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# Indoor and Outdoor Air Quality Assessment of Four Wastewater Treatment Plants

Joung Ae Lee,<sup>1</sup> John C. Johnson,<sup>2</sup> Stephen J. Reynolds,<sup>3</sup>  
Peter S. Thorne,<sup>1</sup> and Patrick T. O'Shaughnessy<sup>1</sup>

<sup>1</sup>Department of Occupational and Environmental Health, The University of Iowa, Iowa City, Iowa

<sup>2</sup>Iowa Department of Public Health, Des Moines, Iowa

<sup>3</sup>Department of Environmental and Radiological Health Sciences, Colorado State University, Fort Collins, Colorado

*The study assessed the air quality of four wastewater treatment plants (WWTPs) by monitoring levels of hydrogen sulfide (H<sub>2</sub>S) and endotoxin. Samples were taken over a 1-year period (2001–2002). The unit operations at each WWTP were categorized as: (a) grit removal, (b) primary clarification, (c) biological treatment, (d) secondary clarification, (e) sludge dewatering, and (f) digestion. Temperature and humidity were monitored simultaneously, whereas airborne H<sub>2</sub>S and endotoxin were monitored at each of the six unit operations in each plant. Carbonaceous biochemical oxygen demand (CBOD) and total incoming flow of the day of visit were also recorded. The geometric means of H<sub>2</sub>S concentration were less than 1 ppm and endotoxin ranged from 6–1247 EU/m<sup>3</sup>. A mixed model analysis of covariance (ANCOVA) was used for the statistical analysis. While temperature was not associated with the levels of both contaminants, humidity was influential on the level of H<sub>2</sub>S ( $p < 0.01$ ) but not of endotoxin. CBOD did not affect the levels of either contaminant; however, incoming flows showed an association with the levels of H<sub>2</sub>S ( $p < 0.05$ ). The concentrations of H<sub>2</sub>S in the six unit operations were statistically different, whereas endotoxin did not show any differences in concentrations between units. Individual comparisons proved that concentrations of H<sub>2</sub>S in the grit removal and sludge dewatering unit operations were statistically higher than the other operations. Overall, the concentrations of H<sub>2</sub>S varied depending on total incoming flow, humidity, and different unit operations. This trend was not observed for endotoxin. The results showed that the factors analyzed affected concentrations of H<sub>2</sub>S and endotoxin differently. Therefore, different control methods for endotoxin and H<sub>2</sub>S need to be considered to effectively reduce their concentrations at WWTPs.*

**Keywords** endotoxin, hydrogen sulfide, wastewater treatment

Address correspondence to: Patrick T. O'Shaughnessy, The University of Iowa, Department of Occupational and Environmental Health, 100 Oakdale Campus, #137 IREH, Iowa City, IA 52242; e-mail: patrick-oshaughnessy@uiowa.edu.

Wastewater treatment plants (WWTPs) are typical work environments where airborne contaminants such as hydrogen sulfide (H<sub>2</sub>S) and microorganisms exist. Particularly, H<sub>2</sub>S and endotoxin are believed to cause various health problems even at low levels, and several epidemiologic studies showed high prevalence rates of respiratory health problems among workers in WWTPs and similar industries.<sup>(1–6)</sup> Although theories regarding a connection between workers' health problems and exposure to contaminants were posed in these studies, adequate information on concentration levels for evaluating the risks has not been sufficiently provided. Concentrations of both H<sub>2</sub>S and endotoxin, particularly in WWTPs, have been rarely determined.

The levels of contaminants common to WWTPs have been reported; endotoxin ranged from below detection levels to 4000 EU/m<sup>3</sup> and H<sub>2</sub>S levels were reported as less than 2 ppm (Table I).<sup>(2,4,7–9)</sup> Previous studies have also suggested that the levels of various contaminants, such as endotoxin and total bacteria, are related to WWTP procedures—the workplaces containing water agitation, such as aeration basins, inlet tunnels, or water sprinkling systems, showed the highest contaminant levels.<sup>(3)</sup> Furthermore, the exposure levels in outdoor workplace areas were lower than indoor areas.<sup>(2)</sup> Likewise, concentrations of airborne bacteria and fungi were correlated with the quantity of sewage treated.<sup>(10)</sup> However, there were a few limitations with these studies: (a) precise levels of H<sub>2</sub>S were not provided, (b) concentration measurements from area sampling devices were rarely reported, and (c) potential factors in association with the contaminants' generation rates were seldom investigated.

The aim of this study was to characterize the levels of H<sub>2</sub>S and endotoxin in WWTPs in a comprehensive manner by monitoring different unit operations at WWTPs and evaluating the influences of other operational and environmental factors on the levels of these airborne contaminants. Specifically, this study investigated three aims: (1) determine whether there is

**TABLE I. Summary of Exposure Levels of Airborne Contaminants Common to Wastewater Treatment Plants**

	Total Bacteria (10 <sup>3</sup> /m <sup>3</sup> )	Spherical Bacteria (10 <sup>3</sup> /m <sup>3</sup> )	Rod-Shaped Bacteria (10 <sup>3</sup> /m <sup>3</sup> )	Gram Negative Rods (10 <sup>3</sup> /m <sup>3</sup> )	Endotoxin (EU/m <sup>3</sup> )	Hydrogen Sulfide (ppm)
Typical levels	520	300	81	NR	300–1000	NR
Ranges	0–9500	0–6900	0–4300	0.01–100	0–4000	<1

Note: NR = not reported in literature.

an association between contaminant levels and both qualitative (carbonaceous biochemical oxygen demands) and quantitative (total incoming flow per day) characteristics of wastewater, (2) determine if environmental factors such as temperature and relative humidity affect concentration levels, and (3) determine if the detected levels are different among the various unit operations.

## METHODS

### Site Description

Four wastewater treatment plants in Iowa were chosen for this study. The four plants received both residential and industrial sewage. Total incoming flows varied from 15,140 m<sup>3</sup>/day to 143,830 m<sup>3</sup>/day and carbonaceous biochemical oxygen demand (CBOD), which is an indicator of the degree of organic contamination of incoming wastewater, ranged from 188 to 550 mg/L (Table II). The unit operations associated with each wastewater treatment plant were categorized as: grit removal, primary clarification, biological treatment, secondary clarification, sludge dewatering, and digestion, although one plant did not have a unit for sludge digestion. The specific operating techniques associated with each unit operation in the four plants are summarized in Table III. Unit operations for grit removal, sludge dewatering, and sludge digestion were located indoors in all four plants and all other units were outdoors.

### Sampling and Sample Analysis

Each of the four plants was visited multiple times during the time period between August 2001 and October 2002. At each of the six unit operations, area samples for H<sub>2</sub>S and endotoxin were taken over a 4- to 5-hour time period along with temperature and relative humidity measurements.

**TABLE II. Average Levels of CBOD and Total Incoming Flows**

Plant	CBOD (mg/L)	Total Incoming Flow (m <sup>3</sup> /day)
Plant I	197	15,140
Plant II	188	18,925
Plant III	550	143,830
Plant IV	208	75,700

Measurement instruments were placed on a large cart (1.2 m high) and placed within 1 m and downwind of the edge of a tank associated with the outdoor unit operations (primary and secondary clarification and biological degradation). The cart was placed as near to the source of open water in indoor operations as possible (grit removal and sludge dewatering). The sludge digestion operation involves enclosed tanks; therefore, sampling took place within the control room for this operation at each plant. However, the biological degradation operation in one plant was not available for sampling and one plant did not have a sludge digestion operation. Therefore, a total of 22 unit operations were sampled, and a total of 105 samples for hydrogen sulfide and 104 samples for endotoxin were collected, with 2 to 8 samples taken per unit operation per plant.

H<sub>2</sub>S was measured using a direct-reading instrument (Jerome 631-X; Arizona Instrument, Phoenix, Ariz.). An annual calibration from the manufacturer was completed for the instrument just prior to the initiation of this study. The instrument was also examined using a functional test module (FTM) to check the accuracy of measurements before each visit. As reported by the manufacturer, the limit of detection (LOD) of the instrument was 3 ppb with an accuracy of ±3 ppb at the level of 50 ppb. The instrument monitored the air for 30 sec at 5-min intervals, with a 0.15 L/min flow rate during the sampling period. The concentrations measured during each sampling period were averaged for the statistical analysis.

Endotoxin was collected using pumps (GilAir; Sensidyne, Clearwater, Fla.) with 37-mm, glass-fiber filters housed in 3-piece cassettes (SKC Inc., Eight Four, Pa.). The sampling flow rate was 2 L/min and pumps were calibrated using a soap bubble flow meter (Gilibator; Sensidyne). The concentration of endotoxin in the air was determined using the Limulus amoebocyte lysate (LAL) assay (QCL-1000; BioWhittaker Inc., Walkersville, Md.) and reported in endotoxin units per cubic meter (EU/m<sup>3</sup>). A 0.05% solution of Tween 20 in pyrogen-free water was used for the filter-extraction. The LOD for the analysis was 0.049 EU/mL of elute solution.<sup>(11)</sup>

Temperature and relative humidity were monitored over the sampling period using a direct reading instrument (Q-trak, model 5881; TSI Inc., Shoreview, Minn.). The accuracy of the temperature and humidity readings were ±0.6°C and ±3%, respectively and were calibrated with a sling psychrometer (Bacharach Inc, Pittsburgh, Pa.).

**TABLE III. Treatment Techniques**

Unit Operation	Plant I	Plant II	Plant III	Plant IV
Grit removal <sup>A</sup>	Bar screening	Bar screening, grit removing chamber	Bar screening	Bar screening
Primary clarification	Settling tank	Settling tank	Settling tank	Settling tank
Biological degradation	Trickling filter	Roughing filter, aeration basin	Trickling filter (Not accessible)	Aeration tank (Not accessible)
Secondary clarification	Settling tank	Settling tank	Settling tank	Settling tank
Sludge dewatering <sup>A</sup>	Sludge pressing	Dissolved air floatation	Incinerator (No dewatering procedure)	Sludge pressing
Sludge digestion <sup>A</sup>	Digestion	Digestion	N/A	Digestion

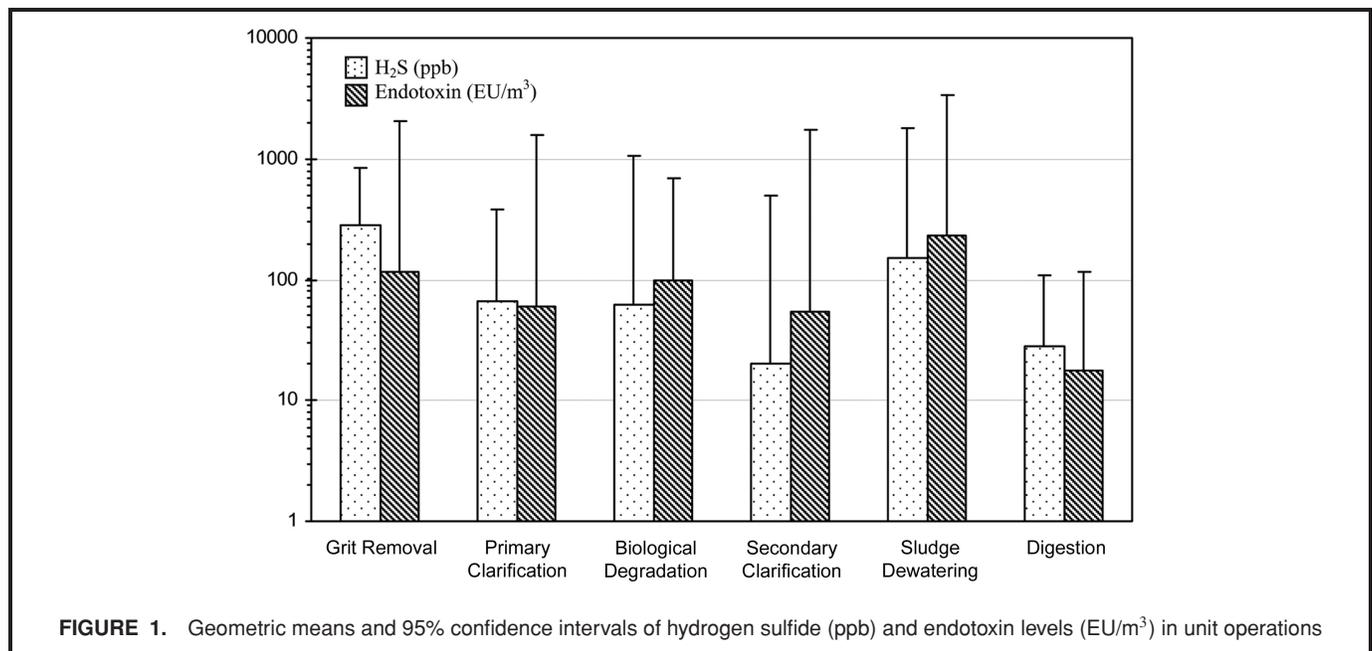
<sup>A</sup>Located indoors.

**Statistical Analysis**

Statistical analyses were performed using SAS 9.1 (SAS Institute). To investigate whether specific unit operations produced higher contaminant concentrations than others, a mixed-model ANCOVA for repeated measurements was used. The variable “six unit operations” was considered a fixed effect, and the variable “plants” was a random effect in the mixed model. Also, CBOD, total incoming flow rates, temperature, relative humidity, and indoor/outdoor were included as covariants to test their influences on the contaminant levels. A normal distribution was required for satisfying criteria needed to use the repeated mixed model; however, the datasets for both contaminants were not normally distributed, and a log-transformation was attempted to normalize the H<sub>2</sub>S and endotoxin concentrations. This effort did not make the datasets normally distributed and they were then rank-transformed as a nonparametric approach. The six unit operations were grouped

as indoor and outdoor units and differences in the levels were tested with the mixed design. As a part of the analysis, an option for a Tukey comparison was applied to identify specific unit operations showing high concentration levels compared to others. A z-test with normal approximation was also used to test the difference in contaminant levels between indoor and outdoor units.

The correlations between factors were analyzed with Pearson correlation coefficients. Significant correlations among the factors were detected; therefore, collinearity was suspected to affect the estimate value of each factor. Hence, the values of the model coefficients were compared with and without variables having correlation to see if there were significant changes in the coefficient values. The results of this analysis indicated there were no significant changes, and collinearity was assumed to have little affect on the results of the regression analysis.



**FIGURE 1.** Geometric means and 95% confidence intervals of hydrogen sulfide (ppb) and endotoxin levels (EU/m<sup>3</sup>) in unit operations

**TABLE IV. Hydrogen Sulfide Concentrations**

	Plant I			Plant II			Plant III			Plant IV		
	N	GM (ppb) (range, ppb)	CV	N	GM (ppb) (range, ppb)	CV	N	GM (ppb) (range, ppb)	CV	N	GM (ppb) (range, ppb)	CV
Grit removal	5	140.9 (3.5~544.6)	0.06	8	940.8 (344.0~3518.6)	<0.01	4	300.8 (35.5~3018.0)	0.04	8	173.8 (3.3~4659.0)	0.08
Primary clarification	4	11.0 (5.5~44.0)	0.24	5	76.1 (11.2~667.0)	0.07	4	314.0 (51.1~2718.5)	0.02	4	73.4 (54.8~145.4)	0.02
Biological degradation	4	5.6 (2.9~11.6)	0.35		N/A <sup>A</sup>		6	506.6 (76.8~7000.0)	0.01	4	30.5 (5.0~76.1)	0.11
Secondary clarification	2	4.9 (4.0~5.9)	0.27	4	2.0 (0.4~7.3)	1.72	4	134.3 (11.6~676.4)	0.04	4	66.0 (3.3~3432.3)	0.28
Sludge dewatering	5	640.2 (89.1~2834.7)	0.01	7	31.6 (1.0~1800.0)	0.40	6	940.8 (46.0~19022.2)	0.01	5	38.8 (5.0~840.0)	0.18
Sludge digestion	4	27.1 (9.1~76.8)	0.09	4	49.6 (13.8~515.9)	0.11		N/A <sup>B</sup>		4	16.4 (11.5~28.4)	0.09

Note: N = sample size; GM = geometric mean; CV = coefficient of variation.

<sup>A</sup>Area not accessible for sampling.

<sup>B</sup>No unit operation of this type at this plant.

TABLE V. Endotoxin Concentrations

	Plant I			Plant II			Plant III			Plant IV		
	N	GM (EU/m <sup>3</sup> ) (range, EU/m <sup>3</sup> )	CV	N	GM (EU/m <sup>3</sup> ) (range, EU/m <sup>3</sup> )	CV	N	GM (EU/m <sup>3</sup> ) (range, EU/m <sup>3</sup> )	CV	N	GM (EU/m <sup>3</sup> ) (range, EU/m <sup>3</sup> )	CV
Grit removal	5	40.70 (3.94~162.57)	0.11	8	103.06 (31.81~22281.46)	0.12	4	534.21 (244.15~2139.82)	0.01	8	58.59 (17.09~319.52)	0.04
Primary clarification	4	66.82 (2.43~1585.70)	0.22	5	60.87 (14.18~155.20)	0.04	4	35.59 (16.38~82.69)	0.06	4	88.79 (3.02~12983.02)	0.39
Biological degradation	4	89.07 (6.53~1394.90)	0.10		N/A <sup>A</sup>		6	147.77 (29.00~4097.26)	0.04	4	62.37 (6.40~312.64)	0.09
Secondary clarification	2	66.00 (65.42~66.58)	0.02	4	38.61 (32.65~51.00)	0.03	4	68.17 (14.44~168.09)	0.04	4	55.85 (0.90~12234.14)	0.97
Sludge dewatering	5	308.47 (70.98~2261.91)	0.01	6	1247.65 (87.1~34727.85)	0.01	6	393.37 (26.13~3201.115)	0.02	5	15.45 (2.60~49.67)	0.19
Sludge digestion	4	34.14 (5.28~594.67)	0.22	4	27.40 (14.58~82.47)	0.08		N/A <sup>B</sup>		4	6.29 (3.30~11.09)	0.27

Notes: N = Sample size; GM = geometric mean; CV = coefficient of variation.

<sup>A</sup>Area not accessible for sampling.

<sup>B</sup>No unit operation of this type at this plant.

**TABLE VI. Mixed Model Analysis**

Effect	Num DF <sup>A</sup>	Den DF <sup>B</sup>	F Value	PR > F
Unit operations	5	76	4.45	0.0017
Total flow	1	76	12.98	0.0006
Relative humidity	1	76	6.27	0.0144

Note: Mixed model analysis: the association between H<sub>2</sub>S concentrations and different unit operations and environmental factors.

<sup>A</sup>Numerator degree of freedom (k-1).

<sup>B</sup>Denominator degree of freedom (n-1).

## RESULTS

In general, geometric means (GMs) of the H<sub>2</sub>S levels at 22 unit operations in the four plants were less than 1 ppm (Figure 1, Table IV), whereas endotoxin showed a very broad range of GMs among the unit operations, ranging from 6–1248 EU/m<sup>3</sup> (Figure 1, Table V). Coefficients of variation (CVs) were calculated to determine seasonal variations. The contaminant levels showed broad ranges in CV values for both contaminants: <0.001 to 1.72, and 0.01 to 0.97 for H<sub>2</sub>S and endotoxin, respectively.

After including temperature, relative humidity, CBOD, and total incoming flow rates into the statistical analysis, the results showed the concentrations of H<sub>2</sub>S were significantly affected by the three factors: unit operations, total incoming flow, and relative humidity (p-values <0.05) (Table VI). However, none of the factors of interest in this study were associated with endotoxin concentrations. Individual comparisons of the six different unit operations were analyzed in the mixed model

to detect specifically which units contained high levels of contaminants compared to the others. According to the results of the Tukey analysis, concentrations of H<sub>2</sub>S in the grit removal unit were statistically higher than all other unit operations except sludge dewatering unit (p-values <0.05). Furthermore, H<sub>2</sub>S levels in the sludge dewatering unit were significantly higher than the monitored levels in the secondary clarification and digestion units (p-values < 0.05). Concentrations of endotoxin were not statistically different among the six unit operations (Table VII).

Relative humidity was generally constant for both indoor and outdoor unit operations over the four different sampling visits, whereas temperatures at outdoor units varied as expected (Table VIII). Pearson correlation analysis revealed that relative humidity and CBOD were significantly correlated (r = 0.31, p < 0.01). The association between CBOD and incoming flow rates also proved to be significant (r = 0.87, p < 0.01). Furthermore, a z-test revealed that H<sub>2</sub>S concentrations were higher indoors than outdoors (p < 0.001) (Table IX).

## DISCUSSION

The statistical analysis to compare the concentrations taken from the different unit operations was performed under two assumptions: (1) the chosen four plants were homogeneous, and (2) that each designated unit operation in each of the four plants involved the same treatment technique.

As shown in Table III, the chosen plants used a variety of treatment techniques for the same unit operation. For example, a trickling filter, roughing filter, and aeration tank were utilized for the biological degradation unit operation. However, one of our aims was to detect whether certain

**TABLE VII. Paired Comparison of Hydrogen Sulfide and Endotoxin Concentrations Among the Six Unit Operations**

Unit Operation		Hydrogen sulfide		Endotoxin	
		DF	t Value	DF	t Value
Grit removal	Primary clarification	70	2.63 <sup>A</sup>	62	-0.57
Grit removal	Biological degradation	70	2.69 <sup>A</sup>	62	-0.78
Grit removal	Secondary clarification	70	3.82 <sup>B</sup>	62	-0.40
Grit removal	Sludge dewatering	70	0.40	62	-0.51
Grit removal	Digestion	70	2.51 <sup>A</sup>	62	0.22
Primary clarification	Biological degradation	70	0.10	62	-0.27
Primary clarification	Secondary clarification	70	1.22	62	0.16
Primary clarification	Sludge dewatering	70	-2.22 <sup>A</sup>	62	0.09
Primary clarification	Digestion	70	0.13	62	0.66
Biological degradation	Secondary clarification	70	1.11	62	0.41
Biological degradation	Sludge dewatering	70	-2.29 <sup>A</sup>	62	0.32
Biological degradation	Digestion	70	0.04	62	0.83
Secondary clarification	Sludge dewatering	70	-3.42 <sup>B</sup>	62	-0.06
Secondary clarification	Digestion	70	-0.97	62	0.52
Sludge dewatering	Digestion	70	2.15 <sup>A</sup>	62	0.65

<sup>A</sup>p-value <0.05.

<sup>B</sup>p-value <0.01.

**TABLE VIII. Geometric Means of Temperature and Relative Humidity for Indoor and Outdoor Unit Operations**

Unit Operations	Period of Visits	Temperature (°C)	Relative Humidity (%)
Indoor <sup>A</sup>	Aug.–Sep.2001	23.3	57.5
	Oct.–Nov. 2001	18.9	52.3
	Feb.–Mar. 2002	10.5	49.8
	Sep.–Oct. 2002	20.5	56.5
Outdoor <sup>B</sup>	Aug.–Sep. 2001	24.1	50.1
	Oct.–Nov. 2001	16.2	57.6
	Feb.–Mar. 2002	1.4	52.8
	Sep.–Oct. 2002	23.5	49.0

<sup>A</sup>Grit removal, sludge dewatering and digestion units.

<sup>B</sup>Primary and secondary clarifications and biological degradation units.

unit operations produced statistically different contaminant concentration levels than the others regardless of technique; thus, the effect of different treatment plants on concentration levels was not considered as a factor of interest but rather taken as a random effect in the statistical analysis for the study. Therefore, the comparison between different unit operations involved a comparison between the combinations of different treatment techniques associated with each unit operation rather than each treatment technique separately.

From the comparison of H<sub>2</sub>S concentrations among unit operations, the levels associated with grit removal and sludge dewatering were statistically higher than the levels of the other units. Normally, grit removal, biological degradation, and sludge dewatering involve a large amount of water movement and resulting turbulence, whereas water is retained in primary and secondary clarification tanks for hours with minimal agitation. Therefore, the physical nature of the unit operations influenced the different levels of contaminants measured during this study. In addition, the significant difference between concentrations of H<sub>2</sub>S at indoor and outdoor units supports the effect of locations. Compared with contaminants at indoor units (which include grit removal and sludge dewatering) contaminants at outdoor units are likely to be diluted by air dispersion. This outcome suggests that the level of control needed to minimize H<sub>2</sub>S levels should be different depending on unit operation type and that the level of control needed to reduce H<sub>2</sub>S exposures is related to the amount of wastewater flow through a plant. In this study, endotoxin outcomes showed no difference between

**TABLE IX. Least Square Means Estimate of H<sub>2</sub>S Levels**

Effect	Estimate	Standard		DF	t Value	Pr >  t
		Error	DF			
Indoor/outdoor units	1.7076	0.5877	67	2.91	<.005	

different unit operations, whereas such a difference was found in another study.<sup>(2)</sup> The lack of an association between unit operation and endotoxin levels in this study may be due to the wide range of variability among the observations.

There are potential health concerns associated with the H<sub>2</sub>S levels measured during this study. In this study, 17% of the 105 measurements were higher than the proposed threshold limit value of 1 ppm for H<sub>2</sub>S. Furthermore, chronic exposures to low levels of H<sub>2</sub>S have been associated with health problems, such as neurological and respiratory symptoms.<sup>(12–14)</sup> Likewise, considering suggested levels of 50 EU/m<sup>3</sup> for endotoxin, the results from this study showed that 60.9% of overall endotoxin levels exceeded the suggested occupational exposure limit level.<sup>(15)</sup> Therefore, further investigations regarding health problems associated with exposures to hydrogen sulfide and endotoxin among wastewater treatment plant workers are warranted.

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