



GEM - Geologic Emissions of Methane, the missing source of the atmospheric methane budget

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Central to any study of climate change is the development of an inventory that identifies and quantifies natural and anthropogenic sources and sinks of greenhouse gases (GHG). Recent studies have demonstrated that geologic emissions of methane (GEM), although not considered in the inventories of the Intergovernmental Panel on Climate Change (IPCC), are an important GHG source. Etiope and Klusman (Chemosphere (2002), 49, 777-789) documented that significant amounts of methane, produced within the Earth crust, are released naturally into the atmosphere through faults and fractured rocks. Major GEMs are related to hydrocarbon production in sedimentary basins (biogenic and thermogenic methane), through continuous exhalation and eruptions from more than 1200 onshore and offshore mud volcanoes (MVs), through diffuse soil microseepage, and shallow marine seeps; secondarily, methane is released from geothermal and volcano-magmatic systems. Minor geologic sources are those related to natural exhalation from coal-bearing rocks (influenced by mining activities), degassing from crystalline basement and mantle. While marine seeps are studied since decades, methane flux from MVs has been object of detailed measurements only starting from 2001, when hundreds of gas flux measurements were performed from vents and soil in the main terrestrial MVs of Europe, in Romania and Italy (Etiope et al., Geoph. Res. Lett. (2003), 30, 1094, doi:10.1029/2002GL016287; and references therein). In 2003 gas flux was measured in Azerbaijan, which hosts the world's biggest MVs and densest MV population (Etiope et al., Geology (2004), in press). In all areas investigated around 10^2 - 10^3 tons of methane per km^2 are annually injected into the atmosphere. The global estimates of GEM from MVs range from 5 to 13 Tg y^{-1} (Etiope and Milkov, Environm.Geology (2004), in press).

It is known that methane flux in drylands is typically negative, in the order of -5 to $-1 \text{ mg m}^{-2} \text{ d}^{-1}$, due to soil methanotrophic consumption. Nevertheless, microseepage

can be higher than soil consumption and positive fluxes can occur, reaching tens or hundreds of $\text{mg m}^{-2}\text{d}^{-1}$ over wide areas in tectonically active and petroliferous regions. In MV zones microseepage reaches orders of $10^3\text{-}10^5 \text{ mg m}^{-2}\text{d}^{-1}$. Currently, soil-gas investigations are carried out in correspondence with deep gas reservoirs of the Corinth Gulf (Greece), where methane emissions are visible from seafloor pockmarks close to the coast, and of the Adriatic side of central Italy, where first data are suggesting a low (a few units to tens $\text{mg m}^{-2}\text{d}^{-1}$) but pervasive emission of methane throughout grasslands along belts of tens km^2 , characterised by neotectonic faults.

Potentially, microseeping areas are all the sedimentary basins with hydrocarbon generation processes at depth: this area was estimated to be around $43,366,000 \text{ km}^2$ leading to a global emission of methane of at least 7 Tg y^{-1} (Klusman et al., Assoc. Petrol. Geochem. Explor., (1998), 11, 1-55). Later, microseepage data from USA, former Soviet Union, Romania and Italy, and recent US Geological Survey GIS data-sets on the largest petroleum systems, were used to estimate a global microseepage area in the order of $8 \times 10^6 \text{ km}^2$ and a global output in the range $14\text{-}28 \text{ Tg y}^{-1}$ (Etiope, Annals of Geoph. (2004), in press). Finally, marine seeps can release globally about 20 Tg y^{-1} (Kvenvolden et al. (2001), EOS, 82, 457) and the geothermal flux around $2.5\text{-}6.3 \text{ Tg y}^{-1}$ (Etiope and Klusman, 2002, ibid). Accordingly the global GEM would amount to $40\text{-}60 \text{ Tg y}^{-1}$.

Thanks to all these new data is now possible to define better the role of GEM in the atmospheric methane budget. This budget seems to not be balanced, suggesting a “missing” source of methane as stated in early 1990s (Crutzen, Nature (1991), 350, 380-381).

The Second Assessment Report of IPCC-1996 defines the atmospheric methane budget by a bottom-up method, in which summing up the individual natural and anthropogenic sources gives the total emission into the atmosphere ($535 (\pm 125) \text{ Tg y}^{-1}$). Geologic emissions are not considered. The total sink was $560 (\pm 100) \text{ Tg y}^{-1}$ and the atmospheric increase $37 (\pm 3) \text{ Tg y}^{-1}$. So, the mean values of total source and sink (plus atmospheric increase) are not balanced.

The Third Assessment Report (IPCC-2001) derives the total methane emission by a top-down method, an inverse method considering a total burden of about $4,850 \text{ Tg}$, lifetime of 8.4 yr , and an imbalance of $+8 \text{ ppb/yr}$. The total source (burden/lifetime) is thus 598 Tg y^{-1} , the sink is 576 Tg y^{-1} and the atmospheric increase is 22 Tg y^{-1} . So, the total source is basically the quantity necessary to balance the sink plus increase, and is far from the sum of all the sources identified. IPCC-2001 reports however seven inventories of methane sources from works published from 1991 to 1999. The sum of the mean values of each source is 548 Tg y^{-1} , with an imbalance of

50 Tg y⁻¹ respect to the total sink plus atmospheric increase. Other bottom-up models also indicate a total source that is much lower than the total sink (e.g., Wuebbles and Hayhoe, *Earth-Science Rev.* (2002) 57, 177-210).

But there are large uncertainties in the individual source strengths and thus the imbalance refers only to the mean values. A more reliable indication of a “missing source” should be searched in radiocarbon isotopic studies. The atmosphere contains about 20% ($\pm 4\%$) of fossil, radiocarbon-free, methane. Thus with a total source of 600 Tg y⁻¹, the “old” methane contributes about 96-144 Tg y⁻¹ to the total methane emission. The best estimates for total anthropogenic fossil methane source (natural gas, coal mining and oil industry) are 74 Tg y⁻¹, indicated by EPA in 2001, following accurate bottom-up procedures and updated inventories from individual countries, and 89 Tg y⁻¹ suggested by Houweling et al. (*J.Geophys.Res* (1999), 104, 26137-26160) with an inverse modelling approach. All these estimates show that the amount of fossil methane in the atmosphere is higher than the amount expected from energy-related emissions, with an imbalance from 7 to 70 Tg y⁻¹. So, if this “hole” is real, where is the missing source?

It is evident that adding GEM (40-60 Tg y⁻¹) in the atmospheric methane budget, the imbalance between total source and sink resulting from bottom-up models is strongly reduced. Global GEM contributes to balance the isotopic budget being more than enough to provide the amount required to account for the suspected missing source of fossil methane. The global GEM estimates are of the same level of or higher than other sources or sinks considered in the IPCC tables, such as biomass burning (40 Tg y⁻¹), termites (20 Tg y⁻¹), oceans (10 Tg y⁻¹), soil uptake (30 Tg y⁻¹). GEMs represent the second most important natural source; it is more than 8% of the total emission, and 20% and 27% of the total natural sources considered by top-down (IPCC-2001) and bottom-up (IPCC-1996) models, respectively.

These results show clearly that GEMs, strictly controlled by geodynamic and tectonic processes, are not a minor source, as generally assumed in the past, but have a primary role in the atmospheric GHG budget, and cannot be disregarded anymore in the next IPCC assessment reports.