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Emissions and Control of Hydrogen Sulfide at Landfills: A Review

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Hydrogen sulfide (H_2S) in landfill gas, formed with the biodegradation of municipal solid waste, is a major odorous component in a landfill. It poses a potential risk to humans and causes odor problems and complaints by the residents near landfills. Many studies have been conducted on landfill gas qualities. Also, various H_2S control technologies have been used in different industrial sectors. However, comprehensive reviews on H_2S issues in landfill gas are rare. An understanding of the formation of H_2S encountered in landfill gas and the emission at which it occurs helps assess risk and problems. The authors review recent publications from different perspectives on H_2S odor in landfills, including H_2S generation the impact on the environment and human health, H_2S levels in the landfill environment, and H_2S control technologies at landfills.

KEY WORDS: hydrogen sulfide, landfills, odor emission, control technologies, environmental and health impacts

1. INTRODUCTION

Landfills are one of the major sources of odorous nuisance in the environment and can cause many complaints from residents living near the facilities (Sadowska-Rociek et al., 2009; Dincer and Muezzinoglu, 2006). To a large extent, the odor problem became one of the main reasons for public opposition

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to locating landfill sites (Asakura et al., 2010). The emission of odorous gases from landfills mainly results from the generation of gaseous compounds during the biodegradation of landfilled waste (Dincer and Muezzinoglu, 2006; ElFadel et al., 1997). More than 100 odorous compounds have been identified as contributors to landfill odors, including hydrogen sulfide (H_2S), methyl mercaptan (CH_3SH), dimethyl sulfide ($(\text{CH}_3)_2\text{S}$), and ammonia (NH_3 ; He et al., 2012; Kim et al., 2006b; Ying et al., 2012; Kim, 2006). Among the sulfur compounds, H_2S has been identified as a major contributor to odors at landfills (Kim et al., 2006a; Lee et al., 2006; Kim et al., 2005).

The emission of H_2S from landfills has become a growing environmental concern (He et al. et al., 2012; Plaza et al. et al., 2007; Xu et al., 2010b; Sironi et al., 2005). H_2S is a poisonous, flammable, colorless gas with a characteristic odor of rotten eggs. It is known that H_2S poses flammability in air at concentrations between 4.3–45.5% by volume. The odor threshold of H_2S ranges from 0.0005 to 0.3 ppm (Agency for Toxic Substances and Disease Registry, 2008). The average odor threshold, which is defined as detection by 50% of the population, is reported to be 7–9 parts per billion (ppb; Ruth, 1986). Because of the low threshold, landfill gas emitted with a low concentration of H_2S can cause odor complaints by neighbored residents. Not only for odor control but also for landfill gas-to-energy projects, H_2S must be removed from the gas because of its corrosiveness to equipment (Fairweather and Barlaz, 1998). Also, H_2S poses a potential health and safety threat to people living or working near facilities releasing the compound (Ying et al., 2012; Li et al., 2008).

Although many studies have been conducted to control H_2S in off-gases from various sources, few comprehensive reviews have been done on landfill H_2S emission and control. To address H_2S odor problems in a landfill, it is necessary to understand H_2S generation and to manage the landfill properly. This paper reviews literature on H_2S in landfills, including H_2S generation, the risk of H_2S exposure, H_2S emission from landfills, and H_2S control technologies for landfills.

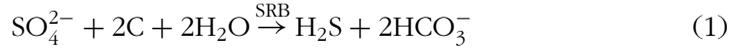
2. H_2S GENERATION IN LANDFILLS

2.1 Generation of H_2S by Sulfate-Reducing Bacteria

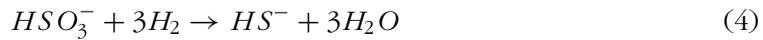
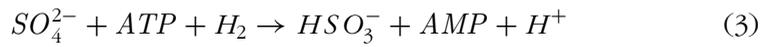
Microorganisms are able to produce H_2S from inorganic sulfur compounds, sulfate and sulfite, through the sulfate assimilation pathway (Thomas and Surdin-Kerjan, 1997) or organic sulfur compounds, and through the catabolism of cysteine and glutathione (Landaud et al., 2008; Winter and Curtin, 2012). Hydrogen sulfide can be produced from both bacterial reduction of sulfate and decomposition of sulfur-containing organic constituents under anaerobic conditions in landfills.

The generation of H_2S from dissimilatory sulfate reduction is carried out by a specialized group of anaerobes, called sulfate-reducing bacteria

(SRB). SRB are widely spread in sewer systems, wastewater sludge, and landfills (Hao, 2003). The population of SRB in a lab-scale simulated landfill reached 8.1×10^6 cells per day (Fairweather and Barlaz, 1998). During the dissimilatory reduction process, SRB obtain energy for growth by oxidation of organic substrates and use sulfate as the terminal electron acceptor as Equation 1 (Liamleam and Annachhatre, 2007):



The generation of H_2S through biological sulfate reduction by SRB mainly include three steps: sulfate activation (Equation 2), sulfate reduction to sulfite (Equation 3) and sulfide formation (Equation 4; Barton and Plunkett, 2002):



SRB are obligate anaerobic bacteria and can only use sulfate in the absence of electron acceptors such as O_2 , H_2O_2 , Cl_2 , NO_3^- , and Fe^{3+} . Postgate (1984) reported that in order to cultivate SRB, the redox potential of the environment must start around -100 mV (Postgate, 1984). The optimal ORP for H_2S generation was reported in the range of -100 mV to -50 mV (Boon, 1995). SRB prefer an environment around a pH of 7 and H_2S generation are usually inhibited at pH values lower than 5.5 or higher than 9 (Hao, 2003).

In addition to sulfate, most SRB can use sulfite or thiosulfate as electron acceptors. In the presence of thiosulfate or sulfite, SRB first disproportionates the thiosulfate or sulfite compounds to sulfate and then oxidizes the organic substrates with the newly formed sulfate as shown in Equations 5 and 6 (Li et al., 2008).



H_2S is also one of the end products resulting from the action of many varieties of bacteria on organic material containing protein, such as food waste. Amino acids containing sulfur, such as, cysteine ($\text{HS-CH}_2\text{-CH(NH}_2\text{)-COOH}$) and methionine ($\text{CH}_3\text{-S-CH}_2\text{-CH(NH}_2\text{)-COOH}$) are recognized as the chief sources of H_2S formed during the putrefaction of food waste (Almy, 1925). Under anaerobic conditions, waste containing organic sulfur compounds (cysteine and methionine) can be decomposed through the desulfurization process (Peu et al., 2012; Schieder et al., 2003; Chen et al., 2011). During the process, sulfur is desulfurized from organic molecules to

generate H₂S gas, as shown in Equation 7.



The putrefaction can play a role in H₂S production in some conditions. However, it is generally believed that the putrefactive H₂S production rate is insignificant compared with inorganic sulfate reduction as discussed above (Li et al., 2008; He et al., 2011).

2.2 Source of Sulfur in Landfills

The main source of sulfur in a landfill includes gypsum drywell (or wall-board), sulfur-containing organic waste (mainly food waste and paper), and sludge of wastewater treatment plants (WWTPs). One of the largest sulfur sources in landfills is gypsum drywall, especially in C&D debris landfills. The utilization of gypsum drywall by SRB to produce H₂S in landfills has been studied (Xu et al., 2010b; Fairweather and Barlaz, 1998). Gypsum is a mineral composed of calcium sulfate (CaSO₄) and water (H₂O; approximately 90% CaSO₄·2H₂O and 10% paper) and is widely used in commercial and residential construction and renovation for interior walls because it is a good fire barrier. Although waste gypsum drywall can be recycled, it often ends up in landfills (Musson et al., 2008).

Food waste is one of the main components of MSW (Fang et al., 2012). Some food waste, such as dairy products and meat products, contain organic sulfur compounds. Currently, the most common method of food waste treatment is landfilling, especially in developing countries (Fang et al., 2012). Typical sulfur content of food waste is known to be as much as 0.4% (by mass) by ultimate analysis (measured as C, H, N, O, S, and ash; Tchobanoglous et al., 1993). Under anaerobic conditions, organic sulfur compounds, such as cysteine and methionine, are biologically decomposed to produce H₂S (Ryckebosch et al., 2011; Wu, Wang et al., 2010; Wang et al., 2001). Paper waste also contains sulfate as measured up to a few tens of millimoles per kilogram of waste paper (Gurijala and Suflita, 1993). Watanabe et al. (2004) measured the sulfur of waste paper ranging from 0.40 to 1.7 mg/g (dry weight of waste paper) in Osaka, Japan. If landfilled MSW contains a large portion of waste paper, the waste paper can be considered an important sulfur source in landfills.

Wastewater sludge is a by-product generated in a WWTP. It is common practice to codispose sludge with MSW in landfills (Fairweather and Barlaz, 1998; Fang, Yang, Cen et al., 2012; Çinar et al., 2004). The sulfur content of wastewater sludge ranges from very little to 2.3% on a dry solid basis (Dewil et al., 2009). As sludge is generally dewatered prior to being landfilled, some chemical conditioners are often added to enhance sludge dewaterability. The use of the conditioners can affect H₂S generation in landfills. Liu et al.

(2012) conducted research to investigate the odorous gas compounds emission from different sludge conditioning processes. The results indicated that H₂S generation decreased with the addition of CaO as conditioner. However, the addition of H₂SO₄ and FeSO₄ for pH adjustment and Fenton reagent not only increased the sulfur content in sludge, but also accelerated the decomposition of sulfur-containing proteins (Liu et al., 2012).

2.3 Factors Affecting the Biological Formation of H₂S

Because H₂S generation mainly is a result of biological processes, its generation is firmly related to the growth of SRB. D'Imporzano et al. (2008) reported there was a good regression between biological activity and the odor molecules ($R^2 = 0.991$) during biodegradation of organic matters. The activity of SRB in landfills depends on many different factors, such as moisture content, temperature, and pH (D'Imporzano et al., 2008).

Moisture is essential for SRB growth. Bergersen and Haarstad (2008) observed that H₂S production significantly increased when waste is wet and water-saturated. It was observed that after a rainfall event the ambient H₂S concentrations were generally higher than those under normal conditions in landfills. The infiltration of rainwater into dry wastes can stimulate the activity of SRB resulting in H₂S generation (Johnson, 1986).

Temperature is another important factor for SRB growth. Sulfate reduction rates typically increase 2- to 3.9-fold with a temperature increase of 10°C (Widdle, 1988). The optimum temperature for most pure cultures of SRB ranges from 28°C to 32°C. The lower optima among SRB are observed with some *Desulfobacterium* strains and a curved *Desulfobacter* strain at 24–28°C, and the highest is around 70°C for *Thermodesulfobacterium commune* (Hao, 2003). Hao et al. (1996) reported that most species of SRB died rapidly at temperatures above 45°C. Xu et al. (2010b) observed that in a field study, H₂S generation underneath a compost layer was reduced to some extent, due to high temperature (about 51°C) in the layer. H₂S production from a landfill varies with seasons likely because of seasonal temperature variation. Kim (2006) measured seasonal variation of H₂S emission from two landfill sites in South Korea. The results indicated that H₂S had the highest emission rate during summer (Kim, 2006). Vasarevicius (2011) also observed that H₂S concentrations in the air near a landfill were related with seasonal changes of ambient temperature, because the generation of H₂S in the top layer of waste was affected by air temperature (Vasarevicius, 2011).

SRB prefer a neutral pH environment and are usually inhibited at pH values lower than 5.5 or higher than 9 (Hao et al., 1996). Research demonstrated that SRB can survive at low pH conditions (less than 4.5) like peat bogs and acid mine water. It was hypothesized that in acidic environments, SRB were present in microniches. In addition, the effect of low pH could be minimized because the sulfate reduction process generates additional bicarbonate alkalinity (Hao, 2003). Changing the pH value by adding acid or

alkali was reported as a method of inhibiting SRB in industrial plants (Postgate, 1984). Since pH of waste may vary with codisposed wastes, it has been recommended that prior to disposing gypsum drywall in landfills, powdered lime could be added to mix with gypsum drywall to reduce H₂S generation (Chalvatzaki and Lazaridis, 2010; Xu et al., 2011).

3. H₂S IN LANDFILL ENVIRONMENT

3.1 Levels of H₂S in Landfill Environments

H₂S measurements have been reported as concentrations in air, in landfill gas samples or flux rates from landfill surfaces. However, there is no standard method to measure H₂S emission from landfills. Various sampling methods have been used to collect landfill gas samples in previous studies. Gas samples often are collected from landfill gas collection systems (active or passive venting systems), the inside of landfills using soil gas probes installed through the landfill cover, ambient air near or over the site, and/or a chamber flux meter to capture H₂S emitted from cover soil (Lee et al., 2006; Xu et al., 2010b; Eun et al., 2007). H₂S measured with a single sampling technique probably is insufficient to describe the entire H₂S emission from a landfill because the levels of H₂S measured vary with different sampling techniques and sampling points. H₂S concentrations in gas collection systems can represent a relative amount of H₂S generation but does not include fugitive emissions because the gas in the collection system usually is treated before being released. Also, the forced extraction of landfill gas can be diluted landfill gas with ambient air that moves through the landfill cover by suction pressure. H₂S measurement using soil gas probes and flux chambers covers only a small portion of a landfill surface. The measured levels may not be representative for the whole area of the landfill surface. Direct measurements of H₂S from ambient air are easily influenced by meteorological conditions (temperature, atmospheric pressure, and precipitation). To properly measure H₂S emissions from a landfill, multiple measurement techniques may be required to understand landfill conditions and metrological parameters.

3.1.1 H₂S CONCENTRATIONS IN LANDFILL GAS

As summarized in Table 1, H₂S concentrations in landfill gas samples often range from under detection limits to thousands part per million. The UK Environment Agency (2002) reviewed trace landfill gas data from 79 sites. In the reviewed data, H₂S concentration was reported as high as about 70,000 ppm. This magnitude of H₂S concentration is not often observed in MSW landfill gases. The median concentration and average concentration of H₂S was 2.0 ppm (2.8 mg/m³) and 96.6 ppm (134.2 mg/m³), respectively (Parker et al., 2002). However, 68% of H₂S concentrations reviewed were under its detection limit. USEPA (1995) conducted an extensive measurement of landfill gases emitted from the Fresh Kills landfill at New York. The

TABLE 1. Health effects of hydrogen sulfide (Agency for Toxic Substances and Disease Registry, 2006)

Exposure limits (ppm)	Health effects
0.008–0.2	Olfactory threshold –“rotten eggs” smell detectable
20	Sense of smell to gas lost Concentrations tolerated for some hours without harm
20–50	Eye irritation
50	Prolonged exposure may cause pharyngitis and bronchitis
60	Prolonged exposure may cause conjunctivitis and eye pain
150	Irritation of upper respiratory tract; sense of smell lost
250	Pulmonary edema with risk of death
500	Very dangerous, evacuation should occur well below this level
1000	Loss of consciousness occurs
1000–2000	Immediate collapse with paralysis of respiration

concentrations of H₂S measured from passive vents at the Fresh Kills landfill ranged from 0.11 to 220 ppm. The ratio of H₂S production rate to methane production from the landfill was estimated to be 2.53×10^{-5} (by mass production rate). The emissions were measured from landfill surface and passive vents and the landfill gas collection system.

3.1.2 H₂S EMISSION FROM LANDFILLS

Few field studies have been performed for H₂S emission. In the U.S. Environmental Protection Agency (USEPA) Fresh Kills landfill study, landfill gas samples were collected from four sections (two closed sections and two active sections) of the landfill using passive vent wells, flux chamber meters, and gas collection system (U. S. Environmental Protection Agency, 1995). Total emission rate for the Fresh Kills landfill was estimated at 0.453 g/s (emissions from landfill surface and passive vents). The observed surface emission ranged from below detection limit (< 0.29 mg/m²-day) to 10,498 mg/m²-day. The H₂S emission factor based on flux chamber measurement was estimated to be 4.34×10^{-10} g/s-kg of MSW. The magnitude of H₂S emission from C&D waste landfill is greater than in MSW landfill if C&D waste contains a high degree of sulfur-containing waste, like gypsum drywall. Eun et al. (2007) observed that H₂S emission rate was proportional to the gypsum drywall content. From measurements of five C&D landfills, they reported that the inverse distance weighting (IDW) mean emission rate ranged from 0.192 (± 0.34) to 1.76 (± 4.15) mg/m²-day. Colledge (2008) presented continuous monitoring data of H₂S from a C&D landfill site in Ohio, USA. The researcher estimated flux rates using Box model and data measured in the landfill to range from 0.005 mg/m²-day to 449.28 mg/m²-day.

3.1.3 LEVEL OF H₂S CONCENTRATION IN AMBIENT AIR AROUND LANDFILLS

H₂S is ubiquitous in ambient air, and concentrations from natural sources range between 0.11 and 0.33 ppb. Typical H₂S background concentrations

are known as less than 1 ppb (Agency for Toxic Substances and Disease Registry, 2006). In literature, H₂S concentrations in ambient air samples collected from landfill areas are much greater than the background level range. Table 1 shows ranges of H₂S concentrations from under detection limits to low 10s ppb. Even though there are some incidences of H₂S exposure with offensive levels, the exposure may not be chronic due to meteorological conditions strongly influencing the level of H₂S in the ambient air near an H₂S source.

3.2 Environmental Factors Influencing H₂S Emission

H₂S emission from a landfill is affected by various environmental factors, such as landfill geometry, waste characteristics (age and composition), the presence of air in the landfill (landfill aeration), and weather conditions. Geometrical H₂S emission from landfills can be related to waste landfill geometry such as area fill, trench fill, and valley fill (canyon fill). The preliminary H₂S production areas in an MSW landfill are an active waste tipping area. Barry et al. (2003) measured landfill surface flux emissions for 21 active landfills in the United Kingdom and found that the emission from landfill sides was greater than the top sides. In contrast, a USEPA report indicated that the average emission factor determined by flux chamber measurements on the side was similar to that on the top, but the total emission rate from the top was greater than that from the side because the surface of the top area was larger than that of the side (U. S. Environmental Protection Agency, 1995). However, the source of H₂S emission is not only landfilled waste itself but also other sources such as waste process area and leachate ponds in a landfill site. Fang et al. (2012) measured H₂S concentration in ambient air samples collected from eight locations in a landfill site accepting MSW and sewage sludge. They also collected gas samples from the gas extraction well of a closed landfill area. High levels of H₂S were measured from the active landfill tipping area (109 ppb), sludge discharging area (48.9 ppb), and leachate storage pool (53.0 ppb).

Waste age is a critical factor for H₂S generation. H₂S levels in gas samples from fresh waste materials are higher than those of old waste. Kim (2006) observed that the H₂S concentrations ranged from 0.4 to 524 ppm in gas samples collected vent pipes of a young landfill (<5 years old, lined landfill) but from 0.23 to 10.2 ppb in an old landfill (5–23-year-old, unlined landfill). In a UKEPA study, the highest concentrations of H₂S were also reported in the early stages of refuse compositions, and the levels of H₂S concentration gradually dropped over time (Parker et al., 2002). This observation may be due to the depletion of sulfur compounds over time by reductive conversions.

The presence of oxygen inside a landfill can influence the formation of H₂S in the landfill. Operating a landfill in aerobic conditions (adding air into

the landfill) is not common. However, aerating waste has been used to abate odor problems during remediation activities (Jacobs et al., 2003). Because hydrogen sulfide is produced under reduced conditions, the aeration creates unfavorable conditions for sulfur reducing bacteria and oxidizes H₂S. Powell et al. (2006) measured hydrogen sulfide reduction in a bioreactor landfill by air injection. The authors listed the causes of the reduction as a change in the reduced conditions by aeration and dilution and decomposition of H₂S by added air.

Meteorological conditions (temperature, atmospheric pressure, wind, and precipitation) influence landfill surface gas emission, H₂S dispersion in the air, and the formation of H₂S in a landfill (McBain et al., 2005; Poulsen et al., 2003; Christophersen et al., 2001). The sensitivity of gas emission from a landfill with change of atmospheric pressure has been studied. Typically, gas emission rates show negative correlation to atmospheric pressure change. Rain events can provide moisture SRB in a landfill, eventually. The moisture content of landfill soil cover also is a key factor of the gas emission. Landfill gas emission decreases with increasing cover soil moisture content (Christophersen et al., 2001). Due to high water solubility of H₂S, when H₂S gas passes through wet cover soil, it would tend to dissolve in water, temporarily reducing H₂S emissions. As water evaporates in cover soils, the accumulated H₂S gas is emitted, resulting in higher ambient H₂S concentration (Panza and Belgiorno, 2010). The impact of temperature on SRB was described in the previous section. Higher H₂S concentrations of the ambient air samples inside landfill footprints are reported during summer than those during fall.

4. IMPACTS ON THE ENVIRONMENT AND HUMAN HEALTH

4.1 Environmental Impact

4.1.1 FORMATION OF AIR POLLUTIONS

The emission of H₂S from landfills can result in the formation of another air pollutant, sulfur dioxide (SO₂). The released H₂S can react with atomic oxygen (O), oxygen (O₂), or ozone (O₃) to form SO₂, as shown by Equation 8 (Bibbero and Young, 1974). SO₂ also has a pungent odor, but its odor threshold (1 ppm) is higher than H₂S (Agency for Toxic Substances and Disease Registry, 2006). In general, the lifetime of H₂S before conversion to SO₂ is on the order of hours (Seinfeld, 1975). SO₂ is eventually removed from the atmosphere through absorption by plants and soils or through precipitation.



In addition, H₂S contained in landfill gas can also cause equipment damage (e.g., corrosion) in the facilities using landfill gas as an energy

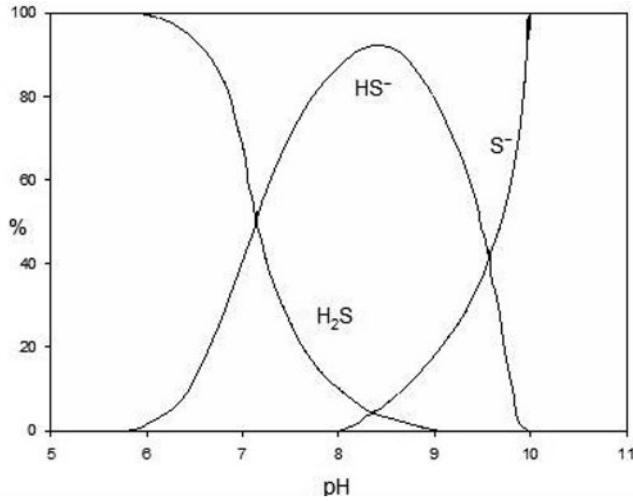


FIGURE 1. Ionic species of hydrogen sulfide (Hao et al., 1996).

source. It was reported that damage caused by H₂S scale is serious in many facilities. Therefore, pretreatment processes are required to remove any H₂S before landfill gas is used to produce heat and electricity (Nam et al., 2011).

4.1.2 GROUNDWATER CONTAMINATION

Compared to other chemical species in landfill gas, H₂S has relatively high water solubility (about 4,370 mg/L at 0°C, and 1,860 mg/L at 40°C). A relatively large amount of H₂S can dissolve into landfill leachate, an important reservoir of H₂S. Bergersen and Haarstad (2008) reported that leachate from Norwegian landfills emitted H₂S with concentrations as high as 5,000 ppm. The existence of ionic species of H₂S in water is highly dependent on pH. Major sulfur species present in water are H₂S, bisulfide (HS⁻) and sulfide (S²⁻). Changes in the concentrations of H₂S, HS⁻, and S²⁻ are highly affected by pH changes. As shown in Figure 1, H₂S is the dominant species at a low pH between 5.0 and 6.0. At a pH of 7.0, the ratio of the concentration of aqueous H₂S to HS⁻ ion is approximately 1. When the pH is increased between 7.0 and 9.0, the HS⁻ becomes the dominant species. At a high pH greater than 9.0, the S²⁻ becomes the dominant species (Thompson et al., 1995).

The generation of H₂S from landfills can cause a serious threat to groundwater resources by two potential pathways to groundwater. Dissolved H₂S can migrate downward with leachate into groundwater, especially in old landfills without bottom liner systems. Otherwise, H₂S in landfill gas can travel through the unsaturated zone and come into contact with groundwater

(ElFadel et al., 1997). Groundwater contaminated by H₂S has an undesirable taste with a rotten egg smell. Ingestion of H₂S contaminated water can result in stomach discomfort, nausea, and vomiting (Thompson et al., 1995; Edwards et al., 2011).

4.2 Human Health Impacts

In addition to the odor problem, H₂S poses adverse impacts on human health (Flynn, 1998; Theakston, 2000; Selene and Chou, 2003; Campagna et al., 2003). The health hazard depends upon H₂S exposure duration, frequency, and concentration. Exposure to low concentrations of H₂S can cause difficulty in breathing for some asthmatics and irritation to the eyes, nose, and throat (Kansas Department of Health & Environment, 2010). Eye irritation has been described as the first health effect to manifest at low concentrations. In community settings, following short-term exposure, 25 ppb H₂S appears to be the lowest concentration observed to irritate the eyes. Serious eye damage is caused by a concentration of 50 ppm (70 mg/m³; Lambert et al., 2006). As H₂S concentrations increase beyond 100 ppm, they quickly paralyze the olfactory senses so that the odor can no longer be recognized as a warning signal and it begins to affect the whole body. Exposure to high concentrations results in depression of the central nervous system and loss of consciousness. Over 500 ppm, H₂S can cause convulsions, respiratory arrest, coma, and even death (Flynn, 1998). Table 2 shows the health effects of respiratory exposure of H₂S at various concentrations.

The health and quality of life in landfill-neighbored communities and landfill workers has been investigated. Heaney et al. (2011) observed that there was a positive association of low-level H₂S exposure from a landfill with reports of malodor when the wind was blowing from the landfill toward the community. The Agency for Toxic Substances and Disease Registry in the United States conducted an exposure investigation of odor complaints from residents living within about a 2.4 km radius of the APAC-Reno landfill. The reported health problems associated with odors included difficulty breathing, asthma exacerbations, headaches, and nausea. Dzaman et al. (2009) investigated the functioning of the taste and smell senses in workers employed at a landfill. The results showed that after working hours, the percentage of workers with smell disorders increased from 21.7% to 38.4% in the group of landfill workers because of the exposure to odor gases in the landfill. Fielder et al. (2000) compared the health conditions of residents living near the Nant-y-Gwyddon landfill site to a population with similar socioeconomic status. The monitoring data demonstrated that the site was responsible for the odor problem, and there was an increased maternal risk of having a baby with a congenital abnormality in residents near the landfill.

TABLE 2. Summary of H₂S levels measured from landfill sites

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference
A landfill near Shanghai, China	MSW (7,600 tons/day) and sewage sludge (2,500 tons/day). speed	May 12 (spring), 2011 21°C, 55–65% RH, 5.7–6.9 m/s wind	Ambient air	TU: active landfill tipping area-U TD: active landfill tipping area-D LS: leachate storage pool LD: leachate disposal pool SD1: sludge discharge area SD2: sludge disposal work place SD3: sludge sun drying site SL: sludge landfill site	TU: 109 ppb TD: 3.9 ppb LS: 53.0 ppb LD: 7.1 ppb SD1: 6.6 ppb SD2: 48.9 ppb SD3: 3.3 ppb SL 2:9 ppb	(Fang et al., 2012)
Jerubaicai landfill, Plunge district in Lithuania	MSW, 14900 m ³ (2.5 thick) with 20–25 cm soil cover	August, November, February, and April	Ambient air	Center of the site, 1m above surface	900 ppb (February) 8600 ppb (August)	(Vasarevicius, 2011)
Five landfills in Beijing, China	MSW landfill	April	Ambient air	Chinese National Criteria, GB3095-1996) and Manual Methods for Ambient Air quality Monitoring (China Environmental Bureau HJ/T194-2005)	< 3.6 ppb	(Li et al., 2008)

Daegu landfill, S. Korea	MSW Daegu Bangchun	January, 2004 Air temperature: -1.2-1.8°C	Ambient air	Landfill boundary and a near residential area These eight locations were representing (1) a residential site that is approximately 1km away, (2) the entrance point of the landfill, (3-7) five points evenly distributed on the landfill borderline, and (8) one in the central landfill area (covered area).	1: 0.001-2.1 ppb 2: 0.026-0.76 ppb 3: 0.25-1.19 ppb 4: 0.78-2.33 ppb 5: 0.61-7.57 ppb 6: 1.50-11.29 ppb 7: 0.43-14.49 ppb 8: 5.46-27.01 ppb	(Shon et al.,
Two landfills in S. Korea	Gunsan landfill (G-LF) Household waste Donghae landfill (D-LF) House hold waste + 20% coal ash	G-LF: May (P1), August (P2), and December (P3) D-LF: May (P1'), July(P2'), October (P3'), and December (P4')	Ambient air (1 m above the ground)	G-LF: G1: The central location, G2: a residential area about 2km away from the LF, G3: a suburban residential area about 1km away from the LF, and G4: a coastal area G4within 3 km of the LF. D-LF: D1: The central location of the LF (i.e., D1), D2- D4: the 3 sites are all within 3 km east of the LF.	G1: < 0.052-1.68 ppb G2: < 0.052-0.297 ppb G3: < 0.052-0.109 ppb G4: < 0.052-0.168 ppb D1:0.0 46-5.396 ppb D2: 0.023-0.101 ppb D3: 0.015-0.135 ppb D4: 0.016-0.194 ppb	(Song et al.,
Florida, USA	10 C&D landfills		Ambient air	Directly measured from ambient air using an analyzer	Average 3-4,000 ppb	(Lee et al., 2006)

(Continued on next page)

TABLE 2. Summary of H₂S levels measured from landfill sites (*Continued*)

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference
Belgium	NA	NA	Ambient air in waste handling and discharging site	Sampling ambient air by pumping 20 dm ³ through adsorbent trap (sampling was performed at 1.5 m height)-	Not detected	(Termonia and Termonia, 1999)
Tianziling landfill, Hangzhou City, China	Domestic waste (not accepted: medical, industrial, and hazardous wastes)	July, 2007–June, 2009 Air temperature: 5.2–28.5°C	Ambient air samples at 6 sampling points	Dumping area Wastewater treatment plant Office area Factory boundary Residential area (affected area by the odor) 165 platform (old dumping site)	369.1 ± 16.8 ppb 350.3 ± 12.6 ppb 147.1 ± 6.6 ppb 128.0 ± 4.1 ppb 40.6 ± 2.0 ppb 129.1 ± 18.0 ppb	(Ying et al., 2012)
Warren recycling landfill, Ohio, USA.	C&D, the operation began in 1994.	Oct., 2003–Sep., 2004 June 9–August 15, 2004 Sep 8–Oct 6, 2004	Ambient air (continuous). Hourly averages (up to 7500 observation in 337 day period)	Two sampling stations (north [N] and northwest [NW]), sampling inlet located at the elevation of approximately 3.5 meters. Six additional sampling locations (Site 1 to 6) Data were normalized to STP (25°C and 29.92”Hg)	N: 0–87 ppb NW: 0–178 ppb Site 1: 0–141 ppb Site 2: 0–71 ppb Site 3: 0–15 ppb Site 4: 0–80 ppb Site 5: 0–9 ppb Site 6: 0–44 ppb	(Colledge, 2008)

MSW regional landfill, NC, USA.	MSW	Jan.- Feb, 2009 and Sep.-Nov. 2009	Ambient air	The community boundary near the landfill using continuous monitoring	Range: 0–14.86 ppb The 15-min average H ₂ S = 0.22± 0.52 ppb	(Heaney et al., 2011)
In the Valencianne Community, Spain		July, and November, 2009	Ambient air	Inside of landfill footprint Summer Autumn Outside of landfill Summer Autumn	3.6–16.3 ppb 2.5–8.8 ppb 0.2–4.3 ppb 0.01–2.9 ppb	(Font et al., 2010)
Jerubaiciai landfill, Plunge district in Lithuania	MSW	August, November, February, and April	Landfill gas	Two gas wells (G1 and G2)	August: 7.4 ppm (G1), 8.6 ppm (G2) November: 5.5 ppm (G1), 8.1 ppm (G2) February: 2.5 ppm (G1), 6.0 ppm (G2) April: 5.0 ppm (G1), 7.3 ppm (G2)	(Vasarevicius, 2011)

(Continued on next page)

TABLE 2. Summary of H₂S levels measured from landfill sites (*Continued*)

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference
A landfill near Shanghai, China	MSW (7,600 tons/day) and sewage sludge (2,500 tons/day).	May 12 (Spring), 2011. 21°C, 55–65% RH, 5.7–6.9 m/s wind speed	Two parallel samples were collected in each sampling point. All Tedlar bags were placed in a vacuum-generating container. Using a vacuum pump connected to the container, five liters of gas sample were drawn directly into the Tedlar bag via Teflon tubing with a 5 mm outside diameter inserted into the vent system. These samples were then transported immediately to the lab for analysis (within 24 h) to minimize the loss of reduced sulfur compounds due to storage.	GW: Closed landfill site, gas extraction wells	GW 16.1 ppm	(Fang et al., 2012)

Four MSW landfills MSW in S. Korea	9 Aug.2003 22-23 July 2003 22-23 July 2003 13-16 Jan. 2004	Landfill gas	Ventpipe (from moderately fresh burial locations) W: Woon Jung landfill (active) N: No Hyung landfill (closed) H: Hoei Chun landfill (active) B: Bang Chun landfill (active)	0.212-681.370 ppm (W) 0.336 ppb ppm (N) 854.580-5,142.9 ppm (H) 89.132-143.091 ppm (B)	(Kim et al., 2005)
Dong Hae, S. Korea	May, July, November, and December	Landfill gas (venting pipe)	YLF: young <5 years, area: 0.35 km ² OLF: old, 5-23 years, area: 0.32 km ²	YLF (ppm) OLF (ppm) May: 2.629-124.41 0.0002-0.002 July: 6.848-523.838 0.00055 Nov.: 0.437-280.97 0.0013-0.005 Dec.: 70.280-181.337 0.00063-0.01	(Kim, 2006)

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TABLE 2. Summary of H₂S levels measured from landfill sites (*Continued*)

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference
A landfill near Shanghai, China	MSW (7,600 tons/day) and sewage sludge (2,500 tons/day).	May 12 (Spring), 2011. 21°C, 55–65% RH, 5.7–6.9 m/s wind speed	Two parallel samples were collected in each sampling point. All Tedlar bags were placed in a vacuum-generating container. Using a vacuum pump connected to the container, five liters of gas sample were drawn directly into the Tedlar bag via Teflon tubing with a 5 mm outside diameter inserted into the vent system. These samples were then transported immediately to the lab for analysis (within 24 h) to minimize the loss of reduced sulfur compounds due to storage.	Closed landfill site, gas extraction wells	GW 16.1 ppm	(Fang et al., 2012)

Four MSW landfills, MSW in S. Korea	9 Aug. 2003 22–23 July 2003 22–23 July 2003 13–16 Jan. 2004	Landfill gas	Ventpipe (from moderately fresh burial locations) W: Woon Jung landfill (active) N: No Hyung landfill (closed) H: Hoei Chun landfill (active) B: Bang Chun landfill (active)	0.212–681.370 ppm (W) 0.336 ppb ppm (N) 854.580–5,142.9 ppm (H) 89.132–143.091 ppm (B)	(Kim et al., 2005)
Dong Hae, S. Korea	MSW Young landfill (YLF) Old landfill (OLF)	Landfill gas (venting pipe)	YLF: young <5 years, area: 0.35 km ² OLF: old, 5–23 years, area: 0.32 km ²	YLF (ppm) OLF (ppm) May: 2.629–124.41 0.0002–0.002 July: 6.848–523.838 0.00055 Nov.: 0.437–280.97 0.0013–0.005 Dec.: 70.280–181.337 0.00063–0.01	(Kim, 2006)
Fresh Kills landfill, United States	Residential waste	Landfill gas (concentration [ppm] and emission rate ($\mu\text{g/s}$), surface emission.	Passive vents (H ₂ S concentrations, flow rates) Surface emission (flux chambers) Gas collection system (one active section out of four sections) Extraction well	Passive Vents Section 2/8: 1.21–104 ppm Section 3/4: 0.11–220 ppm Section 1/9: 5.3–35 ppm 15.1–427.5 ppm	(Parker et al., 2002)
Sudokwon landfill, Incheon city in S. Korea	MSW	Landfill gas			(Shin et al., 2002)

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TABLE 2. Summary of H₂S levels measured from landfill sites (*Continued*)

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference
Biovale landfill			Landfill gas	Landfill gas measured from landfill gas collection system	100 ± 10 ppm	(Ducom et al., 2009)
Florida, USA	10 C&D landfills		Landfill gas	Existing gas collection system or soil vapor probe (0.3 m depth)	< 0.003–12,000 ppm	(Lee et al., 2006)
Belgium	NA	NA	Landfill gas (bio-gas)	Sampled at a flare station	0.072 ± 0.029 ppm	(Termonia and Termonia, 1999)
UK codisposal site	MSW (67%) and trade waste (23%) Young waste (3 years) Old waste (17 years)	September and March in 2001 and 2002.	Gas wells	Landfill gas	1.2–5.4 ppm 5.1–16.4 ppm	(Parker et al., 2002)
Five waste disposal sites at Mexico City		September–November, 2003	From drilled holes through the cover or From vent pipes/burners	Operating controlled dump Operating open dump Operating landfill Closed open dump Closed controlled/open dump	18 ± 5 ppm 50 ± 10 ppm 95 ± 10 ppm 120 ± 20 ppm 150 ± 20 ppm	(de la Rosa, Velasco, Rosas and Volke-Sepulveda, 2006)
Shanghai solid waste landfill, Shanghai, China		December, 2007, air temperature 14–18°C	Landfill gas, installing soil gas probes (80 cm depth) under the soil cover.	Cell 18 (< 1 year old) Cell 56 (2 years after the closure) Cell 52 (5 years after the closure) Cell 40 (13 years after the closure)	Cell 18: 84–3,400 ppm Cell 56: 0–67 ppm Cell 52: 0–4 ppm Cell 40: not detected	(Xiaoli et al., 2011)

Location	Site description	Flux chamber	Subsurface	Inverse distance weighted mean	Reference
Florida, USA	Site A: opened in 1991, 4% drywall	Flux chamber	Subsurface	Inverse distance weighted mean	(Eun et al., 2007)
	Site B: opened in 1990, 10% drywall				
	Site C: 35 m deep; 0.6-m final cover				
	Site D: opened early 1980s				
	Site E: closed in 1983				
Dong Hae, S. Korea	MSW	May, July, November, December	YLF: young <5 years, area: 0.35 km ²	YLF: 232–278,211 g/year	(Kim, 2006)
	Young landfill (YLF) Old landfill (OLF)	Total emission rate (estimated using H ₂ S concentrations measured in landfill gas from venting pipes)	OLF: old, 5–23 years, area: 0.32 km ²	OLF: 0.03–3.72 g/year	

(Continued on next page)

TABLE 2. Summary of H₂S levels measured from landfill sites (*Continued*)

Site, Region	Waste composition/Landfill type	Season	Sample type	Sampling location/description	H ₂ S concentration/Emission	Reference	
Fresh Kills landfill, MSW NY, United States		Four sections: 3/4 section: closed (being retrofitted with: 1) passive vents, 2) PVC cover, and active gas collection wells. 2/8 section: closed (being retrofitted with: 1) passive vents, 2) PVC cover, and active gas collection wells.) 6/7 section: active (covered with 0.3–0.6 m soil cover, no active gas collection, passive vents or PVC cover.) 1/9 section: active (active gas collection cover 2/3 of the section)	Landfill gas from passive vents or using flux chamber	Passive vents (emission rates were calculated with H ₂ S concentrations and flow rates)	Section 2/8: (n = 10) 0.0, 30–3.99 mg/m ² -day Section 3/4: (n = 11) 0–7.55 mg/m ² -day Section 1/9: (n = 10) 0.19–10.94 mg/m ² -day Section 6/7 (n = 43): 0.45–10497.6 mg/m ² -day	(Parker et al., 2002)	
					Surface emission (flux chambers)		

Warren recycling landfill, Ohio, USA.	C&D, three phases. 70 acres for each phase. The operation began in 1994.	Oct. 17, 2003-Sep 17, 2004	Ambient air	AERMOD steady-state plume model was used to estimate downwind concentrations from landfill area sources. A simple box model (Box A-D) was used to back-calculate flux rates of on-site area sources. Continuous 5-minute ambient H ₂ S measurements were used.	Box A: 27.13 mg/m ² -day (Colledge, 2008) Box B: 29.72 mg/m ² -day Box C: 19.01 mg/m ² -day Box D: 210.82 mg/m ² -day
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TABLE 3. Summary of methods for controlling H₂S in landfill gas

Purpose	Media	Category	Method	Examples
Inhibition of H ₂ S generation	Solid waste	Waste composition	Sulfur source removal/modifying waste composition	Sorting sulfur containing waste materials such as drywalls
	Solid waste	Biological treatment	Increasing pH (> pH9), increasing redox potential greater than -100 mV	NaOH, Ca(OH) ₂ , adding alkaline wastes like concrete
		Chemical treatment	Inhibition of SRB activities with chemical inhibitors	Adding alternative electron acceptors
Removal of H ₂ S formed	Landfill gas captured	Thermal treatment	Incineration	Flare stack
		Physicochemical treatment	Dry H ₂ S removal	Solids adsorbents (iron oxides, zinc oxides, and alkaline solids) or adsorbents (zeolites and activated carbon)
			Liquid H ₂ S removal	Redox processes, alkaline solution processes, and amine solution processes
	Biological treatment	Biological oxidation H ₂ S	Bioscrubbers Biofilter Trickling biofilter Membrane bioreactors Activated sludge	
	Landfill gas fugitive emission	Alternative cover materials	Biofilters Metal oxide filters	Compost
	Masking agent	Deodorants or Neutralizer	Fly ash Spray chemicals around or over odor source	

5. H₂S EMISSION CONTROL TECHNOLOGIES

Suppressing H₂S generation and/or emission from a landfill can be achieved by inhibiting H₂S generation and removing H₂S from landfill gas. Table 3

summarizes different categories of H₂S reduction technologies. Options for H₂S control in landfills vary. Control techniques can be applied to remove the sulfur source from waste, to inhibit SRB growth in landfilled waste, and to remove H₂S from the landfill gas. H₂S removal in landfill gas methods also differs depending on the presence of a landfill gas collection system and the use of collected landfill gas.

5.1 Source Control

Controlling waste composition is one of the most effective methods for reducing H₂S generation from a landfill. The landfilling of biodegradable waste materials with high sulfate content has been prohibited in England and Wales since July 2005 (UK Health Protection Agency, 2011). Banning sulfur-containing waste such as drywall (usually 90% CaSO₄·2H₂O and 10% paper by mass composition) can inhibit SRB activities inside the landfill. Codisposal of C&D waste with high pH waste can reduce H₂S production. For example, waste such as concrete impacts H₂S generation by increasing pH greater than the optimum pH ranges (pH 6~9) for SRB and by absorbing H₂S that is present in landfill gas (Kenton et al., 2006).

H₂S formation in landfilled waste is influenced by codisposed materials. Yang et al. (2006) showed that H₂S generation is affected by the presence of codisposed wood and concrete. The organic acids leaching from codisposed wood lowered the pH of the leaching solution and decreased SRB activity. Concrete plays a role in the reduction of H₂S production by increasing leachate pH from optimal pH range of SRB and reacting with H₂S as the following chemical reaction (Equation 9). It was reported that mixing lime with gypsum drywall prior to disposing in landfills effectively reduced H₂S generation (Johnson, 1986; Xu et al., 2011).



5.2 Use of Inhibitors for SRB

As H₂S from landfills mostly is produced biologically, creating artificial environments to inhibit SRB is an effective way to minimize H₂S emission. There are various ways to create unfavorable environments including changing redox potential, increasing pH, stimulating growth of a competing group of anaerobic bacteria, and adding SRB inhibitors.

The redox potential of the environment should be -100 mV or less to cultivate SRB (Postgate, 1984). The redox potential can be adjusted by adding oxidizing agents such as oxygen and nitrate. The inhibition of sulfate reduction could occur by increasing dissolved oxygen in wastewater greater than 1 mg/L (Hao et al., 1996). The addition of oxygen (air) not only increases

redox potential but also causes the abiotic oxidation of H₂S. Duangmanee (2009) studied sulfide removal from biogas of anaerobic digesters using precisely regulated air injection, referred to as microaeration, to both the gas and liquid phases (effluent). As a result of the investigation, more than 98% of sulfide in biogas was removed and 95% of the H₂S removal was caused by abiotic oxidation.

The addition of a chemical (an electron acceptor for a group of anaerobic bacteria outcompeting SRB) can result in inhibiting H₂S generation. The chemicals include nitrate and ferric compounds. The addition of nitrate into an aqueous environment also inhibits SRB by increasing the redox potential higher than -100 mV (Hao et al., 1996). In addition, it also stimulates the growth of nitrate-reducing bacteria (NRB), which have a large thermodynamic advantage over the SRB for the same substrates (Eckford and Fedorak, 2002). The addition of ferric iron can stimulate the growth of iron-reducing bacteria (IRB) which can outcompete SRB by maintaining concentrations of substrates at levels lower than thresholds required by SRB (Lovley, 1991). Lovley and Phillips (1986) found that the addition of ferric iron to sediment inhibited sulfate reduction by 86–100% (Lovley and Phillips, 1986). Du et al. (2014) reported that with the addition of Fe(OH)₃, the emission of H₂S decreased about 95% from biodegradable sulfur-containing waste, indicating iron has the potential role to control H₂S emission in landfills (Du et al., 2014). Xu et al. (2011) demonstrated the addition of ferric chloride (lowering pH) or hydrated lime (increasing pH) out of the optimum range for SRB can effectively inhibit H₂S generation. SRB is usually inhibited at pH values lower than 5.5 or higher than 9 (Hao et al., 1996).

Saleh et al. (1964) reviewed and summarized chemical inhibitors of SRB activities. The chemical inhibitors include antibiotics, detergents, dyes, mercurials, metal ions and complexes, nitro compounds, phenolic substances, sulfate analogs, and sulfonamides. Analogs of sulfate ion such as molybdate, chromate, tungstate, and selenate inhibit sulfate reduction. Among these metal ions, molybdate has been extensively used as an SRB inhibitor in lake and marine sediment and anaerobic digesters. Patidar and Tare (2005) showed that molybdate supplementation inhibited SRB cavity in biomass over 85% at 3mM. They found that the higher molybdate dose needed as the COD/SO₄²⁻ ratio increased could stimulate or inhibit the total methanogenic activity, depending on other factors. The concentration of formaldehyde (a range of 12–19 mg/L) effectively reduced sulfide generation up to 90% of the total sulfide with negligible impacts on COD removal, nitrification rate and oxygen uptake rates under anaerobic conditions (Zhang et al., 2008). However, the authors pointed out that the use of formaldehyde for an SRB inhibitor should be performed under caution because of the toxicity of the chemical to humans. Xu et al. (2011) observed considerable inhibitive effects of sodium molybdate (Na₂MoO₄) on H₂S generation from flasks with 100 g gypsum drywall incubated for 30 days (under 35°C). The average H₂S

concentration with 2 mM sodium molybdate dose was about 8.6 ppm, but the H₂S concentration in flasks without sodium molybdate showed up to 67,500 ppm (Xu et al., 2011). Even though the chemical inhibitors successfully reduced H₂S generation from simulated landfills, for the application of chemical inhibitors in landfills some issues should be addressed, including homogeneous contribution of inhibitors on waste, cost-effectiveness, and environmental impacts.

5.3 Removal of H₂S Formed in Landfill Gas

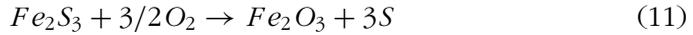
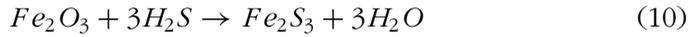
5.3.1 REMOVAL OF H₂S IN CAPTURED LANDFILL GAS

There are various methods for treating H₂S in landfill gas including thermal, physicochemical, and biological methods to treat H₂S in landfill gas. A typical modern sanitary landfill is required to be equipped with a landfill gas collection system and the final cover system. The most common option for collecting landfill gas is gas incineration on a candlestick flare system. This option results in converting H₂S to SO₂ or H₂SO₄.

If landfill gas is used for biogas to energy, it may be necessary to remove the impurities (including H₂S) of the gas below certain levels. Diverse H₂S removal processes for off-gases are developed and commercialized. Various processes to remove H₂S in biogas have been well reviewed by researchers (Jensen and Webb, 1995; Syed et al., 2006; Abatzoglou and Boivin, 2009). To select an appropriate H₂S removal technology for a landfill site, multiple variables such as concentration of H₂S gas in the bio gas; pressure, temperature and composition of the gas; ratio of H₂S to CO₂; volume of gas, total H₂S load, gas specifications for the process output, economic considerations, and environmental implications should be considered (Gómez and Cantero, 2007). In this section, common physicochemical and biological technologies to remove H₂S in biogases are summarized.

Physicochemical processes for removing sulfur compounds in gas (not only H₂S) include dry removal processes and liquid removal processes. In the dry processes, H₂S is removed by solid adsorbents. The dry absorbent processes are cost-effective with low sulfur loading but not economical for high sulfur loading because of spent waste production. Iron oxides are commonly used as H₂S adsorbents with an activator (0.125–5% weight basis; Abatzoglou and Boivin, 2009). Iron sponge (powdered iron oxide adhered to wood shavings) has long been used for the H₂S removal from the gas phase. To remove H₂S, gas is forced to a container or a series of containers filled with iron sponge. The iron oxide of iron sponge reacts with H₂S to form iron sulfide. The used iron sponge can be regenerated back to iron oxide by exposing the bed to oxygen. Xu et al. (2010a) used oxidized tire-derived steel, a by-product of automobile tire recycling, as alternative cover material for H₂S removal. The oxidized steel showed a much stronger H₂S

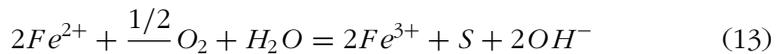
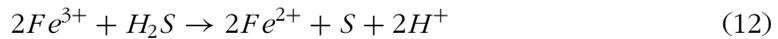
removal ability than sandy soils. With an initial concentration of 550 ppm, over 98% H₂S was removed in 2 min by 20 g of the steel, compared to only 50% H₂S removal in 60 min by the same amount of sandy soil (Xu et al., 2010a). Chemical reaction involved in iron oxide adsorbents are as shown in Equation 10 and Equation 11.



Commercially available iron oxide absorbents include SulfaTreat (MI SWACO), Sulfur-Rite (Merichem), Media-G2 (ADI International Inc.), and GAS RAP. Few cases of the use for treating landfill gas were reported (UK Environment Agency, 2010). Abtzipglou and Boivin (2009) compared the performances and costs of these iron sponges.

Impregnated carbon is also widely used in gaseous H₂S removal processes, which combines both physical adsorption and chemical reaction. Carbon source, such as coconut shell, serves as an adsorbent for H₂S, while the chemicals, such as metal oxide and alkaline materials, react with H₂S. Other absorbents, such as silica gel, alumina, zeolites, etc., can also be used to remove H₂S in landfill gas (Xiao et al., 2008; Elsayed et al., 2009).

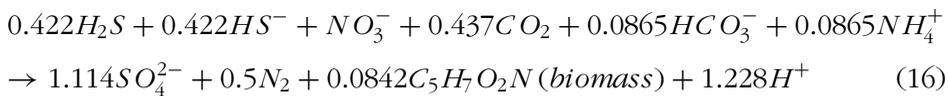
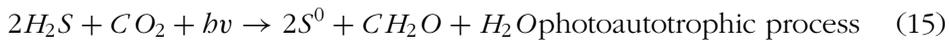
Liquid H₂S removal processes include liquid redox processes, alkaline solution processes, and amine solution processes. In liquid redox processes, iron-chelated solutions convert sulfide to element sulfur by the reactions expressed as shown in Equations 12 and 13.



LO-CAT (Shell/Dow) and Sulferox (US Filter/Merichem) processes are the commercial names of typical liquid redox processes. LO-CAT has been applied for treating H₂S in landfill gas in Florida, USA (UK Environment Agency, 2010). The economical range of the sulfur loading for LO-CAT and Sulferox is higher than 100–20,000 kg sulfur/day and more than 200 kg sulfur/day, respectively (Abatzoglou and Boivin, 2009). Alkaline solution processes are not economically appropriate for landfill gas because this process produces a large volume of waste stream, which is nonregenerable by the reaction of alkaline salts and CO₂ of the landfill gas. Amine solution processes are used in applications processing very large quantities of sulfur in gas (>27,000 kg/day), such as petroleum refinery processes (Baspinar et al., 2011). Other liquid-based techniques applied to reduce H₂S in landfill gas include water scrubbing (Sonzy landfill, France) and the use of Selexol (liquid solvent; UK Environment Agency, 2010).

Estrada et al. (2011) compared different odor-treatment technologies for wastewater treatment plants in terms of environmental performance, process economics, and social impact using the IChemE Sustainability Metrics. In their study, biological treatment technologies showed the highest investments but the lowest operating cost. However, the investment costs per unit flow rate treated decreased dramatically when the design flow rates increased. Meanwhile, physical/chemical technologies exhibit higher environmental impacts than biological technologies in terms of energy consumption, material and reagents consumption, operational risk, and hazardous waste production (Estrada et al., 2011).

Biological oxidation of sulfide into elemental sulfur can be conducted by chemoautotrophic or photoautotrophic bacteria, as shown in Equations 14 and 15. Also, oxidation of sulfide can occur in the absence of oxygen with nitrate (Equation 16; Tichý et al., 1998).



Biological H₂S removal processes with various microorganisms are well documented by Jensen and Webb (1995) and Syed et al. (2006). Syed et al. (2003) suggested that the use of phototrophic bacteria (e.g., *Cholorobium limicola*) for the removal of H₂S in biogas was beneficial for the anaerobic nature of the gas, even though light supply is a key constraint of phototrophic oxidation of H₂S. However, chemoautotrophic thiobacteria more often are used to treat H₂S in biogas. Among the chemoautotrophic oxidation bacteria, *Thiobacillus* sp. are commonly used in H₂S removal studies. *Thiobacillus* sp. have the ability to grow under various environmental stress conditions such as oxygen deficiency, acid conditions, and etc. The common biological treatment processes for gases purification include biofiltration, bioscrubbing, and biotrickling filtration (Zdeb and Pawlowska, 2009).

5.3.2 H₂S REMOVAL IN FUGITIVE LANDFILL GAS

5.3.2.1 Use of Alternative Covers. Landfill cover is an essential component for controlling gas emission from a landfill. Contrasted with conventional cover soil (earth material), various alternative cover materials have been proposed to abate odor emission. Suggested alternative cover materials by previous studies include compost, wood chips, fine waste materials (construction and demolition fines, commercial and industrial fines, and crushed concrete), fly ash, bottom ash (wood ash, coal ash, and municipal waste incinerator bottom ash), quicklime, hydrated lime, calcium carbonated materials, and waste steel (Plaza et al., 2007; Xu et al., 2010a; Xu et al., 2010b;

Solan et al., 2010). Each alternative cover material was tested independently or by mixing with other material. Physical adsorption, chemical reactions, and biological oxidation are the main mechanisms of H₂S reduction of alternative landfill cover materials. However, the capacity of H₂S removal with alternative cover materials can vary with experimental conditions and physicochemical properties of the materials.

Biofilter materials (e.g., wood chips, yard waste, compost) are often available in landfill sites that process these wastes. Hurst et al. (2005) examined the ability of municipal waste compost as a daily cover material for reducing odorous emission. In their study, the level of H₂S (70 ppm) in simulated odor gas considerably decreased by more than 95% with the first 10 cm of compost (with a bulk density 590 kg/m³). Bergersen and Haarstad (2008) compared various filter materials (spruce bark, wood chips, sludge compost, the mixture of bottom ash to iron oxide [3:1], and iron oxide) for H₂S removal in the gas generated from simulated columns with different water levels. They concluded that metallic filter materials had greater adsorption potentials than those of organic filter materials. The sulfide adsorption of different alternative cover materials for odor reduction is summarized in Table 4.

MSW incineration bottom ash has been shown to be useful as sorbing agent for remove H₂S from landfill gas (Mostbauer et al., 2008). Ducom et al. (2009) observed that 1 kg of MSW incinerator bottom ash could remove more than 3.0 g of hydrogen sulfide. H₂S retention capacity of bottom ash relied on moisture content and contact time (gas flow rates) in their experiments. They also explained the retention mechanisms of H₂S by bottom ash as acid-basic reactions (sulfide ion [S²⁻] in the basic medium reactions with metal cations in solution). He et al. (2011) tested adsorption removal of H₂S with waste biocover soil collected from a rural waste bioreactor and sieved to separate large pieces, with different particle sizes (≤ 0.45 mm to 5 mm), temperature (4–45°C), moisture (2–60%), pH (3–11), and oxygen content (0–21%). They observed that the absorption capacity of the biocover increased with decreasing particle size, increasing moisture content, and increasing pH. Also, they found that H₂S removal varied with different oxygen contents. Peak absorption capacities were observed at 35% moisture content and 10% oxygen content in the tested ranges. The influence of oxygen on H₂S removal with alternative cover materials was observed in other research (Xu et al., 2010a). By the adsorption isotherm, the authors concluded that H₂S concentration, the characteristic of water film on adsorbents (pH, temperature and ionic strength), and the characteristics of adsorbents (surface area, organic matter, mineral composition, and pore size) played important roles in H₂S adsorption characteristics.

The amendment of cover soil also is another option to reduce H₂S emission. Mixing on-site soil with alkaline material(s) such as quicklime, hydrated lime, and/or fine concrete materials, has been tested in the laboratory and

TABLE 4. H₂S removal using alternative cover materials

Materials	Sulfide adsorption (mg/kg) or H ₂ S removal	Experimental conditions	Reference
Spruce bark	215 mg S/kg	Column tests by adding a filter layer (5–10 cm)	(Bergersen and Haarstad, 2008)
Wood chips	387 mg S/kg		
Sludge compost	762 mg S/kg		
Bottom ash/iron oxide 3:1	3,345 mg S/kg		
Iron oxide	983 mg S/kg		
Waste biocover soil	3–56 mg/kg (particle size <0.45 to >25 mm) 8–168 mg/kg (pH 3–11) 34–60 mg/kg (O ₂ 0–10%) 383 ± 40 mg/kg (saturation)	Batch experiment Particle size, <0.45 to >25 mm Water content: 2–60%, pH 3–11, O ₂ 0–10%	(He et al., 2011)
Coal bottom ash pH 6.2–8.6, surface area (m ² /g) 7.2–15.5	10.5 mg H ₂ S/g bottom ash	Gas adsorption column. Gaseous H ₂ S (9000 ppm) mixed with pre-humidified air	(Lin et al., 2001)
MSW incineration bottom ash	3g H ₂ S/kg bottom ash	The experiment was designed for removing sulfur gas in landfill gas 90–150 mg Nm ⁻³ H ₂ S (landfill gas) 13 kg bottom ash (MC 15%), flow rate 879–1710 NLhr ⁻¹	(Ducom et al., 2009)
Red clay	36.04 H ₂ S mg/g	Batch test. A 20 g sample with 5,090 ppm H ₂ S in 275 mL bottle (for 60 min)	(Liu et al., 2011)
Compost	25.97 H ₂ S mg/g		
Waste wire	29.82 H ₂ S mg/g		
Sand	17.39 H ₂ S mg/g		
Sandy soil	50% removal	20 g sample in 550 ppm H ₂ S containing 275 mL bottle (tested for 60 min)	(Xu et al., 2010b)
Tire-derived steel	99% removal (7.5 mg H ₂ S/g tire-derived steel) [†]		
MSW Compost with a density of 590 kg/m ³ and 740 kg/m ³ (moisture content 35–50%)	More than 90% H ₂ S and 95% H ₂ S in standard gas (70 ppm) was removed with the first 10 cm of 590 kg/m ³ and 740 kg/m ³ , respectively. 1500–1600 mgH ₂ S/kg compost [†]	Column test. Odor reduction was measured in different depths (10–60cm) of compost layer using standard gas (a gas mixture of odor compounds) using a flow rate of 250 mL/min for 96 min	(Hurst et al.,

[†]Calculated adsorption capacity with given information but may not be at equilibrium conditions.

field (Plaza et al., 2007; Xu et al., 2010b). Xu et al. (2010b) found that fine concrete, compost, sandy soils mixed with 10% CaCO₃, 1% Ca(OH)₂, or 3% Ca(OH)₂ effectively reduced H₂S emission from a C&D landfill in their field and laboratory tests. These alkaline materials not only remove H₂S in landfill gas by the reactions to form sulfide minerals but also possibly reduce H₂S generation by creating alkaline environments under the amended cover materials.

Some organic filter waste from production of mineral wool also showed good H₂S removal capacity (Bergersen and Haarstad, 2008). Bergersen and Haarstad (2014) investigated the efficiency of H₂S removal from landfill gas using metal rich mineral wool wastes. Results show that the H₂S gas can effectively be removed by mineral wool waste and the estimated sulfide removal potential was 14 g/kg for mineral wool and 261 g/kg for rod mill waste, respectively (Bergersen and Haarstad, 2014).

5.3.2.2 Use of Masking Agents. Use of masking agents (neutralizing agents and pleasant smelling agents) in landfill areas or near residential areas is a common approach to abate odor from the landfill. For example, terpenes can be added to discontinuous or small volumes of odorous emissions in order to overcome their environmental nuisance (Smet et al., 1998). There are a number of masking agents that are used for this purpose and in most cases privately owned. These masking agents are used to lower odor but not to remove odor compounds.

A few researchers have tested bacterial deodorants using sulfur oxidizing bacteria (Hirano, Kurosawa et al., 1996; Kurosawa et al., 1997). Snyder (2009) introduced a liquid deodorant composed of *Bacillus amyloliquefaciens* strain NRRL B-50141 with a range of concentration between 1×10^5 /mL to 1×10^{10} /mL, propylene carbonate, sodium citrate, sodium bicarbonate, and sodium carbonate. Hirano et al. (1996) used *Thiobacillus thiooxidans* with sodium citrate buffer to remove H₂S and reported that the removal of H₂S (20 μ L/L) in the gas phase ranged from 80% to 100% in the pH range of 2 to 7. A deodorant composition in concentrate form for abating the odor of organic refuse comprises isopropanol (solvent, 5–15% w/w) a glycol (antifreeze and a stabilizing agent, 2–8%), water (30–60%), an emulsifying and surfactant agent (20–40%), a binding agent (0.1–1.0%), and an essential oil (eucalyptus oil, lemon grass oil, and juniper berry oil, 5–15%; Roychowdhury, 1989). The inventors suggested that using 0.28–0.57 m³ of a deodorant composition per acre of the landfill area would be effective for the abatement of landfill odor for a period of 3–5 days. Also, plant-derived oils were used to reduce odor from livestock manure (Varel, 2002). Carvacrol and thymol were found to inhibit odor generation by reducing anaerobic microbial activities. However, misting deodorant in landfills is a temporary solution for the odor reduction.

5. CONCLUSION

The control of H₂S from landfills is important for public health and environmental protection. The authors reviewed the current academic development on the formation of H₂S in landfills, the environmental risk of H₂S, the levels of H₂S in the landfill environment, and H₂S control. H₂S emission varies with waste composition, waste age, and environmental conditions for sulfur reducing bacteria, landfill geometry, and meteorological factors influencing gas emission.

Reducing sulfur sources in waste materials to be landfilled is the most effective option for lowering H₂S generation in landfills. When treating H₂S in collected landfill gas, physicochemical and biological technologies are commonly used. In addition to H₂S removal efficiency, cost is another most important consideration for choosing treatment technology. Physicochemical H₂S removal processes using absorption removal in iron-chelated solutions and iron-based adsorbents are mature technologies. Biological H₂S treatment technologies need high initial investments but their operating costs are lower than the physicochemical processes. The use of alternative cover is the most commonly used method to remove H₂S in fugitive landfill gas. Even though the range of H₂S adsorption capacity retention of the alternative cover materials varies with testing conditions, the alternative cover using recycled waste is a good option to the control surface emission of H₂S in landfills from both odor control and waste recycling perspectives.

It should be noted that although there are many methods available for H₂S control in landfills, the selection of technologies should be based on many considerations, such as waste composition, landfill location, climate condition, removal efficiency, and the initial and operational cost etc. In addition, other odor-causing compounds also need to be counted for odor control in landfills.

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