

Methane emissions and global methane budget

Emisiile de metan și bugetul global de metan

MSc Manuel Valer HERLO^{1,a}, Pedro ALVES ^a

¹ Renewable Energy Specialist - OGAUS Technology, Arad, Romania

^a PhD candidate in Climate Change and Sustainable Development Policies - University of Lisbon, Portugal

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Abstract: The levels of greenhouse gases in the atmosphere are increasing since the industrial revolution. The main cause is associated to anthropogenic activities. Contrarily to what has been observed to atmospheric carbon dioxide levels, the evolution of the atmospheric methane concentration is still not fully understood. A relative stagnation of the methane concentration was observed from 2000 to 2006. After that, the levels of methane continue to increase. The global methane budget is assessed using two different approaches. It is verified the existence of inconsistencies between the results obtained from them. A better understanding of the methane cycle is needed to reduce the discrepancy and uncertainties associated to the methane sources and sinks and its quantification. This would allow a more reliable image of future scenarios.

Keywords: Methane emissions, Methane cycle, Methane budget

Rezumat: Nivelurile de gaze cu efect de seră din atmosferă sunt în creștere de la revoluția industrială. Cauza principală este asociată activităților antropice. Contrar celor observate la nivelurile de dioxid de carbon atmosferice, evoluția concentrației de metan atmosferic nu este încă înțeleasă pe deplin. S-a observat o stagnare relativă a concentrației de metan din 2000 până în 2006. După aceea, nivelurile de metan continuă să crească. Bugetul global al metanului este evaluat folosind două abordări diferite. Se verifică existența unor inconsistențe între rezultatele obținute din acestea. O mai bună înțelegere a ciclului metanului este necesară pentru a reduce discrepanța și incertitudinile asociate cu sursele și rezervele de metan și cuantificarea acestuia. Aceasta ar permite o imagine mai fiabilă a scenariilor viitoare

Cuvinte cheie: emisiile de metan, ciclul metanului, bugetul metanului.

1. Introduction

The majority of climate scientists (97%) agree that the primary cause of climate change, since the beginning of the industrial revolution, is due to human activities (anthropogenic activities) [1]. These activities are generally associated with the over-exploitation of ecosystem services and natural resources, for the sake of satisfying the excessive demand of goods and services. These actions lead to environmental

degradation and the emission of greenhouse gases to the atmosphere. Today, we listen a lot about the increase in carbon dioxide (CO₂) levels in our atmosphere/ However, this is not the only greenhouse gas that is increasing.

More powerful greenhouse gases (i.e., with higher global warming potential) like methane (CH₄) and nitrous oxide (N₂O) are also increasing. The atmospheric methane concentration varied during the time, rising from an estimated of 375 ppb 21000 years ago [2], to 772 +- 25 ppb before the industrial revolution. At that time, the methane production by the wetlands contributed to 70% of its total atmospheric level. After the year 2000 is considered that 50 - 70% of the total atmospheric methane is produced by anthropogenic activities [3].

Anthropogenic methane, mainly resultant from the extraction and use of fossil fuels, entered in the methane cycle after that period. Between 2000 and 2006, the levels of methane in the atmosphere stayed relatively constant. Some authors suggest that this plateau in atmospheric methane levels may be related to an increase in hydroxyl radical (•OH) concentration, the main sink of methane, in the atmosphere. However, from 2006 up to now, measurements have shown the rise of the atmospheric methane levels. Studies suggest that the increase in methane levels may be related with an increase in fossil fuel emissions [4] and with the expansion of tropical wetlands as well as the intensification of agricultural activity after 2006 [5]. The increase in the atmospheric methane levels has also contributed around 17% of the radiative forcing from well-mixed greenhouse gases since 1750 [2]. However, contrarily to the carbon dioxide case, the exact drivers of the global methane concentration growth are still debated [6]. The carbon dioxide cycle is already well known. On the other hand, the full picture of the methane cycle is still not completely understood. A better comprehension of the sources and mechanisms which increase the concentrations of methane in the atmosphere translates in a better comprehension of the methane cycle, which in turn is fundamental to reduce the uncertainties in the global methane budget, allowing to trace more plausible scenarios of methane future emissions.

2. The greenhouse effect and radiative forcing

Since the last seventy decades, greenhouse gases (GHGs) start to become more famous due to the global warming discussion. The most well-known greenhouse gases are the carbon dioxide, methane, nitrous oxide, ozone, and water vapor. The molecular structure of these gases allows them to vibrate at different frequencies within the infrared (IR) spectrum. Therefore, these gases are allowed to interact with infrared radiation, i.e., they will absorb and emit this type of radiation. It is fundamental to have these gases in the atmosphere (troposphere and stratosphere).

Without them, the planet would be colder [7] (average surface temperature of 255 K) and would not have the ozone protective layer against ultra-violet radiation. The radiation emitted by the Sun is the main contributor to the warming of the planet. The presence of greenhouse gases in the atmosphere is another factor that contributes to warm the planet. These interact with the infrared radiation emitted by the Earth's surface. Part of that radiation goes to space, while another part is absorbed and re-emitted by the greenhouse gases back to the Earth. This fraction warms the planet.

This effect is called the greenhouse effect (GHE), which was firstly proposed in 1824 by Joseph Fourier. The presence of the greenhouse gases at the atmosphere, establishes an energetic imbalance since the energy flux entering the planet is higher than the energy flux leaving it. This difference is called radiative forcing and is, typically, given in units of Wm^{-2} . This quantity may be positive or negative depending if the energetic flux entering is higher or lower compared to the energetic flux leaving the planet, respectively. Greenhouse gases are considered a climate forcing factor (climate driver) since they influence this balance, and so Earth's climate. The climate forcing factors can have a natural or anthropogenic origin. Natural climate forcing factors are, for example, the variations in Sun's energy output, variations in the Earth's orbital parameters (Milankovitch cycles), emission of particles to the atmosphere resulting from volcanic eruptions. Anthropogenic ones are like the emissions of greenhouse gases to the atmosphere and land-use change [8].

Like the other greenhouse gases, methane is a colourless and odourless gas that is found in nature, as a product of nature or as a product of human activity. The methane is lighter than air, having a lower density than normal atmospheric gases. In the following points some facts about the methane are given:

- after the carbon dioxide (60%), methane is the second main responsible for global warming (17%) [9];
- it has a smaller lifetime (time it takes for a molecule of to be removed from the atmosphere) in the atmosphere (12 years) compared with carbon dioxide (>100 years);
- its concentration in the atmosphere is about 220 times smaller than the concentration of carbon dioxide;
- it has a global warming potential (GWP), i.e., effectiveness in absorbing infrared radiation, about 28 times higher than carbon dioxide in an interval of time of 100 years [10].

3. Methane cycle

Knowing what are the sources and sinks of methane is fundamental to compute the methane budget, try to understand what was the leading cause of the increase of the methane levels in the atmosphere since 2005, as also as to make predictions about possible future scenarios.

3.1 Methane sources

As for the carbon dioxide greenhouse gas, methane production may result from natural or anthropogenic processes. Natural sources of methane are wetlands, or other inland water systems, offshore and onshore geological sources, biomass and biofuel burning, termites, wild animals, oceans, and permafrost. The methane is mainly produced through the decomposition of organic matter by bacteria when submitted to proper conditions like in anaerobic environment, specific temperature and pressure, and wet conditions. The quantity and efficiency in producing methane depend on these conditions as well as in the available amount of organic matter. However, recent studies have discovered that methane can also be produced in aerobic environment in a

near- ambient condition [11]. Additionally, human activities like agriculture, extraction and use of fossil fuels, livestock, waste storage and incomplete combustion of biomass are some of the main anthropogenic sources of methane.

Contrarily to carbon dioxide production, the main process behind methane comes from biotic processes. These processes are still not yet fully understood. An example is the "ocean methane paradox" [12], that states that the surface of oceanic waters are supersaturated with the greenhouse gas, although most bacteria that can generate the gas can not survive in oxygen-rich surface waters. New evidences suggest the possibility of methane production at well-oxygenated zones, in the sea and lakes, typically close to the water-air interface.

3.2 Methane sinks

Methane can be removed from the atmosphere primarily in two ways. The first and main process to sink the methane from the atmosphere results from the interaction between methane and the hydroxyl radicals. These reactions occur mainly in the troposphere (88%) and much less frequently in the stratosphere (7%) [3]. Depending on the compounds presented in the atmosphere, ozone, water vapour and carbon dioxide may result from secondary reactions triggered by the initial reaction between methane and the hydroxyl radicals. Therefore, the remove of methane from the atmosphere is highly dependent on the hydroxyl radical concentration. If hydroxyl radical concentration is lower/higher the atmospheric methane concentrations will tend to be higher/lower. Some authors suggested that the stagnation observed in atmospheric methane levels were a result of an increase of the hydroxyl radicals levels at the atmosphere [3]. Given the role of the hydroxyl radical in the methane cycle, is fundamental to understand what are the factors which influence its concentration. Various studies affirm that hydroxyl radical concentrations are dependent on the flux of ultraviolet radiation, on the water vapour concentration, as well as nitrous oxide and carbon dioxide emissions. Given the highest ability of the troposphere to remove the methane, its importance as chemical composition, especially the hydroxyl concentration is understandable. The second main sink of methane (5%) are the soils. The methanotrophic bacteria living in the soils can remove methane from the atmosphere by oxidising it, in a process known as "high-affinity methane oxidation", to produce energy. However, the contributions of methane sources and sinks to the computation of the methane budge still have several uncertainties associated, since neither the biotic processes associated to methane production nor the sink in the atmosphere are still not fully understood, what makes the methane cycle assessment a real challenge.

4. Global methane budget

The measurement of the total methane emissions and the concentration in the atmosphere is essential for calculating the global methane budget. A global methane budget is an important component in understanding and quantifying the methane sources and sinks and assessing the concentrations of the gas in the atmosphere. A higher concentration of methane in the atmosphere results in a higher radiative forcing, which results in warming of the climate. A global methane budget is also a

powerful tool for designing realistic pathways to mitigate climate change. Studies [3] have tried to quantify the total budget, using different approaches, but because of the uncertainties in the methane formation and emissions, the methods used to assess them had led to different results. The whole processes that influence the methane formation and the sinks are still under debate.

4.1 Measurement approaches

To calculate the global methane budget, the total methane emissions are measured. The measurements are also useful in assessing the reduction potential, by identifying the sources and the amount of greenhouse gases that can be avoided. Methane emissions are measured using two different approaches [13]: top-down and bottom-up and the most common techniques. Both top-down and bottom-up measurements are useful in calculating the total emissions and can be used in assessing from a small-scale individual source to large-scale global assessment, over short timescale or for a total annual. The large-scale global assessments involve the use of models and assumptions, and considers the emissions from all the sources, while small-scale local emissions are calculated from source-specific data and the measurements may not account for all the sources. Top-down techniques are mainly used for large-scale emissions assessment and bottom-up techniques are mainly used for specific local or facility area emissions assessment.

4.2. Methane budget

The global methane budget for year 2008 - 2017, was estimated at 572 Tg CH₄/yr (a range between 538–593 taking into account uncertainties) with the top-down approach and at 737 Tg CH₄/yr (a range between 538–880 taking into account uncertainties) with the bottom-up approach [3]. The difference in the global methane emissions between the top-down approach and the bottom-up approach is mainly because of the uncertainties in the bottom-up approach, with overestimating emissions from the natural sources. Based on the top-down approach, from the total budget, 60% are attributed to anthropogenic sources mainly from fossil fuels and 40% are attributed to natural sources. However, in the bottom-up approach, the ratio between anthropogenic and natural sources is balanced (50%-50%). The natural emissions are dominated by natural wetlands and other inland water systems. Uncertainties are smaller for anthropogenic sources than to natural sources. One of the largest uncertainties in the assessment of the methane budget is associated with the mechanisms of methane formation from the natural sources, mechanisms which are not yet fully understood. However, the anthropogenic sources have also a grade of uncertainty, for both unintentional and intentional emissions, like leaks from fossil fuel production or agriculture emissions.

Future trends are difficult to anticipate, not only because of the uncertainties in the natural processes of methane, but also because of uncertainties in future trends of human behaviour. The increase in population and the increase in energy and food demand, or the climate change impact like global temperature increase and rising ocean levels can make it much more difficult to predict the upcoming methane levels

and concentration in the atmosphere and its future impact on the global methane budget.

5. Final remarks

According to the current assessment, it was noticed that the methane role in climate science is still under many uncertainties and still a lot of gaps are to be filled with future research. Many of the gaps are related to the knowledge of the methane cycle, mainly the methane emissions from different sources and the quantification, and also the methane removal from the atmosphere. This creates further uncertainties for the computation of the global methane budget. To assess the global methane budget, different approaches are used, which can result in different conclusions, dependent on the given situations. A top-down approach is favourable compared to a bottom-up approach when a regional area needs to be assessed. A top-down approach can give a broader view of the methane emissions in the atmosphere and it is based on inversion modelling. Inverse modelling is a reliable technique for temporal and spatial assessment of atmosphere methane emissions [13]. A bottom-up approach is preferable for local assessments, and it is based on estimations and representative samples, which can have a high level of uncertainty. Because of the existence of a knowledge gap in the methane cycle, the assessment of the specific sources or the global methane budget will have discrepancies between the two different approaches. An alternative strategy is the use of a mix approaches, in order to optimize the methane assessment process. Also, a proper assessment of the anthropogenic sources, using a suitable approach, it will be possible to have a better picture of the nature contribution to methane emission in the atmosphere. To have a more exact global methane budget, reducing the uncertainties is a priority. Having a clearer picture of the global budget will show the total methane emission level that cannot be exceeded, in order to keep a balance between the sources and the sinks. Uncertainties can be reduced for both anthropogenic and natural methane sources. Methane emissions from natural gas and oil are equal to 6% of the total greenhouse gas emissions in the energy sector, mainly caused by deliberate or accidentally leaks [14]. Taking into account that natural gas demand and oil demand increased and it is expected to increase further more [15], makes it important to develop a proper mechanism in order to detect and reduce the amount of leaks from the fossil fuel sector which will reduce the uncertainties in the methane emission assessment. The contribution associated to the natural methane sources is related to biotic processes. Uncertainties are high because of the lack of a complete understanding of the natural processes which lead to methane emissions. A better assessment of the aerobic and anaerobic processes in different environments is required.

Also, there are uncertainties in the atmospheric chemistry. The concentration of hydroxyl radicals influences the ability of the atmosphere to reduce the methane levels and the variation of hydroxyl in the lower levels of the atmosphere is not well known.

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