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Determination of volatile organic compounds from biowaste and co-fermentation biogas plants by single-sorbent adsorption

J.I. Salazar Gómez*, H. Lohmann, J. Krassowski

Fraunhofer Institute for Environmental, Safety, and Energy Technology (UMSICHT), Osterfelder Strasse 3, 46047 Oberhausen, Germany

HIGHLIGHTS

- A correlation between TVOCs and substrate composition and feeding amount was determined.
- Longer residence times in the digester appear to reduce the emissions of VOCs.
- Terpenes and ketones were found to be the predominant VOCs in all three biogas plants.
- In situ desulphurisation reduces the amounts of hydrogen sulphide but shows little influence on sulphides and thiols.
- A simple sampling method allowed the quantification of a broad range of VOCs.

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ABSTRACT

Characterisation of biogases is normally dedicated to the online monitoring of the major components methane and carbon dioxide and, to a lesser extent, to the determination of ammonia and hydrogen sulphide. For the case of Volatile Organic Compounds (VOCs), much less attention is usually paid, since such compounds are normally removed during gas conditioning and with exception of sulphur compounds and siloxanes represent a rather low risk to conventional downstream devices but could be a hindrance for fuel cells. However, there is very little information in the literature about the type of substances found in biogases generated from biowaste or co-fermentation plants and their concentration fluctuations. The main aim of this study was to provide information about the time dependencies of the VOCs in three biogas plants spread out through Germany from autumn until summer, which have different process control, in order to assess their potential as biofuels. Additionally, this study was an attempt to establish a correlation between the nature of the substrates used in the biogas plants and the composition of the VOCs present in the gas phase. Significant time-dependent variations in concentration were observed for most VOCs but only small changes in composition were observed. In general, terpenes and ketones appeared as the predominant VOCs in biogas. Although for substances such as esters, sulphur-organic compounds and siloxanes the average concentrations observed were rather low, they exhibited significant concentration peaks. The second biogas plant which operates with dry fermentation was found to contain the highest levels of VOCs. The amount of total volatile organic compounds (TVOCs) for the first, second and third biogas plants ranged from 35 to 259 mg Nm^{-3}, 291–1731 mg Nm^{-3} and 84–528 mg Nm^{-3}, respectively.

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1. Introduction

The utilisation of biogenic gases such as, landfill gas (LFG) (Dernbach, 1985; Franzius, 1985; Aitchison, 1996), sewage gas (Friedrich et al., 2000; Rossol et al., 2003; Tsagarakis, 2006) or farm biogas from energy crops or agricultural wastes (biowastes) (Raven & Gregersen, 2007; Weiland, 2010; Murphy & McCarthy, 2005) as renewable sources of energy has become a common practice in the European context and has been in recent years increasingly supported by the introduction of incentives and subsidies (Weiland, 2000, 2003; Graβmann, 2011). The political decision taken by the German government in June 2011 to back out completely of the nuclear energy programme by the year 2022, decision mainly motivated by the unfortunate natural disaster in Fukushima, Japan,

* Corresponding author.
E-mail addresses: jsalazar@gmx.net (J.I. Salazar Gómez), heiko.lohmann@umsicht.fraunhofer.de (H. Lohmann), joachim.krassowski@umsicht.fraunhofer.de (J. Krassowski).

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has enormously pushed on the utilisation of biogas in particular from agricultural sources. This decision supports the modern trend of a more decentralised power supply system and the search for alternative sources of energy in order to improve the current power and heat efficiencies (Staffell et al., 2008), which will help to mitigate the dependency to non-renewable sources of energy (e.g. coal and oil). According to the German Biogas Association, in the year 2013 the number of biogas plants in Germany accounted for 7850 plants, which generated around 3543 MW electricity from farm biogas. This helped to mitigate the emission of about 16,8 million tonnes of CO_2 (German Biogas Association). Based on data of the German Ministry of Environment (BMU), in 2010 the savings of CO_2 emissions through the utilisation of sewage and landfill gas were around 1,11 million tonnes and 0,65 million tonnes of CO_2, respectively (German Ministry of Environment). Data from the European Biogas Association (EBA) show that, in the European scenario, Germany appears as the main biogas producer followed by the UK (European Biogas Association).

1.1. Biogas production and composition

The term "Biogas" is widely understood as the gas produced by the decomposition of organic matter under anaerobic conditions where the organic materials undergo basically four steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Appels et al., 2008). Hydrolysis is generally regarded as the rate-limiting step (Scherer, 2001) since high-molecular weight molecules e.g. polysaccharides, lipids and proteins have to be degraded to smaller compounds. Fig. 1 shows the widely accepted sequence of anaerobic degradation processes.

Methane and carbon dioxide appear as the main constituents of the different biogenic gases, with concentrations ranging between 40-60% and 40–55% (Rasi et al., 2010), respectively. For the case of LFG, methane and CO_2 concentrations depend strongly on the age of the landfill (anaerobic methanogenic steady phase) (Farquhar & Rovers, 1973), the composition of the waste and partly on the type of the collection system. Moreover, among all biogenic gases LFG appears as the most contaminated gas due to the higher number of harmful VOCs (e.g. halogenated compounds), followed by sewage gas, whereas farm biogas seems to be the less contaminated one (Rasi et al., 2007). Apart from the major components, farm biogas exhibits low concentrations of ammonia and is saturated with water vapour but may also contain relatively high amounts of hydrogen sulphide (H_2S). Depending on the substrates used it may exhibit some similar VOCs as in landfill and sewage gas such as, ketones, alcohols, terpenes, alkanes, esters, furans, mercaptanes, sulphides and aromatics (Smet et al., 1999). Halogenated compounds and siloxanes are in principle not expected in farm biogas. As pointed out by Rasi et al. (2011) their concentrations in farm biogas are apparently very low and their presence in biogas would depend on the nature of the feedstock used for co-fermentation. Farm biogas from exclusively energy crops may exhibit H_2S concentrations as high as 2000 ppm. In contrast, farm biogas from biowastes may exhibit H_2S concentrations even over 20,000 ppm.

The presence of trace compounds in LFG and sewage gas is well documented in the literature but for farm biogas there is little information about the type of VOCs that can be found, their concentration ranges and possible seasonal fluctuations. Therefore, it is the aim of this article to show the results gathered in a research project. The main goal of the present study was to determine the concentration and the seasonal variations of typical VOCs present in the biogas of three different biogas plants spread out throughout Germany, which are fed with very different substrates in order to help make decisions about adequate upgrading processes.

2. Materials and methods

2.1. Biogas plant description

For the determination of VOCs in biogas, three different biogas plants (BP) were selected and monitored for a period over six months (from autumn until summer) in order to check for eventual concentration peaks, since some substrates commonly used in biogas plants are season dependent. The biogas plants are located in different federal states throughout Germany in order to avoid similar composition of the substrates used and differ in their technical design and process control. The first biogas plant is located in the federal state of North Rhine-Westphalia. The composition of the substrates used at this site comprises typically 45% glycerine, about 40% of fatty sludge from the biodiesel production and 15% cattle manure.

The second biogas plant is located in the federal state of Saxony-

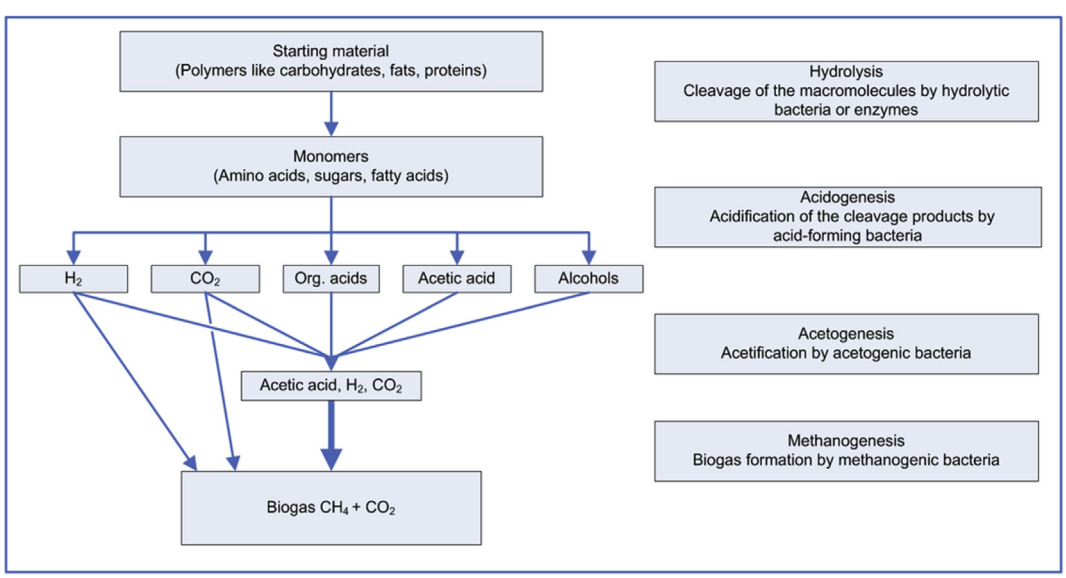

Fig. 1. The anaerobic degradation process after (Bischoffsberger & et al., 2005).

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