

# A full-scale integrated-bioreactor with two zones treating odours from sludge thickening tank and dewatering house: performance and microbial characteristics

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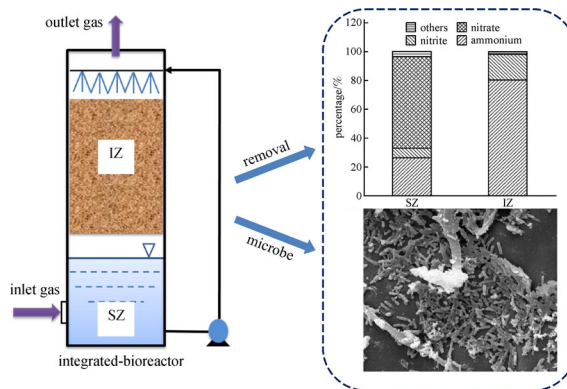
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## HIGHLIGHTS

- The integrated-bioreactor consists of a suspended zone and an immobilized zone.
- H<sub>2</sub>S and NH<sub>3</sub> from WWTP were effectively eliminated by the integrated-bioreactor.
- Different microbial populations dominated in the individual zones.
- Most of the H<sub>2</sub>S was bio-oxidized into elemental sulfur and sulfate in IZ.
- Large amount of NH<sub>3</sub> was converted into nitrate and nitrite in SZ.

## GRAPHIC ABSTRACT



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## ABSTRACT

A full-scale integrated-bioreactor consisting of a suspended zone and an immobilized zone was employed to treat the odours emitted from a wastewater treatment plant. The inlet concentrations of H<sub>2</sub>S and NH<sub>3</sub> were 1.6–38.6 mg·m<sup>-3</sup> and 0.1–6.7 mg·m<sup>-3</sup>, respectively, while the steady-state outlet concentrations were reduced to 0–2.8 mg·m<sup>-3</sup> for H<sub>2</sub>S and 0–0.5 mg·m<sup>-3</sup> for NH<sub>3</sub>. Both H<sub>2</sub>S and NH<sub>3</sub> were eliminated effectively by the integrated-bioreactor. The removal efficiencies of H<sub>2</sub>S and NH<sub>3</sub> differed between the two zones. Four species of microorganisms related to the degradation of H<sub>2</sub>S and NH<sub>3</sub> were isolated. The characteristics and distributions of the microbes in the bioreactor depended on the inlet concentration of substrates and the micro-environmental conditions in the individual zones. Product analysis indicated that most of the H<sub>2</sub>S was oxidized into sulfate in the immobilized zone but was dissolved into the liquid phase in the suspended zone. A large amount of NH<sub>3</sub> was converted into nitrate and nitrite by nitrification in the suspended zone, whereas only a small amount of NH<sub>3</sub> was transferred to the aqueous phase mainly by absorption or chemical neutralization in the immobilized zone. Different microbial populations dominated the individual zones, and the major biodegradation products varied accordingly.

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## 1 Introduction

Odours produced and emitted from wastewater treatment systems and sludge handling processes can become a nuisance to adjacent areas and are considered atmospheric pollution. With the significant expansion of communities

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in recent years, some older wastewater treatment plants (WWTPs) are now adjacent to neighborhoods. Effects from the odours can worsen during summer or with shifting wind direction. Odour control has been a well-researched concern regarding waste and wastewater treatment processes for many years. Biological technologies such as biofilters [1,2], bioscrubbers [3], and biotrickling filters [4] have been preferentially applied in WWTPs for odour control because of their efficiency, cost effectiveness, and environmental acceptability [5,6]. About 200 biofilters, including large full-scale and simple open systems, have been installed to eliminate odours and VOCs since the late 1980s [7,8]. Most of the odours come from sulfur- and nitrogen-based compounds, of which hydrogen sulfide and ammonia are the predominant species [9,10]. Removal of the major odorants hydrogen sulphide ( $H_2S$ ) and ammonia ( $NH_3$ ) during waste bioprocessing has been studied extensively [11,12]. Rabbani et al. reported on the simultaneous removal of  $H_2S$  and  $NH_3$  by a biofilter in a WWTP. The biofilter achieved removal efficiencies greater than 90% for both the gases [13]. A pilot scale biotrickling filter was applied to treat  $H_2S$  released from a WWTP and more than 90% of the  $H_2S$  was removed [14].

Within bioreactors, microorganisms produce enzymes that biodegrade malodorous pollutants into less hazardous products. A short adaptation period can be achieved by inoculating the reactors with the specialized microorganisms. This strategy improves odour control while maintaining the maximum efficiency of the biofilters. Various methods have been used to monitor microorganism variation; in specific, the plate count method has been employed for non-acidophilic and acidophilic *Thiobacillus* whereas the most probable number (MPN) method has been utilized for ammonium- and nitrite-oxidizing bacteria [15,16]. *Thiobacillus* sp. isolated from bioreactors that treated  $H_2S$  [17–19], could act as electron acceptors and were the main functional bacteria present during in sulfur-containing compounds removal [16,20].

The processes of contaminants biodegradation within a bioreactor involves three stages. The contaminants first transfer from air to the water phase, then sorb into the biofilm and onto the filter medium, and finally undergo biodegradation [21]. Generally, biofilters are adapted to treat both hydrophilic and hydrophobic substances, while biotrickling filters are more suitable for the removal of water soluble substances. Only readily soluble substances with Henry coefficients  $< 0.01$  can be effectively removed by bioscrubbers [22].

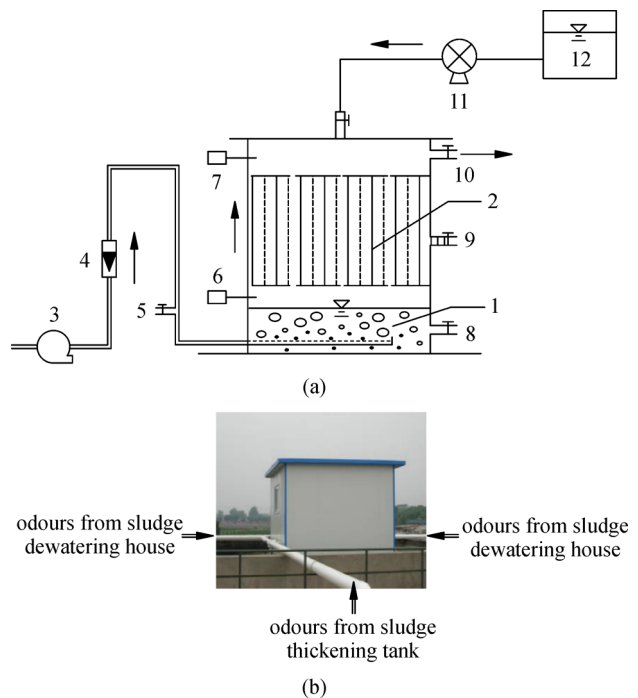
Gaseous effluents from WWTPs include a complex mixture of odorous compounds [23]. Contaminants can not be simultaneously and completely degraded by using only one type of microorganism. In the present study, a full-scale integrated-bioreactor was developed to eliminate the unpleasant odours emitted from a municipal WWTP. This bioreactor consisted of two reaction zones: a suspended

zone (SZ) with suspended growth microorganisms and an immobilized zone (IZ) packed with materials for biofilm formation. Different microorganisms can be set to dominate in the individual zones by controlling their micro-environmental conditions in such zones. The performance of the integrated-bioreactor was monitored continuously for 7 months. Microorganisms related to odor biodegradation were isolated from the integrated-bioreactor. The characteristics and distribution of the microorganisms and products produced in each zone of the bioreactor were observed during the treatment process. The objectives of this work were to 1) investigate the performance of the integrated-bioreactor for odours treatment; 2) assay the microbial population in each reaction zone, and 3) provide an effective method for the removal of unpleasant odours caused by pollutants with different water solubilities.

## 2 Materials and methods

### 2.1 Integrated-bioreactor set up

A full scale odour control system consisted of gas collection pipes, blower, integrated-bioreactor, and detectors (Fig. 1(a)) was assembled to eliminate odours emitted from a municipal WWTP in Beijing. This treatment plant was built in 1996 and has been operational for more than



**Fig. 1** (a) Schematic diagram of the treatment process: (1) the suspended zone, (2) the immobilized zone, (3) air pump, (4) flowmeter, (5) sampling port 1, (6) monitor instrument 1, (7) monitor instrument 2, (8) sampling port 2, (9) sampling port 3, (10) sampling port 4, (11) meter pump, (12) the nutrient solution tank; (b) the integrated-bioreactor for odour control

20 years. Nearly 20000 m<sup>3</sup> of domestic wastewater from communities was treated daily by a sequencing batch reactor in this plant. The moisture content in the excess sludge was reduced to below 80% by a belt-type sludge-dehydrating machine in the dewatering house.

The main sources of unpleasant odours in the WWTP were a sludge thickening pond and a sludge dewatering facility. The components of the odour emitted from the sludge treatment facility were sulfur-containing, nitrogen-containing, and volatile organic compounds (Table 1). We measured 0.1–7.8 mg·m<sup>-3</sup> of H<sub>2</sub>S and 0.1–3.2 mg·m<sup>-3</sup> of NH<sub>3</sub> emitted from the sludge thickening pond. Meanwhile, we noted 1.5–38.8 mg·m<sup>-3</sup> of H<sub>2</sub>S and 1.0–6.5 mg·m<sup>-3</sup> of NH<sub>3</sub> generated from the sludge dewatering facility. Therefore, H<sub>2</sub>S and NH<sub>3</sub> were selected as the predominant pollutants for removal. Aside from their difference in water solubility, the two pollutants also vary in pH; H<sub>2</sub>S is acidic, whereas NH<sub>3</sub> is alkaline [24]. Considering the characteristics of the pollutants, we designed and applied an integrated-bioreactor. This bioreactor (Fig. 1) consisted of two reaction zones: a suspended zone (SZ) with suspended growth microorganisms and an immobilized zone (IZ) packed with materials for biofilm formation. Each zone had different micro-environmental conditions, such as pH and relative humidity. Hence, the types of microorganisms dominating each zone were also expected to differ.

Odours from these two treatment processes were collected by pipes and were introduced into the treatment system through a blower. The effective volume of the integrated-bioreactor was 3.5 m<sup>3</sup>, and the volumes of the suspended and immobilized zones were 0.85 and 2.65 m<sup>3</sup>, respectively. Polyurethane foam cubes of 1–2 cm<sup>3</sup> were packed in the immobilized zone as the packing material. The total flow rate was 250 m<sup>3</sup>·h<sup>-1</sup>, and the corresponding empty bed residence time was 60 s. The bioreactor was inoculated with *Thiobacillus thiooxidans*, which was previously isolated and mass cultivated in the laboratory. The amount of the *Thiobacillus thiooxidans* was about 9.0 × 10<sup>6</sup> CFU·(g dry packing material)<sup>-1</sup>. The filtered effluent from this WWTP was sprayed into the bioreactor periodically to maintain an adequate supply of nutrient and moisture for microorganism growth. A thermal insulation building was installed to protect the biofilter from rain (Fig. 1(b)).

## 2.2 Analytical methods

### 2.2.1 Chemical analysis

Different samples, including the gas samples from the inlet and outlet, liquid samples from the SZ and IZ, and solid samples from the packing media, were picked up periodically to monitor bioreactor performance.

H<sub>2</sub>S concentrations were determined using a gas chromatography (GC) (Agilent 6890 N, USA) with a flame photometric detector (FPD) and a DB-1701 capillary column (30 m × 0.32 mm × 0.25 μm; Hewlett Packard, USA). Nitrogen with a flow rate of 65.6 mL·min<sup>-1</sup> was used as the carrier gas. The oven, injection and detector temperatures were 45°C, 100°C and 200°C, respectively. NH<sub>3</sub> was collected from the sampling ports and was absorbed by a 5 mol·L<sup>-1</sup> sulfuric acid solution for 10 min, then a standard analysis method was selected for NH<sub>3</sub> concentration analysis, as described in a previous report [25,26].

The concentrations of nitrate, nitrite, and sulfate in the liquid phase were measured using ion chromatography (ICS-1000, Dionex ion chromatography system, USA) [26]. Sulfate ions were determined by an ion analyzer (Leici, PHS-3C, INESA, Shanghai, China). The sulfur-containing compounds in the packing material were observed through scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) (HITACHI S-3000N /EDAX Inc., Japan). The pH values in each zone of the bioreactor was measured by a pH meter (pH-3C, Shanghai, China). A Dewpoint Thermohygrometer (WD-35612, OAKTON, Germany) was used to measure temperature and relative humidity of each zone. The flow rate of gas was determined using a flow meter.

### 2.2.2 Microbial assay

One gram of polyurethane foam cubes (or 1.0 mL of liquor samples) was periodically collected from each sampling port for microbial enumeration, and then were mixed with 100 mL of sterile water and was agitated for 10 min. Thiosulfate and modified Waksman media were used to culture non-acidophilic and acidophilic *Thiobacillus*,

**Table 1** Main composition of odors

compounds	molecular	OTCs <sup>a)</sup> /ppm	H <sub>i</sub> <sup>b)</sup> /(25°C)	concentrations /(mg·m <sup>-3</sup> )	
				ST <sup>c)</sup>	SD <sup>d)</sup>
hydrogen sulfide	H <sub>2</sub> S	0.18	0.92	0.1–7.8	1.5–38.8
ammonia	NH <sub>3</sub>	0.90	0.005	0.1–3.2	1.0–6.5
methyl mercaptan	CH <sub>4</sub> S	0.10 × 10 <sup>-3</sup>	IS <sup>f)</sup>	0–0.001	0–0.001
methyl sulfide	C <sub>2</sub> H <sub>6</sub> S	U <sup>e)</sup>	IS	ND <sup>g)</sup>	0–0.001
styrene	C <sub>8</sub> H <sub>8</sub>	0.15–25	IS	0.01–0.05	0.01–0.10

Notes: a) OTCs, odor threshold concentrations; b) H<sub>i</sub>, Henrys' coefficient; c) ST, sludge thickening tank; d) SD, sludge dewatering facility; e) U, unknown; f) IS, insoluble in water; (g) ND, not detected

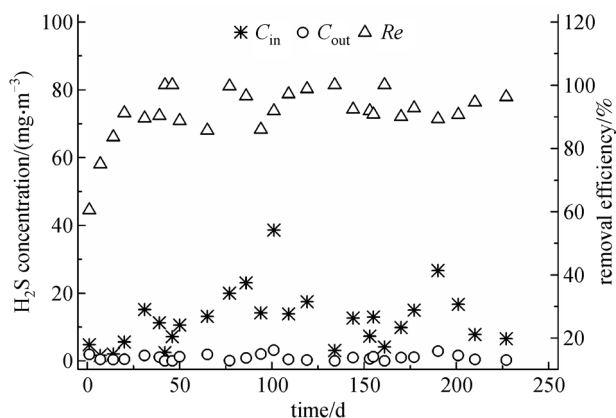
respectively [16]. Inoculation was conducted in triplicates, and the average value for each sample was calculated. The number of microorganisms was expressed as CFU·g<sup>-1</sup> of dry packing medium or CFU·mL<sup>-1</sup> of liquor. The number of ammonia- and nitrite-oxidizing bacteria was determined according to the MPN–Griess counting method described by Both et al. [15]. The bacteria were incubated in the dark at 28°C for 30 days. The counts of ammonia- and nitrite-oxidizing bacteria were presented as MPN·g<sup>-1</sup> of dry packing medium (MPN·g<sup>-1</sup>) or MPN·mL<sup>-1</sup> of liquor.

Samples were obtained from the IZ to observe the penetration of biofilms of microorganisms into the packing material by a scanning electron microscope (HITACHI S-3000N/EDAX Inc., Japan). The procedures of the samples preparation for microbial observation were described in previous reports [27].

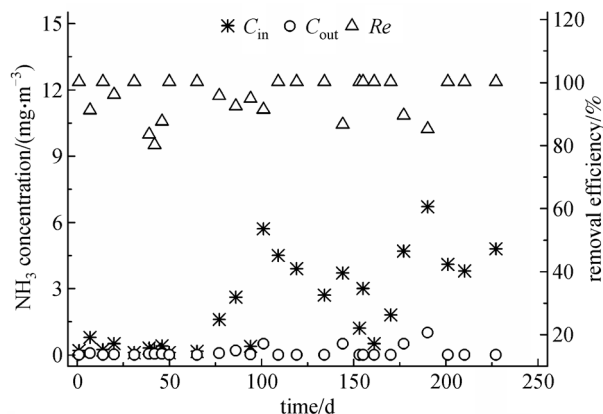
### 3 Results and discussion

#### 3.1 Performance of the integrated-bioreactor for odours removal

The present study evaluated the performance of the integrated-bioreactor for more than 227 days. The inlet and outlet concentrations and removal efficiencies of H<sub>2</sub>S and NH<sub>3</sub> in the integrated-bioreactor are shown in Figs. 2 and 3, respectively. The inlet concentrations of H<sub>2</sub>S were 1.6–38.6 mg·m<sup>-3</sup>, while the steady-state outlet concentrations were reduced to 0–2.8 mg·m<sup>-3</sup>. The concentrations of H<sub>2</sub>S in the outlet stream changed with that in inlet stream (Fig. 2). After adaptation, the removal efficiency of H<sub>2</sub>S increased gradually from 60% to over 90% and was maintained at this level for nearly 6 months. The concentration of NH<sub>3</sub> in the inlet stream was much lower than that of H<sub>2</sub>S. High removal efficiency (95.3% on average) was obtained for NH<sub>3</sub> (Fig. 3). The inlet loads of



**Fig. 2** Inlet and outlet concentration and removal efficiency for H<sub>2</sub>S. *C<sub>in</sub>*: inlet concentration; *C<sub>out</sub>*: outlet concentration; *Re*: removal efficiency



**Fig. 3** Inlet and outlet concentration and removal efficiency for NH<sub>3</sub>. *C<sub>in</sub>*: inlet concentration; *C<sub>out</sub>*: outlet concentration; *Re*: removal efficiency

H<sub>2</sub>S and NH<sub>3</sub> were 0.11–0.86 g·m<sup>-3</sup>·h<sup>-1</sup> and 0.007–0.16 g·m<sup>-3</sup>·h<sup>-1</sup>, respectively. The maximum elimination capacities were 2.53 g·m<sup>-3</sup>·h<sup>-1</sup> for H<sub>2</sub>S and 0.41 g·m<sup>-3</sup>·h<sup>-1</sup> for NH<sub>3</sub>. Both compounds were removed effectively in the integrated-bioreactor during the operational period.

When the odorous compounds reached the suspended zone (SZ), the soluble gas compounds (i.e., NH<sub>3</sub>) in the gases transferred immediately to the liquid phase. Both compounds were biodegraded subsequently by bacteria. The gases were also humidified simultaneously in this zone. Next, the gases migrated to the immobilized zone (IZ), and the microorganisms growing on the packing materials in this zone removed the less water-soluble compounds (i.e., H<sub>2</sub>S). Therefore, various components in gas can be removed by the integrated-bioreactor.

The NH<sub>3</sub> removal efficiency was much higher in the SZ (63.2%) than in the IZ (32.1%). By contrast, most of the H<sub>2</sub>S (66.0%) was eliminated by the IZ. This phenomenon indicates that the removals of NH<sub>3</sub> and H<sub>2</sub>S were different in the SZ than they were in the IZ. Bioreactor performance often depends on the interactions of the contaminant characteristics including adsorptive power, solubility, and potential biodegradability, and the operating conditions of the system. The hydrophilic compound NH<sub>3</sub> exhibited a higher removal efficiency in the SZ than in the IZ. The SZ was maintained with a set volume of liquor that contained the suspended growth bacteria. Hence, the soluble NH<sub>3</sub> could easily transfer from the gas phase to the liquid phase and could be effectively biodegraded effectively in this zone. The IZ contained the packing material on which the microorganisms attached and grew. The porous packing material supplies a large surface area, in which microorganisms can come into contact with the odorous compounds flowing through the bioreactor. Therefore, the less water-soluble compound H<sub>2</sub>S achieved a higher removal efficiency in the IZ than it did in the SZ. Odour containing compounds with different water solubilities can

be treated effectively through the synergistic action of the two zones in the integrated-bioreactor. The size and ratio of each zone could be designed and optimized based on the dominant odorous contaminant characteristics.

### 3.2 Microbial characterization for the integrated-bioreactor

#### 3.2.1 Biomass observations

Gas-phase bioreactors utilize microbial metabolic reactions to remove contaminants from air. A community of microorganisms populates the reactor, utilizing the contaminants for respiration and metabolization [21]. Therefore, dense biomass presents on the packing materials' surfaces. SEM provided detailed images of the distribution of microorganisms from the surface to the inside of a polyurethane foam cube used as packing material (Fig. S1). Large amounts of bacillus, coccus, and fungi mycelium accumulated on the surface, forming a biofilm. The number of microorganisms was decreased with the depth in the foam cube, and there were only a few bacilli appeared at the center. Concentrations of H<sub>2</sub>S and NH<sub>3</sub> varied as air passed through the cubes. Concentrations were much higher on the surface than they were at the center.

#### 3.2.2 Functional bacteria

Four species of microorganisms were isolated from the integrated-bioreactor arranged to operate for more than 6 months. Non-acidophilic and acidophilic *Thiobacillus* consumed the H<sub>2</sub>S, whereas ammonium- and nitrite-oxidizing bacteria oxidized the NH<sub>3</sub>. In the SZ,  $(9.85 \pm 0.99) \times 10^6$  CFU·mL<sup>-1</sup> of non-acidophilic *Thiobacillus* and  $(9.50 \pm 1.01) \times 10^5$  CFU·mL<sup>-1</sup> of acidophilic *Thiobacillus* were found, whereas  $(2.13 \pm 0.37) \times 10^6$  CFU·g<sup>-1</sup> of non-acidophilic *Thiobacillus* and  $(1.32 \pm 0.32) \times 10^7$  CFU·g<sup>-1</sup> of acidophilic *Thiobacillus* were present in the IZ. Strains of *Thiobacillus* have often been isolated and identified as efficient degraders of sulfur-containing compounds in biofilters [16,28–30]. Wang et al. reported that *Thiobacillus* dominated in four reactors for H<sub>2</sub>S removal [20]. *Thiobacillus thiooparus* acted as desulphurizing bacteria during biodegradation of dimethyl sulphide and methanethiol [28]. The results in Table 2 show that non-acidophilic and acidophilic *Thiobacillus*

dominated both zones.

Competition may occur when mixed contaminants are treated within one bioreactor. The inlet concentrations of H<sub>2</sub>S were 1.6–38.8 mg·m<sup>-3</sup>, which were considerably larger than those of NH<sub>3</sub> (0.1–6.7 mg·m<sup>-3</sup>). The degrading microorganisms will grow rapidly and become abundant when an air contaminant becomes a dominant substrate. Therefore, sulfur-oxidizing bacteria dominated both zones. A similar phenomenon was reported in a previous study on the removal of dichloromethane and toluene. The species that degraded the substrates dominated in separate volumes within one biofilter [31].

The treatment of contaminants relies on developing enhanced microbial communities within the bioreactor. Mixed contaminants may require different microorganisms species and many metabolic steps in the transformation. The different removal characteristics of H<sub>2</sub>S and NH<sub>3</sub> were due to the different types, numbers, and activities of the microorganisms formed in the two zones of the integrated-bioreactor. More ammonium- and nitrite-oxidizing bacteria were found in the SZ than in the IZ, so that NH<sub>3</sub> removal efficiency was much higher in the SZ than in the IZ. The abundance of non-acidophilic and acidophilic *Thiobacillus* led to good H<sub>2</sub>S removal efficiency in the IZ.

### 3.3 Characteristics of the degradation products

#### 3.3.1 Products of H<sub>2</sub>S degradation

The main products of H<sub>2</sub>S bio-oxidation are elemental sulfur and sulfate [32,33]. S<sup>2-</sup> could also be detected in the bioreactor when H<sub>2</sub>S was dissolved in the liquid phase. The concentrations of SO<sub>4</sub><sup>2-</sup> and S<sup>2-</sup> in the integrated-bioreactor were analyzed periodically. *Thiobacillus*, which degrades sulfur-containing compounds, was also observed concurrently.

In the SZ, SO<sub>4</sub><sup>2-</sup> concentrations increased slightly, from 0.15 to 1.00 mg·L<sup>-1</sup>, whereas the S<sup>2-</sup> concentration was stable at 4–5 mg·L<sup>-1</sup> (Table 3). Most sulfur-containing compounds were present in the form of S<sup>2-</sup> in this zone. The *Thiobacillus* species that oxidize H<sub>2</sub>S generally exhibited optimum activities at acidic pH. The pH in the SZ was 6–8 during the operation of the bioreactor (Fig. 4). The acidophilic and non-acidophilic *Thiobacillus* lacked optimum conditions for the growth and metabolism. Hence, most H<sub>2</sub>S dissolved in the liquid phase, and only

**Table 2** Microorganisms in the integrated-bioreactor at steady-state

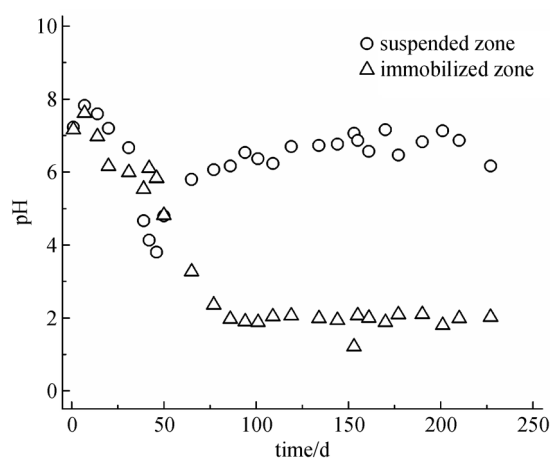
species	the suspended zone	the immobilized zone
acidophilic <i>Thiobacilli</i>	$9.50 \pm 1.01 \times 10^5$ CFU <sup>a</sup> ·mL <sup>-1</sup>	$1.32 \pm 0.32 \times 10^7$ CFU·g <sup>-1</sup>
non-acidophilic <i>Thiobacilli</i>	$9.85 \pm 0.99 \times 10^6$ CFU·mL <sup>-1</sup>	$2.13 \pm 0.37 \times 10^6$ CFU·g <sup>-1</sup>
ammonia oxidizing bacteria	$1.70 \pm 0.26 \times 10^4$ MPN <sup>b</sup> ·mL <sup>-1</sup>	$6.97 \pm 0.95 \times 10^2$ MPN·g <sup>-1</sup>
nitrite oxidizing bacteria	$5.60 \pm 0.87 \times 10^4$ MPN·mL <sup>-1</sup>	$4.59 \pm 0.44 \times 10^3$ MPN·g <sup>-1</sup>

Notes: a) CFU, colony forming units; b) MPN, most probable numbers

**Table 3** Changes of *Thiobacilli* and products in H<sub>2</sub>S removal

time /d	non-acidophilic <i>Thiobacilli</i> /(CFU <sup>a</sup> ·mL <sup>-1</sup> )	acidophilic <i>Thiobacilli</i> /(CFU·mL <sup>-1</sup> )	SO <sub>4</sub> <sup>2-</sup> /(mg·L <sup>-1</sup> )	S <sup>2-</sup> /(mg·L <sup>-1</sup> )
suspended zone				
10	1.9±0.39 × 10 <sup>4</sup>	9.5±1.85 × 10 <sup>3</sup>	0.15±0.02	4.46±0.11
35	1.5±0.36 × 10 <sup>4</sup>	5.44±0.69 × 10 <sup>4</sup>	0.61±0.13	5.01±0.13
105	not detected	not detected	0.82±0.03	4.96±0.20
155	9.85±0.99 × 10 <sup>6</sup>	9.5±1.01 × 10 <sup>5</sup>	1.00±0.18	4.15±0.13
immobilized zone				
10	4.57±0.61 × 10 <sup>5</sup>	1.32±0.14 × 10 <sup>4</sup>	0.18±0.01	3.06±0.12
35	8.6±0.82 × 10 <sup>5</sup>	4.17±0.35 × 10 <sup>6</sup>	1.54±0.10	1.23±0.19
105	not detected	not detected	3.71±0.08	0.24±0.05
155	2.13±0.37 × 10 <sup>6</sup>	1.32±0.32 × 10 <sup>7</sup>	2.55±0.07	0.28±0.03

Notes: a) CFU, colony forming units

**Fig. 4** Changes of pH in the suspended zone and the immobilized zone

a small portion of the H<sub>2</sub>S was bio-oxidized by non-acidophilic *Thiobacillus* in this zone.

In the IZ, the S<sup>2-</sup> concentration decreased with the formation of SO<sub>4</sub><sup>2-</sup>. Most of the sulfur-containing compounds existed in the form of SO<sub>4</sub><sup>2-</sup>. The counts of acidophilic *Thiobacillus* exceeded those of non-acidophilic *Thiobacillus* (Table 3). High removal efficiencies for H<sub>2</sub>S can be obtained in acidic conditions [34]. H<sub>2</sub>S oxidation generated sulfuric acid and reduced pH. The pH in the IZ gradually declined from pH 7 to less than 2.0. Acidic environments favored the growth of acidophilic *Thiobacillus*, which has the highest metabolic capability for biodegrading H<sub>2</sub>S in low pH conditions. As shown in Table 3, (1.32±0.32) × 10<sup>7</sup> CFU·(g dry packing material)<sup>-1</sup> of acidophilic *Thiobacillus* was presented after 155 days, which was considerably greater than the amount of non-acidophilic *Thiobacillus* ((2.13±0.37) × 10<sup>6</sup> CFU·(g dry packing material)<sup>-1</sup>) determined for the same day. Most of the H<sub>2</sub>S was bio-oxidized by acidophilic

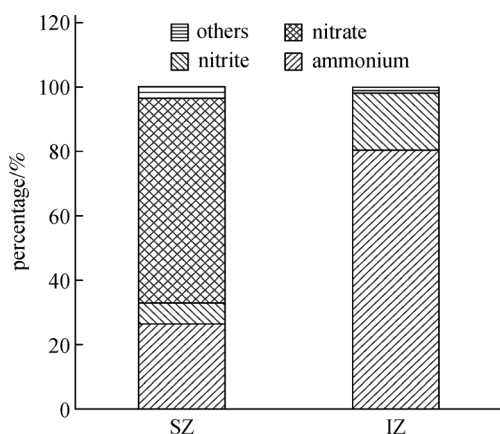
*Thiobacillus* and produced sulfate in the IZ. Similar results were obtained by other studies. Shinabe et al. found a low optimal pH of 2.5 during H<sub>2</sub>S removal by *Thiobacillus thiooxidans*, and did not also detect the inhibition of H<sub>2</sub>S biodegradation at pH 1 in a trickling biofilter [30].

Porous polyurethane foam cubes were packed in the IZ to support the microorganisms and supply them with easy access to contaminants in the airflow. The microorganisms were distributed from the surface to the inside of the packing material. Sulfur filaments can grow either on the surface or within the packing material. Most of the H<sub>2</sub>S was degraded into sulphates in the IZ, which then accumulated on the packing material. SEM images of the distribution of total sulfur on the packing materials were obtained. A microphotograph of the new packing materials lacking sulfur-containing compounds is shown in Fig. S2(a). After 155 days of bioreactor operation, the packing materials were sampled for microscopic observations. The Figs. S2(b) and S2(c) exhibit the distribution of total sulfur from the surface to the core of the polyurethane foam cubes. The surface had an accumulation of 3.01% of sulfur-containing components (Fig. S2(b), Table S1), and the amount of sulfur-containing compounds decreased with foam cube depth. Only a few sulfur containing compounds (1.87%) were found at the center (Fig. S2(c), Table S1). The distributions of microorganisms (Fig. S1) and biodegraded products followed similar patterns.

### 3.3.2 Products of NH<sub>3</sub> degradation

Chemical processes (e.g., chemical neutralization), physicochemical processes (e.g., absorption or adsorption), and biodegradation may have contributed to the removal of NH<sub>3</sub> in the integrated-bioreactors. The products of these processes could be nitrite, nitrate, or NH<sub>4</sub><sup>+</sup>. The operational conditions and microbiological characteristics differed in each zone and resulted in the formation of various products.

The percentages of various nitrogen-containing compounds were calculated for each zone, as shown in Fig. 5. As displayed, 64.07% nitrate and 26.32% NH<sub>4</sub><sup>+</sup> were present in the SZ, indicating that nitrate was the main inorganic nitrogen-containing compound present in this zone. The pH in the SZ was maintained at 6 to 8 (Fig. 4). Most of the ammonia absorbed or adsorbed was ultimately converted into nitrate in the SZ. The production of nitrite or nitrate and the dissolution of H<sub>2</sub>S can lead to pH reduction, while NH<sub>3</sub> dissolved in the liquid phase can neutralize acidic chemicals. The pH in the SZ depended on the amount of nitrite and nitrate produced, as well as the amount of H<sub>2</sub>S and NH<sub>3</sub> dissolved. NH<sub>3</sub> neutralization in a biofilter for odour removal was also reported by Yang et al. [35]. NH<sub>3</sub>-oxidizing bacteria prefer neutral or alkaline environments. The ammonium- and nitrite-oxidizing bacteria detected in this zone were  $(1.70 \pm 0.26) \times 10^4$  MPN·(mL liquid)<sup>-1</sup> and  $(5.60 \pm 0.87) \times 10^4$  MPN·(mL liquid)<sup>-1</sup>, respectively, indicating that most of the NH<sub>3</sub> in the inlet stream was transformed into nitrites and nitrates by ammonium- and nitrite-oxidizing bacteria in the SZ. Only a small fraction of NH<sub>3</sub> transferred to the liquid phase via adsorption, absorption, and chemical neutralization in this zone.



**Fig. 5** Percentage of various nitrogen containing compounds in individual zone

Less nitrite and nitrate were produced in the IZ than in the SZ. The percentage of NH<sub>3</sub> dissolved was as high as 80.33% in the IZ. The pH in the IZ dropped to less than 2.0 because of the accumulation of sulfate, the main product obtained from H<sub>2</sub>S biodegradation. The ammonium- and nitrite-oxidizing bacteria preferred an alkaline environment and did not thrive in an acidic environment. The populations of ammonium- and nitrite-oxidizing bacteria in the IZ were rather small. A substantially larger amount of NH<sub>3</sub> dissolved into the liquid phase and only a small portion of NH<sub>3</sub> was removed by nitrification because of the lack of nitrite-oxidizing bacteria in this zone.

The performance of the bioreactor was affected by the

accumulation of sulfate and nitrate. Excessive sulfate and nitrate exited out from the bioreactor in spray effluent and were treated by discharging into waste water treatment facilities in the same WWTP.

## 4 Conclusions

With the synergistic action of the two zones, the H<sub>2</sub>S and NH<sub>3</sub> generated from the gravity sludge thickener and sludge dewatering facility could be reduced effectively in the assembled integrated-bioreactor. During steady treatment, the outlet concentrations reduced to 0–2.8 mg·m<sup>-3</sup> for H<sub>2</sub>S and 0–0.5 mg·m<sup>-3</sup> for NH<sub>3</sub>. The extents of H<sub>2</sub>S and NH<sub>3</sub> removal differed between the immobilized and suspended zones, and the bioreactor can be optimally designed in accordance with the contaminant characteristics in the odours. Different microbial populations dominated each zone and accordingly generated the distinction in the major biodegradation products between the zones. Most of the H<sub>2</sub>S was bio-oxidized by acidophilic *Thiobacillus* in the immobilized zone, whereas most of the NH<sub>3</sub> was removed by nitrite-oxidizing bacteria in the suspended zone. The integrated-bioreactor contains multiple degraders and thus can remove more than one pollutant from air.

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