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Environmental Impact of Methane from Geogenic Sources*

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Abstract:
Recent studies have shown that methane emissions from geological sources can reach important values in sedimentary basins containing hydrocarbons. The flux of methane from soils situated in the perimeter of hydrocarbon bearing basins is greater than in regular soils. Invisible methane degasifications, known as diffuse soil microseepage from soils related to hydrocarbon production in sedimentary basins, increase the atmospheric methane budget. Methane is formed in a natural way, but it can also be the result of several human activities. In nature, this gas is formed as a result of many processes in the biosphere, atmosphere and geosphere.

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Methane – as a greenhouse gas

Methane is the third most important greenhouse gas responsible for the intensification of the greenhouse effect, after water vapours and carbon dioxide (CO_2), thus being among the gases accountable for the Planet's climatic changes. Methane has a global warming potential 25 times higher than carbon dioxide (GWP for a time horizon of 100 years). Its concentration is almost 200 times lower than that of the carbon dioxide. The lifetime of CH_4 in the atmosphere varies between 7–12 years, the mean lifetime being around 8.4 years.

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The atmospheric abundance of methane has had different values in the course of time, significant increases being recorded in the atmosphere parallel with the economic progress of society. Between 1000 and 1800 A.D., the methane abundance had stable values around 695 ppb. Later, the methane abundance in the atmosphere increased by a factor of 2.5 as compared to that of the pre-industrial era, as the measurements of CH_4 in ice cores and firn show.[1]

Methane's globally averaged atmospheric surface abundance continued to increase from approximately 1520 ppb in 1978 to 1745 ppb in 1998,[2] reaching 1774 ± 1.8 ppb in 2005.[3]

The average growth rate of CH_4 in the troposphere was 4.9 ppb/year between 1992 and 1998, corresponding to an average annual increase of 14 Tg in the atmospheric burden. The total amount of methane in the atmosphere reached 4850 Tg in 1998[4] and 4932 Tg in 2005.[5]

During the last two decades the growth rate of atmospheric methane decreased, maybe due to the disequilibrium between sources and sinks, though the exact cause has not yet been discovered.[6]

Atmospheric methane sources

Atmospheric methane originates from both non-biogenic and biogenic sources. The main sources of atmospheric methane, representing more than 70% of the global total, are biogenic.[7] These sources include wetlands, rice cultures, ruminants, landfills, forests, the ocean and termites. Non-biogenic sources include emissions from mining activities and the burning of fossil fuels (natural gas, oil and coal);

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[1] D. M. Etheridge et al., "Atmospheric Methane between 1000 A.D. and Present: Evidence of Anthropogenic Emissions and Climatic Variability", *Journal of Geophysical Research*, 103 (1998), 15979–15993.

[2] IPCC 2001, *Climate Change 2001: The Scientific Basis.* Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, ed. J. T. Houghton et al. (Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, 2001), 244.

[3] IPCC 2007, *Climate Change 2007: The Physical Science Basis*. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, ed. S. Solomon et al. (Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, 2007), 541.

[4] IPCC 2001, *Climate Change 2001,* 248.

[5] IPCC 2007, *Climate Change 2007,* 541.

[6] IPCC 2007, *Climate Change 2007,* 541.

[7] IPCC 2007, *Climate Change 2007,* 539.

biomass burning; waste treatment and geological sources (fossil methane from natural gas seepage in sedimentary basins and from geothermal/volcanic systems).

Methane sinks

The main process responsible for destroying atmospheric methane is its reaction with tropospheric OH, which is the most significant process of sinking CH_4 in the atmosphere (~85–90%).

$$\mathbf{OH + CH_4 \rightarrow CH_3 + H_2O}$$

This way a mean global loss rate of 506 Tg (CH_4)/yr [1] or 511 Tg (CH_4)/yr [2] is obtained.

The remaining 10–15% of methane is divided between other sinks, such as:

- Reaction with Cl atoms in marine environments constitutes probably less than 2% of the total consumption.[3] This additional source is estimated to be approximately 19 Tg (CH_4)/yr by some authors.[4]
- Minor methane amounts are also consumed in the stratosphere through the reaction with OH, Cl, O (^{1}D), resulting in a loss of 40 Tg (CH_4)/yr.[5]
- In the soil, methane is consumed by the activity of methanotrophic bacteria ~ 30 Tg (CH_4)/yr.[6] The methane consumption in the soil is considered to be an entirely biological process,[7] several classes of aerobic (methanothropic) bacteria being responsible for it. Zhuang et al., 2008 estimated that the global methane sink in the soil was 35 Tg (CH_4)/yr in the 1990s,

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[1] IPCC 2001, *Climate Change 2001,* 250.

[2] IPCC 2007, *Climate Change 2007,* 542.

[3] H. B. Singh et al., “Tetrachloroethylene as an Indicator of Low Cl Atom Concentrations in the Troposphere”, *Geophysical Research Letters*, 23, (1996), 1531.

[4] IPCC 2007, *Climate Change 2007,* 541.

[5] IPCC 2001, *Climate Change 2001,* 248.

[6] IPCC 2007, *Climate Change 2007,* 542.

[7] C. L. Curry, D. Van der Kamp, “Modelling the Soil Consumption of Atmospheric Methane”, in: *Non-CO_2 Greenhouse Gases (NCGG-4)*, ed. by A. van Amstel (Rotterdam: Millpress, 2005), 361.

with a loss rate that increased with 0.01 Tg (CH_4)/yr during the last century.[1]

The annual sink rate of methane from the atmosphere (**Figure 1**) reaches the value of 576 Tg,[2] respectively 581 Tg.[3]

Geologic methane emissions

Methane emissions from different sources contribute to a global total with approximately 600 Tg/yr,[4] out of which 60% are due to human activities such as agriculture, the use of fossil fuels and waste deposits.

The total methane source (the ratio between burden and lifetime) is 598 Tg/yr, the sink being 576 Tg/yr, and the atmospheric increase resulting from the difference between the two values is 22 Tg/yr.[5]

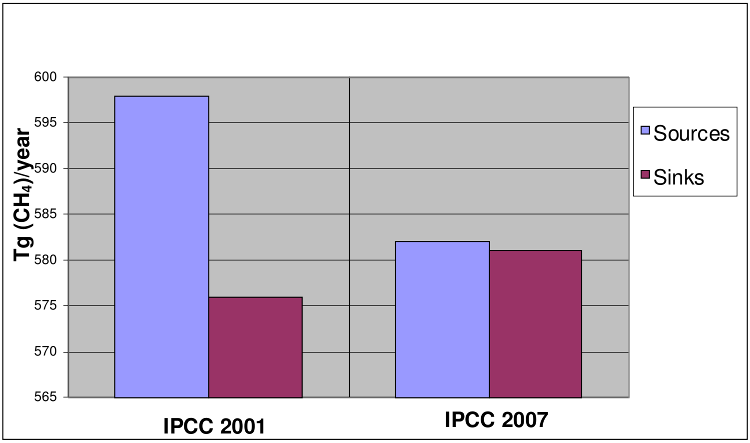

Fig. 1. Comparison between methane sources and sinks according to IPCC 2001; 2007

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[1] Q. Zhuang, K. Xu, Jinyun Tang, "Responses of Global Soil Methane Consumption to Changes of Climate, Land-use and Land-cover, and Atmospheric Chemistry Deposition during the 20th Century", *Geophysical Research Abstracts*, Vol. 10, European Geosciences Union, General Assembly 2008, Vienna, Austria, 13–18. April 2008.

[2] IPCC 2001, *Climate Change 2001,* 250.

[3] IPCC 2007, *Climate Change 2007,* 542.

[4] IPCC 2001, *Climate Change 2001,* 250.

[5] IPCC 2001, *Climate Change 2001,* 250.

Nevertheless, during 2000–2005 the average yearly emission was approximately 582 Tg (CH_4)/yr. The consumption of atmospheric methane was 581 Tg (CH_4) yr^{-1}, resulting in a methane surplus of only 1 Tg (CH_4)/yr.[1]

The estimations made by the Intergovernmental Panel on Climate Change in 2001 highlight the existence of a methane surplus which is attributed to no source. Therefore, this disequilibrium must be due to an important methane source, such as emissions from geogenic sources, not taken into consideration[2] until recently when the geological sources of methane were recognized as an important component in the atmospheric budget by the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (2007).

Estimations related to geological methane sources having an impact on the budget of this greenhouse effect gas have been made by researchers as Etiope and Klusman,[3] Judd et al.,[4] Etiope,[5] Kvenvolden and Rogers[6].

These studies suggest that significant amounts of CH_4, produced within the Earth's crust (mainly by microbial and thermogenic processes), are released into the atmosphere through faults and fractured rocks, mud volcanoes on land and the seafloor, submarine gas seepage, microseepage over dry lands and geothermal seeps. The emissions from these sources are estimated to be as large as 40 to 60 Tg (CH_4)/yr.

In 2007, the Intergovernmental Panel on Climate Change accepted the contribution of geogenic methane emissions among the sources which increase the global methane budget. The emissions originating from geologic sources are estimated to be between 40 and 60 Tg (CH_4)/yr.[7]

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[1] IPCC 2007, *Climate Change 2007,* 541.

[2] G. Etiope, "New Directions: GEM–Geologic Emissions of Methane, the Missing Source in the Atmospheric Methane Budget", *Atmospheric Environment* 38 (19) (2004), 3099–3100.

[3] G. Etiope, R. W. Klusman, "Geologic Emissions of Methane to the Atmosphere", *Chemosphere* 49 (2002), 777–789.

[4] A. G. Judd et al., "The Geological Methane Budget at Continental Margins and Its Influence on Climate Change" *Geofluids*, 2 (2002), 109–126.

[5] Etiope, "New Directions: GEM…", 3099–3100.

[6] K. A. Kvenvolden, B. W. Rogers, "Gaia's Breath – Global Methane Exhalations", *Marine and Petroleum Geology*, 22 (2005), 579–590.

[7] IPCC 2007, *Climate Change 2007,* 541.

Geogenic methane emissions in the Transylvanian Basin

Methane emissions from lithosphere to atmosphere (geogenic emissions) are common in hydrocarbon-prone areas or in geothermal areas. The Transylvanian Basin is well known for its highly purified methane reservoirs.[1] The methane related to gas reservoirs often finds ways to escape.

The processes that can be seen with the naked eye (mud volcanoes, everlasting fires, gas emissions in water) are named macro-seeps, while those that cannot be observed are called microseepages (diffuse emissions, small fluxes). The microseepages can be detected with the closed chamber technique, widely used for soil respiration or methane flux from wetlands or rice paddies.

The studies regarding the estimation of methane emissions having an impact on the environment and originating from geogenic sources in the Transylvanian Basin are quite new.

On relatively large surfaces that superimpose on Transylvanian hydrocarbon basins, there is a positive methane flux from the soil to the atmosphere. Macro-seeps connected to gas reservoirs are widespread in the basin.

The Everlasting Fires of Sarmasel (Mures County) are, by far, the most prolific single source of methane in the Transylvanian Basin. This single macro-seep injects hundreds of tones of methane into the atmosphere each year.

Deleni macro-seep (Mures County) is situated above one of the largest gas structures in the Transylvanian Basin. Salty water and gas are seeping in two main vent zones, producing swampy areas.

Methane emissions in water, situated in the Corund Creek on the SW of Salt Hill, have been studied at Praid (Harghita County).

More than 70 mud volcanoes are known in the Transylvanian Basin, and even if they are not as big or active as those existing in Berca-Arbănaşi area (Buzău County), they release an important methane flux into the atmosphere.

Some of the active investigated mud volcanoes that emit methane are located in Monor (Bistriţa-Năsăud County), Cobăteşti, Filiaş, Porumbenii Mici (Harghita County), Băile Homorod (Braşov County), Vlişoara (Mureş County) and Boz (Alba County).

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[1] C. Beca, D. Prodan, *Geologia zăcămintelor de hidrocarburi* (The Geology of Hydrocarbon Deposits) (Bucharest: Editura Didactică şi Pedagogică, 1983), 202.

All these combined locations release a positive methane flux into the atmosphere that cannot be neglected in the local budget of greenhouse gases emitted into the terrestrial atmosphere from a hydrocarbon basin.

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