Anthropogenic emission is the main contributor to the rise of atmospheric methane during 1993–2017

Zhen Zhang (张臻)1, *, Benjamin Poulter2, Sara Knox3, Ann Stavert4, Gavin McNico5, Etienne Fluet-Chouinard6, Aryeh Feinberg7, Yuanhong Zhao (赵园红)8, Philippe Bousquet9, Josep G. Canadell4, Anita Ganesan10, Gustaf Hugelius11, George Hurtt1, Robert B. Jackson6,12, Prabir K. Patra13, Marielle Saunois9, Lena Höglund-Isaksson14, Chunlin Huang (黄春林)15, Abhishek Chatterjee16,17 and Xin Li (李新)18

1Department of Geographical Sciences, University of Maryland, College Park, MD 20742, USA; 2Biospheric Sciences Laboratory, NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA; 3Department of Geography, University of British Columbia, Vancouver V6T 2W5, Canada; 4Global Carbon Project, CSIRO Oceans and Atmosphere, Canberra, ACT 2601, Australia; 5Department of Earth and Environmental Sciences, University of Illinois Chicago, Chicago, IL 60607, USA; 6Department of Earth System Science, Stanford University, Stanford, CA 94305, USA; 7Institute for Data, Systems and Society, Massachusetts Institute of Technology, Cambridge, MA 02139, USA; (Continued on next page)

*Corresponding author. E-mail: yuisheng@gmail.com

ABSTRACT

Atmospheric methane (CH4) concentrations have shown a puzzling resumption in growth since 2007 following a period of stabilization from 2000 to 2006. Multiple hypotheses have been proposed to explain the temporal variations in CH4 growth, and attribute the rise of atmospheric CH4 either to increases in emissions from fossil fuel activities, agriculture and natural wetlands, or to a decrease in the atmospheric chemical sink. Here, we use a comprehensive ensemble of CH4 source estimates and isotopic δ13C-CH4 source signature data to show that the resumption of CH4 growth is most likely due to increased anthropogenic emissions. Our emission scenarios that have the fewest biases with respect to isotopic composition suggest that the agriculture, landfill and waste sectors were responsible for 53 ± 13% of the renewed growth over the period 2007–2017 compared to 2000–2006; industrial fossil fuel sources explained an additional 34 ± 24%, and wetland sources contributed the least at 13 ± 9%. The hypothesis that a large increase in emissions from natural wetlands drove the decrease in atmospheric δ13C-CH4 values cannot be reconciled with current process-based wetland CH4 models. This finding suggests the need for increased wetland measurements to better understand the contemporary and future role of wetlands in the rise of atmospheric methane and climate feedback. Our findings highlight the predominant role of anthropogenic activities in driving the growth of atmospheric CH4 concentrations.

Keywords: greenhouse gas, carbon cycle, climate mitigation, wetland, methane isotope

INTRODUCTION

Stabilizing atmospheric methane (CH4) emissions from anthropogenic activities is a critical component of climate change mitigation [1]. The atmospheric CH4 concentration has increased ~2.5-fold from 731 ppb (parts per billion) in 1750 (pre-industrial reference year [2]) to 1890 ppb in 2020 [3]. Meanwhile, over the past century, atmospheric δ13C-CH4 values increased from ~−49.0‰ in 1912 to −47.2‰ in 2007 due to increasing emissions of isotopically 13C-enriched (i.e. isotopically heavy) fossil fuels [4]. Despite the importance of understanding the temporal changes in atmospheric CH4, the drivers of changes in the growth rate of atmospheric CH4 over recent decades remain poorly understood [5]. The increase in atmospheric CH4 slowed in the early 1990s and was followed by a so-called stabilization period during 2000–2006 [6]. Since 2007, global atmospheric CH4 concentrations have begun to rise again, accompanied by a decline in δ13C-CH4 values from −47.2‰ in 2007 to −47.4‰ in 2017 [3]. The cause of this change has been studied recently using atmospheric inversion models [7–10], atmospheric box models [11–15] and...
Sources of the global CH₄ budget are mainly determined by three broadly defined groups: (i) thermogenic sources from industrial fossil fuel (e.g. coal, oil and natural gas; IFFCH₄) and geological sources (GEOCH₄); (ii) biogenic sources from livestock, rice agriculture, landfills and waste (AGWCH₄), and natural wetlands (WETCH₄); and (iii) pyrogenic sources from wildfires and biomass burning (BBCH₄). The primary sink for CH₄ is reactions with tropospheric OH, soil microbial uptake and a natural wetlands sinks, and reactions with chlorine (Cl), stratospheric OH [5]. OH plays a significant role in driving the observed atmospheric trend; it difficult to estimate due to its complicated chemistry, i.e. non-linear chemical feedback and short lifetime [30,31]. For example, estimates of interannual variability (IAV) in global mean OH are significantly higher in the empirical box-model estimates that use CO and methylchloroform (MCF) constraints [32] than estimates based on chemical transport models (Fig. S1).

A thorough investigation of these hypotheses in a clearly defined framework is essential to help resolve the unexplained change in the growth rate of atmospheric CH₄. Here, we use an isotopic mass balance approach to attribute drivers of the growth rate of atmospheric CH₄ using a large ensemble of scenarios to represent different combinations of emission hypotheses (denoted emission scenarios, see Materials and Methods) from a comprehensive set of updated bottom-up estimates representing anthropogenic emission inventories and spatially explicit signatures for major CH₄ sources. Each emission scenario is composed of a time series of sectoral CH₄ fluxes and their hemispheric emission-weighted δ¹³C-CH₄ values. A globally representative database and spatially resolved distributions of δ¹³C-CH₄ values for the major CH₄ sources [36–38] were used to evaluate the temporal and regional variability in observed δ¹³C-CH₄ values. Monte Carlo techniques were applied to explore the uncertainty in δ¹³C-CH₄ estimates with full consideration of the spatial heterogeneity in CH₄ sources and their δ¹³C-CH₄ signatures. Scenario-specific parameters for the time series of the CH₄ removal rate driven by OH variations and run-specific CH₄ fractionation factors were derived by inverting an atmospheric two-box model (see Methods and Supplementary Data). We then evaluated the emission scenarios against observed δ¹³C-CH₄ values for 1993–2017 by running the two-box model in the forward mode. To test the hypothesis of a large increase from wetland CH₄ emissions, the idealized wetland scenarios (i.e. without process-based constraint) were then calculated to reproduce the temporal pattern of δ¹³C-CH₄. The comparison against atmospheric isotopic observations allowed us to select the most likely set of emission scenarios, which are defined as the first percentile cut-off of the lowest mean squared difference (MSD) in simulated δ¹³C-CH₄ values.
RESULTS AND DISCUSSION

Temporal variations in the atmospheric CH$_4$ concentration and its $\delta^{13}$C-CH$_4$ value

The ensemble simulations for the 96 emission scenarios (4 IFF$_{CH4} \times 3$ AGW$_{CH4} \times 2$ WETCH$_{CH4} \times 2$ BB$_{CH4} \times 2$ GEO$_{CH4}$) reproduced the observed atmospheric CH$_4$ concentration (Fig. 1A) using the corresponding optimized time series of OH derived from running the box model in inverse mode (Fig. S1). We accounted for the uncertainty in source signatures of $\delta^{13}$C-CH$_4$ by resampling 1000 sets of $\delta^{13}$C-CH$_4$ signature time series for each emission scenario ($n = 96 \times 1000$), which resulted in a wide range of modeled atmospheric $\delta^{13}$C-CH$_4$ values with some closely reproducing the observations. However, most emission scenarios tended to generate more enriched $\delta^{13}$C-CH$_4$ trends for 1993–2017, suggesting that existing bottom-up inventories overestimate the increase in IFF$_{CH4}$ during the study period, especially during slowdown and stagnation periods (Fig. 1B). Furthermore, to balance the rise in CH$_4$ sources, the increases in OH levels also led to positive trends in atmospheric $\delta^{13}$C-CH$_4$ values during these two periods (Fig. S1). Note that the large increase in coal-related emissions since 2003 has increased the $\delta^{13}$C-CH$_4$ values of the sources, which contribute to the divergence between the model and observations. The timing of this divergence is consistent with a rapid increase in methane emissions from China (mainly coal emissions) as reported by the inventories [23] and inversions [39]. For most of the emission scenarios, the estimated temporal variations in
Figure 2. Performance of emission scenarios in simulating atmospheric δ^{13}C-CH₄. (A) Taylor diagram illustrating the similarity between individual time series from the 96,000 simulations of different emission scenarios (colored dots) to the observed atmospheric δ^{13}C-CH₄ for 1993–2017. The black solid dot refers to the observed global average δ^{13}C-CH₄ for 1993–2017. Each dot symbol indicates the correlation value (angle), the standard deviation (SD, radial distance to the origin point) and the root mean square error (RMSE, distance to the black solid dot), with different colors representing the mean global average δ^{13}C-CH₄ value of the source. (B) Histograms of RMSE between simulated δ^{13}C-CH₄ values and observations grouped by bottom-up estimates with different colors for major CH₄ source categories. The solid lines represent the fitted density distribution after spline smoothing.

OH for 1993–2017 fall within the 1-σ range of the Bayesian inversion from Ref. [14] and are in good agreement with Ref. [35] for the post-2000 period. The EDGARv4.2-based emission scenarios have the largest mismatch with the inversion-based OH anomaly (relative to a global mean concentration of 1e⁶ molecules (molec)/cm³), confirming the known higher bias in EDGARv4.2 than in other inventories (Fig. S2).

The time series of CH₄ sources for 1993–2017 (Fig. 1C) suggests that decadal-scale variations in atmospheric CH₄ are dominated by anthropogenic emissions from both agricultural and fossil fuel activities. However, there is high uncertainty across IFFCH₄ inventories, with a sizable (>40 Tg CH₄ yr⁻¹) difference in magnitude and a large difference in temporal trends between inventories (i.e. EDGARv4.2, EDGARv4.3.2 and GAINS) and an atmospheric-observation-constrained approach (i.e. SCH150, which hypothesizes that IFFCH₄ is underestimated but does not increase over time). The temporal variation in AGWCH₄ exhibits a lower discrepancy than IFFCH₄ in the inventories, whereas WETCH₄ and BBCH₄ are more constrained. The IAV and magnitude of our estimates for WETCH₄, calculated using the process-based model LPJ-wsl, are comparable to the ensemble mean of multiple wetland models [40] and a global wetland CH₄ emission model ensemble for use in atmospheric chemical transport models (WetCHARTs) [41]. The wetland CH₄ estimates derived from driving the wetland model with a ground-measurement-based meteorological dataset from the Climate Research Unit (CRU) yield a small increase (<1 Tg CH₄ yr⁻¹), whereas the same model with climate reanalysis (REN) has an ~7.3 Tg CH₄ step increase from tropical wetlands between 2000–2006 and 2007–2017 [22].

Evaluations of proposed CH₄ hypotheses using emission scenarios

Figure 2A shows the distribution of residual bias in the individual box model simulations in terms of how they reproduce the observed δ^{13}C-CH₄ values. In the Taylor diagram [42] the global average δ^{13}C-CH₄ values of the sources before fractionation by chemical sinks range from −53‰ to −55‰ over 1993–2017, and a correlation coefficient lower than 0.6 is obtained for all of the simulations for 1993–2017. The low agreement suggests that the biases in the inventories and the wetland models contribute to the discrepancies in reproducing the δ^{13}C-CH₄, which is likely due to the overestimated increase in inventories, especially the coal emission that has a relatively heavy isotopic signature, as found by previous atmospheric inversion studies [16]. In addition, some of the simulations can reproduce similar IAV in atmospheric δ^{13}C-CH₄ values with root mean
square errors (RMSEs) from 0.05% to 0.5%, but \(\sim 85\%\) of simulations tend to produce higher IAV than observed. Although the global average \(\delta^{13}C\)-CH\(_4\) value was regarded as observational ‘truth’, this reference has an uncertainty of 0.04%, attributed to variability in measurements across all the stations and uncertainty from scale conversions between networks [43].

The density distributions of RMSEs grouped by different bottom-up CH\(_4\) estimates show the divergent performance of emission scenarios in reproducing observed atmospheric \(\delta^{13}C\)-CH\(_4\) values (Fig. 2B). Among the four IFF\(_{CH4}\) inventories, 80% of the simulations using EDGARv4.2 generated more positive trends of \(\delta^{13}C\)-CH\(_4\) values than the other three inventories, with no EDGARv4.2-based runs located in the most likely set of emission scenarios (Fig. S3). This result corroborates previous studies that also suggest that EDGARv4.2 tends to overestimate fossil fuel growth [44]. The more recent EDGARv4.3.2 has been improved, with 95% of the simulations located within the RMSE range from 0.1% to 0.3%, mainly due to improved emission factors and revised statistics of CH\(_4\) sectors [23]. SCH150 produces lower agreement than EDGARv4.3.2 partly due to the low IAV of SCH150, as SCH150 focuses on the long-term trends in IFF\(_{CH4}\). The GAINS inventories generated better performance: 78% of the simulations in the first percentile of MSD are based on GAINS IFF\(_{CH4}\). Note that this does not rule out the IFF\(_{CH4}\) scenarios that have flat or insignificant trends (e.g. SCH150), as \(^{13}C\)-enriched BB\(_{CH4}\) estimates in this study show declining trends over recent years, which would allow for compensation by increasing emissions from IFF\(_{CH4}\) to meet the decreasing atmospheric \(\delta^{13}C\)-CH\(_4\) values. Generally, IFF\(_{CH4}\) has a more pronounced impact in determining the past trends in \(\delta^{13}C\)-CH\(_4\) changes than the other major CH\(_4\) sources.

The contribution of combined agriculture, landfills and waste included in AGW\(_{CH4}\), which together represent 50%–62% of all anthropogenic sources, again reveals a higher bias of EDGARv4.2-based simulations compared to the other two inventories (Fig. 2B). Agricultural emissions dominate AGW\(_{CH4}\), with an average contribution of 77.5% to the total AGW\(_{CH4}\) over the study period. The lower MSD scores using EDGARv4.3.2 and WOLF2017 emission scenarios show improved reconciliation of estimates for the AGW\(_{CH4}\) source relative to the isotopic budget. This result supports the hypothesis that the global livestock estimates based on the 2006 IPCC Tier 1 guidelines underestimate livestock CH\(_4\) emissions at the national or state level [45], which is potentially attributable to outdated information used to develop the emission factors. However, there is no clear signal to distinguish whether EDGARv4.3.2 or WOLF2017 has a lower a priori bias, suggesting the need for further regional and global assessments by spatially explicit 4-D atmospheric models.

Our calculations suggest that, in contrast to anthropogenic sources, wetland CH\(_4\) emissions play a limited role in reproducing the decadal trend in atmospheric \(\delta^{13}C\)-CH\(_4\) (Figs 1 and 2B). Both REN and CRU demonstrate that wetland CH\(_4\) emissions appear to have contributed little to the renewed growth in atmospheric CH\(_4\). However, wetland emissions help explain the IAV in the atmospheric CH\(_4\) growth rate via its pulsed responses to climate dynamics, such as the El Niño–Southern Oscillation [46]. The latitudinal gradient of the growth rate for CH\(_4\) sources (Fig. S4) suggests that WET\(_{CH4}\) in the tropics has an important impact on the IAV of the CH\(_4\) growth rate, albeit the current limited understanding of WET\(_{CH4}\) is due to a significant deficiency in WET\(_{CH4}\) measurements in the tropics, especially for Africa [21].

The density distribution of RMSE grouped by BB\(_{CH4}\) and GEO\(_{CH4}\) (Fig. 2B) suggests that the recent hypotheses regarding a larger decrease [13] in BB\(_{CH4}\) and overestimated contemporary GEO\(_{CH4}\) [27,28] have a good agreement with the isotopic budget. The lower RMSE of Worden (2017)-based scenarios supports the hypothesis of a decreasing trend in BB\(_{CH4}\) during the post-2007 period, as suggested by inversion modeling based on satellite measurements of carbon monoxide [47]. The low GEO\(_{CH4}\) scenarios, which assume a geological source of 15 Tg CH\(_4\) yr\(^{-1}\) with upward-revised IFF\(_{CH4}\) (see Methods and Supplementary Data), yield lower RMSEs than the conventional high-GEO\(_{CH4}\) scenarios in which GEO\(_{CH4}\) was set to 52 Tg CH\(_4\) yr\(^{-1}\). These findings support the hypothesis that the current bottom-up estimates of anthropogenic fossil fuel CH\(_4\) emissions are underestimated and that geological emissions are overestimated.

Changes in the trends of \(\delta^{13}C\)-CH\(_4\) source signatures

A change in source signature (Fig. 3A) suggests varying global-emission-weighted average sources driven by the change in spatiotemporal distribution of CH\(_4\) source estimates for the four major CH\(_4\) categories. When considering spatial heterogeneity in the source signature, the globally representative \(\delta^{13}C\)-CH\(_4\) values tend to suggest a larger variation than previous assumptions that use globally uniform values [7,13]. The IFF\(_{CH4}\)
Figure 3. Changes in $\delta^{13}$C-CH$_4$ values and in contribution of $^{13}$CH$_4$ mass to the annual total of $^{13}$CH$_4$ mass for major source categories during 1993–2017. (A) Density function of the mean emission-weighted $\delta^{13}$C value of the sources for four time periods from Monte Carlo accounting ($n = 96,000$): decreased atmospheric growth during 1993–1999, relative stabilization during 2000–2006, renewed growth during 2007–2012 and accelerated growth during 2013–2017. While the emission-weighted $\delta^{13}$C-CH$_4$ signatures may change over time, it is the combination of these signatures with their respective emission amounts that determines the atmospheric isotopic trend. (B) Density distribution of changes in the contribution of $^{13}$CH$_4$ mass from major CH$_4$ source categories to annual global $^{13}$CH$_4$ mass. The contribution of $^{13}$CH$_4$ mass is calculated as the average percentage change of the ratio of average annual total $^{13}$CH$_4$ mass in the source during 1993–1999, 2007–2012 and 2013–2017 relative to the CH$_4$ plateau period 2000–2006.

The change in the $\delta^{13}$C-CH$_4$ contribution from individual sources does not necessarily imply the same trend in the global average signature. Theoretically, even if the tropical wetland signature becomes more positive, the increased proportion of wetland-contributed $^{13}$CH$_4$ mass to the total $^{13}$CH$_4$ mass can still result in a shift towards a more negative global signal, as the biogenic signature is considerably lighter than the global atmospheric $\delta^{13}$C-CH$_4$ value ($\sim −53.6 \permil$) before fractionation. This is the case in some paleoclimate studies [52] where tropical wetlands and other natural sources (e.g. biomass burning) dominated the annual CH$_4$ budget. However, the role of human activities has
Figure 4. Idealized wetland emissions that reproduce the observations of atmospheric CH₄ and δ¹³C-CH₄ for 2000–2017. (A) Time series of anomalies of idealized WETCH₄ with a 7-year moving window (colored lines: ensemble mean grouped by IFFCH₄) in comparison to the two emission scenarios (CRU and REN) applied in this study and the estimates from Ref. [53] (denoted Zhang2017) and WetCHARTs [41]. All estimates are anomalies relative to 2001. The min/max range of idealized wetland emissions is shown as the gray area. (B) Trend of wetland emissions for 2000–2017 computed using linear regression. Significant trends at the 95% confidence level are denoted by ‘*’. 

Idealized wetland emission scenarios that reproduce the decrease in atmospheric δ¹³C-CH₄ values

Beyond our wetland-model ensemble, we created scenarios to investigate the possible involvement of rising WETCH₄ in the decrease of atmospheric δ¹³C-CH₄ values. To do so, we performed a sensitivity test by running the box model in inverse mode for each individual run to calculate idealized WETCH₄ given the other sources, varying OH concentration, atmospheric CH₄ and δ¹³C-CH₄ observations, and the isotopic signatures of the sources, which thus linearizes the problem. The results suggest that the magnitude of increase in idealized WETCH₄ largely depends on the hypothesis of IFF sources, where greater wetland increases are required to compensate for the large increase in the IFFCH₄ scenarios (Fig. 4A). Note that all the idealized WETCH₄ scenarios are higher than the two WETCH₄ scenarios in this study (i.e. CRU and REN) or WetCHARTs, a wetland CH₄ product that is based on satellite-derived surface water extent and precipitation reconstruction and an ensemble of ecosystem respiration estimates. One process-based WETCH₄ that overlaps the increases in idealized wetland scenarios is the ensemble mean of LPJ-wsl simulations for a future projection under the climate scenario RCP8.5 [53] (denoted Zhang2017). RCP8.5 is considered the upper bound of wetland CH₄ feedback to rising temperature in LPJ-wsl because the strong and steady increase in temperature in RCP8.5 is higher than that determined from actual observations. Note that this scenario would occur only in combination with the hypothesis that IFFCH₄ has had no significant trends in recent years.

The comparison of 2000–2017 trends in WETCH₄ (Fig. 4B) suggests that to reproduce the magnitude of the observed decrease in atmospheric δ¹³C-CH₄ values, the required emission increase from natural wetlands would need to be much higher than the current estimates from process-based wetland models. The trend of CRU is consistent with the ensemble estimate of global wetland model simulations [5,40], while that of REN is at the higher end of the trends that consider the potential inundation increase due to enhanced tropical precipitation [22]. Note that the range of idealized increases in WETCH₄ is in line with two recent inversion studies [8,10] based on GOSAT CH₄ measurements, which suggests a positive wetland trend of 2–3 Tg CH₄ yr⁻¹ yr⁻¹ for 2010–2018. However, to produce such a significant trend, the Q10 parameter (temperature sensitivity of CH₄ emissions) in the wetland models would need to be much higher than the range of 2–3 from LPJ-wsl and WetCHARTs or the measurement-based average of 2.57 from FLUXNET-CH₄ [54]. In addition, a recent multi-model ensemble inversion study [55] suggests that the observation-constrained wetland CH₄ feedback to rising temperature is lower than that of Zhang 2017. Despite this, there are considerable uncertainties in modeled WETCH₄ due to scarcity of measurements for the tropics [40]. We conclude that the hypothesis that a large increase in natural wetlands drives the decrease in atmospheric δ¹³C-CH₄ values cannot be reconciled with process-based wetland CH₄ models.
Table 1. Statistics of global representative average $\delta^{13}$C-CH$_4$ values for the major CH$_4$ categories used in the Monte Carlo simulations for 1993–2017. The interannual variability (IAV), uncertainty propagated from bottom-up estimates, and the full uncertainty that considers both uncertainty from emissions and spatially resolved distribution of source signatures for different CH$_4$ categories are listed. See Table S2 for CH$_4$ sectors that are not shown here.

<table>
<thead>
<tr>
<th>CH$_4$ category</th>
<th>Mean (‰)</th>
<th>IAV (‰)</th>
<th>Emissions uncertainty</th>
<th>Full uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal</td>
<td>-45.8</td>
<td>0.71</td>
<td>1.23</td>
<td>2.57</td>
</tr>
<tr>
<td>Oil &amp; gas</td>
<td>-43.8</td>
<td>0.06</td>
<td>0.61</td>
<td>0.93</td>
</tr>
<tr>
<td>Livestock</td>
<td>-65.4</td>
<td>0.16</td>
<td>0.32</td>
<td>0.86</td>
</tr>
<tr>
<td>Wetlands</td>
<td>-59.6</td>
<td>0.10</td>
<td>0.15</td>
<td>0.20</td>
</tr>
<tr>
<td>Biomass burning*</td>
<td>-23.9</td>
<td>0.38</td>
<td>0.03</td>
<td>0.14</td>
</tr>
</tbody>
</table>

*The higher IAV of BB$_{CH4}$ than the uncertainty range is due to the spikes during some extreme El Niño years, which are one order of magnitude higher than that of most years.

**Attributions of the CH$_4$ rise based on the most likely scenarios**

Our Monte Carlo estimation (Table 1) suggests that the largest uncertainties in global representative source $\delta^{13}$C-CH$_4$ values are in industrial fossil fuel activities, providing clues for future studies. Our estimated global representative values for total CH$_4$ source signatures are within the uncertainty of recently compiled databases [12,36] but are lower than the value used in previous inverse studies (see Fig. S6 for references). Among the major CH$_4$ emission sectors, the global average emission-weighted $\delta^{13}$C-CH$_4$ signature for coal has the highest IAV, which is mainly due to the large deviation in country-level data in coal emissions [39] and the heterogeneous distribution of different coal ranks [56]. Note that the low-rank coals tend to produce isotopically lighter CH$_4$ [36] with a potentially biogenic origin [57], indicating that the proportion of consumption of different coal types may have a significant impact on atmospheric $\delta^{13}$C-CH$_4$ values.

We calculate the most likely scenarios based on the agreement of bottom-up estimates with isotopic observations (Fig. 5). The results suggest that the agricultural, landfill and waste sectors account for 53 ± 13% (21.0 ± 0.8 Tg CH$_4$ yr$^{-1}$; 1-σ) of renewed growth over the period of 2007–2017 compared to 2000–2006, with industrial fossil fuel sources and wetland sources contributing 34 ± 24% (13.7 ± 8.8 Tg CH$_4$ yr$^{-1}$) and 13 ± 9% (5.3 ± 3.5 Tg CH$_4$ yr$^{-1}$), respectively. The decreasing emissions from fossil fuel sectors in 1993–1999 compared to 2000–2006, combined with the increasing OH anomaly (Fig. S8), may have contributed to the CH$_4$ stabilization period (Fig. 5 and Fig. S7). The increases in methane emissions (mainly from the fossil fuel, agriculture and waste sectors) combined with a step increase from wetland CH$_4$ and small decreases in OH levels led to renewed growth in methane during 2007–2012. Moreover, the higher CH$_4$ emissions from mainly anthropogenic activities, i.e. coal, oil, gas, livestock, landfill and waste sectors, drove the accelerated increase in atmospheric CH$_4$ during 2013–2017. These sectoral emission increases are consistent with economic activity data (Table S2; Table S3) showing that, in the past decade, coal production has increased by 41.7% globally (International Energy Agency, https://www.iea.org/topics/coal/statistics/) and that the populations of major livestock species (e.g. swine, chickens and ruminant animals) have increased by 22.5% (FAOSTAT, http://www.fao.org/faostat/). Although coal production exhibited a temporary decline during 2014–2016 (Statistical Review of World Energy 2020, https://www.bp.com/content/dam/bp/business-sites/en/global/corporate/pdfs/energy-economics/statistical-review/bp-stats-review-2020-full-report.pdf), average coal emissions during 2013–2017 were higher than those during 2007–2012, indicating that coal mining emissions continued to grow, with a higher contribution to the increase in atmospheric CH$_4$.

**CONCLUSIONS**

Our analysis shows that a comprehensive evaluation of hypotheses regarding the attribution of rising
atmospheric CH₄ based on a combination of bottom-up approaches and isotopic values can reconcile multiple lines of evidence into a robust global CH₄ budget. However, we acknowledge that there are some biases and uncertainties in the bottom-up estimates and that our exploration of possible emission scenarios does not cover all potential scenarios. This study clearly suggests that the proposed hypotheses are influenced by the choice of a priori estimates, indicating that the high-bias a priori estimates of trends applied in some earlier studies are some biases and uncertainties in the bottom-up approaches and isotopic values can reconcile multiple lines of evidence into a robust global CH₄ budget. However, we acknowledge that there are some biases and uncertainties in the bottom-up estimates and that our exploration of possible emission scenarios does not cover all potential scenarios. This study clearly suggests that the proposed hypotheses are influenced by the choice of a priori estimates, indicating that the high-bias a priori estimates of trends applied in some earlier studies have led to equally biased conclusions regarding the attribution of atmospheric methane rise. Our results suggest that decreasing emissions from coal, oil and gas from 1993–1999 to 2000–2006, combined with the increasing OH anomaly, likely contributed to the methane stabilization period. Anthropogenic sources were the most likely major contributor to the recent rising trend in atmospheric CH₄. Our findings improve our understanding of the causes of changes in atmospheric CH₄ over the past 25 years, enabling the development of more targeted mitigation strategies and policies to stabilize and ultimately reduce key contributing emission sectors.

**MATERIALS AND METHODS**

**Model descriptions**

The model was developed from previous studies and consists of two perfectly mixed boxes representing the troposphere in the northern and southern hemispheres. The changes in CH₄ concentration are calculated using the following equations:

\[
^{12}\text{CH}_4(t + \Delta t) = ^{12}\text{CH}_4(t) + \left( \sum_i ^{12} S_i^N(t) + \sum_j k_j ^{12} \text{CH}_4^N(t) \right) \Delta t,
\]

\[
^{13}\text{CH}_4(t + \Delta t) = ^{13}\text{CH}_4(t) + \left( \sum_i ^{13} S_i^N(t) + \sum_j k_j ^{13} \text{CH}_4^N(t) \right) \Delta t,
\]

where \(^{12}\text{CH}_4\) is approximated by CH₄ and \(^{13}\text{CH}_4\) by CH₄ and \(^{12}S_i\) and \(^{13}S_i\) represent the annual source strength of the source in the northern hemisphere and southern hemisphere, respectively. \(k^{12}\) is the first-order removal rate coefficient for the sinks. The interhemispheric exchange time \(\tau\) is set to a constant value of 1 yr given that the overall methane CH₄ concentration and OH anomalies are largely unaffected by the interhemispheric exchanges [15].

The \(^{13}\text{C} / \text{CH}_4\) isotopic signatures of the different source categories \(i\) and the kinetic isotope effect (KIE) in the individual sink reactions \(j\) are used to calculate the sources \(^{13}S_i\) and removal rate coefficients \(^{13}k_j\) for \(^{13}\text{C} / \text{CH}_4\) values.

These terms are then used to derive the mixing ratio changes in \(^{13}\text{C} / \text{CH}_4\):

\[
^{13}\text{CH}_4(t + \Delta t) = ^{13}\text{CH}_4(t) + \left( \sum_i ^{13} S_i^N(t) + \sum_j k_j ^{13} \text{CH}_4^N(t) \right) \Delta t,
\]

\[
^{13}\text{CH}_4(t + \Delta t) = ^{13}\text{CH}_4(t) + \left( \sum_i ^{13} S_i^N(t) + \sum_j k_j ^{13} \text{CH}_4^N(t) \right) \Delta t.
\]

The mixing ratios of the individual isotopologues are converted to \(\delta\) values as follows:

\[
\delta^{13}_C = \left( \frac{^{13}\text{CH}_4 / ^{12}\text{CH}_4}{^{13}R_{\text{std}}} - 1 \right),
\]
where $^{13}R_{\text{tot}} = 1.12372\%$ is the $^{13}\text{C}/^{12}\text{C}$ ratio of the international reference material Vienna Pee Dee Belemnite (VPDB).

The soil sink is considered to have a low IAV, as suggested by biogeochemical models [62,63], despite a recent study [64] based on a few site-level measurements suggesting a decline in the soil sink in temperate forests in recent decades. For soil CH$_4$ uptake, we use climatology from a process-based model [63] in the calculation of the hemispheric net CH$_4$ source (see equation 6 in Materials and Methods). The contributions of CI sink and stratospheric loss to the removal of CH$_4$ in the troposphere are highly uncertain and not well constrained by direct observations, but have a strong kinetic isotope effect on $^{13}$CH$_4$. Given the large uncertainty in CI and stratospheric sinks and the lack of available datasets, the magnitudes of these two sinks were not explicitly considered in the calculation of the hemispheric CH$_4$ budget. We assume that the annual methane removal rate is driven solely by OH variability, while other minor sinks are kept constant over the study period. Because sensitivity tests [65] suggest that the uncertain magnitude of CI fields leads to a wide range of simulated $^{13}$C-CH$_4$ values given its strong ‘isotope leverage’ effect [66] (−60 ± 1‰) on total ε, the sink-weighted average fractionation factor ε is highly uncertain. The approach in this study is to estimate the total ε value for each box model run that optimizes the match between atmospheric observations and simulation at the onset of the study period. The optimized ε values were derived from running the box model in inverse mode by matching the observed global average [11]. This allows us to explore the uncertainty in ε based on bottom-up source aggregation and the uncertainty in $^{13}$C-CH$_4$ values. Figure S6 shows that the estimated fractionation factors for the full ensemble and first percentile ensemble are broadly in agreement with previous studies [11–13,60,66–70]. The distribution of the mean methane lifetime (Fig. S9) over the study period is slightly lower than the estimated 9.1 ± 0.9 yr from Ref. [71] and is comparable in magnitude to that between atmospheric chemistry-transport models in the recent model intercomparison [31,32,72]. Here, we evaluate the global results from the box model, instead of hemispheric results, to minimize the potential influence of uncertainty in IAV from inter-hemispheric transport on model performance, as suggested by a recent study [73]. See Supplementary Data for details about the model strategy.

### CH$_4$ source estimates

To test all the proposed competing hypotheses, we carried out simulation experiments using box modeling for different emission scenarios based on a suite of bottom-up datasets. We first list all the possible options for the CH$_4$ inventories by five CH$_4$ source categories (i.e. IFF$_{CH4}$, AGW$_{CH4}$, WET$_{CH4}$, BB$_{CH4}$ and GEO$_{CH4}$) and then generate emission scenarios with combinations of CH$_4$ inventories. The assignment of the inventory (i.e. EDGAR)-specific sectors into the main categories IFF$_{CH4}$, AGW$_{CH4}$ and BB$_{CH4}$ follows the criteria from Supplementary Table S4 in Ref. [5]. Anthropogenic CH$_4$ emissions related to fossil fuels from exploitation, transportation and usage of coal, oil and natural gas are defined as IFF$_{CH4}$. For methane sectors related to enteric fermentation and manure, landfills, waste and rice agriculture are defined as AGW$_{CH4}$.

### Spatially resolved $^{13}$C-CH$_4$ and uncertainty estimation

Spatially resolved distributions of $^{13}$C-CH$_4$ source signatures for the following major methane categories were applied in this study: coal, natural gas/oil, livestock, wetlands and biomass burning. For the other sources, including agricultural waste, rice, geological sources, termites, freshwater systems and wild animals, we use a globally averaged value (Table S4) from a global inventory database that collected isotopic source signatures based on literature values [36,66,68].

### Emission scenarios

An emission scenario is a combination of the individual CH$_4$ source estimates listed in Table S1. Annual total net CH$_4$ sources can be expressed as follows:

$$S_{\text{tot}} = S_{\text{IFF}} + S_{\text{AGW}} + S_{\text{WET}} + S_{\text{BB}} + S_{\text{GEO}},$$

$$+ S_{\text{OTH}} - S_{\text{soil}},$$

where S represents the individual CH$_4$ source from Table S1 and $S_{\text{soil}}$ is a constant soil sink. The total number of emission scenarios is 96, calculated as 4 IFF$_{CH4} \times 3$ AGW$_{CH4} \times 2$ WET$_{CH4} \times 2$ BB$_{CH4} \times 2$ GEO$_{CH4}$. For each emission scenario, we use Monte Carlo techniques to estimate the uncertainty in the source signature propagated from bottom-up estimates and the spatial variability of the source signature. A set of 1000 random maps of $^{13}$C-CH$_4$ values for each major CH$_4$ source (Table 1) were generated based on the uncertainty maps in this study assuming a Gaussian distribution. For CH$_4$ sources that are not spatially resolved, 1000 samples of the global-representative signature values are calculated with mean and 1-standard deviation defined by observations from the compiled databases (Table S4). One thousand sets of emission-weighted hemispheric time series of $^{13}$C-CH$_4$, which were
calculated with bottom-up estimates depending on emission scenarios, were used as inputs for the box model. For each emission scenario, the simulated time series of $\delta^{13}$C-CH$_4$ values covers the uncertainty range of spatial variability in the isotopic signatures of major CH$_4$ categories.

**DATA AVAILABILITY**

All data needed to evaluate the conclusions in this paper are present in the paper and/or the Supplementary Data. Additional ancillary data are available from the corresponding author upon request.

**SUPPLEMENTARY DATA**

Supplementary data are available at NSR online.

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**Conflict of interest statement.** None declared.

**REFERENCES**


