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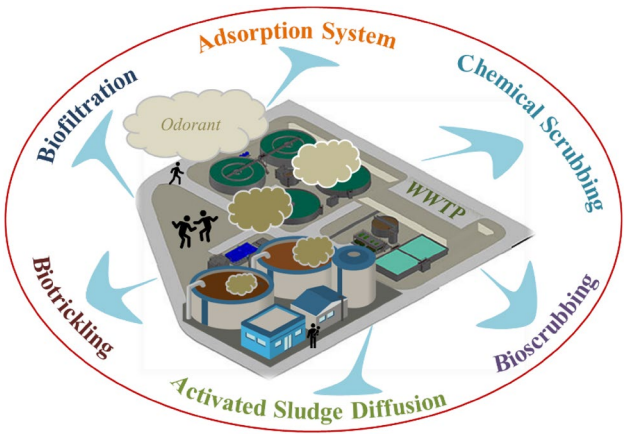
Current Status and Outlook of Odor Removal Technologies in Wastewater Treatment Plant

Baiming Ren · Yaqian Zhao · Nathalie Lyczko · Ange Nzihou

Abstract

The ever increasing public complaints arising from wastewater treatment plants (WWTPs) grow since sewerage treatment has been associated with nauseous odorous on account of the anaerobic decomposition process and emission. Various physical/chemical and/or biological methods were used for abating odours worldwide. Thus, an updated comprehensive review for the WWTP odor abatement technologies is urgently required. This study reviews the new achievements of odor abatement technologies (adsorption, chemical scrubbing, biofiltration, biotrickling, bioscrubbing, activated sludge diffusion) in WWTPs and then identifies a new aspect for the future studies. Overall, hybrid technologies (physical/chemical + biotechnologies) attract increasing attention since their highly reliable removal efficiency for various odorants, however, the high costs for investment and O&M (operation & maintenance) of the adsorption part and the complexity and variability of odorants are still the major challenging for wide engineering application and technological innovation. Thus, developing the cost-effective, environmentally friendly odor control technologies, like using the alum sludge (waterworks residue) based adsorbents/media, in terms of using "waste" for waste treatment, could be a highly promising prospect.

Graphical Abstract



Keywords Adsorption · Alum sludge · Hydrogen sulfide (H_2S) · Review · Wastewater treatment plants (WWTPs) · Odor treatment/abatement

Statement of Novelty

This study provided the updated information for the WWTPs odor management as well as to identify the promising areas of further research and development. Given that alum sludge (waterworks residues) could be the promising low-cost

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adsorbent for odor removal even to the media in the biofilters and biotrickling due to its inherent characteristics, developing the low-cost adsorbents/media for WWTPs odor treatment could be a vital innovative aspect of the environmentally friendly development as well as the "Blue Economics". It not only opens the new prospects for the waterworks sludge management but also fits in the circular approach of using a waste for waste gas control.

Introduction

In recent years, the release of unpleasant odours from wastewater treatment plants (WWTPs) has attracted broad attention, especially among the high population density countries. For technological reasons, WWTPs always occupy a large surface area, ranging from several to more than a dozen hectares and, as a result, are often considered responsible for odor emissions [1]. They not only have a negative impact on the local population but also represent a significant contribution to photochemical smog formation and particulate secondary contaminant emission [2–4]. Odorous compounds in sewage mainly originate from two processes: anaerobic decomposition of biodegradable materials in the wastewater or direct emission of specific chemicals with wastewater discharges [5, 6]. Furthermore, odours emanating from WWTPs are composed of a mixture of various chemical compounds including ammonia (NH_4), hydrogen sulfide (H_2S) and butanone etc., 78 kinds of main odor-producing compounds relating to wastewater collection and treatment facilities have been reported [7–10], such as various volatile organic compounds (VOCs), indoles, skatoles, mercaptans etc. [11]. Stuetz and Frechen [12] had summarized a range of odorous emissions from WWTPs (Table 1). Significantly, H_2S is considered the most important cause for both odor emission and corrosion in wastewater collection and treatment facilities [12]. H_2S is a toxic, flammable, and colourless gas with an unpleasant smell, similar to that of rotten eggs. It can be smelled at low concentrations (about 0.5 ppb) [13]. Based on the European, American, and Chinese standards, the maximum permissible H_2S emission through a chimney is 5–10 ppm [7].

It is worth noting that H_2S is a key element in sulphide chemical species according to the equilibrium:

$$H_2S\,(g) \overset{Water-air}{\Leftrightarrow} H_2S\,(aq) \overset{pK_{a1}=7.0}{\Leftrightarrow} HS^- \overset{pK_{a2}=14}{\Leftrightarrow} S^{2-} \quad (1)$$

where, the equilibrium constants K_{a1}, K_{a2}, determine the ratio between the concentrations, C, at equilibrium:

$$K_{a1} = \frac{C_{H^+} \times C_{HS^-}}{C_{H_2S(aq)}} \quad (2)$$

$$K_{a2} = \frac{C_{H^+} \times C_{S^{2-}}}{C_{HS^-}} \quad (3)$$

The H_2S release to the atmosphere is thereby strongly dependent on the pH (C_{H+} in Eqs. (2) and (3)) [12]. Simultaneously, another substance that produces odor in WWTPs is ammonia, which is caused by bacterial decomposition of urea produced in the sewage networks. Ammonia possesses a low evaporation temperature, it is therefore easily evaporated and leads to the release of wastewater odours in the environment [15–19].

Consequently, a careful management of WWTP odor is required to avoid the annoyance and to meet the strict regulations. Until now, odor treatment technologies can be classified into physical/chemical and biological methods. Adsorption and chemical scrubbers are among the physical/chemical methods, while biofilters, biotrickling filters, bioscrubbers, and activated sludge diffusion reactors etc. are biological approaches for odor control. Physical/chemical technologies have been broadly implemented since their rapid start-up, low empty bed residence time (EBRT) and extensive experience in design and operation [20–24]. These techniques are often based on the transfer of odorants from the gas emission to either a solid (adsorption) or liquid (absorption) phase. These pollutants can be further

Table 1 A range of odorous emissions from WWTPs. Reproduced with permission from [12, 14]

Compound	Odor description	Chemical formula	Odor threshold (ppb)
Ammonia	Sharp, pungent	NH_4	130–15300
Butanone	Sweet, minty, green apple	$CH_3C(O)CH_2CH_3$	270
Dimethyl sulfide	Decayed vegetables	$(CH_3)_2S$	0.12–0.4
Geosmin	Earthy, musty	$C_{12}H_{22}O$	4
Hydrogen sulfide	Rotten eggs	H_2S	0.5
Indole	Fecal, repulsive	C_8H_7N	0.3–1.4
Methyl mercaptan	Decayed cabbage, garlic	CH_4S	0.0014–18
Skatole	Fecal	C_9H_9N	0.006
Sulfur dioxide	Pungent, acidic	SO_2	9
Thiophenol	Garlic, stench	C_6H_6S	0.064

transformed into by-products according to their reactivity with the chemicals used. However, in the last decades biological systems have been increasingly implemented due to their ability to efficiently treat malodorous emissions at lower operating costs. The main merits of biotechnologies compared to their physical/chemical counterparts derive from their low generation of secondary wastes and low demand of resources, such as chemicals or adsorbent media. On the other hand, biological processes often require larger EBRT (2–120 vs. 1–5 s) and associated footprint than physical/chemical alternatives at similar odor removal efficiencies [25–28].

Although various technologies have been widely reported in the past [2, 29], few comprehensive review was made to analyse and compare complete odor treatment technologies in spite of the recent report of [2, 7, 30]. Moreover, a cost-effective and environmentally friendly abatement of odours is still urgently needed as the increasingly public concerns as well as the stringent legislation. Thus, the aim of this study is trying to conduct a comprehensive review on the current six main odor treatment technologies emerged in the last 30 years. It is expected to provide an updated guideline for the WWTPs odor management as well as to identify the promising areas of further research and development.

Odor Treatment Technologies in WWTPs

Physical/Chemical Technologies

Physical/chemical technologies consist of two types of reactors, namely adsorption systems and chemical scrubbing, as illustrated in Fig. 1. Either of them is commonly used in practice for odor removal in WWTPs because of their low EBRT, extensive experience in design and operation, and rapid start-up, etc. [2].

Adsorption Systems

Adsorption systems generally consist of static beds of granular materials in vertical cylindrical columns (Fig. 1). Accordingly, the odorous air steam enters the column, process in the direction of air flow, and continues until odor "break through" at the exit end. Its efficiency is severely limited by the high moisture content prevailing in WWTPs malodors emissions. Moreover, capacity, temperature etc. are the vital characteristics of a robustness adsorption system [31]. Several sorbents have been studied, including fly ash, carbon, activated carbon, polymers, carbon-coated polymers, ceramics, micro- and mesoporous materials, metal organic frameworks, natural zeolites, and synthetic zeolites [32]. It's worth noting that specific surface area, pore structure together with surface chemical functional groups are three crucial factors of adsorbents, which would directly determine their performance on odorants adsorption [33]. However, it is difficult to find an adsorbent with all the features of an excellent adsorbent, some of the adsorbent properties must be compromised, such as removal capacity, regeneration, or cost impact [34].

Activated carbon (AC) is widely used as adsorbent for odor control in WWTPs. In the initial stage, AC was impregnated with caustics (NaOH or KOH) for H_2S control. Both NaOH and KOH reacted with atmospheric CO_2 to form the corresponding carbonates, thus facilitating H_2S removal. Other impregnated sorbents for H_2S removal are carbons impregnated with heavy metal salts such as copper sulphate or lead acetate. These media are usually classified as hazardous materials because of their content of heavy

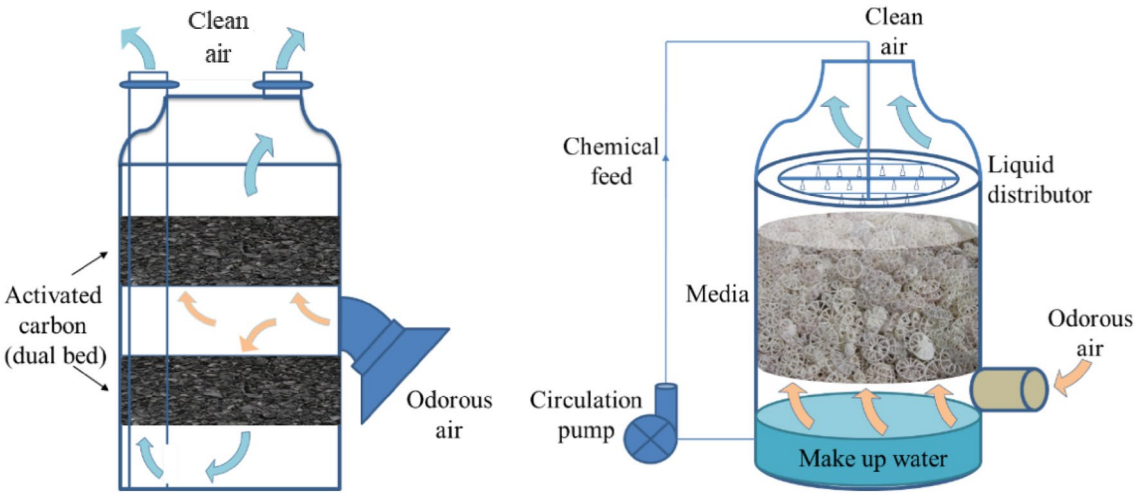


Fig. 1 Physical/chemical abatement technologies: adsorption system (left), chemical scrubber (right) (Reproduced with permission from [12])

metals. Among non-carbonaceous sorbents, activated alumina impregnated with potassium permanganate has also been used for oxidative removal of H_2S [12]. However, it has been demonstrated by Bandosz et al. [35] that unmodified carbon can provide enough capacity to efficiency removal of H_2S from effluent gas in WWTPs. It is worth noting that AC adsorption presented the highest operating costs (0.45 € per 1000 m^3 treated) among the odor abatement technologies [2]. This is because the AC filtration presented the highest annual packed-bed-material requirements and the 6 months replacement short life span as well as the needs for specific management procedures (regeneration or disposal as hazardous waste). Thus, although lots of efforts have been made in terms of the development of different sorbents in WWTPs odor treatment, the high cost of operation continues to be their major drawbacks for wide engineering application of the AC system. As such, generation of high performance sewage sludge-based ACs is continuously studied worldwide, while Yang et al. [36] reported a new generated sewage sludge-based ACs, which possesses excellent adsorption capacity (259.9 mg/g) of methyl mercaptan (CH_3SH). Additionally, Aziz and Kim [34] investigated the removal of VOCs (benzene, toluene, ethylbenzene, and p-xylene) by Na-ZSM-5 and H-ZSM-5 (a synthesized material have a high ratios of SiO_2/Al_2O_3). NaZSM-5 rapidly removed 75% of toluene and ethylbenzene and 50% of Benzene, but xylene was not efficiently removed. On the other hand, HZSM-5 could remove all of the VOCs. Furthermore, Sempere et al. [37] reported that a AC adsorption combined with biotechnologies to form the hybrid technology becomes a popular odor control approach in WWTPs. Overall, adsorption-based systems provide an excellent performance in the treatment of highly hydrophobic odorants (90–99%) [32, 38].

Several mechanisms have been proposed for the adsorption reactions. However, it is not yet fully understood. Generally, the mechanism of odor adsorption consists of seven steps as follows: (i) transport of the gas from the bulk of a mixture to a solid particle, (ii) transport of the reactants in the pores of the adsorbent particles to an active site, (iii) adsorption of the reactants to the active site via Van der Waals forces, (iv) reaction of reactants to form an adsorbed product, (v) desorption of the product from the active site, (vi) transport of the products in the pores of the catalytic particle out of the particle, (vii) and transport of the products from the particle to the bulk of the mixture [39].

Chemical Odor Scrubbing

Chemical scrubbers (CS) are among the most commonly employed abatement techniques in WWTPs due to the extensive experience and high robustness as well as the short gas retention time (as low as 1–2.5 s) [40]. Various types of CS include the counter-current scrubber, cross-flow scrubber, and venture scrubber have been reported [7]. The most common configuration (Fig. 1) is a vertical shell with gas flow going up through packing and the liquid solution (depending on the target compounds) going down. Liquid solution is usually circulated over the packing by pumping from a collection sump in the bottom of the tower, while chemicals are added either in the sump or in the recirculation piping.

For best performance, a multi-stage scrubber is often used. Yang and Chen [41] investigated the oxidation of 150 ppm nitric oxide with a two-stage chemical scrubber using the dc corona as an alternative for one of the scrubbing chemicals. Bandyopadhyay and Biswas [42] studied the scrubbing of sulfur dioxide in a two-stage hybrid scrubber using water and dilute sodium alkali. Chen et al. [43] reported a novel two-stage wet scrubbing for composting gases treatment, which consisted of acidic chlorination followed by alkaline sulfurization. However, a water/oil emulsion for VOCs removal has recently been developed by Hariz et al. [44], it is an alternative that reduces the number of towers and thus reduces the investment and operation costs. Significantly, thermal regeneration of the VOC-saturated oil was reported in the first time to reduce the solvent consumption and to reduce the impact on the environment.

Generally, since a portion of solution is continually wasted to remove the accumulated contaminants, CS required the large annual amounts of chemical reagents and higher quality (preferably softened) potable water. The purchase of chemicals accounts for the highest contribution (69%) to the CS operating cost and followed by the energy consumption (22%) as a result of the high liquid recycling rates [45]. Moreover, NaClO as the commonly used oxidant in CS is likely to form chlorinated by-products which are harmful for human health and to produce another pungent odor, hypochloric acid [46, 47]. Alfonsín et al. [27] also pointed out that CS presented the highest impacts in freshwater eutrophication, photochemical oxidant formation, human toxicity and ecotoxicity due to the use of large amounts of chemicals. H_2O_2 is a promising oxidant except for its instability in basic aqueous solution and the low efficiency of CH_3SH removal [48, 49]. The emerging oxidants of CS like peroxymonosulfate (PMS) has a similar structure with H_2O_2 but the oxidation–reduction potential is stronger than H_2O_2 [50]. However, Talaiekhozani et al. [7] pointed out that Venturi scrubbers can be a highly cost-efficient candidate for many industries, due to their simplicity of construction and usage, the ability to purify large volume of air in a short time and to simultaneously remove particles and gases. Significantly, Venturi scrubbers can even be distinguished from biological methods, such as biofilters, since it can quickly reach the maximum efficiency comparison with biofilters, while the alkaline substances, such as limewater, can convert sulfur into calcium sulfide and inhibit the reproduction of hydrogen sulfide etc. On the other hand,

CS showed the lowest geographic dependence worldwide, because chemicals, which constitute the main cost in these systems, were considered to be part of a global market and their prices did not depend on the geographical location [51]. Overall, despite the high removal efficiencies (> 99%) achieved for H_2S [50, 52], CS presents serious limitations in the elimination of hydrophobic VOCs (high Henry Law constants) as they are finally based on odorant transfer to an aqueous solution of oxidant. In addition, the hazardous nature of the chemical reagents employed and by-products generated represent a serious challenge to its supremacy in a world increasingly devoted to sustainable development.

Biotechnological Treatment

Biotechnologies, particularly hybrid technologies (physical/chemical + biotechnologies) become the main approach in the last decades for odor control, since their low-cost and environmentally friendly nature. To date, four types of reactors are commonly used in WWTPs [53]: biofilters (BF), biotrickling filters (BTF), bioscrubbers (BS) and activated sludge diffusion reactors (ASD), as illustrated in Fig. 2.

Biofiltration

BF is indisputably the most commonly employed biotechnology for odor treatment in WWTPs [29]. In a biofilter system (Fig. 2a), the humified odorant is forced through a packed bed (compost, peat, bark or a mixture of these) on which the microorganisms are attached as a biofilm [54–59]. The pollutants are sorbed by the filter material and degraded by the biofilm. However, to control the key parameters such as pH and moisture content within the packed bed, and to avoid the accumulation of inhibitory by-products are still the technical difficulties and limitations [37, 60, 61].

Performance data of a series of lab-scale BF and engineering applications are summarized in Table 2. An important advantage of biological treatment methods over physical and chemical technologies is that the biological processes can be conducted at moderate temperatures (10–40 °C) and atmospheric pressure. Moreover, microbial degradation processes are generally oxidative in nature and produce compounds such as carbon dioxide, water, sulfate and nitrate that are ecologically safe [54]. The microorganisms are the engine of the biotreatment process. Biofilms contain a mixture of bacteria, fungi, yeasts, ciliated protozoa, amoebae, nematodes and algae. Bacteria and fungi are the two dominant microorganisms groups in biofilters, however as bacteria populations (primary degraders) grow they can sustain yeasts and other fungi, algae and higher organisms such as protozoa, rotifers, nematodes, etc., the optimum pH for microorganisms is 7 [53].

By providing support for microbial growth, BF media are very often regarded as the key issue to determine removal

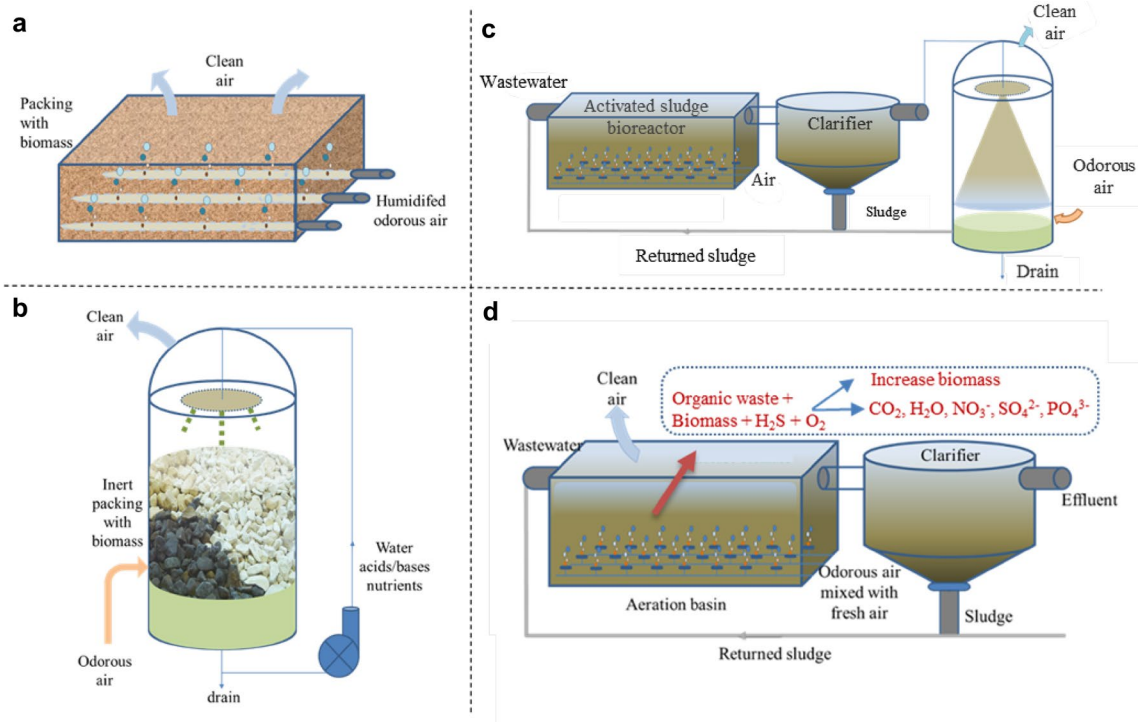


Fig. 2 Biotechnological treatment: **a** open biofilter; **b** biotrickling filter; **c** bioscrubber; **d** activated sludge diffusion (Reproduced with permission from [12])

Table 2 Performance of BF in lab and on-site systems

Reference/location[a]	Media	Parameters				Odorant	Concentration (mg/m^3)	RE (%)
		EBRT (s)	pH	Temp. (°C)	Nutrients			
[69]	Perlite, polyurethane foam, compost, wood chips, straw	–	7–7.5	20	$NaNO_3$, KH_2PO_4, Na_2HPO_4, $MgSO_4$, $CaCl_2{\cdot}2H_2O$, $FeSO_4{\cdot}7H_2O$, $ZnSO_4{\cdot}H_2O$, $MnSO_4{\cdot}H_2O$, $CuSO_4{\cdot}5H_2O$, $CoCl_2{\cdot}6H_2O$, H_3BO_3, $Na_2MoO_7{\cdot}2H_2O$	Acetone, n-butanol methane ethylene ammonia	23.9 7.0 21.7 ppm 20.2 ppm 10–30 ppm	100 100 0 95 85–98
[75]	Pine bark, composted wood mulch	70–290	2.7–8.7	Room temp	K_2HPO_4 $(NH_4)_2SO_4$ Na_2CO_3	H_2S DMDS ethanethiol	3.8–7.6	100 95 82
[70]	Compost & woodchip	34	6.1–8.1	Room temp	None	Ammonia	40 ppm	35–63
[74]	Wood bark	1.6–3.1	–	11.3–30.8	None	Ammonia H_2S	0.25–75.00 ppm 0.05–8.00 ppm	95.2–97.9 95.8–100
Los Angeles, U.S	Compost, perlite, oyster shell	14–69	7–9	–	None	Benzene Xylenes Toluene Dichloro- Benzene H_2S Carbon disulfide, Methyl mercaptan Dimethyl sulfide Carbonyl sulfide	0.002–0.003 0.18–0.66 0.077–0.23 0.024–0.049 10–50 0.02–0.03 0.30–0.33 0.02–0.03 0.05–0.13	0–50 40–75 42–86 43–60 > 99 32–36 91–94 0–21 30–35
Ojai Valley, U.S	Lava rock	18–54	7.9–8.1	–	Nitrogen Phosphorous Potassium	MTBE Acetone Toluene Xylenes Dichloro- methane Chloro- form H_2S	1.8 1.6 2.3 1.3 3.5 0.3 0.01–42	20 80 60 40 30 15 > 90
Carson, U.S	Compost, wood chips, oyster shell, perlite	45–180	–	–	None	Benzene Toluene m,p-Xylene o-Xylene H_2S	3.0 4.0 1.1 0.4 13.9	83–95 88–97 88–93 88–91 > 99
Yarmouth, U.S	Compost, bark mulch, wood chips	45	–	Low temp	Chemical misting	α-Pinene β-Pinene Dimethyl sulfide DMDS D-Limonene Carbon disulfide Methyl mercaptan	675 ppb 345 0.02 0.16 70 0.01 0.006	100 100 100 100 97 100 100
Tampa, U.S	Top soil, peat, mulch	60	–	–	None	H_2S	7–120	100
Albany, U.S	–	150	–	–	–	H_2S Dimethyl sulfide Methyl mercaptan	200 8.8 22	100 21 66
Hillsborough, U.S	Pine bark	115	–	–	None	H_2S DMDS Carbon disulfide Methyl mercaptan	140 936 618 330	99.5 97 82 100

Table 2 (continued)

Reference/location[a]	Media	Parameters				Odorant	Concentration (mg/m^3)	RE (%)
		EBRT (s)	pH	Temp. (°C)	Nutrients			
Boca-Grande, U.S	Peat, wood chips, top soil	130	–	–	None	H_2S	140	100
Charlotte, U.S	Wood chips, compost, perlite, granular fill	111	–	–	–	Dimethyl sulfide Carbon disulfide	625 448	100 100
Fountain Valley, U.S	Two units with GAC and yard waste compost	17–70	1–2.7	–	–	Benzene Toluene Xylenes Dimethyl sulfide Chloroform TCE PCE Total gaseous nonmethane organics H_2S	0.01 0.1 0.08 0.07 0.06 0.01 0.37 26 ppm 4.3	36–93 24–99 96 35 11 82 98 99 99
Martinez, U.S	Wood chips, yard waste, compost, lime	38	–	–	–	H_2S Dimethyl sulfide DMDS Methyl mercaptan	0.11 0.03 0.01 0.054	95 68 41 90
Renton, U.S	Bark, topsoil, compost, peat, moss, oyster shells	40–60	–	–	–	Acetone H_2S Benzene Mercaptans Amines TCE PCE Chloroform	0.03–0.09 1.5–34 0.01–0.25 0.16–3.8 ppm 2.5–6 ppm 0.02–0.05 0.02–0.5 0.10–0.21	55 97 25 62 > 60 44 40 43

[a]Reproduced with permission from [27, 62, 63]

efficiencies and BF lifespan [13, 64]. Nontoxic organic or inorganic supports with high specific surface areas and porosities, good water retention capacities, high buffer capacities, and high nutrients content have provided the best odor removal performances. Organic media provide an extra C source necessary to maintain microbial activity, likely challenged by the extremely low C concentrations present at the biofilm-water/air interface [65–67]. On the other hand, inorganic materials such as ceramic, plastics, lava rock, and activated carbon provide an extra structural stability, which increases BF lifespan [68, 69].

Moisture content of the packing is the most critical parameter to control in BF. Indeed, many references listed in Table 2 mentioned system upsets causing excessive drying of the packed bed and declining performance. Although the relative humidity of the air undergoing treatment is often over 80% at WWTPs, the waste air is frequently humidified in packed towers before entering the BF [70].

As shown in Table 2, BFs at industrial applications are operated at EBRTs from 20 to 200 s. Removal of H_2S is generally between 90 and 100%, indicating the effectiveness of BF. On the other hand, removal of odorous compounds like dimethyl sulfide, DMDS, and methyl mercaptan is often instable, with reported removal efficiencies ranging from about 20 to 100% [71–74]. However, Jaber et al. [75] pointed out the accumulation of sulfuric acid, the most abundant product of the biological oxidation of sulfur compounds in the packing material caused the pH decreasing, led to a reduction of the elimination efficiencies of DMDS etc., while the microorganisms involved in H_2S degradation appeared active in a large pH range, from 3 to 9.

Because of the need to maintain low pressure drops across the packing bed and the high EBRT needed for efficient odor treatment, BF presented the highest land requirements [76]. This footprint was 7 and 25 times higher than that of the BTF and CS, respectively, and can limit the application of this biotechnology during plant upgrading in WWTPs when facing land limitations [40, 77, 78].

Biotrickling

To overcome the disadvantages of BF, more sophisticated filtration equipment called biotrickling has been developed. In BTF or fixed-film bioscrubber (Fig. 2b), the odorous gas

is forced through a packed bed filled with a chemically inert carrier material which is colonized by microorganism, similar to trickling filters in wastewater treatment. The liquid medium is circulated over the packed bed and the pollutants are first taken up by the biofilm on the carrier material and then degreed by the microorganisms. The liquid medium can be recirculated continuously or discontinuously and in co-or countercurrent to the gas stream. Flow directions will not affect the efficiency of the process.

Performance data of BTF in lab and WWTPs are given in Table 3. Various types of packing materials have been used: inorganic salts, polyurethane foam, activated carbon fibers, multi-surface hollow balls etc. The high porosity of these packings causes less headloss compared to that of BF with organic packings, even though BTF are operated at a higher gas velocity [62], It is thus noted that BTF footprints are comparable to those of physical/chemical technologies partly as a result of their relative higher media depth. Moreover, a distinctive feature of BTF is the continuous trickling of liquid over the packing, which allows for improved control of nutrient addition, pH, acid product neutralization, end product removal, and (potentially) temperature [79]. The composition of liquid phase can affect the efficiency of BTF process, thus the trickling liquid is continuously enriched with elementary mineral nutrients containing nitrogen, phosphorus, potassium, and trace elements. Usually, BTF is operated in the temperature range between 10 and 40 °C, which is characteristics of the mesophilic microorganisms growth. The efficiency of biotreatment process may be limited by both biological reaction rate and the mass transfer rate. Therefore, it is important to notice that temperature can affect either these limitations [63]. Hydrocarbon vapors

Table 3 Performance of BTF in lab and on-site systems

Reference/location[a]	Media	EBRT (s)	Odorant	Concentration (mg/m^3)	RE (%)
[71]	A special inorganic salt	9.6	Dichloromethane	0.7–3.12	72–99
[37]	Polypropylene rings	25–60	Ethanol & ethyl acetate	50–90	46.6–68.9
[84]	Activated carbon fibers & multi-surface hollow balls	28–56	Chlorobenzene	878.53–1522.48	91.34
[85]	Polyurethane foam cubes	4–84	Methyl mercaptan, toluene, α-pinene, hexane	0.75–4.9	> 90
[76]	Tween-20 & Zn(II)	15–60	Ethylbenzene	64.8–189.0	54–94
[73]/Penn Valley, California, U.S	Seashell	19/13 1st /2nd stage	H_2S Methyl mercaptan Dimethyl sulfide	10–18 ppm – –	> 99
Headworks, Los Angeles, U.S	Structured PVC	24	H_2S Xylenes Toluene Methyl mercaptan Dichlorobenzene	10–50 0.18–0.66 0.077–0.23 0.30–0.33 0.024–0.049	> 99 0–23 0–17 64–72 0–6
Headworks, Los Angeles, U.S	Lava rock	14	H_2S	14–100	99
Primary clarifier; Fountain Valley, U.S	Continuous synthetic type	11–20	Benzene H_2S Xylenes Toluene 1,1,1,-Trichloroethane Carbon tetrachloride Chloroform Dichloromethane TCE PCE Vinylchloride	0–0.11 1.8–16 0.08–0.42 0.10–0.74 0.08–0.64 0.003–0.012 0.05–0.17 0.07–0.57 0.01–0.04 0.36–4.8 0.003–0.02	19–29 87–99 6–57 50–74 0–38 2–15 0–25 0–61 0–24 0–8 0–13
Industrial wastewater treatment; San Diego, U.S	Random inorganic	36	Benzene H_2S Xylenes Toluene MTBE Chloroform Dichloromethane	0.03 0–2 3.5 0.7 0.09 0.01 1.2	59 > 99 92 85 60 3 11

[a]Reproduced with permission from [27, 62, 63]

are removed by aerobic heterotrophic microorganisms that utilize the vapors as a source of carbon and energy [80, 81]. In the case of odorous waste air containing reduced sulfur compounds, production of sulfuric acid with declining pH and/or accumulation of sodium sulfate (after neutralization with caustic soda) is an important design parameter. Performance data for BTF indicate that these reactors are capable of efficient removal of high concentrations of H_2S at relatively low EBRTs. Thus, BTF appear to be a good option when the gas to be treated contains high concentrations of H_2S and possibly other reduced sulfur compounds. BTF performance could be limited by mass transfer of oxygen into the biofilm because oxygen solubility in water is low [82, 83]. However, there are still some drawbacks of BTF, such as the problem of gas transfer arising from the necessity of dissolving the gaseous pollutants in the aqueous phase, the biofilm development on the carrier surface which progressively reduces the empty volume of the filter bed and may lead to excessive pressure drop even the complete clogging of the bed [63].

Bioscrubbers

BS or suspended growth BS could be a solution of the limited suitability of BTF for handling high pollutant concentrations and large gas flows. In the early 1980s, the first BS units were applied to the treatment of waste gases [63]. In BS (Fig. 2c), the pollutant is adsorbed in an aqueous phase in an absorption tower then converted by the active microorganisms into CO_2, H_2O, and biomass in a separate activated sludge unit, the effluent is circulated over the absorption tower in a co-or countercurrent way to the gas stream. Meanwhile, Bowker [86] has proofed that bubble size in activated sludge unit is an important factor, since the reduction of H_2S and odors in fine-bubble diffusers can be higher (above 99.5%) than in coarse-bubble diffusers (95% for odors and 92% for H_2S).

Most existing BSs are designed for the removal of a single pollutant. For example, Nisola et al. [87] developed a single BS for ammonia removal; Potivichayanon et al. [88] investigated a fixed-film BS for hydrogen sulfide removal. Contrarily, various design modifications have been reported to handling pollutant mixtures, such as sorptive-slurry BS, anoxic BS, two-liquid phase BS, airlift BS, spray column or two-stage BS [89]. Friedrich et al. [90] investigated a 3-stage system (two identical BS + dry chemical scrubber) for the abatement of odors from sludge thickeners. Over 90% abatement efficiency was observed from the first BS and the removal efficiency exceeded 99% when applied two series-connected BS; Liu et al. [91] reported a two-series connected field-scale BS for simultaneous removal of NH_3 and CH_4 from the animal houses. Furthermore, BS offers operational stability and effective control of operating parameters such as pH and nutrients dosage, relatively low gas pressure drop and small space requirement. For example, Hansen and Rindel [92] pointed out the pH values of 8.5–9.0 as the optimal range that facilitates maintaining a high biological activity and at the same time, ensuring effective absorption of H_2S. Compared to BTF, the risk of clogging of the packing material by growing biomass is avoided, large gas flow rates and high pollutant concentrations can be handled, moreover, as reaction products are removed by washing, concentrations of toxic byproducts generated in the reactor can be maintained at low levels. However, the separate treatment system of the liquid phase increases the initial costs of establishing such kind systems. BS also possess low capacity for treatment in the removal of poorly soluble contaminants, such as hydrogen sulfide [79], thus, adding chemical compounds such as sodium hydroxide or lime water to the circulating liquid provides higher removal rates for such kind of compounds. Alternatively, the choice of microorganisms could be considered, since immobilized cells of *Chlorobium limicola* have been identified to transform H_2S into elemental sulfur in an autotrophic reaction, and heterotrophic Xanthomonas species are also known to remove H_2S from gas streams [93]. Efficient degradation of sulfur-containing compounds by certain strains of *Thiobacillus* and *Hyphomicrobium* has also been reported by Tóth et al. [94]. The mechanisms of odorous compounds removal in BS involves mainly the physical and biochemical processes such as absorption, biodegradation or biotransformation, significantly, biodegradation is the main process for the removal of the pollutants [63].

Activated Sludge Diffusion

The economical and practical odor abatement choice is simply moving them from the gaseous phase to the liquid phase [63]. A variety of systems can be employed for this purpose, for WWTPs, ASD offers a low-cost alternative. As shown in Fig. 2d, by collection of the odorous gas and its diversion into an activated sludge aeration basin, odours can be eliminated using relatively technology. Complete mixing ensures an adequate food supply for the microbial cells and maximizes the oxygen gradient to optimize mass transfer and disperse the products of metabolism from inside the flocs, wastewater entry displaces mixed liquor into a clarifier, where the flocculated biomass separates into sludge and clarified effluent.

Lebrero et al. [40] compared BF with ASD for the synthetic odor (H_2S, butanone and toluene) removal. The results confirmed ASD being a robust and efficient technology with over 95% removal efficiency for synthetic odor. Recently, Rodríguez et al. [95] investigated the microbial community and bioreactor function relationships at different EBRTs in an ASD, when treating a

synthetic malodorous (H_2S, toluene, butanone and alpha-pinene). A stable and efficient abatement performance of H_2S, butanone and toluene was observed, regard less of the EBRT and fluctuations applied, while no clear positive or negative relationship between community characteristics and bioreactor functions was confirmed. The most abundant groups namely Actinobacteria, Proteobacteria and Fungi (Hypocreales, Chaetothyriales) play a putative key role in the degradation of butanone and toluene. On the other hand, Barbosa et al. [96] reported the effects of H_2S diffusion into ASD on odor and VOC concentrations in offgas, indicating a negative effect of H_2S on VOC removal and minimal effect on odor emissions. Moreover, Barbosa and Stuetz [97] identified hydrogen sulfide can act as an energy source for microorganisms, while the carbon sources can be provided by compounds such as glucose, methanol or untreated domestic sewage. Since the solubility of hydrogen sulfide in water with pH of 7 and temperature of 19 °C is 4 g/l [7], ASD are recommended in treatment of this contaminant [98]. Additionally, Blonda et al. [99] pointed out that there is no significant effects of sulphide on COD removal efficiencies as well as nitrification and denitrification in ASD. Contaminant removal mechanisms in ASD include absorption, adsorption (high molecular mass compounds with low solubility adsorb onto flocs) or condensation (VOCs in warm air condense on contact with the cooler mixed liquor), followed by biodegradation [8]. ASD is used as an alternative to more established bioreactors for waste gas treatment, such as BF, BS, BTF. Despite ASD systems have been used for over 30 years with high H_2S removal efficiencies, their widespread implementation is still limited by the lack of reliable data concerning its performance during the treatment of odorous volatile organic compounds (VOCs).

Comparison of Odor Control Technologies in WWTPs

Summary of Different Odor Control Technologies

The benefits and disadvantages of various odor treatment technologies in WWTPs are compared in Table 4. It should be noted that none of the six main odor abatement technologies could be "one-size-fits-all". In the last decades, lot of efforts have been done in the selection of various odor control technologies. Among the efforts, life cycle assessment (LCA) could be a promising solution which has been proofed by [14, 27, 45] etc.

Table 4 Summary of different odor control technologies in WWTPs

Technology	Benefits	Disadvantages
Adsorption system	– High odor removal efficiency – Simplicity of mechanism	– Short life span – Air humidity and small molecule size – High reduction costs – Efficient only in small concentrations of contaminant
Chemical scrubbing	– Simple, effective, efficient, reliable & proper utilization – Low maintenance costs	– High costs than biotechnologies – Secondary treatment of leftover sludge
Biofilter system	– Low investment & operating costs – Absence of secondary waste streams – Low pressure drop & suitability for treating large volume of low concentration odorous gases	– Low efficiency of the treatment of high concentration pollutants – Difficult control of moisture and pH – Filter bed replacement every 2–5 years & risk of bed clogging by particulate matter
Biotrickling	– Simple and low-cost technology – Medium capital, low operating costs – Effective removal of pollutants including acid-producing ones – Low pressure drop	– When treating high pollutant concentrations, too high nutrient doses may lead to filter-bed clogging by growing biomass
Bioscrubber	– Stability of operation – Proper biological parameters control, including pH, temperature, nutrients – High pressure drop – Harmful environmental compounds are easily eliminated – Avoids inhibitory effects	– Low efficiency in case of poorly soluble substances – Treatment efficiency is reduced by special contact area of gas/liquid – High pressure drop
Activated sludge diffusion	– Utilization of available equipment (lower costs) & easy operation – Higher management capability for larger loadings – Simultaneous treatment of air and wastewater	– Complex steering by experts – Tower efficiency due to the limits of gas/liquid transference – Possibility of corrosion in equipment

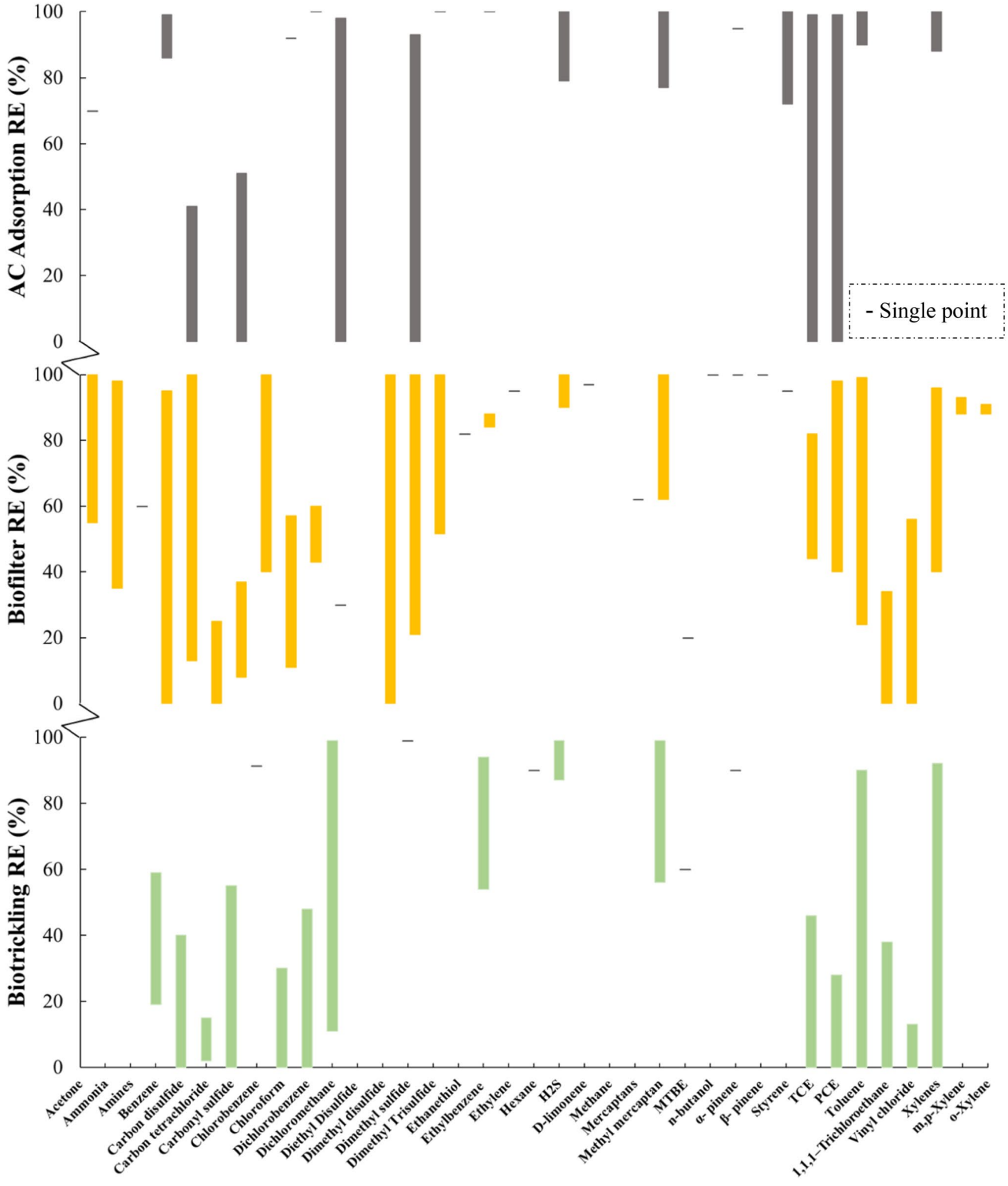


Fig. 3 The range of various odorant removal efficiency by AC adsorption, BT, BTF in WWTPs from literature [62, 63, 69, 75, 79, 89, 100–103]

Comparison of Different Odor Removal Performance

The range of 37 kinds of odorant removal performance in AC adsorption, BF and BTF are reviewed and summarised in Fig. 3. The single point represents the only one available data from the literature, indicating that there are less studies on the relevant odorant.

Generally, the performance data of odorant removal in AC adsorption were less reported than the relevant biotechnologies, 19 kinds of odorants were investigated by AC adsorption in the lab and/or field tests, H_2S and methyl mercaptan are well removed by AC, however, dimethyl disulfide was poorly removed since its generation from the breakdown of methyl mercaptan on AC systems [104]. Moreover, the removal of benzene, xylene, toluene, TCE and PCE reduced with the active lifetime decreased of the AC systems. Although AC has a longer history than BTF, in-depth studies on the performance of field units generally rely on one or two samples to justify removal. It's worth to note that there is little data available on the long-term performance of these units, particularly with respect to removal efficiency other than H_2S. The majority odorants could be removed by BF and nearly 35 kinds of different odorants were reported. BF is good at removing H_2S (RE range from 90 to 100%), Toluene (20–100%) while dimethyl disulfide and carbonyl sulfide exhibit poor removal efficiency, higher retention times might facilitate dimethyl disulfide and carbonyl sulfide removal. Twenty-one kinds of odorants were removed by BTF in the past. BTF was able to remove H_2S at lower EBRT than in BF due to the high solubility of the pollutant. They were widely used in the higher load of H_2S. However, VOC removal is poorer in BTF than BF which could be due to the low solubility of many VOCs and the continuous trickling water layer acting as a barrier to VOC bioreaction in BTF.

Perspective

In the last 30 years, hybrid technologies (physical/chemical with biological technologies) have become popular, which agrees with the robustness evaluation [51] as well as the life LCA study [27]. Although it presents a highly reliable removal efficiency for various odorants, a large amount of media replacement cost in hybrid technologies, significantly, the replacement of AC in adsorption system still the major concerns. Consequently, development of low-cost and environmental friendly adsorbents/media for odor adsorption system/biotechnologies is urgently needed in the future study. Notwithstanding various media/sorbents for odor treatment have been reported in previous studies, such as peat, zeolite, silicon, sewage sludge derived sorbents, etc. To the best of our knowledge, none of the studies ever chose the waterworks residue (alum sludge) as the low-cost adsorbent even the media in odor control of WWTPs, even it has been intensively studied and demonstrated to be a good material for wastewater treatment [105, 106].

Alum sludge is an inescapable by-product of the processing of drinking water in waterworks where aluminium salt is used as the coagulant for purifying raw water [107]. It has been advocated that waterworks sludge could be a potential recyclable product, offering promising potential as a low-cost adsorbent for various pollutants immobilization with unique feature of using "waste" for wastewater treatment [108, 109]. Thus, it has been widely reused in wastewater treatment, building & construction material, etc. moreover, alum sludge is a locally, easily and largely available material in towns, cities and metropolis and free of charge for the moment, unfortunately treated as a waste for landfilling [106]. Table 5 summaries the major characterization of two sources alum sludge which have been collected from Dublin (Ireland) and Carmaux (France), it is worth to note that alum sludge has a large specific surface area (40–300 m^2/g) and porous structure, contains various metal elements like Al, Ca, Fe, etc., particularly it has been successfully reused in constructed wetlands as substrate for high P-containing wastewater treatment and our group is a leading group in the study of alum sludge reuse [110–112].

Therefore, it is highly expected and reasonable to believe that alum sludge could be a good material/adsorbent for adsorption system. Preliminary trials have been conducted, two sources of alum sludges collected from Carmaux (France) and Dublin (Ireland) have demonstrated the H_2S adsorption effectiveness, significantly, the H_2S adsorption breakthrough capacity of Carmaux sludge (78 mg/g after 90 h) at ambient condition is several times than the capacity of calcium carbonate based solid wastes, sewage sludge based sorbents, even few active carbons [113–115]. Therefore, developing the alum sludge based adsorbent and figuring out the potential effectiveness for various odorants removal even the mechanisms and kinetics for WWTPs odor control should be a promising perspective.

Table 5 Characterization of two sources alum sludges

Sludge	S_P (BET) (m^2/g)	pH	Al (mg/kg)	Ca	K, Fe, Mo	Si, P
Dublin	47	6.9	17,983	2673	4210	106
Carmaux	257	10.0	17,581	21,156	9483	3015

Conclusion

Six odor abatement technologies were reviewed. Initially, adsorption systems and CS were developed and gradually replaced by the low-cost and environmental friendly biotechnologies. BF has dominated odor treatment applications and later, more sophisticated types of filtration equipment such as BTF and BS have been developed. However, the usage of water and the regular replacement of media is still a big concern. Particularly, the high costs of adsorption system limited their wide engineering application. Subsequently, hybrid technologies (adsorption + biotechnologies) present an environmentally friendly solution. Further research is required on the selection of low-cost sorbent to ensure a high efficiency of pollutant removal and satisfactory process robustness. Alum sludge due to its inherent characteristics could be a promising replacement to the traditional activated carbon in the adsorption system even to the media in the biofilters and biotrickling. Developing the alum sludge-based adsorbents/media for WWTPs odor treatment could be a vital innovative aspect of the environmentally friendly development as well as the "Blue Economics". It not only opens the new prospects for the waterworks sludge management but also fits in the circular approach of using a waste for waste gas control. It is hoped in this review, a clear overview of the state of the art of odor treatment technologies as well as a future study prospect was provided.

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