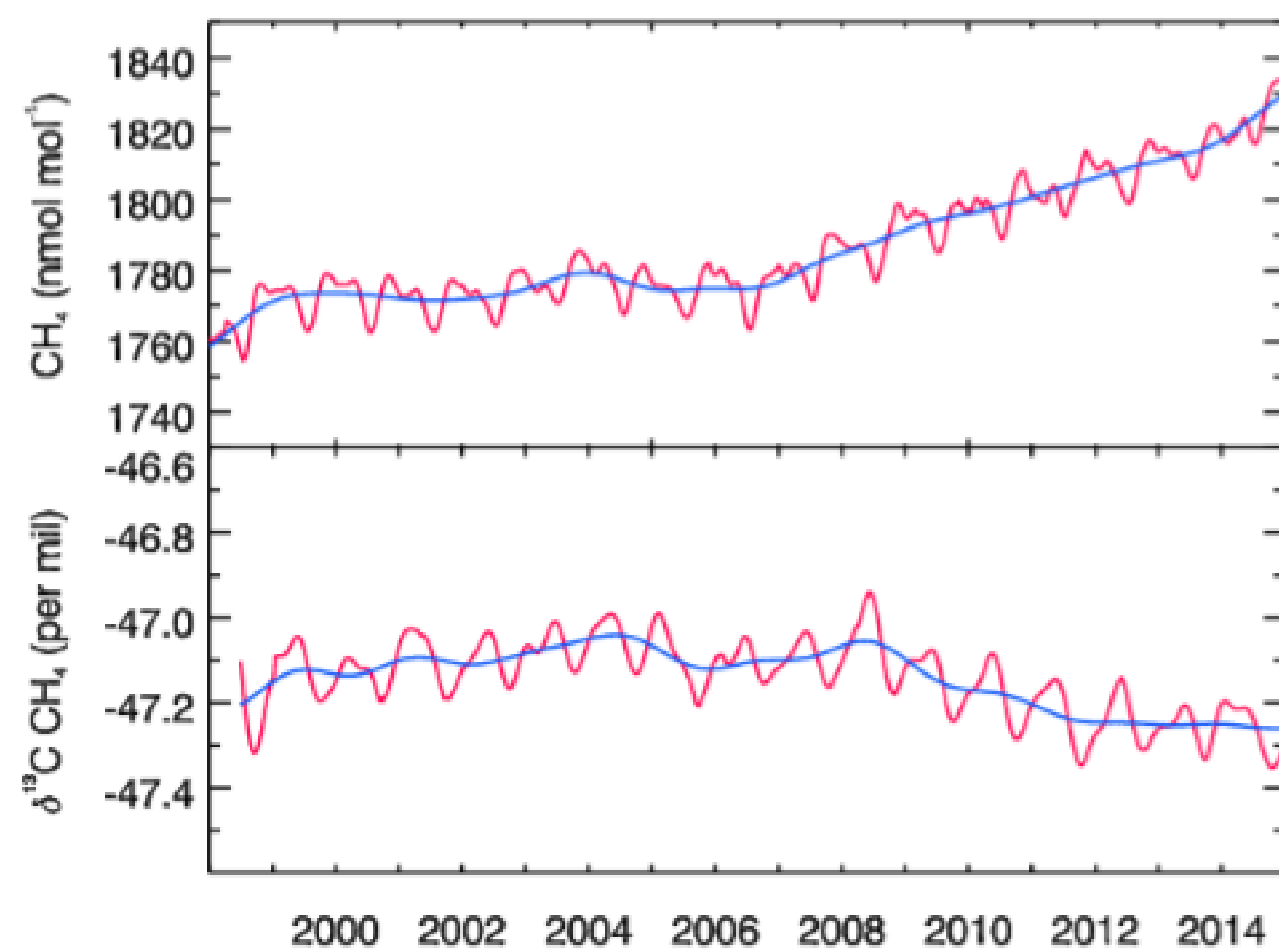
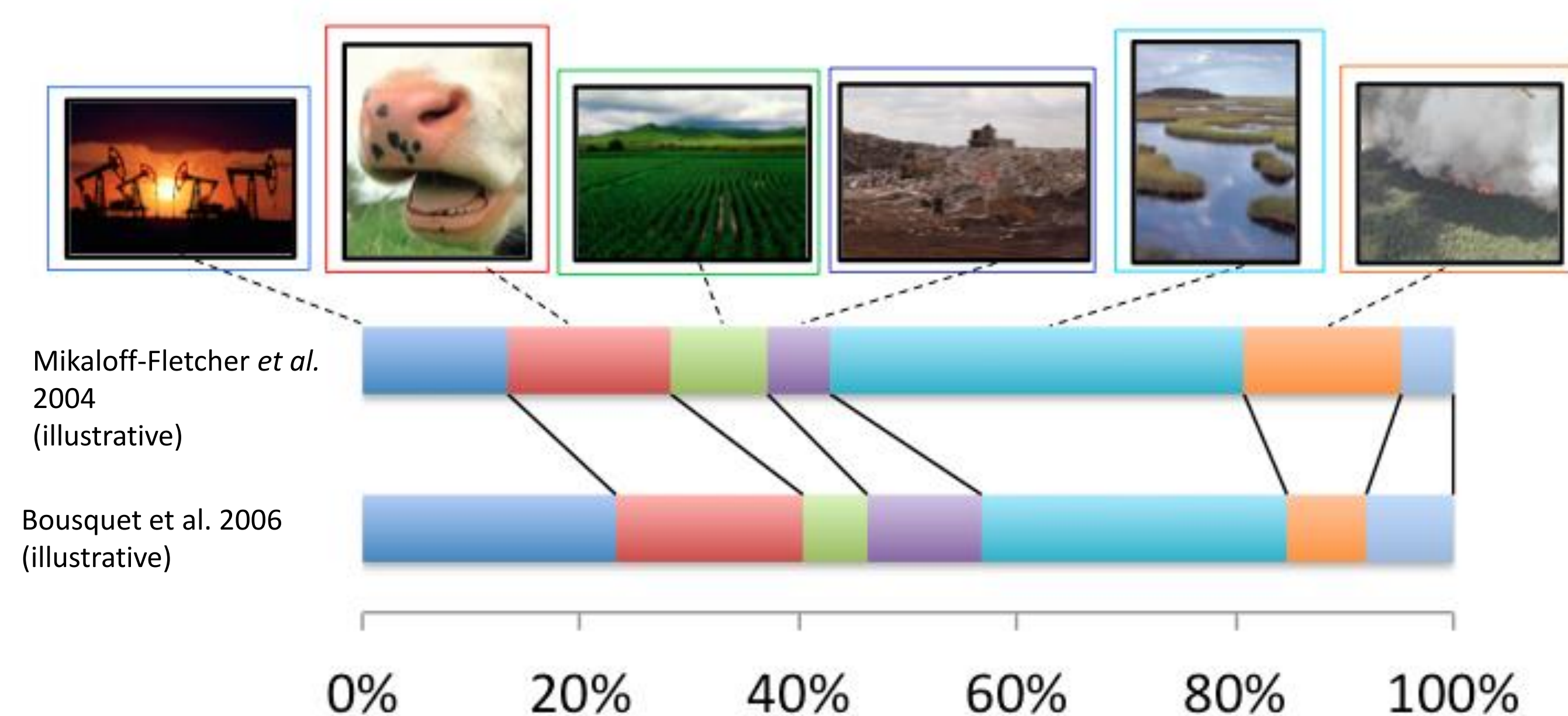


## Introduction

Methane (CH<sub>4</sub>) is a potent greenhouse gas (GHG) which accounts for ~20% of the GHG radiative forcing over the industrial era (Dlugokencky et al., 2011). The rise in atmospheric CH<sub>4</sub> has been non-linear, with a period of relative stability from 1997-2007, followed by a renewed increase since 2008 (Nisbet et al., 2014; Fig. 1). The reasons for these variations are not completely understood, partly because specific contributions from natural and anthropogenic sources remain unclear (Fig. 2).



**Figure 1:** Global time series plots of CH<sub>4</sub> and  $\delta^{13}\text{C}$  of CH<sub>4</sub> from 1999-2014. The recent rise in CH<sub>4</sub> is concurrent with a decrease in  $\delta^{13}\text{C}$  of CH<sub>4</sub>.



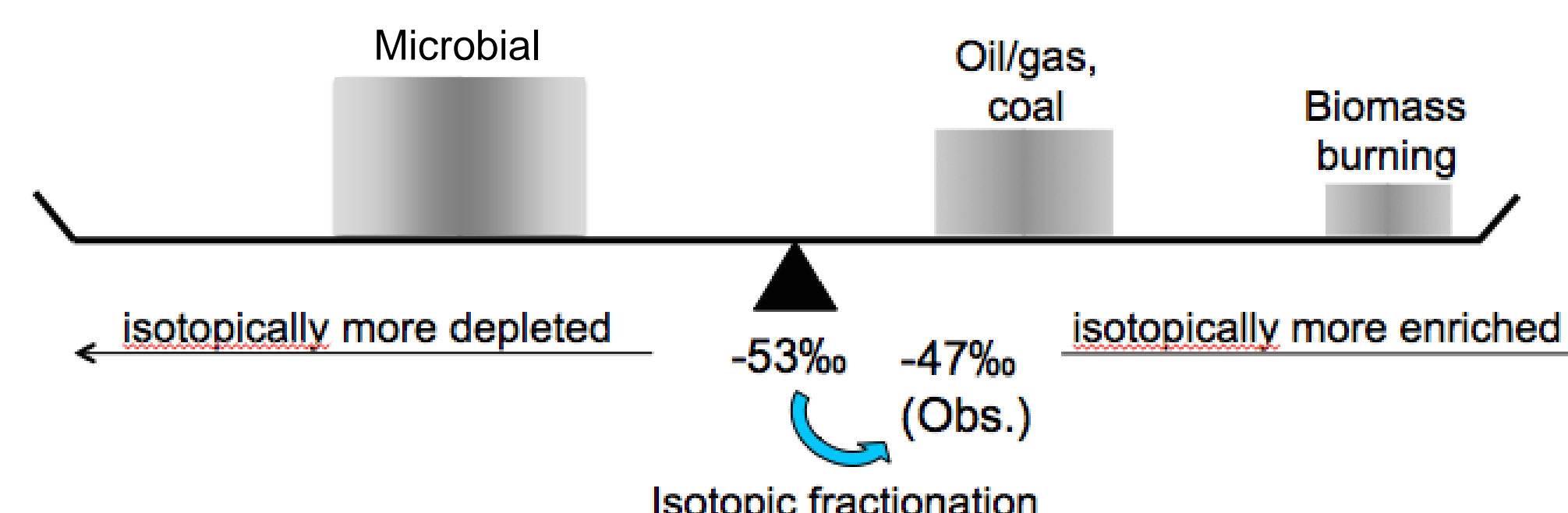
**Figure 2:** Illustration of uncertainties in attribution of global CH<sub>4</sub> sources. L to R: fossil fuels, ruminants, rice production, landfills, wetlands, and biomass burning.

## References

Dlugokencky EJ et al., (2011). Phil. Trans. Royal Soc. A369 (1943), 2058–2072.  
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Schwietzke et al., (2014). Environmental Science & Technology. 48(14): 7714-7722.  
We thank Martin Schoell, Guisepe Etiope and Edward Dugloencky for assistance with data.

## Why $\delta^{13}\text{C}$ end-members?

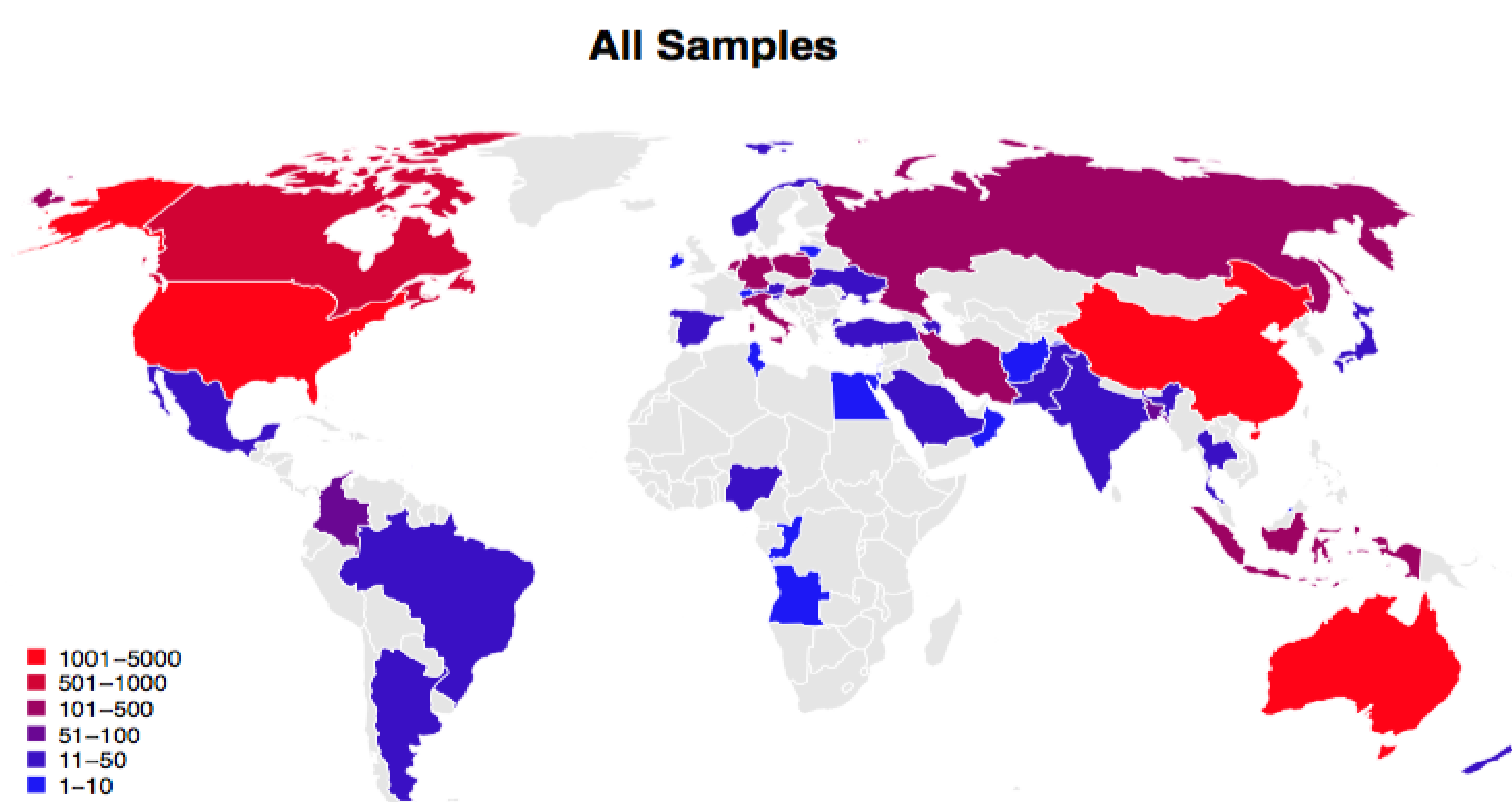
Measurements of  $\delta^{13}\text{C}$  can be used to constrain global methane sources (Fig. 1). However, the global-weighted end-member values are not well characterized. By knowing where the end-member values fall horizontally on the scale (Fig. 3), we can estimate how much each of the sources contribute to overall methane concentrations in the atmosphere.



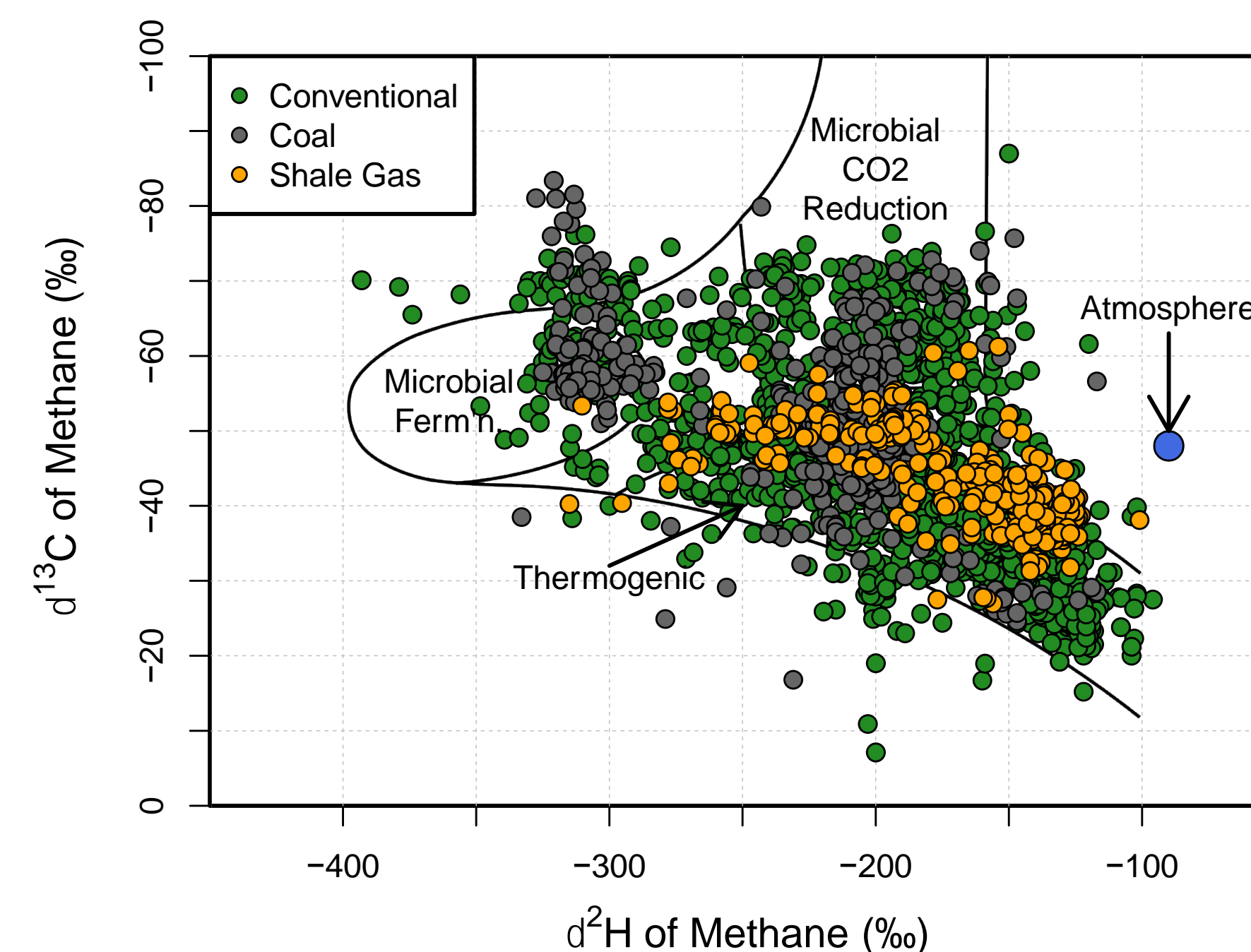
**Figure 3:** Mass balance of three CH<sub>4</sub> source categories. “Microbial” includes wetlands, ruminants, rice, landfills, and termites.

## Database

We compiled a global inventory of natural gas molecular and isotopic measurements from the peer-reviewed literature and government reports. The inventory contains data from 45 countries, 179 basins, 597 geological formations, and 10790 unique samples. On a country-level basis, the data represent 79% of world natural gas production.



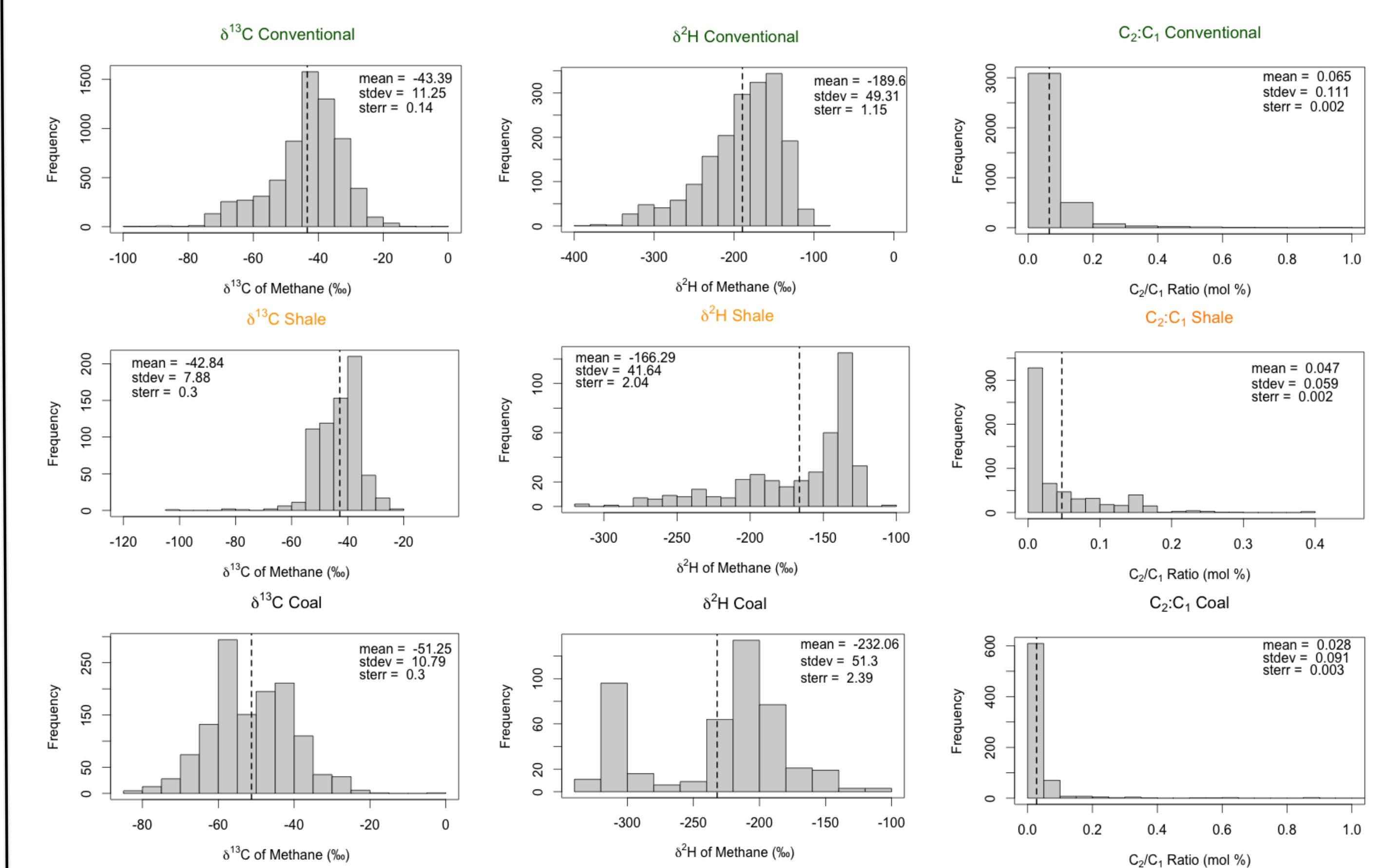
**Figure 4:** Global sample count of CH<sub>4</sub>- $\delta^{13}\text{C}$  for all types of natural gas and coal production.



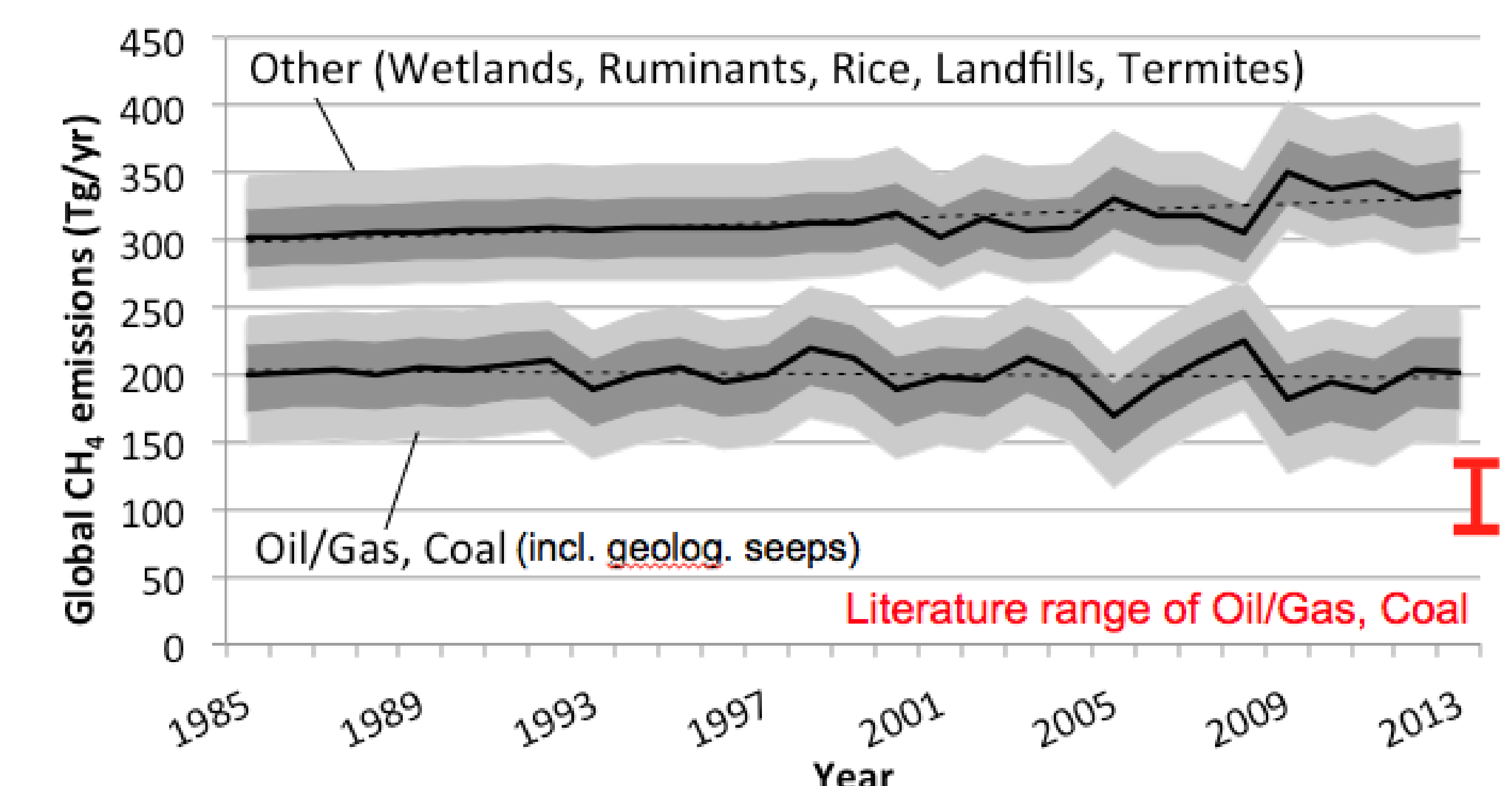
**Figure 5:** Generic classification of natural gases in the database, according to Whiticar (1999).

## Results

Data for different types of natural gas production (conventional, coal and shale gas) are presented in Figs. 5 and 6. Note how CH<sub>4</sub>- $\delta^{13}\text{C}$  values are skewed to more negative values, because of the importance of isotopically-depleted microbial gas. By weighting the data by continent-level gas production (BP, 2015) integrated over the years 2000-2014, we obtain a global, production-weighted average of  $-43.9 \pm 0.3$  ‰ (bootstrapped 95% confidence intervals) for the CH<sub>4</sub>- $\delta^{13}\text{C}$  of conventional gas. This value is considerably lower than the value (-40 ‰) typically used in global, top-down models of the global CH<sub>4</sub> budget. This could have a major consequence in methane emissions estimates (Fig. 7)



**Figure 6:** Visual representations of three analyzed parameters ( $\delta^{13}\text{C}$ ,  $\delta^2\text{H}$ , and C<sub>2</sub>:C<sub>1</sub>) and three types of production (conventional, shale, coal) in the global database. Statistical weighting of the geochemical parameters by basin-level production is still in progress.



**Figure 7:** Time series plot of modeled CH<sub>4</sub> emissions from different sources. The recent increase in global CH<sub>4</sub> can be attributed primarily to microbial emissions. Model adapted from Schwietzke et al. (2014) assuming -45 ‰ as the value for integrated fossil fuel methane emissions.