrenko et al. 2009, Nisbet & Chappellaz 2009). Methane's growth record during glacial-interglacial Terminations V to I (Fig. 8 and Table 3) is preserved in bubbles in ice cores (Spahni et al. 2005, Wolff et al.

2009, Möller et al. 2013, Bock et al. 2017). The methane record correlates with monsoonal patterns as observed in oxygen isotope records in stalagmites (Cheng et al. 2009).

During Termination IA ending the Younger Dryas, methane rose abruptly by ~250ppb, with essentially synchronous high-latitude and low-latitude climate change (Capron et al. 2021). Ice core data imply emissions increases as large as 6 to 21 Tg/year causing methane growth rates as high as 6 ppb/yr (Rhodes et al. 2015). It is likely methane's rapid growth was primarily driven by rising tropical emissions, responding to abrupt changes in tropical rainfall (Schaefer et al., 2006; Möller et al. 2013, Bock et al. 2017, Riddell-Young et al. 2023).

Feedback loops in the methane budget rapidly respond to global-scale climate change. It is likely that both warming and climate shifts such as intensification of ENSO events would have especially driven emissions in the tropics where both wetlands and regional land vegetation respond quickly to increased warmth and rainfall, wetland productivity increasing synchronously with parallel expansion of methane-emitting (Clauss et al. 2020) populations of ruminants (buffalo, antelope) and pseudo-ruminants (hippopotamoi).

Figure 8 shows the changes in methane and $\delta^{13}\text{C}_{\text{CH}_4}$ in selected past terminations, showing data updated from Bock et al. 2017; see also Möller et al. (2013). Initially, at the start of a typical termination, there was a gradual rise in methane and CO_2 . This rise was accompanied by a negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$, which covaried with CO_2 , as was characteristic during glaci- als, and with CH_4 . This gradual phase lasted a few thousand years.

In typical terminations the brief abrupt phase, with sharp growth in methane, In typical terminations the brief abrupt phase with its sharp growth in methane by several 100 ppb did not leave a significant imprint in the $\delta^{13}\text{C}_{\text{CH}_4}$ record (Schaefer et al. 2006; Möller et al. 2013, Bock et al. 2017) indicated with arrows in Fig. 8. This pattern is consistent with the interpretation that methane emissions growth was dominated by equatorial wetland which has $\delta^{13}\text{C}_{\text{CH}_4}$ close to the global bulk source (Brownlow et al. 2017, Ganesan et al, 2019, Nisbet et al. 2021a,b, France et al. 2021, Oh et al. 2022). Methane's sharp growth in the abrupt phase is thus best explained by a rapid increase in emissions from tropical wetlands emitting methane with $\delta^{13}\text{C}_{\text{CH}_4}$ close to that of the bulk atmosphere (i.e. especially in the equatorial zone) (Bock et al. 2017).

Switching on tropical wetlands was likely associated with a rapid shift in the ITCZ that coincided with resumption of a strong Atlantic Meridional Overturning Circulation (AMOC) at the start of interglacial climate conditions, initiating a warmer northern hemisphere and generally stable climate with subdued millennial-scale variability. The AMOC 'flip' is denoted in Fig. 8 by vertical dashed lines, and essentially marked the crux of the termination, although it took millennia for sea level to rise as the great ice domes in Canada and Scandinavia melted. During the 'flip' phase, there was typically a rapid increase of about 200 ppb in methane mixing ratio, but only a relatively small change in $\delta^{13}\text{C}_{\text{CH}_4}$.

AMOC changes and consequential meridional heat transport anomalies have been a feature of past episodes of abrupt global climate shifts (Cheng et al. (2007). For example, Riveiros et al. (2013), and Rhodes et al. (2015) showed that AMOC slowdown is capable of producing atmospheric responses that extend throughout the Northern Hemisphere and into the tropics, with strong coupling between the two regions. Though very different from today's changes, the 32 ppb rise in atmospheric methane during the HS 1 event reported by Rhodes

et al. (2015) has interesting parallels with modern shifts, including both weakening of AMOC and southern expansion of the ITCZ and Hadley cell. However, there are also large differences: in HS1 there was boreal cooling as the ITCZ moved southward; in the present event the north is warming strongly.

Although there are many possible explanations of this sequence of events, the pattern is consistent with a wider scenario beginning with initial modest Hadley cell expansion during interdecadal variation, bringing about more methane emission from wetter outer tropical swamps ($\delta^{13}\text{C}_{\text{CH}_4}$ perhaps very roughly -60‰ in cool conditions). Then in response, changes in AMOC, the Walker circulation and the Indian Ocean Dipole (Abram et al. 2020, Brierley et al. 2023), may have brought about intensification of equatorial rainfall and consequent rapid growth in equatorial wetland emissions, initiating methane's abrupt rise.

Thus in this scenario of the abrupt phase of a glacial termination, the growth of wetlands under strong equatorial rainfall will rapidly increase the atmospheric methane burden but be accompanied by only subdued initial leverage on $\delta^{13}\text{C}_{\text{CH}_4}$. Then, as the boreal realm warms, northern methane sources with more negative $\delta^{13}\text{C}_{\text{CH}_4}$ begin to contribute. Note that in an end-glacial scenario the presence of large ice domes over Canada and Scandinavia inhibited development of boreal vegetation: in these areas boreal wetlands were not functioning until millennia later when the ice melted. Today, the boreal and Arctic wetlands, which emit methane with markedly negative $\delta^{13}\text{C}_{\text{CH}_4}$, are very productive and are experiencing strong warming (Previdi, et al. 2021).

Figure 8. Ice core records from Terminations I, II and V. Modified from Bock et al. 2017 and updated. Note synchronicity of CH_4 rise with negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$ (plotted upwards on y-axis). Solid arrows denote the CH_4 from the start of a Termination to the point of the growth flip. Dashed arrows mark the corresponding CH_4 change in the abrupt flip. CO_2 record is from Bereiter et al. (2015) and CH_4 from Loulergue et al. (2008) with updates from Bock et al. (2017).

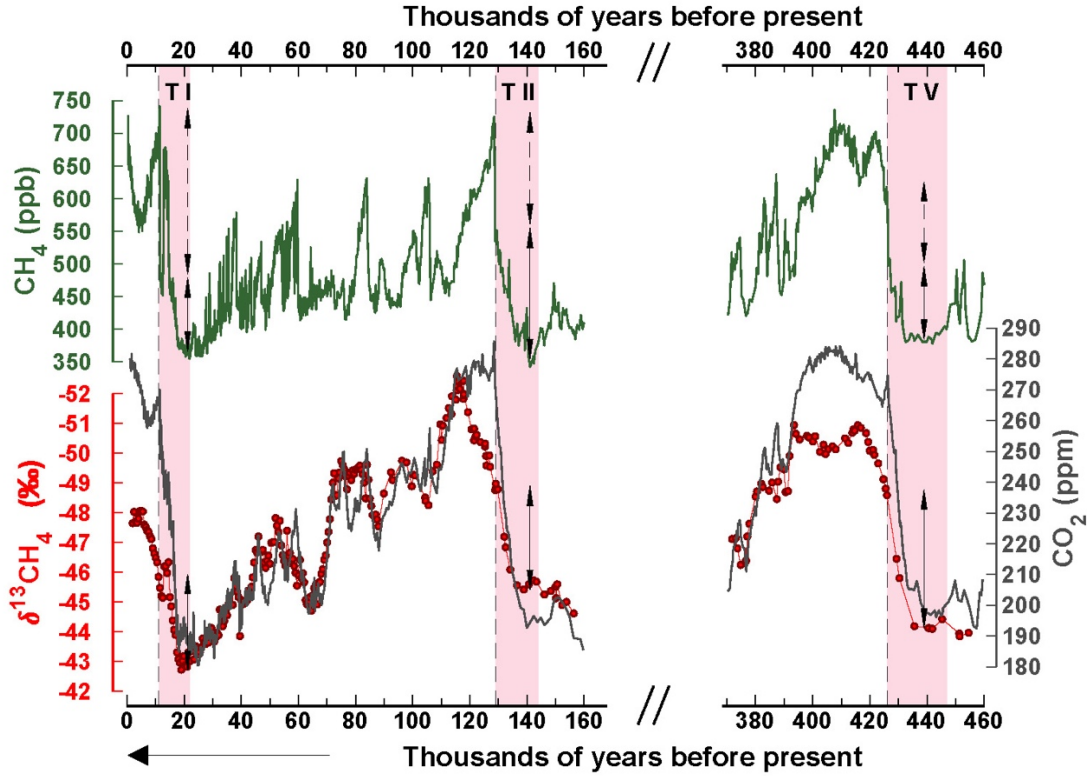


Table 3: Synopsis of Termination changes

Summarised here from methane records detailed in Section 7, particularly Bock et al. 2017. Assumes 1 ppb is equivalent to 2.77 Tg methane.

Event	Approximate Date (ka BP) of abrupt phase	Change over whole termination ppb (approx.)	CH ₄ Change over whole termination Tg (approx.)	Maximum Growth rate ppb/yr (approximate)	δ ¹³ C _{CH₄} shift over whole termination (approximate)
PAST					
Termination IA	11.64	250	700-	> 2.5	-3.5‰
Termination IB	14.65	150	400-450	> 2	
Termination II	131	350	900-1000	> 2.5	-4.5‰
Termination III	250	300	800-850		
Termination IV	345	400	1100		
Termination V	430	350	900-1000		-5‰
MODERN					
Modern	2006-2022-?	135	374 318 (biogenic 85% component)	~ 9 (Between 6 and 18 ppb)	-0.7‰
Model to 2035	2006 to 2035	573	1584	10	-1.1‰ (roughly equivalent to -4.4‰ if injected into much smaller glacial burden)

Over full terminations, negative drops in $\delta^{13}\text{C}_{\text{CH}_4}$ ranged between -5‰ to -3‰ (Bock et al. 2017). For example, in Termination V, $\delta^{13}\text{C}_{\text{CH}_4}$ dropped from about -44‰ to -49‰, while in Termination II there was a similar negative shift (Bock et al. 2017). Again, at the start of the Preboreal Holocene $\delta^{13}\text{C}_{\text{CH}_4}$ shifted from about -43‰ in full glacial conditions, to about -46.3‰ (Fischer et al. 2008, Melton et al. 2012; Bock et al. 2017).

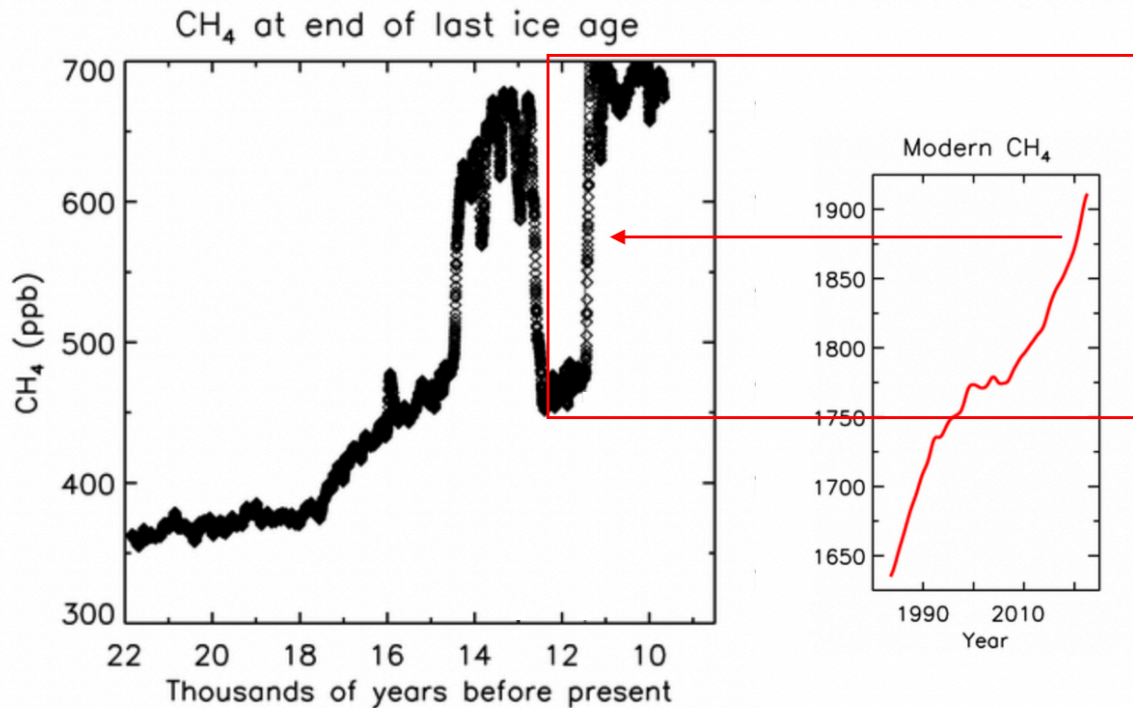
To consider the older events first, the abrupt phase of Termination II took place between 130.9 ± 0.9 and 130.7 ± 0.9 ka ago (Moseley et al., 2015), with contemporary strengthening of the monsoon. In Terminations II and III the methane burden rose by roughly 500-600Tg in sudden growth events. During similar abrupt rises in Terminations IV and V, methane rose from about 400ppb to 700ppb, or more, increasing the methane burden by 800-900Tg.

The most recent termination was in multiple phases (Fig.9 left panel). In the abrupt part of Termination 1B, which ended the last major glaciation, the Greenland Summit climate rapidly warmed by $9 \pm 3^\circ\text{C}$, and 20-30 years later methane rose abruptly (Severinghaus and Brook, 1999). Methane rose quickly over the sixty years between 14,655 and 14,595 BP, with especially rapid growth in the decade from 14,655 - 14,645 BP (Severinghaus and Brook 1999). This change initiated the Bølling–Allerød interstadial, until 12.87 thousand years BP, when the Younger Dryas began, with sudden cooling of the North Atlantic realm linked to a drop in methane to 450-500 ppb. Then, ending the Younger Dryas, the abrupt warming event of Termination 1A took place between $\sim 11,700$ and 11,610 B.P, with the fastest change about 11.64 thousand years ago (indicated by vertical line in Fig. 8). About 0-30 years after this warming began, methane rose sharply to above 700 ppb (Severinghaus et al 1998; Cheng et al. 2020). In all, methane rose as much as 250 ppb in little over a century, an increase of about 700 Tg in the burden.

Figure 9. Comparison between Methane's record during Termination 1 and methane's growth since 1980s. Note – these plots are not to the same time scale.

Left panel: Methane record during Termination 1. Prior to about 17ka before the present, methane was below 400 ppb (from various sources: see text). Methane rose in Termination 1B, especially in the abrupt phase about 14.65 kyr ago. The Younger Dryas. 12 ka ago, saw a return to cold conditions, especially in the northern North Atlantic, abruptly ended in Termination 1A, 11.64 kyr ago, date shown by vertical line.

Right panel: modern methane record. Growth prior to 2006 shows convex equilibration trend; growth from 2006 shows concave (accelerating) trend (NOAA data). Note time scales not the same.



There is evidence for strong growth in tropical (30°S to 30°N) methane emissions during Termination 1 (Riddell-Young et al. submitted 2023, Baumgartner et al 2012), as tropical wetlands responded to an abrupt expansion of tropical rain belts. Methane output may have risen from perhaps 60 Tg/yr during glaciation to 120 Tg/yr or more in the post-termination Holocene. That global increase of 60 Tg across the whole event can be compared to the present-day estimate derived above that 'natural emissions' have increased by about 35-45 Tg since 2006. Riddell-Young et al. (submitted 2023) and Baumgartner et al. (2012) also found evidence for a supplementary role of emissions from boreal methane sources, findings prescient of the present-day results of Qu et al. (2022). Similar patterns are also inferred from the weak interpolar gradient for methane in Termination 1B which suggests methane growth was driven by a major tropical input (Severinghaus and Brook 1999).

In addition to termination events, examples of rapid changes in atmospheric methane, in which methane rose by 32-53 ppb, are also recorded in Heinrich events during glacial periods. Though not analogous to present-day conditions, the rapid methane changes are instructive as they show how responsive methane emissions are to climate shifts. Heinrich events are marked by layers of ice-rafted debris in the North Atlantic seabed record, that must have been dropped by armadas of icebergs breaking off the ice sheets over Greenland and North America. Studying the HS1 event (16.15 kyr BP), which occurred near the end of the last major glacial period, Rhodes et al. (2015) inferred a marked southerly shift in the Inter-Tropical Convergence Zone had taken place in response to Northern Hemisphere cooling and greater sea-ice cover, causing a major expansion in southern hemisphere wetlands, which in turn drove a rapid increase in methane emissions by 29 Tg/yr.

To conclude, the methane and $\delta^{13}\text{C}_{\text{CH}_4}$ records of glacial/interglacial events are consistent with the hypothesis that the abrupt phase of methane growth was driven by a very rapid biogenic response, mainly from tropical wetlands.

8. Meteorological Changes

Meteorologically, past termination events may have originated in specific regional locations and then spread globally. For example, in the North Atlantic realm, the rapid meteorological changes in the most studied abrupt termination event, the end of the Younger Dryas (T1A), may have been a consequence of a set of changes originating in the tropics and then spreading to include the North Atlantic, the Asian Monsoon and the Westerlies (Cheng et al. 2020). Moreover, model experiments typically show a spin-up time before any change has impact on the global methane budget: thus for a methane signal to have become identifiable in the period 2007-2022, the changes triggering the necessary spin up would have been perhaps a decade or more ago.

If the post-2006 methane growth represents more than multi-decadal variability and is indeed the start of a major long-term reorganisation of the planetary climate system, then two questions arise: Can such global-scale modern meteorological changes be identified, given that both short-term change and sustained regional multi-decadal variability are characteristic of the Holocene climate (Wanner et al, 2008)? Are there today any direct signs of incipient global-scale meteorological reorganisation?

Recent meteorological changes, though different in style and regional impact, may indeed be similar in speed and planetary scale to global changes during termination events. During 2006-2020 the total Earth system has experienced a remarkable heating of $0.76 \pm 0.2 \text{ Wm}^{-2}$ with about 89% of the heat stored in the ocean, 6% on land, 4% in the cryosphere and 1% in the atmosphere (von Schuckmann et al. 2023). Even a small increase in the partitioning of this heat into the air will have dramatic impact on global weather.

Arguably the most profound global-scale evidence for present-day climate warming comes from the heat content of the upper part of the oceans (von Schuckmann et al. 2023), which not only has a sustained and strong warming trend, but the rate of warming is accelerating (Cheng et al 2023). Warming has been particularly anomalous in the South Atlantic, in the North Atlantic offshore North America and in the Indian Ocean offshore Southern Africa. Ocean heat produces evaporation and thus precipitation: wetlands will respond. The 'triple-dip' La Niña of the first years of this decade must be a factor (McPhaden et al. 2023), while multi-centennial variability of internal origin may also be important.

In recent years, there has been marked weakening of the Atlantic Meridional Overturning Circulation (AMOC), in particular since 2005 (WMO 2023, Bonnet et al, 2021). Concurrently, changes in the Walker circulation (El Niño /Southern Oscillation, ENSO) and the Indian Ocean dipole have had profound impacts on tropical precipitation and temperature on land. Hadley cell expansion has shifted the zone of tropical cyclones (Sharmila and Walsh 2018), with poleward shifts in extra-tropical storm tracks and jets, and upper tropospheric warming. As during glacial/interglacial transitions, and particularly in the abrupt 'flip' phase, the Hadley circulation is expanding again today and the extension of the subtropical edge of the Hadley circulation has already had impact on a wide range of extratropical biological productivity (e.g. Pandey et al. 2017, Osland et al 2021). The poleward shifts of the planetary circulation and the concurrent expansion of tropical weather systems are now becoming well-documented (Staten et al. 2020, Grise et al. 2019, Voight et al. 2021, Studholme et al. 2022).

Tropical weather systems are tightly interlinked (Cai et al. 2019, McPhaden et al. 2023), with widespread major recent changes. Geng et al (2022) have suggested that increased ENSO changes may emerge much sooner than hitherto expected, while Wang et al. (2019)

presented evidence for major shifts in the origination and strength of El Niño events, finding more frequent occurrence of extreme events since the 1970s and changes in ENSO onset from originating in the eastern Pacific to the western Pacific. McPhaden et al. (2023), Hasan et al. (2022) and Jo et al. (2021) suggested linkage between the Indian Ocean Dipole and Atlantic and Pacific conditions. Sun et al. (2022) present evidence for substantial increases in early onset of monsoon weather and positive Indian Ocean Dipole events, with dramatic impacts on regional flooding. For example, catastrophic rains and floods in East Africa in 2019 came from a remarkable concatenation of meteorological factors, including record levels of the Dipole Mode Index, ENSO effects, anomalous zonal winds over the central Indian Ocean, very warm sea surface temperatures in the eastern Atlantic and Indian Oceans, and unusual zonal circulation (Nicholson et al 2022).

There have been recent large-scale shifts in the Indian Ocean Dipole (IOD) and unusual ENSO changes (Pandey et al. 2017). These weather changes have profound impacts on methane emissions. Feng et al. (2022) attributed much of the change in methane emissions over tropical South America and tropical Africa to IOD and ENSO shifts. Dramatic changes may be affecting the Indian Ocean Dipole, which plays a major role in controlling rainfall over the wetlands of east sub-Saharan Africa (Feng et al, 2022), with recent trends towards more frequent and intense positive events (Abram et al. 2020). During strong La Niña episodes, rainfall can deeply fill wetlands, while during El Niño episodes various parts of Amazonia and East Africa have experienced changes likely related to climate warming, with longer dry- and shorter wet-seasons in some places, and fires spreading to areas that have not burned for millennia; the opposite taking place in other regions. In particular, rains in Eastern Africa have strong links to the ENSO cycle and are dependent on changes in the Walker circulation (Nicholson 2015) and Indian Ocean dipole, together with the strength of pulses of the Madden-Julien oscillation.

Polar climates are also changing rapidly (Łupikasza & Cielecka-Nowak, 2020) especially with Arctic amplification of warming (Previdi et al. 2021). Arctic sea ice is in very rapid retreat (Kim et al. 2023). Arctic amplification may be linked to a negative Northern Hemisphere Annular Mode, large scale circulation changes, and increased durations of weather patterns and extreme precipitation over the Northern Hemisphere (Liu et al. 2021). Weather patterns are shifting in continental interiors (IPCC 2021 their Fig. 8.11), with regional warming across Siberia (Wang et al 2022). Regional climate warming has had significant impact on plant productivity, promoting the greening of the tundra (Berner et al. 2020). Boreal and Arctic warming have had marked effects on temperature and precipitation over large swampy areas, for example in western Siberia (Gorbatenko et al 2020), presumably by increasing supplies of organic detritus and thus methanogen productivity.

These profound ongoing global-scale changes in the modern ocean and atmospheric circulation patterns have many parallels in past termination events (see Section 7 and Fig. 8), though with the caveat that the modern world is profoundly different, especially in the absence of the major meteorological impacts of the large ice domes over Canada and Scandinavia. Thus any global-scale meteorological changes that occur in the next few decades will clearly be very different from the glacial/interglacial transitions. Yet perhaps there are analogies too, particularly in the paleoclimatological evidence that the global climate system can reorganise very rapidly indeed, over as short a time period as a few decades.

A further caveat: although the present-day meteorological changes are widespread and profound, clearly on an inter-hemispheric scale, and likely involving multiple interlinked teleconnections, they may yet still be within the multi-decadal or centennial spectrum of Holocene variability. Many major meteorological circulation patterns have only recently been identified and their record of change compared to the recent historic past remains poorly documented, although the IPCC (2021) Summary for Policymakers commented that evidence of observed change has strengthened since the previous assessment report. It is not clear that current climate extremes, though large, abrupt, and ongoing, and with potent impact on methane emissions (Feng et al 2022), are great enough in sum to be evidence for an ongoing global-scale meteorological reorganisation. A grand-scale meteorological progression of the type identified by Cheng et al. (2020) would be hard to identify from weather data while it took place, and likely only recognisable in retrospect. The evidence remains unclear.

Moreover, apart from the measurement of heat stored in the oceans (Cheng et al. 2023, von Schuckmann et al. 2023), it is difficult to quantify the total sum of change, nor to quantify the collective impact on global climate. But equally it is plausible that the multiplicity of recent events, their abruptness, and their power are circumstantial signs a major reorganisation of the global climate system has already begun. Like the heat of the oceans, the biogenic methane signal may be a telling summation, a bellwether that signals the onset of permanent global change, the end of the Holocene.

To summarise the results in Sections 1-8: In the period 2007-2022, methane's growth rate has been very high, with an overall concave (accelerating) trend and a significant negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$. If the estimates given above of the growth in agricultural emission are accepted, then much of the new emissions that are driving this growth come from wetlands and other natural sources, as found by Basu et al (2022), Qu et al. (2022), Feng et al. (2022, 2023), Peng et al (2022) and Rosentreter et al. (2021). If the "85% biogenic" estimate (Basu et al. 2022) continues to apply in the year 2022, growth from specifically biogenic emissions alone (i.e. growth from climate change feedbacks, both wetland and agricultural and waste) may have been 15 ppb, implying the present-day 'non-fossil fuel' growth trend by itself is significantly faster than the maximum growth rates in the ice core records (Table 2).

9. Modelling

9.1 Modelling Methodology

Is methane's growth since late 2006 now becoming comparable in scale and in cause to methane's growth during a termination event?

To address this question, modelling focussed on the question: what would happen to the methane burden and its $\delta^{13}\text{C}_{\text{CH}_4}$ during the abrupt phase of a termination event, if the abrupt growth took place today? This is not a model to replicate the palaeoclimate record but to elucidate modern trends. Thus the model is set up with a modern distribution of wetlands, including both tropical and boreal wetlands (which were largely absent in glacial periods). The model is designed to match century-scale trends over a conceptual abrupt growth phase and to follow major emission variations.

A modified version of the model used in Nisbet et al. (2019) was used. The model is a running budget model with 4 semi-hemispheres and exchange rates between them consistent with the observed interhemispheric rate of mixing. Methane removal uses $[\text{OH}]$

destruction from Spivakovsky et al. (2000) with kinetic isotope exchange from Cantrell et al (1990), Cl removal from Hossaini et al. (2016), and soil removal from Curry (2007). The model was designed to examine changes in methane mole fraction and in methane's isotopic content, averaged over four semi-hemispheric regions (30–90°S, 0–30°S, 0–30°N, and 30–90°N), with exchange rates between them consistent with the interhemispheric transport time. Fitting methane's carbon isotopic record is central to the modelling because of the powerful insights $\delta^{13}\text{C}_{\text{CH}_4}$ provides into changing methane sources (Craig et al. 1988).

We first modelled an abrupt ('flip') phase patterned on a the abrupt phase of a 'typical' termination event, to understand its dynamics. Then, having characterized the abrupt phase of a typical event, we used the model to match the current (21st century) growth record. The model tracks the impact of a sharp increase in biogenic methane, to test whether it is comparable to the sudden changes in a typical termination.

9.1.1 – Termination event model – changes in methane burden and $\delta^{13}\text{C}_{\text{CH}_4}$

We simulated modern-type emissions growing over a time period of 30 years, into a putative model of methane's glacial-like atmospheric burden to see if a 'termination-like' pattern developed, not just replicating the growth in methane burden but also to investigate the $\delta^{13}\text{C}_{\text{CH}_4}$ isotopic consequences.

Biogenic sources used in each semi-hemisphere for the “Ice-age” and “Termination 1” periods are scaled down from a modern scenario fitting recent CH_4 and $\delta^{13}\text{C}_{\text{CH}_4}$ data (Nisbet et al 2019) and are not from glacial-period biosphere reconstructions, but the fossil fuel contribution, taken as 30% of the total source at present (Lowe et al. 1988; Lassey et al. 2007), was omitted. Relative to the total, the distribution of CH_4 sources by semi-hemisphere was: 12.4% in 30-90°S (HSH); 27.7% in 0-30°S (LSH); 31.1% in 0-30°N (LNH) and 28.8% in 30-90°N (HNH). The analysis of geographic spread was not expected to be very sensitive to the latitudinal distribution of fossil methane sources but the omitted recent fossil fraction was taken as being distributed 20% : 40%: 40% in LSH : LNH : HNH. Omitting the fossil fuel emissions source leaves a distribution for the other sources as: 17.7% in HSH; 31.0% in LSH; 27.3% in LNH; and 24.0% in HNH.

These proportions according to latitudinal zone applied to a global total source consistent with sources scaled to give a global mean CH_4 mixing ratio of 700 ppb for "post-transition" and 400 ppb for 'initial' conditions. The initial scenario also reduced the 30-90°N (HNH) region source further because cold temperatures restricted land plants and especially wetland areas. Global average results are not particularly sensitive to how much the HNH region is reduced relative to the global total source, but results are shown for the HNH region source being reduced from 24% to 16% of the total with the other 8% distributed equally over LNH and LSH.

The isotopic signatures of the “Initial” (i.e. pre-transition) sources (S), here termed $\delta^{13}\text{C}_\text{S}$, are taken as having $\delta^{13}\text{C}_\text{S}$ 4‰ heavier than present sources, and the source pulse of increased emission leading into the transition is taken as having $\delta^{13}\text{C}_\text{S} = -59\text{‰}$, a value typical of modern outer (seasonal) tropical wetlands (France et al. 2021, MOYA/ZWAMPS Team 2021). The transition between the "interglacial" and "glacial" states of the global methane budget is done with a constant rate of change in the source, and its $\delta^{13}\text{C}_{\text{CH}_4}$, over 30 years. Mixing ratio, source emissions, Src (Tg/yr), source signatures for $\delta^{13}\text{C}_\text{S}$, and atmospheric $\delta^{13}\text{C}_{\text{CH}_4}$, here termed $\delta^{13}\text{C}_\text{M}$, are summarised in Table 4.

Table 4: Source emissions, $\delta^{13}\text{C}_\text{s}$, mixing ratios and atmospheric $\delta^{13}\text{C}_\text{M}$ in modelled 'Initial' (pre-transition) to post-transition ('Post') values

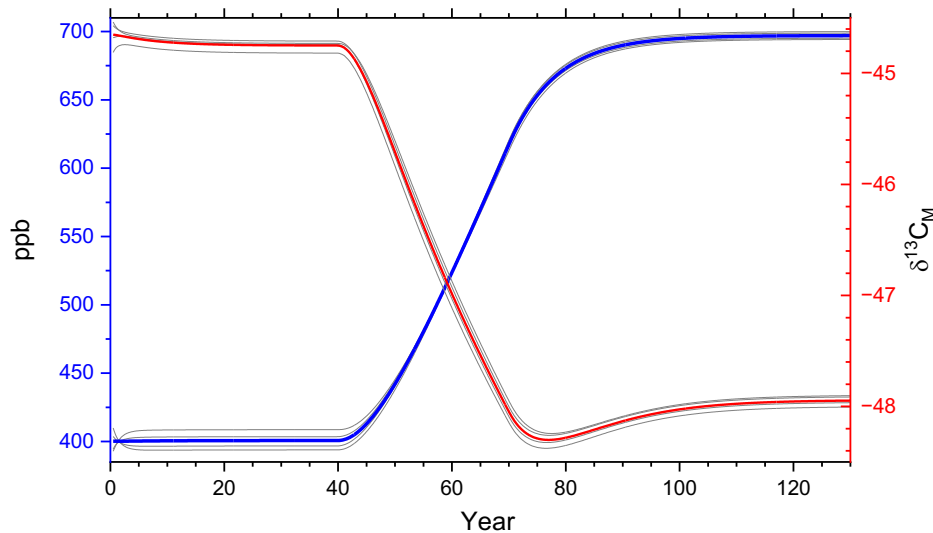
	Source Tg/yr (running annual average)					Source $\delta^{13}\text{C}_\text{s}$ ‰				
	30-90°S	0-30°S	0-30°N	30-90°N	Global	30-90°S	0-30°S	0-30°N	30-90°N	Global
Initial	23.6	46.7	41.8	21.4	133.5	-49.8	-50.8	-51.7	-56.4	-51.8
Post	30.9	78.2	79.6	45.1	233.7	-53.8	-54.3	-54.8	-57.5	-55.0
	Mixing ratio ppb (running annual average)					Atmospheric $\delta^{13}\text{C}_\text{M}$ ‰				
	30-90°S	0-30°S	0-30°N	30-90°N	Global	30-90°S	0-30°S	0-30°N	30-90°N	Global
Initial	408.6	403.5	396.7	393.8	400.7	-44.7	-44.7	-44.7	-44.82	-44.75
Post	694.2	695.1	698.9	699.9	697.0	-47.9	-47.9	-48.0	-48.0	-48.0

9.1.2 Modelling Results.

Plots of the changes in methane mixing ratio and $\delta^{13}\text{C}_{\text{CH}_4}$ in the modelled 'Source-jump' Scenario (Table 4), are shown in Figures 10a and 10b. This sequence of events is broadly similar to the real records of the methane burden in typical glacial terminations. The isotopic results reflect the very rapid growth of emissions from all latitude zones, both tropical and boreal. The isotopic results show the 'Tans effect' (Tans 1997), with changes in $\delta^{13}\text{C}_\text{M}$ overshooting followed by a slow recovery – i.e. with $\delta^{13}\text{C}_\text{M}$ taking longer to equilibrate than the mixing ratio. Inter-hemispheric spatial gradients take time to adjust after major changes in emissions and Tans (1997) pointed out that the timescale for the $\delta^{13}\text{C}_{\text{CH}_4}$ ratio is also longer than for methane's abundance so provides complementary information.

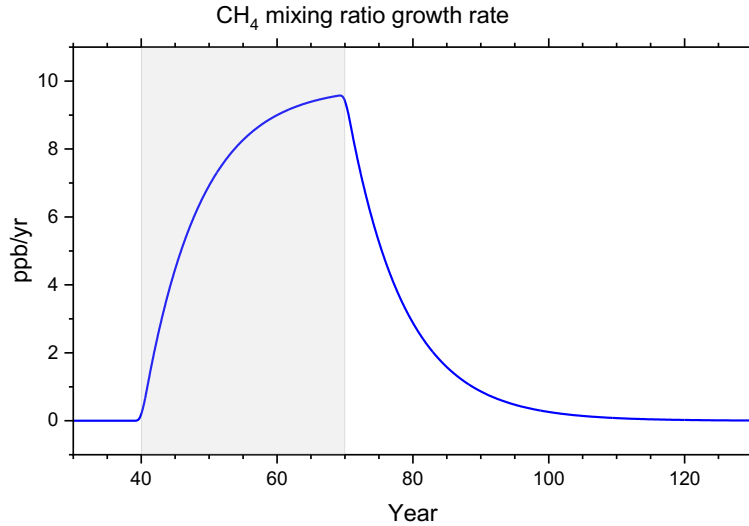
In glacial terminations, the boreal wetlands took longer to deglacierate and hence the dominance of tropical emissions with $\delta^{13}\text{C}_{\text{CH}_4}$ close to the bulk atmospheric source led to a brief hiatus in $\delta^{13}\text{C}_{\text{CH}_4}$ while the burden grew. In the model, unlike the real past record, the abrupt growth of boreal sources produces a significant negative shift in $\delta^{13}\text{C}_{\text{CH}_4}$.

Figure 10a. Methane mole fraction ($\delta^{13}\text{C}_\text{M}$) in ppb and $\delta^{13}\text{C}_{\text{CH}_4}$ from the 4-box running budget analysis with a 30-yr transition from 'Initial' to 'Post' sources. Blue is the global average mole



fraction and thin lines are for the four semihemispheres. The latitudinal distribution of the CH_4 mole fraction reverses from being lowest in 30-90°N to being the highest there. Red is the global average $\delta^{13}\text{C}_M$ and thin lines show values for each semihemisphere and this change in $\delta^{13}\text{C}_{\text{CH}_4}$ is uniform across all semihemispheres.

Figure 10b. Growth rate (ppb/yr) of the global average CH_4 mixing ratio in Fig. 8a.



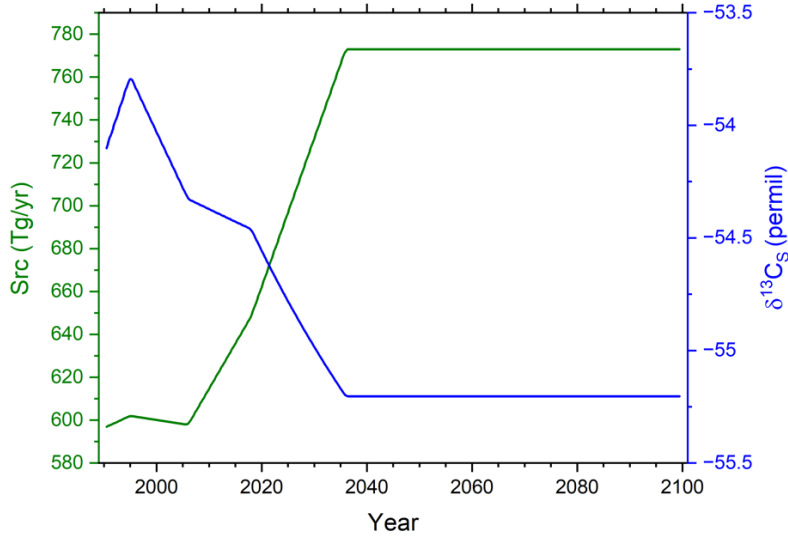
9.2. Present-day scenario: Modelling methane's modern growth and predicting the near future

To predict the impact if current trends continue for a full span of 30 years, from end-2006 to 2037. the second model experiment focussed on matching the current growth in methane emissions and then projecting into the future until the change is assumed to be completed after 2036. This is an optimistic scenario that assumes good progress in the Global Methane Pledge to reduce methane emissions from purely abiogenic (fossil fuel) sources, and that biogenic emission growth then slows in a stable future climate.

9.2.1 – Modern event methane growth model – changes in methane burden and $\delta^{13}\text{C}_{\text{CH}_4}$

The 'modern-day' model run is for a 30-year period 2007-2037, together with initialising years and years of post-change stability in emissions budgets. After the 30-year period, emissions stabilise. In this model, sources are fitted to observations with a simple piecewise linear fit of both source emissions, Src (Tg/yr) and $\delta^{13}\text{C}_s$ up to 2022 (Fig. 11). Then the projections in Figs 11a and 11b that go beyond 2022 extend the source flux growth that was fitted to the most recent data, and continue for the rest of a 30 year transition period. The isotopic signature $\delta^{13}\text{C}_s$ is determined by assigning a constant $\delta^{13}\text{C}$ lighter than the current average $\delta^{13}\text{C}_s$ to the Src increase.

Fig. 11. 'Modern-day' model for a 30 year transition period 2007-2037 assuming a change from earlier global climate state ending in late 2006 to new stable climate state beginning in 2037. Source flux (Src) and source isotopic signature $\delta^{13}\text{C}_s$ modelled to fit to measurement data (see Fig. 11a) up to 2022, thereafter projected to end of 30 year period, after which Src and $\delta^{13}\text{C}_s$ remain constant.



For the *Present-day Scenario*, Figure 12a shows NOAA methane mixing ratio and atmospheric $\delta^{13}\text{C}_{\text{CH}_4}$ data together with an extrapolated extension of the running budget analysis in Nisbet et al. (2019). The fit-to-data follows a smoothed version of the observed mixing ratio, shown in Figure 12a, with a maximum growth of 14.5 ppb/yr in 2022 (Figure 12b), when the observed rate was actually about 14 ppb, following an 18 ppb rise in 2021. This fit-to-data is up to August 2022 for the observed mixing ratio and $\delta^{13}\text{C}_{\text{CH}_4}$ measurements, and then extrapolated to 2037, for a full 30-year period of emissions growth. The scenario has an overall mean growth rate over 30 years of 10.5 ppb/yr. *Figure 12a. Change in global mean mixing ratio (blue) and $\delta^{13}\text{C}_{\text{CH}_4}$ (red) together with NOAA data to August 2022 and $\delta^{13}\text{C}_M$ data (green) to 2018. Compare with Fig. 10a.*

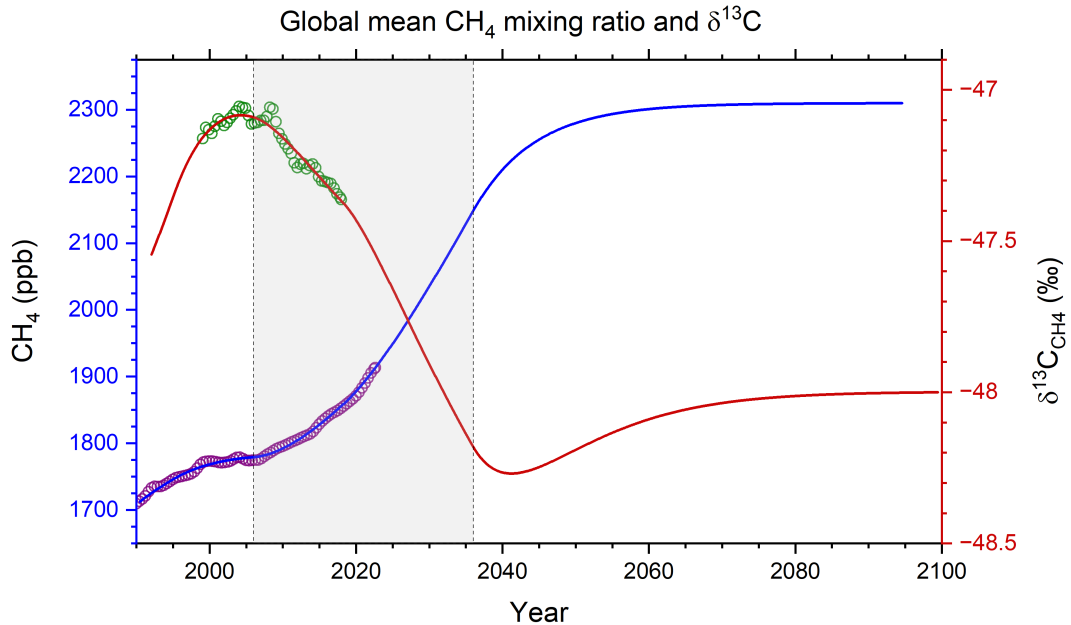
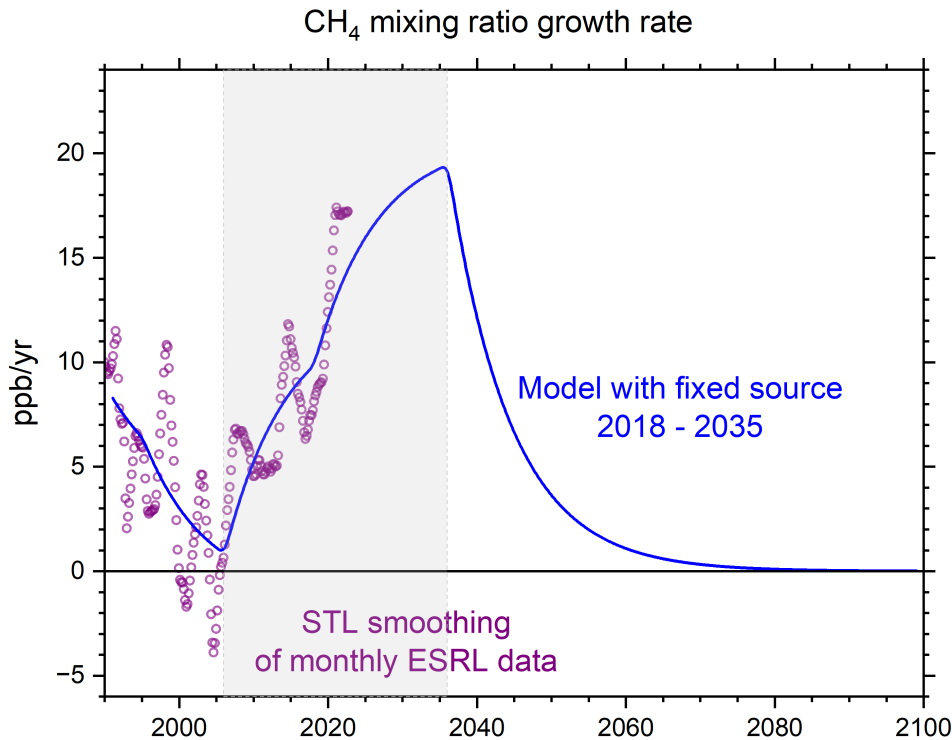


Figure 12b. Observed methane growth rate in ppb/yr from the ESRL data and from the model using a fixed source strength up to 2037, as shown in Fig 11. The measurement-based

1037 growth rate determination uses the Seasonal Trend Loess (STL) analysis of Cleveland et al
 1038 (1990) to separate a changing seasonal cycle from a changing trend. Compare with Fig. 10b.
 1039



1040 This analysis successfully tracks the observed inflexion and then decrease in atmospheric
 1041 $\delta^{13}\text{C}_{\text{CH}_4}$ (see Fig. 4 and green circles on red curve in Fig 12a), observed until 2022. In the
 1042 projection part of the model analysis, the global mean CH_4 mixing ratio reaches 2348 ppb
 1043 and stabilises around the year 2055. The budget analysis also follows the observed decrease
 1044 in $\delta^{13}\text{C}_\text{M}$, which reaches a minimum in the year 2040 followed by some recovery.
 1045 Stabilisation of $\delta^{13}\text{C}_\text{M}$ takes longer than for the mixing ratio because of the Tans effect, with
 1046 $\delta^{13}\text{C}_{\text{CH}_4}$ eventually reaching a minimum in 2040, followed by some limited recovery as the
 1047 burden equilibrates, while the global mean $\delta^{13}\text{C}_\text{s}$ of source emissions decreases from -
 1048 54.3‰ in 2006 to -55.2‰ in 2099.

1049 Our modelling suggests that the modern pulse of new biogenic methane, despite ongoing
 1050 anthropogenic fossil fuel inputs with relatively positive $\delta^{13}\text{C}_{\text{CH}_4}$, is capable of shifting the
 1051 $\delta^{13}\text{C}_{\text{CH}_4}$ of the much larger whole modern atmospheric burden by, say, -1.1‰ over the full
 1052 30 year period 2007-2037. That includes inputs of equatorial methane with little isotopic
 1053 leverage, outer seasonal tropical methane with some isotopic leverage, and boreal/Arctic
 1054 methane with strong isotopic leverage. In the real record of the contemporary modern
 1055 atmosphere, prior to the onset of growth in 2006, the mean atmospheric methane burden
 1056 was slightly over 4900 Tg. In 2022, the mean burden had risen to 5300 Tg, a net increase of
 1057 roughly 400 Tg during 2006-2022 inclusive. The accompanying $\delta^{13}\text{C}_{\text{CH}_4}$ shift of about -0.55‰
 1058 includes a 'positive' isotopic input from thermogenic sources such as anthropogenic fossil
 1059 fuel emissions and biomass burning with isotopic signatures more ^{13}C rich than the bulk
 1060 atmospheric source. Had there been no such positive leverage, the total biogenic-driven
 1061 $\delta^{13}\text{C}_{\text{CH}_4}$ shift over 2007-2022 that we would now be measuring would have been perhaps -

0.7‰. Note that this shift has occurred despite the new emissions being diluted into a global burden five times larger than in glacial periods. A comparable pulse into the glacial air would shift $\delta^{13}\text{C}_{\text{CH}_4}$ by 3 to 4‰ over a complete termination, including turn-on of boreal sources.

To conclude: If methane emissions, primarily from biogenic methane sources, continue to rise at a rate similar to the one observed in recent years, the modern rise since 2006 would be large enough to be identified as the first part of a 'termination-scale' event in the ice core record of atmospheric methane. The conclusion also holds if only 85% of the current growth is considered, given that 15% of the ongoing rise is likely from fossil emissions that may be mitigated in the near future. Even if livestock emissions growth as fast as 1.4 Tg/yr/yr (Zhang et al., 2022: see above) is deducted, the remaining 'non-fossil fuel, non-livestock' growth rate still bears comparison with abrupt growth in a termination-scale event, the more so because the productivity of tropical livestock depends on vegetation growth that in turn follows climate-driven rain and warmth.

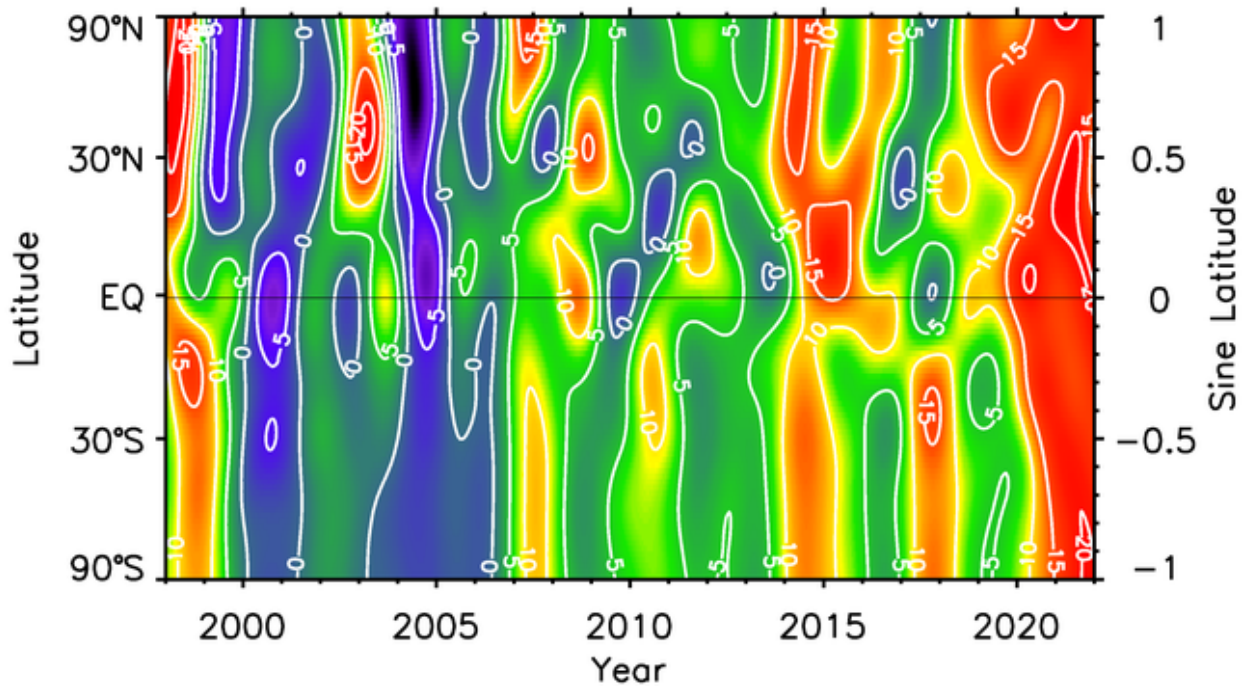
10. Discussion of Comparison

To summarize, methane's mole fraction growth in the past 15 years is comparable to growth in termination events, and it is likely that ongoing changes in methane emissions from present day wetlands are comparable to feedbacks during the past global change events. Even if only 85% of modern methane growth is biogenic, and part of that biogenic growth is due to anthropogenic inputs like increased cattle populations and more use of nitrogen fertilisers, methane's non-anthropogenic growth in the past 15 years has been at least comparable to growth in past climate change events. Allowing for the difference in the mass of the pre-transition burden, the associated isotopic shift is also comparable to shifts recorded (Fig. 8) across whole termination events (i.e. after the boreal wetlands had become fully active).

It remains unclear how much the biogenic changes to the methane budget in recent decades are within the natural range of responses to decadal or centennial-scale climate variability.

The current methane growth event started in late 2006 in both northern and southern high latitudes (Fig. 13). In Arctic summer, southerly winds blew high-methane air poleward from boreal zones. From then on, the tropics became the main zone of methane growth, especially in 2014/5, but with marked growth in the northern temperate zone in 2014 and in the southern outer tropics around in 2018 (e.g. Pantanal wetland). Then came the extraordinary ongoing growth surge of 2019-2022. The sine-latitude-time methane map in Figure 13 (updated from Lan et al. 2021b) reflects this history, though with the obvious caveat that changes in natural sources and sinks are added to by changes in human emissions from the fossil fuel industry, agriculture and waste.

Figure 13 –Methane growth rate averaged by latitudinal zone, from 2000 through 2021, updated from NOAA data. Red 'warm' colours – growth. Blue 'cool' colours – decline. The vertical axis denotes the sine of latitude, which weights each latitude band by its atmospheric mass. The ITCZ migrates seasonally from roughly 30°N to 30°S (sine latitude 0.5 to -0.5), while the Arctic zone is the band north of sine latitude 0.91. The current growth event began in late 2006. Note the remarkable growth since 2020.



Although the ongoing increase in global biogenic methane emission might be within the multi-century range of historic emissions variability (Fig. 3) (Ferretti et al. 2005, Mischler et al. 2009), the speed-of-change of the modern trend is extreme, much more rapid than methane's medieval changes or the post-1800 rise driven by fossil fuel use. The inference is that the biogenic component of methane's current (2007-2022) change is indeed likely outside the range of natural variability.

Methane's modern growth, in ppb/yr, and isotopic shift are more rapid than in glacial-interglacial terminations (Table 3), though not necessarily faster than the maximum decadal change rates during some terminations. Thus there is evident similarity between the past increases in tropical biogenic sources that drove the glacial-interglacial terminations and the present-day biogenic inputs driving the current 2006-2022 rise in methane emissions. Though the events are very different, in the 15 years of growth to date the modern rise does indeed bear comparison with the scale and speed of the abrupt phase of a termination event.

Previous orbitally driven natural transitions from cold glacial periods to warmer interglacial periods took place in the presence of large ice caps and widespread sea ice. In contrast, the modern climate change is from a warm Holocene (i.e. with widespread northern wetlands) to a new, presumably even warmer future. The comparison between modern growth and past analogues should not be taken too far. A modern 'termination-style' event marking the reorganisation from a Holocene warm climate to some new post-Holocene yet-warmer climate will be very different from any of the end-glacial events of the past half-million years.

Moreover, today's transition is anthropogenically driven, in an atmosphere where CO₂ is much higher and rapidly growing, and where crop wastes and fertilizer run-off are

widespread. Thus it is possible that the wetland response today has already surpassed the wetland response in past glacial interglacial terminations.

Yet it is indeed possible to imagine a new, modern step-change reorganisation of the planetary climate in a global 'termination-scale' event – this could include marked ocean warming and expansion of the ITCZ, and changes in the behaviour of the Indian Ocean Dipole as the Hadley circulation expands coupled with alteration in the strength and frequency of ENSO variations. Such changes could be accompanied by rapid Arctic land-surface warming and sustained ocean circulation responses, including changes in the Atlantic Meridional Overturning Circulation, on centennial-level timescales (Matsumoto, 2007).

Compared to the 'natural' CO₂ budget, which has a major component in the deciduous vegetation of the temperate northern hemisphere, changes in the biogenic methane signal reflect more global distribution of sources and sinks. In past palaeo-climate transitions, large changes occurred in the CO₂ budget, in response to changing latitude distribution of insolation or meteorology and warming oceans. But such feedback-related changes in CO₂ are hard to identify. In the present-day record there is not yet clear evidence for sustained changes in the CO₂ burden that directly relate to climate change feedbacks and which cannot be explained within the uncertainty of quantifying anthropogenic fossil fuel emissions and land use changes. In contrast, given methane's lifetime, it can serve as a 'tallying-together' of ongoing climate change impacts, a representative bellwether.

Methane is a good integrative signal of the state of global wetlands, and methane's global budget – its growth or decline when the burden in the air is not in equilibrium with the net flux from sources and sinks – sums the changing breath of the whole land biosphere. This includes not only changing outputs from biogenic sources (nearly all terrestrial or coastal, not oceanic) such as tropical and boreal wetlands, grass and tree growth for ruminants, and growth in biomass fire fuel loads, but also changes in the meteorologically sensitive oxidative capacity of the atmosphere involving [OH] Cl, and soil uptake. All natural sources and sinks are highly variable with climate fluctuation. Wetland and ruminant emissions vary with plant growth, rapidly changing as precipitation and temperature shift in ENSO and Indian Ocean Dipole cycles, and as CO₂ varies. Methane removal by [OH] (especially in the tropical mid-troposphere) and Cl fluctuates with the intensity of the Inter-Tropical Convergence and the trade winds in the ENSO cycles. Hydroxyl is also sensitive to atmospheric NO_x, that historically came from dry season fires, usually human-lit. Soil sinks in aerated soils, capable of breathing as pressure fluctuates under frontal passage are climate-dependent, as bacterial methanotrophy needs the right conditions, neither too dry nor too wet.

If the climate is constant, then the methane burden equilibrates and is steady. When the climate changes, natural methane sources and sinks respond and there is consequent disequilibrium in the methane budget (i.e. growth or decline of the burden). Sustained disequilibrium in the methane burden thus signals ongoing change in the biological productivity of the entire planet's land surface. With the isotopic spin-up taking several decades, a long-sustained growth or decline indicates a multi-decade change that is outside decadal variability.

The abruptness, power and global scale of past glacial/interglacial terminations provide palaeoclimatological standards for comparison with what is taking place today (Tzedakis et

al. 2009). The Eemian interglacial (130-115 kaBP, Marine Isotopic Stage 5, initiated by Termination II) was warmest around 125,000 years ago, with sea level perhaps 6 to 9 m above present-day level, implying significant melting both in Greenland and Antarctica compared to today (Dutton and Lambeck, 2012). The Eemian 'green Sahara' had abundant wetlands and great lakes (Larrasoana et al. 2013), very different from the current climate. However, the Eemian may be a poor guide to the future. Such past records may give little predictive help: the driving force today is anthropogenic, the speed of growth in CO₂ and CH₄ burdens is unprecedented, as is global land use change, so what is to come may be warmer than recent interglacials, even the Eemian.

The model growth curve in Figures 12 a,b assumes growth that may be below what will occur in the next decade if both natural and anthropogenic emissions rise strongly. However, assuming the emission growth rates in the model, the increased radiative forcing of the climate from this extra methane input from the years 2021-2036 can be estimated at roughly 0.17 W/m². To put this in context, that is about equivalent to five years of present-day growth in CO₂ forcing. There are important consequences for climate modellers. Using a methane-enabled Earth System Model, Kleinen et al. (2021) found methane mixing ratios and hence radiative forcing substantially higher than assumed in the scenarios used for the Coupled Model Intercomparison Project (CMIP6); assuming methane growth is only due to anthropogenic causes may lead to underestimation of future change.

The question remains open. The observed post-2006 CH₄ growth (Fig. 1) is comparable to growth during the abrupt phase of a termination. Top-down and bottom-up estimates of modern emission growth rates are comparable to or greater than growth in termination events. Modelling shows that the modern record is comparable to termination events both for the change in CH₄ abundance driven by natural sources and that there are parallels between the observed modern $\delta^{13}\text{C}_{\text{CH}_4}$ record and the isotopic records during full terminations.

Methane's unexpected rise means achieving the Paris Agreement's targets will be very difficult (Mikaloff-Fletcher and Schaefer 2019, Nisbet et al. 2020) and will likely demand accelerated switching from fossil fuels to renewable energy sources, in addition to rapid elimination of methane emissions from fossil fuel use. Reducing the methane burden to its pre-industrial size would have beneficial impacts not only on climate, but also on air quality, increasing the oxidising capacity of the atmosphere (Staniaszek et al. 2022). Shooting the methane messenger will not eliminate methane's message. But it would help.

A casual coincidence is not necessarily a causal connection. That we are in the midst of a rapid methane growth event, substantially driven by natural sources, does not necessarily mean the onset of global climate change. There is still the possibility that we are simply experiencing decadal- or centennial-scale variability. But past decadal- and centennial- scale Holocene variability, while significant, is smaller and much less abrupt than the typical abrupt climate change. Moreover, rapid growth in natural methane emissions does not happen unless climate is changing also: thus methane may indeed be the bellwether, the first indicator, the messenger of climate change.

There are limits to comparison. Applying the designation 'termination' to current changes is not strictly correct in the modern usage of Terminations IX to I. Yet Broecker and Denton (1990) originally recognised terminations in the wider sense as "massive and abrupt reorganizations of the ocean-atmosphere system". In this sense, such an event may indeed

be taking place. The remarkable shifts in the biogenic methane budget since the end of 2006 may indeed be a first indicator of a large-scale reorganisation of the climate system on a scale that matches past great changes. Any current or near-future warming transition will differ greatly from the past glacial-interglacial Terminations IX to I. There is no Roman number zero. But then, zero is different from all other numbers. The question is valid: have we already entered Termination Zero?

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Contributions

EJD, XL, SEM, DL, REF, JLF, TR, JS and EGN collected samples, made primary measurements, participated in development of the study and edited the manuscript. SEM performed $\delta^{13}\text{C}_{\text{CH}_4}$ measurements. MRM carried out the modelling in Section 9 and he and PP had input on meteorological and modelling discussion, especially in the tropics. XL and HDvdG estimated emissions growth from EDGAR data. JS and MND guided the input on ice core records and past terminations. Figures, including updates and modifications: 1 EJD,XL,(NOAA), EGN; 2 PIP; 3 JS, MRM; 4 SEM,MRM,GD, TB; 5 SEM,MRM,GD, TB; 6 XL, HDvdG; 7 JS; 8 EJD; 9, 10, 11 MRM; 12 JLF with input from TR and SEM; 13 EJD, XL; SI 1 SEM, EJD, SI 2,3,4 MRM, GB,TB SI 5 XL and HDvdG,. Table 1 EGN; Table 2 HDvdG; Table 3 EGN; Table 4 MRM. Conceptualisation and manuscript preparation mainly by EGN, MRM and EJD, with input from all authors. This paper and all methane papers depend on the impeccable quality of NOAA data (EJD, XL, SEM). Eric Wolff and Mark Lunt, and especially Vas Petrenko are thanked for their thoughtful, critical, and extremely helpful comments. We thank the most helpful anonymous reviewers and the editor for their kind and perceptive assistance over this very long and demanding study.

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Supplementary information

Figure SI 1. NOAA $\delta^{13}\text{C}_{\text{CH}_4}$ record 2007-2022 from Ascension Island (8°S in the equatorial zone). Note variability range of short-term weather changes and sustained negative shift. Ascension Island is the key monitoring station representing the southern hemisphere Atlantic tropical and trade wind zones.

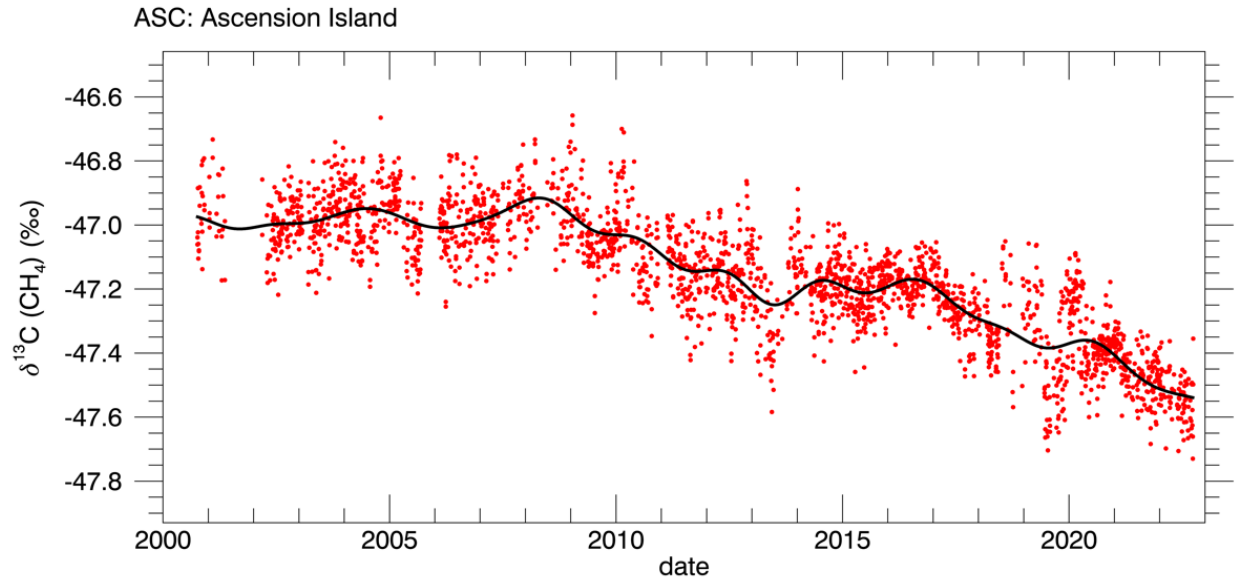


Figure SI 2. Polar record for CH_4 and $\delta^{13}\text{C}_{\text{CH}_4}$, Alert and Barrow. NOAA data. See Fig. 4 caption in main text.

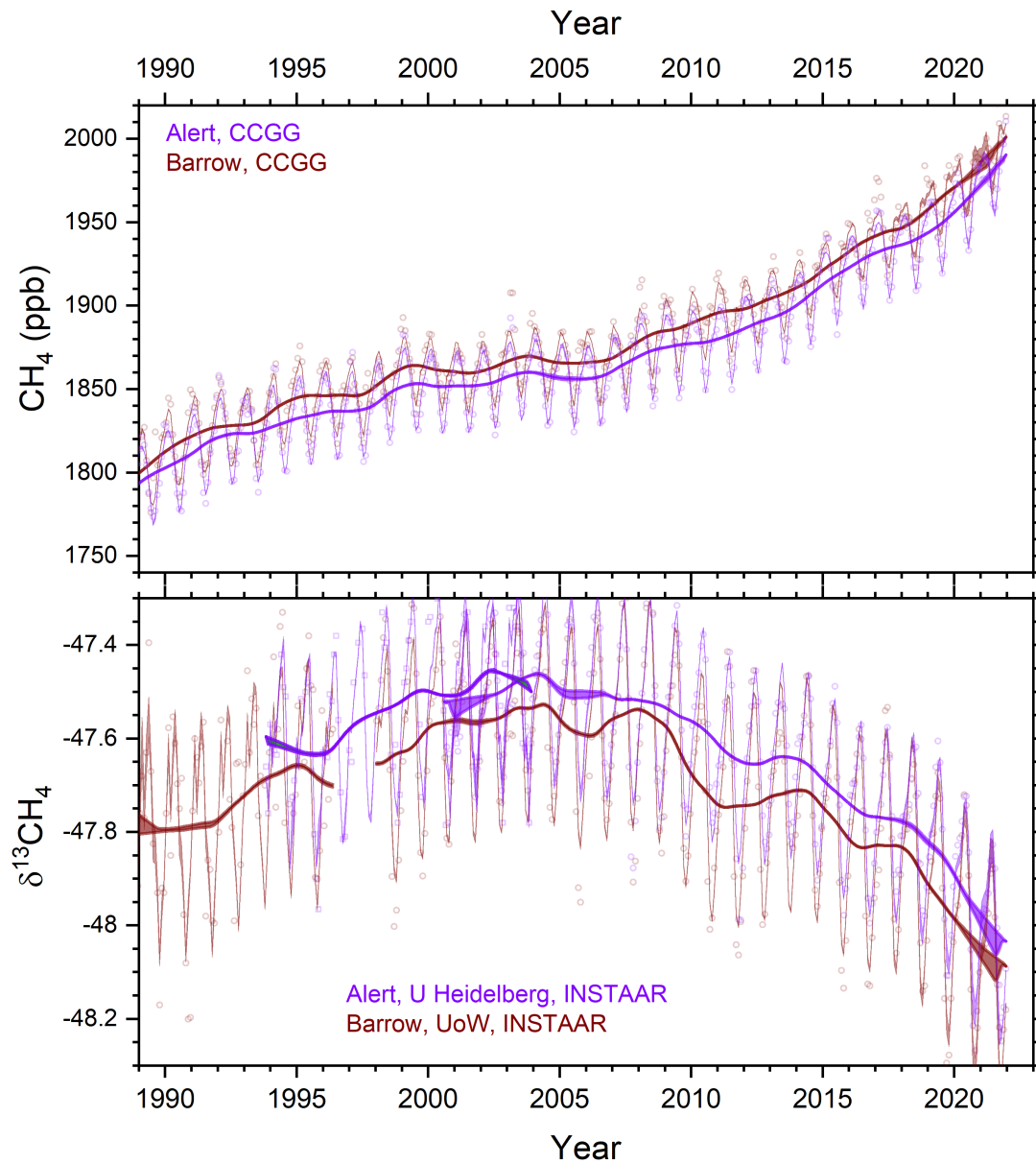
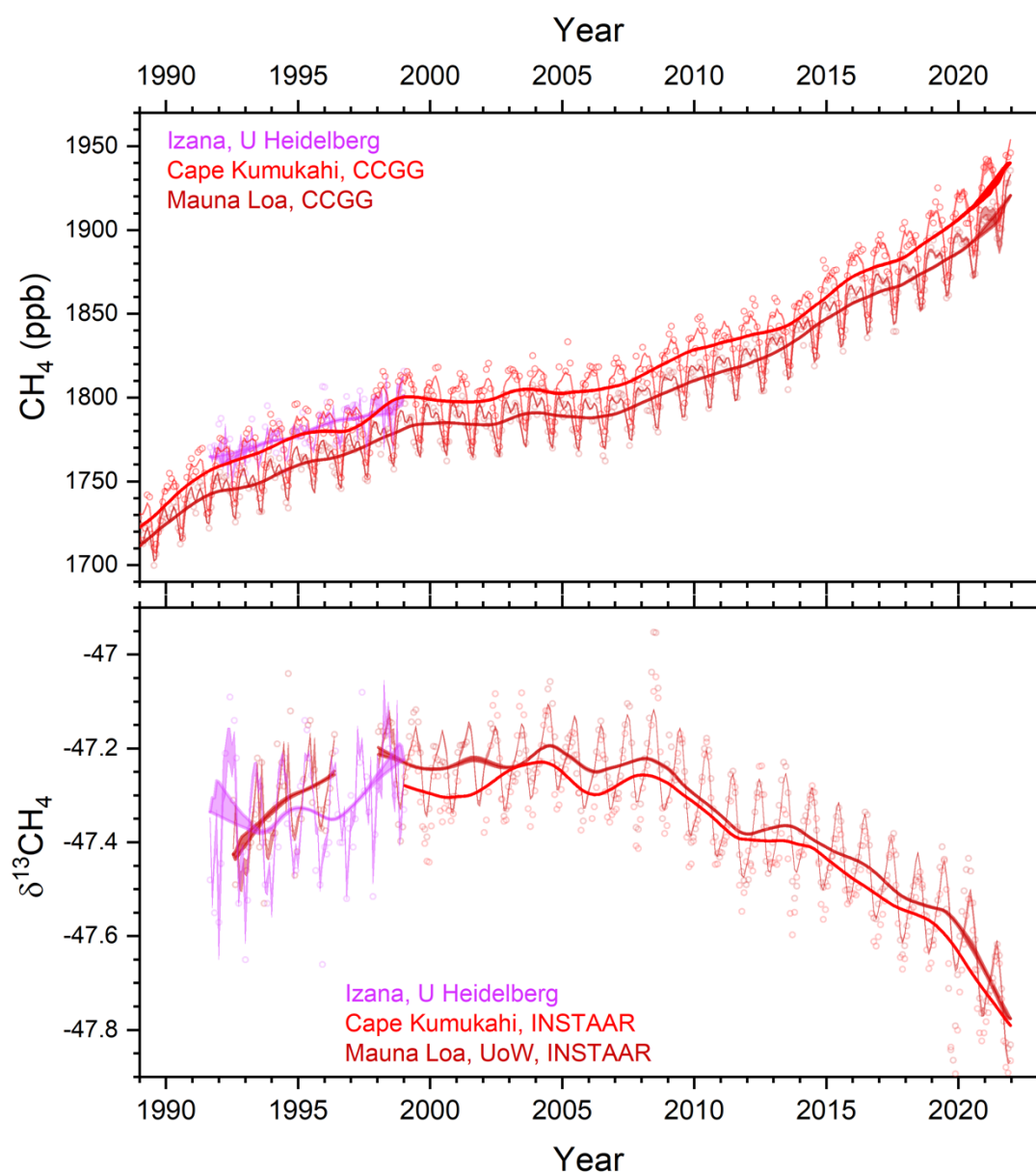
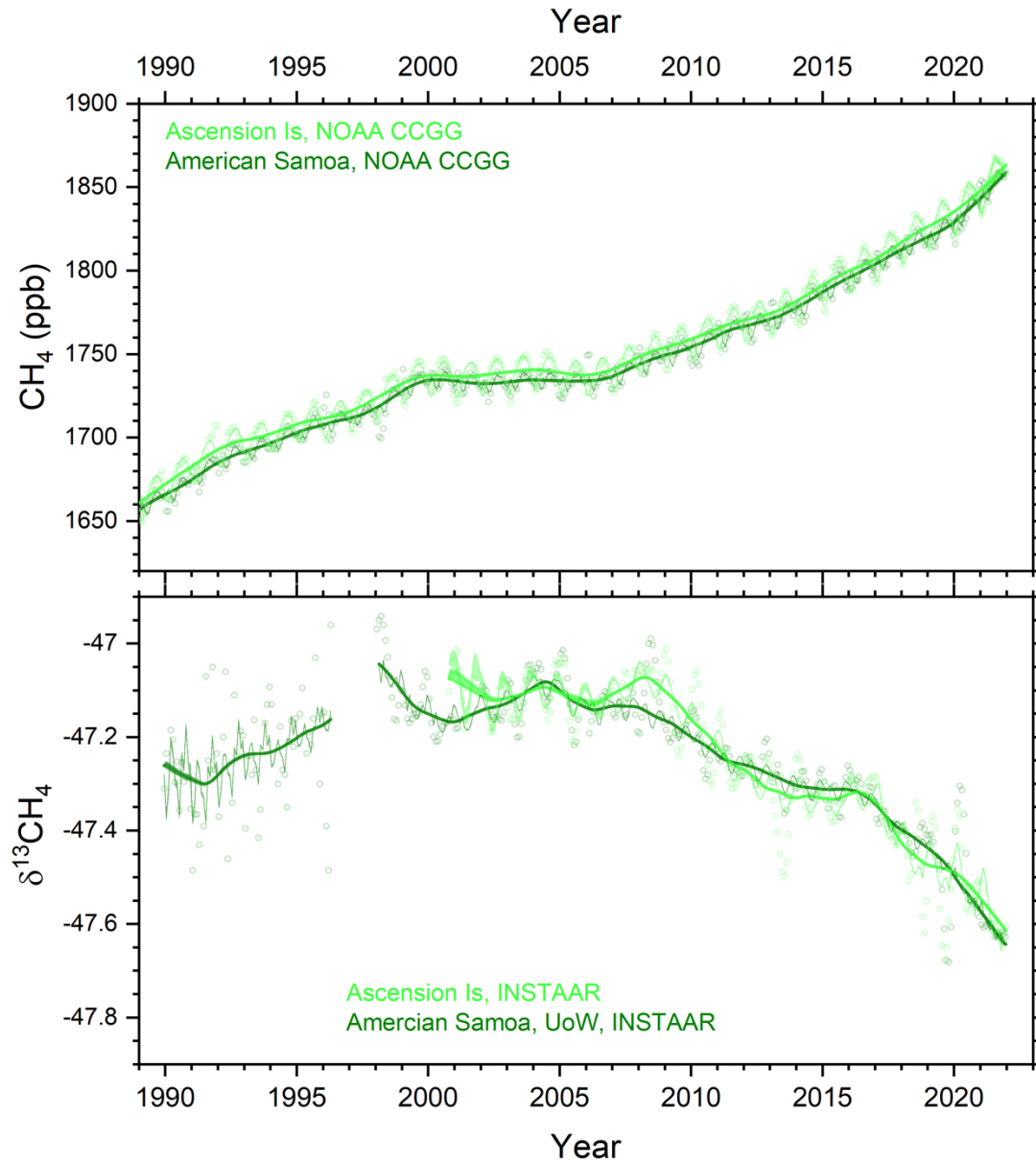


Figure SI 3 Northern mid-latitude record for CH_4 and $\delta^{13}\text{C}_{\text{CH}_4}$. Data from World Data Center for Greenhouse gases and NOAA (see Fig. 4 caption in main text).



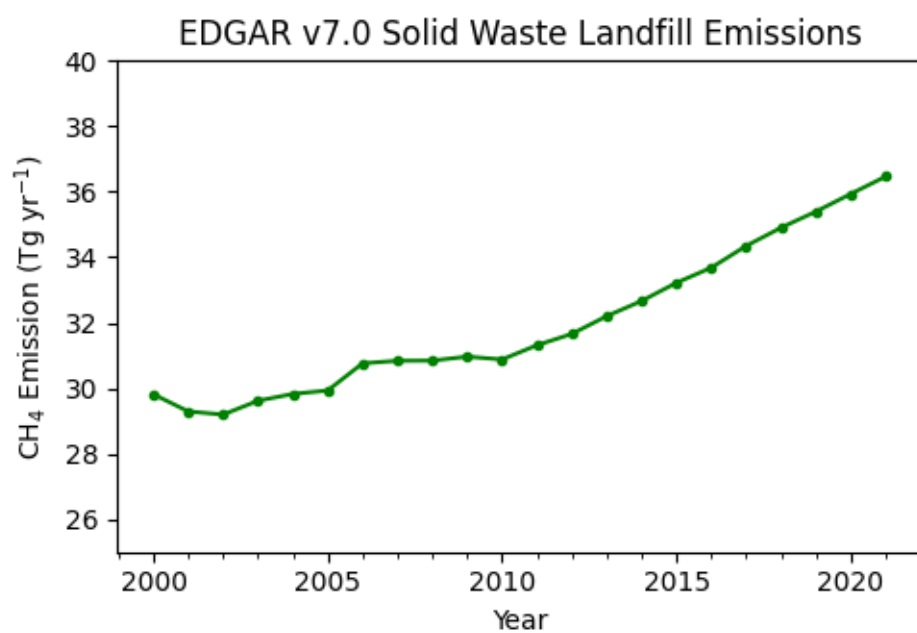
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1824 *Figure SI 4. Equatorial record for CH_4 and $\delta^{13}\text{C}_{\text{CH}_4}$, Ascension and American Samoa. NOAA*
 1825 *data. See Fig. 4 caption in main text.*

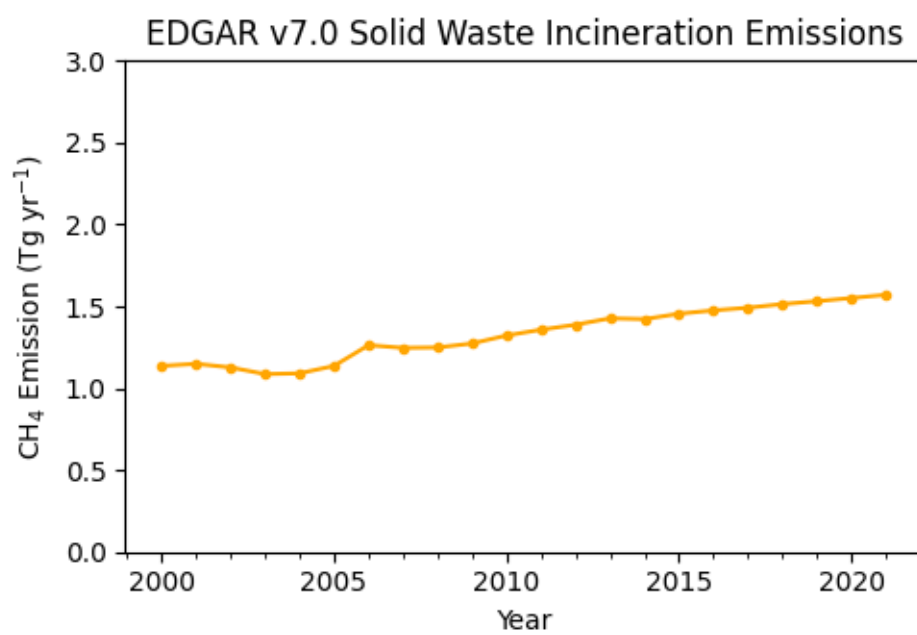


1826 *SI 5 EDGAR v. 7.0 (Emissions Database for Global Atmospheric Research) estimates of*
 1827 *growth in emissions from a) solid waste and landfill; b) solid waste incineration; c) Manure*
 1828 *management; d) Waste water; e) Ricefields.*
 1829 *From EDGAR (2023). https://edgar.jrc.ec.europa.eu/dataset_ghg70*

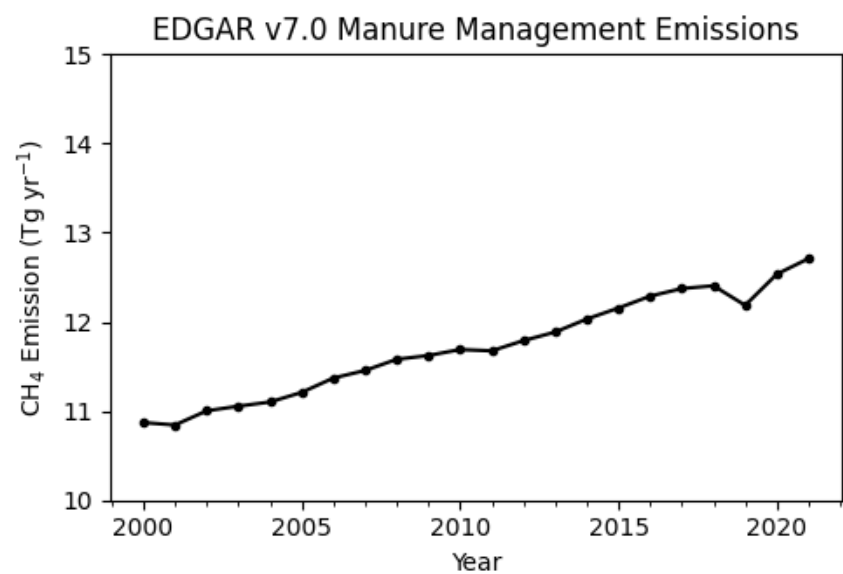
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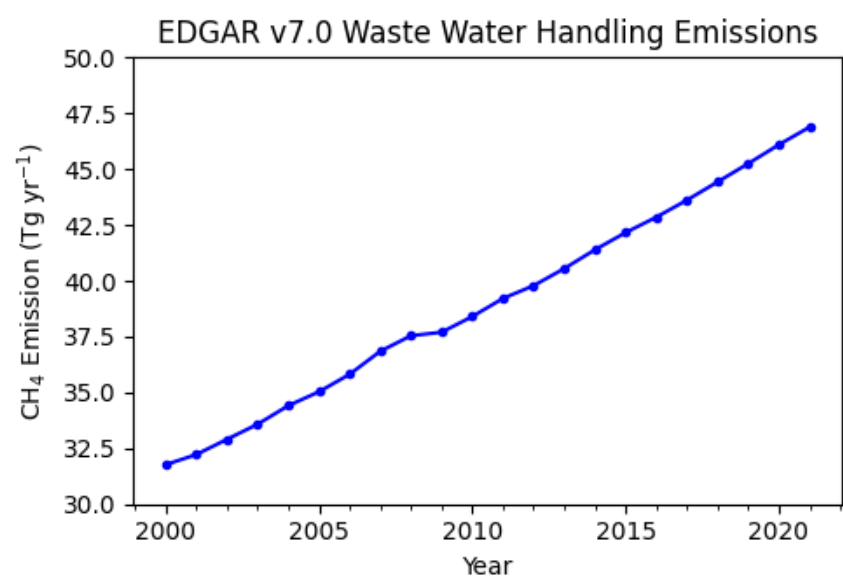
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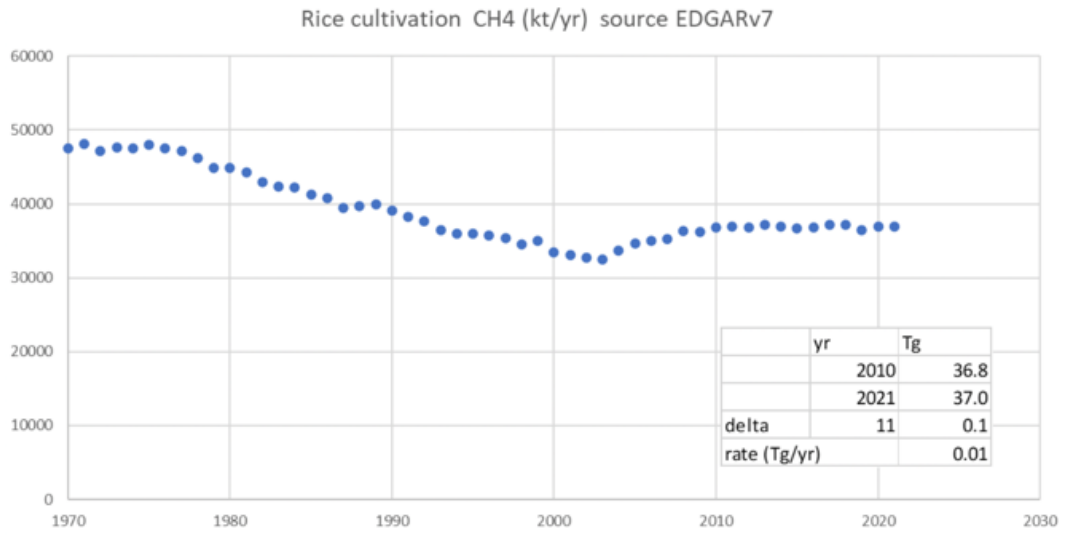


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1836 *Fig. SI.5e Methane from rice cultivation: minimal recent (2010-2021) growth rate.*



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Fig SI 6. CH₄ mole fraction values in ppb from the 4-box running budget model for sources that fit measured values to 2017 and then give 10 ppb/yr growth until 2035. Orange is 0-30°N. Black is the global average and overlays the 30-90°N case. Blue is 30-90°S and green 0-30°S. Jump in emissions takes place in 2007; equilibration is mostly complete by 2045. Note that CH₄ mixing ratio is higher in the 0-30°N semihemisphere before and after this transition.

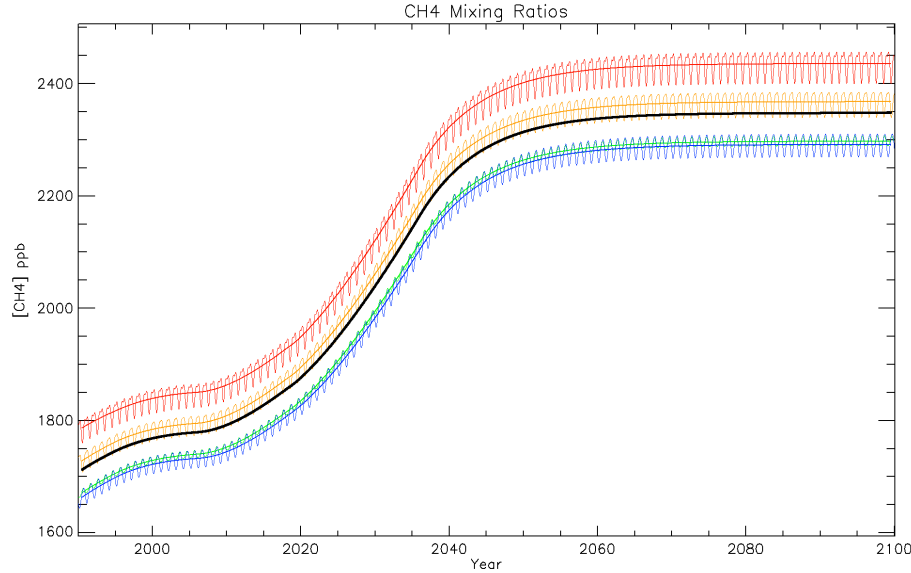
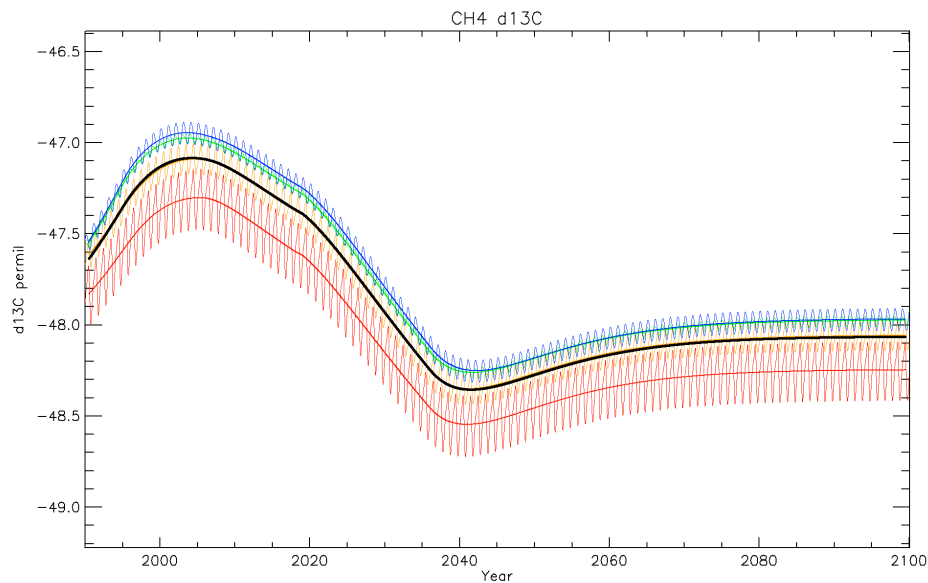


Fig SI 7. $\delta^{13}C_{CH_4}$ (‰) in the 4 semi-hemispheres for the inputs and colours shown in Fig SI2.



Main Text Figures 4 and 5

Changes in CH₄ mole fraction and $\delta^{13}C$ have been taken from ten sites and data from the World Data Center for Greenhouse Gases (<https://gaw.kishou.go.jp/>), NOAA, INSTAAR, NIWA, University of Washington and University of Heidelberg (IZO from Levin et al., (2012). Changing trends in the data have used the Cleveland *et al* STL (Seasonal Trend Loess) method with monthly data for each site. This approach has been extended to deal with missing monthly data values by optimising these to produce a smooth STL fit. Periods where the number of months is not a multiple of 12 are dealt with by merging a series of STL fits

that cover sub-periods that are a multiple of 12 months and retaining the full range of these partial fits across common months. The result is a spread in trends at the begin and end of the data series and around regions where there are significant groups of missing data. The same colour code is used in the following plots as in Figure 4 of the main text. The $\delta^{13}\text{C}$ values shown have used the recent intercomparison of $\delta^{13}\text{C}_{\text{CH}_4}$ (Umezawa et al, 2018) to adjust reported values and make them all consistent with the reference level used in that paper. Notably, the degree of agreement between different organisations that is shown here demonstrates the great and continuing value of that painstaking study by Umezawa *et al.* (2018).