

Seasonal and influent characteristic effects on hydrogen sulfide generation at a water reclamation plant

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ABSTRACT

Correlations between sulfide generation and seasonal influent wastewater characteristics were identified based on a long-term monitoring program in summer and winter at a water reclamation plant. During summer, the emission rates of hydrogen sulfide (H₂S) from the liquid treatment processes increased substantially compared to those during winter due to the increased wastewater temperature. The open tanks/clarifiers were the least significant H₂S emission contributors throughout the year. For solids-handling processes, the H₂S emission rates did not change during the year due to similar sludge characteristics in the different seasons. The fate of sulfide in liquid treatment processes was investigated as an alternative to estimation of H₂S emissions. H₂S emission from the wet well and screens was proven to be robustly associated with the wastewater temperature, flow rate, 5-day biochemical oxygen demand and total Kjeldahl nitrogen levels. However, the correlation between influent parameters and H₂S emission from aerated grit chambers was not statistically significant.

Key words | hydrogen sulfide emission, odor prediction, seasonal variation, wastewater influent characteristics

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INTRODUCTION

Wastewater treatment and solids-handling processes at water reclamation plants (WRPs) have the potential to generate malodors and cause negative impacts on the surrounding community. The odor profiles vary depending on the influent wastewater characteristics and specific plant operating conditions. Among the numerous odorous compounds, hydrogen sulfide (H₂S) has been suggested as a strong odor indicator because of the low odor detection threshold of 0.5 ppbv and large quantity of emission from treatment plants (Gostelow & Parsons 2000). H₂S can also be easily correlated to sensory odor measurements determined by olfactometry. It has been shown that the odor perception due to different odorants in air from WRPs could be primarily attributed to the main odorant, H₂S, in the air (Wang *et al.* 2011). Therefore, H₂S has been considered as an appropriate indicator for odor emissions and widely used as the criteria for odor control in WRPs and sewer systems. However, odor is not necessarily synonymous with H₂S.

Successful odor control strategies in WRPs consist of odor minimization in wastewater, odor containment and gas phase odor treatment. Considerable efforts have been

directed at odor minimization in wastewater by pretreatment of industrial wastewater before discharging it into the sewer system, prevention of anaerobic conditions and treatment of sulfide compounds (Park & Novak 2013). Gas phase odor treatment has been accomplished by various physical, chemical and biological processes (Kraakman *et al.* 2014). Determination of odorant emission rates into the atmosphere, along with the typical values for odorant concentrations in liquid phase, is critical to evaluate the dosage for chemical treatment, to design the appropriate odor treatment units and to develop proper inputs to computational dispersion modeling (Ranzato *et al.* 2012).

While odor prevention and treatment technologies continue to be developed, very little attention has been given to odor prediction by using influent wastewater characteristics, operating conditions, or meteorological conditions. Associating odorant emissions with influent characteristics could allow plants to adjust odor control measures to either reduce energy/chemical consumption or enhance the magnitude of odor control in response to fluctuations in odorant emission rates.

The specific objective of this study was to evaluate H₂S emissions and sulfide concentrations in a WRP for various influent characteristics and process operations. H₂S emissions from the headworks could be correlated to the wastewater influent characteristics to provide early-stage warning for the implementing of odor control strategies based on seasonal patterns.

MATERIALS AND METHODS

The WRP investigated in the study

The WRP investigated is a design average flow facility of 114,000 m³/day located in Illinois, USA, treating mostly domestic wastewater. The solids-handling processes comprise gravity belt thickeners, mesophilic anaerobic digesters and solid-bowl centrifuges for dewatering. This plant has typical unit processes that are widely employed by many treatment plants in the Midwest of the USA. Although odor emissions from different treatment plants vary, the general findings from this plant can be extrapolated to other similar plants with a good level of confidence. The sources of odor and sampling locations in this study are listed in Table 1.

Table 1 | Sources of odor emission and sampling locations in the WRP

Facility	Source of emission	Sampling location
Pretreatment building	Wet well and coarse screens	Vent of local exhaust system ^a (5,4) ^c
	Fine screens	Roof exhaust fans ^a (5,4)
Closed tanks	Aerated grit chambers	Vent of local exhaust system ^a (5,4)
Open tanks	Primary clarifiers	Sedimentation zone ^b and vent on the cover over the effluent weir ^a (4,4)
	Aeration tanks	Water surface ^b (4,4)
	Secondary clarifiers	Water surface ^b (4,4)
Thickening building	Gravity belt thickeners	Vent of local exhaust system ^a (5,4)
	Sludge storage tanks	Vent of local exhaust system ^a (5,4)
Dewatering building	Solid-bowl centrifuges	Roof exhaust fans ^a (5,4)

^aPoint sources.

^bArea sources.

^cNumbers in parentheses represent number of sampling events (winter, summer).

Sampling methods

Sampling campaigns for most of the odor studies reported in the literature were restricted by the project time frame and budgetary constraints, often allowing no more than four discrete sampling events for a study (Chen et al. 2011). In order to obtain more representative H₂S emission values and avoid underestimating the emission rates from all the sources in this study, 5-day sampling events were performed during winter from February to April, 2010, while 4-day sampling events were carried out during summer from June to July, 2010. The 5-month sampling took into account seasonal variations of emission rates as the result of changes of influent wastewater characteristics.

Measuring H₂S from point sources was accomplished by collecting the air from the exhaust fans or vents of local exhaust systems into 10-L Tedlar bags (Jensen Inert Products, FL) by using a vacuum chamber (St. Croix Sensory Inc., MN). H₂S emission rates were determined as a product of concentration and air flow rates from sources. The air flow rates vary between 0.58 and 2.87 m³/s. In the case of area sources, H₂S emission rates from area sources were measured by using a flux chamber (St. Croix Sensory, Inc., MN). The wind velocity within the chamber should be matched to that outside to simulate field conditions. The review by Hudson & Ayoko (2008) identified that sweep air flows could differ from 1.8 to 4.8 L/min even though the same sampling device was used in different studies. In this study, the sweep air flow was chosen to be 3.25 L/min, as recommended by the chamber's operation manual. A minimum of 45 min was allowed to pass for the air in the flux chamber to reach steady state before the samples were collected. The emission rates of H₂S were estimated by the method recommended by the chamber vendor, by normalizing the surface area of the flux chamber to the entire water surface area. Each sampling event required about 4 hours for 15 point sources and 6 hours for three area sources.

Analytical methods

Gas phase samples were analyzed using a Jerome 631-X H₂S analyzer (Arizona Instrument LLC, AZ) at ambient pressure. The detection limit for H₂S was 1 ppbv. Measurement of total sulfide concentration in the liquid phase was carried out according to the methylene blue method described in *Standard Methods* (APHA 2005). The detection range for total sulfide was 0–0.60 mg/L as S²⁻.

Characteristics of the sludge in the thickening and dewatering processes were obtained from the plant operation reports.

RESULTS AND DISCUSSION

H₂S emissions from the unit processes

It is clear from Figure 1 that the H₂S emission rates from the primary and secondary treatment units of the WRP were negligible compared to those from other treatment units. Although the clarifiers and aeration tanks have large surface area, the H₂S emissions from these area sources were very limited as a result of extremely low H₂S flux rates (0.0001–0.003 mg/hr-m²). Due to the high cost associated with covering the open tanks, the benefit/cost ratio for odor control of such open tanks is not attractive at this WRP.

During winter, the total emission rate of H₂S from the headworks (wet well, coarse screens, fine screens and grit chambers) was 5,900 mg/hr, which was approximately the same as emissions from the sludge thickening (6,600 mg/hr) and dewatering processes (5,700 mg/hr). During summer, there were significant increases in H₂S emissions for each of the headworks units. The average emission rates from the wet well/coarse screens, fine screens and grit chambers were 10, 10 and 20 times higher, respectively, in summer than in winter. The enhanced H₂S emission also increased the load of H₂S on the odor treatment units, such as biofilters and ozone oxidation equipment. Odor

treatment units are more efficient for low contaminant concentrations. Increase of H₂S load requires either longer reaction time or higher chemical dosage (Rehman *et al.* 2009; Shareefdeen *et al.* 2011; Zhang & Pagilla 2013). The H₂S emissions from the thickening and the dewatering processes were similar in summer and winter. It was therefore concluded that the liquid treatment system was the predominant contributor to total H₂S emission rate (85% of total) in summer compared to 32% of total in winter. From the monthly operating data and field measurements, the average wastewater and ambient temperatures were 8 and 11 °C higher, respectively, in summer than in winter. Increased water temperature enhances the production of H₂S by increased microbial activity in sewers and in WRPs. At the same time, an increase in wastewater temperature reduces the solubility of oxygen in wastewater. Therefore, these two factors promote oxygen depletion and development of anaerobic conditions in raw wastewater, favoring formation of sulfide in liquid. An increase of the Henry's constant for H₂S due to the increased wastewater temperature also favors volatilization of H₂S to the air. Turbulence and agitation in the headworks assist the stripping of H₂S from the liquid phase.

In contrast, the H₂S emissions from the sludge thickening and dewatering processes during summer were not significantly higher than those measured during winter. It appears that the seasonal variation of wastewater and ambient temperatures have very limited effects on H₂S generation from the solids-handling processes. These results were unexpected. It was hypothesized that the uniform sludge characteristics throughout the year caused the consistent emission rates. H₂S emitted from the thickening process was measured from the thickeners and the sludge storage tanks before and after the thickeners. H₂S emission from the thickener is due to turbulent discharge of filtrate below the belt combined with the exposed surface area of thickened sludge. H₂S was released from the sludge storage tanks due to the decomposition of organic matter under anaerobic conditions developed during storage. Of the total H₂S emission from the thickening process, only 11.5% was emitted from the thickeners. This indicates that the H₂S generated from waste activated sludge under predominantly aerobic conditions is very limited compared to that generated due to the anaerobic conditions in the sludge storage tanks. During the thickening process, the flow rates and total solids (TS) concentrations of the feed sludge remained at approximately the same level throughout the year. For the thickened sludge, there were no significant differences in TS and volatile solids (VS) content between

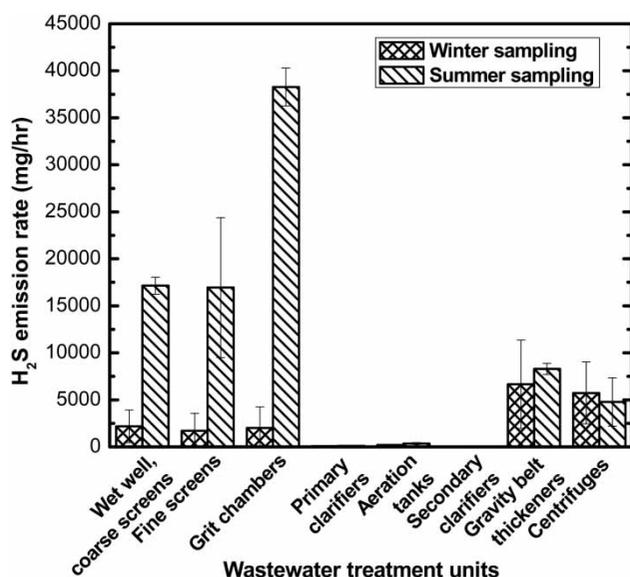


Figure 1 | H₂S emissions from the various treatment units in the WRP.

the two seasons. Therefore, in both feed and thickened sludge, approximately the same amount of organic matter was present throughout the year, with similar potential for organic matter decomposition and H₂S generation. Anaerobic digesters are sealed in this plant and do not have any known sources of gas emissions.

The emissions of H₂S and other volatile sulfur compounds after the dewatering process have been correlated with the bioavailable protein content, which is further affected by factors such as the anaerobic origin of the feed sludge, polymer use and dewatering equipment (Kim et al. 2011). The VS content of the digested sludge in the WRP did not change throughout the year, being 64 and 61% of TS in winter and summer, respectively. In addition, the same type of conditioning polymer was added to dewatering at a constant concentration as 0.5% of TS regardless of time of the year. Solid-bowl centrifuges continuously produce the biosolids with TS content of 25–27%. Since digested sludge feed, polymer use and the performance of dewatering equipment did not vary over time, it was believed that bioavailable protein content and subsequently H₂S emission rates did not vary seasonally.

To design effective odor treatment units, both average and maximum H₂S emission rates should be included as design criteria. Odor treatment units designed according to average H₂S concentrations will control the emission most of the time. However, odor complaints are likely to occur when higher H₂S concentrations are present. These concentrations may exceed the treatment capabilities of the control units. In view of the big standard deviations from Figure 1, especially for winter, the maximum H₂S emission rates could be significantly higher than the average values at the plant. Thus, in order to reduce odor complaints to a minimum number, engineers should pay more attention to the difference between the maximum and the average H₂S concentrations. Minimizing this difference will reduce the probability of emitting untreated odorous air. Importantly, due to the limited sample size ($n = 9$), the maximum emission rates quantified from the sampling campaign may be lower than the values that could be identified if more extensive sampling were undertaken. Consequently, a safety factor should be applied to these emission rates if they are used to design odor treatment units.

Sulfide in liquid treatment processes

Measurement of sulfide concentrations in liquid phase is a simple and inexpensive method and can be used as an alternative for measurement of the gas phase H₂S emission

rate. Understanding the fate of H₂S in liquid treatment processes is also critical to implement strategies for odor prevention in the liquid phase and odor treatment in the gas phase. As shown in Figure 1, in summer, approximately 85% of H₂S emission was contributed by the liquid treatment processes. Thus it was recommended that the liquid treatment processes should receive the highest priority for odor control in summer. All the unit processes in liquid treatment processes require optimal hydraulic residence time and tank depth. Therefore, these unit processes are characterized by an emitting surface (the tank surface) that is proportional to the plant capacity. Consequently, the sulfide emissions and reaction rates obtained from this study could be applied to other WRPs, as the H₂S emission rates vary proportionally to the plant size. The mass balance equation of total sulfide compounds in each unit process can be expressed as:

$$\text{Sulfide}_{\text{in}} = \text{Sulfide}_{\text{out}} = \text{Sulfide}_{\text{emitted}} = \text{Sulfide}_{\text{reacted}}$$

Five wastewater samples were collected on 5 different days between 10.00 and 15.00 hrs by grab sampling from each liquid treatment process in June and July to demonstrate the sulfide profile. The results in Table 2 provide a general indication of the severity of odors from each unit process. It can be seen that total sulfide was present at higher levels in the preliminary and primary treatment but at much lower levels in the secondary treatment. Sulfide_{emitted} was calculated based on the H₂S concentration and flow rate of the odorous air collected from the sampling

Table 2 | Sulfide profile through unit processes during summer

Unit processes	Sulfide _{in} (μg/L)	Sulfide _{out} (μg/L)	Sulfide _{emitted} (μg/L) ^a	Sulfide _{reacted} (μg/L) ^a
Wet well + screens	91 ± 11	140 ± 18	9	-58 ^b
Aerated grit chambers	140 ± 18	118 ± 10	8	14
Primary clarifiers-settling zone	118 ± 10	79 ± 4	0.02 (0.016) ^c	39
Primary clarifiers-effluent weir area	NA	NA	0.04	NA
Aeration tanks	17 ± 7	5 ± 3	0.07 (0.051) ^c	12
Secondary clarifiers	5 ± 3	4 ± 3	0.01 (0.007) ^c	1

^aBased on the influent flow rate of 1.14×10^5 m³/day.

^bNegative value indicates sulfide generation.

^cValues measured by Jeon et al. (2009).

locations listed in Table 1. The reaction term, Sulfide_{reacted} was obtained with the other three known terms. The reaction term of sulfide in liquid phase includes anaerobic reduction of sulfate to sulfide, methylation of sulfide to organic sulfur, and oxidation of sulfide to sulfate under aerobic conditions. It can be seen from the small standard deviation of sulfide levels that the total sulfide concentration of the wastewater is relatively constant, which makes it possible to use measurement of sulfide and pH from liquid samples to assess the odor generation rate and to use liquid sampling as an economical measure of supplementing the odor emission database.

As the influent wastewater passes through the wet well and screens, the anaerobic conditions prior to the aerated grit chamber keep the wastewater septic, promoting the production of sulfide. The two main mechanisms responsible for H₂S generation are decomposition of sulfur-containing organic compounds and anaerobic reduction of sulfate to sulfide by sulfate-reducing bacteria. Approximately 58 µg/L of sulfide was produced after the wastewater flowed into the plant and 9 µg/L was emitted to the air. The air emissions are due to stripping by turbulence in the hydraulic channel and the transfer points of the headworks.

In the grit removal process, the sulfide was reduced by 22 µg/L due to aerobic conditions (average dissolved oxygen (DO) concentration = 7.4 mg/L); 14 µg/L of sulfide was oxidized into sulfate and 8 µg/L of the sulfide was stripped out to the atmosphere because of the turbulence caused by the aeration. In view of the standard deviation of sulfide values used to calculate Sulfide_{reacted}, the oxidation process and stripping process in grit chambers have equal potential for the removal of sulfide from the liquid.

In the primary clarifiers, the DO concentrations of wastewater in the feed ring and effluent weir were about 5.5–5.8 and 0.6–0.9 mg/L, respectively. Aerobic conditions in the clarifiers cause oxidation of sulfide, reducing the concentrations from 118 to 79 µg/L. Only a negligible amount of sulfide (0.05% of sulfide removal) was stripped into the atmosphere. In the aeration tanks, sulfide concentration reduced from 17 to 5 µg/L. More than 99% of the sulfide removal was due to the oxidation of sulfide into sulfate and the metabolism by active biomass under aerobic conditions. Thus installing cover over the aeration tanks should not be considered as a cost-effective method for H₂S emission control. In the secondary clarifiers, sulfide was present at very low level. Only 1 µg/L sulfide was consumed and 0.01 µg/L sulfide was stripped into the atmosphere, indicating very limited microbiological activity and negligible volatilization of H₂S.

Impact of influent wastewater parameters on H₂S emissions from the headworks

During summer more than 85% of plant-wide H₂S emissions originated from the headworks (Figure 1). As raw influent wastewater receives very limited treatment when passing through the headworks, H₂S emission is more likely to be affected by the influent characteristics instead of the headworks. It was hypothesized that the emission of H₂S from the headworks is affected by several routinely-monitored wastewater parameters, including daily average wastewater temperature, flow rate, TS, 5-day biochemical oxygen demand (BOD₅) and total Kjeldahl nitrogen (TKN) concentrations. In view of the small sample size (nine samples), normal distribution of H₂S emission rates or other variables cannot be assumed. Therefore, Spearman's rho correlation (a non-parametric correlation) was applied on the association of H₂S emission rates and influent characteristics. The hypothesis (H₀: ρ = 0 against H₁: ρ ≠ 0 with α = 0.05) was tested for each of the influent parameters. The P-values for wastewater temperature, BOD₅, TKN and flow rate are lower than 0.05. Therefore, we failed to reject the null hypothesis H₀, and concluded that the emissions of H₂S were associated with wastewater temperature, BOD₅, TKN and flow rate.

Positive correlation was established between the H₂S emission rate and the BOD₅ value of raw wastewater (Table 3). This is due to the fact that higher BOD₅ concentration in wastewater can create higher potential for anaerobic conditions that cause sulfide generation. The correlation between TKN and H₂S emissions indicated that as TKN content in wastewater increases, the emission rate of H₂S also increases from the Pretreatment Building. TKN is a measure of a reduced form of nitrogen species, including organic nitrogen and ammonia from urine, proteins and

Table 3 | Spearman's rho correlations between emission rates of H₂S from pretreatment building/aerated grit chambers and selected influent characteristics

Influent characteristic	Pretreatment building		Aerated grit chambers	
	Correlation coefficient	P-value	Correlation coefficient	P-value
Water temperature	0.928	0.008	0.653	0.127
TS	0.371	0.468	0.124	0.582
BOD ₅	0.943	0.005	0.532	0.252
TKN	0.886	0.019	0.477	0.366
Flow rate	-0.886	0.019	0.707	0.108

amino acids. Following the hydrolysis of proteins to amino acids, amino acids can be decomposed anaerobically by various microorganisms to organic nitrogen and ammonia. Therefore, TKN can be an indicator of amino acid decomposition. In turn, sulfur is a component of some amino acids, including cysteine, cystine, methionine and taurine. As more amino acids are decomposed, more organic sulfur associated with nitrogen is released to the wastewater and subsequently converted into H₂S.

As mentioned above, enhanced H₂S emission rates during the summer sampling campaign were associated with higher wastewater temperature. Based on Henry's constant of H₂S, microbial activity and DO levels, a correlation between H₂S emission and wastewater temperature should be expected (P -value = 0.008). However, TS concentrations appeared to have no influence on H₂S emission (P -value = 0.468). This might be due to the significant variation of solids content and the fraction of odor-causing components in solids. In contrast to other influent parameters, a negative correlation coefficient for influent flow rate indicates that H₂S emission rates from the headworks decreased as influent flow rate increased. Flow rate increases are associated with precipitation events. Runoff introduces more DO into wastewater, allowing increased oxidation of microbially produced inorganic sulfide. Larger flow rates also dilute the sewage, decelerating oxygen depletion and the incidence of anaerobic conditions.

Spearman's rho correlation was also performed to associate the H₂S emission from the aerated grit chambers with the influent parameters (Table 3). None of the P -values was less than 0.05, indicating the influent parameters are not the predominant factors affecting H₂S emission from the grit removal process. The most significant processes are oxidation and stripping, as the aeration significantly increases the DO level (from <1.0 to 7.45 mg/L) to oxidize sulfide and creates turbulence to strip out H₂S. Aeration appears to be more important than the influent parameters in affecting H₂S emission rates from aerated grit chambers.

The above discussion indicates that wastewater temperature, flow rate, BOD₅ and TKN are correlated to H₂S emission rates from the wet well and screens, but not from aerated grit chambers. The Spearman correlation between H₂S emission rate and each of these characteristics is robust and so is the direction of the association. Therefore, a warning system could be based on these routinely measured characteristics, increasing the opportunity to initiate early odor control based on liquid phase measurements of influent characteristics or on-line monitoring of

the same. However, it is not feasible to use BOD₅ as a predictive tool because of the time required for analysis.

CONCLUSIONS

During summer, both the average and the maximum emission rates of H₂S from liquid treatment processes increased significantly compared to those measured during winter. However, for solids-handling processes, the emission rates remained constant because sludge characteristics did not vary throughout the year. H₂S emissions from area sources in the WRP were limited compared to those from point sources. This was principally a consequence of low H₂S flux rate occurred regardless of the season.

Total sulfide was present at higher levels in preliminary and primary treatment units than in the secondary treatment units. The wet well/screens and aerated grit chamber had approximately the same potential for sulfide emissions. H₂S emission from wet wells and screens was correlated to several routinely-monitored influent parameters, except TS concentration. The results suggest the use of wastewater temperature, TKN and flow rate as indicators of H₂S emission rates.

REFERENCES

- APHA, AWWA, WEF 2005 *Standard Methods for the Examination of Water and Wastewater*. 21st edn, Washington, DC.
- Chen, Y., Higgins, M., Beightol, S., Murthy, S. & Toffey, W. 2011 [Anaerobically digested biosolids odor generation and pathogen indicator regrowth after dewatering](#). *Water Research* **45** (8), 2616–2626.
- Gostelow, P. & Parsons, S. A. 2000 Sewage treatment works odor measurement. *Water Science and Technology* **41** (6), 33–40.
- Hudson, N. & Ayoko, G. A. 2008 [Odor sampling 2: Comparison of physical and aerodynamic characteristics of sampling devices: A review](#). *Bioresource Technology* **99**, 3993–4007.
- Jeon, E. C., Son, H. K. & Sa, J. H. 2009 [Emission characteristics and factors of selected odorous compounds at a wastewater treatment plant](#). *Sensors* **9**, 311–326.
- Kim, J., Park, C. & Novak, J. 2011 [Combination of coagulating agents \(aluminum sulfate and cationic polymer\) for biosolids dewatering and its impact to odors](#). *KSCE Journal of Civil Engineering* **15** (3), 447–451.
- Kraakman, N., Lebrero, R., Cesca, J. & Muñoz, R. 2014 [Evaluating odor control technologies using reliability and sustainability criteria – a case study for water treatment plants](#). *Water Science and Technology* **69** (7), 1426–1433.

- Park, C. & Novak, J. 2013 [The effect of direct addition of iron \(III\) on anaerobic digestion efficiency and odor causing compounds](#). *Water Science & Technology* **68** (11), 2391–2396.
- Ranzato, L., Barausse, A., Mantovani, A., Pittarello, A., Benzo, M. & Palmeri, L. 2012 [A comparison of methods for the assessment of odor impacts on air quality: Field inspection \(VDI 3940\) and the air dispersion model CALPUFF](#). *Atmospheric Environment* **61**, 570–579.
- Rehman, Z. U., Farooqi, I. H. & Ayub, S. 2009 Performance of biofilter for the removal of hydrogen sulfide odor. *International Journal of Environmental Research* **3** (4), 537–544.
- Shareefdeen, Z. M., Ahmed, W. & Aidan, A. 2011 [Kinetics and modeling of H₂S removal in a novel biofilter](#). *Advances in Chemical Engineering and Science* **1**, 72–76.
- Wang, T., Sattayatewa, C., Venkatesan, D., Noll, K., Pagilla, K. R. & Moschandreas, D. 2011 [Modeling indoor odor-odorant concentrations and the relative humidity effect on odor perception at a water reclamation plant](#). *Atmospheric Environment* **45**, 7235–7239.
- Zhang, Y. & Pagilla, K. R. 2013 [Gas-phase ozone oxidation of hydrogen sulfide for odor treatment in water reclamation plants](#). *Ozone: Science and Engineering* **35** (5), 390–398.

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