

Retrofitting existing chemical scrubbers to biotrickling filters for H₂S emission control

David Gabriel* and Marc A. Deshusses†

Department of Chemical and Environmental Engineering, University of California, Riverside, CA 92521

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Biological treatment is a promising alternative to conventional air-pollution control methods, but thus far biotreatment processes for odor control have always required much larger reactor volumes than chemical scrubbers. We converted an existing full-scale chemical scrubber to a biological trickling filter and showed that effective treatment of hydrogen sulfide (H₂S) in the converted scrubber was possible even at gas contact times as low as 1.6 s. That is 8–20 times shorter than previous biotrickling filtration reports and comparable to usual contact times in chemical scrubbers. Significant removal of reduced sulfur compounds, ammonia, and volatile organic compounds present in traces in the air was also observed. Continuous operation for >8 months showed stable performance and robust behavior for H₂S treatment, with pollutant-removal performance comparable to that achieved by using a chemical scrubber. Our study demonstrates that biotrickling filters can replace chemical scrubbers and be a safer, more economical technique for odor control.

There are ≈16,000 publicly owned treatment works in the United States (1, 2) and probably >50,000 worldwide, and emission of objectionable odors from these facilities is a major problem. Odors emitted by publicly owned treatment works are typically a complex blend of H₂S, reduced sulfur compounds (RSCs), amines, and low molecular weight carboxylic acids (3, 4), all with very low odor thresholds (5, 6). For example, the odor threshold for H₂S is 0.5–2 parts per billion by volume (ppb_v), 1–2 ppb_v for methyl mercaptan, and ≈0.1 ppb_v for dimethyl sulfide. H₂S resulting from the bacterial reduction of sulfate is by far the largest contributor to odor nuisances and is the principal compound targeted for control by wastewater-treatment plants (7, 8). Controlling H₂S is usually achieved in wet or chemical scrubbers in which H₂S is either absorbed in a caustic solution or absorbed and oxidized in a caustic-hypochlorite or caustic-peroxide solution (8). Chemical scrubbing in packed-bed towers is an established technique and is effective at gas contact times as short as 1.3–2 s. However, chemical scrubbing suffers from important drawbacks such as high operating costs, generation of halomethanes that are known air toxics (9), and the requirement for hazardous chemicals, which pose serious health and safety concerns.

In the past decades, biological techniques have been increasingly applied for air-pollution control (10–13). In biological trickling filters or biotrickling filters, the waste air stream is passed through a packed bed on which pollutant-degrading organisms are immobilized as biofilms. As shown schematically in Fig. 1, the contaminant is transferred from the gas to the liquid phase and subsequently to the biofilm, or it is transferred directly from the gas to the biofilm, where it is degraded biologically to harmless compounds. In general, biodegradation of the contaminant is mediated by a mixed culture of bacteria. In the particular case of H₂S control, autotrophic organisms mainly of the *Thiobacillus* genus (14, 15) oxidize sulfide to sulfate while using carbon dioxide as a source of carbon for growth (chemoautotrophic microorganisms). *Thiobacillus* species are slow-growing organisms and are resistant to low pH (14). Because of the importance of finding alternative treatment techniques for H₂S control, numerous studies have reported the removal of H₂S in

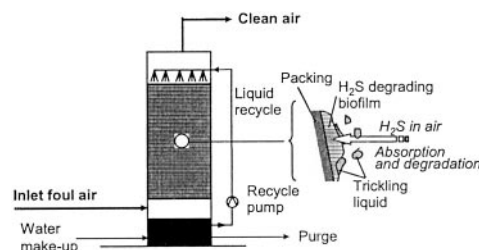


Fig. 1. Sketch of a biological trickling filter for air-pollution control. Gaseous pollutants are absorbed and degraded by immobilized organisms.

vapor-phase bioreactors such as biofilters and biotrickling filters (15–19). However, before this study, successful biotreatment of H₂S required gas contact times ranging from 10 to 30 s (15–19). This is much longer than the contact time in chemical scrubbers; consequently, biotreatment has been impractical for the control of very large air streams.

We report on a recent field project in which we converted a chemical scrubber to a biotrickling filter keeping the original contact time of 1.6–2.3 s. The scrubber vessel was reused, and the conversion procedure was simple and relatively inexpensive. The biotrickling filter successfully treated H₂S at rates comparable to those of chemical scrubbers. It seems that a combination of high mass transfer rate and optimum operating conditions was responsible for the unprecedented performance. The results of the study suggest that a large number of chemical scrubbers now used for H₂S control could be converted to biotrickling filters, with major cost savings and substantial environmental benefits.

Materials and Methods

Field Conditions. The chemical scrubber that was converted is located at the Orange County Sanitation District (OCSD) in California. The OCSD manages two wastewater-treatment plants that treat a total average flow of 910,000 m³ of wastewater per day. Dosage of chemicals in the trunk lines and a total of 37 chemical scrubbers are used for treatment of odor emissions from both facilities with an associated cost of U.S. \$3.5 million per year. The scrubber that was converted treats odorous air from the influent sewer trunk line and from a sewage-pump vent. Trunk-line scrubbers act as first-stage roughing scrubbers to reduce the concentration before further downstream treatment by other scrubbers. A picture of the converted scrubber is provided in Fig. 6, which is published as supporting information on the PNAS web site, www.pnas.org.

Scrubber Characteristics. The converted scrubber is 9.75 m high, 1.82 m in i.d., and made of fiberglass-reinforced plastic with a nominal packed-bed height of 2.8 m and bed volume of 7.3 m³

Abbreviations: RSC, reduced sulfur compound; ppb_v, parts per billion by volume; ppm_v, parts per million by volume; OCSD, Orange County Sanitation District.

*Present address: Escola Tècnica Superior d'Enginyeria, Universitat Autònoma de Barcelona, Bellaterra, 08193 Barcelona, Spain.

†To whom correspondence should be addressed. E-mail: mdeshusses@enr.ucr.edu.

(see Fig. 6). The packing is supported by a multibeam packing support. The trickling liquid-distribution system uses parting boxes and weir troughs and is situated 0.80 m above the top of the bed. The water-trickling flow rate is $4.5 \text{ m}^3 \text{ h}^{-1}$, corresponding to a velocity of 1.7 m h^{-1} . Foul air is fed to the scrubber by a 30-kW fixed-speed centrifugal blower at an average flow rate of $16,300 \text{ m}^3 \text{ h}^{-1}$, resulting in an empty bed gas contact time of 1.6 s. The bottom of the scrubber serves as a liquid sump (1.73-m^3 volume) and includes a 2.13-m-high air plenum for air and make-up water feed. Both pH control and nutrient supply to the process culture is achieved by the continuous supply of plant water ($0.23 \text{ m}^3 \text{ h}^{-1}$), thus alleviating the need for any chemical feed. The purge from the biotrickling filter is continuously directed to the sewer; it is acidic but does not pose any special concern, because it is greatly diluted with the sewage being treated. Plant water samples ($n = 10$) contained $1.5 \pm 0.6 \text{ g of N-NO}_3^- \text{ m}^{-3}$, $16.7 \pm 8.8 \text{ g of N-NH}_4^+ \text{ m}^{-3}$, $7.5 \pm 1.9 \text{ g of PO}_4^{3-} \text{ m}^{-3}$, $58.4 \pm 11.4 \text{ g of C m}^{-3}$ as inorganic carbon, $18.5 \pm 6.0 \text{ g of C m}^{-3}$ as total organic carbon, and $0.8 \pm 0.2 \text{ g of Cl}_2 \text{ m}^{-3}$ as main parameters. Trace ions were found at levels between 50 and 10 mg m^{-3} . No supply of any additional nutrient source was necessary to sustain effective biotrickling-filter operation, which demonstrates that the process culture could thrive under minimum nutrient supply. The scrubber is equipped with continuous monitoring of multiple parameters including pH and H_2S influent and effluent concentrations (see *Analytical Methods*).

Development of the Chemical Scrubber Conversion Procedure. A conversion procedure was developed after initial laboratory research in a pilot-scale biotrickling filter and after considering the usual characteristics of scrubbers used for H_2S control (20). In short, we operated a laboratory pilot unit under conditions similar to those expected at a publicly owned treatment work and tested selected packing materials for their suitability for biotrickling filtration. A random dump packing (3.5" Tri-Packs, Jaeger Products, Houston) commonly used in chemical scrubbers with a surface area of $125 \text{ m}^2 \text{ m}^{-3}$ proved unsuitable, probably because of its low surface area, which resulted in insufficient bacterial attachment. Open-pore polyurethane foam (M+W Zander, Nuremberg, Germany), which had been used as a packing support in several biotrickling-filter applications, exhibited the best performance. The packing has a surface area of $600 \text{ m}^2 \text{ m}^{-3}$ and a density of 35 kg m^{-3} and is cut in cubes of 4 cm by side. The foam cubes are used as random dump packing. The laboratory phase also revealed that a trickling velocity of $\approx 1.7 \text{ m h}^{-1}$, i.e., 20 times lower than the trickling rate of chemical scrubbers, was appropriate for biotrickling-filter application. The lower liquid trickling rate allowed replacement of the original 5.6-kW recycle pump of the chemical scrubber that was converted at the OCSD with a 0.4-kW pump and lower the liquid recycle rate to $4.5 \text{ m}^3 \text{ h}^{-1}$. Thus, the conversion of the scrubber at the OCSD simply consisted of (i) replacing the existing packing, which had a low interfacial area and was not suitable for microorganism attachment; (ii) replacing the liquid recycle pump with a smaller one; (iii) disconnecting the chemical feeds; and (iv) modifying controls of the reactor. The converted scrubber was similar in principle to the flow sheet shown in Fig. 1.

Analytical Methods. H_2S was monitored online by using electrochemical sensors (Vapex Sentinel System, Vapex/Quality Air Products, Ocoee, FL) with independent, interchangeable sensors connected to an inlet and an outlet-sampling port in the scrubber. The H_2S detection limit for the inlet and outlet sensors was 0.1 and 0.07 parts per million by volume (ppm_v), respectively. Online Vapex units were configured to display H_2S inlet and outlet concentrations every 4 s and to store the average of 12-min segments. Inline pH in the biotrickling filter was measured with

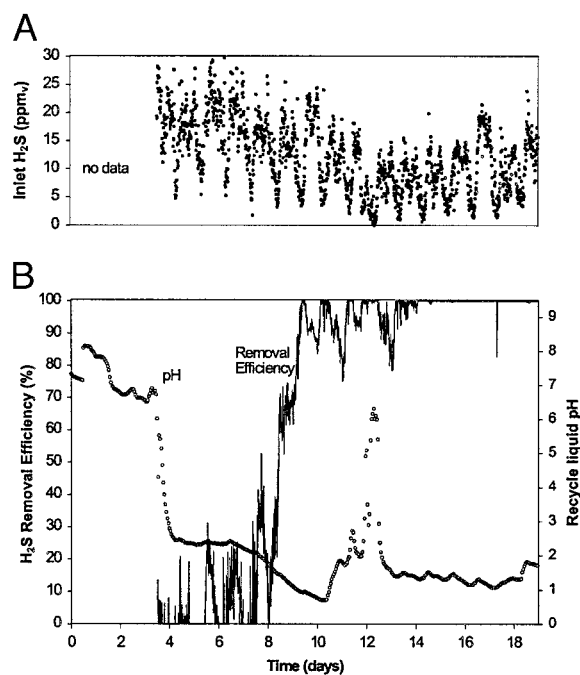


Fig. 2. Startup of the biotrickling filter. (A) Inlet concentration over time. (B) Removal efficiency and liquid recycle pH over time. H_2S measurements were unavailable until day 3 and unreliable until day 6. Between days 10 and 13, the makeup water feed was adjusted, resulting in pH changes.

a pH sensor (Rosemount Analytical, Irvine, CA) installed in the liquid recycle line of the reactor and connected to the supervisory control and data acquisition system of the facility.

H_2S data were supplemented periodically with grab-sample measurements made with a Jerome 631X series meter (Arizona Instruments, Tempe, AZ). Gas samples for offline analysis were collected in 1-liter Tedlar bags (SKC, Eighty Four, PA). RSCs (carbonyl sulfide, sulfur dioxide, methyl mercaptan, ethyl mercaptan, dimethyl sulfide, carbon disulfide, *n*-propylthiol, *iso*-propylthiol, and dimethyl disulfide) were analyzed offline by gas chromatography with a 60-m, 0.32-mm i.d. capillary column and a sulfur chemiluminescence detector (Sievers 350) and photoionization detector. Gaseous ammonia was measured by using a PhD Ultra atmospheric monitor instrument (Bacou USA Safety, Middletown, CT). Carbon dioxide was measured by using a chemical sensor connected to a portable data logger (Vernier Software & Technology, Beaverton, OR). Airflow was measured with a Pitot tube inserted in the inlet duct of the biotrickling filter, and cross-sectional data were corrected by using the log-linear method. Odor was assessed by olfactometry tests with the triangular forced-choice method using a AC'SCENT olfactometer (St. Croix Sensory, Lake Elmo, MN). Standard methods (21) were used for nitrogen, phosphate, organic matter, and chlorine analysis for water-sample characterization. Organic and inorganic carbon in water samples were measured in a TOC-5050 total organic carbon analyzer (Shimadzu). Ions in make-up and trickling water were measured by the inductively coupled plasma technique.

Results and Discussion

Biotrickling-Filter Performance During the Startup. Before startup, the reactor sump was inoculated with 0.8 m^3 of activated sludge from the wastewater-treatment plant. Analysis of the H_2S -removal performance during the startup phase (Fig. 2) revealed that the pH started to decline after 3 days of operation to reach a value of 2 at 7 days after startup. The decline in pH (from the

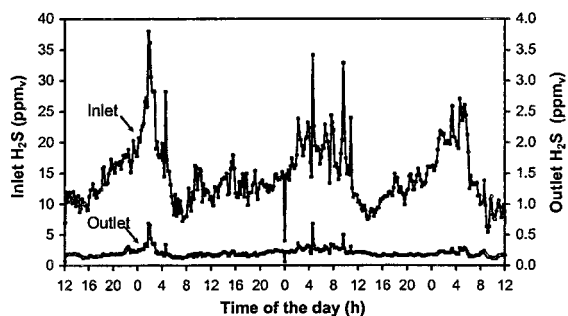


Fig. 3. Representative profiles of inlet (Upper) and outlet (Lower) H₂S concentrations. Each datum point plotted corresponds to a 12-min averaged value of readings at 4-s intervals performed by independent sensors.

production of H⁺ and sulfate from the oxidation of H₂S) correlated with the increase in H₂S-removal efficiency. Sulfate production was in quantitative agreement with H₂S removal. Acclimation lasted ≈10 days, after which H₂S removal was in excess of 99% for H₂S inlet concentrations ranging from 5 to 25 ppm_v and remained high for the remainder of the study.

Although the low pH is not favorable for H₂S absorption, it provides favorable conditions for the sulfide oxidizing process culture (14). In Fig. 3 we show a plot of fluctuations of H₂S concentrations for a 36-h period. Although a highly fluctuating inlet concentration was repeatedly observed each day, the biotrickling filter was robust enough to consistently absorb and treat all concentration peaks. Fluctuations in pH were modest because of the large volume of the liquid sump and the buffer capacity of the secondary effluent used for trickling. Hence no specific adjustment of the make-up water flow was necessary to control the pH between 1.5 and 2.2, thus eliminating the need for detailed pH control. Operation at neutral pH for periods of <24 h did not affect H₂S removal markedly.

Long-Term Performance. The long-term performance of the biotrickling filter is reported in Fig. 4. The elimination capacity (EC) is defined as the amount of pollutant degraded per unit of time, normalized by the volume of packed bed. Plotting EC vs. pollutant loading (Fig. 4) allows for the comparison of bioreactors of different sizes operated under different conditions. In the

present study, the airflow was essentially constant, and the loading changes were a result of the fluctuations in H₂S inlet concentration. At loadings between 15 and 95 g of H₂S m⁻³ h⁻¹, the system consistently degraded >97% of the influent H₂S. Above 95 g of H₂S m⁻³ h⁻¹, breakthrough of H₂S occurred, whereas a quasizero order degradation regime was observed at loadings >105 g of H₂S m⁻³ h⁻¹, with removal efficiencies gradually decreasing to <80%. Removal efficiencies <90% were also found at inlet loadings <5–6 g of H₂S m⁻³ h⁻¹, which corresponded to inlet H₂S concentrations of ≈2 ppm_v. At such low concentrations, the H₂S outlet data were scattered due to limitations of the analytical equipment, resulting in some uncertainties on the actual H₂S removal. Still, the reduction in the H₂S removal at low loadings was significant. Possible reasons include diffusional and biological kinetic limitations that are typical for the biotreatment of low contaminant concentrations (10). Even so, the biotrickling-filter effluent air was always in compliance with local regulations (a maximum limit of 1 ppm_v as average outlet concentration over 24 consecutive hours for H₂S discharge). Typically, removal efficiencies of 98% were commonly reached for inlet H₂S concentrations as high as 30 ppm_v when the system was operated at gas contact times as low as 1.6–2.3 s. Such performance is exceptionally high compared with other biofilters or biotrickling filters removing a low concentration of H₂S even at higher gas contact times (17, 19, 22). Probability plots of inlet and outlet H₂S concentrations (Fig. 5) indicate that although the inlet concentration was highly fluctuating, the outlet was maintained well below the discharge limit level a high percentage of the time that the reactor was operated. The few outlet H₂S data >1 ppm_v corresponded to transient periods of rapidly increasing H₂S inlet concentrations. The concentration distribution clearly depended on the ambient temperature, because temperature affected H₂S generation in the trunk lines. In Fig. 5B we show that during the cold season, the reactor operated at lower H₂S inlet concentrations, which resulted in lower discharge concentrations a higher fraction of the time. In both cases, however, the results shown in Fig. 5 demonstrate the robustness of the system over a wide range of operating conditions.

Simultaneous Removal of Other Air Pollutants. We also investigated the simultaneous removal of trace RSCs and the reduction of odor (Table 1). Three of nine RSCs analyzed were routinely

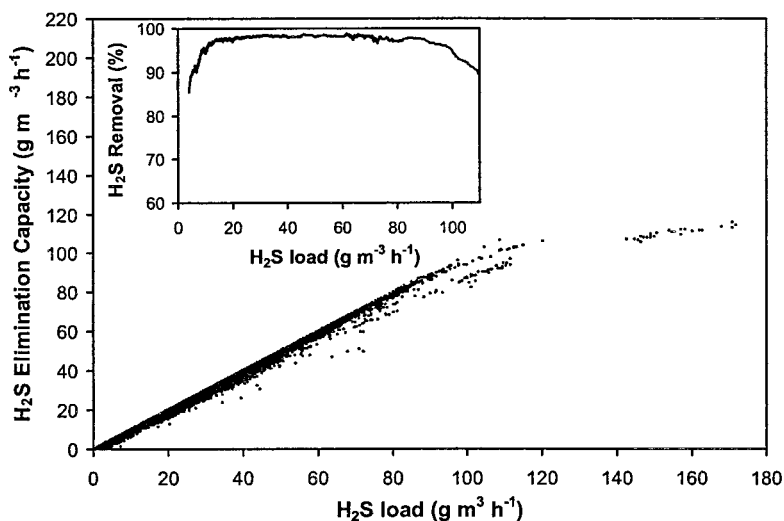


Fig. 4. Elimination capacity of H₂S versus H₂S loading in the biotrickling filter. (Inset) Removal efficiency as a function of the loading, with the removal plotted as moving average to remove scattering. Data include >8 months of continuous operation. Loadings >110 g of H₂S m⁻³ h⁻¹ were achieved by spiking the inlet air with H₂S from a compressed gas cylinder.

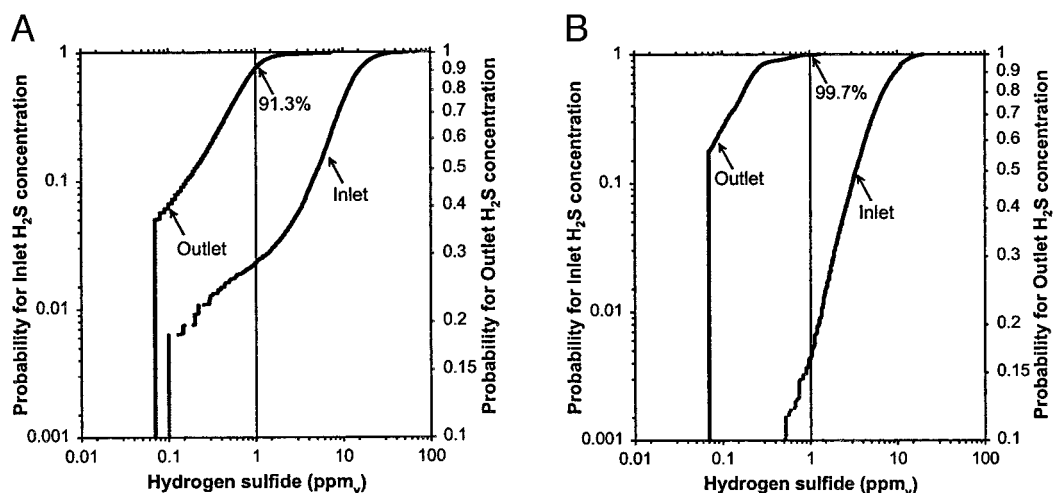


Fig. 5. Normal probability plot of inlet and outlet H_2S concentrations during routine operation of the reactor for all data acquired between August 8 and November 30, 2001 (A), and December 1, 2001, and March 31, 2002 (B). The vertical bar at 1 ppm_v indicates the average discharge limit not to be exceeded for >24 consecutive hours. Note the different scales used for inlet and outlet probabilities.

detected in the inlet air of the biotrickling filter. Similar to other studies (3), we observed a high spread of RSC inlet concentrations, which led to a high variability in RSC removal. Noticeable removal of RSCs started only ≈ 1 month after the system startup, indicating a slower growth and acclimation of the autotrophic/heterotrophic mixed population degrading RSCs (23). This was most probably caused by the low concentrations of pollutant substrate and both kinetically and mass transfer-limiting conditions. Odor reduction evaluated by odor panels was mostly because of H_2S removal, although residual odor was not negligible. Under these circumstances, the posttreatment downstream of the biotrickling filter was maintained. Improvement in terms of odor reduction needs to deal with defining better conditions for RSCs and other potential remaining odorous compound degradation in the system.

Factors Influencing Performance. Overall, the results of the present study show performances well exceeding previous reports. Most earlier studies require 15–40 s of gas contact time for the efficient removal of 20–100 ppm_v of H_2S , resulting in elimination capacities ranging from 10 to 40 g of $\text{m}^{-3} \text{h}^{-1}$ (17–19, 22). A few studies show higher performance. Among them, Yang and Allen (16) operated a biofilter in which a maximum elimination capacity (EC_{max}) of 140 g of $\text{H}_2\text{S} \text{m}^{-3} \text{h}^{-1}$ was reached; however, the inlet concentration of H_2S was extremely high (up to 2,000 ppm_v) so that the high performance was clearly linked to the high concentration. Koe and Yang (19) treated lower concentrations of H_2S (20–100 ppm_v) in a biotrickling filter operated at a gas residence time of 5–15 s. Significant breakthrough of H_2S was

observed at 5 and 7 s of contact time for loadings of 40–50 g of $\text{H}_2\text{S} \text{m}^{-3} \text{h}^{-1}$.

We determined that the high performance observed at the OCSD is the result of a combination of factors. First, the packing material, which is known to play a key role (24), provided a large surface area for high bacterial attachment and pollutant mass transfer. With respect to pollutant mass transfer, the air linear velocity in our system was ≈ 20 times higher than previously reported cases. A higher air velocity implies a more turbulent regime and higher gas–film mass-transfer coefficients. This was proven to have a major impact on H_2S treatment by performing independent experiments with packing material removed from the OCSD biotrickling filter (25). In short, the volumetric H_2S elimination capacity of single cubes of packing was determined at various air velocities in a lab-scale differential biotrickling filter. The elimination capacity of H_2S was found to increase as much as 5- to 10-fold when increasing the air velocity from $<200 \text{ m h}^{-1}$, i.e., the usual values in biofilters, to $6,300 \text{ m h}^{-1}$, i.e., the air velocity imposed on the OCSD biotrickling filter (25). Another factor contributing to the outstanding performance of the field biotrickling filter is the ample supply of micronutrients (via the secondary effluent feed) and CO_2 ($>4,000 \text{ ppm}_v$ in the foul air) to the process culture such that they were never limiting the degradation of H_2S . Finally, both liquid and air were distributed homogeneously across the section of the scrubber with no evident gas short-circuiting or extensive dry or dead zones in the packed bed. Therefore, optimum use of the biologically active section was achieved.

Impact

The demonstration that H_2S can be effectively treated biologically at contact times comparable to those of chemical scrubbers has substantial implications for odor control. An overall cost–benefit analysis of the scrubber that was converted at the OCSD shows that total annual savings in operating costs (essentially chemicals and electricity) are \approx U.S. \$30,000 per year for the biotrickling filter compared with chemical scrubbing. The estimated commercial cost of converting the chemical scrubber to a biotrickling filter was \approx U.S. \$40,000–60,000, which compares well with the annual savings in operating costs. It is difficult to generalize to other scrubbers, because the costs of conversion and the operating-cost savings depend on each application (size, concentration of H_2S , chemical feed, etc.). Still, we estimate that an average converted chemical

Table 1. Inlet concentrations and removal efficiencies of odor and RSCs

Compound	Inlet concentration, ppb _v	Removal efficiency, %
Carbonyl sulfide	67.2 \pm 7.7	44 \pm 11
Methyl mercaptan	192.5 \pm 34.1	67 \pm 11
Carbon disulfide	70.3 \pm 20.5	35 \pm 5
Odor	1,980 \pm 480 D/T	65 \pm 21

Data are mean \pm SD; $n = 21$ for RSCs and 9 for odor panels. Conditions were representative of the biotrickling filter operated at gas contact times between 1.6 and 3.4 s. D/T, dilution-to-threshold ratio as measured by standard odor panel.

scrubber in the United States would save between U.S. \$10,000 and \$50,000 worth of chemicals and electricity per year. Note that environmental health and safety benefits are not factored in the above savings estimates, but they may be of the same order of magnitude. An estimated 10,000–40,000 scrubbers for odor control operate at publicly owned treatment works in the U.S. and probably >100,000 scrubbers worldwide. Many of those scrubbers treat H₂S only and, hence, based on the results of this study, could potentially be converted to biotrickling

filters. If one assumes that 25–40% of the chemical scrubbers worldwide could be converted to biotrickling filters, this would represent a total market of U.S. \$1–3 billion and would result in net energy and chemical savings of U.S. \$0.25–2 billion per year.

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