Odor Removal Technologies

Subjects: Engineering, Environmental
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Comparing various methods of odor removal, undoubtedly biological methods of pollution degrada-tion have an advantage over others—chemical and physical. This advantage is manifestedmainly in ecological and economic terms. The possibility of using biological methods to remove H2S and NH3, 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 advan-tageous 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.

biodegradation odors H2S NH3

1. 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 [1][2]. 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 [3].

1.1. 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 [4]. 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+}) [5]. 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 [6]. Chemical absorption in many cases is used as a pre-treatment method of emitted gases characterized by a high concentration of odors [1].

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 [5]. Adsorbents used for deodorizing the emitted gases are in the form of powder (8–80 µm), granules (200 µm 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 [6].

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^3, 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 [7]. For this purpose, a common practice is to combine combustion processes with adsorption [8]. 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 [6][5].

1.2. 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 [3][9]. 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 [10][11][12].

2. Effectiveness of H_2S and 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 [1]. Among the biological methods of air purification, biofilters are relatively simple and the longest used methods; hence, also, the best known [13], there are many literature reports confirming the use of biofilters for odor removal.

2.1.Application of Biofilters to Remove H_2S and NH_3

Chung et al. [14] studied the degradation of H_2S and NH_3 using a biofilter. Impurities in the form of H_2S and NH_3 were administered in various proportions. Their biodegradation efficiency was on the average level of over 95%, regardless of the H_2S and 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 H_2S can inhibit NH_3 removal, while NH_3 concentration has only a negligible effect on H_2S removal.

Whereas Choi et al. [15] tested the 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—pearlite (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. [16] also investigated the biodegradation efficiency of NH_3 with an open biofilter, but in this case the source of 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^2. The efficiency of degradation of pollutants in the biofilter was tested in fivephases, in the initial phase of the experiment (after fivedays 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 NH_3 removal during the experiment.

Pagans et al. [17] also investigated the effectiveness of 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 NH_3 biodegradation was observed when its concentration at the inlet to the biofilter increased to over 2000 mg/m^3.

While Rehman et al. [18] investigated the performance of biofilters intended for H_2S 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 H_2S with the subsequent phases. It was found that the biofilter most effectively removed H_2S 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 H_2S concentration of 50 ppm.

In turn, the aim of the research by Omri et al. [19] was to investigate the degree of H_2S 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 H_2S in the inlet gases ranged from 200 to 1300 mg/m^3, while the efficiency of H_2S removal reached 99%.

Kavyashree et al. [20] 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 500–700µg/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 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. [21] investigated the effectiveness of removing H_2S present in synthetic biogas using a biofilter filled with sawdust. The tests were carried out for 37 days, with an average H_2S removal efficiency of 75 ± 13%, while the maximum efficiency was 97%.

Rabbani et al. [22] investigated the effectiveness of H_2S and 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 NH_3 from gases. Thus, in the gases entering the biofilter, only H_2S was present, which as a result of biological oxidation formed H_2SO_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 H_2S and to generate an appropriate amount of H_2SO_4 accumulated at the bottom of the biofilter to remove NH_3 in stage II. In turn, in the second stage of the experiment, gases containing a mixture of H_2S and NH_3were introduced into the same biofilter, this stage lasted sevenweeks. The average H_2S removal efficiency was 91.96% and NH_3 100%. At the bottom of the biofilter, a small amount of effluent (0.2 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 [23] was the degradation of various odor compounds, including H_2S and NH_3, emitted by the wastewater treatment plant with the use of a biofilter. The concentrations of H_2S and NH_3 at the inlet to the biofilter were 154 µg/m^3 and 1799 µg/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. [24] compared the removal efficiency of odors consisting mainly of H_2S and 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 NH_3, while the H_2S 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 (H_2S and NH_3) in a chemical scrubber was over 97%, while in the biofilter it was 92% for H_2S and 99.5% for 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. [25] 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.

2.2.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. [26] 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 [27]. Gabriel, Cox, and Deshusses [28] 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. [29] 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 −4600ppmv and 120 s residence time and 982 ppmv and 45 s residence time, than at the lower concentrations when the H_2Sremoval efficiency was 47%.

Ramirez et al. [30] 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 [3]. 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 [31]. 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, Poland), Reproduced from [32], Industrial varnishing: 2020.

The latest published results of Kasperczyk et al. [3] 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_2Sremoval 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. [33] 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. [34]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. [35] 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_2S biodegradation efficiency was 86.1%.

Chen et al. [36] tested the biodegradation efficiency of H_2S 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_2S 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_2S—considered to be the most persistent representative of odors. There are also many publications on the simultaneous removal of H_2S and VOCs as components of odors. An equally persistent and harmful odor compound emitted by sewage treatment plants is ammonia NH_3.

Sakuma et al. [37] investigated the NH_3 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_3 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_3 was treated in the last reactor. $NH_3$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. [38] investigated the removal of NH_3 in a biotrickling filter that developed a simultaneous nitrification/denitrification process. The bioreactor was filled with polyurethane foam, while the desired concentration of NH_3 flowing into the reactor was obtained by adjusting the air and NH_3 streams by trial and error. The results showed that this bioreactor would be able to completely remove 100ppmNH_3 from the polluted gas with a 98.4% efficiency.

Huan et al. [39] investigated the efficiency of removing both H_2S and NH_3 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_2S was 98.25% and NH_3 was 88.55%.

Ying et al. [40] tested the ability of H_2S and NH_3 biodegradation in a laboratory scale biotrickling filter, packed with porcelain Raschig rings and ceramsite. The maximum degree of H_2S and NH_3 removal was over 99%.

Liu et al. [41] conducted research on integrated reactors in full-scale to determine the degree of odor removal (mainly H_2S and NH_3), 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^3/h, while the concentration of odors in the polluted air was recorded: H_2S from 0.95 to 41.26 mg/m^3 and NH_3 from 0.91 to 21.37 mg/m^3.

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