


Review

# Biological Waste Air and Waste Gas Treatment: Overview, Challenges, Operational Efficiency, and Current Trends

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**Abstract:** International contracts to restrict emissions of climate-relevant gases, and thus global warming, also require a critical reconsideration of technologies for treating municipal, commercial, industrial, and agricultural waste gas emissions. A change from energy- and resource-intensive technologies, such as thermal post-combustion and adsorption, as well to low-emission technologies with high energy and resource efficiency, becomes mandatory. Biological processes already meet these requirements, but show restrictions in case of treatment of complex volatile organic compound (VOC) mixtures and space demand. Innovative approaches combining advanced oxidation and biofiltration processes seem to be a solution. In this review, biological processes, both as stand-alone technology and in combination with advanced oxidation processes, were critically evaluated in regard to technical, economical, and climate policy aspects, as well as present limitations and corresponding solutions to overcome these restrictions.

**Keywords:** biological waste air treatment; biological waste gas treatment; overview; challenges; costs; trends; sustainability; VOC; UV oxidation; non-thermal plasma

## 1. Introduction

Studies of the International Energy Agency (IEA) and British Petroleum showed that by 2040 the global gross domestic product is expected to double, while greenhouse gas emissions will rise by around 30% [1,2]. However, even emissions at the current level are already leading to unquestionable environmental changes and global warming. According to a study by the German Advisory Council on Global Change (WBGU) [3], a reduction of CO<sub>2</sub> equivalents of industrial countries from approximately 12 t CO<sub>2,eq</sub>·a<sup>-1</sup>·inhabitant<sup>-1</sup> to less than 1 t CO<sub>2,eq</sub>·a<sup>-1</sup>·inhabitant<sup>-1</sup> by the year 2050 is strictly required to achieve the international objective of limiting global warming to 2 °C, equivalent to a reduction by more than 90%. In order to achieve this goal, it is inevitable that agricultural, municipal and industrial waste gas applications have to be addressed through innovative energy and resource-efficient technologies. Biological processes already meet these requirements but show restrictions in case of treatment of complex volatile organic compound (VOC) mixtures and space demand.

In the following sections, biological waste gas treatment processes are firstly presented in an overview and then with regard to technical, economic, and ecological aspects. Based on the current market situation for these technologies and the emerging markets of emission treatment of sewage sludge drying plants, the challenges for the application of this technology are also presented. This review

is concluded by presentation of current trends in the innovative combination of biological processes with advanced-oxidation technologies to overcome the listed restrictions. This process combinations show high energy and resource efficiencies and are therefore sustainable process solutions. The application of these sustainable process combinations will be necessary to meet the requirements in reduction of greenhouse gas emissions, to overcome current restrictions of biological waste gas processes, and to increase the applicability of bio-based waste gas treatment.

## 2. Overview and Application

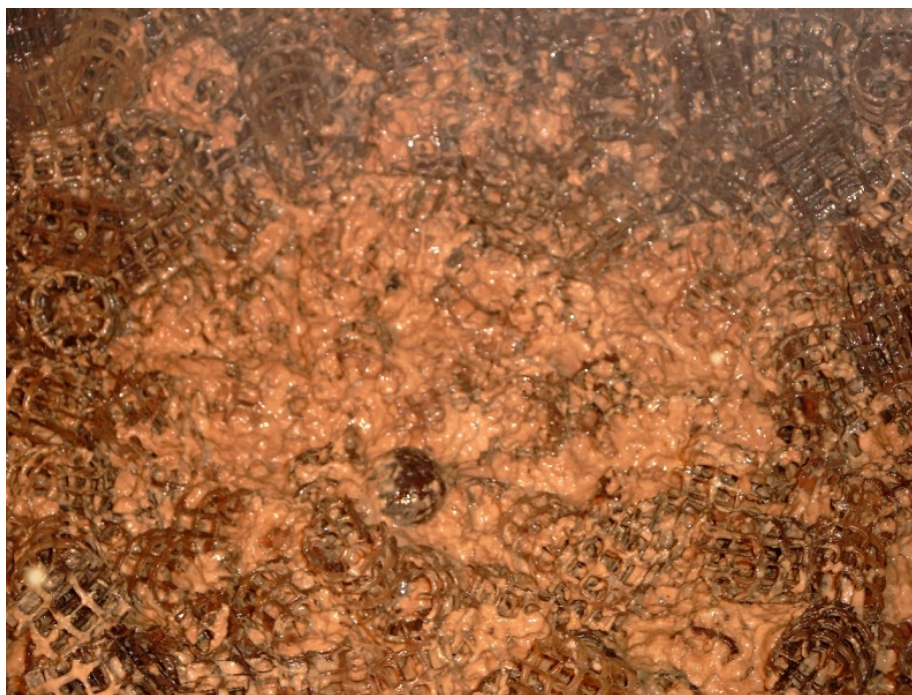
Besides competing waste gas and waste air purification techniques, like adsorption, physical or chemical scrubbing, membrane processes, condensation, thermal oxidative combustion processes, UV oxidation, non-thermal plasma (NTP), or thermal plasma, biological processes represent an important technology established at large-scale in treatment of volatile organic compounds (VOCs) and odors in agricultural, municipal, and industrial applications and are used either as stand-alone technology or in combination with the mentioned non-biological approaches. They can be subdivided into the three basic processes of bioscrubber, biofilter (BF), and biotrickling filter (BTF), in which the respective application, in particular, is defined by the polarity and, thus, the water solubility of the waste gas components to be treated [4–7]. Their process structure, function, applicability and technology-specific pros and cons are as follows.

### 2.1. Bioscrubber

The bioscrubber is a wet, two-stage system consisting of a column for physical absorption of waste gas compounds and a regenerator for biological regeneration of the loaded liquid phase [6,8]. Within the absorber, the waste gas is commonly conducted in counterflow to the dispersed liquid phase in order to enhance the number of separation stages and, thus, the purification efficiency of the column [6]. The purification efficiency can be further increased in a first step by installation of package materials to enhance the degree of turbulence and, thus, the contact time of both phases. The loaded phase is purified by biotransformation and mineralization processes of the suspended microorganisms in the aerated regenerator and subsequently dispersed in the absorber via nozzles in a closed loop operation, consisting of regeneration and loading again during contact with the waste gas [8].

The loading capacity and loading kinetics of the liquid phase depend on substance-specific properties (water solubility, Henry coefficient, pollutant concentration) [6], as well as process-specific parameters (residual load, spray density, exhaust air temperature, salinity, spray pattern, droplet size, degree of turbulence, contact time, number of separation stages, concentration of co-components, etc.). The requirements of the absorber stage (low pollutant load) conflict with the requirements of the regenerator for a high transformation kinetics (high pollutant load), so that in real operation a self-regulating operating point is usually established. Due to the wet operation of the system, bioscrubbers prove to be well suited for the treatment of polar components [4,6,7]. Since the distribution equilibrium of polar compounds is on the side of the aqueous phase, and thus the tendency of desorption is rather low, the bio-scrubber can be used for comparatively high pollutant concentrations of up to  $2 \text{ g C}\cdot\text{m}^{-3}$  [7]. Due to the high swamp volume and the wet mode of operation, it also allows a shaving of pollutant peaks, as well as a simple dosage of alkaline, solutions of nutrients, or even nitrification inhibitors for neutralization and bacterial growth [6,7,9,10]. On the other hand, the bioscrubber is not suitable for the treatment of waste gas with a high proportion of lipophilic components due to its design as a wet system. Since the 1970s, approaches with solubilizers mainly based on silicone oils were successfully introduced, creating a smooth transition to the biotrickling filter, but failed due to the high persistence of these compounds and the resulting disposal problems [11–16]. Current advances favor phthalic acid esters or polyethylene glycols on a technical scale. However, the very high biodegradability requires, on the one hand, a semi-continuous addition of these solubilizers into the aqueous phase and, on the other hand, an increased elutriation rate with higher wastewater

volumes due to the strongly increasing biomass growth in the system. The process also requires a comparatively complex control and regulation technology and the expensive provision of compressed air (approximately  $3 \text{ kW} \cdot \text{kg C}^{-1}$ ) [5,7,10,17]. The most common reason for an insufficient cleaning performance of an established bioscrubber is an insufficient oxygen input [4], caused either by a too high biomass concentration or by aging effects at the aeration elements (clogging of ceramic aerators, pore expansion or cracks in membrane aerators). As a consequence, increased foam formation due to cell lysis, a drop in pH combined with acidic odor emissions, and migration of the suspended microbial consortium onto the absorber's packing often occur (see Figure 1).



**Figure 1.** Clogging of the package material by enhanced migration of the biomass out of the swamp phase onto the packing material. Source: self-developed.

## 2.2. Biofilter

In terms of polarity, water regime, control, and regulation technology, as well as investment and operating costs, the biofilter is the counterpart of the bioscrubber [5,7]. The biofilter is a biologically active fixed bed where waste gas compounds are firstly absorbed and then mineralized by a fixed community of bacteria and fungi. As filter bed, inexpensive organic packing materials, such as compost, bark materials, forest residues, like root wood, peat, heather, coconut fibers, sawdust, or wood chips, inorganic materials, such as lava or marble quarry, or mixtures of these materials are mostly used [5,9]. Torn burl wood or a filter mat is often drawn in as the lowest layer to prevent blocking of the gas distributor. Due to the dry operation mode and to delay compaction of the package material, irrigation of the package material is strictly limited and waste gas humidification is mainly performed by a pre-scrubber in case of unsaturated waste gas conditions. Additionally, the mass transfer of mainly lipophilic waste gas compounds via the liquid phase/biofilm into microbial cells is considerably increased. Thus, biofilters are mainly used for applications dominated by lipophilic waste gas compounds in the concentration range up to  $500 \text{ mg C} \cdot \text{m}^{-3}$  and for odor abatement [5]. Here, the adsorptive properties of the organic materials have a positive effect on the purification performance, as the components present in the trace range accumulate and reach concentration levels which allow induction of microbial degradation pathways in terms of energy.

The inhomogeneous packing material and related problems are the main cons of the biofilters, where inhomogeneity leads to an inhomogeneous gas distribution and results in inactive volume

fractions of the packing material of about 40–60% [6]. The active volume fraction can be further reduced by the formation of additional flow channels and partial drying or wetting of the system. Hence, performance levels of biofilters are generally lower than those of bioscrubbers or biotrickling filters and tend to decline at long-term conditions [5,7]. This process-related lower degradation performance is usually countered by larger-volume plants. Particularly during the cold season, the filter tends to be more susceptible to wetting and has a considerable amount of highly contaminated condensate.

As wooden packing material may act as a carbon source in microbial deficiency situations, the microbial attack in combination with acidification leads to a sprouting of calcium carbonate fixed in the wood (approximately 23–48% in the ash) [18]. Hence, the wood becomes soft, decomposes more easily, and tends to intensified compaction and clogging by the fine fractions produced. The lifetime of wooden materials is approximately 7 years under mesophilic conditions but drops to less than 2 years under thermophilic conditions [7].

A significant problem of the biofilter is pollutant-induced acidification [4,7]. Usually, acidification occurs to a greater extent with alcohols, aldehydes, organic acids and esters, and to a lesser extent with ketones, ethers, aromatics, and unsubstituted aliphatics, since the volatile components of the exhaust air are biotransformed into organic acids as storage substances [4]. Acid formation also occurs during the biodegradation of organosulfur compounds, such as methanethiol, dimethyl sulfide, and dimethyl disulfide, which are mineralized either via H<sub>2</sub>S or dimethyl sulfoxide to sulfuric acid as the final product [19]. Due to the already acidic filter material, as well as a pollutant-induced acidification of the filter material, the microbial flora in biofilters has a significantly higher fungal content, which can lead to typical fungal odor.

### 2.3. Biotrickling Filter

The biotrickling filter takes an intermediate position between the bio-scrubber and the biofilter, consisting of an absorber column and a rudimentary swamp. Besides an increase in turbulence and contact times for enhanced VOC mass transfer, the package material, mostly made of plastics, serves as growth surface for the microbial biofilm. Here, biotransformation of contaminants dominantly takes place [6]. The commonly not aerated swamp phase serves as a reservoir for the circulation pump but may be slightly aerated to exclude potential odor problems.

In industrial applications, irrigation can be continuous or intermittent and the flow rate can be both adapted to hydrophilic or lipophilic characteristics of the waste gas compounds and the wetting behavior of the package [20,21]. In case of agricultural or municipal applications, which are often characterized by NH<sub>3</sub> emissions, continuous spraying is strictly necessary to minimize N<sub>2</sub>O emissions, since otherwise an average of 26% of the NH<sub>3</sub>-N load is released as N<sub>2</sub>O-N into the clean gas [22,23]. Since the flow rate is still only approximately 1–10% of that of the bioscrubber, this procedure is called semi-wet system, but it can still be used for dosage of nutrients or alkaline for neutralization, as well as pre-humidification of the waste gas [5,9]. Likewise, a temporary increase of the irrigation density with simultaneous elutriation allows a limited discharge of excess biomass, potentially toxic intermediates or excess salts to avoid salination [9]. In case of continuous irrigation, a countercurrent operation must be considered.

The gas velocity in the column is usually 0.1 m·s<sup>-1</sup> for industrial applications and is thus higher by a factor of 3–6 than for biofilters, which means that biotrickling filters can be built up more compactly. Biotrickling filters in agricultural applications operate at even higher gas velocities of 0.3–1.4 m·s<sup>-1</sup> [24]. However, the more compact design leads to higher specific loads, which may lead to clogging [5,9,25]. For example, Kan and Deshusses [26], Wang et al. [27] and Ryu et al. [28] describe the use of biotrickling filters for treatment of highly loaded toluene-containing waste gas with volume-specific loads of up to 4000 g toluene·m<sup>-3</sup>·h<sup>-1</sup>. The maximum elimination rates achieved here were 408, 480, and 3700 g toluene·m<sup>-3</sup>·h<sup>-1</sup>, respectively. Assuming a yield of 0.2 and appearance of first clogging phenomena, like inhomogeneous flow distributions and partial drying/wetting from approximately 60 kg·m<sup>-3</sup> onwards, these elimination rates result in a calculated operating time of only

80–735 h excluding increasing effects of conservative metabolism. In the case of Ryu et al. [28], a real operating time of 145 days (corresponds to a yield of only 0.01) was achieved at extremely short contact times of 3.5 s. Datta and Philip [29] described a rotating disc reactor for treatment of a mixture of methyl ethyl ketone, methyl isobutyl ketone, ethylbenzene, o-xylene, and toluene and achieved almost complete conversion with a maximum total elimination performance of  $569 \text{ g C}\cdot\text{m}^{-3}\cdot\text{h}^{-1}$ . However, specific loads or elimination rates in the range of  $10\text{--}100 \text{ g C}\cdot\text{m}^{-3}\cdot\text{h}^{-1}$  should be aimed for in order to ensure stable long-term operation [30].

Further disadvantages of biotrickling filters are the low buffer capacity against pollutant peaks and low moisture content of the plastic package materials due to the lack of adsorption capacity of the filter material. The use of plastic carriers also causes problems during time intervals without pollutant loads, as no metabolizable carbon source is available. Artificial addition of pollutants either into the swamp phase or into the conventional exhaust air route is a feasible solution.

### 3. Technical, Economic, and Climate Policy Evaluation of Waste Air Treatment Techniques

#### 3.1. Technical Aspects

Biological waste air processes are particularly used in agricultural, municipal, and industrial waste air applications, especially for issues arising from husbandries, animal meal and sewage sludge drying, composting, landfilling, biogas treatment, and production processes in chemical, plastics, paper, and wood industries. Waste air from agricultural and municipal applications is typically characterized by large-volume flows (approximately  $10,000\text{--}100,000 \text{ m}^3\cdot\text{h}^{-1}$ ) at variable concentrations of VOCs (approximately  $10\text{--}300 \text{ mg C}\cdot\text{m}^{-3}$ ), ammonia (up to  $500 \text{ mg NH}_3\cdot\text{m}^{-3}$ ), nitrous oxide ( $5\text{--}120 \text{ mg N}_2\text{O}\cdot\text{m}^{-3}$ ), odorous compounds (over  $20,000 \text{ OU}\cdot\text{m}^{-3}$ ), germs ( $10^3\text{--}10^5 \text{ CFU}\cdot\text{m}^{-3}$ ), and possibly methane ( $10\text{--}150 \text{ mg C}\cdot\text{m}^{-3}$ ) and, to a lesser extent,  $\text{H}_2\text{S}$ . Industrial production processes are also characterized by high volume flows (approximately  $10,000\text{--}500,000 \text{ m}^3\cdot\text{h}^{-1}$ ) with enhanced concentrations of VOCs (approximately  $100\text{--}500 \text{ mg C}\cdot\text{m}^{-3}$ , in some cases up to  $2000 \text{ mg C}\cdot\text{m}^{-3}$ ) [7,31]. The typical spectrum of waste gas compounds ranges from polar compounds, such as alcohols and ketones (e.g., in the printing and paint industry), mixtures of polar and lipophilic components to situations with solely lipophilic compounds, like styrene and toluene derivatives (e.g., in trickle resins, film, and plastic production). Depending on the production processes, additional inorganic compounds, like  $\text{NH}_3$ ,  $\text{N}_2\text{O}$ , or  $\text{H}_2\text{S}$ , may occur at concentrations above  $1 \text{ g}\cdot\text{m}^{-3}$  (e.g., at pig bristle hydrolysis). Furthermore, waste air flows are characterized by highly variable dust loads, humidity (0–100% of relative humidity), and ranges in temperature (ambient level up to approximately  $60 \text{ }^\circ\text{C}$ ). These operational parameters typically exclude treatment processes, such as condensation, membrane processes, thermal plasmas, or oxidative catalytic processes, from application. Alternative processes, like absorption, thermal oxidative processes, non-thermal plasma, UV-oxidation, and adsorption, are not suitable as stand-alone single-stage systems under these circumstances, as well. A summary of the suitability and limits of the respective processes is given in Table 1.

As shown in Table 1, physisorption is no suitable technique in presence of poorly water-soluble components and permanent gases due to its limitation to hydrophilic components. Although chemisorption permits improved elimination of  $\text{H}_2\text{S}$ , odors, germs, and individual aromatic components, particularly when oxidizing agents are added, lipophilic components with a low tendency to oxidize also prove unsuitable for this process. The suitability of non-thermal plasma, as well as UV-oxidation, highly depends on substance properties. An efficient treatment of the waste gas with low specific input energy (SIE) values requires an increased reactivity of the waste gas components (e.g., aldehydes) or the presence of an unsaturated bond (e.g., alkenes, styrene or similar). Although adsorption is, meanwhile, also suitable for the treatment of polar components due to the development of impregnated carbons, the specific loadability is still many times lower than for lipophilic components. Especially in dusty exhaust air streams with high relative humidity, the process

requires preconditioning or has high operating costs. However, thermal oxidative processes have proven to be suitable and therefore strongly dominate this field of industrial applications.

**Table 1.** Suitability of waste air treatment processes based on nearly 70 in-house industrial studies. +: high suitability, +/-: moderate suitability, -: no/low suitability.

Parameter	Phy. Scrubber	Chem. Scrubber	Incineration	NTP	UV-Oxidation	Adsorption	Bioscrubber	Biotrickling Filter	Biofilter
Aliphatics	-	-	+	+/-	+/-	+	-	+	+
Alcohols	+	+	+	+/-	+/-	+	+	+	+/-
Aldehydes	+	+	+	+	+	+/-	+	+	+
Ketones	+	+	+	+/-	+/-	+	+	+	+
Organic acids	+	+	+	+	+	+	+	+	+/-
Esters	+	+	+	+	+	+	+	+	+/-
Ethers	-	-	+	+/-	+/-	+	-	+	+
Aromatics	-	+/-	+	+/-	+/-	+	-	+	+
Sulphurorganics	-	+	+/-	+/-	+/-	+	-	+	+/-
Haloorganics	-	-	+/-	+/-	+/-	+	-	+	+/-
CH <sub>4</sub>	-	-	+	-	-	-	-	-	+/-
NH <sub>3</sub>	+	+	+/-	+	+	-	+	+	+
N <sub>2</sub> O	-	-	+/-	-	-	-	-	-	+/-
H <sub>2</sub> S	+/-	+	+/-	+/-	+/-	+/-	+	+	+/-
Odor	+/-	+	+	+	+	+	+/-	+/-	+/-
Germs	-	+	+	+	+	+	-	+	+
Dust	+	+	-	-	-	-	+	+/-	-
Humidity	+	+	+/-	-	-	-	+	+	+

Similar to the non-biological scrubbers, the bioscrubber is also restricted to polar waste gas compounds [6,7]. Due to low irrigation density, however, both biofilter and biotrickling filter are eligible for treatment of a wide range of polarities and are therefore suitable for various applications [5]. In particular, the biotrickling filter benefits from the variable water management, which allows the discharge of acid reaction products, like H<sub>2</sub>SO<sub>4</sub>, HCl, or HF, or the addition of alkalines for neutralization [5,9,32].

### 3.2. Economic Aspects

From an economic point of view, biological processes are clearly preferable to post-combustion processes, as their investment costs reach only 20–50% of the costs of a post-combustion plant. As incinerators operating at non-adiabatic conditions require both primary and electrical energy, they have high operating costs. In contrast, the operating costs of biological treatment processes are mainly defined by the electrical energy demands of the compressor and, if necessary, the circulation pump [5,7–9]. Operational costs for dosage of alkalines and nutrients are normally below 1000 €·a<sup>-1</sup> for treatment plants of a capacity up to 20,000 m<sup>3</sup>·h<sup>-1</sup>. An overview of investment and operating costs of biofilters, biotrickling filters, thermal oxidation, and adsorption, as well as non-thermal plasmas as comparative processes, presented in literature, is compiled in Table 2. The calculations of this study base on electrical energy costs of 0.15 €·kWh<sup>-1</sup> and natural gas costs of 0.416 €·m<sup>-3</sup>.

**Table 2.** Compilation of investment and operational costs of waste air treatment processes. Calculations of this study base on electrical energy costs of  $0.15 \text{ €} \cdot \text{kWh}^{-1}$ , natural gas costs of  $0.416 \text{ €} \cdot \text{m}^{-3}$ , and a minimum size of  $10,000 \text{ m}^3 \cdot \text{h}^{-1}$ .

Process	Investment Costs ( $\text{€} \cdot \text{m}^3/\text{h}^{-1}$ )	Operational Costs ( $\text{€} \cdot 1000 \text{ m}^{-3}$ )	Reference
Biofilter	1.53–5.11	0.44–0.48	[33]
	0.90–1.50	0.70–1.00	[34]
	4.10–12.80	0.25–0.77	[35]
	2.80–4.00	0.25	[36]
	2.50	0.23	[37]
	-	0.24	[38]
Biotrickling filter	13.00–15.00	0.71	[39]
	2.81–8.13	0.04–1.27	[40]
	15	0.13–0.45	[38]
	-	0.12	[41]
	2.50	0.20	[37]
	1.41–18.78	0.14–0.31	This study
Adsorption	2.50–5.00	3.00–3.50	[42]
Chemical scrubber	0.70–1.90 (odor)	0.40 (odor)	[34]
	18.85–23.20 (VOC)	0.50 (VOC)	
	4.10–25.60	0.50–2.55	[35]
Thermal oxidation	13.00–15.00	2.27	[39]
	12.8–25.6	1.50–7.67	[35]
	34.83	1.62	This study
Non-thermal plasma	5.0–8.0	0.30–0.84	[42]
	1.20–1.40 (odor)	0.30 (odor)	[34]
	10.00–12.00 (VOC)	1.00 (VOC)	
	8.52–30.68	0.19–0.68	[43]
UV	13.65	2.62	[44]

As shown, the investment costs for biotrickling filters are in the range of 2.50–18.78 € per cubic meter of hourly waste air capacity, depending on the size of the plant, while the investment costs for thermal oxidation are between 12.80–34.83 € per cubic meter of hourly waste air capacity [35,37,38] this study. The operating costs of biotrickling filters are in the range of 0.12–0.45 €·1000 m<sup>-3</sup> and, thus, a factor of 12–17 below those of thermal oxidation processes (1.50–7.67 €·1000 m<sup>-3</sup>) [35,37,38,41]. Due to lower requirements in treatment of agricultural exhaust gases (target set to 70% in minimum for ammonia, dust, and odor), biofiltration processes in agricultural applications are operated at significantly higher filter volume loads ( $100\text{--}175 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$  in industry vs.  $1000\text{--}11,000 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$  in agricultural applications), which allows a more compact design [24]. Accordingly, the specific operating costs are significantly reduced by a factor of 2. For clarification, the investment and operating costs of a biotrickling filter versus combustion process for an industrial styrene-containing exhaust air stream (20–25 °C; 50% r.H.;  $150\text{--}500 \text{ mg styrene} \cdot \text{m}^{-3}$ ,  $13,000 \text{ m}^3 \cdot \text{h}^{-1}$  gas flow) and of a biotrickling filter for a pig fattening site (1000 places,  $91,000 \text{ m}^3 \cdot \text{h}^{-1}$  gas flow) are summarized in Table 3. In summary, it can be pointed out that biofiltration processes with optional pre-treatment processes are competitive technologies compared to conventional thermal oxidation processes at lower investment and operational costs. Furthermore, in combination with additional advanced oxidation processes, like NTP or UV-oxidation, bioavailability and biodegradability and, finally, the elimination efficiency may be enhanced, leading to an additional reduction in investment and operating costs [45] (see Section 6).

**Table 3.** Cost comparison between biotrickling filter (BTF) and a regenerative combustion (RPC) for treatment of an industrial styrene-containing air of 13,000 m<sup>3</sup>·h<sup>-1</sup> and of a biotrickling filter for treatment of pig fattening waste gases of 91,000 m<sup>3</sup>·h<sup>-1</sup>, based on actual quotes. Calculations use electrical energy costs of 0.15 €·kWh<sup>-1</sup> and natural gas costs of 0.416 €·m<sup>-3</sup>.

	BTF Resin	RPC Resin	BTF Pig Fattening
RPC ( $p = 262$ kW, $\Delta T = 35$ K)	-	250,000 €	-
BTF	151,000 €	-	49,000 €
Bypass during production shutdown	-	30,000 €	-
Fresh air damper	-	2400 €	-
Pre-filter	-	11,700 €	-
Burn-out function	-	9500 €	-
Dryer for compressed air	-	1900 €	-
Compressor (at 6 bar)	3500 € (1.3 L·s <sup>-1</sup> )	5400 € (2 L·s <sup>-1</sup> )	-
Ventilator	4500 € (3500 Pa)	21,500 € (for 100 °C, 5500 Pa)	18,600 € (2100 Pa)
Chimney	-	20,500 €	-
Assembly and commissioning	18,000 €	35,000 €	10,500 €
Transport and crane	5000 €	9500 €	4000 €
Foundation	30,000 €	30,000 €	20,000 €
Waste air pipes (25 m)	2500 €	2500 €	3500 €
Natural gas connection	-	15,000 €	-
Water connection (25 m, of which 10 m ice-free-proof)	1750 €	-	3000 €
Waste water connection (10 m, ice-free-proof)	1200 €	-	2000 €
pH dosage	5000 €	-	5000 €
Circulation pump	800 €	-	500 €
Nutrient dosage	1500 €	-	-
Level control	2500 €	-	2500 €
Control cabinet	7000 €	8000 €	9500 €
Isolation and ice-free installations	10,000 €	-	-
<b>Total investment</b>	<b>244,250 €</b>	<b>452,900 €</b>	<b>128,100 €</b>
Electricity demand ventilator	130,000 kWh	389,000 kWh	648,000 kWh
Electricity demand pump	11,200 kWh	-	9300 kWh
Electricity demand compressor	3800 kWh	5600 kWh	-
Electricity demand pipe heating (10 W·m <sup>-1</sup> )	43 kWh	-	43 kWh
Peripheral electricity demand	550 kWh	-	300 kWh
Annual electricity demand (kWh)	145,593 kWh	394,600 kWh	657,643 kWh
Annual electricity demand (€)	21,840 €	59,190 €	98,650 €
Natural gas	-	94,200 €	-
Chemicals	1000 €	-	550 €
Parts of annual replacement	1000 €	8500 €	250 €
Annual maintenance	1500 €	5000 €	700 €
Staff costs (on-site)	6500 €	6500 €	4000 €
Interest costs (2.5%)	6200 €	11,500 €	3200 €
<b>Annual operational costs</b>	<b>35,540 €</b>	<b>184,890 €</b>	<b>107,350 €</b>
<b>Specific investment costs (€·m<sup>-3</sup> hourly air capacity)</b>	<b>18.78</b>	<b>34.83</b>	<b>1.41</b>
<b>Specific operational costs (€·1000 m<sup>-3</sup>)</b>	<b>0.31</b>	<b>1.62</b>	<b>0.14</b>

### 3.3. Ecological Aspects

From an ecological point of view, the application of biofiltration processes is also clearly preferable to combustion processes, since thermal oxidative processes are characterized by high greenhouse gas emissions (CO<sub>2</sub>, CH<sub>4</sub> as slip gas). It is also known that N<sub>2</sub>O emissions are formed during thermal oxidative processes, which contribute to a considerable greenhouse potential. Assuming the German energy mix of 2019 (production of 0.401 kg CO<sub>2</sub>·kWh<sup>-1</sup>) [46], secondary emissions of a thermal incineration process reach 58,477 kg CO<sub>2,eq</sub>·a<sup>-1</sup> for treatment of a waste air volume flow of 1000 m<sup>3</sup>·h<sup>-1</sup> at 8000 h per year. Since the energy demand of biofiltration processes is quantitatively defined by the energy requirements of the compressor and N<sub>2</sub>O emissions of converted ammonia-based fertilizers, emissions of CO<sub>2</sub> equivalents for biological processes are approximately 3565–3939 kg CO<sub>2,eq</sub>·a<sup>-1</sup> and, thus, approximately 6–7% of the global warming potential of thermal incineration processes. However, this advantage might be completely negated in case of incorrectly operated plants for NH<sub>3</sub> elimination, as secondary emissions of 1–11% of NO or 3–29% of N<sub>2</sub>O of the original ammonia load can be formed by bioconversion processes of NH<sub>3</sub> [47–51]. Distinguished by source the emissions of CO<sub>2</sub> equivalents of an incineration process and of a non-thermal plasma process with post plasma catalyst (PPC), as discussed in Section 7, are presented in Table 4. Compared to the thermal process, the NTP process reveals a lower emission load of 67.3% or 39,359 kg CO<sub>2,eq</sub>·a<sup>-1</sup> but is up to 15,598 kg·a<sup>-1</sup> above



the biofilter. Since the balancing of Table 4 for NTP processes assumes a complete mineralization of the pollutant by a stand-alone NTP process, a process combination of NTP and biotrickling filter (BTF) appears as an ecologically advantageous alternative, since the pollutants are then hydrophilized by partial oxidation in the NTP stage and can thus be degraded by a smaller BTF stage (see Section 7).

**Table 4.** Balancing of emissions of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O as greenhouse gases for combustion and non-thermal plasma (NTP) processes for treatment of a gas flow of 1000 m<sup>3</sup>·h<sup>-1</sup> for 8000 h·a<sup>-1</sup>. Data based on CO<sub>2</sub> emissions of the German energy mix in 2019 (0.401 kg CO<sub>2</sub>·kWh<sup>-1</sup>). \* analytical data of the authors; \*\* experience values for combustion. \*\*\* calculated value based on a concentration of 10 mg·L<sup>-1</sup> of ammonia as fertilizer in the swamp phase at pH 7.8 and ammonium/ammonia distribution of Kollbach [52].

Parameter	Incineration		Non-Thermal Plasma		BTF	
	Quantity	In CO <sub>2,eq</sub>	Quantity	In CO <sub>2,eq</sub>	Quantity	In CO <sub>2,eq</sub>
Natural gas	12,729.5 m <sup>3</sup> ·a <sup>-1</sup>	25,003 kg·a <sup>-1</sup>	0 m <sup>3</sup> ·a <sup>-1</sup>	0 kg·a <sup>-1</sup>	0 m <sup>3</sup> ·a <sup>-1</sup>	0 kg·a <sup>-1</sup>
Methane emissions	24.0 kg·a <sup>-1</sup> (3 mg C·m <sup>-3</sup> **)	504 kg·a <sup>-1</sup>	3.31 kg·a <sup>-1</sup> (0.58 ppm *)	69.6 kg·a <sup>-1</sup>	0 kg·a <sup>-1</sup>	0 kg·a <sup>-1</sup>
Energy demand fan	17.6 MWh·a <sup>-1</sup> (Δp = 5000 Pa)	7040 kg·a <sup>-1</sup>	8.8 MWh·a <sup>-1</sup> (Δp = 2250 Pa)	3520 kg·a <sup>-1</sup>	8.8 MWh·a <sup>-1</sup> (Δp = 2250 Pa)	3520 kg·a <sup>-1</sup>
Energy demand compressor	4 MWh·a <sup>-1</sup> (P = 0.5 kW)	1600 kg·a <sup>-1</sup>	0 kWh	0 kg·a <sup>-1</sup>	0 kWh	0 kg·a <sup>-1</sup>
Energy demand NTP	0 kWh	0 kg·a <sup>-1</sup>	32 MWh·a <sup>-1</sup> (4 kWh·1000 m <sup>-3</sup> )	12,800 kg·a <sup>-1</sup>	0 MWh·a <sup>-1</sup>	0 kg·a <sup>-1</sup>
N <sub>2</sub> O formation	78.5 kg·a <sup>-1</sup> (5 ppm **)	24,330 kg·a <sup>-1</sup>	8.8 kg·a <sup>-1</sup> (0.57 ppm *)	2728 kg·a <sup>-1</sup>	0.15–1.35 kg·a <sup>-1</sup> (0.32 ppm ***)	45–419 kg·a <sup>-1</sup>
<b>Sum</b>		<b>58,477 kg·a<sup>-1</sup></b>		<b>19,118 kg·a<sup>-1</sup></b>		<b>3565–3939 kg·a<sup>-1</sup></b>
<b>Reduction potential</b>		<b>0 kg·a<sup>-1</sup></b>		<b>39,359 kg·a<sup>-1</sup> or 67.3%</b>		<b>54,538–54,912 kg·a<sup>-1</sup> or 93.3–93.9%</b>

#### 4. Current Market Position of Biological Waste Air Treatment Processes

As shown before, biological waste air treatment processes prove to be particularly suitable for applications, like agricultural animal husbandry, drying of animal meal and sewage sludge, waste composting, or applications in similar industrial sectors, but they particularly compete with scrubber processes (agriculture) and thermal incineration or adsorption processes (industrial sectors). In agricultural application, both regular surveys of the German Federal Statistical Office on livestock [53] and a study by Hahne [54] allow a good estimation of the market share of biological waste air treatment processes in this market segment. In Germany, at the recording date of 1 March 2016, there were about 120.966, 40.267, 19.556, 9.808, 49.093, and 42.145 farms for cattle, pig, sheep, goats, poultry, and horses/donkeys, respectively. Emissions of these farms are characterized by low carbon emissions but considerable odor emissions, mainly caused by NH<sub>3</sub>, which is solely emitted to more than 500,000 t·a<sup>-1</sup> with further upward trend by stock farming.

Nevertheless, the installation of an exhaust air purification plant is not generally required and only demanded in specific individual cases of the approval procedure, for example when the distance to residential buildings is below minimum requirements [55]. Accordingly, the installation of waste air treatment systems is being carried out tentatively in Germany. In the period from 1997 to 2010, only 1014 waste air treatment plants were installed (pigs: 762, poultry: 170, others: 82) [54]; hence, only 0.4% of all farms were equipped with treatment technologies in 2011. Single-stage biofilters were installed in 26.4% of the cases, single stage biotrickling filters in 24.0%, and multi-stage biological systems in 43.2%. Only in 6.4% of the cases non-biological systems, such as chemical scrubbers, were implemented. However, a considerable expansion of installations may be expected after the entry into force of the federal filter decrees of North Rhine-Westphalia (2013), Lower Saxony (2013), Schleswig-Holstein (2014), Thuringia (2016), and Brandenburg (2017), since, in the case of new installations, as well as considerable constructive changes to existing pig farms, the installation of waste air treatment plants is generally required [56]. If necessary, installation can also be imposed on smaller farms, but state-specific

installation obligation varies between 1000 (Thuringia) and 10,000 (Brandenburg) fattening places. Nevertheless, the number of waste air treatment systems has thus risen sharply since 2013, but no current market figures are available. Since cattle farming causes approximately 51% of agricultural ammonia emissions, the expansion of these decrees to this application is expected in the next few years.

Biological waste air processes are widely used in the environmental and waste management industry, but also in some other fields of industrial and manufacturing application (see Table 5) [57,58]. Further applications are known from automotive industry, cement industry, energy production, printing industry, the re-food sector, and other industrial applications. In all market sectors, they are mainly used for odor abatement, although isolated applications for the treatment of high carbon loads are also known.

**Table 5.** Industrial fields of application and shares of biological waste air treatment in Austria [57,58].

Sector	Share (%)
Municipal wastewater treatment	48.0
Industrial wastewater treatment	14.0
Food industry	10.6–14.1
Composting	6.8
Chemical industry	3.4–8.4
Commercial	4.6–6.5
Landfills	4.6
Rendering plants	4.6
Plastics industry	1.9–3.4
Pulp and paper industry	0.0–2.7
Electronics industry	0.0–1.1
Ceramics industry	0.0–1.1
Petroleum industry	0.0–0.8
Wood processing industry	0.0–0.8
Leather processing industry	0.0–0.8

The share of industrial waste air applications with VOC contents of several  $100 \text{ mg C}\cdot\text{m}^{-3}$  and potentially odorous compounds at temperatures less than  $50 \text{ }^\circ\text{C}$  can be estimated at 10–15% of the total number of applications where waste air treatment is required, but only about 1% of these applications are assumed to be treated by biological waste air treatment processes. In this respect, there is a growth potential of 1400% for biological processes, which might be further increased by marketing of intelligent and economically interesting combined processes as described in Section 7 [59].

## 5. Biofiltration in Sludge Drying Processes

In recent years the drying of municipal and industrial sludges can be observed as a new market segment for biological exhaust air treatment. This development is based on the following three factors [60–62]:

1. according to German legislation sewage sludge may no longer be applied to agricultural fields;
2. in the course of the pending German coal exit, sewage sludge is increasingly seen as a substitute for lignite; and
3. sewage sludge ashes are rich in phosphates, which is why sewage sludge is becoming increasingly important as a source of phosphorus, despite the high costs of recovery. However, recovery requires a thermal pretreatment.

In the drying process, solar-supported low-temperature drying (up to approximately  $50 \text{ }^\circ\text{C}$ ) in greenhouse-like halls, as well as high-temperature processes ( $80 \text{ }^\circ\text{C}$  to approximately  $300 \text{ }^\circ\text{C}$ ) in the form of belt dryers, plate dryers, fluidized bed dryers, drum dryers, and paddle dryers, have been established as technical processes. Biofilters with one or more preconditioning stages have become established for the waste gas treatment at most drying locations. Due to the strongly

varying drying temperatures, this results in strongly varying treatment times in the dryer (minutes in the high-temperature dryers, up to seven days in low-temperature dryers) [60], as well as high variations in both waste gas temperature and pollutant concentrations [63–65]. For selection of the right waste gas treatment technology, preliminary tests are therefore necessary to characterize the actual emissions. Typical plant configurations for low-temperature drying include a simple plant combination of acidic scrubber for ammonia elimination and a biofilter for odor elimination at volume loads of  $250 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$ , while high-temperature processes require more complex treatment systems consisting of at least two upstream scrubbers and a thermophilic biofilter with a volume load of less than  $100 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$ .

It should be pointed out that these plant configurations are suitable for compliance with the limit values of the German Technical Instructions on Air Quality Control (TA Luft). As an exception, methane emissions cannot be effectively treated by these processes (see Section 6.7).

## 6. Marketing Obstacles and Challenges

Despite considerable economic and ecological advantages, as well as a significant underrepresentation of biological processes in waste air treatment applications according to Section 4, biological waste air treatment processes still represent a niche solution due to the following technical limitations, which must be overcome to establish these processes successfully.

### 6.1. Limitation in Efficiency

Well-adjusted biofiltration processes may achieve stable efficiencies of about 85% for VOCs and above 95% for odor abatement. Further increases in efficiency are associated with an exponential increase in maintenance.

### 6.2. Limitation in VOC Crude Gas Levels

According to the German Technical Instructions on Air Quality Control clean gas levels of VOCs are restricted to  $50 \text{ mg C} \cdot \text{m}^{-3}$  in total. If an efficiency of 85% for treatment of VOC emissions according to 5.1. is considered, biological processes are firstly restricted to crude gas concentration of  $333 \text{ mg C} \cdot \text{m}^{-3}$  in maximum.

### 6.3. Limitation in Space Requirement

Higher crude gas levels can be successfully treated by increasing the empty bed residence time (EBRT), thus reducing volume specific VOC loads. However, the lower the specific loads, the higher the space requirements of the biofiltration processes are [5,7]. Based on process specific gas velocities ( $3\text{--}5 \text{ m} \cdot \text{s}^{-1}$  for non-biological processes ex. adsorption; approximately  $1.5 \text{ m} \cdot \text{s}^{-1}$  for bio-scrubber;  $0.3\text{--}1.0 \text{ m} \cdot \text{s}^{-1}$  for biotrickling filter; less than  $0.3 \text{ m} \cdot \text{s}^{-1}$  for biofilter and adsorption), the required plant volume of biological processes is higher by a factor of 6–10 than the plant volume of non-biological processes.

### 6.4. Clogging

Biofiltration processes normally operate at volume-specific loads of  $125 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$  for VOC elimination and  $175 \text{ m}^3 \cdot \text{m}^{-3} \cdot \text{h}^{-1}$  for odor abatement, equivalent to VOC elimination efficiencies up to  $100 \text{ g C} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$ , respectively. By reducing the space requirements, and hence the plant volume, the volume-specific loads increase. During biodegradation VOCs are converted both to mineralization products, like  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{NH}_3$ ,  $\text{HCl}$ , or similar, and especially energy, which is partly used in biomass formation. An increase in the volume-specific load considerably accelerates the total biomass production. If approaches to get rid of excessive biomass are not successful, the risk of biomass-induced clogging of the package increases considerably and operational time of the plant decreases dramatically [66,67]. Such approaches might be adequate operation parameters of

the package materials (material, size, structure), physical and mechanical approaches (changes in irrigation or flow pattern, flushing, fluidization, high pressure nozzles, periodical or continuous extraction of the packing material and subsequent cleaning, submerged system, rotating contactors, squeezing techniques, or temperature shifts), chemical methods (shifts in pH or salinity, further addition of chemicals), advanced oxidation processes (NTP, UV oxidation, ozonation), biological approaches (shifts in microbial communities, addition of predators), or limitations (nutrients, starvation periods) [25]. With the exception of starvation and nutrient limitation, only a few studies have dealt with the validation of these methods for clogging prevention in pilot-scaled or industrial-scaled waste air treatment systems at gas flows of  $100 \text{ m}^3 \cdot \text{h}^{-1}$  or higher [25,68–70]

### 6.5. Limitation in Transformation Kinetics

A wide range of xenobiotic chemicals is used in industrial and commercial processes. By increasing the xenobiotic character of the chemicals, the biological transformation kinetics and, thus, the volume specific elimination efficiency decreases [71]. Therefore, larger plant sizes and, thus, more expensive installations are required for successful treatment of corresponding waste gas flows [5,71]. The low transformation kinetics of xenobiotic compounds can be based either on a low affinity of the relevant enzymes to their new substrates or on low induction by these new substrates during transcription of the associated gene sequences. The same applies to lipophilic waste gas compounds, as well as components with very high vapor pressures which, due to the limited water solubility, are only present at low concentrations in the aqueous phase or biofilm, and therefore show low bioavailability. Additionally, biodegradation follows a 1st order reaction kinetics. However, the objective of both compact and cost-effective biological waste gas treatment systems seems realistic in case of processes combining non-biological and biological approaches, in which the upstream non-biological stage enhances hydrophilicity and, thus, bioavailability of waste gas compounds by partial oxidation reactions (see Section 7 for details).

### 6.6. Grade of Persistence

The xenobiotic character of chemicals increases by higher ratio of anthropogenic substituents and binding types that do not occur naturally in the environment. Examples are halogen substituents, nitro groups or nitrile bonds. Since the exposure of microorganisms to these substituents and bonds has taken place for a short time from an evolutionary point of view, microorganisms generally have not adapted/optimized corresponding degradation enzymes nor established new types of highly efficient pathways. The absence of suitable enzymes may lead to failure of the application.

On the other hand, this postulate was contradicted by various biodegradation studies of xenobiotic compounds. As examples, biodegradation of 2-halobenzoates [72], 3-fluorobenzoates [73], 2,4-dichlorophenoxyacetate and 2-chloro-4-methylphenoxyacetate [74,75], fluorene and other PAHs [76,77], aryl, alkyl, biaryl and cyclically condensed biaryl ethers [78], fluorobenzene [79,80], 2-chlorotoluene [32,81], and triclosan [82–84] were shown successfully.

### 6.7. Limitation in Degradation of VOC Mixtures

With increasing complexity, number and variation of chemical substituents, the complexity of regulatory mechanisms both at gene and protein levels in biodegradation of waste gas compounds also increases. VOCs that belong to the same chemical group or lead to the same degradation pathway can usually be degraded microbially without any loss of enzymatic performance when presented as a mixture (e.g., alkyl aromatics, like toluene, styrene, and xylene isomers, can be degraded simultaneously by the same enzyme set) [85,86]. A comprehensive compilation of test series for the simultaneous degradation of aromatics can be found in [85]. However, simultaneous biodegradation of VOCs belonging to different chemical groups is more difficult, i.e., treatment of 1,2-dichlorobenzene in mixture with toluene and o-xylene [85], alcohols and ethers [87], methoxypropyl acetate and xylene isomers [88], or *tert*-butanol and acetone [89], as shown by different studies of our research group. Moreover,

this effect was even observed in case of closely related groups of substances, like benzene in mixture with toluene, styrene, and xylene isomers [86]. While a simultaneous degradation of toluene, styrene, and xylene isomers at degradation rates corresponding to their specific water solubility occurred, benzene was not degraded due to a different initial ring oxidizing enzyme system.

Even though a natural compound, it should be mentioned that methane reveals poor biodegradability due to its high vapor pressure, low solubility in water, and high chemical stability. Especially in concentrations less than 100 ppm, and therefore relevant for the majority of large-scale applications, methane conversion was observed at very high EBRTs of 72–96 h, and performance further declined in presence of VOCs [90–93].

Hence, besides the affiliation to individual substance groups, the water solubility of components plays a decisive role in microbial biodegradation, as water solubility and, thus, bioavailability decreases with increasing lipophilicity. Accordingly, multiple studies pointed out the preferential degradation of hydrophilic VOCs in mixtures of both hydrophilic and lipophilic compounds. As an example, enhanced input loads of n-hexane and benzene in a ternary mixture of methanol, n-hexane, and benzene had no significant effect on methanol removal efficiency (>95%), while the removal efficiency of n-hexane was severely affected by changes in methanol loads (drop from 75% to 32%) [94,95]. Comparable results were observed for mixtures of BTEX and n-hexane or benzene and n-hexane, where the removal efficiency of n-hexane as a very lipophilic compound decreased in presence of these aromatic compounds [96,97].

Therefore, simultaneous degradation of VOCs representing different chemical groups is often not possible and sufficient degradation may not be achieved. However, this problem mainly occurs in bioscrubber systems with homogeneous distribution of microbial consortia and pollutants, while this problem is of less relevance in biofiltration processes due to different established microbial consortia along the height of the packing material, i.e., hydrophilic and therefore highly bioavailable compounds are commonly mineralized near to the crude gas inlet of the packing, while lipophilic compounds are degraded downflow.

### 6.8. Limitation in Gas Temperature

The vast majority of microbial primary degraders grow under mesophilic conditions, i.e., approximately 10–40 °C. The transformation optimum is usually around 30 °C. Additionally, thermotolerant or thermophilic microorganisms were also applied in treatment processes for waste air temperatures of 50 °C or higher, like odor emissions of a cocoa factory [31], emissions of sludge drying processes [98], wood processing industry [31,99,100], or treatment of VOC mixtures, like benzene, n-hexane, and toluene [101]. Even though the process adapted microorganisms exhibit higher transformation kinetics due to the higher temperature according to the van't-Hoff equation, biomass yield decreases simultaneously due to increasing maintenance metabolism [102]. The start-up process of industrial plants is thus considerably longer and more difficult. In particular, stable operation of such thermophilic plants is fundamentally more challenging, since they require constant temperatures above 45 °C, which commonly cannot be guaranteed during maintenance cycles. Besides recurring changes between mesophilic and thermophilic conditions, increases in vapor pressures and, thus, lower bioavailability, as well as toxicity, are the main reasons for operational problems. Tests in our research group showed that the biodegradability of formaldehyde as a VOC compound in wood processing industry emissions particularly depends on the waste gas temperature and therefore exhibits high toxicity at thermophilic conditions [103]. In contrast, degradability of methanol and  $\alpha$ -pinene as concomitant substances was successfully shown in literature [99,100]. However, significant formaldehyde emissions were exemplarily observed in hot waste gas streams of biogas plants, combined heat and power plants, wood and pulp industry, plastic coating, and ceramics industry [104].

## 7. Combination of (Non-)Biological Processes

With increasing complexity of waste gas situations multi-stage processes and process combinations are required for successful treatment. For example, three-stage process combinations of acid scrubber, alkaline scrubber and biofilter (rarely a condenser as alternative) represent the state of the art in waste air treatment of sewage sludge drying plants. The waste gas at temperatures of 20–150 °C (depending on the drying process) is firstly cooled adiabatically and cleaned of ammonia, as well as dust particles, in the acidic stage. Separation of H<sub>2</sub>S, mercaptans, and organic acids takes places in the downstream alkaline scrubber, in which cleaning efficiency can be considerably improved by addition of oxidizing agents. The residual VOCs in the waste gas are treated by a biofilter as third stage.

### 7.1. Innovative Combinations of Advanced Oxidation and Biological Processes

In addition to this conventional-looking process combination of two scrubbers and a biofilter, research recently focused on innovative process combinations of biological treatment and advanced oxidation technologies, like non-thermal plasma or UV oxidation. Depending on the sequence of the processes either a partial oxidation of waste gas compounds or a combined finishing and disinfection of the waste gas can be achieved. In the former case, VOCs are partially oxidized by the upstream advanced oxidation process; thus, biological availability of the waste gas compounds is achieved by increasing the water solubility of the intermediates formed [105]. In the latter case, waste gas compounds are mainly mineralized by the biological process and the downstream advanced oxidation process is used as finishing and disinfection step to enable recirculation of the treated air. UV process combinations have recently been used in treatment of chlorobenzene [106,107], dichloromethane [108], ethylbenzene [109], n-hexane [110], PCE [111],  $\alpha$ -pinene [112–114], styrene [39,115], TCE [111], toluene [116,117], and *o*-xylene [118] as single substances or for mixtures of toluene and *o*-xylene [119], a mixture of ethyl acetate, toluene, ethylbenzene, xylene, ethyltoluene, and trimethylbenzene [120], and in a mixture of toluene, ethylbenzene, and xylene isomers [121]. NTP-based combinations were applied in treatment of chlorobenzene [122,123], 1,2-DCE [124], dimethyl disulfide [125], and limonene [126] as single substances and in a couple of different VOC mixtures [34,43,45,105,124,127–132]. Additionally, first tests with magnetic fields as activation step were performed [133,134]. Details of these studies are listed in Table 6 (magnetic fields), Table 7 (UV), and Table 8 (NTP). Data of Tables 6–8 shows that in principle all three process combinations (biological process and magnetic field, UV, or NTP) reveal a higher removal efficiency compared to the sole processes. This achieved efficiency is also above the cumulative efficiencies of the individual processes, i.e., the waste gas components are first partially oxidized in the advanced oxidation process and can then be degraded much more efficiently in the downstream biological stage due to the increased hydrophilicity and, thus, bioavailability of the intermediates. Comparing advanced oxidation processes, the use of these processes leads to an increase in efficiency of 3–15% (magnetic field), 20–50% (UV oxidation), and 25–80% (NTP) compared to the efficiency of the biological process alone. The total achievable efficiencies are the highest in the combination of NTP and a subsequent biological stage. Finally, it should be mentioned that a combination of non-thermal plasma and physical scrubbers for elimination of lipophilic VOCs was also presented [135].

**Table 6.** Recent examples of volatile organic compound (VOC) removal combining static magnetic field and biofiltration.

Compound	Concentration	Efficiency (%)			Magnetic Flux Density (mT)	Gas flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT (s)	Combination	Literature
		$\eta_{\text{Magnetic field}}$	$\eta_{\text{Bio}}$	$\eta_{\text{Combination}}$					
TCE	53.6–326.8 mg·m <sup>-3</sup>	-	29.8–89.8	45.5–92.2	60	0.048	202.5	MF + BTF	[134]
TCE	1000 mg·m <sup>-3</sup>	-	39.4	52.9	20	0.024	405	MF + BTF	[133]

The use of these process combinations appears to be of high interest especially with respect to biologically persistent compounds, since the partial oxidation of the persistent pollutants can significantly increase the biodegradability. For example, in the case of freon R-22, dehalogenated reaction products were identified after exposure to non-thermal plasma, which tend to exhibit higher biodegradability than the original substance [136].

In addition to the requirements of a multi-stage process due to complex waste gas situations, process combinations of UV/NTP with biofiltration processes are often used to reduce both investment costs and operating costs of the overall waste air treatment concept. Compared to a concept based purely on UV oxidation or NTP processes, the process combination has considerably lower electricity costs, since waste gas contaminants are only partially oxidized and intermediates formed can be further treated by biofiltration processes without significant operating costs. Compared to a sole biological process, the treatment of partially oxidized intermediates allows a significantly higher bioavailability and, thus, transformation kinetics compared to the original waste gas contaminants. Hence, the bioprocess can be built significantly smaller and thus more cost-efficient [45]. Furthermore, more stable reactive compounds of the pre-treatment, like ozone, affect the microbial community in the biofiltration process [108,122,125,137]. Thus, a continuous presence of the stressor ozone leads to a lower biomass yield, lower pressure losses, and, thus, a lower tendency to clogging [114,119,122,138].

## 7.2. Combination of Biological Processes

Besides combinations of non-biological and biological processes, multi-stage process combinations of biological processes are also used. These combinations are commonly used in case of VOC mixtures of strongly divergent polarity with high pollutants loads, so that in case of a single-stage biotrickling filter or biofilter long-term stability seems to be questionable due to expected clogging. As an example, a combination of BTF and BF was used for treatment of H<sub>2</sub>S, dimethyl sulfide and dimethyl disulfide [139]. The water-soluble hydrogen sulfide was completely mineralized in the BTF, while the degradation of the two water-insoluble organosulfur compounds took place in the biofilter. Gerl et al. [88] examined biodegradation of a mixture of polar methoxypropyl acetate and lipophilic xylene isomers at a 1:1 ratio of the ester and the aromatics by a combination of bioscrubber and biotrickling filter in comparison to a biofilter as reference system. While the performance of the biofilter was approximately 20% with increasing pressure losses as secondary problem, the combined process revealed long-term stability in operation at elimination efficiencies or more than 70% at the same EBRT.

**Table 7.** Recent examples of VOC removal combining UV oxidation and biofiltration.

Compound	Concentration	Efficiency (%)			Specific Input Energy(SIE, kWh·1000 m <sup>-3</sup> )	Gas Flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT UV (s)	EBRT Bio (s)	Combination	Literature
		$\eta_{UV}$	$\eta_{Bio}$	$\eta_{Combination}$						
Chlorobenzene	250–1500 mg·m <sup>-3</sup>	-	5.4–81.8	31.6–100	45–230	0.1–0.3	28–83	41–122	UV + BF	[106]
Chlorobenzene	250 mg·m <sup>-3</sup>	79–90	30–49	86–100	76.7	0.3	27.6	40.8	UV + BF	[107]
Dichloromethane	420 mg·m <sup>-3</sup>	-	54.6	81.8				60 <sup>(2)</sup> /70 <sup>(3)</sup>		
	650 mg·m <sup>-3</sup>	-	36.3	85.4	44.4	0.81	10	60 <sup>(2)</sup> /70 <sup>(3)</sup>	UV + BTF	[108]
	410 mg·m <sup>-3</sup>	-	4630	66.0				36 <sup>(2)</sup> /46 <sup>(3)</sup>		
Ethylbenzene	3000 mg·m <sup>-3</sup>	36	47	83	2240	0.007	95.3	60	UV + BF	[109]
n-Hexane	33.3 mg·m <sup>-3</sup>	21.5	50–55	65–70	-	0.013	60	34.2	UV + BF	[110]
PCE	32 ppm	96.8	72.8	98.5	44.4	0.09	4	120	UV + BTF	[111]
$\alpha$ -Pinene	130 ppm	20–50	20–50	100	-	0.3–0.39	-	-	UV + BF	[112]
	45 ppm	73.3–94.3	-	-						
	95 ppm	49.5–86.7	-	-	9.7 W·cm <sup>2-2</sup>	0.064–0.151	1.5–3.5	- <sup>(1)</sup>	UV + BF	[113]
$\alpha$ -Pinene	145 ppm	36.2–66.7								
$\alpha$ -Pinene	600–1500 mg·m <sup>-3</sup>	-	25–76	57–98	18–40	0.9–2.0	18	17/23/39	UV + BTF	[114]
$\alpha$ -Pinene	1500–1600 mg·m <sup>-3</sup>	15	70	80	80	0.45	18	27/40/81	UV + BTF	[114]
		15	71	84					UV+BF	
Styrene	2.11–2.525 g·m <sup>-3</sup>	15–16	33–34	57–59	44.4	0.81	10	30	UV + BTF	[115]
Styrene	2–4 g·m <sup>-3</sup>	-	27.2–61.8	64.8–87.3	44.4	0.81	10	20/30/60 <sup>(2)</sup> 30/40/70 <sup>(3)</sup>	UV + BF vs. BF	[115]
Styrene	107 mg·m <sup>-3</sup>	8	93	93–98	1.85	67	10	31	UV + BTF	[39]
TCE	307.5 ppm	97.5	50	94–100	44.4	0.09	4	120	UV + BTF	[111]
Toluene	650 mg C·m <sup>-3</sup>	59	51–69	92	31.6	1.14	2.7	45	UV + BF	[116]
	1200 mg C·m <sup>-3</sup>	74	74–79	84						



Table 7. Cont.

Efficiency (%)										
Compound	Concentration	$\eta_{UV}$	$\eta_{Bio}$	$\eta_{Combination}$	Specific Input Energy(SIE, kWh·1000 m <sup>-3</sup> )	Gas Flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT UV (s)	EBRT Bio (s)	Combination	Literature
Toluene	210–500 mg·m <sup>-3</sup>	53–58.2	92.3	94.1–96.7	40	0.1	13.3	108	UV + BF	[117]
o-Xylene	60–220 mg·m <sup>-3</sup>	10–35	50–60	73–99	41.7–86.0	0.186–0.384	0.5/1.2	35/75	UV + BF	[118]
Toluene, o-Xylene	44.7–121.4 ppm 44.7–121.4 ppm	16–36 10–14	42–77 22–27	90–100 92–99		0.38–0.56	0.5–1.5	45 30	UV + BF	[119]
Ethyl acetate, Toluene, Ethylbenzene	2.4–24.8 5.4–63.9 38.4–150.7	94.5 75.0 81.6	76.5 88.5 72.8	99.0 98.0 94.7						
Xylene Ethyltoluene	21.3–233 0–61.9	93.2 91.0	75.0 64.1	98.3 97.6	0.32	3000	10.8	7.2	BTF + UV	[120]
Trimethylbenzene VOC total	0.6–32.5 46.8–485.9	88.8 86.8	71.4 74.1	97.6 96.6						
Toluene, Ethylbenzene, m-Xylene, o-Xylene, p-Xylene	- - - - -	72.7 77.2 71.9 74.8 60.0	- - - - -	>99 >99 >99 >99 >99	-	-	-	-	UV + BTF	[121]

-: no data; <sup>(1)</sup>: installation of biofiltration was suggested but not implemented; <sup>(2)</sup>: BF or BTF combined with UV; <sup>(3)</sup>: BF or BTF solely installed.

**Table 8.** Recent examples of VOC removal combining NTP and biofiltration.

Compound	Concentration	Efficiency (%)			Specific Input Energy (SIE, kWh·1000 m <sup>-3</sup> )	Gas Flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT NTP (s)	EBRT Bio (s)	Combination	Literature
		$\eta_{\text{NTP}}$	$\eta_{\text{Bio}}$	$\eta_{\text{Combination}}$						
Chlorobenzene	500 mg·m <sup>-3</sup>	58	68.7	97.6	972			60		
	500 mg·m <sup>-3</sup>	88.1	68.7	98.8	3194			60		
	1000 mg·m <sup>-3</sup>	-	57.8	94.5	3194	0.15	4.32	60	NTP + BTF	[122]
	1000 mg·m <sup>-3</sup>	-	43	67.2	3194			30		
Chlorobenzene	750 mg·m <sup>-3</sup>	73–100	-	-	972–3200	0.011–0.033	5–15	- <sup>(1)</sup>	NTP + BTF	[123]
	200 mg·m <sup>-3</sup>	-	95	95–100						
1,2-DCE	800 mg·m <sup>-3</sup>	-	39	85	0.97	0.053	3.24	90	NTP + BTF	[124]
Dimethyl disulfide	76.2	16.1–24.5	5.8–15.9	87.7–96.3	-	0.1	10.6	10.3	NTP + BTF	[125]
Limonene	0.5 ppm	100	-	-	-					
	30 ppm	30	-	-		20	-	-	NTP + BF	[126]
1,2-DCE, n-Hexane	200 mg·m <sup>-3</sup>	-	70	92–95						
	200 mg·m <sup>-3</sup>	-	60	82–85	0.97	0.053	3.24	90	NTP + BTF	[124]
Toluene, p-Xylene, Ethylene	116.5 ppm	-	86–94	95.2						
	132.4 ppm	-	66–70	95.8	-	1.2	-	-	NTP + BTF	[127]
	127.5 ppm	-	50–54	81.3						
Toluene, n-Heptane, p-Xylene, Ethylbenzene, Benzene	95.6	-	60	>93						
	49.4	-	23	81–91						
	60.8	-	87	>93	25.6	0.15	6	60	NTP + BF	[105]
	47.3	-	76	>93						
	36.6	-	28	>93						



Table 8. Cont.

		Efficiency (%)			Specific Input Energy (SIE, kWh·1000 m <sup>-3</sup> )	Gas Flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT NTP (s)	EBRT Bio (s)	Combination	Literature
Compound	Concentration	$\eta_{\text{NTP}}$	$\eta_{\text{Bio}}$	$\eta_{\text{Combination}}$						
Ethanol,	233–604	15–35	-	-						
Ethyl acetate,	216–778	7–8	-	-						
Benzene,	6–72	4–6	-	-	25.6	3·10 <sup>-4</sup>	0.98	- <sup>(1)</sup>	NTP + BF	[130]
Toluene,	53–344	14–20	-	-						
Octane	65–352	22–32	-	-						
Ethanol,	233–604	83–88	-	-						
Ethyl acetate,	216–778	49–90	-	-						
Benzene,	6–72	35–64	-	-	125	3·10 <sup>-4</sup>	0.98	- <sup>(1)</sup>	NTP + BF	[130]
Toluene,	53–344	68–91	-	-						
Octane	65–352	80–94	-	-						
Ethanol,	233–604	84–95	-	-						
Ethyl acetate,	216–778	82–100	-	-						
Benzene,	6–72	65–92	-	-	250	3·10 <sup>-4</sup>	0.98	- <sup>(1)</sup>	NTP + BF	[130]
Toluene,	53–344	96–100	-	-						
Octane	65–352	97–100	-	-						
n-Butanol,	4 mg C·m <sup>-3</sup>	50–60	-	93–100						
2-Butoxyethanol,	1 mg C·m <sup>-3</sup>	100	-	100	2.2	750	0.3	10.6	NTP + BTF	[45]
Odor	700–3600 OU·m <sup>-3</sup>	42.8–91.7	-	94.3–97.2						

Table 8. Cont.

Compound	Concentration	Efficiency (%)			Specific Input Energy (SIE, kWh·1000 m <sup>-3</sup> )	Gas Flow (m <sup>3</sup> ·h <sup>-1</sup> )	EBRT NTP (s)	EBRT Bio (s)	Combination	Literature
		$\eta_{\text{NTP}}$	$\eta_{\text{Bio}}$	$\eta_{\text{Combination}}$						
Butanone,	54 mg C·m <sup>-3</sup>	40.7	100	100	2.73	60	-	9	BF + NTP	[131]
Ethyl acetate,	15 mg C·m <sup>-3</sup>	53.3	100	100						
Dimethyl disulfide,	1 mg C·m <sup>-3</sup>	100	100	100						
Toluene,	12 mg C·m <sup>-3</sup>	41.7	71.4	83.3						
Limonene	59 mg C·m <sup>-3</sup>	89.8	33.3	93.2						
Dimethyl disulfide,	1.4 mg·m <sup>-3</sup>									
Carbonyl sulfide,	0.04 mg·m <sup>-3</sup>									
Dimethyl sulfide,	<0.2 mg·m <sup>-3</sup>	57–72	52	80	-	160	12	-	BF + NTP	[132]
Dimethyl trisulfide,	<0.2 mg·m <sup>-3</sup>									
Odor	86,900–97,900 OU·m <sup>-3</sup>									
VOC mixture	80–160 mg·m <sup>-3</sup>	25	25	50	0.7	250	4.5	-		
Odor	9200–14,200 OU·m <sup>-3</sup>	25	25	50	0.7	250	4.5	-	NTP + BF	[34]
	65,000 OU·m <sup>-3</sup>	40	-	-	0.04	14,000	1.5	-		

-: no data; <sup>(1)</sup>: installation of biofiltration was suggested but not implemented; <sup>(2)</sup>: BTF not installed due to high performance of NTP.

## 8. Conclusions

International studies expect a further increase in greenhouse gas emissions over the next 20 years despite increasing efforts to improve energy efficiency in technical processes. In order to meet the international goal of limiting global warming to 2 °C, industrialized countries will have to reduce emissions of CO<sub>2</sub> equivalents by more than 90%. That way, mankind simply can no longer afford to generate immense CO<sub>2</sub> emissions by waste gas treatment technologies. Development and application of sustainable, energy and resource efficient processes for waste air treatment is obligatory. By a large extent, existing biological waste air treatment techniques meet these requirements. According to the limitations mentioned above, these processes reveal moderate performance levels in case of simultaneous degradation of VOC mixtures and, therefore, have high space requirements. These problems and corresponding marketing obstacles, as well, can be counteracted by the innovative approach of combining advanced oxidation and biofiltration processes, so that compact, energy-efficient, and low-emissions technologies are available for agricultural, municipal, and industrial waste gas applications. Although biological processes, UV oxidation, and NTP processes, as stand-alone processes, have been intensively described, combinations of these processes have rarely been discussed. Experiences in the semi- and large-scale technical scale are even more limited. Future research will therefore have to deal with the process stability (NTP process), the stable transformation efficiency (UV process), the influence of interfering components (especially dusts and aerosols in water vapor saturated emission situations; NTP and UV processes), and the in-depth effect of reactive oxidative components on the performance and growth of the microbial consortium of the subsequent biological stage.

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