

# Revisiting enteric methane emissions from domestic ruminants and their $\delta$13CCH4 source signature

Jinfeng Chang, Shushi Peng, Philippe Ciais, Marielle Saunois, Shree Dangal, Mario Herrero, Petr Havlík, Hanqin Tian, Philippe Bousquet

▶ **To cite this version:**

Jinfeng Chang, Shushi Peng, Philippe Ciais, Marielle Saunois, Shree Dangal, et al.. Revisiting enteric methane emissions from domestic ruminants and their $\delta$13CCH4 source signature. Nature Communications, 2019, 10 (1), 10.1038/s41467-019-11066-3 . hal-02895160

**HAL Id: hal-02895160**
**https://hal.science/hal-02895160**

Submitted on 17 Sep 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

nature COMMUNICATIONS

ARTICLE

https://doi.org/10.1038/s41467-019-11066-3 OPEN

# Revisiting enteric methane emissions from domestic ruminants and their $\delta^{13}C_{CH4}$ source signature

Jinfeng Chang[1,7], Shushi Peng[2], Philippe Ciais[1], Marielle Saunois[1], Shree R.S. Dangal[3,4], Mario Herrero[5], Petr Havlík[6], Hanqin Tian[3] & Philippe Bousquet[1]

Accurate knowledge of $^{13}C$ isotopic signature ($\delta^{13}C$) of methane from each source is crucial for separating biogenic, fossil fuel and pyrogenic emissions in bottom-up and top-down methane budget. Livestock production is the largest anthropogenic source in the global methane budget, mostly from enteric fermentation of domestic ruminants. However, the global average, geographical distribution and temporal variations of the $\delta^{13}C$ of enteric emissions are not well understood yet. Here, we provide a new estimation of C3-C4 diet composition of domestic ruminants (cattle, buffaloes, goats and sheep), a revised estimation of yearly enteric $CH_4$ emissions, and a new estimation for the evolution of its $\delta^{13}C$ during the period 1961–2012. Compared to previous estimates, our results suggest a larger contribution of ruminants' enteric emissions to the increasing trend in global methane emissions between 2000 and 2012, and also a larger contribution to the observed decrease in the $\delta^{13}C$ of atmospheric methane.

[1] Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ/IPSL, Université Paris Saclay, 91191 Gif sur Yvette, France. [2] Sino-French Institute for Earth System Science, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China. [3] International Center for Climate and Global Change Research and School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL 36849, USA. [4] Woods Hole Research Center, Falmouth, MA 02540, USA. [5] Commonwealth Scientific and Industrial Research Organization, St Lucia, QLD 4067, Australia. [6] International Institute for Applied Systems Analysis, A-2361 Laxenburg, Austria. [7] Present address: International Institute for Applied Systems Analysis, A-2361 Laxenburg, Austria. Correspondence and requests for materials should be addressed to J.C. (email: changj@iiasa.ac.at)

Methane has important anthropogenic emissions, and is the second largest driver of global radiative forcing ($0.97 \pm 0.16$ W m$^{-2}$) after $CO_2$[1]. Understanding the global methane budget and its sources is crucial for climate mitigation efforts. Both process-based (bottom-up) and atmospheric-based (top-down) methods are used to constrain the sources and sinks of methane. However, large uncertainties exist in both approaches, which limits the complete understanding of the global methane budget.

Measurements of atmospheric methane concentrations, including their trend and gradients between stations of the surface in situ network, together with a priori spatial and temporal patterns of source type information are used by atmospheric inversion systems to produce optimized estimates of broad source categories (the Global Carbon Project[2]) and of the global budget, including surface sources and atmospheric sinks. The measurements of the $^{13}C$ stable isotope composition of atmospheric methane (i.e., $\delta^{13}C_{CH4\text{-}atm}$) bring additional constraints for attributing methane emission sources[3–6]. The $^{13}C/^{12}C$-ratio in atmospheric $CH_4$ ($\delta^{13}C_{CH4\text{-}atm}$; expressed in δ-notation relative to Vienna Pee Dee Belemnite (VPDB)-standard) is controlled by the relative contributions from different source types with distinctive isotope signatures, $\delta^{13}C_{CH4\text{-}sources}$, and by the isotopic fractionation during reaction with atmospheric OH and chlorine radicals. Reference [6] and further ref. [7] revised the $\delta^{13}C$ of methane sources and concluded that microbial sources, including wetlands, rice paddies, ruminant enteric fermentation and waste emissions, have a mean $\delta^{13}C_{CH4}$ of ~ $-61.7 \pm 6.2$‰, fossil-fuel sources have a mean $\delta^{13}C_{CH4}$ of ~ $-44.8 \pm 10.7$‰, and pyrogenic sources from biomass burning have a mean $\delta^{13}C_{CH4}$ of ~$-26.2 \pm 4.8$‰. The uncertainty and variability (in space and time) of these signatures directly affects the accuracy of source attribution by inversions. For example, the observed plateau of atmospheric methane concentration during 1999–2006, the renewed concentration-rise after 2006, and associated $\delta^{13}C_{CH4\text{-}atm}$ changes were used to quantify the role of different sources[5]. However, biases in the mean isotopic signatures of individual sources and how they change with time translate into potentially large uncertainties on the inferred trends of emission in this approach[8].

Livestock production is the largest anthropogenic source in the global methane budget (103 [95–109] Tg $CH_4$ yr$^{-1}$ during 2000–2009[2]). Enteric fermentation from ruminants dominates this source and accounts for emission of 87–97 Tg $CH_4$ yr$^{-1}$ during 2000–2009[9–12]. Livestock manure management has a smaller contribution. Cattle, buffaloes, goats, and sheep are the main ruminant livestock types emitting $CH_4$ and altogether represent 96% of the global enteric fermentation source[9]. Several methodologies are recommended by IPCC[13] (Vol. 4, Chapter 10.3) to estimate national to global enteric methane emissions. The Tier 1 method that uses the livestock population data and default emission factors, the Tier 2 approach uses a more detailed country-specific data on gross energy intake and methane conversion factors for specific livestock categories, and the Tier 3 approach allows detailed parameterization of rumen fermentation. Given the large magnitude of ruminant emissions ($F_{CH4\text{-}ruminant}$), its $\delta^{13}C$ ($\delta^{13}C_{CH4\text{-}ruminant}$) needs to be assessed as precisely as possible regionally for constraining the global mix of emissions using inversion models driven by atmospheric $CH_4$ and isotope data.

Photosynthesis pathways differentiate C3 and C4 plants[14]. C4 plants contain more $^{13}C$ than C3 plants relative to $^{12}C$. This difference in isotopic ratio causes methane emissions from ruminants with a higher C4 diet to be isotopically heavier (less negative $\delta^{13}C_{CH4}$) than those with a C3 diet. Therefore, to assess the $\delta^{13}C_{CH4\text{-}ruminant}$, it is critical to first differentiate the C3 vs. C4 feed composition in ruminant diet. To our knowledge, the proportions of C3 vs. C4 crops fed to ruminants (as concentrate feeds), as opposed to pig and poultry and their temporal changes, have not been investigated at national and global scale, since FAOSTAT only provides total feed crops for all livestock types grouped together. In addition, given the fact that C4 photosynthetic pathway predominates in warm season/low precipitation grass species, the strong increase in livestock number in tropical regions, such as South America, Africa, and South and Southeast Asia, should also increase the value of $\delta^{13}C_{CH4\text{-}ruminant}$ (i.e., heavier). Few studies have considered the impacts of shifting C4–C3 diet composition in $^{13}C$ constraints on the global $CH_4$ budget. Reference [6] estimated a global weighted mean $\delta^{13}C_{CH4\text{-}ruminant}$ value of $-66.8 \pm 2.8$‰ using an observation-based C4–C3 diet fraction of the United States only and made assumptions for the rest of the world. The spatial distribution and temporal trends of livestock $\delta^{13}C_{CH4\text{-}ruminant}$ is thus a research gap.

In addition, atmospheric isotope signatures of $CO_2$ ($\delta^{13}C_{CO2\text{-}atm}$) decreased by $-1.3$‰ from 1960 to 2012 (see observations from the Scripps $CO_2$ Program; http://scrippsco2.ucsd.edu/; data compiled in ref. [15]) due to the increasing combustion of fossil carbon. This trend can cause the synchronized decrease of $\delta^{13}C$ in plants[16]. Therefore, in addition to the shifting C4–C3 diet composition, the trend of $\delta^{13}C$ in both C4 and C3 feeds will affect the temporal trends of livestock $\delta^{13}C_{CH4\text{-}ruminant}$.

In this study, we establish a global, time-dependent dataset at national scale of the C3–C4 diet composition of domestic ruminants, the enteric methane emissions ($F_{CH4\text{-}ruminant}$), and the flux weighted isotopic signature of the methane emissions ($\delta^{13}C_{CH4\text{-}ruminant}$) over the period between 1961 and 2012. First, we separate the crop concentrate feeds consumed by ruminant vs. pigs and poultry using commodity and animal stocks statistics from FAOSTAT[9]. A simple feed model[17] is used for this separation. Then we estimate the quantity of grass and occasional fodder and scavenged biomass based on ruminant energy requirement and grass-biomass use from previous studies, all with a distinction between C3 and C4. Then, using a relationship between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ constructed in this study from $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ observations, we estimate the national- and time-dependent weighted isotopic signature of ruminant enteric methane emissions ($\delta^{13}C_{CH4\text{-}ruminant}$) for the period of 1961–2012. Finally, we quantify the impact of the revised $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ on $\delta^{13}C_{CH4\text{-}source}$, and use a one-box model to quantify their effects on atmospheric $CH_4$ concentration and $\delta^{13}C_{CH4\text{-}atm}$. Table 1 provides a glossary of terms as used in this study.

## Results

**Feeds for domestic livestock**. Annual concentrate feed commodities for livestock increased from 373 million-tons dry matter (Mt DM) in 1961 to 1186 Mt DM in 2012 (Fig. 1a). The inter-annual variation of the feed could be due to many factors, such as market price of feed (supply-side) and livestock products (demand-side), climate (mainly supply-side), and even epidemic disease (demand-side). We will only focus on decadal average and long-term trend in this study. The feed model estimated that poultry and pigs consumed about half (53%) of the concentrate feeds in 1960s, the rest being for ruminants. In 2000s, 68% of the concentrate feeds were used for poultry and pig production against 32% for ruminants. The increasing share of concentrate feeds for poultry and pigs could be due to the larger increase in the production of poultry and pigs (increased by 11.9, 5.1, and 4.5-folds for poultry meat, eggs and pig meat production, respectively, between 1961 and 2012) compared with that of ruminant (increased by 2.1-fold between 1961 and 2012), and the

**Table 1 Glossary of terms as used in this study**

| Terms | Units | Explanations |
|---|---|---|
| $\delta^{13}C_{CH4}$ | ‰ | The $^{13}C$ isotopic signature of methane; i.e., the $^{13}C/^{12}C$-ratio of $CH_4$ expressed in $\delta$-notation relative to Vienna Pee Dee Belemnite (VPDB)-standard |
| $\delta^{13}C_{CH4\text{-}atm}$ | ‰ | The $^{13}C$ isotopic signature of atmospheric methane |
| $\delta^{13}C_{CH4\text{-}sources}$ | ‰ | The $^{13}C$ isotopic signature of methane from different source types, such as microbial sources, fossil-fuel sources, and biomass burning |
| $\delta^{13}C_{CH4\text{-}ruminant}$ | ‰ | The $^{13}C$ isotopic signature of ruminant enteric fermentation methane emissions |
| $\delta^{13}C_{diet}$ | ‰ | The $^{13}C$ isotopic signature of ruminant diet |
| DM | kg | Dry matter |
| $E_{CH4}$ | MJ (kg $CH_4$)$^{-1}$ | The energy content of methane |
| $E_{GE}$ | MJ (kg dry matter)$^{-1}$ | The gross energy content of feeds |
| $F_{CH4\text{-}ruminant}$ | Tg $CH_4$ yr$^{-1}$ | The annual ruminant enteric fermentation methane emissions |
| FCR | kg dry matter (kg live-weight gain or kg eggs production)$^{-1}$ | The feed conversion ratios |
| $f_{dressing}$ | % | The dressing percentage of livestock |
| $f_{intensity}$ | % | The farming intensity |
| $f_{DE}$ | % | The digestible fraction of gross energy contained in feeds (i.e., an indicator of digestibility) |
| GE | MJ | The gross energy intake/requirement by ruminants |
| ME | MJ | The metabolizable energy intake/requirement by ruminants |
| Q | kg dry matter | The total feed quantity used for different animal types |
| REM | % | The fraction of digestible energy available in diet used for maintenance |
| Weight | kg live-weight gain or kg eggs production | The total live weight of slaughtered animals or total weight of eggs production |
| $Y_m$ | % | The methane conversion factor |

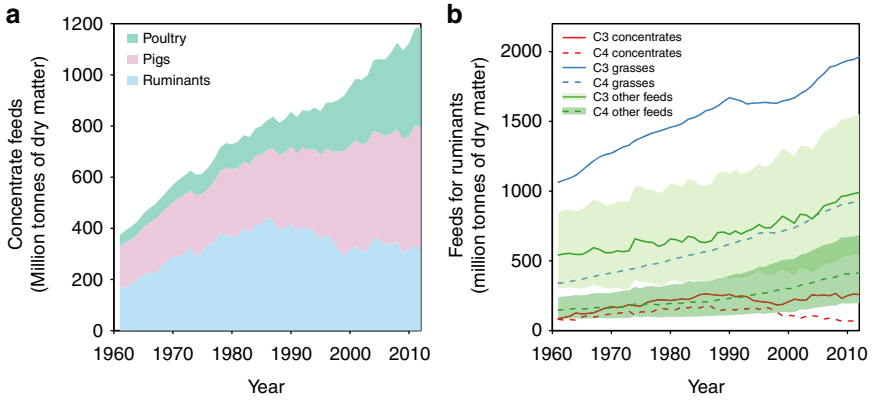



**Fig. 1** The changes in concentrate feeds for poultry, pigs, and ruminants, and in the composition of feeds for ruminants over the period of 1961–2012. The concentrate feeds for poultry, pigs, and ruminants are presented as stacked area chart in (**a**). The feeds for ruminants are concentrates (including all crop concentrate feed commodities for ruminants, C3-based or C4-based), grass (including C3 and C4 grasses), and other feeds (i.e., stover and occasional including C3 and C4 part) following ref. [18]. The light and dark green lines in (**b**) show the mean amount of C3 and C4 other feeds consumed by ruminants estimated in this study derived from Monte Carlo ensembles ($n = 1000$) from the range of uncertainty reported on feed digestibilities (i.e., $f_{DE\text{-}s+o}$, $f_{DE\text{-}concentrates}$, and $f_{DE\text{-}grass}$; see Methods section) and on the fraction of digestible energy available in diet used for maintenance (REMs; REM parameter values themselves dependent on $f_{DE}$; see Methods). The green shaded areas show the 95% confidence interval of the estimates. Source data are provided as a Source Data file

farming intensity change. Annual concentrate feeds consumed by ruminants represented 213 Mt DM yr$^{-1}$ in the 1960s, peaked at 402 Mt DM yr$^{-1}$ in the 1980s and then decreased to 332 Mt DM yr$^{-1}$ in the 2000s. The C4 concentrate feeds comprise 35% (1980s) to 37% (1990s) of the total concentrate feed commodities for livestock.

C3-based concentrate feeds constitute more than half of the ruminants' concentrate feeds over the past five decades. The share of C4 in total ruminant's concentrate feeds decreased from 43% in 1960s to 28% in 2000s. Concentrate feeds comprised only 8.0% [6.5–9.6%] of total dry matter consumption by ruminants in the 2000s, compared with 11.9% [9.9–13.9%] in the 1980s.

Grass-biomass is the largest share of ruminants' dry matter consumption comprising about 63% [52–74%] of the total dry matter intake. It is noteworthy that the share of C4 grasses in grass feed increased significantly from 24.3% in 1960s to 31.3% in 2000s mainly due to the rapid C4 grass feed increase in Latin America and Caribbean (Supplementary Fig. 1).

Other feeds represent the second largest share of total dry matter consumption by ruminants, ranging from 25.8% [14.0–38.4%] in 1980s to 29.1% [15.3–42.3%] in 2000s (Fig. 1b). The share of C4 other feeds in total other feeds follows that of the grass feed, given the assumptions made in Methods (the C3:C4 ratio of other feeds the same as the ratio of grasses for each country).

In total, the C4 diet of ruminant weighted by the fraction of each type of feed increased from 25.2% [24.8–25.8%] in the 1960s to 30.3% [30.2–30.4%] in the 2000s.

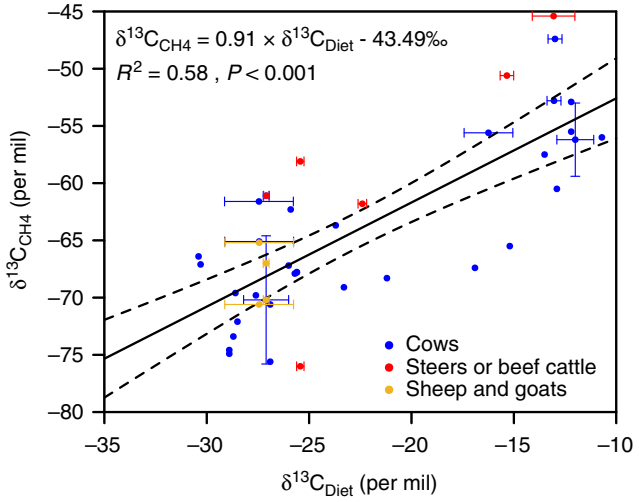



**Fig. 2** Linear regression between $\delta^{13}C$ of diet ($\delta^{13}C_{diet}$) and $\delta^{13}C$ of $CH_4$ from enteric fermentation of ruminants ($\delta^{13}C_{CH4\text{-}ruminant}$). The dots denote 43 observations from six published documents[54–59]. $\delta^{13}C_{diet}$ are either obtained directly from literature or calculated based on the feed composition and the $\delta^{13}C$ uncertainties of different feed categories (Supplementary Table 1 and 2; see Methods for detail). In the latter case, to account for the effect of decreasing $\delta^{13}C_{CO2\text{-}atm}$ on $\delta^{13}C$ of feeds, we adjust the calculated $\delta^{13}C_{diet}$ to the year when the $\delta^{13}C_{CH4\text{-}ruminant}$ was measured assuming that the biomass used for feed grows in the same year as $\delta^{13}C_{CH4\text{-}ruminant}$ measurement and follows the annual $\delta^{13}C$ of atmospheric $CO_2$. The solid line represents the best-fit correlation from the observations considering uncertainties of $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$, and the dashed lines represent 95% confidence interval. The standard errors of the fitted slope and intercept are 0.12 and 2.86‰, respectively

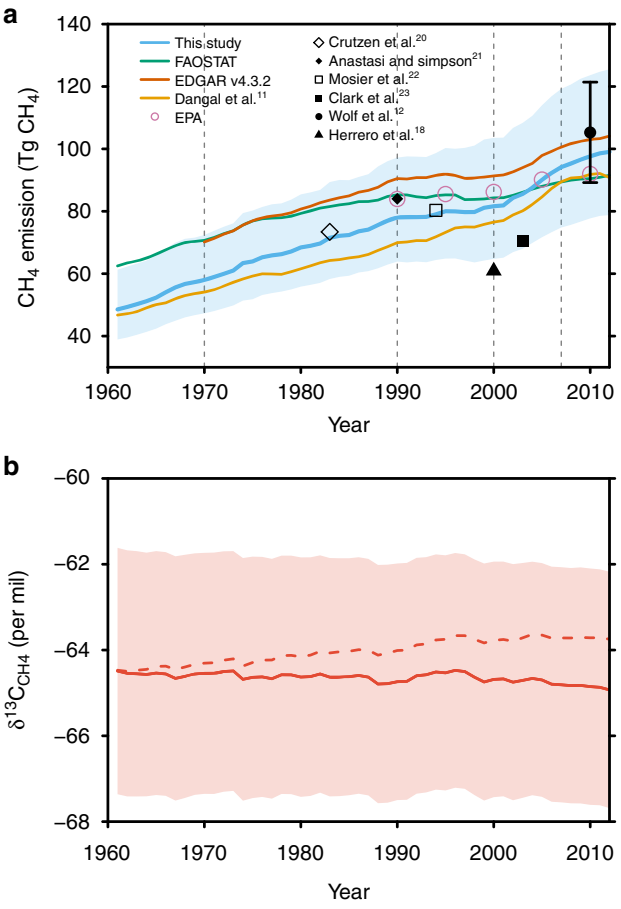



**Fig. 3** The changes in the global methane emissions from enteric fermentation of ruminants ($F_{CH4\text{-}ruminant}$), and their globally weighted average isotopic signatures $\delta^{13}C_{CH4\text{-}ruminant}$ over the period of 1961–2012. The blue (**a**) and red (**b**) shaded areas show the 95% confidence interval of the estimates of $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$, respectively. The red dashed line in (**b**) shows the $\delta^{13}C_{CH4\text{-}ruminant}$ without accounting for the effect of $\delta^{13}C_{CO2\text{-}atm}$ trend (see Methods). The dashed gray lines in (**a**) separate the periods that we consider the trend of $F_{CH4\text{-}ruminant}$ in main text and Table 2. Source data are provided as a Source Data file

**Relationship between $\delta^{13}C_{CH4\text{-}ruminant}$ and $\delta^{13}C_{diet}$.** Figure 2 shows the empirical relationship between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ extracted from literature data (see Methods; Supplementary Tables 1 and 2). Here, we apply a linear regression, which results into the following equation:

$$\delta^{13}C_{CH4} = 0.91 \times \delta^{13}C_{diet} - 43.49‰ (R^2 = 0.58, p<0.001) \quad (1)$$

with the standard errors of the fitted slope and intercept being 0.12 and 2.86‰, respectively (Fig. 2).

**Changes in $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$.** We estimated that global $F_{CH4\text{-}ruminant}$ has doubled from 48.5 ± 5.6 Tg $CH_4$ $yr^{-1}$ (mean ± 1-sigma standard deviation) in 1961 to 99.0 ± 11.7 Tg $CH_4$ $yr^{-1}$ in 2012 (Fig. 3a). The emissions' increase mainly took place in the Latin American and Caribbean countries (+13.8 ± 2.0 Tg $CH_4$ $yr^{-1}$), East and Southeast Asia (+8.8 ± 1.3 Tg $CH_4$ $yr^{-1}$), Sub-Saharan Africa (+8.4 ± 0.7 Tg $CH_4$ $yr^{-1}$), Near East and North Africa (+5.6 ± 0.6 Tg $CH_4$ $yr^{-1}$), and South Asia (+11.1 ± 1.4 Tg $CH_4$ $yr^{-1}$) (Fig. 4). By contrast, $F_{CH4\text{-}ruminant}$ decreased in Europe and Russia during the period of 1990–2012 by 31% and 54%, respectively, making the emissions of 2012 lower than those of 1961 in these two regions.

The global mean $\delta^{13}C_{diet}$ decreased a little from −23.05‰ [−25.45 to −20.66‰] in 1961 to −23.53‰ [−25.74 to −21.31‰] in 2012 (data not shown). This global diet change together with the decreasing $\delta^{13}C$ of feeds due to decreasing $\delta^{13}C_{CO2\text{-}atm}$ caused marginal change in the global mean $\delta^{13}C_{CH4\text{-}ruminant}$ (ranging from −64.49‰ [−67.36 to −61.62‰] in 1961 to −64.93‰ [−67.68 to −62.17‰] in 2012; Fig. 3b). However, $\delta^{13}C_{CH4\text{-}ruminant}$ has noticeable changes in several regions. There are $\delta^{13}C_{CH4\text{-}ruminant}$ increases in Near East and North Africa, Latin America, and Caribbean. Decreases in $\delta^{13}C_{CH4\text{-}ruminant}$ are found in North America since 1990, and in East and Southeast Asia during 1992–1996 caused by an increased reliance of C3 vs. C4 concentrates feed (in relative share; Supplementary Fig. 1).

Figure 5a shows the global distribution of national average $\delta^{13}C_{CH4\text{-}ruminant}$ in 2000s. Countries in tropical regions tend to have isotopically heavier ruminant methane emissions (less negative $\delta^{13}C_{CH4\text{-}ruminant}$) due to a higher C4 plants proportion in the diet of animals. Country-level $\delta^{13}C_{CH4\text{-}ruminant}$ due to diet shift only shows large changes in opposite sign (heavier or lighter; Fig. 5b). Within the major livestock producing countries, $\delta^{13}C_{CH4\text{-}ruminant}$ decreased by −0.3‰ and −1.9‰ in the United States and China, respectively. In these two countries, the increase in poultry and pig numbers consumed most crop feeds, including maize, so that only few C4 crops feed became allocated to ruminants. The lower fraction of C4 diet explained the decrease of $\delta^{13}C_{CH4\text{-}ruminant}$ in these two countries. In Indonesia and Malaysia, the average $\delta^{13}C_{CH4\text{-}ruminant}$ showed a strong decrease of −1.8‰ and −1.2‰, respectively. More C4 crop feed was used there to feed the increasing poultry numbers (i.e., less C4 crops left for ruminant). By contrast, $\delta^{13}C_{CH4\text{-}ruminant}$ significantly

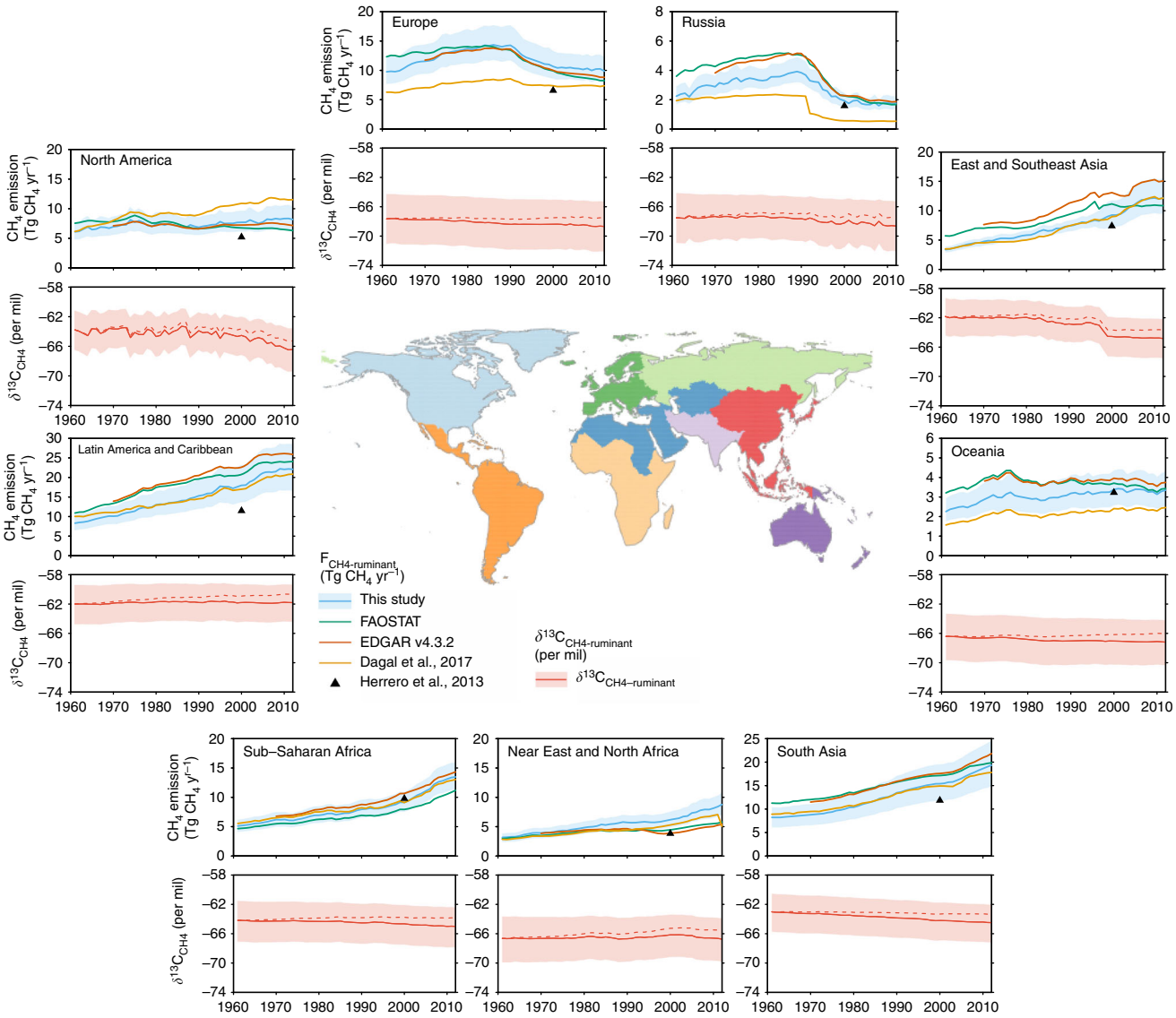



**Fig. 4** The changes in the regional methane emissions from enteric fermentation of ruminants ($F_{CH4\text{-}ruminant}$), and their weighted average isotopic signatures $\delta^{13}C_{CH4\text{-}ruminant}$ over the period of 1961–2012. The blue and red solid lines show the mean value of livestock emissions and their $\delta^{13}C_{CH4}$ from this study, respectively. The blue and red shaded areas give the 95% confidence interval of those estimates. The red dashed lines show the $\delta^{13}C_{CH4\text{-}ruminant}$ without accounting for the decreasing $\delta^{13}C_{CO2\text{-}atm}$ trend (see Methods). Regions are classified following the definition of the FAO Global Livestock Environmental Assessment Model[66]. Western and eastern Europe are combined as Europe. The vertical-scale of the regional methane emissions has been adjusted so that the changes can be easily seen

increased from the 1960s to the 2000s in Brazil (+0.3‰), Argentina (+0.5‰) and Australia (+0.5‰), which are major livestock producing countries. This increase in the source signature is explained by the combined effect of increased C4 crop feed and C4 pasture grazing.

## Discussion

Global livestock feed data, including concentrates, grasses, stover, and occasional feeds, are available only for the year 2000[18]. In this study, we reconstruct the consumption of concentrate crop feeds consumed by ruminant, the grazing of grass-biomass, and inferred the consumption of stover and occasional (other feeds) biomass as a residual to meet the metabolic energy requirement of the whole ruminant production sector over the period of 1961–2012 (Fig. 1). For 2000, we estimate that ruminants consumed 33% of the total concentrate feeds (317 Mt), 11% higher than the value given by ref. [18] (284 Mt). The difference could be due to the uncertainty in the feeds estimate for poultry and pigs in this study. The uncertainty could come from the feed conversion ratios (FCR) used here. For developed countries, we applied the FCR of poultry production derived from the industrial broiler system of the United States (intensive system with relatively low FCR). The FCRs of developing countries are assumed to be 20% higher than those of developed countries (derived from the FCR difference for pig production between the United States and China). On one hand, the FCR of poultry production other than broiler (like other types of chicken, duck, and turkey) is assumed to be the same as that of industrial broiler, which could bring uncertainty to the feed estimates. On the other hand, industrial and smallholder systems are assumed to have the same FCR for production in the simple feed model, due to the lack of the FCR information for smallholder system. Usually, smallholder system tends to have higher FCR than industrial

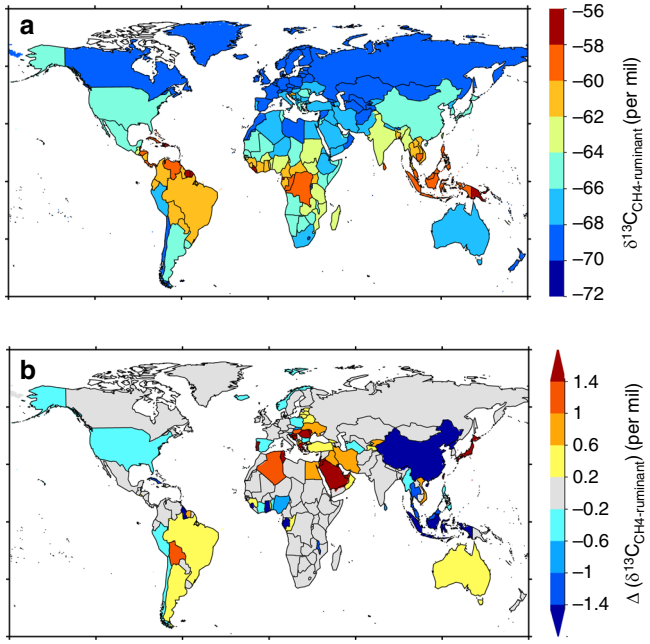



**Fig. 5** Country average $\delta^{13}C_{CH4\text{-ruminant}}$ in 2000s, and the changes in $\delta^{13}C_{CH4\text{-ruminant}}$ between 1960s and 2000s ($\Delta$ ($\delta^{13}C_{CH4\text{-ruminant}}$)). The $\Delta$ ($\delta^{13}C_{CH4\text{-ruminant}}$) is here calculated from the diet shift only (i.e., changes in the relative C3–C4 fraction in feeds) without accounting for decreasing $\delta^{13}C_{CO2\text{-atm}}$ incorporated in the biomass of grazed plants (see Methods). A positive $\Delta$ ($\delta^{13}C_{CH4\text{-ruminant}}$) indicates an increase of $\delta^{13}C_{CH4\text{-ruminant}}$ from 1960s to 2000s from a higher fraction of C4 in the diet

system. This will cause the low feed requirement estimated by the simple feed model. In addition, the uncertainty could be due to the assumed proportion of backyard production that did not receive feed commodities collected in statistics (see Methods). In a simulation assuming that all poultry and pig productions receive feed commodities, the simple feed model estimates poultry and pigs consumed 78% of the total concentrate feeds (1013 Mt), with 22% eaten by ruminants (110 Mt).

Nevertheless, concentrate feeds comprised <10% of the total dry matter consumption by ruminants in the 2000s. The grass feed fraction in 2000 used in this study was directly derived from ref. [18]. Stover and occasional feed used by ruminants is clearly the most uncertain term of the feed equation. They were estimated to be 1098 Mt in average for 2000, close to the estimate of ref. [18] (1131 Mt). But our estimates have large uncertainty ranging from 477 to 1976 Mt (95% confidence interval) due to the large uncertainty in digestibility of feeds. In summary, the simple feed model and assumptions on feed digestibility of ruminants' feed can generally reproduce livestock feed consumptions at global scale well compared with ref. [18].

In this study, annual $F_{CH4\text{-ruminant}}$ was calculated based on IPCC Tier 2 methods. Compared with Tier 1 method that used the livestock population data and default emission factors, Tier 2 approach uses a more detailed country-specific data on gross energy intake and methane conversion factors for specific livestock categories, allowing the consideration of diet quantity and quality[13]. Table 2 shows the comparison of the global $F_{CH4\text{-ruminant}}$ estimated in this study and those from previous studies for the contemporary period (1980–2012; see also Fig. 3a). Our global estimates are generally within the range of previous estimates using IPCC Tier 1 and Tier 2 methods[9–12,19–23]. However, discrepancies exist between estimates using different methods and input data. IPCC Tier 2 method considers gross and net energy requirement and associated feed intake by different livestock categories (cattle, buffaloes, sheep, and goats) and subcategories (dairy of non-dairy), and digestible energy availability of the diet (i.e., feed digestibility). For IPCC Tier 1 method, default emission factors for non-cattle livestock are from literature with indicated live-weight (see Tables 10.10 of ref. [13], Vol. 4, Chapter 10, pp. 10.28), and default region-specific emission factors for cattle used in a Tier 1 method (Tables 10.11 of ref. [13], Vol. 4, Chapter 10, pp. 10.29) are derived in fact from Tier 2 method and the data in Tables 10 A.1 and 10A.2 of ref. [13], Vol. 4, Chapter 10, pp. 10.72–10.75, which embodies livestock characteristics adjudged by expert opinions. Expert opinion necessarily limits confidence in those default emission factors to be representative of all cattle in large regions such as Africa, Middle East, and Asia. Compared with Tier 1 method, Tier 2 method (if related active data like body weight and feed digestibility are used) should allow a more accurate estimate of feed intake which is an important variable in estimating methane production from enteric fermentation (ref. [13], Vol. 4, Chapter 10, pp. 10.10). Reference [18], using Tier 3 approach with herd dynamics (i.e., age of first calving, replacement, and live-weight gain) and detailed parameterization of feed intake and rumen fermentation, estimated lower emissions in the year 2000 than all other studies (including our study; Fig. 3a). The highest emission comes from ref. [12] based on Tier 1 and accounting for recent changes in animal body mass, feed quality and quantity, milk productivity, and management of animals and manure.

The linear trend of global $F_{CH4\text{-ruminant}}$ found in this study ($0.89 \pm 0.11$ Tg $CH_4$ $yr^{-2}$) is much higher than that from FAOSTAT[9] (0.50 Tg $CH_4$ $yr^{-2}$), and is not different from that of ref. [11] (0.86 Tg $CH_4$ $yr^{-2}$) for the period of 1961–2012. For the period of 1970–1989, we estimate a trend of $F_{CH4\text{-ruminant}}$ ($0.95 \pm 0.09$ Tg $CH_4$ $yr^{-2}$) similar to that from EDGAR v4.3.2[10] (0.94 Tg $CH_4$ $yr^{-2}$) and high than those from FAOSTAT[9] and ref. [11] (Table 2). During 1990s, the emission trend from this study ($0.33 \pm 0.14$ Tg $CH_4$ $yr^{-2}$) is larger than those from FAOSTAT[9] ($-0.14$ Tg $CH_4$ $yr^{-2}$) and EDGAR v4.3.2[10] (0.05 Tg $CH_4$ $yr^{-2}$), but lower than that in ref. [11] (0.74 Tg $CH_4$ $yr^{-2}$). For the period of 2000–2012, we estimate a trend of ($1.61 \pm 0.20$ Tg $CH_4$ $yr^{-2}$) which is higher than those from FAOSTAT[9], EDGAR v4.3.2[10] and ref. [12], but similar to that from ref. [11] (1.47 Tg $CH_4$ $yr^{-2}$) using IPCC Tier 2 method. The differences of the trend estimates could come from several factors such as trends in milk yield and carcass weight, the statistics of ruminant numbers, and the assumptions of digestibility.

First, the trends from FAOSTAT[9] are lower than all other estimates, mainly due to the fact that the estimate of FAOSTAT did not consider any trend of carcass weight and milk productivity. The increasing trend of carcass weight and milk productivity resulted into higher emissions per unit livestock as in all estimates other than those from FAOSTAT[9] (see Methods). The EDGAR v4.3.2 inventory[10] applies IPCC Tier 1 methods, but uses country-specific milk yield and carcass weight trend for cattle emissions (not for other animal types like sheep and goats; i.e., a hybrid Tier 1 method). This study and ref. [11] both account for country-specific milk yield and carcass weight trend for all ruminants.

Second, ruminant livestock numbers used in this study and EDGAR v4.3.2[10] are all from FAOSTAT[9], while ref. [11] used additional data from subnational administrative regions in several countries, including the United States, Australia, Brazil, Canada, China, and Mongolia. This will affect the $F_{CH4\text{-ruminant}}$ and its trend.

Third, the method used to calculate $F_{CH4\text{-ruminant}}$ can affect the trend estimate. Time invariant default emission factors of IPCC Tier 1 method were used by FAOSTAT[9] and EDGAR v4.3.2[10]. This study and ref. [11] applied IPCC Tier 2 method considering gross and net energy requirement, and digestible energy

**Table 2 Comparison of the global methane emissions from enteric fermentation of ruminants ($F_{CH4\text{-}ruminant}$) and its trend**

| Year | This study (Tg $CH_4$ $yr^{-1}$) | Estimates from previous studies (Tg $CH_4$ $yr^{-1}$) | Method/emission factors | Sources |
|---|---|---|---|---|
| Global $F_{CH4\text{-}ruminant}$ | | | | |
| 1983 | 71.5 ± 7.8 | 73.4 | IPCC Tier 2 | Crutzen et al.[20] |
| 1990 | 78.0 ± 8.4 | 84 | IPCC Tier 1 | Anastasi and Simpson[21] |
| 1994 | 79.0 ± 8.6 | 80.3 | IPCC Tier 1 | Mosier et al.[22] |
| 2000 | 81.6 ± 9.6 | 60.9 | IPCC Tier 3 | Herrero et al.[18] |
| | | 86.3 | Mixed IPCC Tiers[a] | EPA, 2012[19] |
| | | 84.3 | IPCC Tier 1 | FAOSTAT[9] |
| | | 91.3 | Hybrid IPCC Tier 1 | EDGAR v4.3.2[10] |
| | | 76.5 | IPCC Tier 2 | Dangal et al.[11] |
| 2003 | 85.6 ± 10.0 | 70.5 | IPCC Tier 2 | Clark et al.[23] |
| 2000s | 88.9 ± 10.4 | 96.0 ± 14.7 | Revised IPCC Tier 1 | Wolf et al.[12] |
| | | 83.8 | IPCC Tier 2 | Dangal et al.[11] |
| 2010 | 97.6 ± 11.4 | 92.0 | Mixed IPCC Tiers[a] | EPA, 2012[19] |
| | | 90.4 | IPCC Tier 1 | FAOSTAT[9] |
| | | 103.1 | Hybrid IPCC Tier 1 | EDGAR v4.3.2[10] |
| | | 105.3 ± 16.1 | Revised IPCC Tier 1 | Wolf et al.[12] |
| | | 91.8 | IPCC Tier 2 | Dangal et al.[11] |
| Trend of global $F_{CH4\text{-}ruminant}$ | | | | |
| Period | This study (Tg $CH_4$ $yr^{-2}$) | Estimates from previous studies (Tg $CH_4$ $yr^{-2}$) | Method/emission factors | Sources |
| 1961–2012 | 0.92 ± 0.12 | 0.50 | IPCC Tier 1 | FAOSTAT[9] |
| | | 0.86 | IPCC Tier 2 | Dangal et al.[11] |
| 1970–1989 | 0.95 ± 0.09 | 0.69 | IPCC Tier 1 | FAOSTAT[9] |
| | | 0.94 | Hybrid IPCC Tier 1 | EDGAR v4.3.2[10] |
| | | 0.70 | IPCC Tier 2 | Dangal et al.[11] |
| 1990–1999 | 0.33 ± 0.14 | −0.14 | IPCC Tier 1 | FAOSTAT[9] |
| | | 0.05 | Hybrid IPCC Tier 1 | EDGAR v4.3.2[10] |
| | | 0.74 | IPCC Tier 2 | Dangal et al.[11] |
| 2000–2012 | 1.61 ± 0.20 | 0.64 | IPCC Tier 1 | FAOSTAT[9] |
| | | 1.19 | Hybrid IPCC Tier 1 | EDGAR v4.3.2[10] |
| | | 1.22 | Revised IPCC Tier 1 | Wolf et al.[12] |
| | | 1.47 | IPCC Tier 2 | Dangal et al.[11] |

Values from this study are shown as mean ± 1-sigma standard deviation
[a]U.S. EPA dataset is based on IPCC Tier 1 calculations supplemented with country-reported inventory data (ref. [19], pp.1), with most of the enteric CH4 emissions being from country-reported inventory data (Appendices of ref. [19], pp. G-8 to G-9). Given the fact that a majority of the reported data were derived from the UNFCCC flexible query system using higher IPCC Tiers, we called the method used by U.S. EPA data Mixed IPCC Tiers

availability of the diet (i.e., feed digestibility). But there are differences in the digestibility used. Here, we consider different digestibility according to time varying changes in the different feed types in each country (Eq. (10)) while ref. [11] used regional average feed digestibility derived from ref. [24] across the history.

Using IPCC Tier 2 method, our estimate and that from ref. [11] both give a large increase in emissions between 2002–2006 and 2008–2012, of 8.4 Tg $CH_4$ $yr^{-1}$ in ref. [12] (including manure management emissions; an increase of 6.9 Tg $CH_4$ $yr^{-1}$ for enteric emissions only), 8.6 Tg $CH_4$ $yr^{-1}$ in ref. [11] and 9.4 ± 2.0 Tg $CH_4$ $yr^{-1}$ in this study. These increases are larger than the estimate of in previous studies[9,10,19] (an increase ranging from 3.6 to 6.5 Tg $CH_4$ $yr^{-1}$; Table S5 of ref. [8] for details). This suggests a larger contribution of enteric emissions to the recent increase in global methane emissions, and also a larger contribution to the recent decrease in the $\delta^{13}C$ of atmospheric methane (lighter $\delta^{13}C_{CH4\text{-}atm}$) given the lighter $\delta^{13}C_{CH4\text{-}ruminant}$ than $\delta^{13}C_{CH4\text{-}atm}$ (see Methods). It should be noted that there is larger trend of $F_{CH4\text{-}ruminant}$ in the period of 1999–2006 (1.7 ± 0.2 Tg $CH_4$ $yr^{-2}$) and that in the period of 2008–2012 (1.0 ± 0.1 Tg $CH_4$ $yr^{-2}$). The plateau of atmospheric methane concentration observed between late-1990s and mid-2000s therefore comes from other sources with larger emission decrease[3] or/and from sink variability[5].

At regional scale, $F_{CH4\text{-}ruminant}$ estimated in this study is generally in agreement with those from FAOSTAT[9] (using IPCC Tier 1 method), EDGAR v4.3.2[10] (using a hybrid IPCC Tier 1 method with partial consideration of the trends in livestock productivity), and ref. [11] (using IPCC Tier 2 method; Fig. 4). One major difference comes from Latin America and Caribbean, where we estimate a lower $F_{CH4\text{-}ruminant}$ than those from FAOSTAT[9] and EDGAR v4.3.2[10]. Our estimate is close to that from ref. [11] using IPCC Tier 2 method, and higher than that from ref. [18] using IPCC Tier 3 method. Differences can also be found among our estimates and those from ref. [11] and ref. [18] in North America, Europe, Russia, and Oceania. This could be due to the methods (IPCC Tier 1, 2 or 3 method), statistics of livestock numbers and live weight, and regional digestibility. For example, low estimates from ref. [11] in Europe, Russia, and Oceania could be due to the high feed digestibility used (derived from Table B13 of ref. [24]). In Russia, the difference is due to the low ratio of non-dairy to dairy cattle in ref. [11] (data not shown), as non-dairy cattle emission intensity is higher than that of dairy cattle. Large differences can be found for emissions of the United States using various methods and statistics. For example, ref. [25] conducted an inventory of emissions in 1990s using subnational cattle numbers of different sub-groups, measured methane emission from each sub-group, and predominant type of diets. They reported that U. S. cattle emitted 6.6 Tg $CH_4$ in 1998, which is similar to the estimates of this study (6.7 ± 0.8 Tg $CH_4$ for all ruminants). Reference [12] recently reported U.S. enteric emission of 6.6 ± 1.0 Tg $CH_4$ in 2012 using revised emission factors, which is similar to

that of ref. [26] (6.2 Tg $CH_4$) and our estimate (7.4 ± 0.9 Tg $CH_4$). Using subnational livestock data and IPCC Tier 2 method, ref. [11] reported emissions larger than other estimates (9.4 Tg $CH_4$ in 1998, and 9.9 Tg $CH_4$ in 2012; also in Fig. 4).

Tropical and sub-tropical regions experienced much higher population growth than temperate regions in the past five decades[27]. People's diet in tropical and sub-tropical regions was also shifted toward a more animal-based protein consumption[9]. Population growth and diet shift together resulted in a larger increase in ruminant numbers and feed requirements in regions like Latin America and Caribbean, Sub-Saharan Africa, Near East and North Africa, South Asia, and East and Southeast Asia (Supplementary Fig. 1). In addition to the local consumption, international trade also contributes to the feed requirement and $F_{CH4\text{-ruminant}}$ increase in regions like Latin America and Caribbean. Net export of ruminant meat increase from 0.65 million ton in 1961 to 2.1 million ton in 2012[9]. Given the fact that concentrate feeds comprised only around 10% of the total ruminant feeds (see Results section), the C4 diet fraction in ruminant diet mainly depends on C3–C4 distribution of local feeds. As a result, larger increase in ruminants over tropical and sub-tropical regions (where C4 plant is relatively more dominant) compared with that of temperate regions, is the main cause of the global increase in the C4 diet fraction in the ruminant diet between 1960s and 2000s.

Latin America and Caribbean has the highest C4 diet fraction (Supplementary Fig. 1), and also has the largest increase in C4 diet fraction (from 43.9% [43.5–45.1%] in 1961 to 54.6% [54.2–54.9%] in 2012. This region makes the major contribution to the global increase in C4 diet fraction. The C4 diet also comprises a significant part of ruminant diet in the United States, Sub-Saharan Africa, South Asia, and East and Southeast Asia. Local C4 feeds are the dominant component of the C4 diet in these regions except the United States and China. C4 concentrates comprise 19% and 21% of the total ruminant feeds in the United States and China during the 1980s, respectively. However, due to the fast growth of poultry and pig numbers, more C4 concentrates in these two countries are used for poultry and pigs with less for ruminants simulated by our simple feed model. Thus, the C4 diet fractions in these two countries decreased in the past two decades.

The regional evolution of $F_{CH4\text{-ruminant}}$ follows the regional growth of ruminant numbers and feed consumptions, which is also a result of population growth and diet shift. In opposite to the vast $F_{CH4\text{-ruminant}}$ increase in most regions, large decreases in $F_{CH4\text{-ruminant}}$ are found in Europe and Russia between 1990 and 2012. For eastern Europe and Russia, the collapse of the Former USSR in early 1990s caused large decrease in livestock numbers and thus $F_{CH4\text{-ruminant}}$. In western Europe, the decrease in livestock numbers and $F_{CH4\text{-ruminant}}$ comes from a series of policies: (1) the European Union has provided various incentives to farmers through the Common Agricultural Policy (CAP[28]) since 1962 to avoid the negative side-effects of some farming practices, and has shifted the incentives from price support to direct aid payments to farmers who withdraw land from production in 1992 (thus reduced livestock stocking levels); (2) in 1984, the European Community introduced milk production quotas that contributed to a reduction in the dairy cow population; (3) in 1991, the Nitrates Directive (91/677/EEC[29]) restricted the application of animal manure in nitrate vulnerable zones to a maximum of 170 kg N $ha^{-1}$, which caps livestock density in pasture at some 1.7 livestock unit per ha (Annex 1 in ref. [30]).

Large scatters exist in the data shown in Fig. 2. Due to lack of a more plausible relationship, the choice of linear relationship is a reasonable assumption and an interpretation of the data. Interestingly, the resulted equation is similar to the usual isotope effects modeled with first-order kinetics[31], which is given by:

$$\alpha = \frac{R_{diet}}{R_{CH4}} \quad (2)$$

where $\alpha$ denotes the isotope effects, where $\alpha \cong 1$ and is denoted as $(1+\varepsilon)$; $R_{diet}$ and $R_{CH4}$ denote the 13C/12C molar ratio of diet (reactant) and ruminant enteric $CH_4$ emissions (products of reaction), respectively. Using $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4}$ ($R_{diet}/R_{VPDB\text{-}standard}-1$ and $R_{CH4}/R_{VPDB\text{-}standard}-1$, respectively) instead of $R_{diet}$ and $R_{CH4}$, the Eq. (2) can be transformed to:

$$1+\varepsilon = \frac{1+\delta^{13}C_{diet}}{1+\delta^{13}C_{CH4}} \quad (3)$$

Eq. (3) can lead to:

$$\delta^{13}C_{CH4} = \frac{\delta^{13}C_{diet}}{1+\varepsilon} + \frac{\varepsilon}{1+\varepsilon} \quad (4)$$

which is usually linearized to:

$$\delta^{13}C_{CH4} = \delta^{13}C_{diet} + \varepsilon \quad (5)$$

Given the uncertainties in our regression coefficient for slope (0.91 ± 0.12), our linear regression (Eq. (1)) is compatible with Eq. (5) with a $\varepsilon = 43.49 \pm 2.86‰$, where the resulted $\varepsilon$ can be seen as the isotopic discrimination factor of the fermentation processes of livestock rumen. The resulted equation reflects the biochemical reactions for stable isotopes, which always produce depleted products ($^{13}C$) while enriching the remaining substrates ($^{12}C$) owing to the preference of enzyme systems to use lighter isotope substrates $^{12}C$.

Different $\delta^{13}C_{CH4\text{-ruminant}}$despite the same $\delta^{13}C_{diet}$ was found in the collected data (i.e., the large vertical scatter in Fig. 2). This isotopic variability could be due to the differences in first, exact feed composition (e.g., C3 feed: barley, wheat, soybean, alfalfa, straw, or C3 grass; C4 feed: maize grain, maize silage, or C4 grass), second, variation of feed $\delta^{13}C$ in space and time, plant isotopic fractionation related to water use efficiency (WUE), and growing season $\delta^{13}C_{CO2\text{-}atm}$ when $CO_2$ is fixed by plants and incorporated into biomass, third, energy content of feed, and fourth, the different ruminant species (cow, steer, goat, or sheep). For example, the feed composition given by the literature is sometimes coarse for some data points (i.e., points reporting a general C3 or C4 diet in Supplementary Table 2). In this study, we estimate $\delta^{13}C_{diet}$ using $\delta^{13}C$ data collected for different feed categories, considering their uncertainty, and adjust them to the sampling year of $\delta^{13}C_{CH4\text{-ruminant}}$ using an adjustment factor derived from historical changes in $\delta^{13}C_{CO2\text{-}atm}$ from ref. [15] (see Methods). This adjustment partly accounts for the $\delta^{13}C_{diet}$ variability caused by different feed composition and different years of measurements. Spatial and temporal variability of $\delta^{13}C$ in feed plants cannot be addressed given the sparse data. Energy content of feed might also affect the isotopic variability, through its relation to gut microbes conversion of intake into $CH_4$. Given the fact that gut microbes preferably break down $^{12}C$ components, high energy content of feed could potentially increase the conversion to $CH_4$ by gut microbes, and cause heavier enteric $CH_4$ emissions. Ruminant species and even the characteristics of individual animal could also be a source of the variations in $\delta^{13}C_{CH4\text{-ruminant}}$. For example, as shown in Fig. 2, between animals fed by diet with similar $\delta^{13}C_{diet}$, the $\delta^{13}C_{CH4\text{-ruminant}}$ can vary between different ruminant species (shown with different colors in Fig. 2) and within the same species (the dispersion of $\delta^{13}C_{CH4\text{-ruminant}}$ with similar $\delta^{13}C_{diet}$ shown with the same color in Fig. 2). The variation in $\delta^{13}C_{CH4\text{-ruminant}}$ caused by different ruminant species was also observed in a study using identical feed conditions for cows, sheep, and camels[32], while the authors did

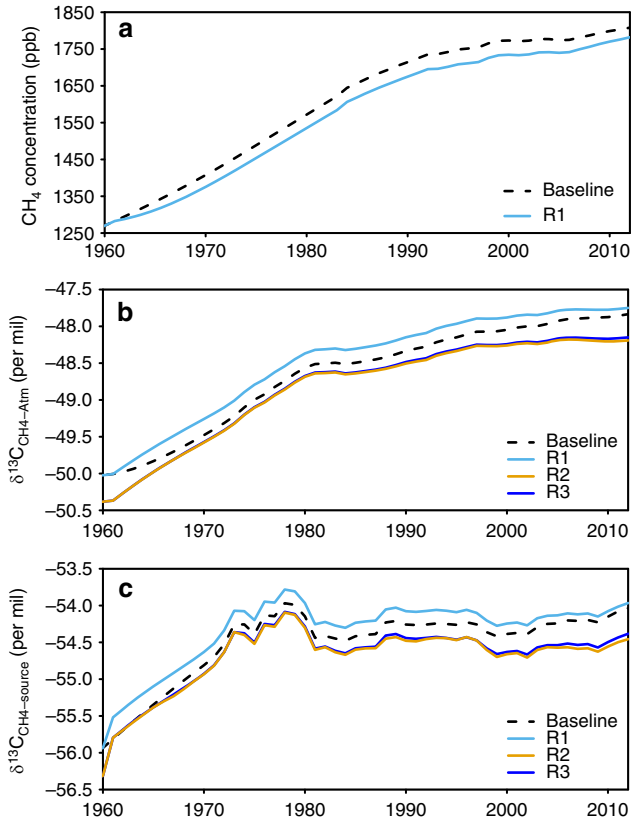



**Fig. 6** Global box model simulations of atmospheric $CH_4$ concentration, the box model results on $\delta^{13}C_{CH4\text{-}atm}$, and the isotopic source signature weighted by all sources ($\delta^{13}C_{CH4\text{-}source}$). In the simulation of R1, only the revised $F_{CH4\text{-}ruminant}$ is used and $\delta^{13}C_{CH4\text{-}ruminant}$ set to default value of −62‰ previously used by ref. [5, 43]; In R2, the revised $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ are used (see Methods). R3 is the same as R2 but with constant $\delta^{13}C_{CH4\text{-}ruminant}$ at −64.49‰ for the period of 1961–2012. The differences between R1 and baseline are the effects of the revised $F_{CH4\text{-}ruminant}$; the differences between R2 and R1 are the effects of the revised $\delta^{13}C_{CH4\text{-}ruminant}$; the differences between R3 and R2 are the effects of the $\delta^{13}C_{CH4\text{-}ruminant}$ variation (i.e., the slightly decrease from −64.49‰ in 1961 to −64.93‰ in 2012). Source data are provided as a Source Data file

not detect significant $\delta^{13}C_{CH4\text{-}ruminant}$ variation within each species.

We estimate that the annual mean $\delta^{13}C_{CH4\text{-}ruminant}$ slightly decreased from −64.49‰ [−67.36 to −61.62‰] in 1961 to −64.93‰ [−67.68 to −62.17‰] in 2012. This net small trend over 50 years is the result of two opposite mechanisms the increasing proportion of C4 grass and feeds (occasional and stover) consumed by ruminants (Fig. 1), and the decreasing $\delta^{13}C_{CO2\text{-}atm}$ that is incorporated in the biomass consumed by ruminants. The first mechanism tends to increase $\delta^{13}C_{diet}$ (less negative) of +0.74‰ over the last 50 years, while the second effect causes a decrease of $\delta^{13}C_{diet}$ of 1.18‰. Note that the magnitude of the decreasing trend of $\delta^{13}C$ of biomass is not only parallel with the decreasing $\delta^{13}C_{CO2\text{-}atm}$, but it is also controlled by trends of WUE over that period. There is evidence for an increase of WUE in temperate, boreal, and tropical forests[33–36] over the last 50 years partly attributed to increasing $CO_2$ in the atmosphere, but less so for C3 crops and grasses consumed by livestock[37]. Thus, in absence of direct observations at appropriate spatial and temporal scales to confirm an increased WUE for C3 plants consumed by livestock and given the fact that C4 plants are not expected to increase their WUE under elevated $CO_2$, we assumed conservatively that WUE is constant, which may overestimate the negative trend of $\delta^{13}C_{CH4\text{-}ruminant}$ reflecting the trend of $\delta^{13}C_{CO2\text{-}atm}$.

Our estimate is significantly lower than the data compilation from ref. [5] (−61‰), higher but within the uncertainty range of ref. [7] (−65.4 ± 6.7‰ with a median of −67.1‰), and at the upper bound of the 1-sigma standard deviation of value estimated by ref. [6] (−66.8 ± 2.8‰). The values given in ref. [7] from $\delta^{13}C_{CH4\text{-}ruminant}$ obtained in local studies were not weighted by the proportion of C3- versus C4-eating ruminants. The value estimated by ref. [6] is derived from data-driven $\delta^{13}C_{CH4\text{-}ruminant}$ estimates of −54.6 ± 3.1‰ for C4 plant-based diet, and of −69.4 ± 3.1‰ for C3 plant-based diet, and a global weighted average C4 diet fraction ranging from 1.5 to 19.6% (uniform distribution). The range is simply estimated by using the estimated C4 emission fraction of the United States (19.6%) as a global upper bound (i.e., the rest of the world have the same C4 emission fraction as the United States) and zero C4 emission fraction except the United States as a global lower bound (i.e., 1.5%). As comparison, our estimate is based on first, the refined national C3:C4 feed fraction and its associated $\delta^{13}C_{diet}$ considering the observation-based uncertainties in the $\delta^{13}C$ of different feed categories and the impact of decreasing $\delta^{13}C_{CO2\text{-}atm}$ on the $\delta^{13}C$ of feeds, and second, a data-driven relationship between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ (see Methods). For the United States, we estimated a C4 feed fraction of 22.2 ± 0.3% for the period of 1980–2012, which is higher but within the uncertainty of the fraction assumed by ref. [6] (19.6 ± 5.9% during 1980–2012).

In this study, we assess the uncertainties of the composition and the $\delta^{13}C$ of ruminant diet ($\delta^{13}C_{diet}$), ruminant enteric methane emission ($F_{CH4\text{-}ruminant}$), and its weighted $\delta^{13}C$ ($\delta^{13}C_{CH4\text{-}ruminant}$) through Monte Carlo ensembles ($n = 1000$) considering the uncertainties of the parameters used in calculation (see Methods and Supplementary Table 3). The parameters' uncertainties considered here include the feed digestibility, the fraction of digestible energy available in diet used for maintenance (REMs), the methane conversion factor ($Y_m$), the $\delta^{13}C$ of feed categories, and the fitted linear regression between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ constructed from observations.

We also acknowledge other uncertainties that are beyond our capacity of more precise estimation at current stage.

When estimating concentrate feeds intake by pigs and poultry, we account for the national livestock productivity, the different feed conversion ratio (converting production to feed requirement) and farming intensity (industrialized vs. backyard production) for developing and developed countries. Uncertainties in the feed conversion ratio have been discussed above. Besides, there could be uncertainties from other aspects. For example, the simple feed model is based on diet composition of Germany, which could cause inevitable uncertainties in the C3–C4 feed composition of pigs and poultry, and further affects uncertainties in C3–C4 concentrate feed for ruminants. There are also uncertainties in our assumptions on the logistic intensification and constant farming intensity of pig and poultry production in developing and developed countries, respectively, which will affect the time evolution of the concentrate feeds consumption by pigs and poultry. However, these uncertainties are currently not accessible due to lack of national-specific information on diet and farming intensity, and their historical changes.

In this study, we account for the impact of the global annual mean $\delta^{13}C_{CO2\text{-}atm}$ trend on the trend of $\delta^{13}C$ of feed, $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$, while the potential effects of the latitudinal gradient and seasonality of $\delta^{13}C_{CO2\text{-}atm}$ (http://scrippsco2.ucsd.edu/graphics_gallery/isotopic_data/global_stations_isotopic_c13_trends) on the $\delta^{13}C$ of feed (plants) are not considered. C4 photosynthesis is

competitive under low atmospheric $CO_2$ or high temperature/low water availability[14]. We constructed gridded C3–C4 grass (and other local feeds) distributions based on growing season temperature[38]. The impact of the rising $CO_2$ (~76 ppm during 1961–2012) on the C3–C4 grass distribution is not considered due to lack of evaluated global estimation. Based on leaf photosynthetic rates, one may expect an increase in C3 grass species due to elevated $CO_2$, thus a lower $\delta^{13}C_{diet}$ and a lower $\delta^{13}C_{CH4\text{-}ruminant}$. But long-term $CO_2$ enrichment experiments have not consistently shown a decrease of C4 species in mixed grasslands[39] suggesting ecosystem-level mechanisms that maintain a fitness of C4 plants to elevated $CO_2$. In addition, $\delta^{13}C$ of C3 plants may have a dependence on mean annual precipitation[40,41], which could potentially affect the spatial patter of the feed $\delta^{13}C$. However, a through meta-analysis on the effect on crops and grasses is needed before such relationship can be applied for assessing $\delta^{13}C$ of feeds.

In the following section, we examine more in details the impact of our revised estimates $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ on the trends of global atmospheric $CH_4$ concentration and its isotopic composition using the time-dependent one-box model of the $CH_4$ budget from refs. [42,43] (see Methods). A baseline simulation was run using the bottom-up reconstructions of the methane sources and tuning the historical atmospheric sink history for $^{12}CH_4$ and $^{13}CH_4$ to match atmospheric observations. Enteric methane emissions from EDGAR v4.3.2 were used in this baseline simulation as it is the most widely used prior inventory of atmospheric inversions[2]. Three perturbed runs were conducted to separate the effects of revised $F_{CH4\text{-}ruminant}$, revised $\delta^{13}C_{CH4\text{-}ruminant}$ and revised $\delta^{13}C_{CH4\text{-}ruminant}$ changes between 1961 and 2012, respectively (see Methods for details in the model and the simulations). The purpose of the box-model simulations is to show the impact of our new estimates of livestock emissions on atmospheric trends, not to provide closure or re-analysis of recent changes in the budget for which adjustment of other sources (wetlands, fossil fuels) would be required.

With our revised $F_{CH4\text{-}ruminant}$ emissions and all other sources and sinks unchanged to their values from the baseline run, we simulate different trends in atmospheric $CH_4$ compared with the baseline (Fig. 6a). During the period 1970–1989, our revised $F_{CH4\text{-}ruminant}$ produces a slightly smaller simulated trend of $CH_4$ (15.8 ppb $yr^{-1}$) than the baseline (16.2 ppb $yr^{-1}$). For the period 1990–1999, our estimate of $F_{CH4\text{-}ruminant}$ (Table 2) produces similar trends of $CH_4$ than the baseline. For the period after 2000, we obtain a larger trend of 4.1 ppb $yr^{-1}$ compared with 3.0 ppb $yr^{-1}$ in the baseline. This result suggests that our revised livestock emissions can explain a larger portion of the observed $CH_4$ increase during 2000–2012, but that another source should be revised downwards by the same amount—or OH sink increased—to match with the observed $CH_4$ trend.

Our revised $F_{CH4\text{-}ruminant}$ (light blue line in Fig. 6c) alone, with $\delta^{13}C_{CH4\text{-}ruminant}$ set the default value from ref. [5,43] (−62‰), implies a higher global isotopic source signature ($\delta^{13}C_{CH4\text{-}source}$ weighted by all sources) due to our lower emissions than in EDGAR v4.3.2[10]. The $\delta^{13}C_{CH4\text{-}source}$ differences with the baseline range from +0.11‰ in the 2000s to +0.18‰ in the 1970s. The smaller $\delta^{13}C_{CH4\text{-}source}$ difference in the 2000s comes from the fact that the revised $F_{CH4\text{-}ruminant}$ is closer to that of EDGAR v4.3.2[10] during that period. Using our new $\delta^{13}C_{CH4\text{-}ruminant}$ value shifts the global $\delta^{13}C_{CH4\text{-}source}$ by −0.38‰ during 1980–2012 (difference between brown and light blue lines in Fig. 6c), which counterbalances the effect of our lower $F_{CH4\text{-}ruminant}$. This result suggests that both $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ revisions have significant impacts on $\delta^{13}C_{CH4\text{-}source}$. Further studies using isotopic mass balances should thus not only include the revised source estimates for ruminants, but also revised $\delta^{13}C_{CH4\text{-}ruminant}$.

Both the revised $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ affect the mean values and trends of $\delta^{13}C_{CH4\text{-}atm}$ in the box-model (Fig. 6b). From 1990 to 2012, compared with the baseline, the revised $F_{CH4\text{-}ruminant}$ alone (light blue line in Fig. 6c) produces a larger mean $\delta^{13}C_{CH4\text{-}atm}$ by +0.15‰. The revised $\delta^{13}C_{CH4\text{-}ruminant}$ alone produces a lower mean value of $\delta^{13}C_{CH4\text{-}atm}$ than the baseline by −0.37‰. The combined revised source and isotopic signature make the mean $\delta^{13}C_{CH4\text{-}atm}$ smaller than the baseline by −0.22‰. However, the decreasing $\delta^{13}C_{CH4\text{-}ruminant}$ has a very small effect on reconstructed $\delta^{13}C_{CH4\text{-}atm}$ (by −0.02‰ only). This finding implies that even with substantial shifts in diet and geographical distribution on ruminant $CH_4$ emissions, the temporal changes in $\delta^{13}C_{CH4\text{-}ruminant}$ in the recent decades alone do not have significant effect on the reconstructed $\delta^{13}C_{CH4\text{-}atm}$.

From 1990 to 2012, the box model prescribed with the revised $F_{CH4\text{-}ruminant}$ simulates a smaller increase in $\delta^{13}C_{CH4\text{-}atm}$ (change of +0.40‰ during 1990–2012; trend of +0.017‰ $yr^{-1}$) compared with the baseline (change of +0.50‰; trend of +0.022‰ $yr^{-1}$). Adding the new $\delta^{13}C_{CH4\text{-}ruminant}$ makes the increase in $\delta^{13}C_{CH4\text{-}atm}$ even smaller (change +0.31‰; trend of +0.013‰ $yr^{-1}$). In other words, as compared with the baseline simulation using emissions from EDGAR v4.3.2[10] and default $\delta^{13}C_{CH4\text{-}ruminant}$ of −62‰, the updated $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ from this study are responsible for a lower $\delta^{13}C_{CH4\text{-}atm}$ by −0.19‰ between 1990 and 2012, and by −0.08‰ after 2006 when the $CH_4$ growth rate became positive again[44,45]. This corresponds to more than half of the observed decrease of $\delta^{13}C_{CH4\text{-}atm}$ (−0.15‰ between 2006 to 2012; derived from Table S4 of ref. [5]). In conclusion, the box-model simulations have two main implications. Firstly, the revised $CH_4$ emissions from ruminants have compensated $\delta^{13}C_{CH4\text{-}atm}$ trend that could have otherwise increased more largely driven by the increasing fossil-fuel-related emissions. Secondly, the revised $\delta^{13}C_{CH4\text{-}ruminant}$ to lower values gives a larger role of ruminant emissions in the recent $\delta^{13}C_{CH4\text{-}atm}$ trend than previously estimated, consistent with the results from ref. [5].

## Methods

**Data**. FAOSTAT—Live Animals and Livestock Primary[9] provides annual national statistics on live animal stocks, numbers of slaughtered animal and laying poultry, milking animals (producing milk) and slaughtered animals (producing meat), and correspondent milk yield for milking animals (dairy cows, sheep, goats, and buffaloes for milk), meat yield (carcass weight) for meat animals (i.e., poultry, pigs, beef cattle, sheep, goats, and buffaloes for meat), and egg yield for laying poultry. In this study, poultry species include chickens, ducks, geese, turkeys, and other birds from FAOSTAT. Data of the period 1961–2012 were used in this study.

Concentrate feeds for livestock over the period 1961–2012 were derived from FAOSTAT—Commodity Balances[9]. In total, 74 feed commodities were presented in FAOSTAT (Supplementary Table 4). These feed commodities were regrouped into seven groups as those in ref. [17] (maize, other cereals, oilseeds, cakes of oilseeds, brans, pulses, and others (mainly starch crops and sugars); Supplementary Table 4) and their dry matter content calculated using dry matter to biomass ratios (Supplementary Table 5).

**Reconstructing the feedstuff for poultry and pigs**. In this study, we adapted a simple feed model to determine the amount of concentrate feeds for poultry and pigs, and by difference to total feed commodities the amount that feeds ruminants. The details of the feed model and the methodology are described in Supplementary Note 1 (also see ref. [17]). The input data for the feed model are concentrate feeds amounts, and animal stocks numbers and yields from FAOSTAT[9]. The 74 feed commodities (in dry matter) were grouped into seven groups of concentrate feeds categories: maize, other cereals, oilseeds, cakes of oilseeds, brans, pulses, and others (mainly starch crops and sugars; Supplementary Table 4). The supply of these concentrate feeds was distributed in priority to poultry and eggs producers, then to pigs according to their specific nutritional and energy demands[46] established in the feed model (see Supplementary Note 1 for detail). Ruminant are assumed to receive all maize, other cereals, cakes of oilseeds, oilseeds, brans, and others feedstuff that are not consumed by poultry and pigs.

National- and time-dependent adjustments for farming intensity were applied to represent the situation that first, a share of the poultry and pig population are

raised by smallholder farms as backyard production, and second, the fraction of backyard production in total production is decreasing in the recent decades following global intensification trend[18]. We assume that all smallholder farms backyard productions are based on local feed resources and did not use feed commodities included in FAOSTAT. In this study, the regional specific farming intensity in the year 2000 was derived from the fraction of smallholder production in literature (see Supporting information Sect. 4 of ref. [18]). For developing countries, we assumed logistic increases in farming intensity during the period of 1960 and 2012, making the intensity value from literature reached by 2000 (Supplementary Fig. 2). The logistic increase is to mimic the intensification of pig and poultry production in developing countries characterized by a fast increase when intensification is low, and a slowing down at later stages when the level of intensification approaches the one of developed countries. An upper bound of 0.95 is set for farming intensity in developing countries, consistent with the maximum intensity indicated by Supporting information Sect. 4 of ref. [18]. Farming intensities for developed countries were assumed to remain at the fraction of 2000 all the years through 1961 to 2012.

The original feed model was constructed based on an average nutrient and energy requirement of animals for a whole year and the animal stocks at the time of enumeration[9]. In this study, we adapted the feed composition (Supplementary Note 1), and calculated feed requirements using animal productions (slaughtered for meat and eggs) and feed conversion ratios (FCR), as given by:

$$Q_{n,i,j} = \mathrm{Weight}_{n,i,j} \times \mathrm{FCR}_n \times f_{\mathrm{intensity},i,j} \quad (6)$$

where $Q_{n,i,j}$ is the total feed quantity (in kg dry matter) used for animal type $n$ (poultry for meat, poultry for eggs, pigs for meat) in country $i$ and year $j$; $\mathrm{Weight}_{n,i,j}$ is the total live weight of slaughtered animals (poultry and pigs for meat) and total weight of eggs in country $i$ in year $j$; $\mathrm{FCR}_n$ defines the feed requirement in kg dry matter per kg body weight gain or per kg egg production for animal type $n$ in year $i$; and $f_{\mathrm{intensity},i,j}$ is the farming intensity in country $i$ in year $j$.

During the recent decades, FCR of farm livestock keep decreasing due to the improvement in factors like nutrition and feeding practices (e.g., diet composition), health condition (reducing mortality), and killing weight (usually efficiency get worse after maturity). For developed countries, FCR = 1.95 kg dry matter (kg live-weight gain or kg eggs production)$^{-1}$ in 2005 with a decreasing rate of −0.01 yr$^{-1}$ for poultry live weight and eggs are used in this study. The FCR in 2005 is derived from the value for the United States Broiler Performance[47], and the decreasing rate is assumed to generally fit the FCR evolution shown in the United States Broiler Performance. FCR = 3.28 kg dry matter (kg live weight gain)$^{-1}$ in 1995 for pig live weight with a decreasing rate of −0.015 yr$^{-1}$ are used in this study. The FCR in 1995 for pigs is derived from ref. [48] (compiled from statistics of the United States). The decreasing rate is roughly estimated using value of 1995 (3.28 kg dry matter (kg live-weight gain)$^{-1}$) and of 2013 (just above 3 kg dry matter (kg live-weight gain)$^{-1}$; see ref. [49]), and assuming a linear change of the FCR. The FCRs of developing countries are assumed to be 20% higher than those of developed countries, given the example that China has 20% higher FCR for pigs than the United States[49].

$\mathrm{Weight}_{n,i,j}$ for poultry for meat ($\mathrm{Weight}_{\mathrm{poultry},i,j}$), pigs ($\mathrm{Weight}_{\mathrm{pig},i,j}$), and eggs ($\mathrm{Weight}_{\mathrm{egg},i,j}$) in country $i$ in year $j$ are calculated as:

$$\mathrm{Weight}_{\mathrm{poultry},i,j} = \frac{\sum N_{k,i,j} \times Y_{k,i,j}}{f_{\mathrm{dressing,poultry}}} \quad (7)$$

$$\mathrm{Weight}_{\mathrm{pig},i,j} = \frac{N_{\mathrm{pig},i,j} \times Y_{\mathrm{pig},i,j}}{f_{\mathrm{dressing,pig}}} \quad (8)$$

$$\mathrm{Weight}_{\mathrm{egg},i,j} = \sum N_{\mathrm{laying},k,i,j} \times Y_{\mathrm{egg},k,i,j} \quad (9)$$

where $N_{k,i,j}$ and $Y_{k,i,j}$ are the slaughtered numbers (in head) and the yield (in kg carcass weight per head) of poultry species $k$ (chickens, ducks, geese, turkeys, and other birds) in country $i$ in year $j$; $N_{\mathrm{pig},i,j}$ and $Y_{\mathrm{pig},i,j}$ are the slaughtered numbers (in head) and yield (in kg carcass weight per head) of pigs in country $i$ in year $j$; $N_{\mathrm{laying},k,i,j}$ and $Y_{\mathrm{egg},k,i,j}$ are the laying numbers (in head) and yield (in kg eggs per head) of poultry species $k$ (including hens and other birds) in country $i$ in year $j$; $f_{\mathrm{dressing,poultry}}$ and $f_{\mathrm{dressing,pig}}$ are the dressing percentage of poultry and pig, respectively, representing the conversion factor between carcass weight and live weight. $f_{\mathrm{dressing,poultry}} = 70\%$ and $f_{\mathrm{dressing,pig}} = 60\%$[50] are used in this study.

**Reconstructing the feeds for ruminants**. After having allocated concentrate feeds to poultry and pigs, ruminants are assumed to receive the remaining quantities of feed commodities ($Q_{\mathrm{concentrates}}$). In addition to feed commodities (concentrate feeds), ruminant livestock are mainly fed on grass but they also receive crop by-products (stover) and occasional feeds[18]. For grass-biomass consumed by ruminants ($Q_{\mathrm{grass}}$), we used the global livestock production dataset of gridded grass-biomass use for the year 2000 from ref. [18] extrapolated by ref. [51] backward and forward in time during 1961–2012 using metabolisable energy (ME) requirement of ruminants in each country. The rest of the consumption of biomass by ruminant livestock is assumed to be met by local stover and occasional feeds (hereafter, as other feeds), following ref. [18].

The quantities of other feeds in country $i$ in year $j$ ($Q_{s+o,i,j}$) are calculated as the solution of the following equation:

$$\begin{aligned} ME_{\mathrm{ruminant},i,j} = {} & Q_{\mathrm{s+o},i,j} \times E_{\mathrm{GE-feed}} \times f_{\mathrm{DE-s+o}} \times REM_{\mathrm{s+o}} \\ & + Q_{\mathrm{concentrates},i,j} \times E_{\mathrm{GE-feed}} \times f_{\mathrm{DE-concentrates}} \times REM_{\mathrm{concentrates}} \\ & + Q_{\mathrm{grass},i,j} \times E_{\mathrm{GE-feed}} \times f_{\mathrm{DE-grass}} \times REM_{\mathrm{grass}} \end{aligned} \quad (10)$$

where $ME_{\mathrm{ruminant},i,j}$ (MJ yr$^{-1}$) is the total ME requirement of domestic ruminants in country $i$ in year $j$ derived from ref. [51]; $E_{\mathrm{GE\text{-}feed}}$ is the average gross energy (GE) density of the feed with a value of 18.45 MJ kg$^{-1}$ of dry matter as suggested by 2006 IPCC Guidelines for National Greenhouse Gas Inventories (ref. [13], Vol. 4, Chapter 10); $f_{\mathrm{DE\text{-}s+o}}$, $f_{\mathrm{DE\text{-}concentrates}}$, and $f_{\mathrm{DE\text{-}grass}}$ (in percent) are the digestible fractions of gross energy contained in other feeds, concentrate feeds, and grass-biomass, respectively. $f_{\mathrm{DE\text{-}concentrates}}$ is set to 80% (mean value) corresponding to the feed digestibility of concentrate diet (ref. [13], Vol. 4, Chapter 10, pp. 14, with a range of 75–85% as 95% confidence interval). $f_{\mathrm{DE\text{-}s+o}}$ and $f_{\mathrm{DE\text{-}grass}}$ are set to 55% (mean value) corresponding to the feed digestibility of medium quality forage (ref. [13], Vol. 4, Chapter 10, pp. 14, range of 45–65% as 95% confidence interval). $REM_{\mathrm{s+o}}$, $REM_{\mathrm{concentrates}}$, and $REM_{\mathrm{grass}}$ (in percent) are the fractions of digestible energy available in diet used for maintenance. REM parameter values themselves depend on $f_{\mathrm{DE}}$ and are calculated following 2006 IPCC Guidelines for National Greenhouse Gas Inventories (ref. [13], Vol. 4, Chapter 10, Eqn. 10.14).

**The isotopic signature of ruminant diet**. The isotopic signature of ruminant diet in country $i$ in year $j$ ($\delta^{13}C_{\mathrm{diet},i,j}$) is determined by the C3 and C4 fractions in ruminant diet, and can be calculated as:

$$\delta^{13}C_{\mathrm{diet},i,j} = \frac{\sum Q_{\mathrm{C3feed},i,j} \times \delta^{13}C_{\mathrm{C3feed}} + \sum Q_{\mathrm{C4feed},i,j} \times \delta^{13}C_{\mathrm{C4feed}}}{\sum Q_{\mathrm{C3feed},i,j} + \sum Q_{\mathrm{C4feed},i,j}} + \Delta_{\delta^{13}C_{\mathrm{CO_2-atm},j}} \quad (11)$$

where $Q_{\mathrm{C3feed},i,j}$ and $Q_{\mathrm{C4feed},i,j}$ are feed quantities (in kg dry matter) of C3-plant sources (including C3 concentrate feeds, C3 grasses, and C3 other feeds) and C4-plant sources (including C4 concentrate feeds, C4 grasses, and C4 other feeds) in country $i$ in year $j$, respectively; $\delta^{13}C_{\mathrm{C3feed}}$ and $\delta^{13}C_{\mathrm{C4feed}}$ are the isotopic signature of different feeds for the year 2012; and $\Delta_{\delta^{13}C_{\mathrm{CO_2-atm},j}}$ is a factor to adjust the $\delta^{13}C_{\mathrm{diet}}$ to year $j$ given the fact that the $\delta^{13}C$ of plant synchronized decreases along $\delta^{13}C_{\mathrm{CO2\text{-}atm}}$ decrease[16]. $\Delta_{\delta^{13}C_{\mathrm{CO_2-atm},j}}$ is given by:

$$\Delta_{\delta^{13}C_{\mathrm{CO_2-atm},j}} = \delta^{13}C_{\mathrm{CO_2-atm},j} - \delta^{13}C_{\mathrm{CO_2-atm,2012}} \quad (12)$$

where $\delta^{13}C_{\mathrm{CO_2-atm},j}$ and $\delta^{13}C_{\mathrm{CO_2-atm,2012}}$ are isotopic signature of atmospheric $CO_2$ for year $j$ and the year 2012, respectively, which are derived from the observations of the Scripps $CO_2$ Program (http://scrippsco2.ucsd.edu/) and compiled in ref. [15]. Given the fact that the biomass used for feed are mainly the products of photosynthesis in the same year or growing season and subject to $\delta^{13}C_{\mathrm{CO2\text{-}atm}}$ of the year, we assumed no time-lag between the changes in $\delta^{13}C_{\mathrm{diet}}$ and that in $\delta^{13}C_{\mathrm{CO2\text{-}atm}}$. For $\delta^{13}C_{\mathrm{C3feed}}$ and $\delta^{13}C_{\mathrm{C4feed}}$, we used the following values derived from literature and adjusted to for the year 2012: −25.10 ± 2.27‰ for C3 concentrate feeds; −28.25 ± 1.68‰ for C3 grasses and other feeds; −12.24 ± 0.34‰ for C4 concentrate feeds (mainly maize); and −13.3 ± 1.1‰ for C4 grasses and other feeds (Supplementary Table 1).

Concentrate feeds (including all feed commodities left for ruminants and their C3 or C4 type), grasses (including C3 and C4 grasses), stover and occasional (including C3 and C4 other feeds) are major components of ruminants' diet to satisfy their ME requirement[18]. Maize ($Q_{\mathrm{maize},i,j}$), millet ($Q_{\mathrm{millet},i,j}$), sorghum ($Q_{\mathrm{sorghum},i,j}$), and sugarcane ($Q_{\mathrm{sugarcane},i,j}$) are the major C4 concentrate feeds for ruminant:

$$Q_{\mathrm{C4concentrates},i,j} = Q_{\mathrm{maize},i,j} + Q_{\mathrm{millet},i,j} + Q_{\mathrm{sorghum},i,j} + Q_{\mathrm{sugarcane},i,j} \quad (13)$$

C3 concentrate feeds include concentrate feeds other than maize, millet, sorghum, and sugarcane:

$$\begin{aligned} Q_{\mathrm{C3concentrates},i,j} = {} & Q_{\mathrm{concentrates},i,j} - Q_{\mathrm{maize},i,j} - Q_{\mathrm{millet},i,j} \\ & - Q_{\mathrm{sorghum},i,j} - Q_{\mathrm{sugarcane},i,j} \end{aligned} \quad (14)$$

where $Q_{\mathrm{concentrates},i,j}$ is the remaining quantities of feed commodities after having allocated concentrate feeds to poultry and pigs. The quantities of millet ($Q_{\mathrm{millet},i,j}$) and sorghum ($Q_{\mathrm{sorghum},i,j}$) are part of other cereals category, and the quantities of sugarcane ($Q_{\mathrm{sugarcane},i,j}$) are part of others category (Supplementary Table 4). In each country each year, the fractions of millet and sorghum in the other cereals category and sugarcane in the others category are assumed to keep the same values than in the initial feed commodities from FAOSTAT (i.e., before applying the simple feed model) and in the feeds left for ruminants (i.e., after applying the simple feed model). Components of ruminant diet other than concentrate feeds (i.e., grass and other feeds) are assumed to be produced and consumed locally. Thus we assumed the C3:C4 ratio of other feeds (i.e., $Q_{\mathrm{C3s+o},i,j}$: $Q_{\mathrm{C4s+o},i,j}$) the same as the ratio of grasses for each country (i.e., $Q_{\mathrm{C3grass},i,j}$: $Q_{\mathrm{C4grass},i,j}$) following a similar geographical C3 vs. C4 distribution[38] (see below).

For grass-biomass consumed by ruminants, we used the global livestock production dataset of gridded grass-biomass use for the year 2000 from ref. [18] extrapolated by ref. [51] at the resolution of 0.5° × 0.5° backward and forward in time

during 1961–2012 using ME requirement of ruminants in each country. To distinguish between C3 and C4 grass-biomass consumed, we used the (0.5° × 0.5°) maps prepared for the MsTMIP model intercomparison[52] of the relative fraction of C3 and C4 grasses. The MsTMIP gridded C3–C4 grass distribution is derived from the approach described in ref. [38] based on growing season temperature (see Sect. 3.6 of ref. [52] for more details). Decadal maps of the relative fraction of C3 and C4 grasses were obtained using gridded CRU–NCEP mean monthly precipitation and temperature data[53] from 1960s to 2000s. The relative fraction of C3 and C4 in each grid cell of the grass-biomass use were averaged at country level to set the C3 ($Q_{C3grass,i,j}$) and C4 grass ($Q_{C4grass,i,j}$) consumed by ruminants.

**Methane emissions from enteric fermentation of ruminants**. Methane emission from enteric fermentation for country $i$ in year $j$ ($F_{CH4\text{-}ruminant,i,j}$) is calculated using Eq. (15) adapted from IPCC Tier 2 algorithms (ref. [13], Vol. 4, Chapter 10, Eqn 10.21):

$$F_{CH4-ruminant,i,j} = \frac{\sum GE_{feed,i,j} \times Y_{m,feed}}{E_{CH4} \times 10^9} \quad (15)$$

where $GE_{ruminant,i,j}$ (in MJ) is the annual gross energy intake by ruminants in country $i$ in year $j$; $E_{CH4}$ is the energy content of methane with value of 55.65 MJ $(kg\ CH_4)^{-1}$; $Y_{m,feed}$ is the methane conversion factor expressed as the percent of gross energy in feed converted to methane; and $10^9$ is to convert the unit of methane emission from kg to Tg (i.e., $10^{12}$ g). In this study, $Y_m = 6.5\% \pm 1.0\%$ (the ± values represent the 95% confidence interval range) is used to represent widespread ruminant fed diets for cattle and mature sheep following IPCC guidelines[13] (Vol. 4, Chapter 10, Tables 10.12 and 10.13), given the fact that globally, only little cattle in some developed countries is feedlot cattle with feed diets contain 90% or more concentrates. $GE_{feed,i,j}$ is calculated as the gross energy content of different ruminant feeds (concentrates, grasses and other feeds):

$$GE_{feed,i,j} = Q_{feed,i,j} \times E_{GE-feed} \quad (16)$$

**$\delta^{13}C$ of diet and $\delta^{13}C$ of enteric methane emissions**. We collected published documents with $\delta^{13}C_{CH4\text{-}ruminant}$ observations and, at the same time, with data on $\delta^{13}C_{diet}$ or information of diet composition (quantities of specific feed categories or proportion of C3 and C4 feeds). Forty-three data from six published documents were collected[54–59], and used to derive the relationship between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$. Within the 43 observations of $\delta^{13}C_{CH4\text{-}ruminant}$, $\delta^{13}C_{diet}$ of 27 observations are obtained directly from the documents. For the rest 16 observations, feed compositions are provided (Supplementary Table 2). We calculated the $\delta^{13}C_{diet}$ as weighted average $\delta^{13}C$ of feed compositions using the $\delta^{13}C$ of different feed categories at the reference year 2012 (Supplementary Table 1), and adjusted the calculated $\delta^{13}C_{diet}$ to the year when the $\delta^{13}C_{CH4\text{-}ruminant}$ was measured (similar to Eqs. (11) and (12)). The $\delta^{13}C$ of different feed categories were originally derived from literature with different years of sampling. To be comparable, they were first adjusted to the same year 2012 using the adjustment factor $\Delta_{\delta^{13}C_{CO_2-atm,j}}$ in Eq. (12). Mean value and uncertainty of the adjusted $\delta^{13}C$ for each feed category was then calculated (Supplementary Table 1), and used to assess $\delta^{13}C_{diet}$ and its uncertainty through Monte Carlo ensembles ($n = 10{,}000$; Supplementary Table 2).

The resulted relationship between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ and their uncertainty (Fig. 2) are used to estimate the $\delta^{13}C_{CH4\text{-}ruminant}$ of the global and national methane emissions from enteric fermentation ($F_{CH4\text{-}ruminant}$) and its evolution since 1961.

**Uncertainty estimates**. In this study, we assessed the uncertainties of the quantity of other feeds for ruminant ($Q_{s+o}$; i.e., feeds other than concentrates and grasses; Eq. (10)), the $\delta^{13}C$ of ruminant diet ($\delta^{13}C_{diet}$; Eq. (11)), ruminant enteric methane emission ($F_{CH4\text{-}ruminant}$; Eq. (15)), and its weighted $\delta^{13}C$ ($\delta^{13}C_{CH4\text{-}ruminant}$, using the fitted equation of $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ obtained in this study; Fig. 2). All the above uncertainties were assessed through Monte Carlo ensembles ($n = 1000$) considering the uncertainties of the parameters used in calculation. The information on the uncertainty assessment in this study is listed in Supplementary Table 3, including the resulting estimates, corresponding equations, and the parameters, and their uncertainties considered in the uncertainty assessment.

For the quantity of other feeds for ruminant ($Q_{s+o}$), we assessed the uncertainties due to feed digestibility (parameters $f_{DE\text{-}s+o}$, $f_{DE\text{-}concentrates}$, and $f_{DE\text{-}grass}$) and associated REMs (REM parameter values themselves dependent on $f_{DE}$). For the $\delta^{13}C$ of ruminant diet ($\delta^{13}C_{diet}$), we assessed the uncertainties due to the $Q_{s+o}$ and the $\delta^{13}C$ of different feeds. For national and global methane emissions from enteric fermentation of ruminants ($F_{CH4\text{-}ruminant}$), we assessed uncertainties due to the $Q_{s+o}$ and the methane conversion factor ($Y_m$). For the weighted $\delta^{13}C_{CH4}$ of ruminants ($\delta^{13}C_{CH4\text{-}ruminant}$), we assessed the uncertainties due to the uncertainties of $\delta^{13}C_{diet,i,j}$ and the fitted linear regression between $\delta^{13}C_{diet}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ constructed from observations.

**Description of the one-box model**. The one-box model used in study is described in detail by ref. [42,43], and was recently used for studying methane sources from observed $\delta^{13}C_{CH4\text{-}atm}$[5]. We commence our model integration at an imposed steady state in 1700 as in refs. [42,43], considering the methane concentration being fairly steady at near ca. 1700. As in ref. [43], we used bottom-up constructions of the methane source inventory (as inputs; see below), and then used the inferred sink history to close the budget for individual isotopologues (i.e., $^{12}CH_4$ and $^{13}CH_4$).

Global methane mass balance can be shown as:

$$\frac{\partial C(t)}{\partial t} = S(t) - \lambda(t)C(t) \quad (17)$$

where $S(t)$, $\lambda(t)$, and $C(t)$ are the global methane source (Tg $yr^{-1}$), sink ($yr^{-1}$), and tropospheric burden (Tg), respectively, at time $t$. The model is performed at 1-year time step, thus ignoring seasonality. Assuming the source and sink are held constant within each time step, the analytic integration of Eq. (17) can be given by:

$$C_{end} = \frac{S}{\lambda} + \left(C_{beg} - \frac{S}{\lambda}\right)e^{-\lambda\Delta t} \quad (18)$$

where the tropospheric burden ($C(t)$; i.e., $C_{end}$ and $C_{beg}$) and source history ($S(t)$) are both specified at annual time steps. $C(t)$ is proportional to global mean concentration (in ppb; see below on the inputs) with a conversion factor of 2.767 Tg $ppb^{-1}$ [60,61]. With bottom-up constructions of the methane source inventory ($S(t)$ as inputs; Supplementary Fig. 3), the mass-balancing sink history ($\lambda(t)$) is deduced by solving Eq. (18) numerically in successive time steps of $\lambda$ (Supplementary Fig. 3c). With the usual assumption of $\lambda_{13} = \alpha\lambda_{12}$, where $\alpha$ is the sink weighted fractionation factor given by $\varepsilon = \alpha - 1 = -6.9‰$[5], we could deduce $\lambda_{13}(t)$ and $\lambda_{12}(t)$ separately from $\lambda(t)$ by imposing mass balance for total methane and for each methane isotopologue (with $S(t)$ and its isotopologue components $S_{13}(t)$ and $S_{12}(t)$ as inputs; Sect. 4 of ref. [43]). $S_{13}(t)$ and $S_{12}(t)$ are calculated from $S(t)$ for each inventory component using a representative $\delta^{13}C$ from Table 1 of ref. [43] (e.g., −62‰ for livestock enteric emissions; Supplementary Table 6).

To quantify the effects of the revised $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ individually or together on atmospheric $CH_4$ concentration and $\delta^{13}C_{CH4\text{-}atm}$, the model was run in forward mode (see below for the details of the simulations). A baseline simulation started in 1700, using the bottom-up constructions of the methane source inventory ($S_{13}(t)$ and $S_{12}(t)$) and the deduced mass-balancing sink history ($\lambda_{13}(t)$ and $\lambda_{12}(t)$) for each methane isotopologue over the historical period (1700–2012). R1 simulation implemented a perturbation in the baseline that commences in 1960 with the revised $F_{CH4\text{-}ruminant}$ and kept the rest sources identical to the baseline simulation (e.g., $\delta^{13}C_{CH4\text{-}ruminant}$ set to default value of −62‰ previously used by ref. [5,43]). In R2 simulation, the revised $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ (including the slightly decrease from −64.49‰ in 1961 to −64.93‰ in 2012) were used. For the spin-up period of 1700–1960 in R2 simulation, the revised $\delta^{13}C_{CH4\text{-}ruminant}$ of 1961 (−64.49‰) was used. To run the R2 simulation, we first re-calculated $S_{13}(t)$ and $S_{12}(t)$ using revised $\delta^{13}C_{CH4\text{-}ruminant}$, and re-deduced $\lambda_{13}(t)$ and $\lambda_{12}(t)$ as inputs. The R3 simulation is the same as R2 but with constant $\delta^{13}C_{CH4\text{-}ruminant}$ at −64.49‰ for the period of 1961–2012. Therefore, the resulted differences between R1 and baseline are the effects of the revised $F_{CH4\text{-}ruminant}$; the differences between R2 and R1 are the effects of the revised $\delta^{13}C_{CH4\text{-}ruminant}$; and the differences between R3 and R2 are the effects of the $\delta^{13}C_{CH4\text{-}ruminant}$ variation (i.e., the slightly decrease from −64.49‰ in 1961 to −64.93‰ in 2012). It should be noted that the purpose of the simulations is not to reproduce the observed $\delta^{13}C_{CH4\text{-}atm}$, but to get a reasonable $\delta^{13}C_{CH4\text{-}atm}$ in 1960, and the purpose of the perturbed simulations R1, R2, and R3 is to test the effects of the $F_{CH4\text{-}ruminant}$ and $\delta^{13}C_{CH4\text{-}ruminant}$ revisions on the post-1960 trends from an unperturbed 1960.

In the original box model, the mass-balancing sink ($\lambda(t)$) during the historical period (1700–2012) was tuned to match exactly atmospheric observations (Supplementary Fig. 3c). Here, we want to illustrate the effect of changing ruminant emission and its $^{13}C$ source signature, and the results from our sensitivity tests thus deviate from atmospheric observations. A re-tuning of the model to match observations of mean concentration (with our revised $F_{CH4\text{-}ruminant}$ for the period of 1961–2012) is beyond the scope of this study, but would only require a change in the global mass-balancing sink of 2.4% during 1961–2012 (from 4.9% in 1961 to <1% after 2010; see difference between red and black lines in Supplementary Figure 3c), which is well within the uncertainty of the atmospheric sink. An additional sensitivity test shows that the re-tuning of the global mass-balancing sink only has marginal effect on the results presented in the Discussion section.

Inputs for the box model include the global methane concentration (Supplementary Fig. 3), and the construction of the methane source inventory from 1700 (Supplementary Table 6). The global methane concentration since 1984 is derived from observations of a globally distributed network (NOAA-ESRL record[44,62]). Data for 1700–1983 are from ref. [63], converted to the NOAA04 scale[64] and adjusted by 45% of the inter-polar gradient to adjust Antarctic to global values[5].

For the construction of the methane source inventory, we combined the natural and anthropogenic emissions of 1700 derived from Table 1 of ref. [43], the historical anthropogenic emissions[65] (1850–2000) used by the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), and the EDGAR v4.3.2[10] for the recent decades (Supplementary Table 6). Five natural emission sources are used in this study following Table 1 of ref. [43]: wetlands, termites, oceans, wild animals, and geologic sources. We assumed that the natural emissions are kept constant across history. Nine anthropogenic sources are used including fossil fuel (industry), waste treatment and landfill, rice cultivation, livestock enteric fermentation,

manure management, and four pyrogenic sources (agricultural waste burning, forest burning, C3 grass burning, and savanna (C4 grass) burning). The emissions of 1700 are derived from Table 1 of ref. [43] except pyrogenic sources (see below). Reference [65] provides gridded (0.5° × 0.5°) historical anthropogenic methane emissions from 1850 to 2000. This dataset includes fossil fuel, waste treatment and landfill, and different pyrogenic sources (agricultural waste burning, forest burning, and grass burning), but gathers rice cultivation, livestock enteric fermentation, manure management as a single agricultural sector source. To separate grass burning into C3 grass burning and savanna (C4 grass) burning, we applied the gridded C3–C4 grass distribution derived from the approach described in ref. [38] based on growing season temperature. The EDGAR v4.3.2[10] provides emissions from fossil fuel, waste treatment and landfill, rice cultivation, livestock enteric fermentation, manure management, and agricultural waste burning for the period of 1970–2012. To separate the agricultural sector emissions from ref. [65] into different components (for the period of 1850–1970), we applied the relative fractions of rice cultivation, livestock enteric fermentation, and manure management from EDGAR v4.3.2[10] for the year 1970. For pyrogenic emissions, we did not use fire emissions (wildfires, forest burning and savanna burning) of 1700 in the Table 1 of ref. [43]. Instead, we used pyrogenic emissions of ref. [65], and assumed that they are constant between 1700 and 1850. For each of the anthropogenic source, we applied linear interpolation between 1700 (Table 1 of ref. [43]) and 1850[65], and between 1850 and 1970 (for each decade) to obtain annual emissions. For the year since 1970, values from the EDGAR v4.3.2[10] were used.

## Data availability

The data that support the findings of this study are available at the public Data Repository of the International Institute of Applied Systems Analysis (IIASA DARE; https://doi.org/10.22022/ESM/06-2019.45). The source data underlying Figs. 1, 3, and 6 are provided as a Source Data file. The source data underlying Fig. 2 are provided by Supplementary Table 2. The source data underlying Figs. 4 and 5 can be found in the IIASA DARE (https://doi.org/10.22022/ESM/06-2019.45).

Received: 5 February 2019 Accepted: 18 June 2019
Published online: 31 July 2019

## References

1. Myhre, G. et al. in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (eds Stocker, T. F. et al.). (Cambridge University Press, Cambridge, UK and New York, NY, USA, 2013).
2. Saunois, M. et al. The global methane budget 2000–2012. *Earth Sys. Sci. Data* **8**, 697–751 (2016).
3. Bousquet, P. et al. Contribution of anthropogenic and natural sources to atmospheric methane variability. *Nature* **443**, 439 (2006).
4. Mikaloff Fletcher, S. E., Tans, P. P., Bruhwiler, L. M., Miller, J. B. & Heimann, M. $CH_4$ sources estimated from atmospheric observations of $CH_4$ and its $^{13}C/^{12}C$ isotopic ratios: 1. Inverse modeling of source processes. *Glob. Biogeochem. Cycles* **18**, https://doi.org/10.1029/2004gb002223 (2004).
5. Schaefer, H. et al. A 21st-century shift from fossil-fuel to biogenic methane emissions indicated by $^{13}CH_4$. *Science* **352**, 80–84 (2016).
6. Schwietzke, S. et al. Upward revision of global fossil fuel methane emissions based on isotope database. *Nature* **538**, 88–91 (2016).
7. Sherwood, O. A., Schwietzke, S., Arling, V. A. & Etiope, G. Global inventory of gas geochemistry data from fossil fuel, microbial and burning sources, version 2017. *Earth Syst. Sci. Data* **9**, 639–656 (2017).
8. Saunois, M. et al. Variability and quasi-decadal changes in the methane budget over the period 2000–2012. *Atmos. Chem. Phys.* **17**, 11135–11161 (2017).
9. FAOSTAT Online Statistical Service. (Food and Agriculture Organization (FAO), 2017); http://faostat3.fao.org.
10. EDGARv4.3.2. http://edgar.jrc.ec.europa.eu/ (2018).
11. Dangal, S. R. S. et al. Methane emission from global livestock sector during 1890–2014: magnitude, trends and spatiotemporal patterns. *Glob. Chang. Biol.* **23**, 4147–4161 (2017).
12. Wolf, J., Asrar, G. R. & West, T. O. Revised methane emissions factors and spatially distributed annual carbon fluxes for global livestock. *Carbon Balance Manag.* **12**, 16 (2017).
13. IPCC. *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (eds Eggleston, H. S., Buendia, L., Miwa, K., Ngara, T. & Tanabe, K.) https://www.ipcc-nggip.iges.or.jp (Institute for Global Environmental Strategies Hayama, Japan, 2006).
14. Ehleringer, J. R. & Cerling, T. E. C3 and C4 photosynthesis. *Encycl. Glob. Environ. Change* **2**, 186–190 (2002).
15. Graven, H. et al. Compiled records of carbon isotopes in atmospheric $CO_2$ for historical simulations in CMIP6. *Geosci. Model Dev.* **10**, 4405–4417 (2017).
16. Francey, R. J. & Farquhar, G. D. An explanation of $^{13}C/^{12}C$ variations in tree rings. *Nature* **297**, 28–31 (1982).
17. Ciais, P., Bousquet, P., Freibauer, A. & Naegler, T. Horizontal displacement of carbon associated with agriculture and its impacts on atmospheric $CO_2$. *Glob. Biogeochem. Cycles* **21**, https://doi.org/10.1029/2006gb002741 (2007).
18. Herrero, M. et al. Biomass use, production, feed efficiencies, and greenhouse gas emissions from global livestock systems. *Proc. Natl Acad. Sci. USA* **110**, 20888–20893 (2013).
19. United States Environment Protection Agency (U.S. EPA). *Global Anthropogenic Non-$CO_2$ Greenhouse Gas Emissions: 1990–2030*. Revised December 2012. https://www.epa.gov/global-mitigation-non-co2-greenhouse-gases/global-anthropogenic-non-co2-greenhouse-gas-emissions (United States Environment Protection Agency, Washington DC, 2012).
20. Crutzen, P. J., Aselmann, I. & Seiler, W. Methane production by domestic animals, wild ruminants, other herbivorous fauna, and humans. *Tellus B* **38**, 271–284 (1986).
21. Anastasi, C. & Simpson, V. J. Future methane emissions from animals. *J. Geophys. Res.: Atmospheres* **98**, 7181–7186 (1993).
22. Mosier, A. R. et al. Mitigating agricultural emissions of methane. *Clim. Change* **40**, 39–80 (1998).
23. Clark, H., Pinares-Patino, C. & De Klein, C. in *Grassland. A Global Resource* (ed. McGilloway) 279–293 (Wageningen Academic Publishers, Wageningen, The Netherlands, 2005).
24. Opio, C. et al. *Greenhouse Gas Emissions from Ruminant Supply Chains—a Global Life Cycle Assessment* 1–214 (Food and agriculture organization of the United Nations (FAO), Rome, 2013).
25. Westberg, H., Lamb, B., Johnson, K. A. & Huyler, M. Inventory of methane emissions from U.S. cattle. *J. Geophys. Res. -Atmos.* **106**, 12633–12642 (2001).
26. Hristov, A. N. et al. Discrepancies and uncertainties in bottom-up gridded inventories of livestock methane emissions for the contiguous United States. *Environ. Sci. Technol.* **51**, 13668–13677 (2017).
27. United Nations, Department of Economic and Social Affairs, Population Division. World Population Prospects: The 2015 Revision, World Population 2015 Wallchart. ST/ESA/SER.A/378 (2015).
28. European Common Agricultural Policy (CAP), https://ec.europa.eu/info/food-farming-fisheries/key-policies/common-agricultural-policy/cap-glance_en.
29. European Commission, Commission staff working document accompanying the document. *Report from the Commission to the Council and the European Parliament on the Implementation of Council Directive 91/676/EEC Concerning the Protection of Waters Against Pollution Caused by Nitrates from Agricultural Sources Based on Member State Reports for the Period 2008–2011 {COM(2013) 683 final}* (European Commission, Brussels, 2013).
30. Webb, J. et al. *Study on Variation of Manure N Efficiency Throughout Europe* (AEA Technology Plc, Didcot, 2011).
31. Farquhar, G. D., J. R. Ehleringer, A. & Hubick, K. T. Carbon isotope discrimination and photosynthesis. *Annu. Rev. Plant Phys.* **40**, 503–537 (1989).
32. Schulze, E., Lohmeyer, S. & Giese, W. Determination of $^{13}C/^{12}C$-ratios in rumen produced methane and $CO_2$ of cows, sheep and camels. *Isot. Environ. Heal. S* **34**, 75–79 (1998).
33. van der Sleen, P. et al. No growth stimulation of tropical trees by 150 years of $CO_2$ fertilization but water-use efficiency increased. *Nat. Geosci.* **8**, 24 (2014).
34. Peñuelas, J., Canadell, J. G. & Ogaya, R. Increased water-use efficiency during the 20th century did not translate into enhanced tree growth. *Glob. Ecol. Biogeogr.* **20**, 597–608 (2011).
35. Giguère-Croteau, C. et al. North America's oldest boreal trees are more efficient water users due to increased $CO_2$ but do not grow faster. *Proc. Natl Acad. Sci. USA* **116**, 2749–2754 (2019).
36. Frank, D. C. et al. Water-use efficiency and transpiration across European forests during the Anthropocene. *Nat. Clim. Change* **5**, 579 (2015).
37. Tang, X. et al. How is water-use efficiency of terrestrial ecosystems distributed and changing on Earth? *Sci. Rep.-UK* **4**, 7483 (2014).
38. Still, C. J., Berry, J. A., Collatz, G. J. & DeFries, R. S. Global distribution of C-3 and C-4 vegetation: carbon cycle implications. *Glob. Biogeochem. Cycles* **17**, 1006 (2003).
39. Reich, P. B., Hobbie, S. E., Lee, T. D. & Pastore, M. A. Unexpected reversal of C3 versus C4 grass response to elevated $CO_2$ during a 20-year field experiment. *Science* **360**, 317–320 (2018).
40. Kohn, M. J. Carbon isotope compositions of terrestrial C3 plants as indicators of (paleo)ecology and (paleo)climate. *Proc. Natl Acad. Sci. USA* **107**, 19691–19695 (2010).
41. Diefendorf, A. F., Mueller, K. E., Wing, S. L., Koch, P. L. & Freeman, K. H. Global patterns in leaf $^{13}C$ discrimination and implications for studies of past and future climate. *Proc. Natl Acad. Sci. USA* **107**, 5738–5743 (2010).
42. Lassey, K. R., Lowe, D. C. & Manning, M. R. The trend in atmospheric methane δ13C and implications for isotopic constraints on the global methane budget. *Glob. Biogeochem. Cycles* **14**, 41–49 (2000).

43. Lassey, K. R., Etheridge, D. M., Lowe, D. C., Smith, A. M. & Ferretti, D. F. Centennial evolution of the atmospheric methane budget: what do the carbon isotopes tell us? *Atmos. Chem. Phys.* **7**, 2119–2139 (2007).
44. NOAA/ESRL. www.esrl.noaa.gov/gmd/ccgg/trends_ch4.
45. Nisbet, E. G. et al. Rising atmospheric methane: 2007–2014 growth and isotopic shift. *Glob. Biogeochem. Cycles* **30**, 1356–1370 (2016).
46. KTBL (Kuratorium fur Technic und Bauwessen in der Landwitschaft). *KTBL Pocket Book Agriculture 2000/01* (in German), 20th edn. (Kuratorium fur Technic und Bauwessen in der Landwitschaft, Munster, 2000).
47. National Chicken Council. https://www.nationalchickencouncil.org/about-the-industry/statistics/u-s-broiler-performance/ (2018).
48. Losinger, W. C. Feed-conversion ratio of finisher pigs in the USA. *Prev. Vet. Med.* **36**, 287–305 (1998).
49. Rabobank Research. https://research.rabobank.com/far/en/sectors/farm-inputs/Pigs_Might_Fly.html (2015).
50. Verheijen, L. A. H. M., Wiersema, D., Hulshoff Pol, L. W. & De Wit, J. Management of Waste from Animal Product Processing, International Agriculture Centre, Wageningen, The Netherlands. http://www.fao.org/WAIRDOCS/LEAD/X6114E/x6114e04.htm (1996).
51. Chang, J. et al. Combining livestock production information in a process-based vegetation model to reconstruct the history of grassland management. *Biogeosciences* **13**, 3757–3776 (2016).
52. Wei, Y. et al. The North American carbon program multi-scale synthesis and terrestrial model intercomparison project—Part 2: Environmental driver data. *Geosci. Model Dev.* **7**, 2875–2893 (2014).
53. Viovy, N. CRU-NCEPv4, CRUNCEP dataset, http://dods.extra.cea.fr/data/p529viov/cruncep/readme.htm (2013).
54. Rust, F. Ruminant methane delta ($^{13}C/^{12}C$) values: relation to atmospheric methane. *Science* **211**, 1044–1046 (1981).
55. Metges, C., Kempe, K. & Schmidt, H.-L. Dependence of the carbon-isotope contents of breath carbon dioxide, milk, serum and rumen fermentation products on the $\delta^{13}C$ value of food in dairy cows. *Brit. J. Nutr.* **63**, 187–196 (1990).
56. Levin, I., Bergamaschi, P., Dörr, H. & Trapp, D. Stable isotopic signature of methane from major sources in Germany. *Chemosphere* **26**, 161–177 (1993).
57. Klevenhusen, F., Bernasconi, S. M., Kreuzer, M. & Soliva, C. R. Experimental validation of the Intergovernmental Panel on Climate Change default values for ruminant-derived methane and its carbon-isotope signature. *Anim. Prod. Sci.* **50**, 159–167 (2010).
58. Klevenhusen, F. et al. Efficiency of monolaurin in mitigating ruminal methanogenesis and modifying C-isotope fractionation when incubating diets composed of either C3 or C4 plants in a rumen simulation technique (Rusitec) system. *Brit. J. Nutr.* **102**, 1308–1317 (2009).
59. Bilek, R. S., Tyler, S. C., Kurihara, M. & Yagi, K. Investigation of cattle methane production and emission over a 24-hour period using measurements of $\delta^{13}C$ and δD of emitted $CH_4$ and rumen water. *J. Geophys. Res. -Atmos.* **106**, 15405–15413 (2001).
60. Fung, I. et al. Three-dimensional model synthesis of the global methane cycle. *J. Geophys. Res. -Atmos.* **96**, 13033–13065 (1991).
61. Dlugokencky, E. J., Masarie, K. A., Lang, P. M. & Tans, P. P. Continuing decline in the growth rate of the atmospheric methane burden. *Nature* **393**, 447–450 (1998).
62. Dlugokencky Edward, J., Nisbet Euan, G., Fisher, R. & Lowry, D. Global atmospheric methane: budget, changes and dangers. *Philos. T. R. Soc. A* **369**, 2058–2072 (2011).
63. MacFarling Meure, C. et al. Law Dome $CO_2$, $CH_4$ and $N_2O$ ice core records extended to 2000 years BP. *Geophys. Res. Lett.* **33**, https://doi.org/10.1029/2006gl026152 (2006).
64. Dlugokencky, E. J. et al. Conversion of NOAA atmospheric dry air $CH_4$ mole fractions to a gravimetrically prepared standard scale. *J. Geophys. Res.-Atmos.* 110, https://doi.org/10.1029/2005jd006035 (2005).
65. Lamarque, J. F. et al. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmos. Chem. Phys.* **10**, 7017–7039 (2010).
66. GLEAM. FAO Global Livestock Environmental Assessment Model; http://www.fao.org/gleam/en/.


## Acknowledgements

J.C. and P.C. acknowledge support from the ERC Synergy grant ERC-2013-SyG-610028 IMBALANCE-P. S.P. acknowledges support from the National Natural Science Foundation of China (grant number 41722101). H.T. and S.D. acknowledge support from NSF (grant numbers: 1903722, 1243232) and NKRDPC (grant number 2017YFA0604702).

## Author contributions

P.C., J.C., and S.P. designed the study. J.C. and S.P. carried out the analysis. J.C., S.P., and P.C. wrote most of the paper. M.S., S.R.S.D, M.H., P.H., H.T., and P.B. contributed significantly to the final revisions of the paper.

## Additional information

**Supplementary Information** accompanies this paper at https://doi.org/10.1038/s41467-019-11066-3.

**Competing interests:** The authors declare no competing interests.

**Reprints and permission** information is available online at http://npg.nature.com/reprintsandpermissions/

**Peer review information:** *Nature Communications* thanks Hinrich Schaefer, Zhiliang Tan and other anonymous reviewer(s) for their contribution to the peer review of this work.

**Publisher's note:** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

**Open Access** This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2019