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OPTIMIZATION OF ENVIRONMENTAL PARAMETERS CONDUCIVE TO METHANE OXIDATION WITH COAL MINE DUMP ROCK

The aim of this paper was to estimate the ability of microorganisms that live in environment of a dump rock from a local coal mine (Bogdanka coal mine, Lublin) to use methane as a nutrient. The kinetics of methane oxidation at different initial concentrations of both methane and moisture was investigated. It was confirmed that the highest rate of methane oxidation, 0.53 μmol g^{-1} day^{-1}, occurred in a sample whose atmosphere was enriched with 10% (v/v) CH_4 at 50% moisture.

1. INTRODUCTION

Methane is a main hydrocarbon present in the atmosphere, its concentration is now ca 1.8 ppmv and it got doubled during last 250 years. Despite a short residence time in the atmosphere, i.e. about 10 years, the ability of methane to absorb infrared radiation makes it 20–30 times more efficient as a greenhouse gas than carbon dioxide [4]. Methane is removed from the atmosphere by reaction with hydroxyl radicals [4]; moreover, about 10% of methane are consumed in a biological way by aerobic oxidation in soil owing to the activity of methanotrophs. In a global methane balance, the soil fulfils two inverse functions being a source of about 40% of released methane and also removing it or reducing its emission into the atmosphere [5]. The ability of soils of different types, such as forest soils [6], arable soils including rice fields and peatbogs [7] and meadows [3] as well in soils from landfill covers [1], [2], to oxidize methane was confirmed in the field and laboratory studies.

The aim of the work was to determine the methanotrophic activity of a coal mine dump rock (originated from Bogdanka coal mine) in different conditions of moisture and initial concentration of methane in the atmosphere.

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2. MATERIALS

The samples of the rock tested were collected directly from a dumping area in the neighbourhood of Bogdanka coal mine (at present: "Lubelski Węgiel" S.A. in Bogdanka) located in the Puchaczów region.

The total area of a waste rock deposition is ca 87.95 ha, while the covered area occupies about 55 ha. The height of dump is above 26 m and its length approaches 1 km. On the top of waste dump there is a lake whose depth is 11 m and volume ca 0.5 million m^3. The production of the tailing waste ranges from ca. 0.8 to 1.2 Mg per year. For the investigations, only the fresh rock (collected directly from the mine) was used.

The rock material was first ground with a roller mill (Testchem, Poland) up to 1 mm grain diameter.

3. METHODS

3.1. WATER CAPACITY

The water capacity of the rock examined was determined gravimetrically by saturating rock samples with water up to a constant weight. The measurements were carried out in triplicate. The total water capacity was 306 $cm^3 \cdot kg^{-1}$.

3.2. METHANOTROPHIC ABILITY

Methanotrophic activity of bacteria was determined by the incubation of 15-g portion of disintegrated rock in dark bottles (60 cm^3) filled up with deionised water to obtain a sample moisture adequate to 50%, 100% and 200% of water capacity.

The bottles were closed with rubber septa, which made taking the gaseous samples possible, capped with aluminium cap and sealed with paraffin. The HNO_3-treated and washing out to neutral pH sand was used as a control sample. The part of air in closed bottles was replaced with methane (gas-tight syringe, Hamilton) to obtain its final concentrations of 0.5, 1, 2, 5, 10, 15 and 30% (v/v) in gaseous phase. The bottles with wetted rock samples and atmosphere diversified in methane concentration were incubated at 20 °C±1 °C. The concentrations of CH_4, O_2 and CO_2 were determined by means of the gas chromatograph GC 3800 (VARIAN, USA). Methane used in the experiment had the purity of 99.99% (Praxair, Poland).

The analyses of gases collected from incubated bottles were carried out until no differences in the atmosphere composition were confirmed.

4. RESULTS

The incubation of rock being in contact with the atmosphere rich in oxygen and methane resulted in the changes of their concentrations. Dynamics of methane and oxygen concentration were represented by decreasing exponential curves, and the concentration of carbon dioxide systematically increased at the same time. The control samples did not show the methanotrophic activity.

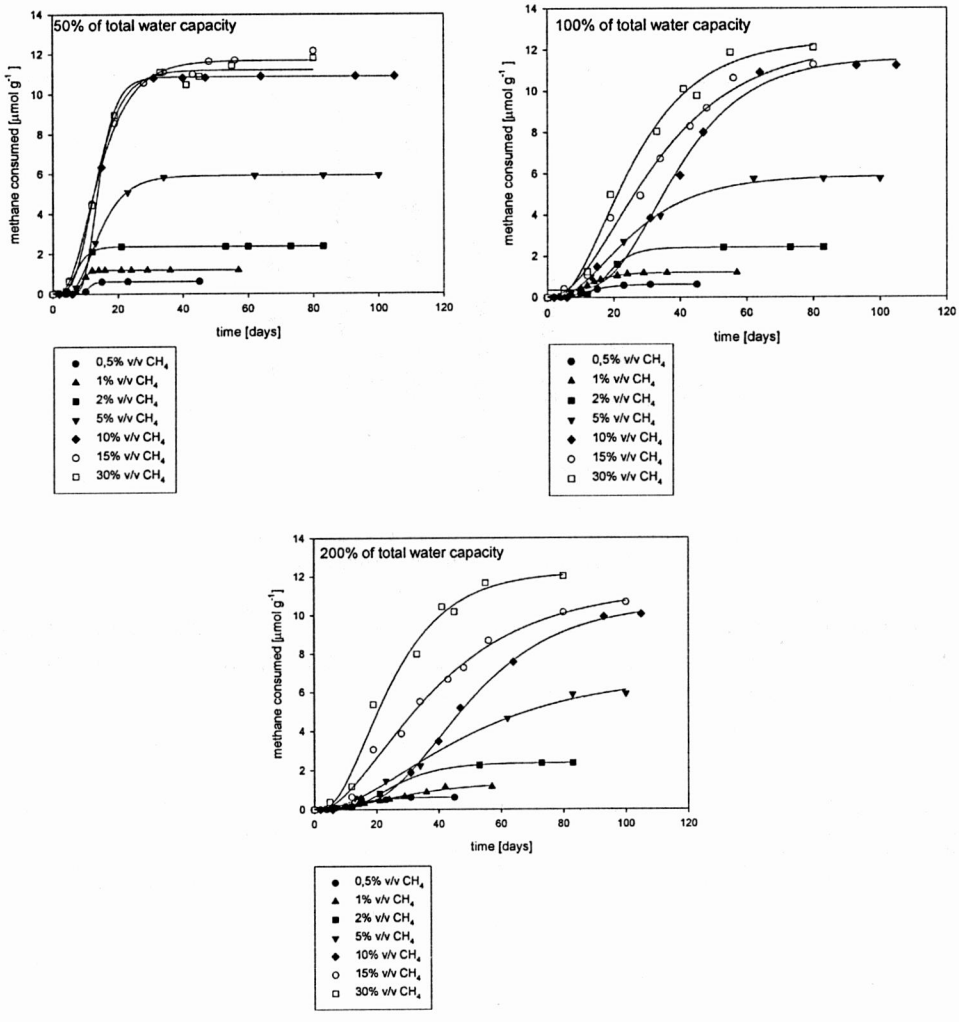

Fig. 1. Kinetics of methane consumption during the incubation of the coal mine waste rock at different initial concentrations of methane and water content. Correlation coefficients r^2 for all curves are > 0.99 ($P < 0.001$)

The oxidation of methane started after 5–12 days, independently of moisture degree, but was evidently dependent on an initial methane concentration (figure 1).

It was confirmed that the rate of methane oxidation increased with substrate concentration, and reached its maximal value of methanotrophic capacity at 10% CH_4 (v/v) and moisture equal to 50% of total water capacity. The maximal values reached 0.53, 0.29 and 0.31 $\mu mol \cdot g^{-1} \cdot day^{-1}$ at 50%, 100% and 200% of water capacity, respectively (figure 2).

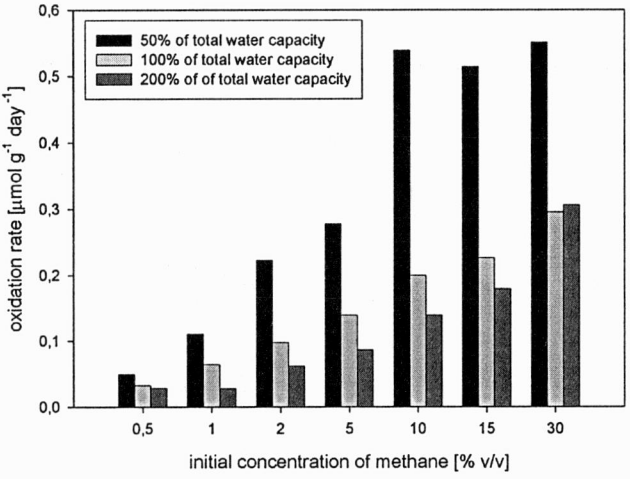

Fig. 2. Rate of methane oxidation by the coal mine waste rock, depending on water content and initial concentration of methane

A complete oxidation was observed at a low initial methane concentration (<10%), and the process efficiency decreased along with an increase in methane concentration approaching the level of 30% (the table).

Table

Efficiency of methane consumption related to water content and initial concentration of methane

Initial concentration of methane [%v/v]	Efficiency of methane oxidation [%]		
	50% of total water capacity	100% of total water capacity	200% of total water capacity
0.5	100	100	100
1	100	100	100
2	100	100	100
5	100	100	100
10	94	99	87
15	64	63	59
30	32	34	34

5. CONCLUSIONS

Based on the experiments performed, the following conclusions can be drawn:

1. The rate of methane oxidation by dump rock was the highest for the samples whose moisture content reached 50% of a total water capacity and then it decreased along with the moisture increase.

2. The maximum rate of methane oxidation occurred at 10% (v/v) concentration of methane and 50% moisture.

3. The methanotrophic capacity of a fresh rock collected directly from the coal mine ranged from 11 to 12 $\mu mol \cdot g^{-1}$ of dry matter.

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REFERENCES

[1] Börjesson G., Chanton J., Svensson B.H., *Methane oxidation in two Swedish landfill covers measured with carbon-13 to carbon-12 isotope ratios*, J. Environ. Qual., 2001, 30, 369–376.

[2] Chan A.S.K., Parkin T.B., *Evaluation of potential inhibitors of methanogenesis and methane oxidation in a landfill cover soil*, Soil Biol. Biochem., 2000, 32, 1581–1590.

[3] Horz H., Raghubanshi A.S., Heyer J., Kammann C., Conrad R., Dunfield P.F., *Activity and community structure of methane-oxidising bacteria in wet meadow soil*, FEMS Microbiol. Ecol., 2002, 41, 247–257.

[4] Le Mer J., Roger P., *Production, oxidation, emission and consumption of methane by soils: a review*, Eur. J. Soil Biol., 2001, 37, 25–50.

[5] Pawłowska M., *The possibility of decreasing methane emission from landfills through biochemical oxidation in a landfill soil cover – laboratory experiments* (in Polish), Wyd. PL, Lublin, 1999.

[6] Philips R.L., Whalen S.C., Schlesinger W.H., *Response of soil methanotrophic activity to carbon dioxide enrichment in a North Carolina coniferous forest*, Soil Biol. Biochem., 2001, 33, 793–800.

[7] Watson A., Stephen K.D., Nedwell D.B., Arah J.R.M., *Oxidation of methane in peat: kinetics of* CH_4 *and* O_2 *removal and the role of plant roots*, Soil Biol. Biochem., 1997, 29/8, 1257–1267.

OPTYMALIZACJA PARAMETRÓW ŚRODOWISKOWYCH SPRZYJAJĄCYCH UTLENIANIU METANU Z UDZIAŁEM SKAŁY PŁONNEJ

Celem pracy było wstępne określenie zdolności metanotroficznej mikroorganizmów żyjących w środowisku skały płonnej towarzyszącej pokładom węgla kamiennego w Bogdance koło Lublina. Dokonano pomiarów kinetyki utleniania metanu dla różnych początkowych stężeń metanu w szerokim zakresie uwilgotnienia. Największą szybkość utleniania metanu wynoszącą 0,53 $\mu mol \cdot g^{-1} \cdot$dzień$^{-1}$ stwierdzono w próbie z atmosferą wzbogaconą w 10% (v/v) metanu przy 50% uwilgotnieniu.