METHANE OXIDATION AND DEGRADATION OF HALOGENATED ORGANIC COMPOUNDS IN LANDFILL GAS AFFECTED SOIL

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Introduction

Waste deposited in a landfill will undergo anaerobic decomposition resulting in generation of landfill gas, which is transported through soil top covers causing emission of gas into the atmosphere. Besides methane (55-60 vol.%) and carbon dioxide (40-45 vol.%) landfill gas also contains numerous trace compounds (up to 5 vol.%) (Brosseuau & Heitz, 1994). The trace compounds originate from hazardous materials deposited in the landfill or from biological/chemical degradation of materials deposited in the landfill.

Emission of methane from landfills accounts for between 7 and 20% of the global anthropogenic sources of methane emissions thus contributing to the global climate change. Emission of trace compounds like benzene and vinyl chloride to the ambient air can be a threat to workers and local habitants, while other trace compounds like freons contribute to the depletion of the ozone layer (Christensen & Kjeldsen, 1995).

Microbial oxidation of methane in aerobic soils plays a significant role in reducing the emission of methane to the atmosphere. In landfill top covers methane and oxygen counter-gradients may appear due to emission of methane from the waste and diffusion of oxygen from ambient air. Oxidation of methane by methanotrophic bacteria in landfill top cover soil has been shown to reduce the amount of methane emitted to the atmosphere. Under methane-oxidating conditions the methanotrophic bacteria is known to co-metabolize a variety of aliphatic compounds including some halogenated hydrocarbons (Oldenhuis et al. 1989).

The objective of this study was to investigate the potential of natural oxidation of methane and halogenated organic compounds in soil exposed to landfill gas. In addition, the impact of individual factors (like soil moisture, soil temperature, co-inhibition and concentrations of oxygen and methane) on methane oxidation and co-oxidation of selected trace compounds was studied. The investigations have been carried out through laboratory experiments. The chosen trace components included six chlorinated ethylenes (perchloroethylene, trichloroethylene, 1,1-dichloroethylene, cis-1,2-dichloroethylene, and vinyl chloride), and two freon compounds (chlorodifluoromethane and dichlorofluoromethane).

Methods

The degradation of trace components was examined in simple batch experiments using soil samples collected from soil covers at Skellingsted landfill, Western Sealand, Denmark. A fixed amount of soil was added to a 117mL batch container equipped with buthyl rubber stoppers, which enabled gas sampling for analysis. Column experiments simulating a landfill top cover soil matrix through in which gas was transported were carried out to examine the degradation process in a dynamic system. The oxidation process was examined in a methane and oxygen counter-gradient system. The columns were packed with landfill cover soil and water, and continuously fed in opposite ends with methane gas containing trace components and air. Sampling ports located along the column length allowed taking gas samples, which were subsequently analyzed by gas chromatography.

Results & Discussion

High methane oxidation potentials were found in batch experiments conducted with soil collected from 5 to 35 cm depth. At 40 cm depth the methane oxidation activity decreased dramatically, but experiments conducted with soil from 90 cm depth showed that there still was low activity at that depth. *Degradation of compounds:* All the chlorinated ethylenes were shown to be degraded in presence of oxygen and methane, and the degradation occurred in parallel with the oxidation of

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methane. However, total transformation of perchloroethylene was not observed. In general, the degradation rates and extent of degradation of the chlorinated ethylenes was inversely related to the chlorine/carbon ratios. HCFC-21 and HCFC-22 were also shown to be degradable. *Impact of individual factors:* The highest degradation rates were obtained in experiments run without methane but containing resting bacteria pre-incubated with methane. Increasing methane concentrations resulted in decreasing degradation rates - a result of the competition between methane and the halogenated compounds for the enzyme methane monooxygenase inducing the oxidation. In batch experiments without oxygen no degradation occurred within the run of the test. The oxidation of methane and halogenated organic compounds was very dependent on temperature, showing optimum rates between 20 and 27°C. However, oxidation occurred at temperatures as low as 2°C. Batches with a water content of approximately 25 %w/w produced maximum oxidation rates. In experiments with air-dried soil it seemed to be difficult the recover the oxidation activity of the bacteria. The column experiments will be finished during the summer and the results have therefore not yet been analyzed.

Simple calculations based on the obtained oxidation rates from the batch experiments and

assuming realistic fluxes of landfill gas and an oxidation zone of 40 cm showed that the

emission of trace components from landfills may be significantly reduced due to

degradation process in the soil top cover.

References

- Brosseau, J. & Heitz, M. (1994): Trace gas compound emissions from municipal landfill sanitary sites. *Atmospheric Environment*, 28, pp 285-293.
- Christensen, T. H. & Kjeldsen, P. (1995): Landfill emissions and environmental impact: An introduction. In: Christensen, T. H., Cossu, R., Stegmann, R. (eds), Sardinia '95, Fifth International Landfill Symposium, S. Margherita di Pula, Italy 2-6 October, Proceedings, Vol. III. Pp. 3-12. CIAS-Environmental Sanitary Engineering Center. Cagliari, Italy.
- Oldenhuis, R., Vink, R. L. J. M., Janssen, D. B. & Witholt, B. (1989): Degradation of chlorinated aliphatic hydrocarbons by *Methylosinus trichosporium* OB3b expressing soluble methane monooxygenase. *Applied and Environmental Microbiology*, Nov. 1989, pp. 2819-2826.