The Influence of Mine Water Rebound on Methane Degassing in Abandoned Coal Mines

Stefan Möllerherm, Christian Melchers

Research Center of Post-Mining at the Technische Hochschule Georg Agricola University, Herner Straße 45, 44787 Bochum, Germany, Stefan.moellerherm@thga.de

Abstract

Hard coal mining is one of the major source for anthropogenic methane emissions. Even after mine closure, methane is still released over a longer period. Experts assume that methane degassing will gradually drop due to the water rising because hydrostatical pressure will reduce the desorption from the seams. However, some scientists have already proved that mine water can contain methane-producing bacteria. This secondary methane geneses potential has not been analysed since then. The authors postulate that bacteria can generate recent methane even in mine water rebound processes. In this paper, they describe their approach to support this thesis.

Keywords: Methane Emission, Abandonment Mines, EU Methane Strategy, Bacteria

EU legislative background

Methane is a powerful greenhouse gas, second only to carbon dioxide in its overall contribution to climate change. On a molecular level, methane is more powerful than carbon dioxide. Although it remains for a shorter time in the atmosphere, it has a significant effect on the climate. Reducing methane emissions therefore contributes to slowing down climate change. The EU Regulation on the Governance of the Energy Union and Climate Action [(EU) 2018/1999)] calls on the Commission to deliver a strategic plan for reducing methane emissions. Furthermore, in the European Green Deal Communication, the Commission indicated that energy-related methane emissions needed to be addressed as part of the commitment to reach climate neutrality by 2050. In this way, policy action to reduce methane emissions will contribute to both the EU's decarbonisation efforts towards the 2030 Climate Target Plan and the EU's zero-pollution ambition for a toxic-free environment [(EU) COM(2019) 640 final].

The EU has reduction targets for 2030 for all greenhouse gases, with anthropogenic methane emissions covered by binding national emission reduction targets under the Effort Sharing Regulation (ESR) [(EU) 2018/842]. However, there is currently no policy dedicated to the reduction of anthropogenic methane emissions. 59% are anthropogenic, of which the largest sources are agriculture (40-53%) – in particular linked to intensive production, fossil fuel production and use (19-30%), and waste (20-26%). In the EU, 53% of anthropogenic methane emissions come from agriculture, 26% from waste and 19% from energy [European Environment Agency (EEA), 2018].

In October 2020, the EU communicated a strategy on reducing methane emissions [(EU) COM(2020) 663 final]. It outlines a comprehensive policy framework combining concrete cross-sectoral and sector-specific actions within the EU, as well as promoting similar action internationally. While in the short-term, the strategy encourages global level voluntary and business-led initiatives to immediately close the gap in terms of emissions monitoring, verification and reporting, as well as reduce methane emissions in all sectors, it foresees EU level legislative proposals in 2021 to ensure widespread and timely contributions towards the aforementioned EU objectives. Now, the Commission is undertaking an open stakeholder consultation process to incorporate the different views and opinions of these parties. The new regulation on methane emissions shall be adopted by end of 2021.

Methane emissions from active and abandoned coal mines

The metamorphosis of terrestrial organic material like peat to coal and anthracite results in the release of water, carbon dioxide, hydrocarbons and nitrogen. Main component of the hydrocarbons is methane. Roundabout 150 – 200 m^3/t of the original organic material originated in the Carboniferous and Permian Age (Gaschnitz 2000). The gas was or is partially absorbed at the coal surface. This methane (also known as mine gas) was generated during the formation of the coal around 300 Mio years ago. Although a huge amount of methane has escaped during millions of years, still significant quantities of methane are stored in the coal deposit.

In active coal mines, methane which is absorbed in the seam and in the strata is released due to the coal extraction process such as longwall mining or room and pillar. The methane is then diluted by fresh air down to a maximum threshold of 1 Vol-% of CH_4 and sucked out of the underground mine by the ventilation air system. In those cases where it is technically not more feasible to dilute the methane to the threshold, the gas is directly drained from the seam or from the strata. The mining company drills boreholes into the strata, sucks the gas and transports it via underground gas pipelines to cogeneration plants on the surface. In these cogeneration plants, the methane is used as a fuel to produce electricity and heat. In the mining business, this type of gas is called Coal Mine Methane (CMM).

While closing a coal mine the release of mine methane will not stop in the underground. Huge amount of gas remains in the strata, in the non-mined areas of the seams and in the over- and underlying seams. Over many years, even decades after mining cessation the coal deposit releases gas into the remaining void, which consists of former galleries and mining-induced excavation-damaged zones. In the sealed areas of the mine, the present air is displaced and the oxygen content is reduced due to oxidation processes. Waterless voids of former coal mining galleries are filled up with oxygen-deficient gas mixtures of methane, nitrogen and carbon dioxide. In general, the content of methane and carbon dioxide will increase after termination of the galleries while at the same time the oxygen content decreases. The gas composition within a closed mine will balance in the long term due to different effects like natural draft or diffusion. Besides the former shafts cracks and fissures in the Coal Formation and in the strata provides another pathway for methane emissions (figure 1). These pathways were created by the

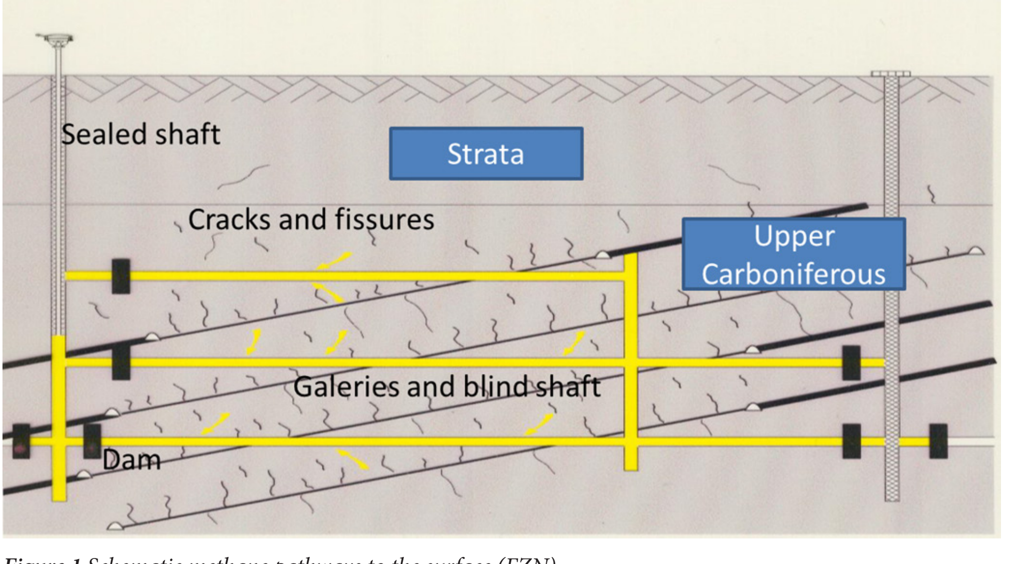

Figure 1 *Schematic methane pathways to the surface (FZN).*

coal extraction and the related deconstruction of the strata due to the ground subsidence.

Therefore, the mining companies at the Ruhr and Saar area are still able to drain methane from this reservoir and utilises the gas in the existing cogeneration plants. Experts call this type of methane Abandoned Mine Methane (AMM). According to the mine authority in North Rhine-Westphalia, roundabout 100 cogeneration plants were running on closed mines in 2020. With an installed electrical capacity of 168 MW 544 Mio kWh of electricity and 108 Mio kWh of thermal heat were produced last year (Wissen 2021).

(Kholod *et al*. 2020) have modelled the global methane emissions from abandoned coal mines. They conclude that the gas from these mines will continue to increase within the next decades even though coal mines will close in the future. Their model estimated global AMM emissions to be 22 bcm in 2010 and forecasts AMM emissions to increase to 75 bcm in 2050 and 162 bcm per year in 2100.

Evolution of the mine degassing process in abandoned coal mines

Several scientists act on the assumption that the methane generation and release process will come to end with flooding the mine (Kholod *et al*. 2020, Imgrund 2020). The mine water rebound will lead to a gradual reduction of the methane release from the strata into the galleries because desorption from the seams will decrease due to the opposing hydrostatical pressure. They assume that the process stops as soon as the hydrostatical pressure exceeds the gas pressure, which correlates with the respective residual gas content of the coal. Furthermore, the water rebound can flood pathways in the galleries and hence interrupt them. This is in particular true for galleries, which were developed on deeper levels to connect different mines. Consequently, these connections between different parts of mines to degassing units or to the cogeneration plants will be inapplicable and hence cannot be degassed or drained any longer. In the course of the water rebound, the gas composition in the void can change too, because the water increase push away methane rich gas mixtures in other voids either in horizontal or vertical directions. This may lead to the fact that the methane content in voids in areas of low or gas free seams will increase. The barometric gas exchange between void and atmosphere will only terminate in case of complete flooding of the mine. Kholod *et al* assume that the gas release process will terminate approximately 6 to 7 years after flooding the mine (figure 2) (Kholod *et al*. 2020).

The diagenesis of coal is not the only source of methane generation. In many coal mining areas one can find evidence for recent methane genesis, which is not of geochemical origin (Rice 1993, Berner & Faber 1996, Zazzeri *et al*. 2016). Bacteria can generate CH_4 out of coal and water under anaerobe conditions. This process has

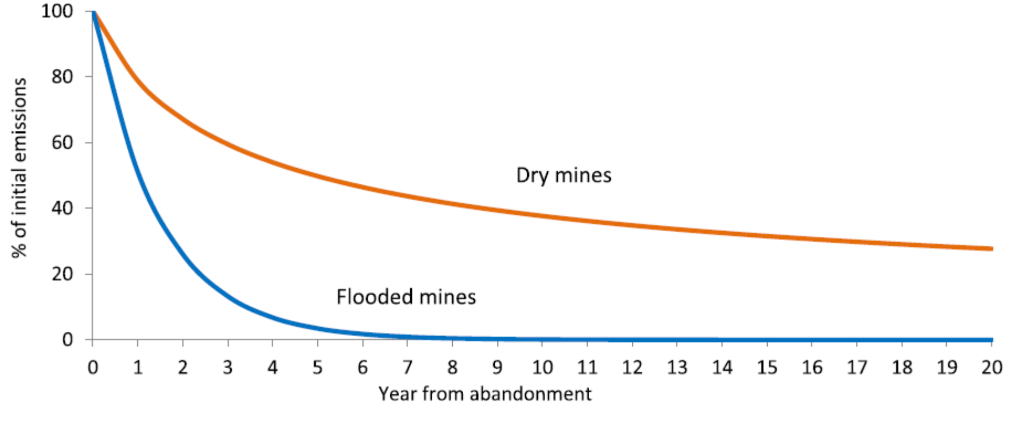

Figure 2 *Assumed AMM emission reductions over time from dry and flooded mines (Kholod et al. 2020).*

taken place also in the Ruhr area and in the Saar area (Thielemann *et al*. 2004, Krüger *et al*. 2008). Thielemann conducted first trials to demonstrate the microbial methane production in the Ruhr area (Thielemann *et al*. 2004). He took samples from 13 sites within 14 months and analysed the components and the isotopes. According to the isotope data, he showed that the methane gas was a mixture of thermogene and by CO_2 reduction generated microbial gas. (Krüger *et al*. 2008) found in mine water samples living methanogens archaea i.e. methane generating bacteria and proved the recent methane gas genesis in abandoned coal mines in the Ruhr area. Despite the indications on secondary genesis of methane, this assumption have not been investigated more in detail in science yet. In addition, a quantitative estimation of a bacterial source term has not been undertaken yet as well. Of particular interest would also be if the mine water rise would lead to an increase of recent methane genesis. Against this background, scientists at the Research Center of Post-Mining in Bochum have initiated the research project "The influence of mine water rebound on methane degassing in abandoned coal mines". The project is conducted in collaboration with the coal mining company RAG, the utility company STEAG and the Federal Geological Survey of Germany (BGR). STEAG is operating cogeneration plants in the former coal mining areas at Ruhr and Saar.

First research results

In 2019 and 2020, the scientists undertook gas-sampling campaigns at the cogeneration plants in the Saar and Ruhr area. The gas from the underground pipeline to the cogeneration plant was sucked via a small injection needle into a vial. At each site, two samples were taken in order to minimize measurement errors. In the Saar area two measurement campaigns at 13 sites have been undertaken, one in 2019 and another one in 2020. At the Ruhr at 29 locations samples have been taken in 2020. Due to the pandemic situation, it was not possible to carry out more campaigns.

Afterwards BGR performed an isotope composition analyses and determined carbon and hydrogen isotopes. The following

***Table 1** Measurement results from the Saar area.*

Site	$\partial_{13}C\ CH_4$ [‰ PDB]	microbial $\delta^{13}C$-CH_4 -80	thermogen $\delta^{13}C$-CH_4 -25	CH_4 (%) 2019	Volume flow (m^3) 2019
Allenfeld	-42,5	44%	56%	75,68	3.789.260
	-43,3	46%	54%		
Altenkessel	-46,4	54%	46%	26,10	1.586.552
	-46,7	54%	46%		
Reden	-41,9	42%	58%	25,52	21.763.508
	-41,9	42%	58%		

***Table 2** Measurement results from the Ruhr area.*

Site	$\partial_{13}C\ CH_4$ [‰ PDB]	microbial $\delta^{13}C$-CH_4 -80	thermogen $\delta^{13}C$-CH_4 -25	CH_4 (%) 2018	Volume flow (m^3) 2018
Blumenthal 3/4	-52,5	69%	31%	15,6	28.301.287
	-52,6	69%	31%		
Hugo 2/5/8	-56,0	78%	22%	18,8	28.463.241
	-54,5	74%	26%		
Minister Stein 4	-54,5	74%	26%	43,5	10.251.754
	-54,4	74%	26%		

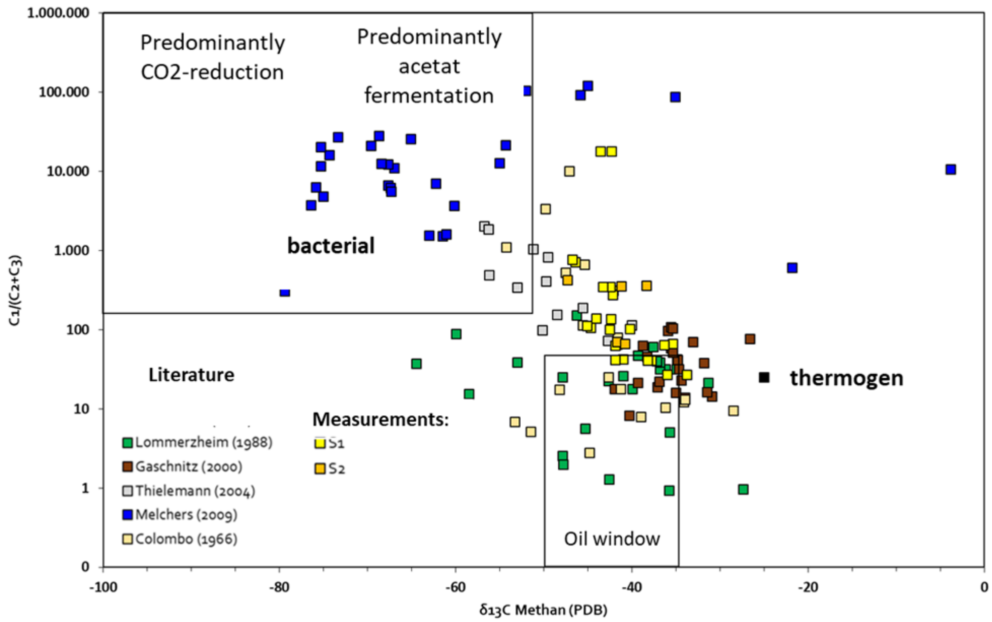

Figure 3 *Plot of the measurement results for the Saar area (FZN).*

tables shows the results for the Saar and for the Ruhr area. (–80) is the end link of the bacterial isotopes and (–25) is the end link of thermogeneous isotopes. It can be seen that the part of microbial isotopes is slightly increasing which is an evidence that bacteria are generating recent methane, in particular in the Ruhr area. Here the percentage of microbial isotope is much higher compared to the Saar.

Afterwards the results were plotted and compared with values taken from literature. Figure 3 shows the results for the Saar area. The x-axis in the chart indicates the $\partial_{13}C$ in methane and the y-axis the ratio of C_1 (methane) to $C_2 + C_3$ (ethane and other hydrocarbons). Thermogeneous methane was generated in the carboniferous era as coal gas. On the other hand bacterial origin refers to the fact that the gas is generated by bacteria which then can continue to exist. The yellow and green dots in the plot represent the respective measurements of the first two campaigns (S1 and S2) in the Saar region. Thermogeneous origin of methane is dominating in the Saar area, however the main number of analyses are close to the transitional phase.

Summary and outlook

The results of the first isotope analyses underpins the assumption that bacteria can generate recent methane in abandoned coal mines. This can be seen in the Ruhr area, where the portion of microbial methane is higher than the portion of thermogen methane. On the other hand, due to pandemic it was not possible to carry out more sampling campaigns. Another one is currently performed at the Ruhr area. Although the contribution to the overall methane degassing from closed mines is relatively low, however, it cannot be neglected and it will continue to be a source of methane in dry mines. More data are required and additional research is needed to quantify the amount of recent methane generation.

Acknowledgement

The authors want to thank RAG-Stiftung for the financial contribution and the partners RAG, STEAG and BGR for their collaboration.

References:

Berner U, Faber E 1996: Empirical carbon isotope/ maturity relationships for gases from algal kerogens and terrigenous organic matter, based on dry, open-system pyrolysis. In: Organic Geochemistry 24 (10-11), S. 947–955. DOI: 10.1016/S0146-6380(96)00090-3

(EU) 2018/1999: REGULATION (EU) 2018/1999 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 11 December 2018 on the Governance of the Energy Union and Climate Action, amending Regulations (EC) No 663/2009 and (EC) No 715/2009 of the European Parliament and of the Council, Directives 94/22/EC, 98/70/EC, 2009/31/EC, 2009/73/EC, 2010/31/EU, 2012/27/EU and 2013/30/EU of the European Parliament and of the Council, Council Directives 2009/119/EC and (EU) 2015/652 and repealing Regulation (EU) No 525/2013 of the European Parliament and of the Council

(EU) COM(2019) 640 final: COMMUNICATION FROM THE COMMISSION The European Green Deal

(EU) 2018/842: REGULATION OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 30 May 2018 on binding annual greenhouse gas emission reductions by Member States from 2021 to 2030 contributing to climate action to meet commitments under the Paris Agreement and amending Regulation (EU) No 525/2013

European Environment Agency (EEA), (2018). EEA greenhouse gas - data viewer. https://www.eea.europa.eu/ds_resolveuid/f4269fac-662f-4ba0-a416-c25373823292

(EU) COM(2020) 663 final: COMMUNICATION FROM THE COMMISSION TO THE EUROPEAN PARLIAMENT, THE COUNCIL, THE EUROPEAN ECONOMIC AND SOCIAL COMMITTEE AND THE COMMITTEE OF THE REGIONS on an EU strategy to reduce methane emissions

Gaschnitz R 2000: Gasgenese und Gasspeicherung im flözflihrenden Oberkarbon des Ruhr-Beckens. Dissertation RWTH Aachen

Imgrund T, Orzol R 2020: Gutachten zur Grubengasgewinnung in Nordrhein-Westfalen. PFG-Nr. 352019 https://www.wirtschaft.nrw/sites/default/files/asset/document/2020-06-0029877.pdf

Kholod N, Evans M, Pilcher Rc, Roshchanka V, Ruiz F, Cote M and Collings R.: Global methane emissions from coal mining to continue growing even with declining coal production, Journal of Cleaner Production (2020), doi:https://doi.org/10.1016/j.jclepro.2020.120489.

Krüger M, Beckmann S, Engelen B, Thielemann T, Cramer B, Schippers A, Cypionka H 2008: Microbial Methane Formation from Hard Coal and Timber in an Abandoned Coal Mine. In: Geomicrobiology Journal 25 (6), S. 315–321. DOI: 10.1080/01490450802258402.

Lommerzheim, A. (1988): Die Genese und Migration von Kohlenwasserstoffen im Münsterländer Becken. – 260 S + 112 S Anh.; 129 Abb., 28 Tab., 50 Taf.; 6 Karten; Münster. [Dissertation]

Melchers, CH. 2009: Methan im südlichen Münsterland – Genese, Migration, Gefahrenpotential. – XVI + 154 S., 62 Abb., 16 Tab., 12 Anh.; Münster. - [Dissertation]

Rice, D D 1993: Composition and origins of coalbed gas. In: Hydrocarbons from coal: AAPG Studies in Geology 38 (1), S. 159–184.

Thielemann T, Cramer B and Schippers A 2004: KOHLEFLÖZGAS IM RUHR-BECKEN: FOSSIL ODER ERNEUERBAR? DGMK-Frühjahrstagung 2004, Fachbereich Aufsuchung und Gewinnung, Celle DGMK-Tagungsbericht 2004-2, ISBN 3-936418-17-9, Seiten 449 bis 459

Wissen M 2021: EU-Methane-Strategy Measures to reduce emissions from abandoned coal mines. Presentation at the EU stakeholder workshop 15. March 2021

Zazzeri G, Lowry D, Fisher R E, France J L, Lanoisellé M, Kelly B 2016: Carbon isotopic signature of coal-derived methane emissions to the atmosphere: from coalification to alteration. In: Atmos. Chem. Phys. 16 (21), S. 13669–13680. DOI: 10.5194/acp-16-13669-2016