

## Review

# Removal of Odors (Mainly H<sub>2</sub>S and NH<sub>3</sub>) Using Biological Treatment Methods

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**Abstract:** This study reviews the available and most commonly used methods of gas deodorization. Comparing various methods of odor removal, undoubtedly biological methods of pollution degradation have an advantage over others—chemical and physical. This advantage is manifested mainly in ecological and economic terms. The possibility of using biological methods to remove H<sub>2</sub>S and NH<sub>3</sub>, as the most common emitted by the municipal sector companies, was analyzed in terms of their removal efficiency. The method of bio-purification of air in biotrickling filters is more advantageous than the others, due to the high effectiveness of VOCs and odors degradation, lack of secondary pollutants, and economic aspects—it is a method competitive to the commonly used air purification method in biofilters.

**Keywords:** biodegradation; odors; H<sub>2</sub>S; NH<sub>3</sub>



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## 1. Introduction

Fragrances, also known as odors, and volatile organic compounds (VOCs) are compounds that play an important role in environmental pollution. They are gaseous pollutants and chemical vapors, characterized by high vapor pressure and low solubility in water. The presence of odors is a problem mainly in urbanized areas, whose source is the combustion of hydrocarbon fuels, chemical and petrochemical industries, mining, municipal, food processing, and agricultural waste [1]. The economic with the highest odor nuisance are primarily waste management plants and wastewater treatment plants, as well as food and agricultural processing plants. Odors are a mixture of volatile chemicals known as odorous gases that can be felt by humans in very low concentrations. They play an important role in the lives of all living organisms, especially humans, causing health problems. Odors are carcinogenic, mutagenic and toxic, and due to odor nuisance, they also affect mental health. These compounds cause irritation of the respiratory system, which results in runny nose, cough, shortness of breath, sore throat, and lacrimation. In many cases, due to people's individual hypersensitivity, psychosomatic symptoms, such as insomnia, reduced psychophysical and emotional performance, and panic attacks also occur [2,3]. The most exposed to the described health problems are people living in the immediate vicinity of the emission sources, as well as employees of odor emitting plants. The incoming new residential and industrial investments cause an increase in the density of development the urban agglomeration, and thus too close proximity to residential buildings with odor-emitting plants, especially wastewater treatment plants, which are very often located in city centers, in the vicinity of large housing estates [4].

## 2. Gases Emitted in the Municipal Sector

Facilities emitting the most persistent harmful gases include companies from the municipal sector, including waste water treatment plants, waste management plants or composting plants. The odor- produced in municipal wastewater treatment plants include

- sulfur compounds, i.e., hydrogen sulfide, thiols, sulfides, and alkyl disulfides,
- nitrogen compounds, i.e., ammonia, and aliphatic amines,
- organic compounds, including aldehydes, ketones, and fatty acids (phenol, cresol, butyric acid, acetic acid, and valeric acid)

Table 1 lists the most common and nuisance odor compounds found in municipal wastewater treatment plants.

**Table 1.** Concentrations of odor compounds above the surface of municipal wastewater [5] (Authors' own study according to [5], Polish Scientific Publishing House: 2012).

Odor Compounds	Substance	Concentration [ppm]	Detection Threshold [ppm]
Compounds with Nitrogen	Ammonia	0.019–5.2	5
	Trimethylamine	1.7	0.00044
	Methylamine	3.3	0.02
	Pyridine	0.013–0.82	0.084
Compounds with Sulfur	Hydrogen sulfide	0.001–0.78	0.008
	Dimethyl sulfide	0.0015–0.02	0.0023
	Diethyl sulfide	0.00025–0.0006	0.004
	Diethyl disulfide	0.000054	0.00043
	Methyl mercaptan	0.0001–0.55	0.001
	Ethyl mercaptan	0.000016–0.074	0.00076
Volatile Organic Compounds	Phenol	0.047–0.65	0.040
	Cresol	0.00047	0.0018
	Butyric acid	0.00028–0.00056	0.004
	Valeric acid	0.0006	0.005

In most wastewater treatment plants, ammonia and hydrogen sulfide have the highest concentrations and amounts of odor compounds in the emitted gases [6,7]. Ammonia is a colorless, corrosive gas with a very pungent and unpleasant odor. Its corrosive and exothermic properties can damage the eyes, skin, and mucous membranes of the mouth and respiratory tract. The effect of ammonia on the human body and the accompanying disease symptoms are presented in Table 2.

**Table 2.** The effect of ammonia on the human body [8] (Authors' own study according to [8]. The National Academies Press: Washington: 2008).

Concentration NH <sub>3</sub> [ppm]	Symptoms
<5–53	Odor threshold
30	Slight irritation after 10 min.
50	Prolonged exposure may cause nausea, tearing of the eyes, Moderate irritation to the eyes, nose, throat and chest after 10 min to 2h
80	Moderate to highly irritation after 30 min to 2 h
110	Highly intense irritation after 30 min to 2h of exposure
140	Unbearable irritation after 30 min to 2h
500	Excessive lacrimation and irritation
570 (21–30 years old) 1000 (60 years old) 1790 (86–90 years old)	Reflex glottis closure—a protective response to inhaling irritant vapors

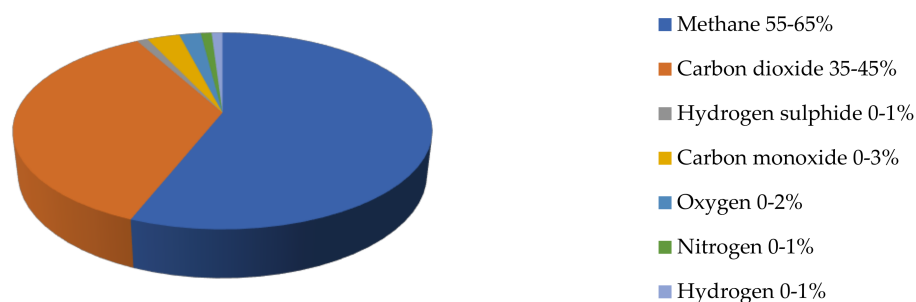
Another very nuisance odor compound, emitted by wastewater treatment plants, is hydrogen sulfide—a colorless, highly flammable and explosive gas, with the smell of rotten eggs, felt in very low concentrations. This gas has a toxic effect on all living organisms,

including humans, causing a number of unpleasant life-threatening ailments. The effect of hydrogen sulfide on the human body along with disease symptoms is presented in Table 3.

**Table 3.** The effect of dihydrogen sulfide on the human body [9].

Concentration H <sub>2</sub> S [ppm]	Symptoms
0.00011–0.00033	Typical background concentrations
0.01–1.5	Odor threshold (rotten egg)
2–5	Prolonged exposure may cause nausea, tearing of the eyes, headaches or loss of sleep
20	Possible fatigue, loss of appetite, headache, irritability, poor memory, dizziness
50–100	Slight conjunctivitis (“gas eye”) and respiratory tract irritation after 1 h. May cause digestive upset and loss of appetite
100	Coughing, eye irritation, loss of smell after 2–15 min (olfactory fatigue). Altered breathing, drowsiness after 15–30 min. Throat irritation after 1 h. Gradual increase in severity of symptoms over several hours. Death may occur after 48 h
100–150	Loss of smell (olfactory fatigue or paralysis)
200–300	Marked conjunctivitis and respiratory tract irritation after 1 h. Pulmonary edema may occur from prolonged exposure
500–700	Staggering, collapse in 5 min. Serious damage to the eyes in 30 min. Death after 30–60min
700–1000	Rapid unconsciousness, “knockdown” or immediate collapse within 1 to 2 breaths, breathing stops, death within minutes
1000–2000	Nearly instant death

The main factors influencing the concentration of odors in the air are the rate of emission and dispersion of gases, which are directly affected by atmospheric conditions (temperature, wind direction, and atmospheric pressure) and geomorphological conditions [10]. The most odor-nuisance areas in the wastewater treatment plant include pretreatment unit—raw wastewater tanks, coarse and fine screen, and sand traps—preliminary settling tanks and sludge management facilities—sludge tanks, and sludge dewatering halls [5,7,11]. Preliminary sedimentation tanks and sewage sludge tanks generate the largest amounts of odors due to the large area of these objects and the emission of gases from their entire surface [12]. Excessive sewage sludge generated in the wastewater treatment process is characterized by a high efficiency of rotting. For their disposal or further use, e.g., for land reclamation, it is necessary to stabilize the sludge and deprive it of pathogenic organisms. For this purpose, the excess sludge is directed to hermetic fermentation chambers, where the four-stage process of anaerobic fermentation takes place, with the participation of several groups of anaerobic bacteria. These bacteria break down complex organic substances (proteins, fats, and carbohydrates) into methane and carbon dioxide. In the fermentation process, the collected biogas can be used to produce heat and electricity [13,14] but before that, it must be purified [15]. Biogas resulting from fermentation consists mainly of methane and carbon dioxide as well as trace amounts of hydrogen, carbon monoxide, nitrogen, oxygen, and hydrogen sulfide (Figure 1), in order to be used for energy production in installations, it must meet the appropriate requirements adapted to such devices as engines and boilers, therefore it is necessary to clean them, also to increase their calorific value [16].



**Figure 1.** Composition of biogas [13,15].

Various techniques of biogas conditioning are used. The most frequently used are desulfurization by help of turf ore, membrane separation, pressure swing adsorption, physical absorption, chemical absorption, and biotechnological methods such as use of biofilters, bioscrubbers, biotrickling filters as well as activated sludge [15,17,18]. The selection of the appropriate technology for biogas purification depends on specific requirements, taking into account the type of installation and local conditions.

### 3. Commonly Used Odor Removal Technologies

Limiting odor emissions generated in wastewater treatment plant consists in preventing the emission of gases directly into the atmosphere, e.g., by hermitization the most odor-troublesome technological devices, and deodorization of exhaust gases. The most frequently used deodorization methods in municipal sector include absorption with the use of reactive oxidizing solutions, adsorption on activated carbon, combustion, and biological methods [7,19]. Recently, biological methods of odor removal have become more and more popular, which using natural reactions occurring in nature, are ecological, effective and inexpensive solutions [20].

#### 3.1. Physicochemical Methods of Odor Removal

The physicochemical methods for deodorizing gases emitted by wastewater treatment plants include absorption, adsorption, and combustion. Air purification by absorption method consists in transferring pollutants from the emitted gas to the liquid and enables the separation of the gas mixture into individual components [21]. In the case of odors from wastewater treatment plants, the absorption efficiency in water is very low, due to the low solubility of most odor pollutants.

In order to increase the efficiency of this process, solutions of oxidants are used as absorption liquids, e.g., ozone  $O_3$ , hydrogen peroxide  $H_2O_2$ , sodium chlorate (I)  $NaOCl$ , under the influence of which organic compounds are oxidized to carbon dioxide ( $CO_2$ ), and hydrogen sulfide to elemental sulfur (S), mercaptans, and sulfides to sulfonic acids or sulfones. These reactions can be accelerated by adding appropriate catalysts (e.g., salts containing iron ions(II)  $Fe^{2+}$ ) [22]. The use of reactive chemicals as absorption liquids requires the use of chemically resistant construction materials to minimize the risk of environmental contamination due to leakage of reagents. The absorption method is an effective solution for removing ammonia ( $NH_3$ ) and hydrogen sulfide ( $H_2S$ ), but it is much more difficult to remove volatile organic compounds, including volatile fatty acids, mercaptans. Moreover, this method generates noxious sewage that must be disposed of [5]. Chemical absorption in many cases is used as a pre-treatment method of emitted gases characterized by a high concentration of odors [7].

Adsorption is a process of inhibiting a pollutant by a solid—an adsorbent. Activated carbon and zeolite are most often used for deodorization, which are characterized by high adsorption capacity in relation to odor compounds [22]. Adsorbents used for deodorizing the emitted gases are in the form of powder (8–80  $\mu m$ ), granules (200  $\mu m$  to 6 mm), compacts (0.8 to 5 mm in diameter and 5 to 20 mm long), pellets (30 to 60 mm in diameter), fibers or fabrics. Except activated carbon and zeolites, diatomaceous and



volcanic earths, sawdust, silica, aluminum oxides, and peat are also used. In addition, clay minerals and polymeric synthetic resins are also used, but this group of adsorbents absorbs odorant molecules much worse. After complete saturation of the bed, its regeneration is carried out to remove adsorbed impurities, depending on their type, various methods are available: thermal, vacuum or chemical regeneration, storage, combustion, but in the case of deodorization, sorbent is usually not regenerated due to the risk of secondary odor emissions and small benefit [5].

In contrast, combustion can be generally divided into thermal and catalytic combustion. Thermal combustion, without the addition of catalysts, requires very high temperatures—in the case of phenol, the combustion temperature reaches 720 °C—which is associated with very high financial outlays. Therefore, the method of catalytic combustion is more widely used—for comparison, the catalytic combustion temperature for phenol is 250 °C. The product of catalytic combustion of hydrocarbons and organic compounds containing oxygen is carbon dioxide and water, and in the case of improperly selected process parameters there is a risk of incomplete combustion and emission of toxic compounds (e.g., aldehydes). The role of catalysts is played by inorganic supports, such as silica, alumina, zeolite, and activated carbon, on which precious metals—platinum, palladium, copper, or vanadium—are deposited. The combustion of low concentrations of odors, about a few mg/m<sup>3</sup>, is in most cases uneconomical, because all the heat needed to heat the gases must be supplied from external sources. In such cases, it is necessary to increase the odor concentration, by concentrating them in order to reduce costs [23]. For this purpose, a common practice is to combine combustion processes with adsorption [24]. First, the adsorbent is saturated with pollutants as a result of odor adsorption on active carbon, and then the pollutants are desorbed from the adsorbent and concentrated in the gas, which is then subjected to the combustion [5,22].

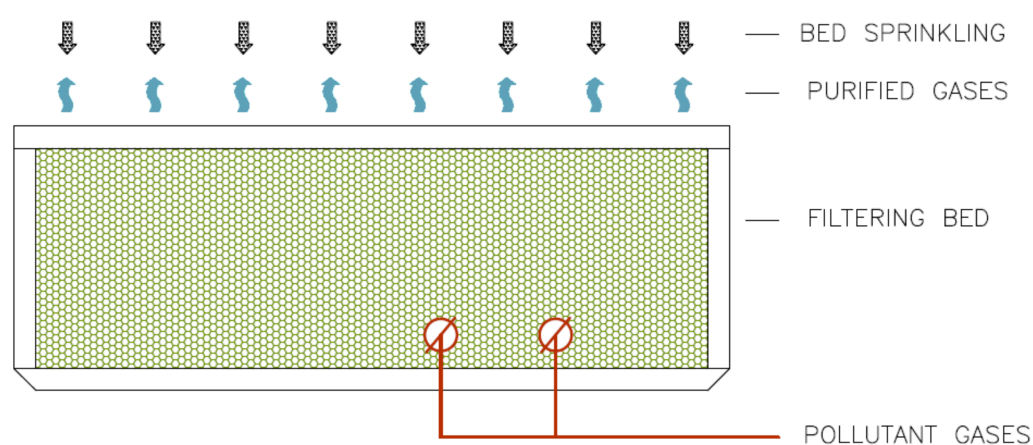
### 3.2. Biological Methods of Odor Removal

Biological methods of gas purification, based on the natural processes of decomposition of organic compounds occurring as a result of the metabolic activity of microorganisms, have gained an opinion in recent decades of the most beneficial methods of pollutant degradation. This opinion results from several significant advantages of biological methods: economy, ecological purity, lack of secondary pollutants, use of processes naturally occurring in nature, and high efficiency of pollution removal [20,25]. There are three main technologies used for air bio-purification: biofilters, bio-scrubbers and biotrickling filters. These methods differ in the type of layers and mobile phases as well as in the location of pollutant-degrading microorganisms [4,26,27].

#### 3.2.1. Biofilters

Deodorization by biofiltration, as shown in Figure 2, consists in passing a humidified, contaminated gas through a solid bed containing microorganisms capable of odors and VOCs degradation. The pollutants are sorbed and then absorbed by bacteria and decomposed into water and carbon dioxide [20]. In most cases, the biofilter bed consists of organic materials: wood bark, peat, straw, loosened soil, compost, coconut fiber, and activated carbon. The biofilter bed is piled up from one to several layers in such a way as to ensure contact of the entire gas stream with the bed and to maintain uniform aeration of the bed in order to prevent the growth of anaerobic bacteria, causing the bed to rot [19]. Bacteria, which have a natural ability to degrade odor and VOCs pollutants, form a biofilm on the surface of the biofilter bed and are selected according to the composition of pollutants present in the gas passing through the biofilter. In addition, they are provided with appropriate conditions for growth and development, e.g., by maintaining an appropriate pH in the bed and regularly supplying nutrients and mineral salts. Microorganisms are selected in such a way as to ensure their greatest diversity, which will enable the degradation of the widest range of pollutants [14]. Biofilters are a commonly used method of purifying gases from odors and VOCs emitted by the municipal sector because this method effectively

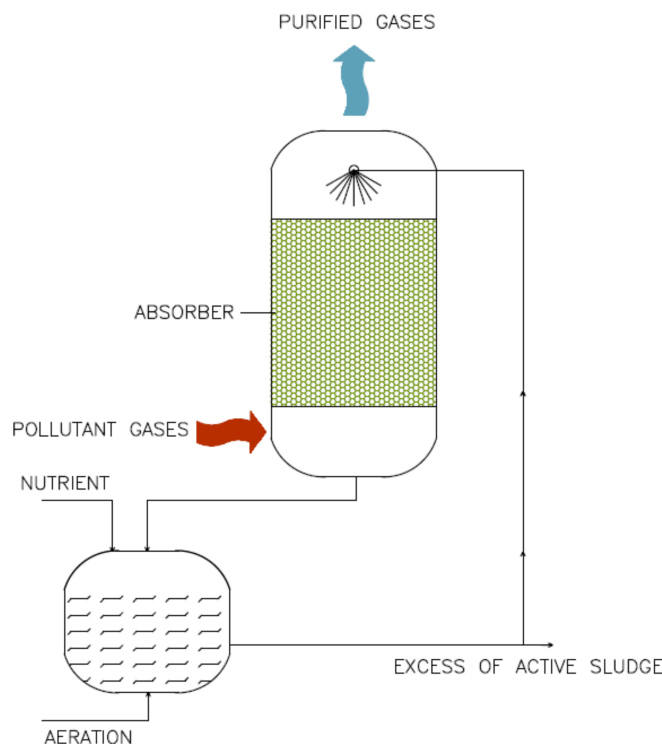
removes both organic pollutants, including aromatic hydrocarbons (toluene and xylene), alcohols, aldehydes, organic acids, and amines as well as inorganic compounds such as hydrogen sulfide and ammonia. However, in the case of inorganic pollutants, it is necessary to control their concentration, due to the products formed as a result of their decomposition, which cause acidification of the biofilter environment. Moreover, the biofiltration method has some limitations on the concentration of pollutants in the treated gas. Biofilters are usually used for treatment of relatively large gas streams [4] (according to practice they are usually used for flows up to 5000 m<sup>3</sup>/h), therefore they require a large mass exchange area and consequently a large size biofilter. The main disadvantages of biofiltration are the difficulty of controlling the process—maintaining the appropriate humidity and pH of the bed (which may become acidic)—as well as clumping of the filter material, relatively large installation size and lower treatment efficiency at high concentrations of pollutants. On the other hand, the advantages include low operating and investment costs, the possibility of purifying large streams of gases at low concentrations of pollutants [4,28].



**Figure 2.** Diagram of a biofilter (own study, based on [5]).

### 3.2.2. Bioscrubbers

The principle of bioscrubber operation is based on two main stages that usually take place in separate devices [25]. In the first tank—the absorber—gaseous pollutants are absorbed into the liquid phase, which then goes to the second tank—the bioreactor. The bioreactor is filled with an aqueous suspension of microorganisms in which biodegradation of pollutants takes place. The liquid circulates through tanks supplied with air, nutrients for bacteria and pH adjusting solutions, while the excess of activated sludge is drained outside the system. The principle of operation and the structure of the bioscrubber are shown in Figure 3. The absorbers are filled with a bed that acts as a carrier for microorganisms. On the surface of the filling, microorganisms form a biofilm consisting of clumped bacterial cells and extra cellular polymeric substances (EPS) [29] capable of colonizing various environments and surfaces [30,31]. The undoubted advantage of bioscrubbers is the ability to control their operating parameters, such as pH, nutrient solution, aeration, which directly stabilizes their work. In addition, the installation is characterized by small dimensions, which is a significant advantage compared to biofilters, and there is no problem of clogging of the filter material. However, bioscrubbers generate large amounts of by-products such as excess sludge and contaminated, recirculated liquid; moreover, the operating costs of maintaining such an installation are much higher than in the case of biofilters [4,32].



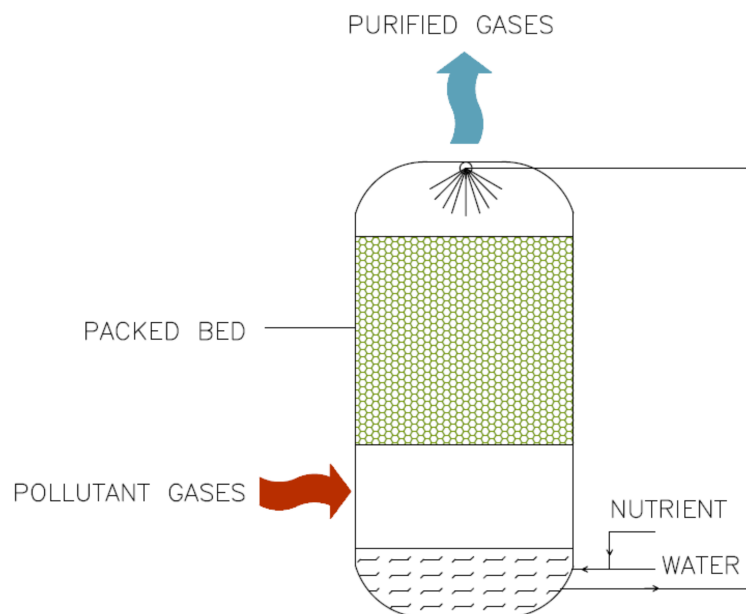
**Figure 3.** Diagram of a bioscrubber (own study, based on [5]).

Bioscrubbers are successfully used to remove odors, in particular  $\text{H}_2\text{S}$ ,  $\text{NH}_3$ , and organic compounds with sulfur. However, due to their acidic nature, these substances cause a significant drop in pH, which may result in acidification of the medium circulating in the installation and a decrease in the efficiency of gas treatment [4].

### 3.2.3. Biotrickling Filters

In biotrickling filters, the process of absorption of pollutants into the liquid phase, and their biodegradation along with further liquid regeneration, takes place simultaneously in one tank [27]. The polluted gas enters the apparatus, in which it flows in the same direction or opposite to the liquid phase, in which the absorption of pollutants takes place. The liquid containing nutrients necessary for the development of microorganisms, along with the absorbed impurities, flows continuously as a thin film on the surface of the bed. As a result, the biofilm layer formed on the bed is constantly wetted, and biodegradation of pollutants to simple products such as water and carbon dioxide takes place there [4,27]. The liquid circulating in the plant is constantly recirculated, so there is no sludge waste. The scheme of biotrickling operation is shown in Figure 4. The bed in this type of installation is made of chemically inert materials, such as activated carbon, ceramic rings, glass balls, and plastic structures [20,27].

One of the advantages of biodegradation of gaseous pollutants in biotrickling filters over other methods is the ability to better control their operating conditions, such as maintaining an appropriate pH and composition of the medium circulating in the reactor. Moreover, the undoubted advantage is that the entire process is carried out in one tank, which saves a lot of space and total costs. On the other hand, the disadvantages that appear during the operation of biotrickling filters may be excessive growth of biomass inhabiting the bed, which may lead to the clogging of the bed and, as a result, a decrease in efficiency [33]. However, there are effective methods to counteract this, e.g., by temporarily increasing the flow of the liquid phase, which will result in breaking a part of the biofilm from the filling [34] or by appropriate selection of microorganisms eliminating excess bacterial biofilm (including protozoa), or by adding appropriate chemical to damage part of the bacterial biofilm [35].



**Figure 4.** Diagram of a biotrickling filter (own study based on [36]).

#### 4. Effectiveness of $\text{H}_2\text{S}$ and $\text{NH}_3$ Removal Using Biological Methods of Odor Degradation

Among the currently used odor removal methods, biological methods turn out to be the most attractive, in particular biofilters and biotrickling filters [7]. Among the biological methods of air purification, biofilters are relatively simple and the longest used methods; hence, also, the best known [14], there are many literature reports confirming the use of biofilters for odor removal.

##### 4.1. Application of Biofilters to Remove $\text{H}_2\text{S}$ and $\text{NH}_3$

Chung et al. [37] studied the degradation of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  using a biofilter. Impurities in the form of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  were administered in various proportions. Their biodegradation efficiency was on the average level of over 95%, regardless of the  $\text{H}_2\text{S}$  and  $\text{NH}_3$  ratios used. The research was carried out in an experimental biofilter in the form of a column, on a laboratory scale. Moreover, it has been found that  $\text{H}_2\text{S}$  can inhibit  $\text{NH}_3$  removal, while  $\text{NH}_3$  concentration has only a negligible effect on  $\text{H}_2\text{S}$  removal.

Whereas Choi et al. [38] tested the  $\text{NH}_3$  removal efficiency in two types of biofilters—with vertical and horizontal gas flow. Mixtures of organic materials such as compost, bark and peat were used as fillings, as well as inorganic material—perlite (perlite). The result of the research was the determination of the ammonia removal capacity with the use of organic and inorganic media used in biofilters in order to select the most efficient filling. The organic packing achieved higher ammonia removal efficiency without significant pressure loss. When testing different types of gas flow, higher contamination removal efficiency was noted for horizontal gas flow reaching 100%.

Tymczyna et al. [39] also investigated the biodegradation efficiency of  $\text{NH}_3$  with an open biofilter, but in this case the source of  $\text{NH}_3$  was a poultry farm. The biofilter bed consisted of fibrous peat, coarse peat, wheat straw, wastewater treatment plant compost, and horse manure and was 1.2 m high, while the biofilter chamber area was 10 m<sup>2</sup>. The efficiency of degradation of pollutants in the biofilter was tested in five phases, in the initial phase of the experiment (after five days from filling the biofilter chamber) the efficiency was low—at the level of 36%, while after three months of biofiltration it increased to 89% and thus this result was the highest efficiency  $\text{NH}_3$  removal during the experiment.

Pagans et al. [40] also investigated the effectiveness of  $\text{NH}_3$  removal, this time from the gases emitted in the composting process, using a biofilter. The ammonia removal efficiency

was nearly 96%. A significant decrease in the efficiency of  $\text{NH}_3$  biodegradation was observed when its concentration at the inlet to the biofilter increased to over  $2000 \text{ mg/m}^3$ .

While Rehman et al. [41] investigated the performance of biofilters intended for  $\text{H}_2\text{S}$  removal. The research was carried out in laboratory conditions, in six phases—starting with feeding only humidified air to the biofilter and gradually increasing the concentration of  $\text{H}_2\text{S}$  with the subsequent phases. It was found that the biofilter most effectively removed  $\text{H}_2\text{S}$  in the concentration range from 10 ppm to 30 ppm, then the efficiency was above 95%, while above these values the efficiency decreased, reaching an efficiency of 85% at an  $\text{H}_2\text{S}$  concentration of 50 ppm.

In turn, the aim of the research by Omri et al. [42] was to investigate the degree of  $\text{H}_2\text{S}$  removal in a biofilter filled with peat. The experiment was conducted on a pilot scale in a wastewater treatment plant in Tunisia. The concentration of  $\text{H}_2\text{S}$  in the inlet gases ranged from 200 to  $1300 \text{ mg/m}^3$ , while the efficiency of  $\text{H}_2\text{S}$  removal reached 99%.

Kavyashree et al. [43] investigated the use of a mixture of manure and rice husk as a filling in a biofilter to remove ammonia emitted by a municipal composting plant at concentrations of  $500\text{--}700 \mu\text{g/m}^3$ . The research was carried out with the use of a biofilter on a laboratory scale, for two variants of the bed depth: 20 cm and 40 cm. The effectiveness of  $\text{NH}_3$  removal for a 20 cm bed depth was 61.5%, while for a 40 cm deep bed it was 71.45%. It was found that along with the increase in the number of bacteria in the deposit, the efficiency of ammonia degradation increases.

Aita et al. [44] investigated the effectiveness of removing  $\text{H}_2\text{S}$  present in synthetic biogas using a biofilter filled with sawdust. The tests were carried out for 37 days, with an average  $\text{H}_2\text{S}$  removal efficiency of  $75 \pm 13\%$ , while the maximum efficiency was 97%.

Rabbani et al. [45] investigated the effectiveness of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  removal from wastewater treatment plants, in a pilot-scale biofilter, under real conditions at the wastewater treatment plant. The experiment consisted of two stages, in the first stage, the biofilter was placed behind a chemical acid scrubber that removed  $\text{NH}_3$  from gases. Thus, in the gases entering the biofilter, only  $\text{H}_2\text{S}$  was present, which as a result of biological oxidation formed  $\text{H}_2\text{SO}_4$ , which was deposited at the bottom of the biofilter. The aim of stage I was to develop a sufficient amount of biofilm to remove  $\text{H}_2\text{S}$  and to generate an appropriate amount of  $\text{H}_2\text{SO}_4$  accumulated at the bottom of the biofilter to remove  $\text{NH}_3$  in stage II. In turn, in the second stage of the experiment, gases containing a mixture of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  were introduced into the same biofilter, this stage lasted seven weeks. The average  $\text{H}_2\text{S}$  removal efficiency was 91.96% and  $\text{NH}_3$  100%. At the bottom of the biofilter, a small amount of effluent ( $0.2 \text{ mlof effluent/L reactor/day}$ ) accumulated in the form of ammonium sulfate. The authors noted that in the case of using biofilters on a full industrial scale, it would be necessary to look at the exact amounts of leachate produced.

Whereas the subject of research by Janas and Zawadzka [46] was the degradation of various odor compounds, including  $\text{H}_2\text{S}$  and  $\text{NH}_3$ , emitted by the wastewater treatment plant with the use of a biofilter. The concentrations of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  at the inlet to the biofilter were  $154 \mu\text{g/m}^3$  and  $1799 \mu\text{g/m}^3$ , respectively, while their removal efficiency was 94% and 91%. However, despite the high efficiency of odor biodegradation, odor has not been completely eliminated.

Alinezhad et al. [6] compared the removal efficiency of odors consisting mainly of  $\text{H}_2\text{S}$  and  $\text{NH}_3$ , emitted by a municipal wastewater treatment plant, using a chemical scrubber and a biofilter. The studies were conducted for 45 days. The biofilter was constantly fed with contaminated gas, while the efficiency of the removal of pollutants in the scrubber was tested only during those times of the day when odor concentrations were at the highest level. Both systems reported almost complete removal of  $\text{NH}_3$ , while the  $\text{H}_2\text{S}$  removal efficiency was 95%. The experiment compared both methods in terms of technology and economy. The technological advantage of the chemical scrubber method over the biofilter was found due to the speed of gas loading and the limitations of the biofilter system. The degradation of both pollutants ( $\text{H}_2\text{S}$  and  $\text{NH}_3$ ) in a chemical scrubber was over 97%, while in the biofilter it was 92% for  $\text{H}_2\text{S}$  and 99.5% for  $\text{NH}_3$ . However, in economic terms,



the biological method of odor degradation in the biofilter turned out to be much more advantageous.

Baltrenas et al. [47] examined the effectiveness of air purification from ammonia in plate biofilters. The research was carried out with the use of different structures—a biofilter with straight lamella plates and a biofilter with wavy lamella plates. Various types of microorganisms were used, including yeast and bacteria. The efficiency of biopurification of air from ammonia was tested at various temperatures ranging from 24 to 32 °C. The best efficiency of ammonia biodegradation was achieved in a biofilter with wavy lamella plates and ranged from 84.2% to 87%.

Due to the simplicity of use and economic advantages for the recipient, biofilters have so far been the most frequently used method to removing odors, and thus the best known. However, for several decades, the odor removal technology in biotrickling filters has become an extremely competitive alternative. Examples of the use of biological degradation methods to remove  $H_2S$  and  $NH_3$  are shown in Table 4. Most likely, this is due to the legal restrictions on odor emissions and the need to find a method whose effectiveness reaches almost 100%, as well as the dynamic development of biotechnological methods of environmental cleaning in recent years.

#### 4.2. Application of Biotrickling Filters to Remove $H_2S$ and $NH_3$

The method of air purification using biotrickling filters has been successfully tested in various technological combinations for both leachate and gas purification (Table 4). Cox et al. [48] tested  $H_2S$  and VOC removal in a biotrickling filters on a pilot scale. Odor removal ( $H_2S$ ) achieved an efficiency of 98%, but the simultaneous removal of VOCs achieved a much lower efficiency, which is influenced, among others, by drop in pH during  $H_2S$  oxidation. Based on the pilot scale studies, it was concluded that the simultaneous removal of VOCs and odors ( $H_2S$ ) is limited, which was not shown in previous laboratory scale studies [49]. Gabriel, Cox, and Deshusses [50] also investigated the removal of  $H_2S$  emitted from wastewater treatment plants under real conditions on a full industrial scale. The results showed a high  $H_2S$  removal efficiency despite the short gas contact time in the bioreactor caused by the high gas flows. These studies looked at only one compound— $H_2S$ .

Aroca et al. [51] conducted experimental studies on  $H_2S$  biodegradation using a laboratory scale biotrickling filter. They investigated the ability to remove  $H_2S$  using two different bacterial strains (*Thiobacillusthioparus* and *Acidithiobacillusthiooxidans*), for different pH values and different concentrations of  $H_2S$  in the inlet gas. The efficiency of  $H_2S$  removal was compared for different concentrations at the inlet to the bioreactor and different contact times—better efficiency of  $H_2S$  removal was noted—nearly 100%—for higher concentrations of  $H_2S$  at the inlet to the reactor —4600 ppmv and 120 s residence time and 982 ppmv and 45 s residence time, than at the lower concentrations when the  $H_2S$  removal efficiency was 47%.

Ramirez et al. [52] also investigated the removal of  $H_2S$  from gases in a Trickle Bed Bioreactor. The research was carried out in stable laboratory conditions on a bench-scale. The  $H_2S$  removal efficiency was 98–99%.

Very broadly, Kasperczyk et al. described the use of Compact Trickle Bed Bioreactors to purify gases from VOCs and odors of various origins. Contaminated gases supplied to the reactor, which are the main source of carbon for bacteria, are absorbed into the liquid phase, and then diffuse into the bacterial biofilm inhabiting the reactor bed. In bacterial biofilm as a result of the metabolic activity of microorganisms, they are transformed into simple products such as water and carbon dioxide [20]. Nutrients needed by microorganisms for proper development are delivered in the form of a solution of mineral salts along with the liquid recirculated in the reactor, which constantly moistens the surface of the bed. An important advantage is the ability to control the conditions in the reactor, such as maintaining the appropriate pH, the composition of mineral salts, which ensure good conditions for the development of microorganisms, and temperature. Moreover, Compact Trickle Bed Bioreactors do not generate additional waste in the form of secondary pollutants,



and are also a relatively inexpensive technology, which is conditioned by their operation at ambient temperature and atmospheric pressure [53]. Figure 5 shows a full-scale industrial Compact Trickle Bed Bioreactors.



**Figure 5.** Compact Trickle Bed Bioreactors in full industrial scale (Compact Trickle Bed Bioreactors, Manufacturer: Ekoinwentyka LTD, Ruda Śląska, Poland), Reproduced from [54], Industrial varnishing: 2020.

The latest published results of Kasperczyk et al. [20] presented the removal of VOC and  $H_2S$  emitted by a sewage treatment plant with the use of a Compact Trickle Bed Bioreactor. The experiment was conducted on a semi-industrial scale, in a wastewater treatment plant. The  $H_2S$  removal efficiency at about 200 ppm concentration on inlet, was over 97%. During the experiment, jumps in  $H_2S$  concentrations from 400 to 600 ppm were noted, which resulted in poisoning the bioreactor. However, after  $H_2S$  concentrations were restored to normal, stable bioreactor operation was achieved within 3 h. Kasperczyk et al. [55] also investigated the biodegradation of a mixture of  $H_2S$  and VOC from copper mines. The research was carried out in a Compact Trickle Bed Bioreactor, on a semi-industrial scale, in a copper mine, 1000 m underground. The bioreactor was filled with polyethylene rings. The efficiency of  $H_2S$  removal was at the level of 80–99%—when the concentration of  $H_2S$  was below 38 ppm, while when jumps in  $H_2S$  concentrations of 40–60 ppm were noted, the efficiency of  $H_2S$  removal decreased to 60–80%.

Sun et al. [56] examined a biotrickling purification filter for the treatment of  $H_2S$  from a municipal wastewater treatment plant. In the research, the culture of microorganisms was excessive sludge, and the filling of the filter was made of polypropylene rings. It has been investigated that in the inoculums which was vaccinated with biotrickling filter there were such microorganism as *Pseudomonas* and *Thiobacillus*. The average  $H_2S$  removal efficiency was 91.8%. In addition Sun et al. [57] also investigated the removal of hydrogen sulfide and volatile sulfur compounds using a two-stage biotrickling system containing acid- biotrickling filter and neutral- biotrickling filter. The contaminated gas came from wastewater treatment plant. Biotrickling filters was filled with polypropylene rings. The

microorganisms most abundantly present in the biotrickling filter system were identified: *Acidithiobacillus* and *Metallibacterium*. The H<sub>2</sub>S biodegradation efficiency was 86.1%.

Chen et al. [58] tested the biodegradation efficiency of H<sub>2</sub>S in biotrickling filter in a pilot scale. The contaminated gas came from the sewage lift station. The biotrickling filter was filled with bamboo charcoal and inoculated with activated sludge from the wastewater treatment plant. During the research the removal rate was 99% with an inlet H<sub>2</sub>S concentration of 5–20 ppmv.

Most of the scientific reports analyzing the use of the method of biotrickling filters for odor removal concern the removal of only H<sub>2</sub>S—considered to be the most persistent representative of odors. There are also many publications on the simultaneous removal of H<sub>2</sub>S and VOCs as components of odors. An equally persistent and harmful odor compound emitted by sewage treatment plants is ammonia NH<sub>3</sub>.

Sakuma et al. [59] investigated the NH<sub>3</sub> removal from polluted air in a system consisting of a biotrickling filter, a denitrification reactor and a leachate treatment reactor (to prevent recycle of the effluent into the biotrickling filter). Composite balls made of ceramics and bovine bones were used as reactor packing. The biotrickling filter and denitrification reactor were inoculated with activated sludge from the wastewater treatment plant. NH<sub>3</sub> absorption and nitrification took place in the biotrickling filter, while nitrates and nitrites were removed in the denitrification bioreactor. Then the excess of dissolved COD and NH<sub>3</sub> was treated in the last reactor. NH<sub>3</sub> was removed effectively, because in the first 15 days of operation the ammonia removal efficiency was 92–96%, while in the further stage of the experiment—after 21 days—the ammonia degradation efficiency did not drop below 96%, reaching 100% in several times.

While Moussavii et al. [60] investigated the removal of NH<sub>3</sub> in a biotrickling filter that developed a simultaneous nitrification/denitrification process. The bioreactor was filled with polyurethane foam, while the desired concentration of NH<sub>3</sub> flowing into the reactor was obtained by adjusting the air and NH<sub>3</sub> streams by trial and error. The results showed that this bioreactor would be able to completely remove 100 ppm NH<sub>3</sub> from the polluted gas with a 98.4% efficiency.

Huan et al. [61] investigated the efficiency of removing both H<sub>2</sub>S and NH<sub>3</sub> using a semi-pilot biological trickling filter reactor. As a filling of the biotrickling filter polyhedral spheres were used and it was inoculated with domesticated activated sludge. Microbiological analysis showed the presence of such microorganisms as *Dokdonella*, *Ferruginibacter*, *Nitrosomonas*, and *Thiobacillus*. The studies were conducted for 61 days and the removal rate of H<sub>2</sub>S was 98.25% and NH<sub>3</sub> was 88.55%.

Ying et al. [62] tested the ability of H<sub>2</sub>S and NH<sub>3</sub> biodegradation in a laboratory scale biotrickling filter, packed with porcelain Raschig rings and ceramsite. The maximum degree of H<sub>2</sub>S and NH<sub>3</sub> removal was over 99%.

Liu et al. [63] conducted research on integrated reactors in full-scale to determine the degree of odor removal (mainly H<sub>2</sub>S and NH<sub>3</sub>), VOC and bioaerosols simultaneously. The polluted air used for the study came from the sludge dewatering room in wastewater treatment plant. The average biodegradation efficiency of the odors was 98.5%, with a flow rate of 5760 m<sup>3</sup>/h, while the concentration of odors in the polluted air was recorded: H<sub>2</sub>S from 0.95 to 41.26 mg/m<sup>3</sup> and NH<sub>3</sub> from 0.91 to 21.37 mg/m<sup>3</sup>.

**Table 4.** Examples of the use of biological degradation methods to remove H<sub>2</sub>S and NH<sub>3</sub>.

Type of Odor	Method of Biological Biodegradation	Type of Microorganism/Bacterial Strain	Parameters (Type of Filling, T, pH)	Efficiency	References
H <sub>2</sub> S	Biofilters	<i>Thiobacillus thioautotrophicus</i> (H <sub>2</sub> S), <i>Nitrosomonas europaea</i> (NH <sub>3</sub> )	30 °C Ca-alginate beads	95%	[37]
		Sulfur Oxidizing Bacteria and microorganisms from compost	Compost pH = 7.5	95%	[41]
		<i>Bacillus</i> sp., <i>Pseudomonas</i> sp., <i>Xanthomonadaceae</i> sp.	Peat	99%	[42]
		<i>Acidithiobacillus thiooxidans</i>	Wood chips	75 ± 13% to 97%	[44]
		Sulfur oxidizing bacteria	Acid resistant polyethylene packing material—AMB Biomedial Bioballs	91.96%	[45]
		-	Pine bark	94%	[46]
		Activated sludge	Pieces of Poly Vinyl Chloride with compost	84–99%	[6]
	Biotrickling filters	Raw influent water from plant (Hyperion treatment plant)	7 layers of a PVC COOLdek™ <sup>tm</sup> Munsters	98%	[48]
		Heterotrophs, yeast, fungi, autotrophic sulfur-oxidizers	Pall rings, I biotrickling filter pH = 4.5, II biotrickling filter pH = 7	~100%	[49]
		Primary and secondary sludge from Orange County Sanitation District	Polyurethane foam, T = 18–24 °C	~98%	[50]
		<i>Thiobacillus thioautotrophicus</i> , <i>Acidithiobacillus thiooxidans</i>	Volcanic stones, polypropylene rings, polyvinylchlorure, pH = 5.5–7	100%	[51]
		<i>Acidithiobacillus thiooxidans</i>	Polyurethane foam	98–99%	[52]
		<i>Pseudomonas fluorescens</i> , <i>Thiobacillus</i> sp.	Polyethylene rings, T = ~30 °C, pH = 5.5–7.5	97%	[20]
		Bacterial strains	Polyethylene rings, pH = 5–7.5, T = ~30 °C	80–99%; 60–80%	[55]
		<i>Pseudomonas</i> sp., <i>Thiobacillus</i> sp.	Polypropylene rings	91.8%	[56]
		<i>Acidithiobacillus</i> sp., <i>Metallibacterium</i> sp.	Polypropylene rings	86.1%	[57]
		Activated sludge from Wastewater Treatment Plant (WWTP)	Bamboo charcoal	99%	[58]
		<i>Dokdonella</i> sp., <i>Ferruginibacter</i> sp., <i>Nitrosomonas</i> sp. and <i>Thiobacillus</i> sp.	Polyhedral spheres	98.25%	[58]
		<i>Acidithiobacillus</i> sp., <i>Thiobacillus</i> sp.	Raschig rings and ceramsite	99%	[62]

Table 4. Cont.

Type of Odor	Method of Biological Biodegradation	Type of Microorganism/Bacterial Strain	Parameters (Type of Filling, T, pH)	Efficiency	References
NH <sub>3</sub>	Biofilters	<i>Thiobacillus thioautotrophicus</i> (H <sub>2</sub> S), <i>Nitrosomonas europaea</i> (NH <sub>3</sub> )	30 °C Ca-alginate beads	95%	[37]
		Activated sludge from Wastewater Treatment Plant (WWTP)	Organic: compost, bark, peat Inorganic: perlite	100%	[38]
		-	Fibrous peat, coarse peat, wheat straw, composts, horse manure	89%	[39]
		Compost		~96%	[40]
		Nitrate oxidizing bacteria ( <i>Nitrosomonas</i> sp., <i>Nitrobacter</i> sp.)—from cattle manure	Cattle manure, rice husk, gravel as a supporting media, 32–39 °C	61.5%—for a bed 20 cm deep, 71.45%—for a bed 40 cm deep	[43]
		-	Acid resistant polyethylene packing material—AMB BiomediaBioballs	100%	[45]
		-	Pine bark	91%	[46]
		Activated sludge	Pieces of Poly Vinyl Chloride with compost	88–99.6%	[6]
		Micromycetes: <i>Acremoniumstrictum</i> , <i>Aspergillus versicolor</i> , <i>Aureobasidium pullulans</i> , <i>Cladosporium</i> sp., <i>Penicillium</i> sp., <i>Gliocladiumviride</i> , <i>Stachybotrys</i> sp., <i>Cladosporiumherbarum</i> ; Yeast: <i>Exophiala</i> sp., <i>Aureobasidiumpullulans</i> ; Bacteria: <i>Rhodococcus</i> sp., <i>Bacillus subtilis</i>	Straight and wavy lamellar plates (hydrophilic synthetic texture), pH = 7, T = 24–32 °C	84.2%–87%	[47]
	Biotricklingfilters	Activated sludge from Wastewater Treatment Plant (WWTP)	Composite balls made of ceramics and bovine bones	92–100%	[59]
		Autotrophic and heterotrophic bacteria	Polyurethane foam	98.4%	[60]
		<i>Dokdonella</i> sp., <i>Ferruginibacter</i> sp., <i>Nitrosomonas</i> sp., <i>Thiobacillus</i> sp.	Polyhedral spheres	88.55%	[61]
		<i>Acidithiobacillus</i> sp.,	Raschig rings and ceramicsite	99%	[62]
		<i>Thiobacillus</i> sp., Ammonia Oxidizing Bacteria, Nitrite Oxidizing Bacteria	Activated carbon fiber	98.5%	[63]

## 5. Directions of Future Research

A review of recent research work underlines the need to use and implement modern, ecological, and cheap tools of biotechnology for odor removal into industrial practice. New physicochemical methods used for odor removal in wastewater treatment plants, such as ozone, UV rays, or non-thermal plasma, despite their high odor removal efficiency, are much more expensive than biological methods for the degradation of odors, and some of them generate emissions of secondary pollutants (including ozone) [23]. Higher operating costs of the above mentioned methods result, from the necessity to supply electricity and its high consumption. The non-thermal plasma method is used to degrade odors occurring in very small amounts (below  $100 \text{ mg/m}^3$ ), when the concentrations of pollutants are higher, the increase in power of the device generates very high costs [23]. This technology causes also the formation of secondary pollutants, which in turn is associated with the need to combine at least two techniques of gas purification, and the resulting significant increase in financial expenditure [64,65]. Hołub et al. [64] achieved 90% odor reduction, but it was noted that not all compounds were removed—aldehydes and other hydrocarbons were removed to a small extent. However, the main advantage of this method is the small size of the installation [23].

The intensive development of modern ecological and innovative biotechnologies, and the steadily increasing amount of research over the last decade testify to the continuous development of this topic, and the focus of research on the possibility of potential implementation to full industrial scale. A review of the literature has shown that biological methods of odor removal give high effects of bio-purification of the air, up to 95–99%; moreover, their advantage is manifested primarily in the economic aspect as well as in terms of environmental friendliness. Therefore, it is necessary to develop and intensify processes based on biological methods of odor removal, in order to implement them to the full industrial scale. An example of the development of research on modern biotechnologies for odor removal is the Compact Trickle Bed Bioreactors, whose results and previous implementation indicate their potential versatility. Therefore, it is necessary to develop research in this area by testing the applicability of such technologies in various industries and in the municipal sector, e.g., wastewater treatment plants or landfills. Furthermore, the impact of extreme conditions and sudden changes of pollutant concentrations on the efficiency of air purification should be investigated, as most laboratory tests do not consider extreme overload conditions. It should be checked what parameters influence the inhibition or intensification of the efficiency of Compact Trickle Bed Bioreactors, e.g., what is the effect of a change in the composition of odors, which depends, among others, on atmospheric conditions and composition of wastewater delivered to the wastewater treatment plant, and how the activity and composition of microorganisms and their adaptation to the removal of variable concentration and composition of odors changes. Therefore, it seems important to study the influence of the parameters of the bio-treatment process and external conditions, in fact often deviating from stable laboratory conditions. In order to implement innovative biotechnological methods of odor removal into industrial practice, it would be necessary to carry out research in real conditions in industrial plants, waste management plants, municipal, and industrial wastewater treatment plants.

## 6. Summary

Among biological odor removal methods, the most commonly used so far is the use of biofilters, which are effective for low concentrations of pollutants in the treated gas, can be used for large streams of polluted gases, are easy to build and operate, and are also relatively cheap. Currently, the method of gas purification in biotrickling filters turns out to be competitive to biofilters. It is a relatively new technology, whose great advantage, is the high efficiency of the biodegradation of pollutants, usually reaching 95–99%, and the ability to control the conditions in the reactor, such as maintaining an appropriate pH, mineral composition, which ensure good environment for the development of microorganisms. In addition, biotrickling filters do not generate additional waste in the form of secondary



pollutants, and are a technology that does not generate further operating costs, which means that there is no need to regenerate the bed, as is the case with biofilters, or to utilize excessive and harmful leachate, such as it is in the case of bioscrubbers. In view of these advantages, biotrickling filters are increasingly used in industry and are the subject of numerous studies.

The studies and results obtained so far show that the research conducted on a laboratory scale does not reflect the actual conditions at a full industrial scale. Significantly higher flows in the gaseous phase, periodic changes, and sudden increases in pollutant concentrations, and the need to maintain an appropriate pH under such conditions are aspects that are not taken into account in laboratory tests because they are difficult to predict. Most of the odor and VOC removal tests performed so far, have been conducted on a laboratory, pilot or semi-industrial scale, while there are only few materials showing the implementation of biotrickling filter technology to a full industrial scale. The results show that the Compact Trickle Bed Bioreactor are a competitive method compared to other odor removal methods. Therefore it is necessary to strive to implement innovative biotechnologies to full industrial scale, which must be preceded by research carried out in real-life conditions at industrial plants and the municipal sector.

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