Fugitive hydrocarbon emissions from Indiana oil/gas wells into the atmosphere



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Introduction

Hydrocarbon gas emissions from oil and gas wells contribute significantly to anthropogenically emitted greenhouse gases, predominantly as methane [1, 2, 3]. Methane flux from oil and gas wells has reached 61 Tg yr⁻¹, which is the largest source of the total anthropogenic methane emission flux of 336 Tg yr⁻¹ [4]. It is essential to reduce methane emissions from oil/gas wells to mitigate global warming.

There are more than 80,000 active, inactive, temporarily abandoned and permanently plugged oil and gas wells in Indiana. The extent of fugitive hydrocarbon emissions from those wells into the atmosphere has not been quantitatively studied. This study is a first attempt to quantify those emissions.

Methanotrophs are methane-oxidizing bacteria which can metabolize methane. Methane in natural seepages can be partly consumed by methanotrophs in soil around seepages before it reaches the atmosphere [5]. Therefore, soils enriched in methanotrophs above hydrocarbon leakages have a potential to mitigate hydrocarbon emissions from leaking oil/gas wells. We evaluate a microbial mitigation strategy with mesocosm and artificial soil mound experiments.

Experiments

1. Research area

We investigated 20 active, temporarily deactivated or plugged-abandoned oil/gas wells across Monroe, Daviess and Sullivan counties in Indiana. Fugitive emissions on the order of liters per hour qualified an active gas well in Monroe County to serve as our research site. 2. Quantification of methane emission

We built a polyethylene tent to enclose fittings of a leaking research well. The bottom of the polyethylene foil was ballasted by stones to seal against the ground. A DC-operated fan homogenized the methane concentration in the tent. Air from within the tent was sampled by a SARAD RTM 2200 instrument and methane concentrations were determined with an Axetris laser **OEM Module LGC F200 CH**₄ detector. The methane emission rate (L*hour⁻¹) equals to k*V*3.6, where k is the slope of methane concentration change with time $(mL^*m^{-3*}s^{-1})$ and V is volume of the tent (m^3) . 3. Mesocosm experiment

We collected topmost soils adjacent to and 20 m away (control soils) from the research well and set up triplicate mesocosm systems of those soils in stoppered 70-mL glass culture bottles in contact with a methane-enhanced standard atmosphere. We measured the remaining methane concentration over time in headspace of culture bottles. We also measured changes in methanotrophic activity of soil as a function of moisture content. The original soil's moisture content was 32 wt. %.





Fig. 2 Polyethylene tent enclosing fittings of leaky research well

Experiments (continued)

4. Soil mound experiment

We tested the feasibility of a "methanotrophic soil mound" over an artificial CH₄ seep to evaluate whether the mound could reduce CH₄ emissions. We used soils from a solid waste landfill in Monroe County to build a "methanotrophic mound" in a bucket and placed it in a 0.903 m × 0.462 m × 0.358 m terrarium. The mound's base was a permeable layer of rocks covered by soil. The height of the mound was 0.15 m with a diameter of 0.258 m. A capillary tube was fixed at the bottom of the rock layer and connected to a CH₄ gas cylinder to continuously emit CH_4 . CH_4 inflow rate from the tube was regulated by a control valve, which was set at 74.34 mL/h (6 bubbles/min). The terrarium was sealed using an adhesive tape and the CH₄ concentration in air was measured by a SARAD RTM **2200.** We also tested if prolonged exposure of a mound (using control soil from the leaking well) to CH₄ supplied in abundance will increase its methanotrophic activity.

Table 1 CH ₄ concentrations in air near investigated oil/gas wells across Indiana			
Well No.	County	Symbol ¹	CH ₄ concentration /ppm
1	Monroe	GSG	1.88-2.08
2	Monroe	OBG	1.12-1.78
3	Monroe	OBS	1.83-12.90
4	Monroe	DRY	1.24-1.98
5	Monroe	OBS	2.40-2.76
6	Monroe	DRY	2.55-2.64
7	Daviess	OIL	1.42
8	Daviess	WIO	1.43-1.53
9	Daviess	OIL	1.93
10	Daviess	OIL	~25
11	Daviess	OIL	1.75
12	Daviess	WIO	1.93-2.35
13	Daviess	OIL	2.45
14	Sullivan	OIL	Saturation ²
15	Sullivan	OIL	0
16	Sullivan	GAS	0
17	Sullivan	GAS	4
18	Sullivan	GAS	0
19	Sullivan	GAS	0
20	Sullivan	AGAS	0

¹ Symbols are defined by Petroleum Database Management System of IGWS. ² Saturation means CH₄ concentration exceeds 96 ppm.

Results

1. Methane emission rates

The measured methane concentrations in air surrounding investigated oil/gas wells are listed in Table 1. We found that methane concentrations in the tent around the research well increased linearly with time. The average methane emission rate was calculated as 1.73 L/hour using R=k*V*3.6 (k is 1.622, 1.323, 1.366 mL*m⁻³*s⁻¹, respectively for the three measurements; V=0.335 m³).





Fig. 4 Artificial soil mound experiments

Results (continued)

2. Mesocosm system

4 g of soil from the leaky well consumed 96.9 % of the initial CH₄ after 138 hours, while the systems with no soil and the same amount of control soil only consumed less than 15 %.

After control soil was continuously exposed to elevated methane concentrations for one month in a culture bottle, its microbes expressed enhanced methanotrophy relative to those in the original control soil.

The order of soil moisture contents from higher to lower methanotrophic activity is 16 >32 >24 >40 >48 =64 >0 wt. %, indicating the optimal moisture content with maximum methanotrophy is 16 wt. %. The microbes in totally dried soil from adjacent to the leaky well "woke up" the fastest, also in the soil remoistened by 16 wt. % added water. **Completely dry soil expresses dormant and** inactive methanotrophs. Water-filled porosity in wet soil does not allow easy access of methane and oxygen from air to enter the soil, and therefore methanotrophy in wet soil is impeded.

3. Soil mound

• Landfill soil mound with higher methanotrophic activity can mitigate methane emission more effectively compared to control soil mound from the leaky research well.

 The methanotrophic activity of the control soil mound can be enhanced over time after being exposed to abundant methane (85 ppm) in the terrarium.

References

[1] Vielstädte L. et al., 2015. Mar. Pet. Geol., 68: 848-860, https://doi.org/10.1016/j.marpetgeo.2015.07.030 [2] Townsend-Small A. et al., 2016. *Geophys. Res. Lett.*, 43: 2283–2290, https://doi.org/10.1002/2015GL067623 [3] Kang M. et al., 2014. PNAS, 111(51): 18173-18177, https://doi.org/10.1073/pnas.1408315111 [4] Van Amstel A., 2012. Journal of Integrative Environmental Sciences, 9(1): 5-30, https://doi.org/10.1080/1943815X.2012.694892 [5] Henckel T. et al., 2000. Appl. Environ. Microbiol., 66(5): 1801-1808, https://doi.org/10.1128/AEM.66.5.1801-1808.2000

ontrol soil after exposed to enhanced metha Fig. 7 Comparison of original control soil and the contr Fig. 6 CH₄ concentration change over time in soil after exposure to elevated CH₄ for one montl 🗕 Blank 🗕 0 % 🚽 16 % 📥 24 % 📥 32 % 📥 40 % 📥 48 % 📥 64 % Fig. 8 CH_a concentration change in mesocosm systems Fig. 9 CH₄ concentration change in mesocosm systems with dried soils re-moistened with water with soils with different moisture contents 🔶 No mound 🔶 control soil mound 🗕 landfill soil mound Fig. 11 CH₄ concentration change with control soil moun Fig. 10 CH₄ concentration change over time in th for three duplicates (1st measured on 9/10/2018, 2nd o im with no mound, control soil mound from 9/11, 3rd on 9/18; the mound had been continuously research well, and landfill soil mound exposed to 85-ppm methane since 9/10)

Conclusions

• The methane flux from the leaky research well is ca. 2 L/hour.

Soils adjacent to leaking wells exhibit significant methanotrophic activity. Prolonged exposure of soil methanotrophs to elevated methane concentrations enhances their methanotrophic activity.

Building a soil mound with high methanotrophic activity above abandoned and leaking oil/gas wells can be a costeffective strategy to microbially mitigate fugitive hydrocarbon emissions from wells.