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# Impact of pH and loading rate on hydrogen sulphide removal efficiency in a biotrickling filter

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**Abstract:** Hydrogen sulphide (H<sub>2</sub>S) removal is a critical aspect of waste gas treatment, particularly in industries, municipalities, and agriculture. This study investigates the impact of pH and loading rate on the efficiency of hydrogen sulphide removal in biotrickling filters. Biological methods, acknowledged as Best Available Techniques (BAT), are gaining prominence due to their advantages over classical physicochemical methods. The research aims to elucidate the influence of pH levels and loading rates on the performance of biotrickling filters in mitigating hydrogen sulphide emissions. The tests were conducted at constant pH values of 1, 2, 3 and 4, which were automatically maintained using a pump dispensing NaOH solution. The H<sub>2</sub>S concentrations at the inlet to the column were selected from a range of 60 – 300 ppm. During the study, the proportions of various groups of microorganisms (mesophilic bacteria for environmental application, potentially pathogenic mesophilic bacteria and microscopic fungi), the loading rate (LR), elimination capacity (EC), and removal efficiency (RE) were determined at pilot-scale level, with gas volume flow rates of 40 L·min<sup>-1</sup> During the series of measurements, a maximum elimination capacity of the biotrickling filter of 224 g H<sub>2</sub>S/(m<sup>3</sup>·h), with nearly 100 % H<sub>2</sub>S removal, was achieved.

# Introduction

Hydrogen sulphide is one of the main odorous air pollutants responsible for the odor impact of municipal facilities and industrial activities. In addition to its unpleasant smell, it poses serious risks to human and animal health (Heaney et al. 2011, Rubright et al. 2017). Among sulphur compounds, H<sub>2</sub>S has been identified as the main contributor to odors in landfills (Ko et al. 2015). Hydrogen sulphide is a colorless, heavierthan-air, flammable gas with a characteristic rotten eggs odor. Moderately soluble in water and other solvents (Rubright et al. 2017), H<sub>2</sub>S has a vapor pressure of 1.83 MPa at 20°C. Due to its high Henry's law constant, hydrogen sulphide readily partitions into the gas phase under equilibrium conditions. However, in aqueous environments, a large fraction of H2S dissociates into HS- ions, depending on the pH, which limits the amount of molecular H2S available for volatilization (Abdollahi and Hosseini 2014). The olfactory detection threshold of hydrogen sulphide is low, ranging from 0.00041 to 0.3 ppm (Dobslaw 2023), which can lead to odor nuisance issues in the vicinity of its emission sources (Ko et al. 2015).

Major natural sources of hydrogen sulphide in the environment include the reduction processes of sulphates occurring during the fermentation of organic material due to the

activity of anaerobic bacteria. Additionally, certain plants may utilize and emit H<sub>2</sub>S during vegetative processes. Significant environmental sources of the gas include locations where organic matter decomposition occurs under anaerobic conditions, such as swamps, hydrocarbon deposits, volcanoes, submarine vents, sulphur springs, and stagnant water bodies (Rubright et al. 2017). Among the most significant anthropogenic sources of hydrogen sulphide are sewage systems and livestock housing associated with animal husbandry. Additionally, H2S can be emitted in industrial processes, including petroleum and natural gas processing, paper production, oil refining, coal gasification, petrochemical production, and food processing (Zhang et al. 2008). Wastewater treatment plants and waste management facilities also contribute significantly to hydrogen sulphide emissions, further exacerbating odor-related issues in urban and industrial areas (Pawnuk et al. 2022, Wiśniewska et al. 2020).

In addition to causing foul odors, hydrogen sulphide has adverse effects on human health. It is recognized as one of the most common hazardous substances associated with acute fatal poisonings in workplace environments (Rubright et al. 2017). Furthermore, it should be noted that hydrogen sulphide readily undergoes oxidation, forming many highly toxic byproducts, such as carbonyl sulphide, sulphur dioxide, sulfuric

acid, sulphurous acid, carbon disulphide, and solid particulate matter (Gupta et al. 2016).

#### The impact of hydrogen sulphide on health

Hydrogen sulphide poses the greatest risk of human exposure in enclosed or poorly ventilated spaces, particularly at ground level or below. Excessive exposure to H<sub>2</sub>S can cause both chronic and acute health effects, such as coma, eye irritation, and respiratory issues (Lambert et al. 2006). The respiratory and nervous systems are particularly susceptible. The health risk depends on the duration, frequency, and concentration of H<sub>2</sub>S exposure (Ko et al. 2015). The effects of hydrogen sulphide exposure depending on its concentration are summarized in Table 1. It is worth noting that hydrogen sulphide is easily detectable at low concentrations by the human nose. However, at higher concentrations (above 100 ppm), it rapidly paralyzes the sense of smell. As a result, the odor ceases to be recognized as a warning signal and begins to affect the entire body. Exposure to high concentrations can cause depression of the central nervous system and loss of consciousness. At concentrations above 500 ppm, H<sub>2</sub>S can lead to convulsions, respiratory arrest, coma, and even death (Ko et al. 2015).

In addition to its olfactory impact and health effects, hydrogen sulphide also has corrosive properties in the presence of humidity, which can damage equipment. Moreover, it can contaminate groundwater and contribute to the formation of acid rain (Zhang et al. 2008).

#### Hydrogen sulphide removal

In light of the above, the implementation of effective methods to limit hydrogen sulphide emissions is necessary. Numerous gas purification methods are available, which can be categorized as physical, chemical, or biological. The selection of an appropriate technique depends on factors such as the properties of the gases to be purified, the concentration of emitted pollutants, the type of emission source, and the required purification efficiency.

Currently, biological methods, considered Best Available Techniques (BAT), are gaining significant popularity in hydrogen sulphide removal, particularly in waste management and intensive poultry and swine farming. In these applications, the air stream often contains high levels of ammonia, which can influence pH conditions inside biotrickling filters. Notably, in a counter-current configuration, ammonia readily dissolves and reacts with acidic components, such as sulphuric acid, in the lower sections of the BTF. This can lead to pH stratification that is not reflected in outlet liquid measurements. This phenomenon is particularly important for process control and should be considered when interpreting operational data and designing control strategies.

These biological methods utilize microorganisms to oxidize both volatile organic compounds (including odorous substances) and inorganic pollutants, primarily hydrogen sulphide and ammonia (Dobrzyniewski et al. 2023). They overcome many of the drawbacks associated with conventional physicochemical methods and offer a wide range of solutions for reducing hydrogen sulphide emissions in the air. Among biological gas purification methods, bioscrubbers, biofilters, and biotrickling filters (BTF) are the most prominent. To a lesser extent, membrane bioreactors and air reactors are also utilized (Wu et al. 2020). Recently, combinations of bioreactors with advanced oxidation processes (AOPs) have become more relevant (Dobslaw and Ortlinghaus 2020).

The operation of a biofilter involves the slow passage of gas through a moist, porous stationary bed. Contaminants are first absorbed in the water that moistens the bed and subsequently into the biofilm, where biodegradation of the absorbed substance occurs. Polluted gas is typically introduced at the bottom of the bed, via a system of perforated pipes as one possible method. The bed is irrigated in a counter-current manner, and nutrients (including biogenic compounds) are supplied with the process liquid to nourish microorganisms.

Biofiltration is a natural process that, due to the selfregeneration of the filtration bed and the possibility of recycling effluents, is practically waste-free. Although all three systems utilize a packed column irrigated with liquid, they differ in operation and microbial configuration. In biofilters, irrigation is intermittent to maintain adequate bed

Tab. 1. Health effects of h	ydrogen sulphide [ATSDR 2008.	Rubright et al. 2017:	: Habeeb et al. 2018]

H <sub>2</sub> S concentrations, ppm	Expected Effects/Symptoms
0.00011 - 0.00033	typical background concentrations
0.0005	lowest concentration detectable by human olfactory senses
0.00041 – 0.3 (1.5)	odor threshold - "rotten eggs" smell
1 – 20	offensive odor, possible nausea, tearing of the nose
20 – 50	nose, throat, and lung irritation and loss of appetite
50 – 100	slight conjunctivitis and respiratory tract irritation after 1-h exposure; ay cause digestive upset and loss of appetite
100 – 200	severe nose, throat, and lung irritation; loss of smelling ability
250 – 500	severe lung irritation, headache, dizziness, staggering, unconsciousness, loss of memory, and death
>500	rapid collapse and death



moisture, and biodegradation takes place in the fixed biofilm formed on the packing material. In BTFs, continuous irrigation with recirculating liquid supports biodegradation within the attached biofilm. Bioscrubbers also use a continuously irrigated packed column; however, here the microorganisms are suspended in the liquid, and the biological degradation of absorbed pollutants occurs in a separate bioreactor connected to the liquid loop.

The central part of the system is a packed bed filled with inert material, typically made of plastics or ceramics, placed on a grid in the form of structured packs or random shapes. The filling elements are covered with a biofilm, where the biodegradation of pollutants occurs. Pollutants may enter the biofilm directly (on unwetted surfaces) or be absorbed by the draining liquid before diffusing into the biofilm (Cox and Deshusses 2001). The bed may be wetted either continuously or intermittently with water or wastewater; in the latter case, inoculation of microflora is unnecessary. Gas purification can be carried out as a standalone process or integrated with wastewater treatment.

A key challenge in BTF operation is excessive biomass overgrowth and uneven gas flow, which can be mitigated by implementing anti-clogging technologies (Dobslaw et al. 2018). BTF can be successfully used for  $\rm H_2S$  removal under both aerobic (Melse et al. 2012, Rodriguez et al. 2014) and anaerobic conditions (Schmidt and Anderson 2017), across a wide range of hydrogen sulphide concentrations at the inlet to the device  $\rm C_{in} = 1,000-10,000~ppm_{v}$ , with efficiencies reaching 99.9% (Quijano et al. 2018). Although such high inlet concentrations are less common in full-scale municipal applications, they have been successfully tested under controlled experimental conditions using selected microbial populations and optimized contact times (Montebello et al. 2014, López et al. 2016, Fortuny et al. 2008).

Determining the operational parameters affecting the efficiency of hydrogen sulphide removal in a trickling biofilter is of key importance when choosing an economically and technologically optimal purification method. The filter bed plays a central role in the gas purification process using biofiltration. Its structure should support the deposition of microorganisms and the formation of biofilm, while minimizing the risk of biomass accumulation and air channel formation, reducing the effectiveness of cleaning. The basic factors influencing bioavailability, defined as the maximum amount of a compound accessible to living organisms at a specific time, include the following (Smreczak et al. 2013): the properties of the pollutants (e.g., molecular structure, water solubility, volatility), the parameters related to the organisms responsible for degradation, the presence of other contaminants, contact time, and environmental parameters (pH, porosity, temperature, and humidity).

The optimal pH for most bacteria is neutral, with their typical activity observed within the pH range of 6.0 to 8.5. At extreme pH values, such as those below 4 or above 11, metabolic processes are severely inhibited and may even lead to bacterial death. In the context of hydrogen sulphide removal, removal efficiency of at least 99% are commonly achieved across a wide range of inlet contaminant concentrations. However, due to the risk of biofilter bed acidification, researchers pay close attention to the necessity of adjusting its pH (McNevin and Barford 2000).

This paper presents the results of a study of a pilot-scale biotrickling filter (BTF) installation conducted under high hydrogen sulphide concentrations and pH conditions ranging from 1 to 4. The test results confirm the effectiveness of the installation and highlight its potential for practical application. A key contribution of this study is the experimental evaluation of BTF performance under strongly acidic conditions (pH 1-4), whereas most existing studies focus on systems operating under neutral or mildly acidic environments. Our results demonstrate that H<sub>2</sub>S removal efficiencies of 98.3 - 100% are achievable even at very low pH levels, opening new possibilities for BTF applications in naturally acidified environments such as anaerobic digesters or wastewater treatment facilities. Notably, in contrast to standard industrial practice where pH control is achieved by increasing irrigation rates – often masking the true impact of acidity due to enhanced mass transfer resistance – our approach allowed for decoupling the influence of pH from irrigation rate. By maintaining a constant irrigation rate and adjusting only the pH, we were able to isolate the effect of acidity on both process performance and microbial behavior. A rarely addressed but valuable aspect of this work is also the microbiological analysis of community structure under different pH levels and at various heights of the column.

#### Materials and methods

#### Experimental set-up

This study focused on the Tholander biotrickling filter research installation at the Department of Environmental Protection Engineering, Wroclaw University of Science and Technology (Romanik et al. 2023). The experimental setup consisted of a vertical biotrickling filter column (see Fig. 1), a trickling liquid reservoir located directly beneath the column, and a separate control panel for operating and monitoring key system parameters. Air was supplied by an oil-free compressor, while hydrogen sulphide was sourced from a gas cylinder (with an H<sub>2</sub>S concentration of 50%v in nitrogen). These gases were blended in a mixer and introduced counter-currently to the liquid flow within the biotrickling filter column. Purified gases exited the system through a mechanical ventilation system. The column was packed with polyurethane foam elements inoculated with microorganisms sourced from activated sludge. Once a week, the filter bed was fertilized with 10 ml of a mineral medium (Substral) with the following composition: NPK (Nitrogen–Phosphorus–Potassium) 7 – 3 – 5% (w/w), total nitrogen (N) 7% (including 1.7% nitrate, 1.8% ammonium, and 3.5% amide), phosphorus pentoxide (P<sub>2</sub>O<sub>5</sub>) 3.0% soluble in water, potassium oxide (K<sub>2</sub>O) 5% soluble in water, copper (Cu) 0.002%, iron (Fe) 0.03%, manganese (Mn) 0.01%, molybdenum (Mo) 0.001%, zinc (Zn) 0.02%.

The complete installation, as shown in Figure 1, had a diameter 0.2 m and a total height of 0.94 m. It was designed for a gas volumetric flow rate of 40 L min<sup>-1</sup>. Polyurethane (PUF) foam cubes with an edge length of 15 mm were used as active packing material.

# Operating procedure

The tests were conducted at constant pH values of 1, 2, 3, and 4, which were automatically controlled using a pump dispensing NaOH solution. In each measurement series, the hydrogen

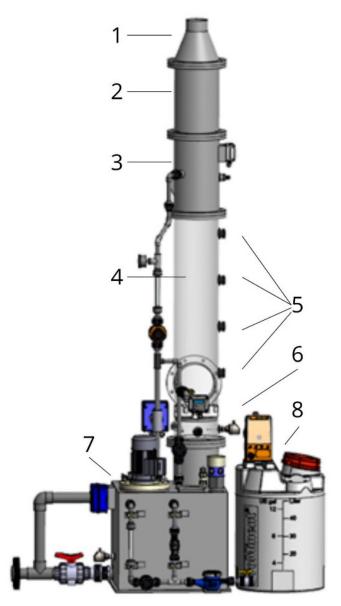


Fig. 1. Vertical biotrickling filter column (Tholander) 1 – gas outlet, 2 – activated carbon filter, 3 – spray nozzle, 4 - biological bed, 5 - sampling ports, 6 - gas inlet, 7 – liquid tank with process monitoring points, 8 - automatic pH control system

sulphide concentration at the column inlet was varied within the range of 60 - 300 ppm. Throughout the study, the proportions of various groups of microorganisms (mesophilic bacteria for environmental application, potentially pathogenic mesophilic bacteria and microscopic fungi), as well as the loading rate (LR), elimination capacity (EC), and removal efficiency (RE), were determined at gas volume flow rate of 40 L·min<sup>-1</sup>. Process parameters such as column pressure, L/G ratio (liquid-to-gas ratio), liquid volume (V<sub>lio</sub>), gas and liquid temperature (T<sub>o</sub>, T<sub>lia</sub>), conductivity (Con.), and oxidoreduction potential (ORP) of the spray liquid were also continuously monitored.

#### Operating characteristics of a biotrickling filter

The performance of the BTF was reported in terms of H<sub>2</sub>S removal efficiency (RE), and elimination capacity (EC) as a function of the loading rate (LR). The basic parameters characterizing the performance of a biotrickling filter include:

• Removal Efficiency (RE) (López et al. 2013) – the percentage of pollutant removed from the gas stream

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \cdot 100 \%$$

 $C_{in}$ - concentration of the pollutant at the inlet of the BTF (g/m<sup>3</sup>)  $\mathrm{C}_{\mathrm{out}}$ - concentration of the pollutant at the outlet of the BTF (g/

• Elimination Capacity (EC) (López et al. 2013) – pollutant degraded per unit of time and volume of the filter bed

$$EC = \frac{Q_g \cdot (C_{in} - C_{out})}{V} g/(m^3 \cdot h)$$

 $C_{in}$  - concentration of the pollutant at the inlet of the BTF (g/m<sup>3</sup>) Courter concentration of the pollutant at the outlet of the BTF

Q<sub>g</sub>- the flow rate of the contaminated stream (m<sup>3</sup>/h) V- the volume of the filter bed (m<sup>3</sup>)

• Loading Rate (LR) (Runye et al. 2015) – the mass of pollutant introduced to the BTF per unit of volume and time  $LR = \frac{Q_g \cdot C_{in}}{v} \ g/(m^3 \cdot h)$ 

$$LR = \frac{Q_g \cdot C_{in}}{V} g/(m^3 \cdot h)$$

 $Q_{\text{g}}$  - the flow rate of the contaminated gas stream (m³/h)  $C_{\text{in}}$  - concentration of the pollutant at the inlet of the BTF (g/m³) V- the volume of the bed (m<sup>3</sup>)

• Empty Bed Residence Time (EBRT) (Iranpour at al. 2005, Runye et al. 2015) – the time requested by the gas stream to pass through the empty volume of the filter bed.

$$EBRT = \frac{V}{Q_g} \text{ (h)}$$

V- the volume of the filter bed (m<sup>3</sup>)

 $Q_g$ - the flow rate of the contaminated stream (m<sup>3</sup>/h)

#### Analyses and sensors

The hydrogen sulphide concentration was measured using a portable Nanosens DP-28 analyzer (Poland), with a measurement range for hydrogen sulphide of 0-5000 ppm. Samples were collected from six locations: the inlet and outlet of the installation, as well as from four measuring ports positioned at specific heights within the bed. Each sample was collected over a period of 90 seconds.

Process parameters, including conductivity, oxidationreduction potential (ORP), inlet gas temperature, spraying liquid temperature, and installation pressure, were measured automatically. Pressure at the inlet and outlet of the installation was measured using a CP110 pressure transmitter. Dosing of aqueous 0.1 mol NaOH solution was performed remotely using a ProMinent gamma/X pump, controlled by a ProMinent DULCOMETER Compact unit that also monitored pH levels. Another ProMinent DULCOMETER Compact controller was employed for Redox/ORP reading. JUMO tecLine HD electrodes were utilized for pH and Redox/ORP measurements and were placed in the tank. Conductivity and temperature were measured using a JUMO CTI-500 inductive transmitter.

All results were collected and managed using the control panel situated in the laboratory and via a dedicated application.



#### Microorganisms and culture conditions

To determine the total number of microorganisms, microbiological tests were conducted using fresh activated sludge obtained from the thickened activated sludge chamber at the sewage treatment plant (serving as a reference value), as well as samples taken after the completion of each test cycle at specific hydrogen sulphide concentration levels at the column inlet. For each concentration level (K1 – K4), one PU foam cube was extracted and placed into 50 ml of 0.98% physiological saline (NaCl CzDA, Chempur). The samples were subjected to ultrasound treatment at a frequency of 45 kHz for 60 seconds using an Ultrasonic Cleaner USC-T (VWR International Sp. z o.o.).

Three dilutions were prepared from the sample as described above. These dilutions were then incubated using the surface method on Sabouraud Dextrose 2% Agar (P-0198) for fungi, and the deep method on Nutrient Agar (P-0049) for bacteria, both supplied by BTL sp. z o.o. Covered plastic Petri dishes were utilized for this purpose. Incubation was performed in a POL-EKO CLW incubator (Poland) under specific conditions: 37°C for mesophilic bacteria, 22°C for potentially pathogenic mesophilic bacteria, and 26°C for microscopic fungi. The results from the three dilutions were counted using

an automatic colony counter (PCC-04, ALCHEM, Poland), with a measurement range of 0-99999. The counts were then averaged for each sample, and the results were expressed as the number of colony-forming units (CFU) per bed element.

# Results and discussion

#### Operation of the biotrickling filter

Table 2 depicts the process parameters recorded during a series of tests conducted at constant pH values. The controlled input parameters included pH, inlet concentration of H<sub>2</sub>S, and consequently, the loading rate. The results are presented in the form of elimination capacity and removal efficiency values. It should be noted that monitored parameters such as gas and liquid temperatures, conductivity, and oxidoreduction potential, varied over time and were primarily dependent on the conditions within the spraying liquid tank.

The comparison of the effect of bed load on the rate and efficiency of hydrogen sulphide removal across all measurement series is illustrated in Figures 2 and 3. A parametric ANOVA of main effects, without interactions, was conducted on the obtained experimental data. The independent variables

Tab. 2. Process parameters of the conducted research series

Parameters	Parameters range				
рН	0.99-1.6	1.99-2.10	2.99-3.20	3.97-4.31	
Inlet concentration of H <sub>2</sub> S, mg/m <sup>3</sup>	68-504	69-502	64-500	75-457	
	K1: 26.1-192.85	K1: 26.64-191.78	K1: 24.51-191.25	K1: 20.78-174.7	
	K2: 12.58-92.94	K2: 12.84-92.43	K2: 11.81 – 92.17	K2: 11.04 – 84.21	
Loading Rate (LR), g/(m³·h)	K3:8.28-61.22	K3:8.46- 60.88	K3: 7.78 – 60.71	K3: -8.46- 55.47	
	K4: 6.18- 45.64	K4: 6.30-45.39	K4: 5.80 – 45.27	K4: 6.8141.36	
	Total: 5.55-41.03	Total: 5.67-40.82	Total: 5.21 – 40.69	Total: 6.12- 37.18	
	K1: 26.1-192.85	K1: 26.64-189.65	K1: 18.1-189.12	K1: 11.72- 174.74	
	K2: 12.58-92.94	K2: 12.84-92.43	K2: 9.76 – 91.91	K2: 11.04 – 84.21	
Elimination Capacity (EC), g/(m³·h)	K3:8.28-61.22	K3: 8.46-60.88	K3: 5.75 – 60.71	K3: 8.46 - 55.47	
	K4:6.18-45.64	K4: 6.30-45.39	K4: 5.80 – 45.27	K4:6.81 – 41.36	
	Total: 5.55-41.03	Total: 5.67-40.82	Total: 5.21 - 40.69	Total: 6.12 – 37.18	
	K1: 97.65-100	K1: 96.47-100	K1: 26.72 - 100	K1: 40.74-100	
	K2: 99.9-100	K2: 97.82-100	K2: 33.59 – 100	K2: 76.19 - 100	
Removal Efficiency (RE), %	K3: 99.9-100	K3: 98.81-100	K3: 53.44 – 100	K3: 95.24- 100	
	K4: 99.9-100	K4: 98.95-100	K4: 71.27 – 100	K4: 99.1- 100	
	Total: 99.9-100	Total: 99.99-100	Total: 75.14 - 100	Total: 96.86- 100	
Liquid volume (V <sub>iiq</sub> ), %	77.1-96.1	77.2-83.1	40-82	77.7-82.2	
Gas temperature (T <sub>g</sub> ), °C	18.2-25.9	18,7-23.3	21.4-27.3	19.7-32.4	
Liquid temperature (T <sub>iiq</sub> ), °C	27.7-31.9	24.5- 30.4	25.6-32.1	17.0 – 34.8	
Conductivity (Con), mS/cm	1.1-126	47.5-77.9	1.8-39.5	0.9-38.9	
Oxidoreduction potential (ORP), mV	216-599	336-463	110-435	119-297	

pH2

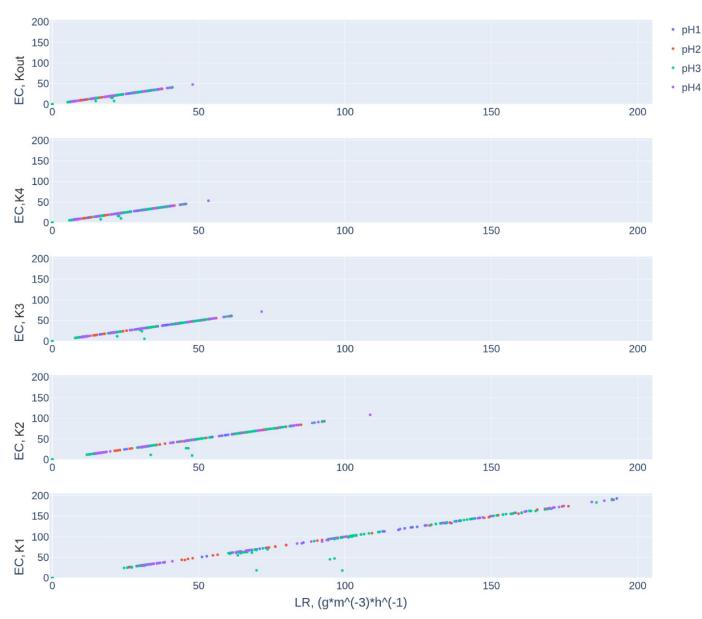


Fig. 2. Effects of H<sub>2</sub>S loading rate on elimination capacity

included pH and the concentration of hydrogen sulphide at the biofilter inlet, while the dependent factor was the biofiltration rate. Based on the research conducted, it was determined that pH did not significantly impact the biodegradation rate, whereas the concentration of contaminant at the inlet exhibited a noticeable influence on the elimination capacity.

The biodegradation of contaminants in biofilm-based systems is often described by the Michaelis-Menten equation:

$$k = k_{max} \cdot \frac{C}{K_M + C}$$

where:

k – reaction rate (g/  $m^3$ ·s)

 $k_{max}^{}$  – maximum reaction rate (g/m<sup>3</sup>·s)

C – substrate concentration (g/m³)

K<sub>M</sub> – Michaelis-Menten constant (g/m<sup>3</sup>)

This model assumes that the reaction rate increases with substrate concentration but gradually approaches a maximum as enzyme saturation occurs. The high values of removal efficiency observed in the studied range (60 – 300 ppm) suggest that the system operated under highly efficient conditions, but without directly indicating the underlying kinetic regime. The nearlinear relationship between loading rate (LR) and elimination capacity (EC) suggests efficient pollutant removal. However, this trend may result from a combination of factors, including biofilm activity, local substrate availability, and mass transfer characteristics, rather than reflecting a specific reaction order. While the Michaelis-Menten model offers a useful starting point for describing biofilm kinetics under moderate substrate concentrations, it fails to adequately capture conditions where inhibition or diffusion limitations prevail.

According to Beigi (2019), the assumptions of the Michaelis-Menten model break down especially at low concentrations (where first-order kinetics dominates), or in systems with thick biofilms, where diffusion limitations reduce local substrate concentrations in the biofilm well below those in the gas phase. Supporting this, Dumont (2017) demonstrated that biofiltration systems often exhibit multiple kinetic regimes coexisting spatially along the packed bed:

pH2

рНЗ

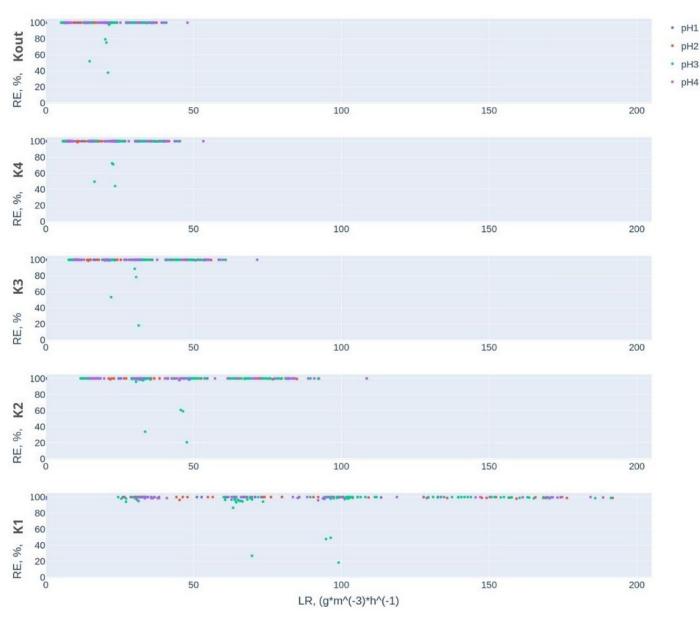


Fig. 3. Effects of H<sub>2</sub>S loading rate on removal efficiency

first-order zones near the outlet (low concentration), reactionlimited zero-order zones near the inlet, and diffusion-limited zero-order zones in the intermediate sections. This multilayer behavior may explain why EC values follow LR so closely even at increasing inlet concentrations. Further insights are provided by Kawase et al. (2014), who investigated ammonia biodegradation in a compact co-current biotrickling filter under varying liquid recycle rates. Their results confirmed that increased irrigation enhances mass transfer and supports higher elimination capacity, and that biodegradation kinetics were best described by the Haldane model, which accounts for both substrate saturation and inhibition effects. Although their study focused on ammonia, the kinetic framework and observed hydrodynamic influences on process performance are relevant to systems treating other gaseous pollutants, including hydrogen sulphide.

An additional consideration is the impact of operating conditions – such as pH and irrigation rate – on H<sub>2</sub>S solubility and bioavailability. As modelled using Onda's approach, thicker liquid films and lower pH values may significantly

reduce the concentration of dissolved H2S available for biodegradation, even when gas-phase concentrations remain high (Beigi 2019).

The Haldane model describes biodegradation kinetics under both substrate-limited and substrate-inhibited conditions. It is expressed by the following equation:

$$r = r_{max} \cdot \frac{C}{K_S + C + \frac{C^2}{K_L}}$$

where:

r – actual reaction rate (g/m³·s)

 $r_{max}$  – maximum reaction rate (g/m $^3$ ·s)

C – substrate concentration in the liquid phase (g/m³)

K<sub>s</sub> – half-saturation constant (g/m<sup>3</sup>)

K<sub>1</sub> – inhibition constant (g/m<sup>3</sup>)

At low substrate concentrations, the Haldane model simplifies to first-order kinetics, while at high concentrations, substrate inhibition causes a decline in reaction rate, distinguishing it from the classical Michaelis-Menten model.

This conceptual understanding of substrate behavior in biofilters is further supported by the kinetic framework proposed by Dumont (2017), which describes the spatial distribution of reaction mechanisms along the packed bed. According to this model, biofilters do not operate under a single kinetic regime. Instead, they consist of three overlapping zones: a region near the gas inlet where reactions are the main limiting step, a central region dominated by diffusion limitations, and an outlet area where reduced substrate concentrations shift the system toward first-order kinetics. Such a stratified profile reflects the interplay between biochemical reaction kinetics and mass transfer phenomena, underscoring the importance of considering these layered dynamics when interpreting overall elimination performance – especially under varying inlet loads and environmental conditions.

In the present study, despite increasing inlet concentrations up to 300 ppm and operation under acidic conditions, no signs of substrate inhibition were observed. The elimination capacity increased proportionally with the loading rate, and removal efficiencies remained high across all tested conditions. These results indicate that neither elevated H2S concentrations nor low pH values negatively affected biodegradation performance within the examined operating range.

During the series of measurements, a maximum elimination rate of 224 g/(m<sup>3</sup>·h), equivalent to the specific inlet load, was achieved. A comparative analysis of literature data revealed that the results obtained for the Tholander research plant fall within the upper range of values reported for BTF systems treating hydrogen sulphide-containing gases. Although direct comparisons between different BTFs are challenging, as system performance is strongly influenced by a wide range of operational and design parameters, including filter media characteristics, contact time, pH control, and microbial composition – the results were compared with those from other studies, taking into account the removal efficiencies achieved by different authors at various hydrogen sulphide inlet concentrations. Examples of effectiveness for other installations are presented below.

For instance, in the study by Ying et al. (2020), a treatment efficiency of 98.3 - 99.8% was achieved with a much lower filter bed load ranging from 2.35 to 17.9 g H<sub>2</sub>S/(m<sup>3</sup>·h). These tests were conducted on a BTF filled with porcelain Raschig rings and ceramsite, and the purified gases, besides hydrogen sulphide, also contained ammonia. Similar to this work, the biofilm was composed of activated sludge sourced from a nitrifying tank. It should be noted that the type of packing material is less important than the specific surface area it provides. A higher surface area per unit volume results in more available surface for biofilm development, thereby increasing the active biomass and enzymatic activity. Reported values of specific surface area vary widely, ranging from approximately  $250-350\ m^2/m^3$  for plastic Raschig rings,  $400-800\ m^2/m^3$ for polypropylene media, and  $600 - 900 \text{ m}^2/\text{m}^3$  for ceramic materials, up to even  $1000 - 1800 \text{ m}^2/\text{m}^3$  for polyurethane

Tab 3 Selected applications of biotrickling filtration (industrial scale)

Gas source	Pollutant	Concentration ranges	RE	Additional information	References
Copper-ore mine	Hydrogen sulphide	5–22 ppm	99%	the gases contained VOCs, mercaptans and hydrogen sulphide	Kasperczyk & Urbaniec 2015
Biogas hy	hydrogen sulphide	12000 ppm	95–99%	biogas from distillery anaerobic digesters in the ethanol industry	Haosagul et al. 2019a
	hydrogen sulphide	2200–2500 ppm	99%	biogas from swine farm	Haosagul et al. 2019b
	hydrogen sulphide	1500–3000 ppm	98%	biogas from WWTP	Rodriguez et al. 2014
Wastewater treatment plant	hydrogen sulphide VOC	200 ppm 25–240 ppm	97% 85–99%		Kasperczyk et al. 2019
	hydrogen sulphide	30 ppm <sup>-</sup>	95% 90%	Gas Contact Time: <sup>*</sup> 1.6 s <sup>**</sup> 2.2 s	Gabriel & Deshusses 2003
	hydrogen sulphide odours **	30–100 ppm 7035–23042 ou <sub>e</sub> /m³	99% 92–96%	"anaerobic tanks of BNR	Lafita et al. 2012b
	hydrogen sulphide	> 300 mgS/m <sup>3</sup>	80%		Spennati et al. 2017
Sewage sludge – processes	odours	121072 ou <sub>E</sub> /m³	97%		Sempere et al. 2018
	hydrogen sulphide VOC	25 ppm 14 ppm	92% 92%		Beigi et al. 2019

(PU) foam. Additionally, the packing material should support microbial colonization while minimizing flow resistance. These aspects - including porosity, hydrodynamics, and mechanical stability – play a key role in determining BTF performance (Sakuma et al. 2006, Danila et al. 2022, Cox and Deshusses 2001, Estrada et al. 2012).

Research on a two-layer BTF with a similarly low bed load  $(15-50 \text{ g H}_2\text{S/(m}^3 \cdot \text{h}))$  was conducted by Chen et al. (2016). In their study, the biotrickling filter was filled with two layers: an upper layer consisting of activated carbon-loaded polyurethane, and lower layer composed of modified organism-suspended fillers. The bed was inoculated with activated sludge and sulphur-oxidizing microorganisms isolated from activated sludge, which were expanded before immobilization. The researchers achieved a hydrogen sulphide removal efficiency of over 96%.

Two other teams of researchers, Tsang et al. (2015) and Fernandez et al. (2013), conducted tests with bed loads of up to 120 g H<sub>2</sub>S/(m<sup>3</sup>·h) and achieved removal efficiencies of 99%. In the study conducted by Tsang et al. a BTF was filled with coal slag and inoculated with sulphide-oxidizing and ammoniaoxidizing bacteria, both prepared from activated sludge. Fernandez et al., on the other hand, conducted experiments using a biotrickling filter packed with PP Pall rings and inoculated with biomass immobilized in previously used BTF studies utilizing open polyurethane foam (OPUF).

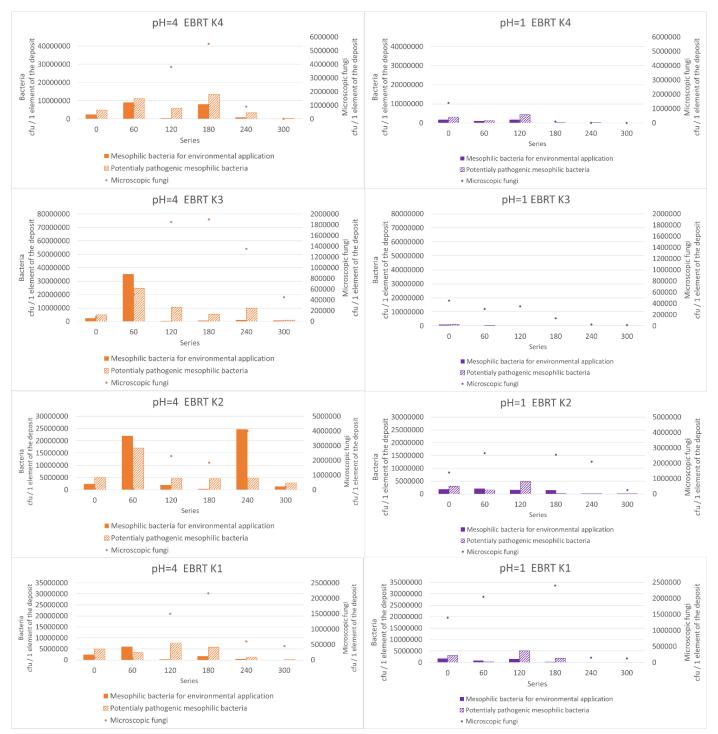


Fig. 4. Distribution of microbial groups at different column levels

Parzentna-Gabor et al. (2023) conducted tests with hydrogen sulphide concentrations ranging from 0 to 150 ppm at pH = 7 and pH = 5. The experiments were conducted at a wastewater treatment plant using a pilot-scale compact trickle bed bioreactor (CTBB). In the liquid phase, pollutant removal efficiency was observed to be higher at pH = 7.0 compared to pH = 5.0. The hydrogen sulphide removal efficiency ranged from 87.5% to 98.9%.

In a study by Wu et al. (2020), biofilter bed loads similar to those used in the present work were applied, ranging from 101 to 210 g  $\rm H_2S/(m^3 \cdot h)$ . The researchers achieved 100% purification efficiency using a biotrickling filter packed with PP cylindrical plastic carriers with internal ribs, which were inoculated with activated sludge.

However, other studies focusing on the application of biotrickling filters in real-world settings (as summarized in Table 3) also demonstrate their effectiveness for biogas purification. These studies report high purification efficiencies even at significantly elevated hydrogen sulphide concentrations. The results obtained in the present study indicate that similarly high purification efficiencies can be achieved at high hydrogen sulphide inlet concentrations in the tested trickling biofilter.

# Comparison of microbial community structure

The microorganisms commonly inhabiting biofilms in biotrickling filters are bacteria and fungi. Their distribution within the packing material depends on various factors, including technological process parameters, bed type, and position relative to the inlet of contaminated gases. As gases flow sequentially through the bed, different environmental conditions develop along the column. Higher concentrations of pollutants at the inlet provide abundant nutrients, leading to intensive biomass growth. During the decomposition of pollutants, only a negligible amount of heat is generated. However, the H<sub>2</sub>S degradation leads to formation of sulfuric acid. Although drying is generally prevented by system irrigation, the resulting pH decrease is significant and promotes fungal growth. This phenomenon was also observed in our own research. Figure 4 illustrates the distribution of individual groups of microorganisms at different levels of the column, for example, at pH 4 and pH 1.

The influence of pH on microbial community structure in biotrickling filters has been described inconsistently in the literature. According to Trisakti et al. (2015), low pH conditions favor autotrophic bacteria that oxidize hydrogen sulphide to elemental sulphur or sulphate, whereas higher pH supports heterotrophic bacteria, which are more effective in VOC removal. However, other authors have reported that autotrophic sulphur-oxidizing bacteria can also dominate under alkaline conditions (Zang et al. 2021; González-Sánchez et al. 2008), achieving high H2S removal efficiencies even at pH 10. The presence of haloalkaliphilic bacteria capable of operating in pH levels ranging from 9 to 11 has also been documented (Sorokin & Kuenen 2005). These contrasting observations suggest that microbial dominance may be highly dependent on operational parameters and the origin of inoculated biomass. Our study, showing an increased abundance of fungi under acidic conditions, is consistent with the idea that low pH environments promote specific microbial shifts, although further analysis is needed to confirm the metabolic roles of the dominant groups.

# **Conclusions**

Under the established operating parameters, hydrogen sulphide (H<sub>2</sub>S) removal efficiency ranging from 98.3% to 100% was achieved. Depending on the specific retention time and the hydrogen sulphide concentration at the column inlet, variations in the proportions of microbial groups were observed both over time and in the vertical profile of the column. As a continuation of this work, further measurement series are planned under controlled pH values and varying inlet contaminant concentrations. The objective is to determine the impact of retention time on hydrogen sulphide removal efficiency and to identify critical parameters for plant operation. Optimization of these parameters could lead to improvements in hydrogen sulphide removal efficiency in biotrickling filters.

The results obtained in this study carry significant practical implications for industrial applications and gas purification processes involving hydrogen sulphide. Our research has validated the effectiveness of the employed method and highlights the potential for reducing the use of alkalis, thereby lowering the operational costs associated with the purification process. On an industrial scale, our findings could serve as a foundation for enhancing current gas purification methodologies and pioneering the development of novel, more efficient technologies.

Moreover, the study opens several promising avenues for further exploration in this field. Continued efforts are needed to optimize operational parameters, such as Empty Bed Residence Time (EBRT), and to systematically evaluate the influence of additional factors on treatment efficiency. A key assumption of this study was that the microbial structure observed under controlled laboratory conditions would be comparable to that in scaled-up systems. Future investigations should aim to verify this assumption under industrial operating conditions, especially when exposed to fluctuating loads or varying compositions of waste gas. While pH was carefully controlled during the experiments, other variables - such as oxygen transfer efficiency, nutrient availability, or biofilm thickness - were either held constant or inferred indirectly. Further research is needed to explicitly assess the interactions among these factors and could examine the interplay of these parameters and their respective contributions to overall system performance. In parallel, delving into alternative technologies and refining current methodologies could support continued progress in the biological treatment of hydrogen sulphidecontained gases.

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# Wpływ pH i obciążenia złoża na wydajność usuwania siarkowodoru w biofiltrze zraszanym

Streszczenie. Badanie wpływu pH oraz obciążenia złoża na skuteczność usuwania siarkowodoru (H<sub>2</sub>S) w biofiltrach zraszanych, z naciskiem na optymalizację parametrów operacyjnych dla procesów oczyszczania gazów. W badaniach zastosowano pilotażową instalację biofiltra zraszanego z wypełnieniem z pianki poliuretanowej. Testy przeprowadzono przy stałych poziomach pH (1, 2, 3 i 4) oraz zmiennych stężeniach H<sub>2</sub>S na włocie (60–300 ppm). Analizowano parametry takie jak szybkość biofiltracji (EC), skuteczność usuwania (RE) oraz strukturę mikrobiologiczną biofilmu. Warunki procesowe, w tym pH, przewodność i potencjał redoks, były automatycznie kontrolowane i monitorowane. Biofiltr zraszany osiągnął maksymalną szybkość biofiltracji wynoszącą 224 g/m³/h przy niemal 100% skuteczności oczyszczania. Zaobserwowano zmienność w strukturze mikrobiologicznej biofilmu wzdłuż wysokości kolumny, zależną od pH oraz stężenia zanieczyszczenia. Stwierdzono, że pH miało minimalny wpływ na szybkość biofiltracji (EC). Biofiltr zraszany wykazał wysoką skuteczność w usuwaniu H<sub>2</sub>S, co ma obiecujące implikacje dla zastosowań przemysłowych. Dalsza optymalizacja parametrów, takich jak czas przebywania gazu w złożu (EBRT) i kontrola pH, może zwiększyć wydajność oraz obniżyć koszty operacyjne.