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# Evaluation of alternative landfill cover soils for attenuating hydrogen sulfide from construction and demolition (C&D) debris landfills

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#### Abstract

Hydrogen sulfide ( $H_2S$ ) generated from C&D debris landfills has emerged as a major environmental concern due to odor problems and possible health impacts to landfill employees and surrounding residents. Research was performed to evaluate the performance of various cover materials as control measures for  $H_2S$  emissions from C&D debris landfills. Twelve laboratory-scale simulated landfill columns containing gypsum drywall were operated under anaerobic conditions to promote  $H_2S$  production. Five different cover materials were placed on top of the waste inside duplicate columns: (1) sandy soil, (2) sandy soil amended with lime, (3) clayey soil, (4) fine concrete (particle size less than 2.5 cm), and (5) coarse concrete (particle size greater than 2.5 cm). No cover was placed on two of the columns, which were used as controls.  $H_2S$  concentrations measured from the middle of the waste layer ranged from 50,000 to 150,000 ppm. The different cover materials demonstrated varying  $H_2S$  removal efficiencies. The sandy soil amended with lime and the fine concrete were the most effective for the control of  $H_2S$  emissions. Both materials exhibited reduction efficiencies greater than 99%. The clayey and sandy soils exhibited lower reduction efficiencies, with average removal efficiencies of 65% and 30%, respectively. The coarse concrete was found to be the least efficient material as a result of its large particle size.

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Keywords: Construction and demolition debris landfills; Hydrogen sulfide; Cover soil emission rate; Landfill gas

#### 1. Introduction

Construction and demolition (C&D) debris represents one of the largest contributors of solid waste in North America (Florida Department of Environmental Protection, 1998). Major components include concrete, asphalt, wood, metal, gypsum drywall, and roofing material produced during the construction, renovation, or demolition of structures. A large fraction of C&D debris is disposed in landfills (US Environmental Protection Agency (EPA), 1998). Given the fact that C&D debris contains much less biodegradable waste in comparison to municipal solid waste (MSW), historic perception has been that C&D debris landfills generate little or no biogas (Flynn, 1998). Gases are, however, produced at these facilities, as evidenced by documented odor problems from reduced sulfur gases such as hydrogen sulfide (H<sub>2</sub>S) (Johnson, 1986;

Musick, 1992; Gypsum Association, 1992; Lee et al., 2006). Although a variety of reduced sulfur compounds (RSC) have been documented in gas from C&D debris landfills, hydrogen sulfide (H<sub>2</sub>S) has been identified as the primary RSC (Lee et al., 2006).

The production of H<sub>2</sub>S at C&D debris landfills results from the biological reduction of sulfate from gypsum drywall. As gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O) is exposed to water in the landfill environment, sulfate and calcium are released into solution. Under anaerobic conditions, sulfate-reducing bacteria (SRB) utilize sulfate as an electron acceptor and produce H<sub>2</sub>S. H<sub>2</sub>S exhibits a distinctive, offensive "rotten egg" odor and may pose a potential environmental and health threat (Agency for Toxic Substances and Disease Registry (ATSDR), 2003; Chou, 2003; Campagna et al., 2004). Most C&D debris landfills do not control gas emissions in the same fashion as MSW landfills (e.g., gas extraction and combustion systems), therefore gas may readily migrate to the surface of the landfill and into the atmosphere. Measurements of gases at C&D debris

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landfills in Florida documented  $H_2S$  concentrations as high as 12,000 ppm in the subsurface of the landfill (Lee et al., 2006). At these facilities,  $H_2S$  concentrations were found to vary dramatically from landfill to landfill, as well as among different locations within the same site.  $H_2S$  concentrations measured in the atmosphere at the surface of these landfills (above the cover soil) were much lower relative to those concentrations within the waste and soil, which may be a result of atmospheric dilution and the barrier provided by the cover material.

In a recent experiment that studied the role of predominant C&D debris components on  $H_2S$  production in landfills, portland cement concrete (PCC) was discovered to play a major role in  $H_2S$  production and fate (Yang, 2000). As an alkaline material, PCC at times increased the pH to the point where  $H_2S$  production was suppressed. In addition, PCC was found to remove  $H_2S$  from the gas phase. The proposed mechanism for the latter was sorption of the  $H_2S$  to the PCC surface and the ultimate formation of sulfide minerals. These results presented the possibility of utilizing PCC (as well as other materials) as a landfill cover layer that attenuates  $H_2S$ .

Landfill operators routinely place soil on top of deposited waste to suppress vector attraction, reduce fire potential, diminish odors, and shed rainfall (Tchobanoglous et al., 1993). It is hypothesized that alternative materials, or amended soils, may serve the dual purpose of creating a barrier to minimize H<sub>2</sub>S emissions and to remove H<sub>2</sub>S from the gas phase. A similar approach has been evaluated for mitigating gaseous emissions at MSW landfills (Whalen et al., 1990; Jones and Nedwell, 1993; Kightley et al., 1994; Boeckx et al., 1996; Barlaz et al., 2004), but most of these studies have focused on the methane oxidizing capacity of the cover soil.

The utilization of specific cover materials to help control H<sub>2</sub>S emissions at C&D debris landfills might prove to be a useful alternative to more expensive gas control techniques such as landfill gas collection systems. This study assessed the performance of various cover materials in the attenuation of H<sub>2</sub>S emissions from C&D debris landfills.

## 2. Materials and methods

## 2.1. Column design

Twelve laboratory-scale columns were constructed using PVC SDR-35 pipes with a diameter of 0.2 m and a length of 1.5 m. Fig. 1 presents a cross section representing a typical column. A PVC end cap was affixed to the bottom of each column and a valve was installed on each bottom end cap for leachate drainage. A drainage layer was placed inside each column in the following order starting at the bottom: a stainless-steel mesh, 5 cm of pea gravel, a second stainless-steel mesh, 15 cm of sand, and a non-woven geotextile. The columns were then filled with size-reduced gypsum drywall to a depth of 1 m. The drywall was purchased from a home supply outlet and was size reduced

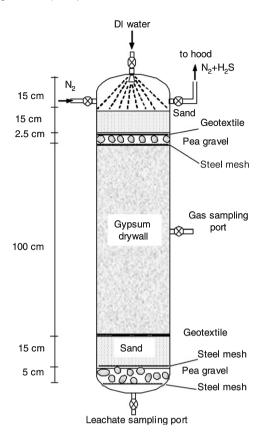


Fig. 1. Cross section of experimental column.

to square pieces  $(5 \times 5 \, \text{cm})$  utilizing a power saw. In the process of adding drywall to the columns, a stainless-steel tube was placed as a sampling port in the middle of the waste layer; this tube was connected to a gas sampling valve on the exterior of the column. The final mass of gypsum drywall in each column was approximately  $10.5 \, \text{kg}$  with an average bulk density of  $314 \, \text{kg/m}^3$ . Once the columns had been loaded, a steel mesh was placed on top of the drywall and  $2.5 \, \text{cm}$  of pea gravel was added to provide additional support and separation between the waste and the cover material. Finally, a non-woven geotextile was placed on top of the pea gravel.

Five different cover materials were examined in this study: a sandy soil, a sandy soil amended with lime, a clayey soil, crushed PCC with a particle size of 2.5 cm or less, and crushed PCC with a particle size of 2.5 cm or greater. Each cover material was placed inside two columns designed as duplicates. Two control columns did not contain cover soil. The sandy and clayey soils were collected from Gainesville, FL (US) in an area identified by local soil scientists. Laboratory analysis was performed to determine the physical and chemical characteristics for the sandy and clayey soils. The sandy and clayey soils had pH values of 6.2 and 5.0, and organic matter contents of 1.33% and 1.16%, respectively. Two different sizes of crushed concrete (fine and coarse concrete) were obtained at a concrete recycling facility in Gainesville, FL and sieve analysis was performed to assess the size distribution for

each sample (Plaza, 2003). The sandy soil was amended with hydrated lime (Ca(OH)<sub>2</sub>) at a ratio of 1 g of lime for every 20 g of sand to obtain a soil pH greater than 12. A total of 15 cm of cover material was loaded into each column.

A PVC end cap was affixed to the top of each column providing 0.15 m of headspace above the cover materials. To maintain a constant headspace in each column, the length of the control columns was reduced accordingly. A valve was installed at the top of the end cap to allow the introduction of water during the course of the experiment. Each valve was connected on the inside of the cap to a sprinkler nozzle that promoted even water distribution. As the objective of the experiment was to measure the amount of H<sub>2</sub>S emitted from the top of each column (through the cover soil, or the surface of the waste in the case of the control columns), Whitey gas valves (model SS-1RS4, Aaron Iron and Steel) and stainless-steel Nupro valves (model SS-4H) were installed at the sides of the caps to serve as a gas inlet and outlet, respectively.

A system was constructed to allow the continuous addition and removal of nitrogen gas to and from the headspace in the columns. The intent of this system was to simulate the removal of gases above the landfill that would normally occur by air flow in the field; nitrogen was used to minimize possible impacts of oxygen on biological activity within the columns. Two nitrogen (N<sub>2</sub>) tanks were connected in series to a mass flow controller to maintain a continuous flow. The tubing coming out of the mass flow controller was split into two lines, which were connected to flow regulators that delivered gas to a group of six columns each. The output valves were connected to stainless-steel tubes directed toward a fume hood. The inlet tubing was constructed of copper and the exhaust tubing was constructed of stainless steel. All lines were cut to the same length to ensure that the losses due to friction would be the same in each line. The nitrogen gas flow was regulated so that each column had the same flow coming out. Flow measurements were performed with the use of a hand-held flow meter (ADM 2000, Agilent) connected to the end of the outlet tubing.

## 2.2. Column operation and monitoring

H<sub>2</sub>S measurements were collected from the columns over a 494-day period. At times during the experiment, a flow of N<sub>2</sub> was maintained through the headspace, while at other times, the system remained closed. Gas was sampled from two primary locations: the headspace and the sampling port in the middle of the drywall. A 100-mL glass syringe was used to extract samples from the middle of the columns. This technique was also used to extract gas samples from the headspace when the columns were closed. Gas samples collected in the syringe were added to one-liter Tedlar<sup>®</sup> bags for analysis. During periods when N<sub>2</sub> was swept through the headspace, gas samples were collected by directing the exit tubing into a Tedlar<sup>®</sup> bag.

Gas samples in the Tedlar® bags were analyzed for H<sub>2</sub>S using a Jerome Meter (model 631-X) from Arizona Instruments. The Jerome Meter has a detection range of 0.003–50 ppm and can only be used for gas phase samples. The instrument requires regeneration before sampling and whenever saturation occurred. During regeneration, the gold film inside the instrument is heated and the hydrogen sulfide is burned off. After regeneration, the instrument is zeroed. Prior to starting analysis, the manufacturer recommends waiting approximately 45 min after regeneration is complete. The instrument's calibration was checked prior to analyzing H<sub>2</sub>S concentrations using a 25 ppm H<sub>2</sub>S gas standard from Air Liquide America Corp. The manufacturer of the instrument does not provide a means for user calibration and recommends sending the unit to them for recalibration on an annual basis.

Tedlar<sup>®</sup> bags containing gas samples were attached to the instrument via a plastic connector. Samples were drawn into the instrument using the devices pump until readings stabilized. In many cases, H<sub>2</sub>S concentrations were greater than 50 ppm, necessitating gas dilution. Most samples taken from the middle of the waste layer needed dilutions of 1/1000 and 1/2000. All dilutions were performed following the same procedure. A 1/1000 dilution was made by filling a clean Tedlar<sup>®</sup> bag with 500 ml of laboratory air using a 500-ml glass syringe (Precision Sampling Corporation). Gas samples of 20 ml were removed Tedlar<sup>®</sup> bag containing the sampled gas using a 100-ml glass syringe; the syringe was pumped several times to provide mixing and to obtain a homogeneous sample. A 3-ml plastic syringe with a gastight valve was then used to extract 0.5 ml sample from the 100-ml syringe. Once again, the 3-ml syringe was pumped several times to extract a homogeneous sample. The 0.5 ml sample was introduced into the Tedlar<sup>®</sup> bag containing the 500 ml of laboratory air. The diluted mixture was left to diffuse for at least 10 min before H<sub>2</sub>S concentrations were measured. A similar same procedure was used to create the 1/200 dilutions, but the clean Tedlar<sup>®</sup> bag was filled with 1000 ml of laboratory air. For most samples this dilution process was performed in duplicate and the results were averaged. The duplicate readings were consistently within 5% of one another.

Throughout the experiment, deionized (DI) water was added regularly to the columns to promote bacterial activity. Water was added via a submersible pump that was attached to the valve at the top of the columns. Concurrently, leachate was drained from the columns. Although examination of leachate quality was not an objective of the experiment, leachate samples were analyzed on occasion for several water quality parameters useful for assessing the conditions within the columns (e.g., pH). A complete description of the leachate analysis performed can be found in Plaza (2003).

Table 1 describes the different operational conditions maintained throughout the experiment. When the experiment began, a constant gas flow of 75 ml/min was maintained. After 87 days into the experiment, the N<sub>2</sub>

flow was stopped to allow confirmation that lower  $H_2S$  concentrations observed in some columns were the result of differences in flow restrictions from the cover soil and not from different rates of biological activity in the columns.  $N_2$  flow was resumed on day 174. In the final stage of the experiments,  $N_2$  gas was not added continuously during the experiment, but was only introduced at the top of the columns prior to sample collection;  $N_2$  was introduced for a period of at least 60 min in order to remove accumulated  $H_2S$ .

## 3. Results and discussion

## 3.1. H<sub>2</sub>S production

H<sub>2</sub>S concentrations were measured from the sampling port located in the middle of the drywall layer for each column type for the entire length of the experiment. Two

Table 1 Column operational phases throughout the experiment

Stage	Period (day)	Flow rate (ml/min)	Flow method
Initial continuous flow stage	1–87	$75\pm3$	Continuous
Static system stage	88-174	0	Closed
Final continuous	179–218	$72\pm3$	Continuous
flow stage Modified flow stage	295–494	75±3	At least 60 min

control columns and two columns for each cover material type were operated. Most columns followed a similar trend with regard to the increase of H<sub>2</sub>S concentration over time. In addition, most of the duplicate columns exhibited similar H<sub>2</sub>S concentrations throughout the experiment. One of the lime columns, however, showed extremely low H<sub>2</sub>S concentrations in the middle of the drywall layer as compared to its duplicate. Upon examination of the pH of the leachate collected periodically from the bottom of the columns, the lime column with the low H<sub>2</sub>S concentrations was found to have a consistently high leachate pH (approximately 12) throughout the experiment. The leachate pH in the other lime column, as well as the two columns containing the fine concrete, started high but ultimately decreased to neutral conditions. Thus, H<sub>2</sub>S production is believed to have been suppressed by the alkaline conditions created by the leaching solution in one of the lime columns. The optimum pH range for SRB activity is approximately 7 and inhibition usually occurs at pH values lower than 6 and greater than 9 (Connell and Patrick, 1968; Widdel and Pfennig, 1984). The difference between the two lime columns is hypothesized to have resulted from differences in water distribution and contact with the lime in the amended soil.

Fig. 2 presents measured  $H_2S$  concentrations from the middle of the waste layer over the course of the experiment. Given that the measurements were similar in magnitude between duplicate columns on any given day, the concentrations for each cover soil scenario are presented as the average of the duplicates. The exception to this is the sandy soil amended with lime scenario; in this case only

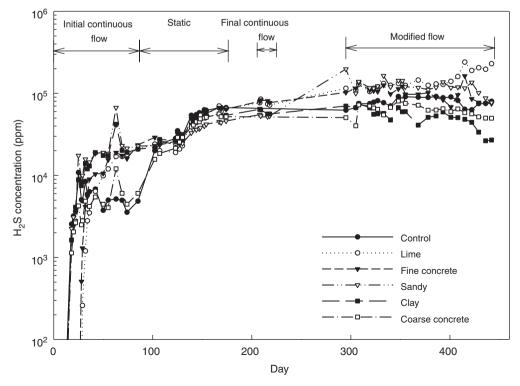


Fig. 2. H<sub>2</sub>S concentrations measured in the middle of the waste as a function of time for each column type.

results from the column where H<sub>2</sub>S production was not inhibited are presented. As expected from previous simulations (Jang, 2000; Yang, 2000), relatively large H<sub>2</sub>S concentrations were observed within the waste layer. At the end of the experiment, H<sub>2</sub>S concentrations in each column were in the range of 50,000 ppm to just over 100,000 ppm (5–10%). Biological sulfate reduction requires an organic carbon source, and as demonstrated in previous research (Yang, 2000), the paper facing and backing from the gypsum drywall provided enough organic carbon to produce large H<sub>2</sub>S concentrations. These concentrations are certainly of environmental significance, as H<sub>2</sub>S concentrations in the range of 500–1000 ppm can be lethal to humans (Flynn, 1998).

H<sub>2</sub>S concentrations within the waste layer increased until relatively consistent concentrations were observed at the end of the experiment. Several interesting observations were noted in the early stages of the experiment. To highlight these differences, Fig. 3 provides a plot of the first 180 days of the experiment on a log scale. During the first days of the experiment, the columns containing limeamended soil and fine concrete lagged in H<sub>2</sub>S production. As mentioned previously, this delay in H<sub>2</sub>S production could be attributed to the initial leaching of alkalinity as water percolated through the cover layers. The higher pH could have created an unfavorable environment for SRB activity. With the exception of one of the lime columns, concentrations within these columns did increase later in the experiment to a similar magnitude as some of the other columns. The pH of the collected leachate decreased to neutral conditions during the same period.

A corresponding lag was not observed in the coarse concrete columns. However, in comparison to the remaining columns, both the coarse concrete columns and the control columns were found to contain lower H2S concentrations in gas samples collected from the middle of the drywall layer. For instance, on day 74 the average H<sub>2</sub>S concentration in these two columns was approximately 4000 ppm, while the average concentration in the other columns was over 18,000 ppm. This occurrence was hypothesized to result from physical differences in gas flow. The control columns contained no cover to act as a barrier for the transport of gases produced in the drywall. Thus, H<sub>2</sub>S did not have the opportunity to accumulate as was the case for the soils and the fine concrete, all of which were fine-grained enough to provide some resistance to flow. The particle size of the coarse concrete was too large to provide any restriction to flow. In order to examine this hypothesis, the continuous flow of N2 through the headspace of the columns was ceased. Once the N2 flow was ceased, H<sub>2</sub>S concentrations within the waste layer of the control and coarse concrete columns quickly increased to levels similar to the other columns (see Fig. 3). In fact, H<sub>2</sub>S concentrations in all of the columns increased given that gas was no longer removed from the system.

During the final stage of the experiment, the  $\rm H_2S$  concentrations in the middle of the waste layer fluctuated within relatively stable ranges. Table 2 presents the average  $\rm H_2S$  concentrations during the modified flow stage of the experiment. The variation in concentrations among the different columns can be attributed to the effect of the cover materials. Due to the large particle size of the coarse

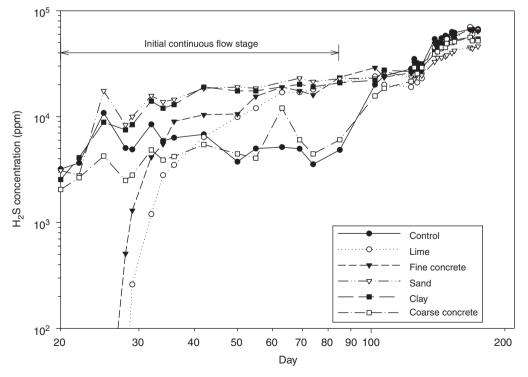


Fig. 3. H<sub>2</sub>S concentrations measured in the middle of the waste as a function of time for each column type (days 20-174).

Table 2
Average H<sub>2</sub>S concentrations of duplicated columns in the middle of the waste layer during the modified flow stage

Cover material	Number of samples	H <sub>2</sub> S concentration range (ppm)		Average H <sub>2</sub> S concentration (ppm)
		Minimum	Maximum	
Lime	22	105,000	240,000	147,000
Fine concrete	22	60,000	242,000	102,000
Clay	22	18,300	90,000	53,100
Sand	22	75,000	195,000	124,000
Coarse concrete	22	29,300	88,000	63,600
Control	22	54,000	99,300	79,100

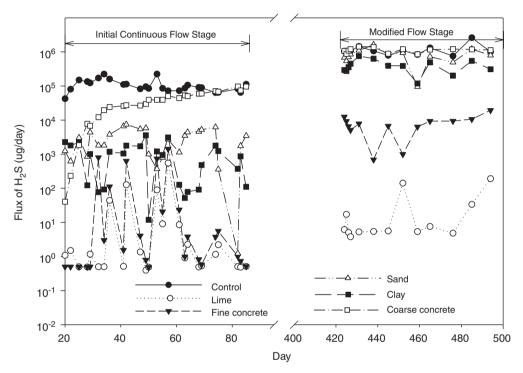


Fig. 4. H<sub>2</sub>S emission rates measured during the initial continuous and modified flow stages.

concrete, H<sub>2</sub>S was able to escape easily from the columns, therefore H<sub>2</sub>S concentrations inside the control and coarse concrete columns were similar and relatively low. The average concentrations within these columns were 79,100 and 62,600 ppm, respectively. Other cover materials with smaller particle size, such as the sand, fine concrete, and the lime-amended soil can act as a physical barrier to reduce gas emissions through the cover layer, which leads to H<sub>2</sub>S accumulation. Some columns exhibited maximum concentrations in excess of 100,000 ppm. Although these large concentrations may not be expected at the surface of C&D debris landfills as a result of the large degree of dilution expected to occur, the data do suggest that caution should be taken workers might be exposed to gas during gas well installation or similar activities that require excavation into the waste. During the modified flow stage, the H<sub>2</sub>S concentration in the clay columns started to slightly decrease. The cause of this observation was not determined.

## 3.2. $H_2S$ emission rates

H<sub>2</sub>S emission rates were calculated by multiplying concentration of H<sub>2</sub>S at the top of the columns by the measured N<sub>2</sub> flow. Fig. 4 presents the H<sub>2</sub>S emission rates (µg/day) for all of the column types during the initial continuous flow stage and the modified flow stage. At times during the initial flow stage, the emission rates for the lime, fine concrete, sand, and clay columns exhibited a considerable degree of variation. This variation was attributed to heterogeneity and preferential pathways in the cover layers, which can be greatly impacted by moisture content. The moisture content of a landfill's cover soil has been cited as the most important internal factor controlling gaseous emissions from landfills (Bogner, 1992; Bogner and Scott, 1995). Lee et al. (2006) observed H<sub>2</sub>S concentrations at the surface of landfills and in the cover soil to be extremely variable. This variation could be partly attributed to the heterogeneous nature of cover soils and the impact of variable soil moisture contents. Gas emission rates measured during the initial phases of the experiment were relatively consistent for the control and coarse concrete columns, as the gas transport in these columns was not impacted by cover heterogeneities or moisture content.

When compared with the initial continuous flow stage of the experiment, the H<sub>2</sub>S emission rates during the final modified flow stage were found to be more stable. During this stage, the difference in emission rates among the various columns was more evident. Water addition into the columns during the modified flow stage was performed less often, and this may have had the impact of creating more uniform soil conditions between sampling events. Fig. 5 summarizes the average emission rates for each column during the initial continuous and modified flow stages of the experiment. The results are presented on a log-scale to account for the substantial difference in measured emission rates. It was observed that the emission rates in each column were greater during the final stage of the experiment, which was a result of the larger concentrations of H<sub>2</sub>S present in the columns at the end of the experiment. This difference could also be due in part to changes in cover characteristics over time and the different methods of adding nitrogen to the columns (intermittent in the final stage versus continuous in the initial stage). During both stages of the experiment, the average H<sub>2</sub>S emission rates were greatest in the control columns, followed by the coarse concrete, the sandy soil, the clayey soil, the fine crushed concrete and the lime-amended sandy soil. The average H<sub>2</sub>S emission rates in each stage were statistically similar between the control and coarse concrete and between the sandy and clayey soils.

Clearly, some cover materials demonstrated a better ability to reduce the emissions of  $H_2S$  than others. To help illustrate this reduction, the  $H_2S$  removal efficiency was calculated utilizing the average  $H_2S$  emission results determined for the modified flow stage. The removal efficiencies of the various cover materials were calculated using the flux rate of the control columns as the standard for comparison:

H<sub>2</sub>S Removal Efficiency

$$= \frac{\text{(Flux of Control Column - Flux of Cover Soil Column)}}{\text{Flux of Control Column}} \times 100\%.$$

The resulting removal efficiencies are presented in Table 3. The lime-amended soil had the greatest reduction efficiency followed by the fine concrete. Both materials showed removal efficiencies greater than 99%. The sandy and clayey soils had reduction efficiencies of 30% and 65%, respectively.

Two primary mechanisms are proposed for the reduction in H<sub>2</sub>S emissions observed in this study. First, the cover materials act as a physical barrier limiting the migration of H<sub>2</sub>S. Given that the volume of gas produced by biological activity within the columns was relatively smaller than the rate of nitrogen flowing across the surface of the column, the primary driving force for gas migration was diffusion. Thus, an increase in the depth of cover material could decrease the diffusive gradient. It is believed that this mechanism was primarily responsible for the H<sub>2</sub>S reduction observed in the sandy and clayey soils. As presented in Fig. 5, the emission rate from the clayey and sandy soils during the initial continuous stage were over an order of magnitude lower than the control column. The reduction became less pronounced during the final stage, possibly

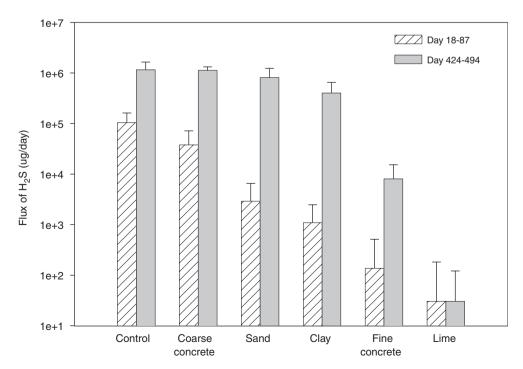


Fig. 5. Average H<sub>2</sub>S emission rates measured during the *initial continuous* and *modified flow stages* (error bar represents one standard deviation).

Table 3 Average  $H_2S$  removal efficiency during the modified flow stage by the different cover materials

Cover material	Number of samples	Flux range of H <sub>2</sub> S (μg/day)		Average H <sub>2</sub> S removal efficiency <sup>a</sup> (%)
		Minimum	Maximum	
Lime	13	1.5	$3.5 \times 10^{2}$	99.9
Fine concrete	13	$2.7 \times 10^{2}$	$2.9 \times 10^{4}$	99.3
Clay soil	13	$9.1 \times 10^{4}$	$1.0 \times 10^{6}$	65.3
Sandy soil	13	$9.9 \times 10^{4}$	$1.7 \times 10^{6}$	29.7
Coarse concrete	13	$7.3 \times 10^{5}$	$1.5 \times 10^{6}$	2.4
Control	13	$6.1 \times 10^{5}$	$3.1 \times 10^{6}$	0

 $^{a}H_{2}S$  Removal Efficiency =  $\frac{(Flux \text{ of Control column} - Flux \text{ of cover soil column})}{Flux \text{ of Control column}} \times 100\%.$ 

because  $N_2$  was only added intermittently, which allowed for gas accumulation. Subsequently, the accumulation of gas inside the columns could have also contributed to advective flux.

Second, as observed previously in an experiment where H<sub>2</sub>S concentrations decreased in the presence of concrete (Yang, 2000), H<sub>2</sub>S can react with alkaline materials and be removed from the gas phase. As a gas molecule diffuses through a porous media, gas molecules adsorb to surfaces of solid materials through processes such as physisorption and chemisorption (Szekely et al., 1976). Because of their alkaline nature, concrete and hydrated lime (Ca(OH)<sub>2</sub>) can react with H<sub>2</sub>S and ultimately be converted to sulfide minerals through reactions such as the following (Borgwardt and Park, 1984):

$$Ca(OH)_2 + H_2S \rightarrow CaS + 2H_2O$$

$$H_2S + CaO \rightarrow CaS + H_2O$$

It was observed that the color of both the fine and coarse concretes changed from gray at the beginning of the experiment to black at the end of the study. The black color in this case is an indicator of metal sulfide compounds. Although the clayey and sandy soils were both considered to be relatively inert materials from a sorption standpoint, changes in color observed in portions of these cover layers are also an indication that some H<sub>2</sub>S removal did occur. Soils naturally contain trace metals that could result in the formation of metal sulfide minerals.

The color change of the coarse concrete indicated that some  $H_2S$  sorption and conversion did occur. The fact that  $H_2S$  emissions were not controlled by this cover material is an indication that any removal that did occur was not enough to make up for its inefficiency as a physical barrier (due to its large particle size). The particle size of cover materials is important for several reasons. The amount of sorption will increase with an increase in available surface area. In addition, small particle size results in lower soil permeability. When gases are transported through the soil via advection, a soil with a lower permeability will result in increased contact time between the gas and the soil. The results of this study indicate that definite reduction in  $H_2S$ 

emissions can be achieved by using cover materials with the potential to remove H<sub>2</sub>S by sorption and subsequent conversion to sulfide minerals. In addition, some degree of emissions reduction should occur even with cover materials having little or no ability to remove H<sub>2</sub>S simply by acting as a physical barrier to gas flow.

## 4. Conclusions

As one of the prime components of C&D debris, gypsum drywall can be used by sulfate reducing bacteria to produce high concentrations of H<sub>2</sub>S in landfill environments. The odor issues and health problems associated with H<sub>2</sub>S emissions are a major concern for owners and operators of C&D debris, as well as other landfills that contain a large percentage of gypsum drywall in their waste stream. This research evaluated the ability of five different landfill cover materials to reduce H<sub>2</sub>S emissions. This study demonstrated that the presence of a cover material can reduce H<sub>2</sub>S emissions and that the performance of each cover material was dependent on its physical and chemical characteristics.

One mechanism by which cover materials can control H<sub>2</sub>S emissions (or emissions of other gas pollutants) is through physical containment. A cover layer can act as a barrier to decrease the pressure and concentration gradients between the waste and the top of the cover soil, thus decreasing gas advection and diffusion through the cover. H<sub>2</sub>S concentrations and pressures in the underlying waste would then increase, which in turn would promote greater H<sub>2</sub>S migration out of the landfill. An effective cover for attenuating H<sub>2</sub>S emissions is one where the cover material both acts to physically limit the advection or diffusion of a gas component and removes the gas component from the gas phase. Another possible mechanism that may impact H<sub>2</sub>S emissions is the creation of higher pH conditions underneath the cover material so that SRB activity becomes limited.

Lime-amended soil and fine concrete demonstrated greater capabilities in reducing  $H_2S$  emissions as compared to clayey and sandy soils. This was hypothesized to result from the sorption of  $H_2S$  to the cover materials and

subsequent conversion to sulfide minerals as a result of their alkaline characteristics. Even non-alkaline materials such as sand and clay reduced H<sub>2</sub>S emissions to some extent as a result of the physical barrier they created and some smaller but measurable degree of removal. In one column, the lime added to the soil suppressed the formation of H<sub>2</sub>S due to the alkaline conditions it created inside the waste environment. The potential utility of using of different cover materials for attenuating H<sub>2</sub>S emissions was demonstrated in the laboratory. The next step should be the evaluation of these materials under field conditions, where a greater number of variables might impact results.

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## References

- Agency for Toxic Substances and Disease Registry (ATSDR), 2003.

  Medical Management Guidelines (MMGs) for Hydrogen Sulfide (H<sub>2</sub>S). Atlanta, GA.
- Barlaz, M.A., Green, R.B., Chanton, J.P., Goldsmith, C.D., Hater, G.R., 2004. Evaluation of a biologically active cover for mitigation of landfill gas emissions. Environmental Science and Technology 38 (18), 4865–4877.
- Boeckx, P., Cleemput, O.V., Villaralvo, I., 1996. Methane emission from a landfill and the methane oxidising capacity of its covering soil. Soil Biology and Biochemistry 28, 1397–1405.
- Bogner, J.E., 1992. Anaerobic burial of refuse in landfills: increased atmospheric methane and implications for increased carbon storage. Ecological Bulletins 42, 98–108.
- Bogner, J.E., Scott, P., 1995. Landfill methane emissions guidance for field measurements. Final Report. International Energy Agency. Expert Working Group on Landfill Gas.
- Borgwardt, R.H., Park, T., 1984. Surface area of calcium oxide and kinetics of calcium sulfide formation. Environmental Progress 3 (2), 129–135.
- Campagna, D., Kathman, S.J., Pierson, R., Inserra, S.G., Phifer, B.L., Middleton, D.C., Zarus, G.M., White, M.C., 2004. Ambient hydrogen sulfide, total reduced sulfur, and hospital visits for respiratory disease

- in northeast Nebraska 1998–2000. Journal of Exposure Analysis and Environmental Epidemiology 14, 180–187.
- Chou, C.H.S.J., 2003. Hydrogen Sulfide: Human Health Aspects. World Health Organization, Geneva.
- Connell, W.E., Patrick, W.H., 1968. Sulfate reduction in soil: effects of redox potential and pH. Science 159, 86–87.
- Environmental Protection Agency (EPA), 1998. Characterization of Building-related Construction and Demolition Debris in the United States. Report no. EPA530-R-98-010.
- Florida Department of Environmental Protection, 1998. Solid Waste Management in Florida. Bureau of Solid and Hazardous Waste Division of Waste Management, Tallahassee, FL.
- Flynn, B.E., 1998. Invisible threat: odor & landfill gas from C&D waste. Waste Age, 91–97.
- Gypsum Association, 1992. Treatment and disposal of gypsum board waste: industry position paper. Construction Dimensions, 29–30.
- Jang, Y.C., 2000. A Study of Construction and Demolition Waste Leachate from Laboratory Landfill-Simulations. Ph.D. Dissertation. University of Florida. Gainesville, FL.
- Johnson, B., 1986. Gypsum wallboard creates landfill odor problem. World Waste, 53–54.
- Jones, H.A., Nedwell, D.B., 1993. Methane emission and methane oxidation in landfill cover soils. FEMS Microbiology Letters 102 (3-4), 185-195.
- Kightley, D., Nedwell, D.B., Cooper, M., 1994. Capacity for methane oxidation in landfill cover soils measured in laboratory scale soil microcosms. Applied and Environmental Microbiology 61, 592–601.
- Lee, S., Xu, Q., Booth, M., Townsend, T., Chadik, P., Bitton, G., 2006.
  Reduced Sulfur Compounds in Gas from Construction and Demolition Debris Landfills. Waste Management 26, 526–533.
- Musick, M., 1992. Recycling gypsum from C&D debris. Biocycle, 34–36.
   Plaza, C., 2003. Evaluation of Cover Materials for the Removal of Hydrogen Sulfide from Construction and Demolition Landfill Gas. Master's Thesis. University of Florida, Gainesville, FL.
- Szekely, J., Evans, J.W., Sohn, H.Y., 1976. Gas–Solid Reactions. Academic Press, NY.
- Tchobanoglous, G., Theisen, H., Vigil, S., 1993. Integrated Solid Waste Management Engineering Principles And Management Issues. McGraw-Hill, New York.
- Whalen, S.C., Reeburgh, W.S., Sandbeck, K.A., 1990. Rapid methane oxidation in a landfill cover soil. Applied and Environmental Microbiology 56, 3405–3411.
- Widdel, F., Pfennig, N., 1984. Dissimilatory Sulfate- or Sulfur Reducing Bacteria. In: Krieg, N.R., Holt, J.G. (Eds.), Bergey's Manual of Systematic Bacteriology. Williams & Wilkins, Baltimore, pp. 663–679.
- Yang, K.J., 2000. Hydrogen Sulfide Generation in Simulated Landfill Columns. Master's Thesis. University of Florida, Gainesville, FL.