Review

Bibliometrics and Knowledge Map Analysis of Research Progress on Biological Treatments for Volatile Organic Compounds

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Abstract: The emission of volatile organic compounds (VOCs) has resulted in increasingly severe harm to the environment and human health. In recent years, biological methods have become the preferred technology for VOC removal due to their environmental friendliness and economic advantages. Based on the theory of bibliometrics, this study analyzed research articles and reviews on biological methods for VOC removal published in the Web of Science Core Collection (WOSCC) database from 1966 to 2021. The knowledge map visualization software CiteSpace was utilized to analyze research progress in different countries, co-citation clustering, co-citation bursts, and keyword clustering in the literature data. The results indicated that early research on VOC biological treatment focused on the removal of odorous gases and single components of volatile organic waste gases. Subsequently, benzene contents (BTEX), hydrophobic VOCs, and multi-component VOCs have gradually become the focus of research. In recent years, improving VOC removal efficiency by studying packing materials and microbial communities has become an important research topic both domestically and internationally. Future research should focus on continuously improving the performance of reactors, developing novel reactors, and investigating technologies for treating complex and recalcitrant VOCs.

Keywords: volatile organic compounds; biological methods; bibliometrics; CiteSpace; clustering analysis

1. Introduction

Volatile organic compounds (VOCs) are a class of gaseous pollutants widely present in the atmosphere. According to the definition of the World Health Organization, VOCs are various organic compounds with boiling points ranging from 50 °C to 260 °C at standard atmospheric pressure. VOCs emanate from a diverse range of sources. Outdoor sources of pollution include effluent gases from chemical, paper, pharmaceutical, electronic, and other industries [1], as well as automobile exhaust produced by transportation. Indoor emissions primarily originate from activities such as coal burning, natural gas combustion, and building decoration. In the atmosphere, VOCs are not only precursors for the formation of secondary pollutants, such as PM2.5 and O3, but also react with other substances in the air to cause air pollution problems, such as haze and photochemical smog [2]. In addition, the majority of VOCs are irritant, toxic, teratogenic, and carcinogenic, not only causing sensory discomfort but also inflicting severe harm. For instance, formaldehyde, a...
pollutant emitted by indoor decorations, can trigger headaches and nausea when present at a concentration above a certain threshold. Moreover, it can lead to coma and harm the human respiratory and nervous systems [3]. Benzene series are very harmful to human blood, viscera, and nerves and can cause teratogenic, carcinogenic, and mutagenic effects in severe cases [4]. In 1979, the United Nations Economic Commission for Europe convened a transnational conference on air pollution in Geneva, which centered on the management of VOCs. Currently, with the aggravation of air pollution worldwide, nations are increasingly recognizing the impact of VOCs. Laws and regulations regarding emissions are being regulated, and treatment technologies are being gradually improved.

VOC treatment technologies are mainly divided into two categories: recovery and removal methods. The recovery methods involve physical techniques, such as adsorption [5] and condensation [6], to adsorb or separate VOCs. However, these methods only transfer pollutants from the gas phase to the liquid or solid phase [7]. Since VOCs cannot be completely eliminated, further treatment is required to mitigate their impact. The removal methods for VOCs comprise combustion [8], photocatalysis [9], plasma technology [10], and biological methods [11]. These methods primarily involve chemical or biochemical reactions that utilize light, heat, microorganisms, and catalysts to degrade pollutants into harmless inorganic substances, such as CO\(_2\) and H\(_2\)O. In recent years, the biological method has undergone widespread development and application due to its low economic cost and environmentally friendly advantages. The primary processes include biofiltration, biotrickling filtration, and bioscrubbing [12]. Among these processes, biofiltration is the most extensively employed and is suitable for treating low flow rates of waste gas [13]. Biotrickling filtration is effective in treating high loads of VOCs and malodorous gases [14]. Bioscrubbing is appropriate for the removal of organic waste gas with good water solubility [15]. Given the rapid development of testing technology and biotechnology, the objects and process technologies of biological methods are also constantly changing. However, the current research still has some limitations. The removal efficiency of complex and difficult-to-degrade acid gas is relatively low; studies on the mechanism of biofilm formation and microbial community evolution in the reactor are not systematic and perfect. Hence, it is necessary to conduct a comprehensive review of related research to provide a reference for the improvement and development of biotechnology.

The traditional literature review involves collecting relevant papers in the field and summarizing, organizing, and reviewing the information. Bibliometrics is the science of quantitatively analyzing knowledge carriers using mathematical and statistical methods, with the primary objects of analysis being the volume of literature, author information, and vocabulary count. This article employs the theoretical framework of bibliometrics and utilizes the knowledge mapping visualization software CiteSpace [16], developed by Professor Chaomei Chen, to quantitatively analyze and model papers published on a global scale and over an extended period in the research field of VOC biological removal. The article outlines the knowledge structure, developmental lineage, and research trends in this field. By mining key studies in the literature and newly published articles, the research focus and development direction in this field are further summarized to provide ideas and references for future research.

2. Data Sources and Analysis Methods
2.1. Data Sources

The literature data used in this paper are from the Web of Science core collection (WOSCC) database, encompassing authoritative and high-impact academic journals across various subject areas. Abstract and keyword information was not included in the WOSCC data for articles published in 1990 and before. As a result, the search strategy was divided into two parts. In the first part, the search term “TS = (biofilter* OR biofiltration*) AND TS = (VOCs* OR volatile organic compounds*)” was used, and the publication types “Article” and “Review” were selected. The literature retrieval period was set from 1 January 1990 to 31 July 2021, and this resulted in obtaining a total of 824 publications. The first part
of the literature was used as the primary data for analyzing the progress of related research using the visualization software CiteSpace. The search strategy in the second part was altered to “TI = (biofilter* OR soil bed)”, with the publication types “Article” and “Review” selected. The retrieval period time was set from 1900 to 1989. A total of 114 search results were obtained, and after manually screening papers related to the topic, 13 papers were selected to understand the developmental history of global biofiltration. As the data in the second part lacked citation information, they were not imported into CiteSpace, and only the relevant contents were directly reviewed.

2.2. Analysis Method

Initially, the number of published papers in the country was manually imported into Microsoft Excel 2019 to calculate the number of published papers all over the world. The WOSCC retrieved documents were downloaded in the format of “full record and cited references” and saved as a plain text (txt) file. The data were imported into the online platform https://bibliometric.com/ (accessed on 9 August 2022) and CiteSpace (5.7.R5). By setting different node types, we were able to visualize the data information, such as the publication volume trends, the cooperation networks of countries and institutions, co-citation clustering, and keyword time zones. We obtained a knowledge map to analyze the distribution characteristics and knowledge structure of papers in the research field, the key research contents and evolution process, the state of research and important studies, as well as the research hotspots and future development trends. The data analysis process is presented in Figure 1.

![Flow chart for the analysis of VOC removal by biological treatment.](image-url)

Note: * refers to any character group, including null characters.

**Figure 1.** Flow chart for the analysis of VOC removal by biological treatment.
3. General Overview

3.1. Number of Published Studies in the Literature

Figure 2 illustrates the annual number of studies published worldwide in the literature retrieved from WOSCC. The results indicate that the number of studies increased slowly until 1997. The initial publication in 1966 by Carlson and Leiser [17] used biofiltration to remove odor and emphasized the crucial role of microorganisms in the soil in odor removal. This was followed by the promotion and application of biological treatment technologies in European countries through the 1980s. From 1997 onwards, there was a substantial increase in the number of published papers, which exhibited an upward trend. Among the published papers, the most-cited article is the review published by Faisal and Alok in 2000, titled “Removal of Volatile Organic Compounds from polluted air” [18]. They presented and evaluated each technology for VOC control in detail and concluded that biofiltration would be the most popular option in the future.

![Figure 2. Annual number of publications (as of 31 July 2021).](image-url)

3.2. Country Cooperation Network

The collaboration network of countries was established based on country collaborations in the cited literature, and collaboration was considered to exist when authors from two different countries contributed to the same article. Based on the publications related to the research topic that appeared between 1900 and 2021, the countries with more than 10 publications and the network of collaborations between them are shown in Figure 3. The circle color scale radiating outward from the center corresponds to the year of publication. The color ranges from dark purple to yellow-green, indicating early to recent years, respectively. In addition, the color of the circle’s center represents the earliest year of publications, while the thickness of the color scale corresponds to the number of publications in the respective years. The United States’ and Canada’s circle centers are dark purple, indicating that relevant research was conducted at the earliest time. The average annual number of publications in the United States is evenly distributed, suggesting that there are continuous research outcomes published in this field. On the other hand, China’s and Spain’s circle centers are dark green, signifying that research in these countries started later. The color levels for China are predominantly from light green to yellow, implying that the main research results have been published in recent years. The size of the circles corresponds to
the total number of publications. As shown in the figure, the number of publications in the United States is significantly higher than that in other countries, indicating that the United States holds a leading position in the field of biological VOC removal research. Additionally, several countries in the figure have nodes encompassed by rosy-red circles outside, which denote points with high intermediary centrality in the network. Intermediary centrality is a commonly used indicator for centrality measurement, representing a pivotal node with strong connections to other nodes. Countries such as the United States, Spain, and Canada have more interactions and collaborations with scholars from other countries in this field.

Figure 3. Country Cooperation Network.

3.3. Research Institutions

Table 1 shows the primary institutions and analytical indicators concerning global research on biological VOC removal from 1990 to 2021. The University of Cincinnati held the top spot in the United States based on the combined count of publications and citations, while Hunan University ranked first in terms of the average number of citations. The institutions in the table are the main forces in the research field and have a strong influence on biological treatment research.

Table 1. Major research institutions.

<table>
<thead>
<tr>
<th>Rank</th>
<th>Institution</th>
<th>Number of Published Articles</th>
<th>Total Number of Citations</th>
<th>Total Citation Rank</th>
<th>Average Number of Citations</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Univ Cincinnati&lt;br&gt;University of Cincinnati (USA)</td>
<td>41</td>
<td>475</td>
<td>1</td>
<td>11.59</td>
</tr>
<tr>
<td>2</td>
<td>Tsinghua Univ&lt;br&gt;Tsinghua University (China)</td>
<td>36</td>
<td>205</td>
<td>6</td>
<td>5.69</td>
</tr>
<tr>
<td>3</td>
<td>Chinese Acad Sci&lt;br&gt;Chinese Academy of Sciences (China)&lt;br&gt;Univ Ghent</td>
<td>34</td>
<td>152</td>
<td>11</td>
<td>4.47</td>
</tr>
<tr>
<td>4</td>
<td>University of Ghent (Belgium)</td>
<td>28</td>
<td>129</td>
<td>15</td>
<td>4.61</td>
</tr>
</tbody>
</table>
### Table 1. Cont.

<table>
<thead>
<tr>
<th>Rank</th>
<th>Institution</th>
<th>Number of Published Articles</th>
<th>Total Number of Citations</th>
<th>Total Citation Rank</th>
<th>Average Number of Citations</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>Univ Valladolid University of Valladolid (Spain)</td>
<td>27</td>
<td>237</td>
<td>5</td>
<td>8.78</td>
</tr>
<tr>
<td>6</td>
<td>Hunan Univ</td>
<td>25</td>
<td>443</td>
<td>2</td>
<td>17.72</td>
</tr>
<tr>
<td>7</td>
<td>Univ Sherbrooke Shebuc University (Canada)</td>
<td>25</td>
<td>308</td>
<td>4</td>
<td>12.32</td>
</tr>
<tr>
<td>8</td>
<td>Univ A Coruna University of A Coruña (Spain)</td>
<td>24</td>
<td>376</td>
<td>3</td>
<td>15.67</td>
</tr>
<tr>
<td>9</td>
<td>Univ Technol Sydney University of Technology Sydney</td>
<td>19</td>
<td>117</td>
<td>18</td>
<td>6.16</td>
</tr>
<tr>
<td>10</td>
<td>Tianjin Univ</td>
<td>18</td>
<td>52</td>
<td>40</td>
<td>2.89</td>
</tr>
</tbody>
</table>

### 3.4. Highly Published Journals

The top 10 journals based on the number of publications are shown in Table 2, with the *Journal of Chemical Technology and Biotechnology* (JCTB) and *Chemosphere* (CHEMOSPHERE) being tied for first place with 43 publications. The total number of citations reflects the academic influence of the journals, as illustrated in the table. The *Journal of the Air Waste Management Association* (JAWMA), the *Journal of Chemical Technology and Biotechnology* (JCTB), and the *Journal of Hazardous Materials* (JHM) are the three most influential journals in this research area. This is important information for new researchers regarding submissions.

### Table 2. Top 10 journals in terms of published articles.

<table>
<thead>
<tr>
<th>Rank</th>
<th>Journal</th>
<th>Number of Published Articles</th>
<th>Total Number of Citations</th>
<th>Total Citation Rank</th>
<th>Average Number of Citations</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td><em>Journal of Chemical Technology and Biotechnology</em> (JCTB)</td>
<td>43</td>
<td>368</td>
<td>2</td>
<td>8.56</td>
</tr>
<tr>
<td>2</td>
<td><em>Chemosphere</em> (CHEMOSPHERE)</td>
<td>43</td>
<td>189</td>
<td>7</td>
<td>4.40</td>
</tr>
<tr>
<td>3</td>
<td><em>Journal of the Air Waste Management Association</em> (JAWMA)</td>
<td>40</td>
<td>401</td>
<td>1</td>
<td>10.03</td>
</tr>
<tr>
<td>4</td>
<td><em>Journal of Hazardous Materials</em> (JHM)</td>
<td>31</td>
<td>343</td>
<td>3</td>
<td>11.06</td>
</tr>
<tr>
<td>5</td>
<td><em>Chemical Engineering Journal</em> (CEJ)</td>
<td>30</td>
<td>296</td>
<td>4</td>
<td>9.87</td>
</tr>
<tr>
<td>6</td>
<td><em>Bioresource Technology</em> (BT)</td>
<td>30</td>
<td>212</td>
<td>5</td>
<td>7.07</td>
</tr>
<tr>
<td>7</td>
<td><em>Environmental Technology</em> (ET)</td>
<td>27</td>
<td>104</td>
<td>18</td>
<td>3.85</td>
</tr>
<tr>
<td>8</td>
<td><em>Water Science and Technology</em> (WST)</td>
<td>25</td>
<td>128</td>
<td>16</td>
<td>5.12</td>
</tr>
<tr>
<td>9</td>
<td><em>Biochemical Engineering Journal</em> (BEJ)</td>
<td>19</td>
<td>133</td>
<td>15</td>
<td>7.00</td>
</tr>
<tr>
<td>10</td>
<td><em>Biotechnology and Bioengineering</em> (BB)</td>
<td>18</td>
<td>198</td>
<td>6</td>
<td>11.00</td>
</tr>
</tbody>
</table>

### 4. Research Themes and Key Studies

#### 4.1. Co-Citation Clustering

Co-citation clustering refers to cases where two or more authors are cited by one or more subsequent papers simultaneously, leading to a co-citation relationship between them. The 30 publications with the highest number of citations every two years were selected in CiteSpace to construct a citation network from 1990 to 2021. Multiple networks were synthesized to obtain a citation network with 263 nodes and 412 connected lines. The clustering function generates knowledge clusters based on the log-likelihood algorithm and assigns labels for the clusters based on the title subject terms of the cited studies in each cluster, as shown in the Figure 4. Clustering aids researchers in identifying the subdomains of a research topic. The modularity and the weighted mean silhouette are the metrics used...
to assess whether the clustering is optimal. In Figure 4, these are 0.84 and 0.94, signifying that the subdomains of clustering and the classification results are evident [19].

Table 3 shows the details of the eight main clusters, including the cluster silhouette, the average year, and the topic identifiers. The cluster silhouette serves as an assessment metric for the clustering function, and the closer it is to 1, the better the clustering performance. The average year represents the period when more research outcomes were published. Cluster identifier words are generated according to the publication titles with the highest number of citations in each cluster. Through the color clustering in the co-citation map combined with the literature reading before 1990, we can understand the development of the research field of biological removal of VOCs.

The first stage, before 1990, was characterized by the nascent phase of biological treatment technology. Even though the concept of biological treatment of waste gas was first proposed by Bach [20] as early as the 1820s, this technology did not receive much attention, and its development remained slow. It was not until the 1970s that the biological method was adopted in industries to address issues related to odor and organic and
inorganic pollutants. In the 1980s, Germany applied the biological method in chemical, printing, coating, and other industries, attaining favorable outcomes [21].

In the second stage, the identifying keywords included “waste gas biological treatment technology” (cluster 6) and “trickle-bed air biofilter” (cluster 2). In this stage, the application of biological methods expanded to include more advanced research areas. The implementation of the Clean Air Act Amendment in 1990 prompted the United States and other developed countries to deepen their research on VOCs, which comprise the vast majority of air pollutants and are categorized into various groups based on their chemical structure, such as alkanes, aromatics, alkenes, halocarbons, esters, aldehydes, ketones, and others. Aromatic compounds, which are complex and difficult to deal with, are commonly found in VOC emissions [22]. Most studies on removing VOCs using biological methods focus on benzene series compounds, particularly toluene. Other commonly studied compounds include chlorobenzene [23], ethyl acetate [24], and methyl tert-butyl ether [25], as well as various mixed organic waste gases. According to a survey, biotechnology accounted for 78% of the German deodorant market in 1994 [26], highlighting the efficacy of biotreatment technologies for waste gas treatment and air pollution control. In their review article “Waste gas biotreatment technology,” Kennes et al. [27] introduced the three most widely used processes in waste gas biotreatment technology and provided application examples with different compounds. Meanwhile, Lu et al. [28] employed a trickle-bed biofilter to effectively remove ethyl acetate under low- to high-load conditions.

The third stage is characterized by the identifying keywords “air stream” (cluster 1), “air pollution control” (cluster 5), and “toluene mixture” (cluster 3). The period for this cluster is 2001–2009, and the research during this stage mainly focuses on the development of bioreactors, including innovative bioreactors in practical applications and laboratory development stages. In his review article “Bioprocesses for air pollution control,” Kennes [29] provides a detailed analysis of the advantages and disadvantages of various reactor installations.

Stage four is characterized by the identifying keywords “biotrickling filter” (cluster 0), “malodorous air” (cluster 4), and “phytoremediation efficiency” (cluster 7). Compared with previous biological methods, this stage emphasizes the importance of low cost and environmental friendliness in effectively removing VOCs. The reactors commonly used during this stage are biotrickling filters, with internal fillers made of inorganic materials, such as ceramics or plastics. A biotrickling filtration system supplemented with a nutrient solution is added to better regulate the growth of microorganisms, which has a good effect on the treatment of high-load organic waste gas, hydrogen sulfide, and malodorous gas [30]. For instance, Yang et al. [31] explored the performance and degradation efficiency of biotrickling filters for treating chemical fiber VOCs and demonstrated that biotrickling filtration is a lower-cost and effective VOC control scheme compared with adsorption technology and regenerative catalytic oxidation technology. Zhang et al. [32] studied the treatment of ethyl acetate organic matter discharged from the pharmaceutical industry by biological drip filters and found that the average removal rate could reach 95% when the residence time of the empty bed was 17 s. In the review article “Technologies for Deodorization of Malodorous Gases”, Wysocka et al. [33] compared various gas deodorization technologies and concluded that biological gas is a relatively low-cost method, but it should be considered when removing gases with high concentrations and complex components. Phytoremediation efficiency is usually used to deal with indoor formaldehyde and other harmful VOCs produced during the decoration process to create a better living environment for people. Irga et al. [34] evaluated the effectiveness of various common plant species in removing two major groups of VOCs. This study provides a baseline indication of the removal efficiency of plant species for modeling hydrophobic and hydrophilic VOCs. The findings show that it is feasible to select plants for target pollutant-dependent plant biofilters.
4.2. Key Studies

The burst detection feature provided by CiteSpace detects publications with a rapid increase in citations over a short period. These are referred to as high-burst papers, reflecting the research topic bias during that particular period. The 10 papers with the highest burst intensity calculated by CiteSpace and their related information are listed in Table 4, all of which relate to the subject of biological VOC treatment. The paper with the highest emergent intensity is “Challenges and solutions for biofiltration of hydrophobic volatile organic compounds” published by Cheng et al. in 2016 [35]. This review paper examined the factors influencing the biological removal of hydrophobic VOCs in five aspects: surfactant incorporation, application of fungal biocatalysts, pretreatment biofiltration, novel bioreactors, and utilization of hydrophilic compounds.

Table 4. High-burst publications.

<table>
<thead>
<tr>
<th>Publication Title</th>
<th>Type of Publication</th>
<th>Specific Information</th>
<th>Total Citations</th>
<th>Strength</th>
<th>Burst Year</th>
</tr>
</thead>
</table>

Papers 3, 5, 6, and 8 in the list are all reviews. Mudliar et al. [37] offered an overview of various bioreactors for the degradation of VOCs and malodorous gases. They provided detailed descriptions of their configurations and designs, operating mechanisms, and microbial degradation processes and of future research and development needs in this field. Barbusinski et al. [39] compared the variations in equipment design and the application scope of different odor treatment technologies by providing an overview of the available technologies, process principles, and characteristics. Yang et al. [40] conducted a review of the interactions between multiple target pollutants in the degradation of VOCs and proposed ways to mitigate the antagonistic effects by improving bioavailability and
biodegradability. Kennes et al. [29] introduced the biodegradation process, as well as the types and relative merits of innovative bioreactors that are in application or still in the experimental stage.

Papers 4, 7, 9, and 10 in the list are studies on the influencing factors and optimization methods in the removal of VOCs by biological methods. Mohseni et al. [38] discovered that the removal efficiency of two pollutants in mixed VOCs influenced each other, and the lipophilic properties of biofilms contributed to the high removal rate in the removal of VOCs. Estrada et al. [41] compared the efficiency of fungal and bacterial biodegradation of VOC mixtures (propionaldehyde, methyl isobutyl ketone, toluene, and hexanol) under the same operating conditions. In general, the removal capacity of fungal biofilters was lower than that of bacterial biofilters, and the implementation of coupled fungal/bacterial systems did not significantly enhance the performance of VOC removal. Rene et al. [42] examined the effectiveness of biological methods for removing a mixture of benzene pollutants and compared the conditions required for achieving optimal removal under various influencing factors. Shareefdeen et al. [43] conducted a parameter sensitivity study using a model and validated the feasibility of the model through experiments on the removal of toluene vapor by gas-phase biofilters.

In addition, the second publication in the list was compiled by Devinny et al. [36] in 1999 and comprises chapters on biofiltration mechanisms, biofiltration media, factors affecting and controlling biofiltration, microbial ecology in biofiltration, and biofiltration models, designs, and costs.

5. Research Hotspots and Trend Analysis

5.1. Analysis of Research Hotspots

A statistical analysis of the co-occurrence of keywords from 1990 to 2021 was performed. The basic information of the 30 most frequently used keywords is listed in Table 5 and can be roughly divided into the following categories:

<table>
<thead>
<tr>
<th>Rank</th>
<th>Keywords</th>
<th>Category</th>
<th>Frequency</th>
<th>Rank</th>
<th>Keywords</th>
<th>Category</th>
<th>Frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Biofiltration</td>
<td>Removal efficiency</td>
<td>510</td>
<td>16</td>
<td>Bioreactor</td>
<td>Reactor type</td>
<td>43</td>
</tr>
<tr>
<td>2</td>
<td>Biofilter</td>
<td>Reactor type</td>
<td>346</td>
<td>17</td>
<td>Odor</td>
<td>Pollutant</td>
<td>40</td>
</tr>
<tr>
<td>3</td>
<td>Volatile organic compound</td>
<td>Pollutant</td>
<td>283</td>
<td>18</td>
<td>Vapor</td>
<td>Removal efficiency</td>
<td>33</td>
</tr>
<tr>
<td>4</td>
<td>Removal</td>
<td>Removal efficiency</td>
<td>266</td>
<td>19</td>
<td>Waste gas</td>
<td>Environmental media</td>
<td>31</td>
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<tr>
<td>5</td>
<td>Toluene</td>
<td>Pollutant</td>
<td>232</td>
<td>20</td>
<td>Packing material</td>
<td>Principle</td>
<td>25</td>
</tr>
<tr>
<td>6</td>
<td>Biodegradation</td>
<td>Principle</td>
<td>206</td>
<td>21</td>
<td>Microbial community</td>
<td>Principle</td>
<td>24</td>
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<td>7</td>
<td>VOC</td>
<td>Pollutant</td>
<td>206</td>
<td>22</td>
<td>Gas</td>
<td>Environmental media</td>
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<td>8</td>
<td>Performance</td>
<td>Removal efficiency</td>
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<td>Model</td>
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<td>9</td>
<td>Air</td>
<td>Environmental media</td>
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<td>24</td>
<td>Activated carbon</td>
<td>Principle</td>
<td>21</td>
</tr>
<tr>
<td>10</td>
<td>Biotrickling filter</td>
<td>Reactor type</td>
<td>132</td>
<td>25</td>
<td>BTEX</td>
<td>Pollutant</td>
<td>20</td>
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<td>11</td>
<td>Degradation</td>
<td>Principle</td>
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<td>Bacteria</td>
<td>Principle</td>
<td>19</td>
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<td>12</td>
<td>Emission</td>
<td>Environmental media</td>
<td>67</td>
<td>27</td>
<td>Abatement</td>
<td>Removal efficiency</td>
<td>19</td>
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<tr>
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<td>Hydrogen sulfide</td>
<td>Pollutant</td>
<td>60</td>
<td>28</td>
<td>Adsorption</td>
<td>Principle</td>
<td>18</td>
</tr>
<tr>
<td>14</td>
<td>Mixture</td>
<td>Pollutant</td>
<td>56</td>
<td>29</td>
<td>Biofilm</td>
<td>Principle</td>
<td>18</td>
</tr>
<tr>
<td>15</td>
<td>Benzene</td>
<td>Pollutant</td>
<td>54</td>
<td>30</td>
<td>Waste gas</td>
<td>Environmental media</td>
<td>17</td>
</tr>
</tbody>
</table>

The classification of environmental media can be identified with keywords, such as “air”, “emission”, “waste gas”, and so forth. VOC pollutants are significant components of air pollution. Their sources can be categorized into natural and anthropogenic. Natural sources include activities such as volcanic eruptions, forest fires, and decay of animal and plant remains. Anthropogenic sources can be further subcategorized into mobile and stationary sources, with mobile sources mainly referring to vehicle exhaust emissions and stationary sources referring to the release of exhaust gases during industrial, agricultural,
and other production processes. VOCs participate in the formation of ozone and secondary aerosols in the atmosphere and act as important precursors to haze and photochemical smog pollution [44]. Due to their strong volatility and human activities, the majority of VOCs accumulate, migrate, and diffuse into the atmosphere, leading to harmful impacts on the environment and human health.

The high-frequency keywords related to reactor types are “biofilter”, “biotrickling filter”, and “bioreactor”. A biofilter typically consists of packing material that offers attachment sites for the growth and reproduction of microorganisms. The exhaust gas flows through the packing bed, comes into contact with the biofilm, and is degraded by microorganisms [45]. The biotrickling filter has a similar structure to the biofilter and is humidified by a circulating water system with a spray at the top. The exhaust gas enters the system from the bottom of the unit and comes into contact with the microorganisms. In a two-phase bioreactor, the gas phase is separated from the aqueous phase in the device by a membrane. The gas phase is contaminated air, which usually contains a carbon source, while the aqueous phase serves as the nutrient source for the biofilm that grows on the water side of the membrane surface [29].

The keywords of the pollutant categories are “toluene”, “benzene”, “BTEX”, “hydrogen sulfide”, etcetera, with toluene being the most frequently occurring pollutant. Toluene is a colorless, volatile liquid with a distinct odor that is widely used in industrial production, decoration products, chemical synthesis, and pesticides [46]. With toluene as a research object, new technologies are being developed. Hybrid biological treatment technologies, such as biofiltration and adsorption, can be effective in treating VOCs [47]. The biofiltration system of Streptomyces griseus immobilized on activated carbon is feasible for removing toluene and increasing the reuse rate, providing a new solution for industrial applications [48]. The microbial activity of the planted biofilter is significantly higher than that of the nonplanted biofilter, and the growth of ryegrass in the biofilter enhances the biodegradation of toluene [49]. The high-frequency keyword “mixture” refers to mixed waste gas, where various pollutants are often mixed in the discharge. Traditional biofilters are inefficient at treating mixed waste gas, so many scholars are investigating ways to enhance the performance of bioreactors for removing mixed waste gas. Some of the findings include: a single-stage biotrickling filter that can effectively treat both hydrogen sulfide and toluene [50]; and that the mixing of hydrophilic and hydrophobic VOCs can improve the mutual degradation rate [51]. Furthermore, the two-stage bioreactor is highly effective in treating mixed waste gases [52].

The keywords to explore the principle of biofiltration are “biofilm”, “adsorption”, and “biodegradation”, to name a few. The basic principle of the biological method includes the following steps: firstly, the organic pollutants in the gas phase are transferred to the liquid phase; these organic compounds react with compounds in the liquid phase or adsorb on the packing materials and the biofilm on the surface of the packing materials through the liquid film; finally, pollutants are decomposed, transformed, or synthesized into part of the cytoplasm through biodegradation, producing intermediate metabolites, water, and carbon dioxide, as well as inorganic small molecules.

The keywords related to exploring the efficiency of the bioreactor are “removal”, “performance”, etcetera. The performance parameters of bioreactor operation include removal efficiency, surface loading, and empty bed residence time. The empty bed residence time refers to the time required for the gas to pass through the bioreactor, while effective residence time refers to the time required for the gas to pass through the filled packing layer. The empty bed residence time is usually considered in the design of bioreactors, and this parameter is related to the removal efficiency and volume of the bioreactor. The surface load is the gas flow rate per unit area of the reactor, and the longer the empty bed residence time, the lower the surface load. The removal efficiency is the most intuitive parameter for evaluating the performance of a bioreactor, and the pollutant removal efficiency of a successful bioreactor is generally 95–99% [53].
5.2. Analysis of Research Trends

The time-zone diagram in Figure 5 also allows for the analysis of research trends related to a particular subject keyword in the development of biological methods for treating VOCs, primarily in terms of bioreactor types, pollutant categories, and research methods.

The type of treatment device shifted from biofilters to biotrickling filters from 1996 to 2002. Traditional biofilters are widely used for odor and VOC removal, having the advantages of simple operation and low operating costs. However, they have limitations in treating large flow rates and hard-to-degrade VOCs. The waste gas must meet the following requirements: hydrophilicity, easy biodegradability, and low concentration of particulate matter [54]. To improve the transfer efficiency of pollutants from the gas phase to the liquid phase, the biotrickling filter was developed. It solves the problems of degradation of packing materials and easy clogging when the pollutant load is high [55]. It can also better regulate the growth of microorganisms.

Around the year 2000, biological methods were primarily applied for the removal of single-component pollutants, such as hydrogen sulfide and toluene. However, with the increase in VOC pollution, the removal of single VOCs is insufficient for pollution control. As a result, benzene pollutants and mixed waste gases gradually became the mainstream targets of biological methods for studying treatments and their functionality. The keyword “BTEX” includes VOCs composed of benzene, toluene, ethylbenzene, xylene, etcetera [56]. BTEX has been a major research hotspot in the field of biological methods in recent years, and related studies are maturing. In industrial waste gas, BTEX is often emitted as a multicomponent mixture; thus, there are synergistic or antagonistic effects among the components in the biodegradation process [57,58]. Susant et al. [59] discovered that the presence of xylene promoted the removal of toluene from the mixture. Raboni et al. [60] achieved an average removal rate of over 96% when treating BTEX with bacteria and fungi in combination. To simultaneously improve the removal of BTEX, Mathur et al. [61] attempted to remove BTEX using a mixture of compost, bagasse, and granular activated carbon filters, and the removal of BTEX was almost 100% under suitable operating conditions. Klapková et al. [62] found that, in the removal of BTEX, bacterial strains...
inoculated with toluene and xylene performed better than the indigenous microorganisms of the packing materials themselves. However, due to the very similar chemical structure and metabolism of BTEX, they showed more mutual inhibition in the process of biodegradation [63]. Inês et al. [64] experimentally demonstrated that the mixing of multiple waste gases in the reactor reduced the biodegradation rate of each pollutant and the overall biodegradation rate. Plessis et al. [65] found that toluene inhibited the removal of benzene and ethylbenzene in a biofilter inoculated with toluene-dominated bacteria.

The keyword at the research method level is “modeling”, which refers to the use of computer and mathematical methods to model the waste gas treatment process and verify the feasibility of the model using experimental results. There have been numerous studies on this subject since the beginning of biological methods. Douglas et al. [66] described the transfer between gas and water phases, and the biodegradation process of substrates. Basil et al. [67] considered the mutual kinetics between pollutants and the effect of oxygen on biodegradation. Based on this, a model was developed for the treatment of mixed gas streams of VOCs in biofilters. Kyungnan et al. [68] utilized mathematical models to determine the results of vinyl acetate removal at different organic loading rates, solid residence times, and dissolved oxygen concentrations. Currently, the method of predicting and evaluating bioreactor operating parameters and effectiveness by constructing models needs further exploration and investigation.

In addition, both the “packing material” and the “microbial community” in the figure are hot topics and major trends of research in recent years. The previous biological methods were limited in treating high concentrations of organic and mixed waste gases. Therefore, the performance of bioreactors has been increasingly researched in recent years, mainly in the exploration of composite functional packing materials and functional microbial communities. The packing material is a solid medium for microbial growth and biofilm formation, and its characteristics, such as its nature, particle size, and moisture content [69], affect the performance of bioreactors. The nutrient level in the packing material [70] guarantees microbial growth and metabolism. The organic packing materials in the biological methods are generally compost [71,72], peat [73], soil [74], etc., while inorganic packing materials include perlite, ceramic, etc. When selecting packing materials, attention should be given to the characteristics of large specific surface area, high porosity, and strong water-retention capacity. To improve the removal efficiency and service life of packing materials, composite materials and new functional packing materials have been continuously developed. Dumont et al. [75] composited calcium carbonate, urea, phosphoric acid, and other materials to form a new type of packing material, with a removal rate of more than 90\% of organic waste gas. A composite material can compensate for the shortcomings of a single material and enhance the efficiency of composite pollutant removal [76]. Han et al. filled a biofilter with perlite and a high-strength 3D matrix material, which not only has a higher mass transfer efficiency [77] but also has a lower pressure drop and can maintain long-term performance [78].

The microbial community is a crucial parameter in bioreactors, and the species and growth of microorganisms are closely related to the degradation of pollutants. Therefore, the analysis of the microbial community in the system is a key point to increase the scope of application and removal efficiency of biological methods. Bacteria and fungi are among the microbial species that are used in biological methods. The genus Pseudomonas is suitable for the removal of VOCs and malodorous gases; the genus Rhodococcus is suitable for the removal of toluene, naphthalene, and other substances [79]. The genera Saccharomyces, Mycobacterium, and Serratia in fungi [80] can also be used in bioreactors. Fungal microorganisms have a filamentous structure and a strong ability to adsorb VOCs, which enhance the removal efficiency of exhaust gases [81]. Currently, the microorganisms used in biological systems are mainly bacteria, but their practical application varies depending on the pollutants to be removed and the environmental conditions. For instance, Zheng et al. [82] discovered that the microbial community in bioreactors depends on the inlet load of pollutants and the characteristics of packing materials. Liu et al. [83] constructed a novel
three-stage biofilter composed of acidophilic bacteria, fungi, and heterotrophic bacteria, which could achieve high and stable removal of ammonia, hydrogen sulfide, and VOCs. With the advancement of biotechnology and new materials, the screening and cultivation of functional microorganisms and the development of 3D porous composites will be future research trends.

6. Conclusions and Outlook

In this paper, the biological removal of VOCs was taken as the subject of research, and the bibliometric software CiteSpace was employed to analyze 837 research articles and reviews in the core WOSCC database. The analysis indicated that between 1966 and 2021, the number of papers in the field of biological methods to remove VOCs increased significantly after 1997. The United States, Spain, and other countries have close cooperation with other countries, and leading research institutions are primarily located in countries such as the United States, China, Belgium, and Spain. They have made significant contributions to the research in this field.

According to the analysis of research hotspots and trends, the field of biological treatment of VOCs concentrated on the removal of malodorous gases and single species of VOCs in the early stage. However, with the growing demand for the treatment of refractory organic waste gases, research on the inadequacy of conventional biofilters for the removal of organic waste gases has intensified and expanded. Benzene, hydrophobic compounds, and multi-component mixed waste gases have been the most prevalent types of pollutants studied. Current research focuses on the interactions of mixed VOCs during the removal process, microbial communities, and packing material innovations.

To achieve more efficient and energy-saving purposes, future research on biological methods for VOC removal should be based on theoretical knowledge, such as the operating mechanisms of bioreactors, combined with practical application experience of existing biological treatment technologies. The focus should be on developing new biological treatment reactors and their processes, as well as improving the efficiency of bioreactors by strengthening the development of new composite packing materials and the screening and cultivation of functional microorganisms for degrading specific pollutants. This will enable us to meet practical application requirements and actively respond to air pollution concerns.

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