



Research article

Potential for anaerobic treatment of polluted sediment

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ABSTRACT

Due to the anaerobic nature of aquatic sediments, the anaerobic treatment of sediments utilizing already present microflora represents an interesting treatment option. Contaminated sediment can contain a variety of organic contaminants, with easily degradable organics usually present in the higher amounts, along with traces of specific organic pollutants (total petroleum hydrocarbons and polycyclic aromatic hydrocarbons). This study applies a comprehensive approach to contaminated sediment treatment which covers all the organic contaminants present in the sediment. The aim of this study was to (1) evaluate the anaerobic treatment of aquatic sediment highly loaded with easily degradable organics via determination of potential biogas and methane production, and (2) assess possibilities of using anaerobic treatment for the degradation of specific organic pollutants in order to reduce the risks posed by the sediment. In order to promote the methanogenic conditions of the indigenous microflora in the sediment, the addition of co-substrates acetate and glucose was investigated. The results, expressed as mL biogas produced per volatile solids content in sediment (VS_{added}) indicate that the addition of the co-substrate has a significant impact on biogas production potential (58.7 and 55.1 mL/g VS_{added} for acetate and glucose co-substrate addition respectively, and 14.6 mL/g VS_{added} for the treatment without co-substrate addition). Theoretical biochemical methane potential was assessed by Gompertz model and Chemical oxygen demand model. The Gompertz model fit better for all the applied treatments, and was capable of predicting the final productivity of biogas and methane in the first 30 days with a relative error of less than 14%. From the aspects of specific organic pollutants, total petroleum hydrocarbon degradation was promoted by co-substrate addition (degradations of 75% and 60% achieved by acetate and glucose co-substrate addition, compared to 45% for the treatment without co-substrate addition). Polycyclic aromatic hydrocarbons were reduced by significant amounts (84–87%) in all the applied treatments, but the addition of co-substrate did not further improve their degradation.

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1. Introduction

Sediment is an essential and dynamic component of all aquatic systems and represents a potential reservoir of toxic substances of anthropogenic origin, due to its tendency for binding pollution. The environmental problems related to contaminated sediment are diverse and sometimes specific with reference to time and space (Juwarkar et al., 2010; Polyak et al., 2018; Ren et al., 2018). Some contaminants flow directly from industrial and municipal waste dischargers, while others come from polluted runoff in urban and agricultural areas. As a consequence, contaminated sediment can contain a variety of organic contaminants, including large amounts

of easily degradable organics (similar to those present in sewage sludge), and specific organic pollutants present in trace amounts such as total petroleum hydrocarbons, pesticides, polycyclic aromatic hydrocarbons, etc. (Agarwal and Liu, 2015; Gong et al., 2014). Today, one of the major problems in the environment is hydrocarbon contamination which is originally from petrochemical industry. Petroleum products are very complex mixtures and hydrocarbons comprise the majority of the components in most of their products and are the compounds that are primarily measured as total petroleum hydrocarbons (TPH). The complexity of petroleum hydrocarbon increases with carbon number (Anon, 1998; Das and Chandran, 2011). Alongside to TPH, polycyclic aromatic hydrocarbons (PAH) represent a large class of organic molecules, which are ubiquitous environmental contaminants derived principally from incomplete combustion of petroleum products. As a

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consequence of their hydrophobicity, TPH and PAH in aquatic environments rapidly tend to be brought down to the bottom sediment as a sink in the aquatic system. Therefore, sediments represent the most important reservoir of PAHs in the aquatic environment (Maletić et al., 2012; Spasojević et al., 2015).

For the remediation of sediments contaminated with organic pollutants, the most often applied techniques are bioremediation under aerobic or anaerobic conditions (Agarwal and Liu, 2015; Gong et al., 2014; Ghattas et al., 2017; Haghollahi et al., 2017; Kuppusamy et al., 2017; Polyak et al., 2018; Ren et al., 2018; Rončević et al., 2016; Spasojević et al., 2015). The anaerobic treatment of sediment is particularly interesting, since aquatic sediments are anaerobic in nature, and the required microflora required for degradation of organic matter may therefore already be present. Numerous studies have proven that many organic pollutants can be degraded under all anaerobic conditions of the thermo-dynamic ladder (Bethke et al., 2011; Bian et al., 2015; Ghattas et al., 2017; Jaekel et al., 2015; Musat, 2015). After oxygen depletion by aerobic respiration, nitrate, manganese, iron and sulfate can be used as an alternative electron acceptor. In the absence of inorganic electron acceptors except for CO₂, methanogenesis plays an important role for the biodegradation of organic matter (Ghattas et al., 2017). As it is well known, main organic matter degradation product in the methanogenic conditions are CO₂ and CH₄. Methane is regarded as the second-most important anthropogenic greenhouse gas, with global emission rates between 500 and 600 Tg yr⁻¹. The contribution of freshwater sediments is estimated to be around 103 TgCH₄ yr⁻¹ (Bastviken et al., 2011; Mach et al., 2015; Maeck et al., 2013, 2014). In addition, if the anaerobic degradation is carried out under methanogenic conditions, there is a possibility for sediments highly loaded with degradable organic contaminants to produce methane in a controlled environment, which can be subsequently be used as an energy source (Musat, 2015; Nazaries et al., 2013).

The controlled production of methane from polluted sediments has been studied in the literature to some extent. The anaerobic degradation of polycyclic aromatic hydrocarbons has been extensively studied under various redox conditions. However, little is known about the anaerobic degradation of petroleum compounds, especially under methanogenic conditions, and most published studies were performed under sulfate reducing conditions (Ghattas et al., 2017). Methanogens cannot directly degrade these complex molecules and thus depend on the supply of electron donors and co-substrate (H₂/CO₂, formate/acetate) by other microorganisms (Ghattas et al., 2017; Nazaries et al., 2013). To our knowledge, the influence of co-substrates such as acetate and similar on TPH and PAH anaerobic biodegradation has not been fully investigated. Additionally, there is a particular lack of data relating to the simultaneous degradation of the large variety of organics present in sediments, which is of great importance for a holistic approach to contaminated sediment treatment.

This study therefore investigates a comprehensive approach to the anaerobic treatment of contaminated sediment, by covering all the organic contaminants present in the sediment. For this purpose, the investigation was directed in two directions: (1) an evaluation of the potential production of biogas and methane during the anaerobic treatment of aquatic sediment highly loaded with easily degradable organics, and (2) an assessment of the possibilities of applying anaerobic treatment for the degradation of specific organic pollutants in order to reduce the risk posed by the contaminated sediment. To promote methanogenic conditions in the indigenous sediment microflora, the addition of co-substrates such as acetate and glucose was also investigated.

2. Experimental and methods

2.1. Contaminated sediment

For this study, sediment highly polluted with organic pollutants was taken from several locations on the Kudos canal (Serbia), which has a long history of contamination from the discharge of untreated municipal and industrial wastewaters (from a tyres factory and tannery). The contamination from this canal is further spread to the river Sava (the biggest tributary of the river Danube). The location of the Kudos canal and Sava river are presented in figure S1 in the supplementary material. The total amount of the sampled sediment was 3 kg. The initial sediment sample (IS) had the following characteristics: volatile solids (VS) 20.8 ± 0.40%, total organic carbon (TOC) 7.38 ± 0.41%, clay 28.3 ± 0.5%, chemical oxygen demand (COD) 155.0 ± 28.2 g O₂/kg, biological oxygen demand after five days (BOD₅) 56.2 ± 15.4 g O₂/kg, total nitrogen 8042 ± 351 mg N/kg, NH₄⁺-N and organic N 7300 ± 235 mg N/kg, nitrate, nitrite N 743 ± 51.4 mg N/kg, total phosphorous 717 ± 65.3 mg P/kg, iron 51.1 ± 3.54 g/kg and manganese 1024 ± 157 mg/kg.

2.2. Experiment setup

Anaerobic biodegradation was carried out with indigenous microorganisms in biometric flasks equipped with a measuring cylinder on top for volumetric measurement of biogas production. Subsamples of sediment (50 g) were placed in biometric flasks with four types of biodegradation conditions (Table 1): abiotic control (ATS-C), a treatment with indigenous microorganisms (ATS-DI), and two treatments with the addition of co-substrates (readily available substrates glucose (ATS-G) and acetate (ATS-A), 40 mM/kg each). Four replicates were prepared for each of these condition types, 16 subsamples in total: the first set of two replicates were used for measuring total biogas production, and the second set of two replicates was used for measurement of methane production. After mixing the sediments with additives, the headspaces of the biometers were flushed with N₂ to exclude oxygen. The biometers were then placed in an incubator at 35 °C in the dark for as long as biogas was produced from the sediment, which lasted for about 40 days. Biogas was monitored daily. Analysis of sediment was carried out for all replicates (for per each treatment), immediately before co-substrate addition (IS), and at the end of the experiment. Sediment sampling and analysis were done for the following parameters: chemical oxygen demand, biological oxygen demand after five days, conductivity (Eh), pH, total organic carbon, volatile solids, polycyclic aromatic hydrocarbons, total petroleum hydrocarbons, semi qualitative analysis of other organic pollutants and enzymatic activity. One way ANOVA (analysis of variance) was carried out on all results using untransformed data.

2.3. Analytical methods

The volume of biogas produced was determined by the wetted displacement of water in the measuring cylinder which was mounted on top of the biometer flasks (biometers are shown in the graphical abstract. The CO₂, H₂S, and NH₃ content in the biogas were determined by its absorption in KOH in the measuring cylinder. The volume of gas which was left after absorption in the KOH was referred to as methane (Magen et al., 2014).

COD in sediment was measured by potassium dichromate oxidation method; test measures the oxygen equivalent of the amount of organic matter oxidizable by potassium dichromate in a 50% sulfuric acid solution (AWWA-APHA-WEF, 2012). BOD₅ was measured by respirometric method using B.O.D. System 6, VELP Scientifica according to the standard method AWWA-APHA-WEF,

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