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# Stable isotopes provide revised global limits of aerobic methane emissions from plants

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## Abstract

Recently Keppler et al. (2006) discovered a surprising new source of methane – terrestrial plants under aerobic conditions, with an estimated global production of 62–236 Tg yr<sup>-1</sup> by an unknown mechanism. This is ~10–50% of the annual total of methane entering the modern atmosphere and ~30–100% of annual methane entering the pre-industrial (0 to 1700 AD) atmosphere. Here we test this reported global production of methane from plants against ice core records of atmospheric methane concentration (CH<sub>4</sub>) and stable carbon isotope ratios ( $\delta^{13}\text{C}_{\text{CH}_4}$ ) over the last 2000 years. Our top-down approach determines that global plant emissions must be much lower than proposed by Keppler et al. (2006) during the last 2000 years and are likely to lie in the range 0–46 Tg yr<sup>-1</sup>.

## 1 Introduction

Atmospheric methane (CH<sub>4</sub>) is an important greenhouse gas that impacts atmospheric chemistry and has almost tripled in abundance since pre-industrial times. The inclusion of large methane emissions from plants via an unknown biological production mechanism as proposed by Keppler et al. (2006) has important multidisciplinary scientific implications. Consequently the discovery is currently subject to methodological scrutiny and requires substantial experimental validation under realistic field conditions. The Keppler et al. (2006) methodology assumed that measured emissions from chambered plants were globally representative and scaleable to annual net primary production (adjusted for seasonal and daylight lengths for different plant types). Extrapolation of their bottom-up measurements resulted in large uncertainties and could overestimate global plant emissions. Recently reported direct measurements of methane production from plants have yielded estimates of 4–38 Tg yr<sup>-1</sup> in Brazilian forests (Carmo et al., 2006) and ~30–60 Tg yr<sup>-1</sup> in Venezuelan savannah and forests (Crutzen et al., 2006). While these estimates may also include some anaerobic methane emissions, they are com-

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parable to satellite measurements of tropical methane emissions (Frankenberg et al., 2005, 2006) and plant emissions reported by Keppler et al. (2006) if comparing tropical regions only. However, alternative calculations to extrapolate the Keppler et al. (2006) results to the global scale, which are based on foliage biomass and photosynthetic rates, estimate that global plant emissions are only  $\sim 10\text{--}60 \text{ Tg yr}^{-1}$  (Kirschbaum et al., 2006) and thus much lower than the  $62\text{--}236 \text{ Tg yr}^{-1}$  reported by Keppler et al. (2006). Although a prominent role of plant emissions in the pre-industrial atmosphere was proposed by Keppler et al. (2006), here we show that plant emissions are likely to be much smaller than they initially proposed and are not essential to close the isotopic mass balance of atmospheric methane.

## 2 Methods

To determine tighter limits on global plant emissions we first postulate fossil and biomass burning emissions in the pre industrial and modern eras then calculate anaerobic and aerobic sources to balance observed atmospheric composition. The atmospheric constraint is given by  $\text{CH}_4$  mass balance and stable carbon-isotope ratios ( $\delta^{13}\text{CH}_4$ ) over the last 2000 years recovered from ice core air bubbles (Ferretti et al., 2005). From independent assessments of fossil and biomass burning we take the lowest reported emissions to constrain the anaerobic/aerobic mix in our “Maximum Estimate” of the plant source, while higher and more probable fossil and biomass burning emissions constrain our “Best Estimate” of the plant source (see Table 1). We allow for  $\delta^{13}\text{CH}_4$  source signature variations and  $\text{CH}_4$  sink fractionation uncertainties (see Table 1).

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### 3 Results and discussion

Our results (Table 1) show that the “Maximum Estimate” of pre-industrial and modern global plant emissions are in the ranges 34–121 Tg yr<sup>-1</sup> and 0–175 Tg yr<sup>-1</sup>, respectively, lower than reported by Keppler et al. (2006). However, global biomass burning emissions at 1700 AD are unlikely to be as low as 5 Tg yr<sup>-1</sup> (Venevsky, 2006) and higher pre-industrial biomass burning and fossil emissions in our “Best Estimate” compare well with other studies (Ferretti et al., 2005; Houweling et al., 2000; Subak, 1994; Scheehle and Kruger, 2006). Thus our “Best Estimate” is a more reasonable methane budget reconstruction, suggesting that pre-industrial and modern plant emissions are more likely to be in the ranges 0–46 Tg yr<sup>-1</sup> and 0–137 Tg yr<sup>-1</sup>, respectively.

Large pre-industrial  $\delta^{13}\text{CH}_4$  variations have been partially explained by natural temperature and precipitation changes causing anaerobic and biomass burning emission variations (Ferretti et al., 2005). However, there is no evidence for temperature dependency of plant emissions over ambient ranges (~10–30°C), nor is there evidence for other climatic or anthropogenic influences on plant emissions during 1000 to 1700 AD, and the relatively small change in “Best-Estimate” plant emissions during 1000 to 1700 AD (see Table 1) is therefore expected. Thus, while Keppler et al. (2006) suggest that pre-industrial  $\delta^{13}\text{CH}_4$  variations (Ferretti et al., 2005) couldn't be reconciled by a wetland-dominated source, our analysis shows that a wetland-dominated pre-industrial source reconstruction with variable biomass burning emissions is more likely to have caused pre-industrial  $\delta^{13}\text{CH}_4$  variations than one controlled by large plant emission variations.

### 4 Conclusions

Our “Best Estimate” of the methane budget suggests that pre-industrial and modern plant emissions are likely to be in the ranges 0–46 Tg yr<sup>-1</sup> and 0–137 Tg yr<sup>-1</sup>, respectively. Therefore, while there is scope in the methane budget for plant emissions, they

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are not essential to reconcile either the pre-industrial or the modern methane budgets. Although this top-down approach allows increased plant emissions during the industrial era, modern plant emissions are likely to be lower than pre-industrial emissions due to the reduction in total biomass that has occurred from anthropogenic deforestation and land use change (Schlesinger, 1991). Therefore, during both the pre-industrial and modern eras, the best estimate of global plant emissions is likely to lie in the range 0–46 Tg yr<sup>-1</sup> and be at least 80% lower than proposed by Keppler et al. (2006). The good agreement between our top-down best estimate (0–46 Tg yr<sup>-1</sup>) and a bottom-up re-assessment of plant emissions (~10–60 Tg yr<sup>-1</sup> Kirschbaum et al., 2006) corroborates our conclusion that plant emissions are likely to be much lower than initially reported by Keppler et al. (2006). The plant source limits are most sensitive to the sink fractionation and if a larger magnitude fractionation is used (e.g. -7.4‰, Ferretti et al., 2005, which is consistent with a global chlorine sink of 25 Tg yr<sup>-1</sup> – see Allan et al., 2006) the upper limits of our best estimate of the plant source would decrease even further. Clearly, a lot remains to be learnt about the pre-industrial and modern methane budgets. Further field studies are needed to better define methane emissions from plants and new ice core records of carbon and hydrogen isotopes in atmospheric methane throughout the Holocene are required to better constrain the pre-industrial methane budget.

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**Table 1.** Upper limits of global CH<sub>4</sub> emissions from plants.

To calculate the “Maximum Estimate” of the plant source, we use lowest reported values of fossil and biomass burning emissions (see notes a and b). The plant source upper limits decrease further in the “Best Estimate” calculations where more likely values are used for fossil and biomass burning emissions (see notes c and d). A C<sub>3</sub>:C<sub>4</sub> plant type ratio of 60:40 is consistent with previous studies (Ferretti et al., 2005; Keppler et al., 2006) and with global  $\delta^{13}\text{CH}_4$  source signatures from biomass burning, plants, and anaerobic sources of  $-20\%$ ,  $-50\%$ , and  $-60\%$ , respectively. Anaerobic and aerobic plant emissions cover a range because we allow for: (i) uncertainties in the weighted-mean value of the CH<sub>4</sub> sink fractionation factor between  $-7\%$  and  $-5\%$  (Lassey et al., 2005); and (ii) variations in the C<sub>3</sub>:C<sub>4</sub> plant type ratio from 40:60 to 60:40 – causing weighted-mean  $\delta^{13}\text{CH}_4$  source signatures of biomass burning, plant, and anaerobic sources to vary by at most 2.6‰.

	Source Type	0 to 1000 AD emissions (Tg yr <sup>-1</sup> )	Changes 1000 to 1700 AD <sup>e</sup> (Tg)	1700 AD emissions (Tg yr <sup>-1</sup> )	2000 AD emissions (Tg yr <sup>-1</sup> )
Maximum Estimate	Fossil <sup>a</sup>	10	(0)	10	82
	Biomass burning <sup>b</sup>	10	(-5)	5	21
	Anaerobic	178–91	(+41)	222–128	437–262
	Aerobic plant	34–121	(-22)	9–103	0–175
Best Estimate	Fossil <sup>c</sup>	19	(0)	19	91
	Biomass burning <sup>d</sup>	25	(-10)	15	26
	Anaerobic	188–144	(+22)	212–166	423–286
	Aerobic plant	0–44	(+2)	0–46	0–137

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**Table 1.** Continued.

<sup>a</sup> The fossil source includes CH<sub>4</sub> emissions from natural geologic and ocean sources together with anthropogenic coal mining and energy use with a  $\delta^{13}\text{CH}_4$  signature of  $-40\%$ . We assume no anthropogenic fossil emissions in the period 0–1700 AD and take the lowest reported estimates of both natural (Judd et al., 1993) and anthropogenic (Scheehle and Kruger, 2006) fossil emissions.

<sup>b</sup> A lower natural limit of  $\sim 1.2 \text{ Pg C yr}^{-1}$  from lightning induced wildfires (Venevsky, 2006) translates to  $\sim 10 \text{ Tg yr}^{-1}$  of CH<sub>4</sub> from biomass burning, using an emission ratio of 162 mol CO<sub>2</sub>/mol CH<sub>4</sub> derived from Andrae and Merlet (2001). We ignore pre-industrial anthropogenic biomass burning emissions. Our lower limit of modern biomass burning emissions is determined by neglecting natural biomass burning emissions and assuming only anthropogenic emissions (Scheehle and Kruger, 2006).

<sup>c</sup> Here we use larger and more commonly reported values for natural fossil emissions (Houweling et al., 2000) although it is possible that natural fossil emissions could be much higher (e.g. Etiope, 2004). We use a conservative estimate for modern anthropogenic fossil emissions (Scheehle and Kruger, 2006).

<sup>d</sup> Here we use larger and more likely pre-industrial biomass burning emissions (Ferretti et al., 2005; Venevsky, 2006; Subak, 1994). We use conservative values for modern biomass burning emissions from natural ( $5 \text{ Tg yr}^{-1}$ ) and anthropogenic (Scheehle and Kruger, 2006) sources.

<sup>e</sup> These source changes are approximate and required to match the  $2\%$   $\delta^{13}\text{CH}_4$  depletion and the  $14 \text{ Tg}$  total source increase between 1000 to 1700 AD determined from ice core data (Ferretti et al., 2005). Note 0 to 1000 AD has a total source of  $232 \text{ Tg yr}^{-1}$  and  $\delta^{13}\text{CH}_4 \approx -49\%$ , 2000 AD has a total source of  $540 \text{ Tg yr}^{-1}$  and  $\delta^{13}\text{CH}_4 \approx -47\%$ .

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