

Article

Comparative Evaluation of Selected Biological Methods for the Removal of Hydrophilic and Hydrophobic Odorous VOCs from Air

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Abstract: Due to increasingly stringent legal regulations as well as increasing social awareness, the removal of odorous volatile organic compounds (VOCs) from air is gaining importance. This paper presents the strategy to compare selected biological methods intended for the removal of different air pollutants, especially of odorous character. Biofiltration, biotrickling filtration and bioscrubbing technologies are evaluated in terms of their suitability for the effective removal of either hydrophilic or hydrophobic VOCs as well as typical inorganic odorous compounds. A pairwise comparison model was used to assess the performance of selected biological processes of air treatment. Process efficiency, economic, technical and environmental aspects of the treatment methods are taken into consideration. The results of the calculations reveal that biotrickling filtration is the most efficient method for the removal of hydrophilic VOCs while biofilters enable the most efficient removal of hydrophobic VOCs. Additionally, a simple approach for preliminary method selection based on a decision tree is proposed. The presented evaluation strategies may be especially helpful when considering the treatment strategy for air polluted with various types of odorous compounds.

Keywords: air deodorization; comparison; biofiltration; volatile organic compound; decision tree

1. Introduction

1.1. Air Deodorization by Biological Methods

Pollution caused by VOCs and other air pollutants, especially odorous compounds, including organic and inorganic compounds, including nitrogen-containing compounds (NH₃, amines) and sulfur-containing compounds (H₂S, mercaptans, sulfides), have adverse effects on both humans and the environment [1–3]. Odorants have been proven to pose toxic effects on human health as well as to negatively influence the quality of life. Thus, a lot of attention has been devoted to the emission control of VOCs and other odorous pollutants in recent years [4,5].

Odorous compounds are emitted from many sectors of human activity, including wastewater treatment plants, communal waste landfills, agriculture and plenty of industrial facilities e.g., crude oil refineries, pulp and paper mills and various chemical industries. Additionally, the need for indoor air treatment is gaining interest [6–11]. Such emissions are controlled by various deodorization techniques. The following methods are most often applied: thermal oxidation, absorption (in water or with chemical reaction), adsorption, masking and biological techniques. The selection of the most appropriate methods is case-specific and depends on the properties of a gas stream, concentration of pollutants, emission source and the desired level of gas deodorization [1,5,12–15].

Among the aforementioned gas deodorization methods, the group of biological methods seems to be superior, especially with the perspective of environment, economy and sustainable development. Biological methods are characterized by low operating costs, low secondary pollution and very high purification efficiency when treating large volumes of gases containing low and medium concentrations of pollutants [16–18]. The process of biological gas treatment is most commonly referred to as “biofiltration”. The most common apparatus intended for air biofiltration include conventional biofilters (BF), bioscrubbers (BS) and biotrickling filters (BTF) [19–21]. Beside the differences in the apparatus design, the mechanism of the air treatment process is similar.

The process of biofiltration is based on the degradation of gas contaminants as a result of biological activity of microorganisms inhabiting the porous packing of the biofilter. The microbes are especially present in so-called biofilm developing over the surface of packing elements. The mechanism of the process consists in the diffusion of pollutants from the gas phase to the biofilm surrounded by a liquid phase. This liquid phase may either be supplied as a trickling liquid (biotrickling filter) or may result from former gas humidification (conventional biofilter). Thus, pollutants from the gas phase are either adsorbed on or absorbed by the biofilm and then undergo biodegradation. The stream of treated (cleaned) air leaves the biofilter together with formation of CO_2 , H_2O and biomass as biodegradation products [5,22,23] (Figure 1).

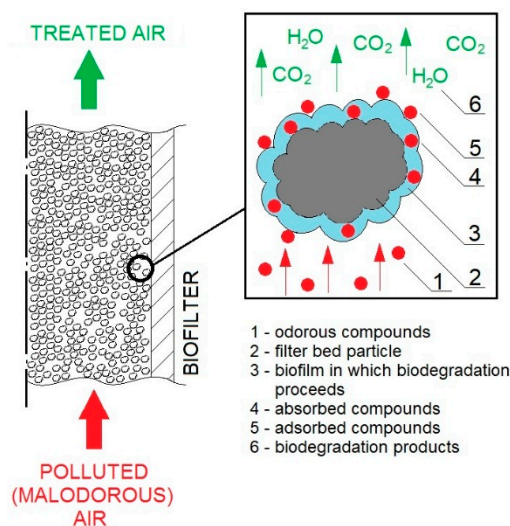


Figure 1. General mechanism of biofiltration.

A conventional biofilter is usually packed with a bed made of organic materials (wood chips, cones, peat) that are naturally colonized by microorganisms capable of degrading various air pollutant impurities. The contaminated gas is humidified in a separate chamber prior to entering the biofilter [24,25].

In the case of BTF, the filter packing is made of inert natural or synthetic materials (ceramic elements, polyurethane foam, lava rock). Such a packing requires inoculation of microbes prior the process start-up and the role of the packing is mainly to give a physical support for the biofilm development. BTF uses a trickling liquid, usually enriched with additional nutrients for microorganisms. Such a configuration enables the pollutants' adsorption/absorption and biodegradation in one apparatus [19,26].

Biological air treatment in bioscrubbers consists in two main processes: absorption of gas contaminants in the liquid phase and biodegradation of these pollutants with the use of additional bioreactors, enabling the liquid regeneration, aeration and circulation [1].

Typical processes of biological air treatment are designed for the removal of water-soluble compounds. The efficiency and the rate of biofiltration of hydrophilic compounds is mainly dependent on the rate of their biodegradation in the biofilm. However, when hydrophobic gas pollutants are considered, the biofiltration efficiency depends drastically on the transfer rate of the components from the gas phase to the liquid, usually aqueous phase [27–29]. This is why biofiltration of hydrophobic

compounds precedes with much lower efficiency than hydrophilic compounds and currently the improvement of the efficiency of hydrophobic compounds biofiltration is a challenge in the design of the biological treatment processes [16,30].

Depending on the air pollutant type and concentrations, one of above discussed biological treatment methods may be chosen. Biological methods of air treatment have been increasingly investigated since the beginning of the 1990s. Firstly, the research concentrated mainly on the effect of basic parameters on the process performance [5,22,31–33]. Furthermore, more in-depth research on the biological aspects have been developing, including the biotechnological assays for the composition of microbial composition of the biofilm. Currently, the research is focused on the improvement of the removal of hydrophobic air pollutants, biofiltration of which is usually limited by low mass transfer rate from the gas to the liquid (biofilm) phase. Parallel to typical experimental investigations, also in the semi-pilot and pilot scales, modeling of these processes is investigated and developed [34–37]. Examples of the latest research include the application of fungi for biofiltration [38], modeling of serial biofiltration unit [39], upgrading of biogas in biofilters [40] or biomass overgrowth control strategies [41]. Research in the field of biotrickling filtration is devoted to e.g., small-scale applications for indoor air treatment [42], application of new strands of microbes [43] or process scaling-up [44]. Examples of current research on bioscrubbers are the emission control of NH_3 from agricultural applications [45] or treatment of air polluted with H_2S and others [46–48].

The selection of the most suitable treatment method is a function of several factors, especially when mixtures of hydrophilic or hydrophobic compounds are considered. In this perspective, a broad set of data or mathematical tools aiding the decision-making may be of importance. In the literature, several papers devoted to comparison of various deodorization methods [12,49–51] may be found. However, these papers mainly present the experimental results and economic analyses or compares different processes/process conditions, leaving the reader with general ideas about the processes discussed. Therefore, the development of a comparative tool for the selection of the treatment method is of both scientific and practical importance.

1.2. Assessment of Biological Methods of Air Treatment

In this paper, two approaches of comparative analysis or selection criteria for the treatment of air polluted with various volatile compounds are investigated. An evaluation methodology based on the procedure described by Oliva et al. [15] is proposed in this paper. The adopted method is derived from a pairwise comparison model, using numerical judgments from an absolute scale of numbers. This method was initially proposed by Henri Lebesgue and it enables the comparison of the examined objects, with the aid of the analysis of their properties and selection of the appropriate scale [52]. It is based on comparing elements, in order to receive their assessment, based on a preference. This method also allows to choose which of the analyzed elements are characterized by a larger number of the selected quantitative properties. Pairwise comparison is often a crucial step in multi-criteria decision analysis [53]. This method was chosen due to the fact that it allows for a fair division and balance of the final value into individual components [54,55]. Similar comparisons are made in other fields of science, e.g., assessment of fuels [56], voting system [57–59], psychology [60,61], artificial intelligence system [62] and others [63–65]. The key element is to provide a tool for making an objective comparison taking into account the division into various aspects of a given field.

Additionally, a decision tree procedure for the selection of a treatment technology is proposed. Decision tree is a graphical method of supporting the decision-making process, which can be used for different types of modeled variable, i.e., continuous or discrete. The goal is to create a model that predicts the value of a target variable based on several input variables [66,67]. The main task of the decision tree method is to generate mutually exclusive regions in which there are as many samples as possible classified into one group. These regions are created by successive divisions of the training set, using binary logical rules [68]. The learning process is carried out to obtain the most homogeneous group of sample sets. As an algorithm output, decision trees can provide two types of information:

the description of which group the examined object is located in or the probability of belonging to a given group [69].

1.3. Aim of Investigations

The aim of this paper is to provide a comprehensive assessment of the selected biological methods for the removal of various volatile compounds from air i.e., biofiltration, biotrickling filtration and biscrubbing with the use of a pairwise comparison model as well as decision tree procedure.

This paper presents three interesting elements from the novelty viewpoint. Firstly, the paper revises the results of selected recent research on the application of biological methods for selected air pollutants, thus it may serve as a source of experimental results. Secondly, the authors adopted a comparison procedure for evaluating the holistic effects of performance, costs and technical aspects of treating air with a given method. Thirdly, a simple tool for preliminary selection of the method is proposed. Such an approach is hardly met in the literature and presents useful way of comparing processes.

2. Materials and Methods

2.1. Data Collection

For the purpose of calculations for a pairwise model as well as the development of a decision tree, literature data was used. For literature search, Science Direct, PubMed and MDPI databases were applied. Articles from last 10 years were selected with the priority of choosing, however older articles were used as well (depending e.g., on the target chemical compound in a given treatment method). Selection of literature data was applied according to the target compound so as to collect data suitable for comparison purposes. Data applied in calculations were taken directly from the literature without normalization procedures.

2.2. Comparative Analysis

A comparative analysis of biological methods in the perspective of the removal of hydrophilic, hydrophobic or inorganic odorous gases is presented with the use of a pairwise comparison model [52]. The numerical procedure used is based on the quantification of a set of parameters previously classified in clusters. It is used to select the best biological process of air purification from impurities. The results obtained on the basis of the semi-quantitative ranking of selected parameters pointed to the advantages and disadvantages of the processes studied. For the purpose of comparison, the focus is on the process performance, technical, economic as well as environmental aspects. The applied method consists in assigning specific values to all highlighted alternatives. Calculations are realized in four main stages i.e., selection of the main criteria determining the deodorization process (C_1 – C_4) as well as selection of sub-criteria affecting the main criteria ($C_{1.1}$ – $C_{4.2}$); assigning weights to each criterion and sub-criterion (w_i and w_{ij}); assigning indicators to sub-criteria (values in the range between 0 and 1); calculation of the results of all alternatives.

The results of all alternatives were obtained using the following equations [28]:

$$R_i = w_{i,j1} \cdot r_{i,j1} + w_{i,j2} \cdot r_{i,j2} + \dots + w_{i,jn} \cdot r_{i,jn}; \quad i = 1, \dots, n; j = 1, \dots, n \quad (1)$$

$$R = w_{i1} \cdot R_{i1} + w_{i2} \cdot R_{i2} + \dots + w_{in} \cdot R_{in}; \quad i = 1, \dots, n; j = 1, \dots, n \quad (2)$$

where:

- $w_{i,j}$ - weight of a given sub-criterion $C_{i,j}$,
- $r_{i,j}$ - result of an alternative to the sub-criterion $C_{i,j}$,
- R_i - result of an alternative to the criterion C_i ,
- w_i - weight of given criterion $C_{i,j}$,

- R - overall result of the alternative,
 n - number of analyzed criteria.

2.3. Decision Tree for the Preliminary Selection of the Deodorization Method

In the second part of this paper, a decision tree procedure was applied. The decision tree has been built based on data from 57 biological processes of removing various chemical compounds from the air i.e., conventional biofiltration, biotrickling filters and bioscrubbers. The data set is presented in Table 5. The following process parameters were selected as input variables: inlet concentration, Henry's constant and empty bed residence time (EBRT). As the result of the decision tree, the most optimal air purification method may be indicated. All calculations were performed in RStudio 1.1.463 [70] using the 'rpart' library [71].

The main advantages of using decision trees are their non-parametric character as well as the automatic identification of the most significant variables by the algorithm and the elimination of statistically insignificant variables. Moreover, the mathematical transformation (e.g., logarithm) of one or more explanatory variables does not change the structure of the tree which is changed only by threshold values. Among the disadvantages should be indicated: a slight modification of the training set (e.g., removal of several observations) can radically change the structure of the tree. In addition, in one step the tree can divide the space only in relation to one variable (in other words: the dividing lines are always perpendicular to the divided axis in the variable space).

2.4. Calculation of Process Performance Parameters

Removal efficiency and empty bed residence time were selected as parameters presenting process performance. These parameters indicate both the degree of the removal of odorous compounds from air as well as gives the information about the rate of the process course based on the air flow rate and the capacity of a bioreactor. Values of RE and EBRT were either taken directly from literature data or calculated according to Formulae (3) and (4):

$$RE = \left(\frac{C_{in} - C_{out}}{C_{in}} \right) \cdot 100\% \quad (3)$$

$$EBRT = \frac{V}{Q} \quad (4)$$

where:

- C_{in} - inlet concentration of target compound,
- C_{out} - outlet concentration of target compound,
- V - volume of the filter bed,
- Q - inlet gas flow rate.

3. Results and Discussion

3.1. Selection of Comparison Main Criteria

In order to characterize biological methods of air deodorization, four main criteria were selected: efficiency/process performance, costs, technical aspects and problems as well as the environmental impact. The aforementioned criteria have been chosen in the perspective of possibly complete evaluation of each analyzed treatment method. The selection criteria are presented in Figure 2.

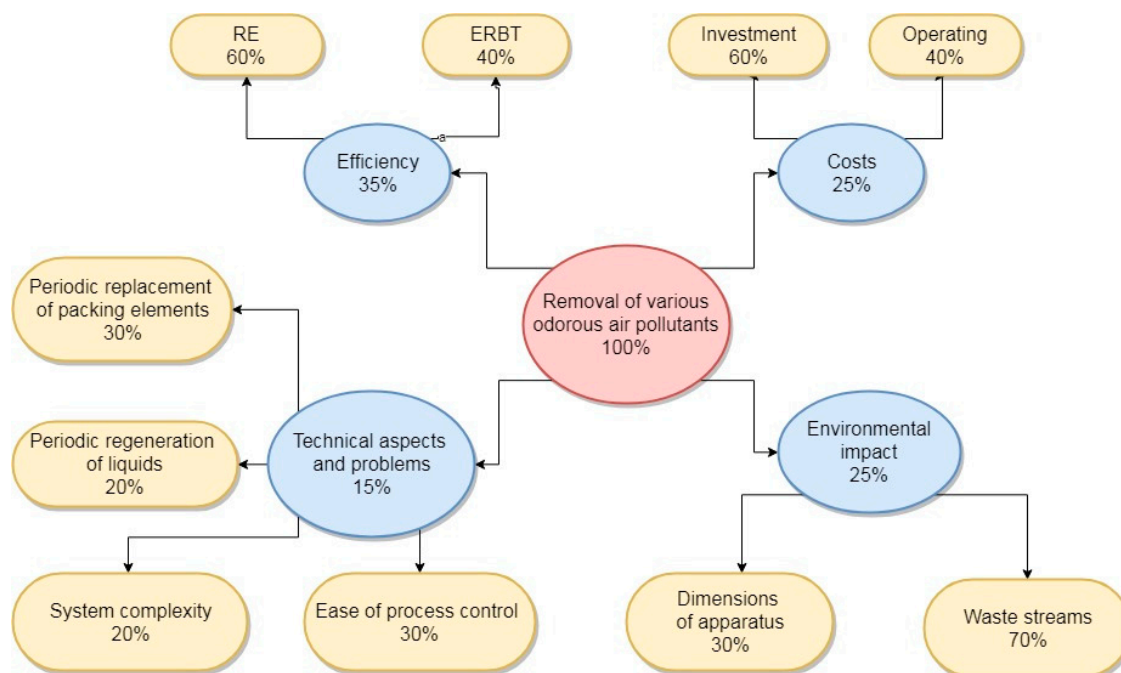


Figure 2. Hierarchy of selected criteria and sub-criteria.

Each of the selected criteria has been divided into second-order criteria. As part of the performance criterion, two sub-criteria were distinguished: removal efficiency (RE) and empty bed residence time (EBRT) [1,4,5,72]. Values of RE inform about the degree of air purification and enable to differentiate among the process performance of analyzed methods. Comparison of the values of empty bed residence time allows for the assessment of the rate of the assessed treatment processes. The criterion related to the costs include investment and operating costs as sub-criteria [12,19,73,74]. These costs are crucial for the process realization, especially when industrial-scale applications are considered.

The authors of this manuscript have proposed two more criteria related to the deodorization strategy i.e., technical problems arising during the process [1,5,19,20,31] as well as the environmental impact of the technology [12,73–75]. Among the technical aspects and possible technical problems, which are thought to greatly affect the long-term process operation, following sub-criteria have been distinguished: periodic replacement of packing elements, the need for periodic regeneration of the liquid, the complexity of the system, as well as the ease of process control and the ability to adapt the system to changes in the charge of current regulation. These sub-criteria have been proposed based on a literature review.

The following sub-criteria have been proposed when the environmental impact of the deodorization technique is considered: dimensions of the apparatus and waste streams generated during the treatment process. Depending on the flow rate of treated air streams, the dimensions of the apparatus may greatly differ (laboratory to industrial scale) [75]. In this perspective, conventional biofilters, especially the open-type, occupy plenty of land area, resulting in a high foot-print. Sizes of biotrickling filters are much smaller. On the other hand, the operation of bioscrubbers generates high volumes of liquid containing compounds absorbed from air and these liquid streams must be further processed [1,5]. This is why the sub-criteria including apparatus size and waste streams generation have been proposed.

3.2. Assigning of Weights to Criteria and Sub-Criteria

The choice of criteria and sub-criteria was made on the basis of a literature review. A literature review was prepared using peer-reviewed journals, other professional literature [72] as well as conference materials. When choosing and proposing the values of the criteria, the authors took

advantage of the process experience of the team [13,21,25,76]. Having the above set of data, the authors attributed the criteria and sub-criteria to the weights, reflecting the importance of the aspect to the entire process of biological purification of air from pollutants. The values of weights are given in Table 1.

Table 1. Weight of the criteria and sub-criteria and ranges of sub-criteria indicators.

Criterion	w_i (%)	Sub-Criterion	$w_{i,j}$ (%)	Range of the Indicators (ID)
C ₁ (Efficiency)	35	C _{1.1} (RE)	60	0 ... 1 (low ... high)
		C _{1.2} (EBRT)	40	1 ... 0 (low ... high)
C ₂ (Costs)	25	C _{2.1} (Investment costs)	60	1 ... 0 (low ... high)
		C _{2.2} (Operating costs)	40	1 ... 0 (low ... high)
C ₃ (Technical aspects and problems)	15	C _{3.1} (Periodic replacement of packing elements)	30	1 ... 0 (slow ... fast)
		C _{3.2} (Periodic regeneration of liquids)	20	1 ... 0 (rarely ... often)
		C _{3.3} (System complexity)	20	1 ... 0 (low ... high)
		C _{3.4} (Ease of process control)	30	0 ... 1 (complex ... simple)
C ₄ (Environmental impact)	25	C _{4.1} (Dimensions of apparatus)	30	1 ... 0 (small ... big)
		C _{4.2} (Waste streams)	70	1 ... 0 (yes ... no)

3.3. Assigning of Indicators

Based on the literature data presented in Table 2; in Table 3, indexes were assigned to indicators taking into account the “bigger is better” principle, i.e., values were assigned from the range from 0 to 1, where the value of 1 is the most favorable considering the whole group of analyzed methods, as given in Table 2. Each criterion and sub-criterion has been transformed into an indicator (values assigned to sub-criteria C_{1.1}–C_{4.2}).

The differences in the hydrophilic character of the compounds were estimated using Henry’s law constant. The greater the Henry’s law constant, the greater the volatility and the lower the solubility of a compound, which is valid for dilute solutions and non-reacting gases at near ambient pressure and temperature. The Henry’s law constant (H_C) can be expressed as the dimensionless ratio between the aqueous-phase concentration C_a of a species and its gas-phase concentration C_g [77]:

$$H_C = \frac{C_g}{C_a} \quad (5)$$

Taking into account Equation (4), values of dimensionless Henry’s constant in Table 2 were calculated using Formula (5):

$$H_C = \frac{RF}{H} \quad (6)$$

where:

RF - recalculation factor equal to 4.03395×10^{-4} , taken from [77],

H - Henry’s constant given as ($\text{mol} \cdot \text{m}^{-3} \cdot \text{Pa}^{-1}$) in Table 2.

Table 2. Literature data for determining the values of sub-criteria 1.1 and 1.2.

Compounds	Conventional Biofilter						Biotrickling Filter				Bioscrubber			
	H (mol·m ⁻³ ·Pa ⁻¹)	H _c (-)	C _{in} (mg·m ⁻³)	RE (%)	EBRT (s)	Reference	C _{in} (mg·m ⁻³)	RE (%)	EBRT (s)	Reference	C _{in} (mg·m ⁻³)	RE (%)	EBRT (s)	Reference
Hydrophilic	-	-	-	99	-	[73]	-	99	-	[73]	-	99	-	[73]
butanol	1.2 [77]	3.4 × 10 ⁻⁴	900–2600	>73	60	[78]	400–1200	15–99	60–124	[79]	-	98–100	48	[80]
aniline	1.1 [5]	3.7 × 10 ⁻⁴	-	-	-	-	300	<99	42–166	[81]	-	-	-	-
isopropanol	1.3 [77]	3.1 × 10 ⁻⁴	1000–8000	81	94.2	[82]	20–65 (g m ⁻³ ·h ⁻¹)	<95	14–160	[83]	200–500	99	-	[84]
ethanol	9.0 [77]	4.5 × 10 ⁻⁵	3700	63–85	101	[85]	470	~80	66	[86]	-	80–99	-	[87]
methanol	2.0 [77]	2.0 × 10 ⁻⁴	-	>95	25	[88]	300–37,000	65	20–65	[89]	50–100	69–81	600	[90]
			0.79–3.3	93.33	38	[91]					-	75	2.5	[92]
Hydrophobic	-	-	-	75	-	[73]	-	50	-	[73]	-	50	-	[73]
hexane	6.1 × 10 ⁻³ [77]	6.6 × 10 ⁻²	500–11,000	79	60	[93]	600	57–91	8–30	[94]	6200	70	420	[92]
methane	1.4 × 10 ⁻⁵ [77]	2.9 × 10 ⁻¹	200–10,000	59–76	60	[95]	0–500	~40	240	[97]	-	5–25	1.6	[98]
ethylene	5.9 × 10 ⁻⁵ [77]	6.8	4581–4908	43	257	[96]	8–100 (g m ⁻³ ·h ⁻¹)	70–95	30	[27,100]	-	-	-	-
α-pinene	2.9 × 10 ⁻⁴ [77]	1.4	331	100	2160	[99]	-	-	14–60	[102]	-	-	-	-
			100–450	90	42	[101]								
			4227	47–67	78	[103]								
styrene	2.7 × 10 ⁻³ [77]	1.5 × 10 ⁻¹	0.1–0.9	90	9–18	[104]	800–3300	95	60–120	[105]	-	-	-	-
			0.85	97	1845	[106]	55–312	90	15–30	[107]	-	-	-	-
			2.3	90	137–825	[108]	2–1128	99	400	[109]	-	-	-	-
toluene	1.5 × 10 ⁻³ [77]	2.7 × 10 ⁻¹	1.9	>80	21.6	[111]	2200	<99	16.2	[112]	3300	89	-	[110]
			6	<98	70	[113]	1000	60	57	[114]				
Inorganic														
ammonia	5.5 × 10 ⁻¹ [77]	7.3 × 10 ⁻⁴	14–350	92–100	17	[115]	9.6	82	1.2	[116]	14	99	142	[117]
			20–100				20–100	99	960	[118]				
H2S	1.0 × 10 ⁻³ [77]	4.0 × 10 ⁻¹	7–3750	100	23–200	[119]	300–650	65–100	53–79	[120]	14–140	98	12–32	[47]

Table 3. Literature data for determining the values of sub-criteria 2.1–4.2 (investment and operating costs are presented for a flowrate 50,000 m³ h⁻¹).

Criteria and Sub-Criteria	Units	BF	BTF	BS	Reference
2 Costs					
Investment					
2.1	(€ per m ³ ·h ⁻¹)	6	11	4	[73]
	(€ per m ³ ·h ⁻¹)	5	10	4	[12]
Operating					
2.2	(€ per m ³ ·h ⁻¹)	2	1.2	3.6	[74]
	(€·10 ⁻⁴ ·m ⁻³)	0.2	0.1	0.28	[73]
3 Technical aspects and problems					
Periodic replacement of element					
3.1	Packing material (years)	2	10	10	[73]
	Annual/material-reagents (kg m ³ ·h)	4	0.1	0.1	[73]
	Packing material (%)	47	44	4	[74]
Periodic regeneration of liquids					
3.2	Annual water consumption (L·m ⁻³ ·h·10 ²)	2.4	6.3	3.3	[73]
	Water that can be replaced with secondary effluent (-)	Possible	Possible	Impossible	[73]
Complexity of the system					
3.3	Basic elements of the apparatus (-)	-Humidification chamber -Packed bioreactor	-Packed bioreactor -Liquid container -Pump	-Absorption column -Pump -Absorbent tank	[5]
	number of basic elements of the apparatus (-)	2	3	3	(-)
Ease of process control					
3.4	impact on the control process	Low	High	Medium	(-)
	Customization at work (-)	Impossible	High	Medium	(-)
4 Environmental impact					
Dimensions of the apparatus					
4.1	The size of the apparatus [m ² ·m ⁻³ ·h·10 ²]	1.75	0.25	0.1	[73]
	Surface area (-)	High	Low	Low	[87]
Waste streams					
4.2	Use of filling (-)	Possible	Impossible	Impossible	[73]
	The possibility of replacing water with sewage (-)	Possible	Possible	Impossible	[12]
	Volume of liquid vol<<VOL	-	vol	VOL	[12]
-	-	Very low	Medium	High	[-]

The values of indicators (Figure 3) are proposed and derived from data presented in Table 2; Table 3 as well as additional literature data [1,19,20,31].

The values of indicators for investment and operating costs are presented in Figure 3a. The obtained results indicate that BSs are characterized by the lowest investment costs (ID = 1), while the highest investment costs are found for BTFs (ID = 0.3). Such a result may be justified by the number of basic elements of apparatus included in the system (BTF contains the highest number of elements, i.e., biofilter chamber, packed bed, circulating pumps, liquid containers, trickling system).

Interestingly, the lowest operating costs are associated with BTF operation (ID = 1), while the highest are attributed to bioscrubbers. The operating costs of BFs (ID = 0.66) are medium in the compared group of methods. It mainly results from the periodic maintenance requirements, as described more precisely by the technical-aspects-and-problems sub-criterion. The BS process is associated with the highest operating costs (ID = 0.33).

Figure 3b shows the values of indicators for periodic replacement of packing elements, periodic regeneration of liquid, complexity of the system and possibility of process control. The literature indicates that the packing material in BTF (ID = 1) should be replaced the least frequently. It is mainly because of the fact that inert, ceramic or synthetic materials are applied as packing materials and their durability is much higher than for natural packing materials, used in BFs [1,5]. By contrast, the results indicate that BF requires the most frequent replacement of the packed bed (ID = 0.2) because BFs are usually packed with natural organic packing materials. The replacement of liquid, due to apparatus construction, does not typically apply for BF (ID = 1). Interestingly, in terms of BS and BTF it is possible to replace water with so called secondary wastewater [73]. Periodic liquid replacement is the biggest problem in the case of BS (ID = 0.3), despite the fact that water consumption compared to the other two processes is at an average level. This results from the inability to use “wastewater” for secondary

use. BTF and BS are characterized by the greatest complexity of the system ($ID = 0.8$). They have three or more basic elements of the apparatus (e.g., bioreactors, packing elements, pumps, trickling system, absorption column etc.). BF, on the other hand, is a relatively simple system ($ID = 1$), having only two basic elements in its construction (humidification chamber and biofilter itself).

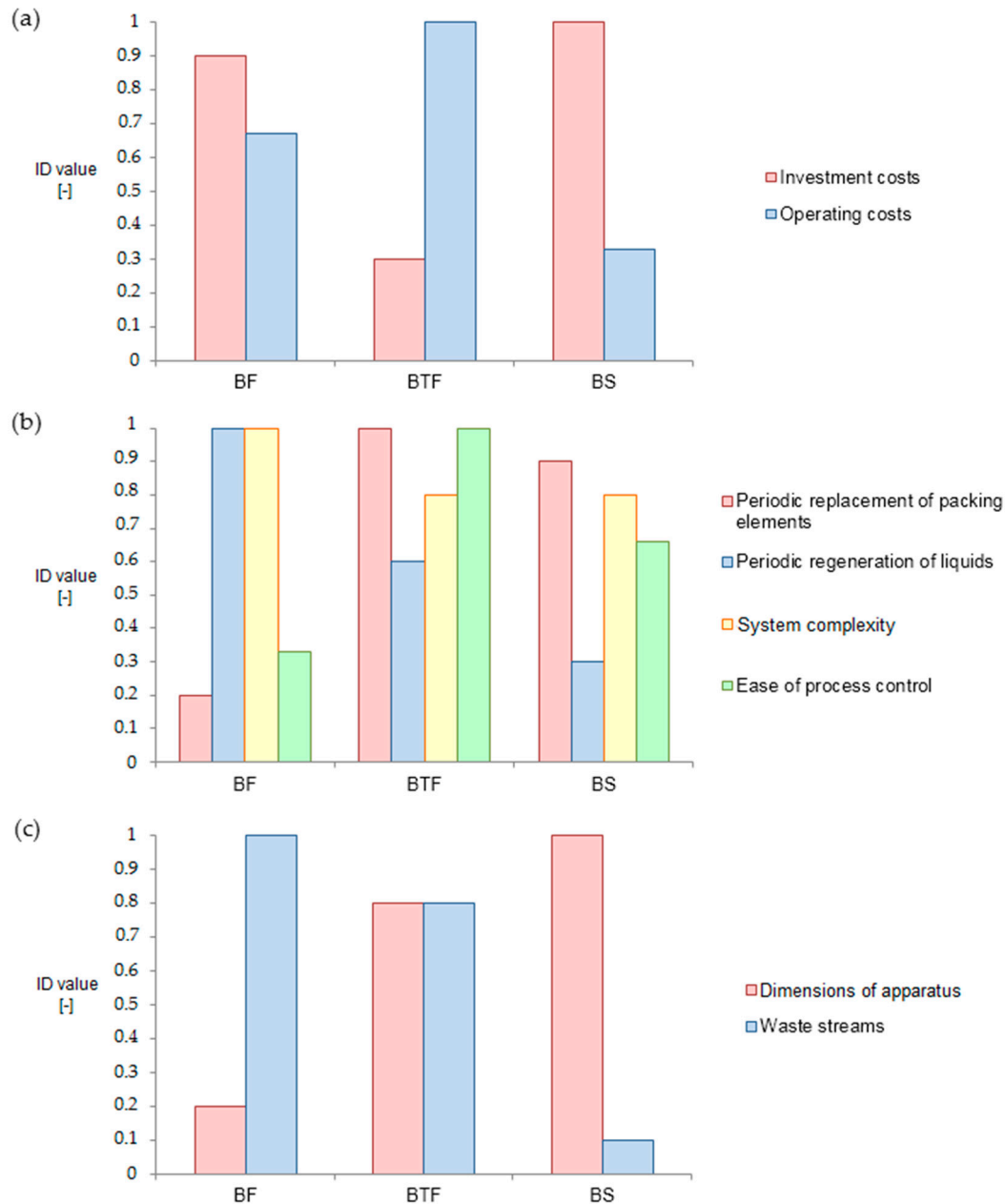


Figure 3. Diagrams presenting indicator values for each sub-criterion: (a) diagram of criterion 2. (b) diagram of criterion 3. (c) diagram of criterion 4.

The results regarding the possibility of a system control indicate that BTFs are characterized with $ID = 1$. This is because this treatment method enables the application of the most complex, effective and quickly responding control system (e.g., control and regulation of trickling liquid frequency, pH, flow rate or composition) [5,31]. The control of process realized in BS is much lower ($ID = 0.67$) and the lowest control possibility is for BF ($ID = 0.33$), indicating high inertness of the process and little regulation possibility in the case of conventional biofilters.



The size of the apparatus (both occupied surface area and capacity) is the largest for BF and this is why the ID calculated for BF is equal to 0.2 (ID = 0.2) (Figure 3c). BS has the smallest area occupied by the apparatus (ID = 1), however the volume of liquid used for the BS process is high. The amount of waste produced is the smallest in the case of BF (ID = 1). Additionally, further reuse of packing material (for another purposes e.g., land fertilizers) as well as the replacement of water by secondary water [12] is possible. The least favorable method in terms of generation of waste streams is BS (ID = 0.1), indicating the need of further processing of generated wastewater for the recovery of absorbed compounds as well as down-stream water purification.

3.4. Results of a Pairwise Comparison

Table 4 was prepared in order to summarize the results collected during a pairwise evaluation procedure. The results contained therein compare the cost-effectiveness of each analyzed method, in general, as well as the given criterion. Table 4 presents the aggregated results for the criterion (C1–C4) and overall result of the alternative (depicted as Summary) to highlight the weaknesses and strengths of the processes studied. The values given in Table 4 were calculated on the basis of Formulae (1) and (2).

The summary results for each investigated treatment method indicate that BTF is the best method for removing hydrophilic compounds. In the case of hydrophobic compounds, BFs are the most convenient, with BTFs presenting very similar efficiency. The removal of inorganic compounds are characterized by the same tendency. In the case of the three methods analyzed, it is least profitable to use BS. These results are supported by the literature data [1,12,19,20,30,73].

Table 4. Scores of the alternatives for each criterion.

Process	Hydrophilic Compounds			Hydrophobic Compounds			Inorganic Compounds		
	BF	BTF	BS	BF	BTF	BS	BF	BTF	BS
C1	0.76	0.93	0.59	0.90	0.88	0.44	0.71	0.67	0.65
C2	0.80	0.58	0.73	0.80	0.58	0.73	0.80	0.58	0.73
C3	0.56	0.88	0.69	0.56	0.88	0.69	0.56	0.88	0.69
C4	0.76	0.80	0.37	0.76	0.80	0.37	0.76	0.80	0.37
Summary	0.74	0.77	0.60	0.78	0.76	0.56	0.72	0.71	0.61

The distribution of the results of individual variants from various criteria is given in Table 4. The results show that BF is a superior technique, among others compared, for the removal of hydrophobic and inorganic compounds from air. BTF proved to be the most beneficial method for removing hydrophilic compounds from air. However, it is of worth to note that very similar results are obtained for BF and BTF for all investigated types of target compounds. The most convenient process in the perspective of exploitation and operating costs is BF, while the least-economic seems to be BTF. BTF and BS presented as the most favorable processes in terms of technical aspects and possibilities of problem elimination. Processes realized in BF and BTF are also the most environmentally-friendly.

Analyzing the obtained results for the removal of hydrophobic compounds, BF is found to be the most efficient. However, BTF attains similar performance as BF in terms of the final summary result.

In terms of efficiency of H₂S and ammonia removal, BF and BTF performance is better than BS. Both methods i.e., BF and BTF are characterized by the lowest negative influence on the environment.

Figure 4 presents the percentage distribution of the results of individual variants from various criteria, based on data from Table 4. The analysis of Figure 4 allows to identify the adequacy of various methods for the removal of odorous compounds.

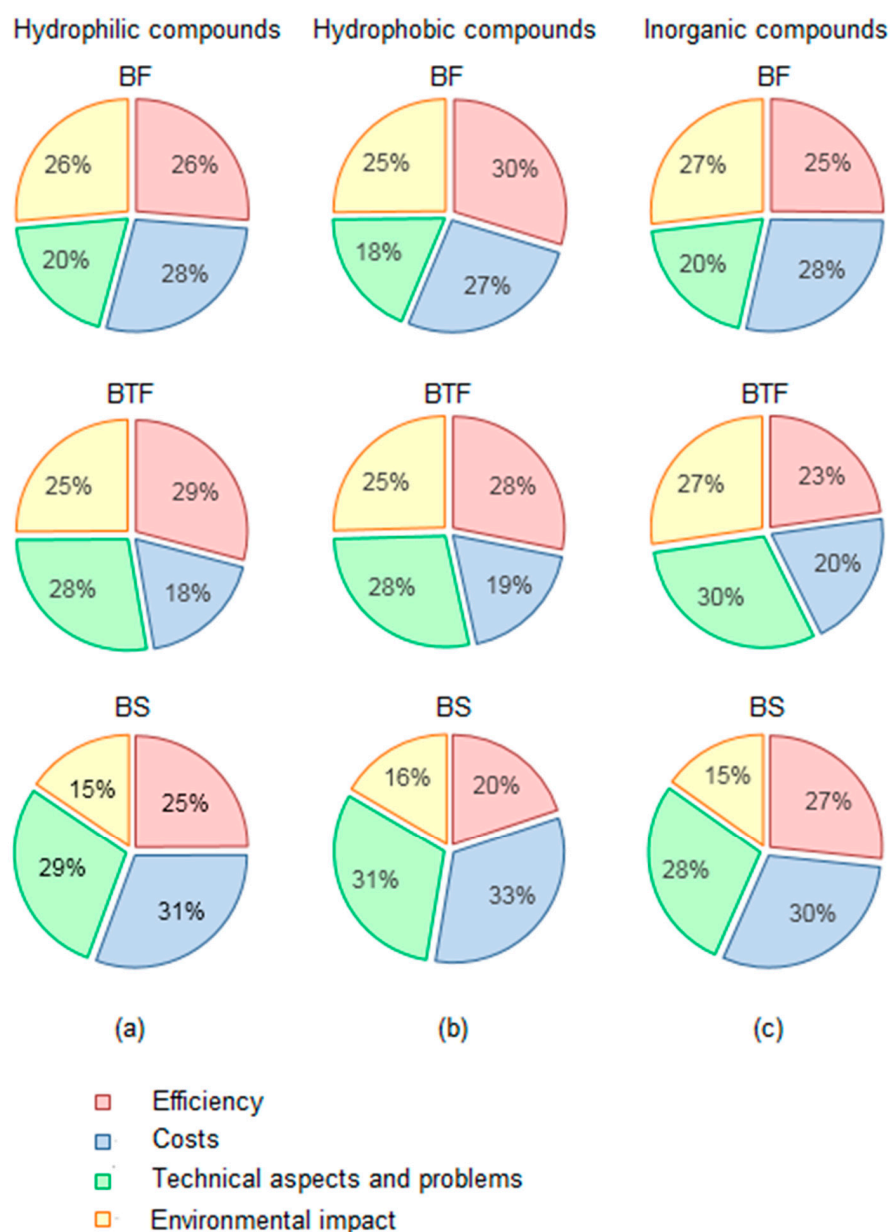


Figure 4. Distribution of alternative results related to different criteria for: (a) hydrophilic compounds, (b) hydrophobic compounds and (c) inorganic compounds.

The results showed that for hydrophilic compounds (Figure 4a), the highest removal efficiency is obtained using BF. BS proved to be the most beneficial due to the costs involved. Similar conclusions for BS concerned technical aspects as well as possible problems faced during the system operation. BF and BTF methods are the most environment-friendly methods. In this respect, BS differs significantly from the other two methods, due to the large production of sewage and the inability of replacing the absorbent by a secondary wastewater.

Analyzing the results for methods of hydrophobic compounds removal from air (Figure 4b), it can be stated that the greatest advantage of the BF method is high removal efficiency and environmental friendliness. The use of the BS method is reasonable when attention is paid to the costs incurred and the technical aspects together with rather low possibility of exploitation problems. In the case of BTF, the results reveal a comparable and even distribution of results for all criteria.

The results obtained for the methods in terms of the removal of inorganic compounds (Figure 4c) revealed that BTF is the optimal method when environmental issues and technical aspects with possible

exploitation problems are considered. On the other hand, BS is outstanding in terms of efficiency and costs of treatment. Due to the fact that BF is characterized by a balanced distribution of results for all analyzed criteria, it may be regarded as a suitable method of purifying air from H₂S and NH₃. Additionally, BF seems to be the best choice when none of the criteria is favored, and only optimal profitability is sought in every respect.

A similar approach of a pairwise comparison was taken up by Oliva et al. [15]. However, the comparison was focused on the advanced oxidation processes, but also included biofiltration methods. In recent years, comparative evaluation of biological methods of air treatment have been proposed by other researchers [12,73,74]. Similar to the results of a pairwise comparison presented in this paper, conventional biofilters seem to be the best choice. Comparing the obtained results with the outcomes of this paper it can be stated that bioscrubbers are the least-favored method of removing compounds of various types from the air.

3.5. Result of Decision Tree for the Preliminary Selection of the Deodorization Method

A decision tree for preliminary selection of an air deodorization method is presented in Figure 5. Table 5 presents the results of investigations used previously for the pairwise comparison procedure as well as additional data collected with the purpose of the tree development. The probability of belonging to a given group of processes is shown at the bottom of Figure 5. The presented decision tree shows, based on the input data used, that only two parameters are important when choosing the proper method i.e., the inlet concentration of a target compound and the hydrophobicity of the compound, represented by Henry's law constant.

Table 5. Data set used for the decision tree development.

Process	Compound	H (mol·m ⁻³ ·Pa ⁻¹)	C _{in} (mg·m ⁻³)	EBRT (s)	References
BF	butanol	1.2	2600	60	[78]
BF	isopropanol	1.3	8000	94.1	[82]
BF	ethanol	9	3700	101	[85]
BF	methanol	2	3.3	38	[91]
BF	hexane	0.0061	10,000	60	[95]
BF	methane	0.000014	4908	257	[96]
BF	ethylene	0.000059	331	2160	[99]
BF	α-pinene	0.00029	450	42	[101]
BF	styrene	0.0027	0.85	1845	[106]
BF	toluene	0.0015	1.9	21.6	[111]
BF	ammonia	0.59	350	17	[115]
BF	hydrogen sulfide	0.001	3750	200	[119]
BF	hexane	0.0061	700	30	[121]
BF	ethanol	9	3700	150	[85]
BF	phenol	0.025	1000	54	[122]
BF	dichloromethane	0.0036	175	60	[123]
BF	methylamine	0.35	136	220	[124]
BF	dimethyl sulfide	0.0056	400	27	[125]
BF	triethylamine	0.066	3000	60	[126]
BF	toluene	0.0015	2800	516	[127]
BF	styrene	0.0027	250	81	[128]
BS	butanol	1.2	1000	48	[80]
BS	isopropanol	1.3	500	60	[84]
BS	methanol	2	100	600	[90]
BS	hexane	0.0061	6200	420	[92]
BS	toluene	0.0015	3300	89	[110]
BS	ammonia	0.59	14	142	[117]
BS	hydrogen sulfide	0.001	140	32	[47]
BS	trichloroethylene	0.0011	300	931	[129]
BS	acetone	0.27	118	195	[130]

Table 5. Cont.

Process	Compound	H (mol·m ⁻³ ·Pa ⁻¹)	C _{in} (mg·m ⁻³)	EBRT (s)	References
BS	1,2-dichloroethane	0.0089	2400	300	[131]
BS	ethyl acetate	0.059	500	84	[132]
BTF	butanol	1.2	1200	124	[79]
BTF	aniline	1.1	300	166	[81]
BTF	isopropanol	1.3	65	160	[83]
BTF	ethanol	9	470	66	[86]
BTF	methanol	2	300	65	[89]
BTF	hexane	0.0061	600	30	[94]
BTF	methane	0.000014	500	240	[97]
BTF	ethylene	0.000059	100	30	[27,100]
BTF	styrene	0.0027	3300	120	[105]
BTF	toluene	0.0015	1128	400	[109]
BTF	ammonia	0.59	100	960	[118]
BTF	hydrogen sulfide	0.001	650	79	[120]
BTF	methyl mercaptan	0.0038	25	50	[28]
BTF	dimethyl sulfide	0.0056	25	123	[120]
BTF	nitrobenzene	0.64	300	24	[133]
BTF	aniline	52	60	42	[81]
BTF	trichloroethylene	0.0011	300	21	[134]
BTF	chlorobenzene	0.0027	1700	60	[135]
BTF	toluene	0.0015	1000	60	[114]
BTF	methyl acrylate	0.049	5000	400	[136]
BTF	methyl acrylate	0.049	5000	200	[136]
BTF	acetone	0.27	8000	137	[137]
BTF	styrene	0.0027	1000	90	[105]
BTF	formaldehyde	3.2	100	80	[138]
BTF	isopropanol	1.3	1000	140	[139]

The decision tree learning algorithm is a non-arbitrary algorithm. A tree was “learned” by splitting the training set into subsets based on an attribute value test. This process was repeated on each derived subset in a recursive manner called recursive partitioning. The recursion is completed when the subset at a node has all the same values of the target variable, or when splitting no longer adds value to the predictions.

In the decision tree model development, only process performance was included, while cost analysis was excluded. Considering such an approach, for high inlet concentrations (higher than 2500 mg·m⁻³), the best treatment option is to use the conventional biofilter. Traditional biofilters based on natural packing materials, for both hydrophilic and hydrophobic compounds, tend to be applied when relatively high inlet concentrations are used [78,82,93,95,96,119]. The use of large EBRT values is necessary for high inlet concentrations, which consequently significantly increases the dimensions of the apparatus. Organic packing materials are cheaper than synthetic ones. The use of a trickling or absorption liquid (BTF and BS) at high concentrations generates the necessity of its frequent replacement (hydrophobic compounds very quickly achieve the saturation state of the liquid), the use of surfactants or increase in the dimensions of the apparatus. The results of applied algorithms show that for the inlet concentrations lower than 2500 mg·m⁻³ and for hydrophilic compounds, biotrickling filtration seems to be the best treatment method. Similar results are obtained for hydrophobic compounds for the inlet concentration range between 475 and 2500 mg·m⁻³. If concentration is lower than 475 mg·m⁻³, the better choice will be the application of a conventional biofilter. Bioscrubbers may be used for compounds characterized by Henry’s law constant between 0.0075 and 0.62 mol·m⁻³·Pa⁻¹ and for inlet concentrations lower than 2500 mg·m⁻³. In this group of compounds, the use of a conventional biofilter should also be considered (due to the low differences in probability of belonging to a given group: 50% and 38% for the bioscrubber and conventional biofilter, respectively).

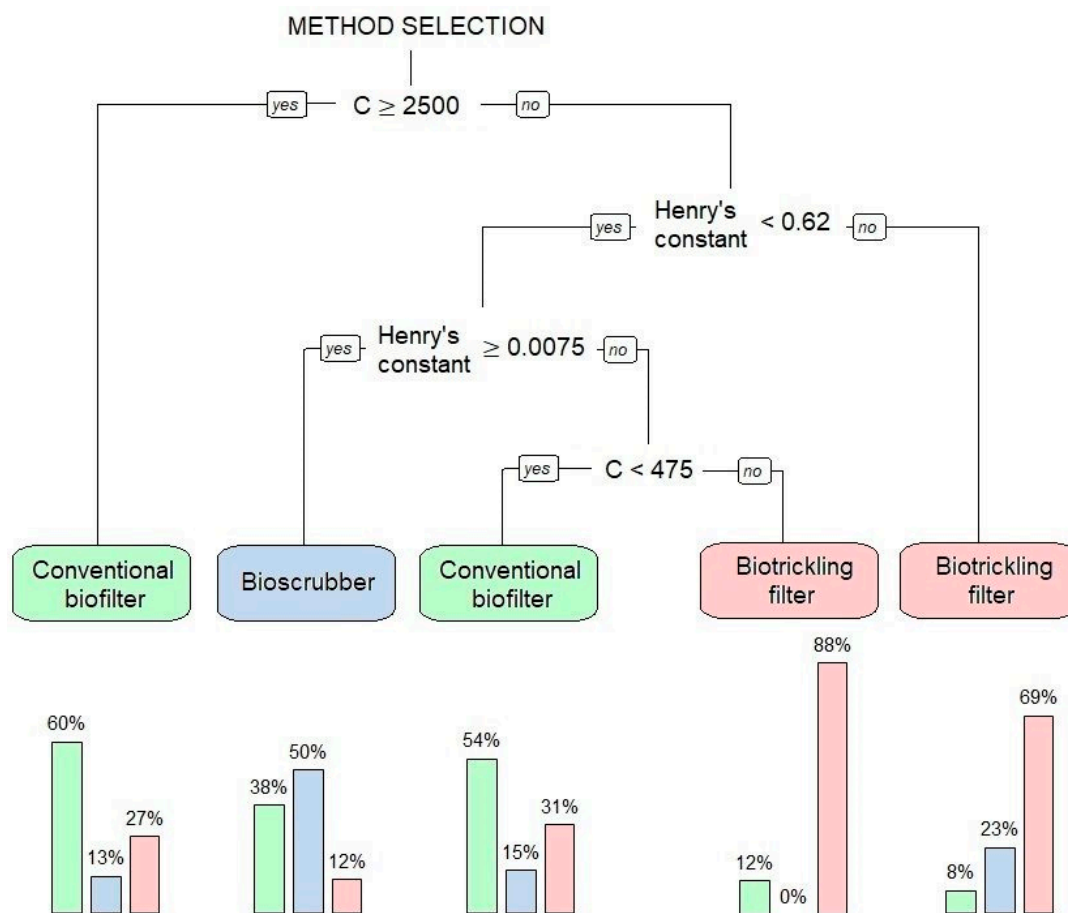


Figure 5. Developed decision tree for the selection of the most suitable biological treatment method (C —inlet concentration, $\text{mg}\cdot\text{m}^{-3}$; Henry's constant, $\text{mol}\cdot\text{m}^{-3}\cdot\text{Pa}^{-1}$).

3.6. Practical Applications and Future Research Perspectives

The presented comparative analysis together with a proposed decision tree model seem to be useful when selecting a treatment procedure for air polluted with odorous compounds. Current development of the legislation regarding the odorous quality of air implies the increased interest in sustainable and efficient deodorization methods, thus the use of biological methods with applications in industrial, agricultural as well as indoor air treatment applications will in particular be increasing in the nearby future. Additionally, currently observed development of these methods, especially when biotrickling filtration is considered, suggests highly probable possibility of eliminating most of the related problems e.g., the effective removal of hydrophobic air pollutants or efficient long-term operation of biological systems. The results of comparative assessment of investigated deodorization methods presented in this paper may, therefore, aid the decision-making process when considering the most efficient biological method of air deodorization.

This paper presents a prototype of a decision model, which after expansion, based on a larger set of input data, will allow for a quick selection of the appropriate method of purification of air polluted with specific compounds. Such an extension of the procedure proposed in this paper is planned by the authors. The future model will take into account the inlet concentrations, geometry as well as the dimensions of apparatus and all important process parameters, including the gas flow rate but also the packed bed material as well as microbial species, especially those selected for the efficient removal of specific compounds from air. In this perspective, the authors believe that a future model will aid and simplify the selection of the treatment method, especially for industrial applications (e.g., pulp and paper, chemical or pharmaceutical) providing that all required input data are available.

4. Conclusions

The results of the comparative evaluation indicate that conventional biofilters and biotrickling filters exhibit similar and good performance of treatment of hydrophobic compounds. Biotrickling filters are superior in terms of the removal of hydrophilic compounds while bioscrubbers present moderate or low performance when compared to BF and BTF. The decision rules obtained from the decision tree method suggest that the most important parameters in the method selection are: inlet concentration and Henry's constant. Based on the literature data presented in this study, the decision tree output suggests using conventional biofilters for the treatment of relatively highly concentrated streams (concentration above $2500 \text{ mg}\cdot\text{m}^{-3}$). For streams with concentrations of odorous compounds lower than depicted, biotrickling filtration is a more suitable method than biofiltration or bioscrubbing. This manuscript reveals the first iteration of the problem related to the selection of the treatment method for hydrophilic and hydrophobic odorous compounds using a proposed decision algorithm. Further expansion of this algorithm is planned in the future and it will be based on more complex input data, including a packing material type or microbial species with the perspective of facilitating the method selection process.

Author Contributions: M.G., P.R., B.S. and J.G. wrote a paper. M.G. revised the literature data, prepared a method of comparison and performed the calculations. P.R. revised the literature for the introduction and discussed the results. B.S. developed a decision tree model. J.G. supervised the preparation of the manuscript, consulted with discussions, and prepared the final conclusions.

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