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A batch assay to measure microbial hydrogen sulfide production from sulfur-containing solid wastes



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- A lab-scale assay to estimate H₂S production from solid wastes was developed.
- High H₂S concentrations depressed both methane production and sulfate reduction.
- Base traps to sequester H_2S reduced its toxic effect.
- H₂S production potential of different solid wastes was measured.
- Not all sulfur in solid wastes is converted to H₂S through biotransformation.



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ABSTRACT

Large volumes of sulfur-containing wastes enter municipal solid waste landfills each year. Under the anaerobic conditions that prevail in landfills, oxidized forms of sulfur, primarily sulfate, are converted to sulfide. Hydrogen sulfide (H₂S) is corrosive to landfill gas collection and treatment systems, and its presence in landfill gas often necessitates the installation of expensive removal systems. For landfill operators to understand the cost of managing sulfur-containing wastes, an estimate of the H₂S production potential is needed. The objective of this study was to develop and demonstrate a biochemical sulfide potential (BSP) test to measure the amount of H₂S produced by different types of sulfur-containing wastes in a relatively fast (30 days) and inexpensive (125 mL serum bottles) batch assay. This study confirmed the toxic effect of H₂S on both sulfate reduction and methane production in batch systems, and demonstrated that removing accumulated H₂S by base adsorption was effective for mitigating inhibition. H₂S production potentials of coal combustion fly ash, flue gas desulfurization residual, municipal solid waste combustion ash, and construction and demolition waste were determined in BSP assays. After 30 days of incubation, most of the sulfate in the wastes was converted to gaseous or aqueous phase sulfide, with BSPs ranging from 0.8 to 58.8 mL H₂S/g waste, depending on the chemical composition of the samples. Selected samples contained solid phase sulfide which contributed to the measured H₂S yield. A 60 day incubation in selected samples resulted in 39–86% additional sulfide production. H₂S production measured in BSP assays was compared with that measured in simulated landfill reactors and that calculated from chemical analyses. H₂S

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production in BSP assays and in reactors was lower than the stoichiometric values calculated from chemical composition for all wastes tested, demonstrating the importance of assays to estimate the microbial sulfide production potential of sulfur-containing wastes.

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1. Introduction

The disposal of solid wastes in engineered landfills remains a common approach for both municipal solid waste (MSW) and a variety of non-hazardous industrial wastes in the U.S. (U.S. EPA, 2014b). When biodegradable wastes are disposed in landfills that are not actively aerated, a series of anaerobic biological reactions occur, and the main end products, methane (CH₄) and carbon dioxide (CO₂), make up the major fraction of landfill gas. In addition to MSW, landfills often receive industrial wastes, some of which contain sulfur. When the sulfurcontaining wastes are co-disposed with MSW in landfills, the oxidized form of sulfur, mainly sulfate, can be reduced to hydrogen sulfide (H₂S) (Fairweather and Barlaz, 1998; Kijjanapanich et al., 2014; U.S. EPA, 2014a; Yang et al., 2006; Zhang et al., 2013, 2014) (Eq. (1))

Organic matter + $SO_4^{2^-}$ + $H_2O \rightarrow H_2S + CO_2$ (1)

where either MSW or MSW leachate serves as the source of organic matter.

The presence of H₂S in landfill gas is problematic for several reasons. First, it is corrosive to landfill gas collection and treatment systems, leading to increased maintenance costs. Second, H₂S has a low odor threshold (0.02-0.13 ppm_v), and its fugitive emissions may result in odor complaints (Beauchamp et al., 1984; OSHA, 2005). Third, it is toxic and presents challenges for occupational safety in enclosed areas such as subgrade elements of a leachate collection system (Selene and Chou, 2003; WHO, 2000). While the toxicity of H₂S to humans is well known, its adverse effect on microorganisms is often overlooked in landfill studies. Although no data are available in landfills, studies conducted in other anaerobic systems (e.g., anaerobic digesters, granular sludge treatment reactors) suggest that H₂S can depress microbial methane production, with inhibitory aqueous concentrations in the range of 100-800 mg/L (3-25 mM) (Parkin et al., 1990). Thus, the H₂S generated in landfills may also have a negative effect on landfill methane generation.

Construction and demolition (C&D) waste is one important source of sulfate in landfills (Lee et al., 2006). C&D waste in the U.S. typically contains drywall (wallboard) that is composed of calcium sulfate. While the composition of C&D waste is highly variable, the drywall content has been estimated at 17-27% by weight (Cochran et al., 2007; U.S. EPA, 1998). A second example of sulfur-containing waste is the fines fraction generated at C&D recycling facilities. When C&D waste is processed for recycling, screens are used to remove small debris such as dirt, stones, and broken concrete. Concurrently, some of the brittle wallboard breaks and passes through the screen. Thus, the fines from C&D recycling facilities are enriched with calcium sulfate. Anderson et al. (2010) reported that C&D fines received at eight Northeastern U.S. landfills contained 0.2–12.0% sulfate. Jang and Townsend (2001) reported sulfate concentrations of 890-1600 mg/L in leaching tests on C&D fines. A third type of sulfur-containing wastes is the ash associated with flue gas desulfurization (FGD) in coal combustion. During FGD, sulfur oxide (SO_x) is captured by the addition of lime (calcium hydroxide) or trona (a sodium carbonate mineral), producing sulfate and sulfite salts. FGD residue may be disposed separately or mixed with fly ash from coal-fired power plants. Other sulfur sources include MSW combustion fly ash and organic wastes such as food waste and biosolids from wastewater treatment plants (Ko et al., 2015).

Prediction of H₂S production potential would allow landfill operators to estimate gaseous H₂S concentrations as a function of the quantity of sulfur-containing waste disposed, help to determine the quantity of wastes that can be accepted and evaluate the need to mitigate gas phase H₂S concentrations. However, studies on H₂S production potential from solid wastes are limited. A method for predicting the H₂S content in biogas produced in anaerobic digesters was developed based on the carbon/sulfur ratio of feedstocks (Peu et al., 2012), but this method is limited to wastes with high organic matter content and therefore no supplemental organic carbon was added. As a result, Peu et al.'s method is not applicable to wastes with high sulfur content but little organic carbon, such as C&D waste and ash. Tolaymat et al. (2013) reported on the H₂S production potential of gypsum, which is the major component in C&D wastes contributing to sulfide production, in a 1.25-L reactor. However, tests were conducted under carbon limited conditions representative of a landfill that receives C&D waste only. Asakura (2015) investigated the relationship between sulfate concentration in gypsum and H₂S production, but about half of the tests were conducted under organic carbon limited conditions, while in MSW landfills, carbon-rich conditions are dominant. Sun and Barlaz (2015) presented a procedure to measure the chemically leachable sulfate content for a range of wastes, including residuals from the combustion of coal, wood and MSW, as well as fines from processing C&D waste. However, the relationship between sulfate content, its mineral form, and the rate and extent of sulfate conversion in landfills has not been established. Given the limitations of these existing methods, an assay to measure the sulfide production potential that is more broadly applicable to wastes with varying C/S ratios is required.

Predicting H₂S production potential is challenging due to the fact that H₂S is also toxic to sulfate reduction (Azabou et al., 2005). In systems where H₂S accumulates, sulfate reduction may be inhibited, resulting in an underestimate of H₂S production potential. In other anaerobic aqueous matrices not limited by organic carbon, aqueous concentrations inhibitory to sulfate reducers have been reported in the range of 64 to 550 mg H₂S/L (2–17 mM) (Okabe et al., 1995 and references therein). Reis et al. (1992) demonstrated that sulfate reduction with lactate as the carbon source stopped when the aqueous sulfide concentration reached ~15 mM, and resumed when the sulfide was stripped to below 1 mM.

This study was conducted to (1) develop a laboratory protocol to estimate the H_2S production potential of sulfur-containing solid wastes without inhibition to the sulfate reducing microbes and under conditions where the availability of an electron donor is not limiting, (2) compare results from this method with data from larger (8L) reactors simulating landfill systems and from chemical extractions, and (3) determine the inhibitory concentration of H_2S on microbial methane production in batch systems.

2. Material and methods

2.1. Test assays for H₂S production

A laboratory-scale batch test protocol was developed using serum bottles to measure the H_2S production potential of sulfur-containing wastes. This protocol will be referred to as a biochemical sulfide potential (BSP) test, analogous to the biochemical methane potential (BMP) test (Shelton and Tiedje, 1984). Compared to larger scale reactor tests, the BSP test is economical, well-controlled, and can be completed in a relatively short period of time, although it has the disadvantage of having a relatively high liquid to solid ratio not representative of landfill conditions.

The BSP procedure was modified from the BMP test (Wang et al., 1994). Each serum bottle included a sulfur-containing solid waste, Download English Version:

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