# Anthropogenic Changes in Atmospheric Methane Concentration

## Overview

Methane is removed from the atmosphere in proportion to its concentration. Unless it is constantly re-supplied, it will exponentially decrease: $X_{CH4}(t) = X_{CH4}(t=0)\,e^{-t/\tau}$, where $X_{CH4}$ is the average volume fraction of atmospheric $CH_4$ and $\tau$ is the exponential decay constant (lifetime) of methane in the atmosphere. When constant (not varying), the atmospheric concentration of methane and $\tau$ determine the emission rate.

This relationship is remarkably useful. For example, the concentration of methane in the atmosphere, measured in dry air mole fraction or parts per billion by volume (ppbv) and known over the past 1000 years from air samples in ice and fern (snow) and from direct measurements, was constant at ~700 ppbv before 1750 AD, and again at ~1774 ppbv around 2000 AD. Taking $\tau$=9.5 years, we can immediately infer that the emissions rate was ~202 Tg $y^{-1}$ in 1750 and 554 Tg $yr^{-1}$ in 2000 AD, and that between 1700 and 2000 AD the emission rate of methane increased by 352 Tg $yr^{-1}$.

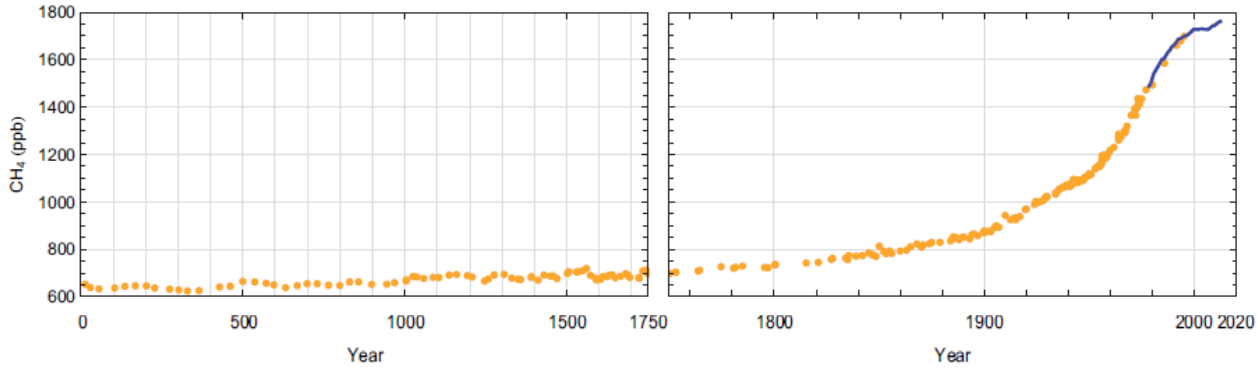


**Figure 1. Methane concentration in dry air mole fraction from Figure 6.11 in IPCC (2013). The atmospheric concentration of methane was constant at ~700 ppbv from 10 to 1750 AD. Over the industrial era it increased rapidly as the result of human-induced land use changes, animal husbandry, and combustion, The rise slowed in the 20th century, was briefly constant between 2000 and 2005 AD, and is presently rising again.**

The isotopic composition of atmospheric methane reflects the relative contribution of biogenic (which contain radiocarbon, e.g., $^{14}C$) and fossil sources (which contain no $^{14}C$), and from the changing proportions of $^{13}C$ in the atmosphere we can infer something about the mix of biogenic sources. Constraints are also provided by direct bottoms-up measurements and local top-down surveys.

Explaining the principles by which methane emissions rates can be estimated from atmospheric methane concentration, refining these estimates, comparing them to direct measurements, and establishing a basis for making future projections of methane emission rates are the purposes of the discussion here.

# Basic methods

## Theory

We obtain the most useful mathematical expression of methane mass balance by taking the derivative of the exponential equation given above:

(1) $$\frac{\partial X_{CH4}[\text{ppbv}]}{\partial t[\text{y}]} = \frac{-X_{CH4}[\text{ppbv}]}{\tau[\text{y}]} + A[\text{ppbvy}^{-1}].$$

Here $X_{CH4}[\text{ppbv}]$ is the concentration of methane in the atmosphere in parts per billion by volume, *t* is time in years, $\tau$ is the decay time of methane in the atmosphere in years, and *A* is the growth of methane in the atmosphere in ppbv per year.

- For a steady source of methane, the concentration of methane will increase until, in about 2$\tau$ years, its removal rate equals its supply rate, and its rate of change is zero. For $\frac{\partial X_{CH4}[\text{ppbv}]}{\partial t[\text{y}]}$=0 in equation 1. $A[\text{ppbvy}^{-1}] = X_{\text{CH4}}^{\text{steadystate}}[\text{ppbv}]/\tau[\text{y}]$ . In other words the emissions rate of methane equals the concentration of methane divided by methane's exponential decay time, $\tau$.
- If methane is increasing in the atmosphere, the emission rate is the sum of that needed to maintain the concentration of methane in the atmosphere and the observed rate of increase at that time: $A[\text{ppbvy}^{-1}] = X_{\text{CH4}}^{\text{steadystate}}[\text{ppbv}]/\tau[\text{y}] + \frac{\partial X_{CH4}[ppvv]}{\partial t}$.
- One ppb of methane in the earth atmosphere equals 2.75 Tg of methane (Tg= terra grams = $10^{12}$ grams of $CH_4$; note IPCC (2013) gives 2.7476 Tg/ppbv, Lassey et al., 2007 use 2.767, so the factor is not as tightly constrained as its decimal places would suggest).

## Example calculations:

1. In pre-industrial times, between 1500 and 1750 in Figure 1, the concentration of methane in the atmosphere was steady at ~700 ppbv. Taking $\tau$=9.5 years, the natural methane emission rate needed to sustain 700 ppbv is 700ppbv/9.5 years = 73.7 ppbv/year. At 2.75 Tg/ppbv methane the emissions rate is ~202 Tg CH4 per year.

2. The concentration of methane in the atmosphere was briefly constant at ~1774 ppbv between 2000 and 2005 AD (IPCC (2013) Table AII.1.1a). Taking t=9.1 years, we see the emission rate was 1774 ppbv/9.1 years = 195 ppbv/year or 535 Tg CH4 per year in this period.

3. In 2010 methane was not constant in the atmosphere, but increasing at a rate of ~ 5 ppbv or 13.7 Tg per year (IPCC, 2013, Figure 6.18). The concentration of methane in

the atmosphere in 2010 was 1795 ppbv, and thus the atmosphere was losing 197 ppbv or 541 Tg of methane per year. The total methane flux to the atmosphere was thus ~554 Tg CH4 per year.

4. If anthropogenic agricultural emissions are escalated in proportion to the human population, and fossil fuel energy emissions are escalated at 1.6% per year, 50 years from now anthropogenic the CH4 emission rate will 758 Tg $yr^{-1}$ and the steady state atmospheric methane concentration will be ~2500 ppbv (=758 Tg $yr^{-1}$ x 9.1 yr / 2.75 Tg/ppbv).

## Refinements

The emission calculated in (3) above is exactly that calculate for 2010 by Prather et al. (2012) and nearly the same as calculated by the IPCC (2013) in Table 6.8 for 2011. However, a good deal more can be learned by considering the isotopic composition of atmospheric methane following Lassey et al. (2007a).

### Changes in the turnover time of methane

There are many sources feeding methane to the atmosphere, but only one major sink: oxidation by the OH radical. OH is generated photolytically at a rate that is influenced by methane, volatile organic compounds, CO, and NOx. Thus, the atmospheric concentration of OH can be expected to change as the concentrations of these pollutants change. How the methane decay time is inferred to have varied with time is shown in Figure 2. Assuming the turnover time, $\tau$, is 7.7 years in 1750 AD and that the atmosphere contained 700 ppbv methane at that time, the total methane emission rate was 252 Tg $yr^{-1}$ (=2.767 Tg/pppv x 700pppv /7.7 y). If anthropogenic sources in 1750 were 30 Tg $yr^{-1}$, the natural sources were 222 Tg $yr^{-1}$. This is well within error estimate and just slightly more than the 202 Tg $yr^{-1}$ calculated above. A reasonable distribution for methane sources in 1750 is shown in Table 1.

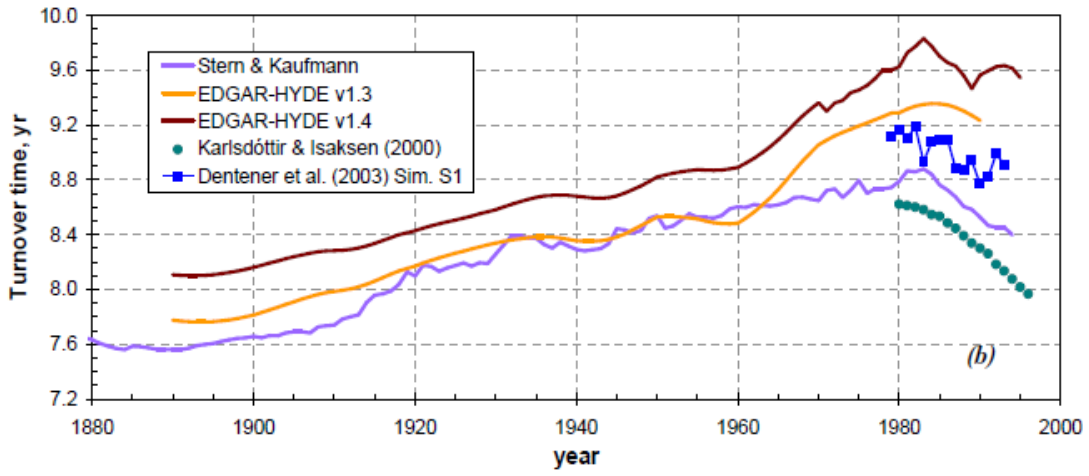


**Figure 2. Change in the methane decay time, $\tau$, (which is the same as the turnover time in this plot) as a function of time as inferred by Lassey et al., (2007a) from laboratory measurements and reconciliation of emission scenarios with the concentration history in Figure 1.**

**Table 1.** An indicative global inventory postulated for 1700 AD, and $\delta^{13}C$ assignments[a].

| Source component | Strength Tg $yr^{-1}$ | $\delta^{13}C$ ‰ |
|---|---|---|
| Natural sources | | |
| Wetlands | 163±78 | –60 |
| Termites | 20±10 | –57 |
| Wildfires | 5±0 | –25 |
| Oceans | 15±10 | –40 |
| Wild animals | 15±0 | –62 |
| Geologic | 4±0 | –40 |
| Natural subtotal | 222 | –57.4 |
| Anthropogenic sources[b] | | |
| Coal mining | 0 | –35 |
| Other fossil | 0 | –40 |
| Farmed livestock | 5±3 | –62 |
| Animal wastes | 0 | –55 |
| Rice cultivation | 10±5 | –64 |
| Forest burning, woodfuel | 5±3 | –25 |
| Savanna burning | 5±3 | –12 |
| Waste treatment, landfills | 5±5 | –55 |
| Anthropogenic subtotal | 30 | –47.0 |
| **Total source** | **252±34** | **–56.1±3.6** |

**Table from Lassey et al. (2007a) showing emissions at the start of the industrial era assuming human sources were 30 Tg $yr^{-1}$. The last column indicates the amount of $^{13}C$ in various sources, expressed as the per-mil (parts per thousand or ‰) deviation from the proportion of $^{13}C$ in a standard.**

## Constraints from the isotopic composition of methane

Methane is composed mainly $^{12}C$, but the atmosphere contains about 1% radioactive $^{14}C$ which decays to $^{14}N$ with a half-life of 5,700 years, and a small amount of $^{13}C$, another stable isotope. Taking into account radioactive decay, the masses of these isotopes are conserved exactly as described by equation 1 above. As indicated in Table 1, various methane sources contain very different amounts of $^{13}C$. Changes in the $^{13}C$ content of the atmosphere (from ice core samples for example) can therefore tell us how methane sources have changed with time.

The radioactive isotope of methane is supplied only by biological sources (burning of wood, methane generated by livestock, rice cultivation, termites, etc.), but there is a lag in this supply because atmospheric carbon can be trapped (in wood, for example) for some time before it is released as methane into the atmosphere. Geological methane sources (e.g., from the burning of fossil fuels or seeping from old strata in the subsurface) contains no $^{14}C$ because there have been geological amounts of time for it to completely decay. Thus $^{14}C$ can indicate how much fossil sources of "dead" methane add to the current mix of biological ("live") sources that are emitting $CH_4$ to the atmosphere in recent times. Working all this out requires finding the appropriate lag time, correcting for bomb $^{14}C$ and the ongoing supply of $^{14}C$ from nuclear reactors, and a chlorine reaction sink with a large isotopic fractionation. Details can be found in Lassey et al. (2007a). The bottom line is that isotopic modeling indicates that 30.0±2.3% of the global methane source between 1986 and 2000 was of fossil origin

(Lassey et al., 2007b). This is a 50% more than previously thought (18±9%), and suggests that natural fossil methane emissions could presently be between 40 and 60 Tg $yr^{-1}$, rather than ~10 Tg yr-1 as previously thought. Measurement of natural fossil methane leakage supports this suggestion (Etiope, 2008).

## Methane emissions from bottoms up measurements

Methane emissions can be measured directly using either "bottoms up" (summing emissions from all the steps in a process such as, for example, fracturing, well production, and leakage from compressors and valves) or "tops-down" (measuring the change in methane concentrations up- and down-wind of a source) methods. Some sources can be highly variable in time. For example, methane emissions from wetlands can vary dramatically from wet to dry years. Methane emissions estimated by these methods compare fairly well to those we have deduced from atmospheric chemistry using the methods discussed above.

Table 2 compiles anthropogenic emissions in 2010 from EPA (2013) and a worldwide assessment by EDGAR. The EDGAR estimates for the U.S. are only slightly different from the EPA (2013). EDGAR estimates the total world anthropogenic emissions in 2010 were 372 Tg, with the U.S. contributing ~6.7% of this total. Assuming natural emissions were the same as in pre-industrial times (222 Tg $yr^{-1}$), the total (anthropogenic plus natural) emissions is 594 Tg (= 372+222 Tg $yr^{-1}$). This is slightly larger than the 554 Tg $CH_4$ per year emissions estimated in (3) above, and mid-way between smaller top-down and larger bottoms-up estimates summarized in Table 6.8 of the IPPC (2013). The EDGAR world energy system emissions of 142 Tg $yr^{-1}$ are within the range of dead carbon emissions (130 to 180 Tg $yr^{-1}$) estimated by Lassey et al (2007b). The different emissions estimates agree reasonably well.

Table 3 groups emissions from Lassey et al. (2007a) and IPCC (2013) into live and dead carbon categories. The data is modified by us in a number of ways: First we add 50 Tg $yr^{-1}$ of geological dead carbon geological methane emissions to Lassey et al's estimate for 1750 (Table 1 above). This brings the 1750 geo-leakage to 54 Tg $yr^{-1}$, the same as IPPC (2013, Table 6.8) estimates for today. We do this because it makes no sense for the geo-leakage to be different in 1750 than today. We keep the wetlands emissions for today the same as in 1750 (163 Tg yr-1), and subsume IPCC lake and river emissions at the low end of their stated range into this wetlands category. The wetland emissions we adopt are at the low end of the top-down IPCC (2013) wetlands estimates, and slightly below the lower end of their bottoms-up estimates range. We average the median values of the IPCC (2013) agricultural sources (which have a relatively small range). The values summarized in Table 3 are reasonable given the uncertainties in the estimates. Table 3 shows anthropogenic methane emissions have increased from 10 to 55% of total methane emissions between 1750 and today. Fossil fuel emissions have increased from 0 to 17% of total emissions, and dead carbon emissions have increased from 18 to 27% in this period. With a turnover time of 8.75 years, a 566 Tg $yr^{-1}$ emission rate will support 1801 ppbv methane in the atmosphere, which is very close to the observed worldwide average 2011 level of 1803 ppbv.

## Future Prediction

Table 3 provides a basis for extrapolating methane emissions into the future. Assuming agricultural emissions increase in proportion to the human population (probably a worst case estimate because farming and animal husbandry methods could be adjusted to reduce methane emissions), agricultural $CH_4$ emissions would increase from 219 to 329 Tg $yr^{-1}$ as the human population increases from the present 7 billion to its peak of ~10.5 billion about 100 years from now. Fifty years from now the agricultural emissions will be ~297 Tg. Assuming fossil fuel emissions grow at 1.6% per year over the next 50 years as we proceed to supply 10.5 billion with a European level of power (7 kW $p^{-1}$), fossil fuel emissions will increase from 95 to 209 Tg $yr^{-1}$ fifty years from now and then decline. Thus 50 years from now the total anthropogenic emissions (agricultural and fossil) could reach ~506 Tg $yr^{-1}$ before they begin to decline. Adding the natural and geologic emissions, which we assume are unchanged at 198+54=252 Tg $yr^{-1}$, the total emissions are 758 Tg $yr^{-1}$. If the methane lifetime remains 8.75 years, the maximum (steady state) concentration that methane could reach in the atmosphere 50 years from now is ~758 Tg $yr^{-1}$ x 8.75 yr /2.75 Tg $ppbv^{-1}$ = 2412 ppbv. Under this plausible scenario methane will increase from current levels of 1803 ppbv by about 36%, and then decline to 2149 ppbv as fossil fuels are replaced by zero carbon fuels, as shown in Table 3.

The future projection in Table 3 is similar to that in the fossil fuel scenario we discuss here. In that scenario total human energy consumption in EJ $yr^{-1}$ is increased at 1.6% per year so that 100 years from now our 10.9 bn human population is supplied with 7 kW of total energy (the current energy consumption of the average Frenchman). Three different scenarios are examined: business as usual, substitute gas for coal and some oil, and proceed to low carbon fuels as fast as possible. In the business as usual scenario The methane emissions are the same as those in Table 3 for a worldwide methane leakage rate of 3.5% of consumption: is 95 Tg $yr^{-1}$ in 2011 (the same as in Table 3 for 2013), and 209 Tg $yr^{-1}$ 50 years hence in 2061.

## Useful Systematics and some discussion

Equation (1) is linear: twice the emission rate will produce twice the equilibrium concentration of methane. Only changes in methane emissions will count; emissions that are steady will have established an equilibrium concentration. We can consider changes from some specific date in terms of the changes in emissions from that date. For example, we can write:

(2) $$\frac{\partial X_{2011}[\mathrm{ppbv}]}{\partial t[\mathrm{y}]} = \frac{-X_{2011}[\mathrm{ppbv}]}{\tau[\mathrm{y}]} + A_{2011}[\mathrm{ppbv\,y^{-1}}],$$

where $X_{2011}$ is the change in atmospheric concentration in ppbv in 2011, when the volume fraction of methane was $X_{2011}$, and $A_{2011}$ is the rate of anthropogenic methane emission. In 2011 methane was increasing at 6 ppbv per year (Figure 1 and IPCC(2013, Table 6.8)) and the concentration in the atmosphere was 1801 ppbv. Thus, taking $\tau$=8.75 yr, from we see that $A_{2011}$=212 ppbv $y^{-1}$. The

equilibrium concentration of methane in 2011, $X^{ss}_{2011} = \tau \times A_{2011} = 1855\, ppbv$, which is 52 ppbv higher than the transient value of 1801 ppbv.

Future equilibrium atmospheric concentrations can be computed as changes from the 2011 equilibrium. For example, if between 2011 and 2061 AD the emissions rate increases by 192 Tg $y^{-1}$ (=192/2.75 = 69.8 ppbv $y^{-1}$) as shown in Table 3, the rise in equilibrium atmospheric methane will be 611 ppbv.

(3) $$\Delta^{ss}X_{2061\text{-}2011}[\text{ppbv}] = \tau[\text{y}]\left(A_{2061}[\text{ppbv y}^{-1}] - A_{2011}\right) = 611 \text{ ppbv}.$$

If the equilibrium concentration in 2011 AD is 1801 ppbv, the concentration in 2061 AD will be 2412 ppbv, as shown in Table 3.

We have implemented these methods to predict climate change from alternative future fuel use scenarios, using both accurate convolution methods (see here), and by making some simplifying simplifications which allow simple but very instructive excel spreadsheet calculation (see discussion here).

A point worth noting is that the 2005 anthropogenic emissions of fossil fuel methane of 62 $Tg_{CH4}$ $yr^{-1}$ calculated from fossil fuel use (assuming 0.18 L[%] $Tg_{CH4}$ $EJ^{-1}{}_{NG}$, 0.12 $Tg_{CH4}$ $EJ^{-1}{}_{coal}$, natural gas leakage of 2.5% of consumption (e.g., L[%]=2.5), and 110 EJ $yr^{-1}$ coal combustion and 110 EJ $yr^{-1}$ natural gas consumption, see Cathles 2012)) are about 65% of the 95 $Tg_{CH4}$ $yr^{-1}$ fossil fuel emissions estimated by Lassey et al. (2007a) in Table 3 below. The larger number assumed by Lassey could reflect natural or equilibrated human-induced methane leakage. Also, the reasons for the difference between the lifetime of methane in the atmosphere of 8.75 years determined by Lassey, and the lifetime used to assess greenhouse forcing by the IPCC(2013) of 12.4 years, is not clear to me at this point (see comments here).

A final point is that the natural methane sources can be quite variable from year to year, depending on rainfall and temperature, and the mix of methane sources is not certain. For example, it has been estimated that the worldwide flux of methane from the atmosphere from dam impoundments might be as large as 100 Tg $y^{-1}$, making it just bigger than the 2011 AD fossil fuel flux of 96 Tg $y^{-1}$ in Table 3).

**Table 2. U.S. anthropogenic CH4 emissions as reported by EPA(2013), and U.S. and world CH4 emissions as reported by Oliver and Janssens-Maenhout 2012 (indicated as EDGAR ). The percentage of world emissions represented by the U.S. for each category of emissions is indicated in the second to last column, and the percentage of total world emissions by the U.S. in each category is given in the last column.**

**Anthropogenic $CH_4$ Emissions of U.S. and World [Tg $y^{-1}$]**

| | -------U.S. 2010------ | | ----------World 2010---------- | | |
|---|---|---|---|---|---|
| | **EPA 2013** | **EDGAR** | **EDGAR** | **% U.S.** | **% U.S.** |
| **Energy Systems (44% U.S., 38% world)** | | | | of category | of total |
| Natural Gas Systems | 6.84 | | | | |
| Coal Mining | 3.45 | | | | |
| Petroleum Systems | 1.47 | | | | |
| Stationary Combustion | 0.30 | | | | |
| Abandoned Underground Coal Mines | 0.23 | | | | |
| Mobile Combustion | 0.09 | | | | |
| Total Energy | 12.38 | 10.05 | 141.9 | 7.1% | 2.7% |
| **Agricultural Systems (33% U. S., 43% world)** | | | | | |
| Enteric Fermentation | 6.54 | | | | |
| Manure Management | 2.48 | | | | |
| Rice Cultivation | 0.31 | | | | |
| Forest Land Remaining Forest Land | 0.68 | | | | |
| Composting | 0.07 | | | | |
| Total Agricultural | 10.19 | 9.31 | 161.4 | 5.8% | 2.5% |
| **Waste Management (21% U.S., 17% world)** | | | | | |
| Landfills | 4.90 | | | | |
| Wastewater Treatment | 0.77 | | | | |
| Field Burning of Agricultural Residues | 0.01 | | | | |
| Total Waste | 5.68 | 5.56 | 61.5 | 9.0% | 1.5% |
| **Other (1% U.S., 2% world)** | | | | | |
| Petrochemical Production | 0.15 | | | | |
| Iron, Steel, Metallurgical Coke | 0.03 | | | | |
| Total Other | 0.17 | 0.05 | 7.21 | 0.7% | 0.0% |
| **TOTAL ANTHROPOGENIC Tg $CH_4$ $y^{-1}$** | **28.4** | **25.0** | **372.0** | **6.71%** | **6.7%** |

Table 3. $CH_4$ emissions in 1750, 2011, 2061 (projected) and 2111 (projected). The 2011 emissions are the average of decadal figures from 1980 to 2010) from IPCC (2013 Table 6.8) except for the wetlands emissions which are from Lassey (1007) and include the IPCC wetlands and lakes and rivers categories. The 1750 emissions are from Table 1 above with a geologic dead carbon emission of 50 Tg $yr^{-1}$ added. The live carbon anthropogenic emissions are increased from 2011 in proportion to population increases. The dead carbon fossil fuel emissions in 2061 are increased from 2011 at 1.6% per year for 50 years preserving today's proportions of fossil fuel and zero carbon energy sources. Fossil fuel uses in 2111 is assumed to be the same as in 2011. Methane concentrations in the atmosphere are calculated using the methane turnover times indicated in parentheses in the last line. They are taken from Figure 2b of Lassey et al., 2007a, with a reasonable projection back to 1750.

| **Years AD** | **1750** | | **2011** | | **2061** | | **2111** | |
|---|---|---|---|---|---|---|---|---|
| **Human Population [bn]** | **0.125** | | **7** | | **9.5** | | **10.5** | |
| **Total Energy consumption [TW]** | | | **15** | | **33** | | **75** | |
| **Live Carbon Emission [$Tg_{CH4}$ $yr^{-1}$]** | | | | | | | | |
| **Natural Sources** | | | | | | | | |
| Wetlands (Lassey, 2007) | 163 | | 163 | | 163 | | 163 | |
| Termites | 20 | | 11 | | 11 | | 11 | |
| Wildfires | 5 | | 3 | | 3 | | 3 | |
| Oceans | 15 | | 6 | | 6 | | 6 | |
| Wild animals | 15 | | 15 | | 15 | | 15 | |
| **Total** | **218** | 72% | **198** | 35% | **198** | 26% | **198** | 29% |
| **Anthropogenic** | | | | | | | | |
| Farmed livestock | 5 | | 86 | | 117 | | 129 | |
| Rice cultivation | 10 | | 38 | | 52 | | 57 | |
| Burning, woodfuel | 10 | | 40 | | 54 | | 60 | |
| Waste treatment, landfills | 5 | | 55 | | 75 | | 83 | |
| **Total** | **30** | 10% | **219** | 39% | **297** | 39% | **329** | 49% |
| **Dead carbon Emissions [$Tg_{CH4}$ $yr^{-1}$]** | | | | | | | | |
| **Geologic** | **54** | 18% | **54** | 10% | **54** | 7% | **54** | 8% |
| **Fossil fuels** | **0** | 0% | **95** | 17% | **209** | 28% | **95** | 14% |
| **Total Emissions [$Tg_{CH4}$ $yr^{-1}$]** | **302** | | **566** | | **758** | | **676** | |
| **Atmospheric $X_{CH4}$[ppb]** | **725** | (6.6 y) | **1801** | (8.75 y) | **2412** | (8.75 y) | **2149** | (8.75 y) |

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