Hydrogen Sulfide Emission and Dispersion at Construction and Demolition Debris Landfills

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Abstract: The emission of hydrogen sulfide from construction and demolition (C&D) debris landfills has resulted in offsite odor complaints at numerous sites in North America. Hydrogen sulfide results from the biological transformation of sulfate from discarded gypsum drywall, a major component of the C&D debris stream. Unlike MSW landfills, C&D debris landfills are not typically constructed with gas collection systems. Hydrogen sulfide concentrations within C&D debris landfills can be very high, but the gas is diluted as it leaves the landfill and mixes with air. The impact of uncontrolled hydrogen sulfide emissions varies as a result of site-specific conditions such as climate, terrain, landfill cover soil and disposal practices. Ambient hydrogen sulfide concentrations that might be typically expected at C&D debris landfills were predicted using an air dispersion model recommended by the EPA, the Gaussian-Plume Multiple Source Air Quality Algorithm (RAM). A range of emission data and weather conditions were used as model inputs. The spatial and time variations of hydrogen sulfide concentrations in an area surrounding a hypothetical landfill cell were examined. The results are presented in a graphical form that provides a means of quickly assessing concentration ranges expected under a variety of conditions. Phenomena related to hydrogen sulfide dispersion at C&D debris landfills are also discussed.

Keywords: hydrogen sulfide; C&D debris landfill; air dispersion model; RAM

INTRODUCTION

Construction and demolition (C&D) debris is one of the largest solid waste streams in the United States (US) and the most common management practice for C&D debris is landfilling (EPA, 1998). One environmental problem associated with C&D debris landfills is odor production (Crosson, 1995; Flynn, 1998; Johnson, 1986). Although many compounds can contribute to malodorous conditions, hydrogen sulfide (H_2S) has been identified as the major contributor to odor problems at C&D debris landfills receiving a large amount of gypsum drywall (Townsend et al., 2000 & 2004). The production of H_2S within C&D debris landfills results from the biological transformation of sulfate (SO_4^{2-}) leaching from gypsum drywall ($CaSO_4$. $2H_2O$), a major component of C&D debris. When gypsum drywall (approximately 90% $CaSO_4 \cdot 2H_2O$ and 10% paper) becomes wet under anaerobic conditions (as expected to occur in most landfills), sulfate-reducing bacteria flourish and use sulfate as a terminal electron acceptor, producing H_2S (Gypsum Association, 1992). Because of its distinctive "rotten egg" smell and low detectable odor threshold (reported as low as 0.5 ppb; Godish, 1991), H_2S emitted from C&D debris landfills has been a major issue at many landfills in Florida and other states in the US with respect to odor, and has raised questions regarding possible impacts on human health (Flynn, 1998).

The issue of H_2S emissions from C&D debris landfills in Florida first surfaced as a possible statewide concern when the Sunset Sand Mine and Landfill, a C&D debris landfill in Central

Florida, was closed in 1995 largely as a result of high concentrations of H_2S in the surrounding neighborhoods (Crosson, 1995). Nearby residents were evacuated twice. One report claimed that H_2S concentrations as high as 3 ppm were measured one quarter of a mile away from the site. Since this time, other sites have also reported odor complaints, and the state has funded several research projects to investigate the issue further. In one study (Townsend et al., 2000), H_2S concentrations in the air directly above a C&D debris landfill were found to be extremely variable, and to range from 3 ppb to above 50 ppm. The study concluded that H_2S concentrations within the waste at C&D debris landfills varied over many orders of magnitude, and that the concentrations experienced off-site would be heavily influenced by the effect of meteorological conditions.

While several investigators have attempted to measure, estimate or model landfill gas emissions from municipal solid waste (MSW) landfills (Perera et al., 2002; Schmidt et al., 1998; Yedla and Parikh, 2002), little research has been conducted to address H_2S emission and dispersion at C&D debris landfills. The objective of the research presented in this paper was to examine the range of H_2S concentrations that might be expected to occur in the ambient air surrounding a C&D debris landfill, and to assess the conditions (particularly meteorological conditions) that would most impact these concentrations. This was performed by modeling ambient H_2S concentrations surrounding two landfills using an EPA recommended air dispersion model (RAM, a Gaussian-plume multiple source air quality algorithm). The two landfill scenarios included a hypothetical site and a site roughly modeled after an existing C&D debris landfill where some ambient H_2S concentrations and surface flux measurements have been collected. The concentrations occurring at any particular site will vary as a function of multiple factors and it is beyond the scope of this paper to address all possible conditions. The paper does, however, provide an overview of what might generally be expected at typical sites and what factors most control expected ambient H_2S concentrations.

METHODOLOGY

Ambient H_2S concentrations were modeled for two different scenarios. A simple hypothetical landfill was used to examine the range of typical ambient air H_2S concentrations and to examine the factors impacting concentrations. A second scenario was based on an actual landfill where the authors have performed a limited amount of H_2S emission monitoring (Reinhart and Townsend, 2003). While not an exhaustive effort, the modeling of the actual site does allow some comparison of the magnitude of modeled and measured concentrations. The methods described below include an overview of the model used, a description of the sites, and the measurement techniques performed at the actual landfill.

RAM Model

RAM is a steady-state Gaussian plume model recommended by the US EPA for evaluating the impact of emission sources on air quality over short-term periods. RAM has the capability to model emissions from point and area sources in urban or rural areas. A total of 250-point sources and 100 area sources can be modeled in one run. One set of meteorological data (e.g., wind speed, wind direction, stability, and mixing height) is considered to represent the entire region being modelled. The major inputs to the model include coordinates describing the emission sources, the emission rate, the source height, the side length of the area source, wind speed and direction, and the Pasquill stability class, among others. The model assumes that dispersion from both point and area sources results in Gaussian distributions in both the horizontal and vertical directions. The narrow plume simplification of Gifford and Hanna is used for area sources. The user's guide of the model should be consulted for more details (Catalano et al., 1987).

Site Description

The two C&D debris landfill scenarios included the hypothetical landfill and the one based upon an actual landfill. The hypothetical landfill was assigned an area of 1,000 ft by 1,000 ft (approximately 23 acres). Figure 1 (a) presents the layout of the site with respect to wind direction. The actual landfill (Figure 1 (b)) used to base the second modeling scenario on is located in Winter Garden, Florida. At the time of this study, the facility was an active C&D debris landfill and included both inactive and active fill areas. The total area of the landfill is approximately 80 acres and the design capacity is 4,000,000 yd^3. The landfill started to operate in 1991 and its annual waste disposal rate is approximately 240,000 tons. It was reported (Chakrabarti, 2002) that gypsum drywall represents approximately 4% of the volume of waste disposed in the facility.

H_2S Sampling and Measurement

An ongoing study is examining the emission rates and control of H_2S from several operating C&D debris landfills in Central Florida (Reinhart and Townsend, 2003) The landfill described above was one of the sites where both H_2S surface flux measurements and ambient H_2S concentrations were measured. The results of this study will be reported in their entirety upon completion. Some of the preliminary results were used as a comparison to the modeling work performed for this paper. At the landfill, a 65-L flux chamber (Odotech Inc.) was used to measure H_2S surface emissions. The rate of H_2S emission may be described as follows:

$$F = (\frac{V}{A}) \times (\frac{\Delta C}{\Delta t})$$

Where: F (g/m^2-s) is the flux of H_2S; V (m^3) is the volume of air within the chamber; A (m^2) is the area of soil surface enclosed by the chamber; and $\Delta c/ \Delta t$ ($g/m^3 s$) is the time rate of change of H_2S concentration. A Jerome Meter (model 631-X) from Arizona Instruments was used to analyze gas samples for hydrogen sulfide concentrations. The Jerome Meter has a detection range of 0.003 ppm to 50 ppm (Arizona Instrument LLC, 2003).

RESULTS AND DISCUSSIONS

H_2S dispersion at C&D debris landfills

The C&D debris landfills were modeled as area sources with continuous H_2S emissions. As H_2S is emitted from the surface of the landfill, it mixes and travels with the wind, which dilutes the H_2S and carries it away from the landfill. Figure 2 show the concentration contour of ambient H_2S for the hypothetical C&D landfill. This particular scenario was created under normal meteorological conditions, a stability class of 3 and a wind speed 4.5 m/s (the average North American ground-level wind speed; Noel, 1995). By considering the thermal buoyancy and

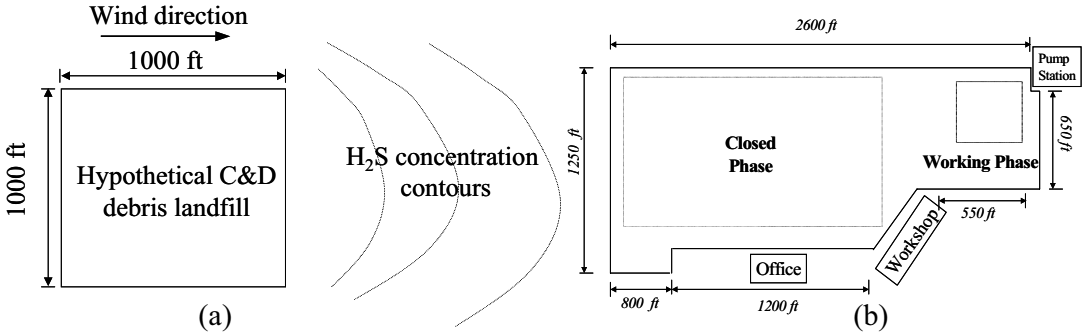

Figure 1. Definition sketch of (a) the hypothetical C&D debris landfill and (b) the scenario modeled based upon an operating C&D debris landfill.

momentum of the emitting H_2S, the mixing height was selected as 1.5 m. The H_2S emission rate, 6.5 μg/s, was calculated by the product of the total area of the hypothetical landfill and a typical flux rate of 7×10^{-8} mg/m^2-s that was measured by the flux chamber described above (Reinhart and Townsend, 2003). The impact of different flux rates will be discussed in greater detail in the next section. It can be seen that the ambient H_2S concentrations increase as the wind travels over the landfill. The maximum concentrations are observed just downwind of the landfill, and eventually these concentrations begin to diminish. Because of model limitations associated with the receptor intervals and the contour plotting method, contour lines show some degree of curvature that is not realistically expected to occur. This figure simply provides a general overview of what typical H_2S dispersion will look like. The factors impacting this dispersion will be discussed in greater detail in the following sections.

It is noted that the modeling work conducted here assumed that H_2S was conserved and not otherwise degraded or transformed. Thus the concentrations presented are conservative. The major sink for H_2S that will result in its removal from the ambient air is atmospheric conversion to sulfur dioxide (SO_2). The released H_2S has been reported to not react photo-chemically (ATSDR, 1999), but instead to react with atomic oxygen (O), oxygen (O_2), or ozone (O_3) to form SO_2 (Bibbero, 1974).

$$H_2S + O_3 \rightarrow H_2O + SO_2$$

Although SO_2 also has pungent odor, its odor threshold (1 ppm) is much higher than H_2S (0.5 ppb) (ATSDR, 2001), which means the SO_2 converted from H_2S will not be detected at concentrations expected to be encountered. In general, the lifetime of H_2S before conversion to SO_2 is on the order of hours (Seinfeld, 1975). While H_2S emitted from the landfill will ultimately be oxidized, this removal process likely does not occur until after the H_2S has been sufficiently diluted.

Effect of wind on H_2S dispersion

Wind speed and direction are among the most important factors influencing H_2S dispersion at landfills. A pre-specified point, 200 ft away from the boundary of the hypothetical landfill, was selected to examine the effect of wind speed on H_2S dispersion. The modeled H_2S concentrations at a variety of different wind speeds are shown in Figure 3 (a). As wind speed increases, the H_2S concentration decreases dramatically, illustrating how a higher wind velocity results in a greater

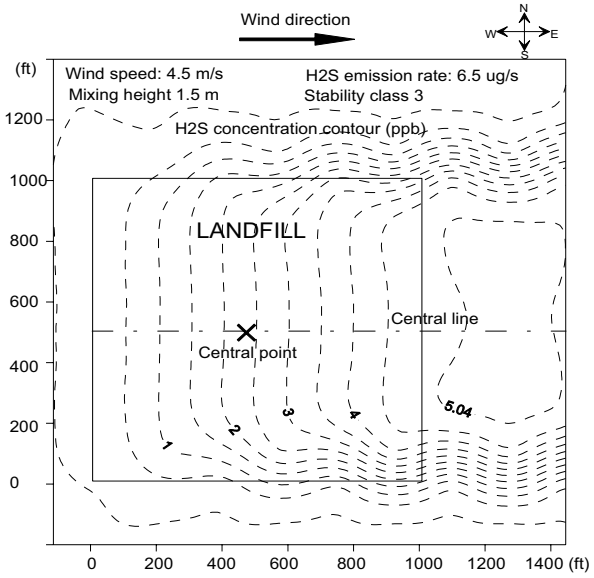

Figure 2 Ambient H_2S concentration contours at the hypothetical landfill

degree of dilution with air. Therefore, the ambient H_2S concentration is an inverse function of the wind speed.

Wind provides the primary driving force for H_2S movement and H_2S concentrations are greatest in the direction where the wind is blowing. Figure 3 (b) presents the pre-specified point of H_2S concentration change for the hypothetical landfill under different wind directions. It can be seen that H_2S concentrations at the same point are variable with the change of wind direction from East to North. The impact of changing wind speed and direction have been noted anecdotally in previous observations where H_2S odors could be detected one minute and not the next at landfills with known odor problems (Townsend et al., 2000).

Effect of maximum mixing height on H_2S dispersion

In addition to the driving force by wind, H_2S dispersion is also affected by convective and turbulent mixing resulting from vertical temperature differences. Because of solar energy, the ground temperature may be different from the temperature of overlying air, which causes convective and turbulent mixing to occur. The maximum mixing height (MMH) represents the maximum height of the convective layer and shows the vertical extent to which emitted H_2S mixing can take place. The MMH ranges from near zero to thousands of meters, depending on the season and the site topography. Figure 4 plots the results of modeled H_2S concentrations as a function of mixing heights. H_2S concentrations decrease with the increase of mixing height. At greater MMHs, more volume of air is available to dilute the emitted H_2S.

Effect of H_2S emission rate on H_2S dispersion

Another factor that obviously influences H_2S dispersion is the H_2S emission rate, which represents the amount of H_2S emitted from the landfill surface in a given time and is the product of flux rate (g/m^2-s) and emission area (m^2). Because of the heterogeneous characteristics of C&D debris, how it is disposed in landfills, differences in rainfall infiltrating into a given landfill, and differences in cover soil types and practices among sites, H_2S flux rates are expected to differ from site to site. Limited data are available describing H_2S flux rates from C&D debris. In a set of laboratory landfill columns, H_2S flux rates were measured and were found to vary over a wide range (three orders of magnitude) (Townsend et al., 2004). The maximum H_2S flux rate, found for a column of size-reduced gypsum drywall with no cover soil, was approximately 0.037 mg/ m^2-s. A similar column that had 15 cm of sandy cover soil placed on the surface had a flux rate of 1.1×10^{-3}mg/ m^2-s. In-situ measurements performed by the authors at several C&D debris landfills found H_2S surface flux rates typically less than 2×10^{-7} mg/m^2-s, with the exception of localized

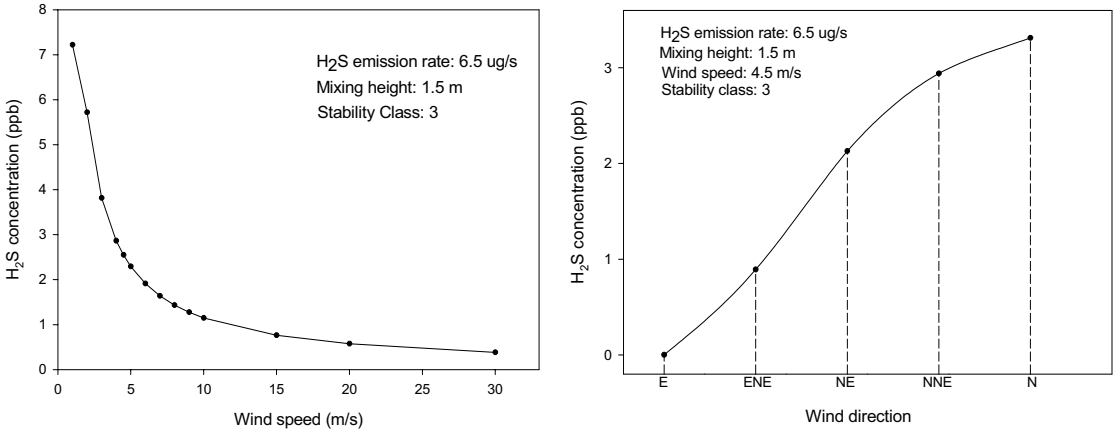

Figure 3 H_2S concentration as a function of (a). Wind speed; (b). Wind direction

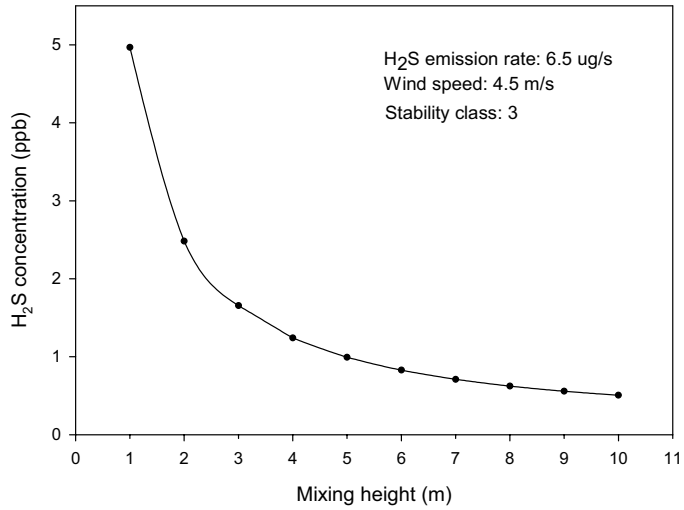

Figure 4 H_2S concentration change as a function of mixing height

"hot spots" where high H_2S emission rates as high as 0.032 mg/m^2-s were measured (Reinhart and Townsend, 2003). However, due to the small emission area, the contribution to these hot spots to the overall emission rate might be low.

Maximum H_2S concentrations were modeled as a series of possible flux rates and wind speeds at the hypothetical landfill (Figure 5). As expected, H_2S concentrations increase with an increase in the flux rate. The results indicate that at flux rates lower than 1.0×10^{-9} mg/m^2.s should not present an odor problem (under the conditions of the hypothetical scenario). At emission rates on the order of 1.0×10^{-8} mg/m^2.s, the odor will likely be noted at lower wind speeds, but should be below the odor threshold at higher wind speeds. At emission rates above 1.0×10^{-7} mg/m^2.s, the H_2S concentrations will likely not be diluted to below odor threshold by wind dilution alone.

Effect of lapse rates on H_2S dispersion

A common observation at C&D debris landfills is that H_2S concentrations (i.e., odor complaints) are greatest in the early morning hours and lowest in the afternoon. This can likely be explained by the diurnal change of mixing height. In general, minimum MMH values occur in the

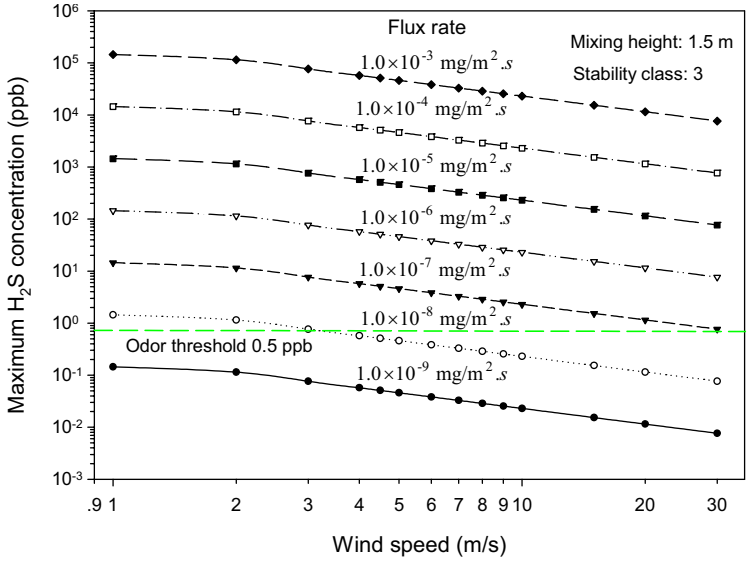

Figure 5 H_2S concentration change as function of emission rate and wind speed

morning before sunrise. As solar energy heats up the ground and warms the air, the MMH expands and rises by convection, resulting in decreasing ambient H_2S concentrations (Figure 5).

Another reason for the temporal changes in odor throughout the day might be the change of lapse rates. The lapse rate is the decreases in temperature with height in the troposphere due to the reduction of heating processes and radiative cooling of air. The environmental lapse rate changes during the whole day because of radiation inversion. The inversion occurs at night and early morning when the ground radiates heat to space, cooling the layer of air above it. When the sun rises, it heats the ground surface, resulting in the lapse rate change (Cooper and Alley, 1990).

Because of the daily change of environmental lapse rate, the H_2S vertical dispersion will also be affected. When the warm gas is emitted from the surface of a landfill, it will rise and undergo adiabatic expansion and then cool. Because the environmental temperature is inverted in the morning (Figure 6 (a) and (b)), if a warm parcel of H_2S gas is moved upward, it will follow the adiabatic lapse rate and be cooler than the surrounding environment. Negative buoyancy will force it back toward its starting spot. If the parcel of H_2S is moved downward, it will be warmer than the surrounding air, and buoyancy will also force it back toward its starting spot. Therefore, the atmospheric condition is very stable and H_2S vertical disturbances are minimized, which means the emitted H_2S cannot be diluted well in the morning. In the afternoon, however, heat from the sun eliminates the temperature inversion (Figure 6 (c) (d)). When a parcel of warm H_2S gas is released into an environment where the temperature decrease with height is greater than the adiabatic lapse, the parcel will rise rapidly. The afternoon atmosphere is unstable and conditions for the vertical dispersion of H_2S is excellent, reducing the ambient H_2S concentrations at landfills.

Effect of rain on H_2S dispersion

Rain can impact H_2S production and resulting odor complaints in several manners. In a laboratory investigation, it was observed that water addition into simulated landfill columns resulted in fluctuating H_2S emissions (Townsend, 2003). It was hypothesized that water can effectively seal the pores in cover soil and thus reduce the vertical H_2S gas permeability of the soil layer. It has been reported that the flow of gas from a landfill is greatly affected by the moisture content of the cover material (Bogner, 1992; Kjeldsen and Fischer, 1995) and the moisture content of the cover soil has been cited as the most important internal factor controlling gaseous emissions from MSW landfills (Bogner, 1992). Another possible reason for reduced H_2S emissions is its solubility in water. Due to the similar structure to water, at 27^0C to 16^0C, the solubility of H_2S in water ranges from approximately 3,018 to 4,033 mg/L (Chwirka, 1990). Therefore, when H_2S gas passes through wet cover soil, it would tend to dissolve into the water, thereby reducing H_2S

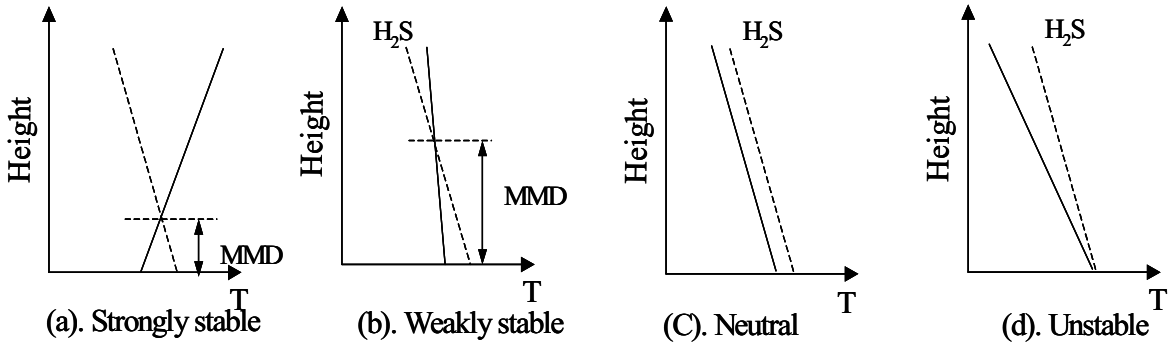

Figure 6 The effect of environmental lapse rate on H_2S vertical dispersion

emissions.

On the other hand, it has been noticed that for several days after a rain event, the ambient H_2S concentrations are higher than before (Johnson, 1986). As mentioned above, H_2S production is the result of gypsum drywall biodegradation by sulfate reducing bacteria (SRB). Sufficient water is needed for SRB survival and plays a very important role in this biochemical reaction. Stimulated by rainwater, the sulfate reducing bacteria may flourish and produce more H_2S. After the evaporation of water held in cover soils, the accumulated H_2S gas is emitted, resulting in higher ambient H_2S concentration. Rainfall might also promote the formation of "hot spots". Hot spots can result from cover soil erosion after a storm and from higher gas pressures that develop when the cover soil is saturated by rainwater. While the rain may seal the cover soil, it may also lead to the formation of distinct gaps or seeps where large amounts of H_2S escape. Those "hot spots" have been observed at many landfills, and are often associated with a black discoloration of in the surrounding soil.

Case study

As shown in Figure 1 (b), the model scenario based upon the real landfill consists of two phases, a closed phase and a working phase. The closed phase and working phase areas are approximately 232,000 m^2 and 33,000 m^2, respectively. Based on a range of in-situ flux chamber measurements at the site, the H_2S flux rate for the closed phase was modeled as 2.9×10^{-8} $mg/m^2.s$ and the emission rate for the working phase was modeled as 1.6×10^{-7} $mg/m^2.s$. Several in-situ ambient H_2S concentration surveys were made under different weather conditions. It was observed that downwind H_2S concentrations were higher than upwind. However, due to the effect of wind, ambient concentration measurements were variable. Therefore, the survey results listed in Table 1 are provided as concentration ranges. The relative wind conditions at the site during the time when the measurements were taken were accounted for in the model by adjusting the wind speed. On days where light wind conditions were noted, a wind speed of 2 m/s was used, while on days where strong wind conditions were observed, a wind speed of 6 m/s was modeled.

Table 1 shows the comparison of in-situ measurements and the calculated results by running the RAM model. Again, the data were not collected for the purpose of validating a dispersion model. These model simulations were performed to provide a rough comparison of what the model predicts versus the magnitude of the measurements at the site. The model results are comparable to the measured results. Both results showed that H_2S concentrations in the closed phase are lower than in the working phase because of cover soil. The cover soil functions as a physical barrier to reduce H_2S emission from landfills. Due to the pressure or concentration gradient, the generated H_2S tends to diffuse through the cover soil, a porous soil matrix, in which some physical or chemical gas-solid reactions would take place to reduce H_2S flux. On the other hand, in the working phase, without a barrier layer, H_2S can constantly flow out without much difficulty, resulting in a higher H_2S flux rate.

One of the reasons for the slight difference between actual measurements and modeled results is the possible change of wind speed or direction during the in-situ survey. Another difference is the minimum H_2S concentration. The Jerome meter has a detection limit of 3 ppb, while the RAM model can predict concentrations lower than 0.5 ppb.

Table1. H_2S concentration comparison between in-situ measurement and modeled results

Weather	Closed phase (ppb)		Working phase (ppb)	
	Measurement	Model	Measurement	Model
29^0C, light wind	4 - 6	2-8	8 - 13	1-13
28^0C, light wind	3 - 4	2-8	5 - 13	1-13
31^0C, strong wind	3 - 4	1-5	5 - 9	0.5-7
30^0C, strong wind	3 - 4	1-5	5 - 9	0.5-7
29^0C, strong wind	3 - 4	1-5	5 - 15	0.5-7

CONCLUSIONS

Odor problems associated with H_2S dispersion are a major concern at C&D debris landfills. H_2S emission and dispersion are affected by many factors, such as meteorological conditions, topography, and composition of the C&D debris. The ambient air H_2S concentrations surrounding these landfills, likewise, tend to be variable over time and from site to site. The RAM (Gaussian-Plume Multiple Source Air Quality Algorithm) model was used to examine the factors influencing H_2S dispersion at a hypothetical C&D debris landfills and at a scenario developed from an actual landfill site. By using the actual H_2S emission rates as model inputs, the results were comparable to the actual in-situ measurements. It is easy to get the conceptual H_2S concentration distribution in C&D debris landfills under different environment conditions, which can be conveniently used for landfill design and operation, health risk assessment, environmental impact analysis and community relations issues.

Wind plays an important role in the factors determining H_2S dispersion. In general, the higher the wind speed, the lower the ambient H_2S concentration because of the mixing and dilution by wind. Moreover, the shifting of wind direction can also reduce H_2S concentration by dispersing H_2S over a larger area. Another important factor is radiation inversion, which daily changes the environmental lapse rate and maximum mixing height (MMH) at landfills, making H_2S concentrations in the afternoon lower than in the morning. An important meteorological factor, rain can reduce H_2S emissions by sealing and dissolving diffused H_2S gas. Rain water can also, also however, stimulate the activity of sulfate reducing bacteria to produce more H_2S, resulting in higher ambient H_2S concentrations, as well as to remove cover soil and create hot-spots on the landfill surface where H_2S flux rates are at their highest.

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