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Quantitative Analysis of the Research Development Status and Trends of Tannery Wastewater Treatment Technology

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Abstract: In order to better grasp the development and trends of tannery wastewater (TWW) treatment research, this paper provides a review of the TWW treatment research dynamics based on the Web of Science (WoS) database and using CiteSpace software. The research dynamics, hot topics, evolutionary history and research trends in this field are revealed. The results showed that research related to TWW treatment has shown a high growth trend in the number of articles in recent years, and India was outstanding in terms of influence in this area. The keyword clustering analysis showed that the main research hotspots in the field of TWW treatment were biological treatment processes (phytoremediation, constructed wetlands, anaerobic treatment and biofilm reactors) and chemical treatment processes (coagulation and flocculation, and advanced oxidation processes). The analysis of new research frontiers showed that the bioremediation and the application of biofuel cells in TWW will become important research directions in the future.

Keywords: tannery wastewater; CiteSpace; biological treatment processes; chemical treatment processes

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Citation: Li, M.; Jia, X.; Wang, J.; Wang, Y.; Chen, Y.; Wu, J.; Wang, Y.; Shen, M.; Xue, H. Quantitative Analysis of the Research Development Status and Trends of Tannery Wastewater Treatment Technology. *Catalysts* **2022**, *12*, 1317. https://doi.org/10.3390/catal12111317

Academic Editors: Xinjiang Hu and Aurora Santos

Received: 26 September 2022
Accepted: 23 October 2022
Published: 27 October 2022

1. Introduction

The tannery industry is one of the traditional industries with high pollution and water consumption. The contamination of tannery wastewater (TWW) mainly comes from raw materials and auxiliary agents (proteins, acids, bases, chromium salts, sulfides, chlorides, tannins, solvents, dyes, auxiliaries and other compounds) that were not transformed into products in the tanning process [1–3]. Therefore, the concentrations of chemical oxygen demand (COD), biological oxygen demand (BOD), total dissolved solids (TDSs), total suspended solids (TSSs), chromium and phenolics in TWW were generally high, with the wastewater having a strong odor and dark-brown color [4,5]. In addition, TWW contains a variety of nutrients such as nitrogen and phosphorus [6–8]. The TSSs (35–250 mg/L), BOD (250–2960 mg/L) and chromium (4.5–15 mg/L) were still detected in the TWW [9–12]. Studies on pollutant hazards, genotoxicity and environmental behavior in TWW have shown [13–15] that TWW is potentially harmful to the ecological environment and human health, and therefore, the pollution problem in the tanning industry is receiving increasing attention. The development of clean production and wastewater recycling technology is an inevitable trend for sustainable and benign development. The reduction in pollutants from tanning was achieved through tanning agent recycling [16], alternative carrier media [17], the enzymatic dehairing process [18], and efficient management [19]. At the same time, the environmental pollution and resource waste in the tannery industry were effectively controlled by developing new processes to improve the treatment efficiency, achieving the efficient treatment and reuse of TWW. However, comprehensive quantitative studies of TWW treatment technology lack systematic summaries, and the characteristics and evolutionary features were less reported.

Scientometric analysis refers to the use of statistical methods to quantitatively analyze the textual information of scientific papers in a certain field, to sort out the research lineage of a certain field, analyze the research hotspots and evolutionary trends, and explore the future research hotspots and directions [20], which is important for a comprehensive understanding of a certain field of research [21–23]. CiteSpace is the main literature information mining software based on scientometrics, data and information visualization techniques [24,25]. It not only performs a co-citation analysis of the literature and mines clustering information, but also visualizes the research status and predicts emerging trends, ultimately revealing the research patterns of a research field in a comprehensive manner.

Therefore, the CiteSpace visualization tool was used in this study to conduct textual information mining on TWW-related studies published in the Web of Science (WOS) from 1991 to 2022. The objectives of the study were divided into the following three directions: (1) to understand the publication trends, sources, publications, disciplinary distribution, and major authors, countries and institutions of TWW research; (2) to identify major national and institutional collaborations in the field of TWW research; and (3) to summarize existing hot issues and explore research prospects and trends based on keyword clustering analysis. The research results can visually show the research hotspots and research trends, major authors, institutions, journals and their collaborations in the field of TWW, which can help us to grasp the content and development trends of TWW research, and thus, clarify the future research direction.

2. Data and Methods

2.1. Data Collection

This research used the materials of the Web of Science Core Collection citation database, including SCI-EXPANDED, SSCI, A&HCI, CPCI–S, CPCI-SSH, BKCI–S, BKCI-SSH, ESCI, CCR-EXPANDED and IC. More than 12,400 authoritative and highly influential international academic journals are included in WOS, covering the natural sciences, engineering, social sciences, and arts and humanities, with content dating back to 1900 [26]. The high-quality documentation provided by WOS is widely used in scientometric studies [27]. Therefore, WOS was chosen as the literature database in this study. The search was conducted on 7 July 2022. The search query used to extract data from the WOS core database was topic terms = ((tannery) AND (wastewater treatment)), with no other restrictions, and 1101 results were retrieved. Finally, 979 pieces of research papers were selected for this study, since we manually excluded conferences and books.

2.2. Methods

The CiteSpace software (Version 5.1) was used for visual analysis. In this software, the options of author, institution, keyword and journal were selected [24,28], and the node strength, threshold and network cropping functional area parameters were set according to the different characteristics of the WOS database for the literature. Finally, the statistics and co-occurrence mapping of TWW treatment research areas were obtained to analyze their research status, hotspots and frontiers. In the knowledge graph, larger nodes or fonts indicate higher frequency of occurrence. The circle layer of nodes represents the chronology, and the purple circle layer is the centrality of the key label (centrality > 0.1). The width of the chronology can refer to the size of centrality as a way to reflect the structure and influence of authors, institutions, keywords, etc. The larger the circle in the graph, the higher the frequency, and the different colors reflect the research years of different countries, whereas the connecting lines also represent the cooperation status between countries.

3. Results and Discussion

3.1. Analysis of Basic Characteristics of Publications

3.1.1. Trend Analysis of Publications

The trend of TWW treatment research can be reflected by the temporal distribution of the number of studies in the literature. In the last 30 years, the number of publications in the field of TWW treatment research has generally shown a significant growth trend (Figure 1). This indicates that TWW treatment research, in general, was evolving, progressing and receiving continued attention.

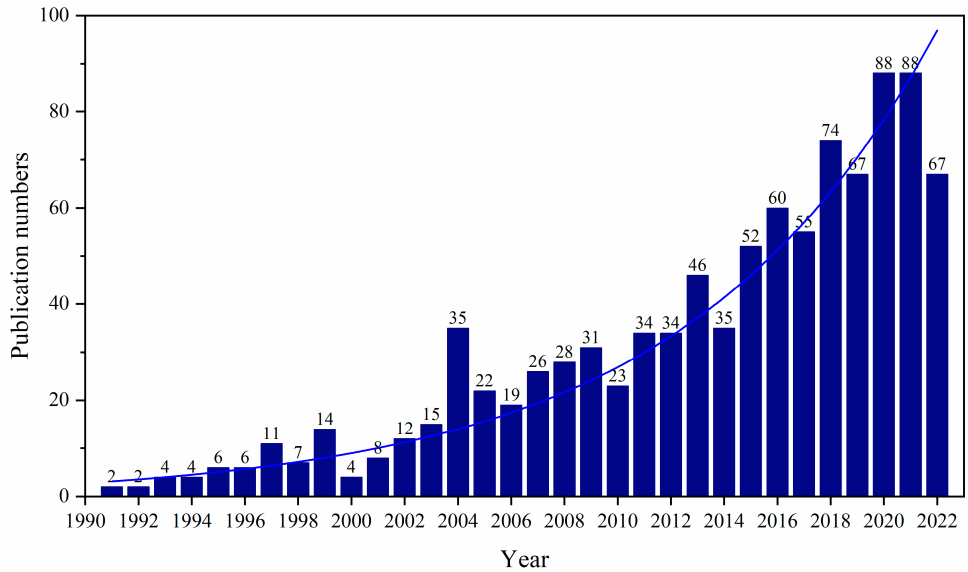

Figure 1. Temporal distribution of published articles on tannery wastewater from 1991 to 2022. Out of the 979 papers published between 1991 and 2022, the number of publications each year on tannery wastewater is shown. The research into tannery wastewater began in 1991, and the number of publications has steadily increased over time. Research on tannery wastewater has been divided into three distinct periods: an "embryonic" phase (1991–2003), a "maturity" period (2004–2012) and an "explosive growth" period (2013–2022). The amount of attention paid to tannery wastewater varies over time, and the number of articles published each year on a particular topic might indicate how far research in that field has progressed.

The average annual number of articles published in the TWW treatment research area reached 30.59, and the total number of articles is still gradually increasing. The current status of TWW treatment research development can be divided into three stages (Figure 1). The first stage (1991–2003) is the embryonic stage of TWW treatment, with a low annual publication volume of 7.3 articles, indicating that the research on TWW treatment had just started, the number of scholars concerned was small and the research topic was relatively single. The second stage (2004–2012) is the maturity period of TWW treatment research, where the number of studies in the literature began to grow steadily (the average annual number of publications was 28), and the number of scholars concerned gradually increased. The third stage (2013–2022) is the explosive growth period of TWW treatment research, with the average annual number of publications exceeding 63 and the research team gradually gaining a certain system and scale. Overall, the trend of TWW treatment research publications showed an exponential growth (R^2 = 0.909), and there will be in-depth research results in this field in the future.

3.1.2. Analysis of Disciplines and Journals

The subject correlation analysis establishes a link between the citing and cited subjects, where the citations form the research frontier and the cited subjects form the knowledge base. By selecting the journal map option from the overlay maps option in CiteSpace, the information service data were overlaid onto the original citation-cited literature discipline base map by adding the overlay, and finally, the dual-map overlay of the discipline distribution involved in the TWW treatment research area was obtained (Figure 2). The left half of Figure 2 shows the distribution of the cited literature by discipline as the current status of TWW treatment research, and the right half shows the research base of the TWW treatment discipline to which the cited literature belongs of. The curves connect the relationship between the research status and the research base, and the inner numbers of the ellipse indicate the number of publications in each discipline [24].

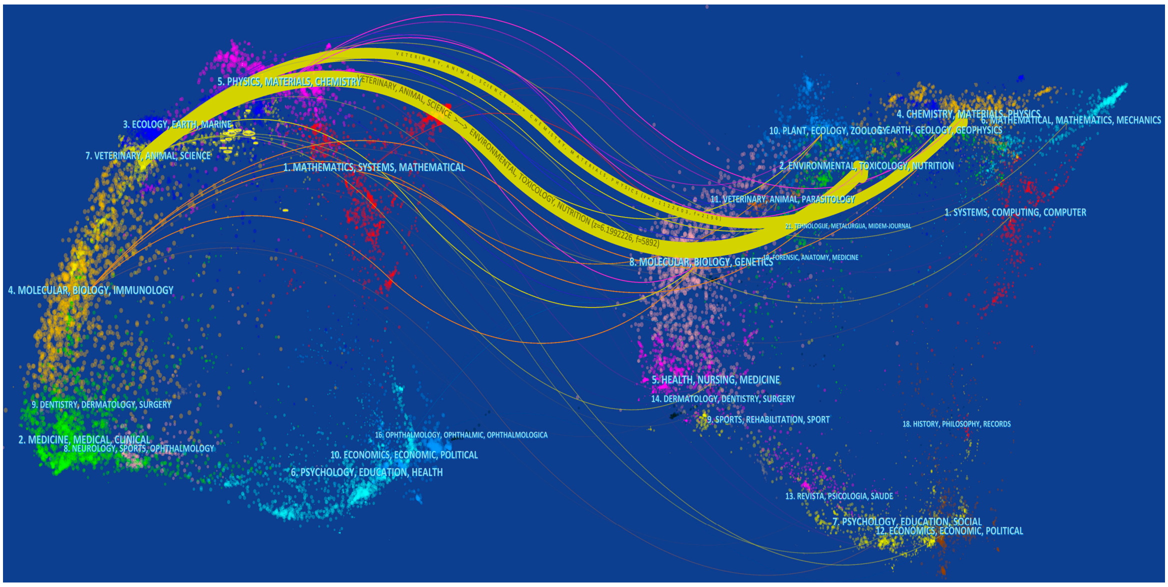

Figure 2. Double-map overlay for tannery wastewater research. Analysis of tannery wastewater using a double-map overlay is illustrated. On the left side of figure, the most widely cited research literature on tannery wastewater covers the fields of veterinary medicine, zoology and science. Environmental science, toxicology and nutrition are all included in the study of tannery wastewater on the right side.

Identifying the distribution of TWW treatment research in different journals and in which journals it is cited can reveal the dynamics of journal knowledge flow in the WOS database. The research in TWW treatment domains was concentrated in the medical, neurological, molecular, mathematical, physical, and ecological journal groups. The citations from the TWW treatment domains were concentrated in the botanical, chemical, mathematical, environmental, systematics, and molecular journal groups. Among them, there were two outward citation paths in veterinary medicine, zoology and science in the citation domain on the left side of the figure, indicating that this group is the most dominant citation group. When the group of veterinary medicine, zoology and science was used as a source journal, the corresponding group of environmental science, toxicology and nutrition has the highest number of citations (Z value of 6.3039).

In this study, the current status of the distribution of the top 10 journals in the field of TWW treatment research was calculated (Table 1). Within the field of TWW treatment research, the number of articles published in the journals *Desalination and Water Treatment*, *Water Science & Technology*, *Journal of the American Leather Chemists Association* and *Journal of Environmental Chemical Engineering* were 48, 46, 27, 26 and 24, respectively. The total publication ratio of the top 10 journals was 44.64%, which indicates that the distribution of the TWW treatment literature was relatively scattered and the research results were not concentrated in a few journals. In addition, the main research directions of the top 10 journals were environmental protection, leather process, chemistry, water treatment, etc.

The above research results show that different scholars have deepened and improved TWW treatment technology from different perspectives and directions.

Table 1. Top 10 productive journals in tannery wastewater research. This list summarizes the ten most prolific journals in the field of tannery wastewater research from 1991 to 2022. *Desalination and Water Treatment* is on top of the list, followed by *Water Science & Technology*.

Serial Number	Journal	Papers	Proportion
1	*Desalination and Water Treatment*	48	4.90%
2	*Water Science & Technology*	46	4.70%
3	*Journal of the American Leather Chemists Association*	27	2.76%
4	*Journal of Environmental Chemical Engineering*	26	2.66%
5	*Desalination*	24	2.45%
6	*Journal of the Society of Leather Technologists and Chemists*	24	2.45%
7	*Journal of Hazardous Materials*	22	2.25%
8	*Journal of Water Process Engineering*	22	2.25%
9	*Water Research*	22	2.25%
10	*Chemosphere*	21	2.15%

3.1.3. Analysis of Countries/Regions

The scientometric analysis of the issuing countries can not only identify the core countries in the research field of TWW treatment, but can also reflect the academic exchanges and cooperation among the research countries in the field. In this study, the country was selected as the analysis object in CiteSpace, and the time slicing was set to “1991–2022”, where the years per slice was set to 2, and the threshold was top 50; finally, the country analysis map was generated (52 network nodes, 54 lines, and a density of 0.0407). This is shown in Figure 3.

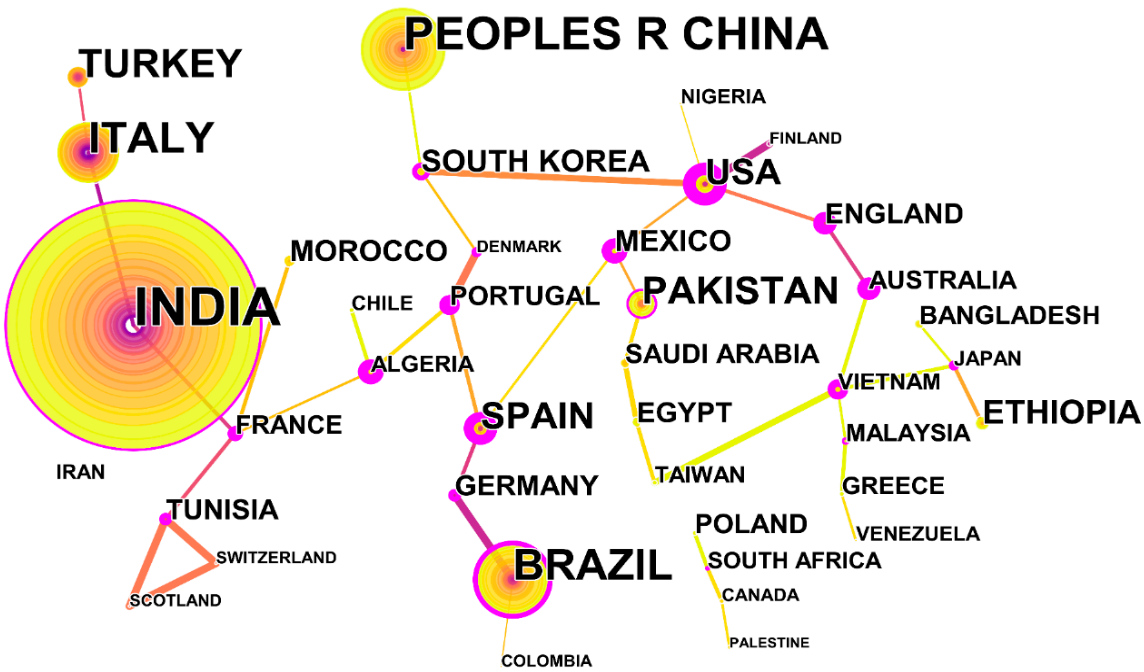

Figure 3. Countries performing tannery wastewater research. Tannery wastewater research is being pursued by governments across the globe, as indicated. In comparison to other countries and regions, India is the most productive country. Cooperative ties between countries and areas are depicted in Figure 3 by the dotted lines. Italy and India have a long history of cooperation. Although showing a low level of cooperation among countries and regions, the weak connections between nodes suggest a future need for further cooperation.

The top 10 countries in terms of frequency are shown (Table 2); India has the highest number of publications in this field with 265, far ahead of other countries, indicating that India is very interested in the TWW treatment field. In addition, the TWW treatment technology in India is also developing relatively fast, helping India gradually become the leader in this field. China ranked second with a frequency of 104, indicating that China has paid increasing attention to TWW treatment technology and is constantly innovating with regard to TWW technology. Italy and Brazil ranked third and fourth, respectively, with frequencies of 83 and 81. For centrality, the USA is the highest with 81, which indicates that the USA is at the heart of the field. The close relationship between the USA and other countries indicates that researchers from other countries tend to collaborate with those from the USA, which may be a result of the help the USA gives to other countries at the technical level. At the same time, 9 of the top 10 countries have publication years before 2005 with the highest frequency, indicating that TWW treatment attracted much attention at the early stage, and that TWW treatment research is a current and even future research direction.

Table 2. Top 10 productive countries in tannery wastewater research. This list summarizes the ten most prolific countries in the field of tannery wastewater research from 1991 to 2022. India is on top of the list, followed by China.

Serial Number	Frequency	Centrality	Country	Year
1	265	0.14	India	1999
2	104	0	China	2001
3	83	0.07	Italy	1997
4	81	0.21	Brazil	2004
5	51	0	Turkey	2000
6	46	0.13	Pakistan	2007
7	39	0.72	Spain	1999
8	36	0.81	USA	2003
9	26	0.07	Ethiopia	2004
10	21	0.29	South Korea	1996

3.1.4. Analysis of Institutions

The institution analysis mapping (132 network nodes, 47 connections, density 0.0054) was obtained by using the institution as the analysis object in CiteSpace (Figure 4). The nodes in the map are scattered, and the cooperation of each institution is in small groups, which indicates that the research institutions in this field are more independent in this research. In addition, most of the research institutions have fixed targets for collaboration, and some of them have obvious geographical characteristics of collaboration. In other words, the cross-institutional cooperation handling TWW research needs to be strengthened and the scope of cooperation needs to be expanded. The top 10 issuing institutions are shown in Table 3. Among them, "year" represents the year of publication, and half-life is the number of years that shows how the literature is calculated forward from the statistical year, accounting for 50% of the total number of citations of the literature up to the statistical year, which represents the classic degree of the literature [29]. The greater the half-life, the longer the influence. Univ Florence had the largest half-life period, indicating that its publications had the longest impact. In terms of frequency, Cent Leather Res Inst has the highest number of articles with 45, because Cent Leather Res Inst is an Indian institute specializing in tanning technology. In addition, Anna Univ is in second place with 29 articles and Sichuan Univ is in third place with 20 articles. For the time of publication, Cent Leather Res Inst, Anna Univ and Istanbul Tech Univ all have their year of publication as 2005, which indicates that these three highly productive institutions started their research on TWW treatment earlier and contributed more, dominating the research in this field. However, Univ Fed Rio Grande do Sul, Univ Agr Faisalabad, Addis Ababa Univ and CSIR Cent Leather Res Inst all initially published in 2015, and later, had a higher number of publications, which indicates that the research content of these four institutions is on current cutting-edge topics in TWW treatment and is more adequately studied.

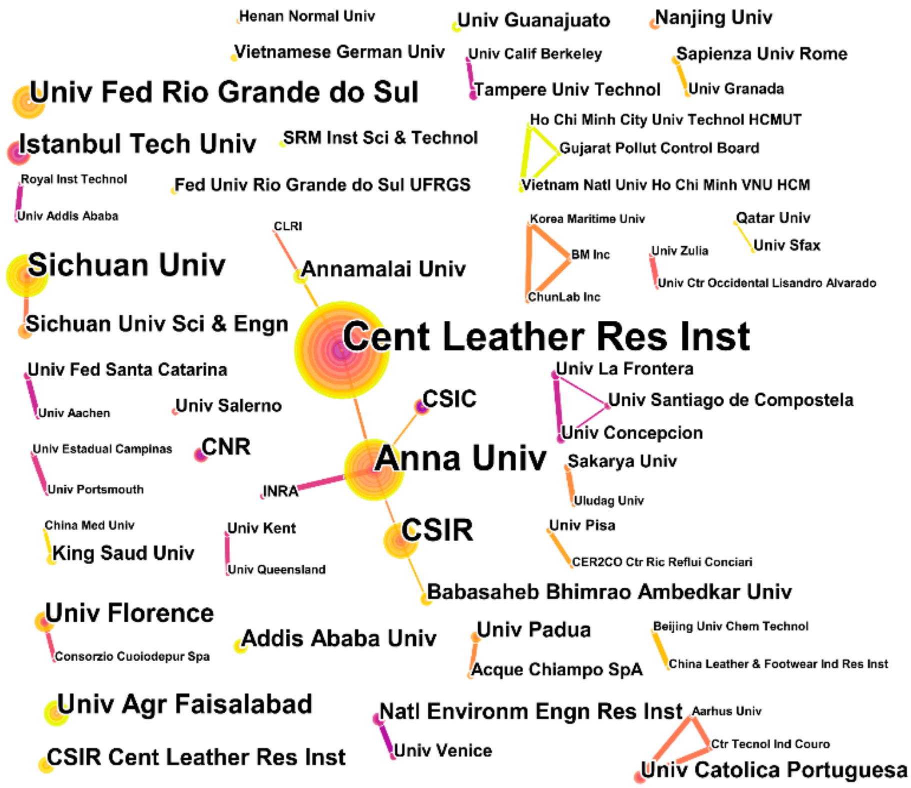

Figure 4. Institutions and their cooperation relationships with regard to tannery wastewater research. Scholars collaborate on research projects via co-authoring articles, which is a significant method of academic cooperation. Because of this, it is critical to know how academics communicate, including information on affiliations and countries, as well as other key author traits. Scholarly collaboration can help research progress. For example, the contributions of diverse students can lead to a better understanding of a particular research area.

Table 3. Top 10 productive institutions in tannery wastewater research. This list summarizes the 10 most prolific institutions in the field of tannery wastewater research from 1991 to 2022. The Cent Leather Res Inst is on top of the list, followed by Anna Univ.

Serial Number	Frequency	Institution	Year	Half-Value Period
1	45	Cent Leather Res Inst	2004	7
2	29	Anna Univ	2005	9
3	20	Sichuan Univ	2011	7
4	17	CSIR	2011	5
5	15	Univ Fed Rio Grande Do Sul	2015	1
6	12	Istanbul Tech Univ	2002	4
7	12	Univ Agr Faisalabad	2015	3
8	10	Univ Florence	2007	10
9	8	Addis Ababa Univ	2017	2
10	8	CSIR Cent Leather Res Inst	2016	3

3.1.5. Analysis of Authors

The network analysis of the authors of publications not only identifies the core authors in the field, but also reflects the academic exchanges and collaborations among researchers. In this study, the author was selected as the analysis object in CiteSpace with a years per slice of 2 and a threshold value of a g-index of 5. The author analysis graph was finally obtained

(170 network nodes, 132 connections and a density of 0.0092). This is shown in Figure 5 and Top 10 productive authors in tannery wastewater research shown in Table 4. The graph of authors is similar to institutions, since the nodes are dispersed and the collaboration of individual authors shows small groups, with relatively independent collaboration between authors. According to Price Law, the total number of authors engaged in TWW treatment research from 1991 to 2022 was 2968, and the square root was 54.48, indicating that the number of core authors in the field of TWW treatment was 55 [28]. Statistically, the number of publications by these core authors was 435, accounting for about 44.43% of the total number of papers, indicating that a stable core group of authors has not been formed in the field of TWW treatment.

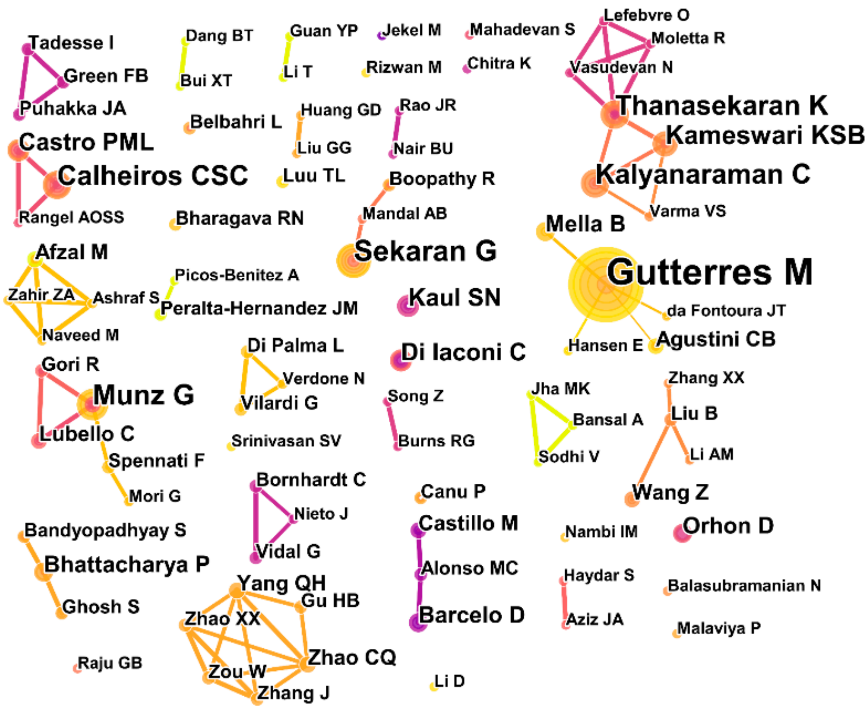

Figure 5. Authors and their cooperation relationship in tannery wastewater research. Scholarly collaboration can help research progress. For example, the contributions of diverse students can lead to a better understanding of a particular research area. The analytical field "author" was used to import the data into the software. A two-year time slice (time slice = 2) and a threshold value of n = 50 were used to determine the time period from 1991 to 2022. Tannery wastewater research collaboration between authors from 1991 to 2022.

Table 4. Top 10 productive authors in tannery wastewater research. This list summarizes the ten most prolific writers in the field of tannery wastewater research from 1991 to 2022. Gutterres M is on top of the list, followed by Sekaran G.

Serial Number	Frequency	Author	Year	Half-Value Period
1	24	Gutterres M	2015	3
2	11	Sekaran G	2012	3
3	10	Munz G	2007	10
4	9	Thanasekaran K	2005	6
5	9	Calheiros CSC	2007	2
6	9	Kalyanaraman C	2011	2
7	8	Kameswari KSB	2011	2
8	7	Kaul SN	2001	3
9	7	Di Iaconi C	2001	2
10	7	Castro PML	2007	2

In addition, a collaborative team with Gutterres M, Sekaran G, Munz G, Thanasekaran K and Calheiros CSC as the main representatives in the field of TWW processing research has been formed. The year of the first publication from the Gutterres M team was 2015, indicating that the team started late, but studied more adequately and produced a large number of results in a short period of time. The year of the first publication of Kaul SN and Di Iaconi C was 2001, which indicates that these two teams started their research on TWW treatment earlier and played a pioneering role. In addition, the half-life (10) of the Munz G team of authors is much larger than that of other authors. This indicates that the research value of the Munz G team is high and has been recognized and paid attention to by many scholars, and there is a great value of references and room for deeper digging. Taken together, the awareness of academic cooperation among scholars from different countries needs to be enhanced, the core team needs to be formed, and the academic influence in the field of TWW processing needs to be enhanced urgently.

3.2. Analysis of Research Hotspots

3.2.1. Analysis of Keyword Network

The hot issues, developmental aspects and knowledge base can be revealed by the analysis and summarization of keywords related to TWW treatment research. In this paper, CiteSpace software was used to analyze keywords in the TWW treatment domain, and the keywords were selected to generate the co-occurrence map as shown in Figure 6 (years per slice is 3, 116 nodes, 137 connected lines and a density of 0.0205).

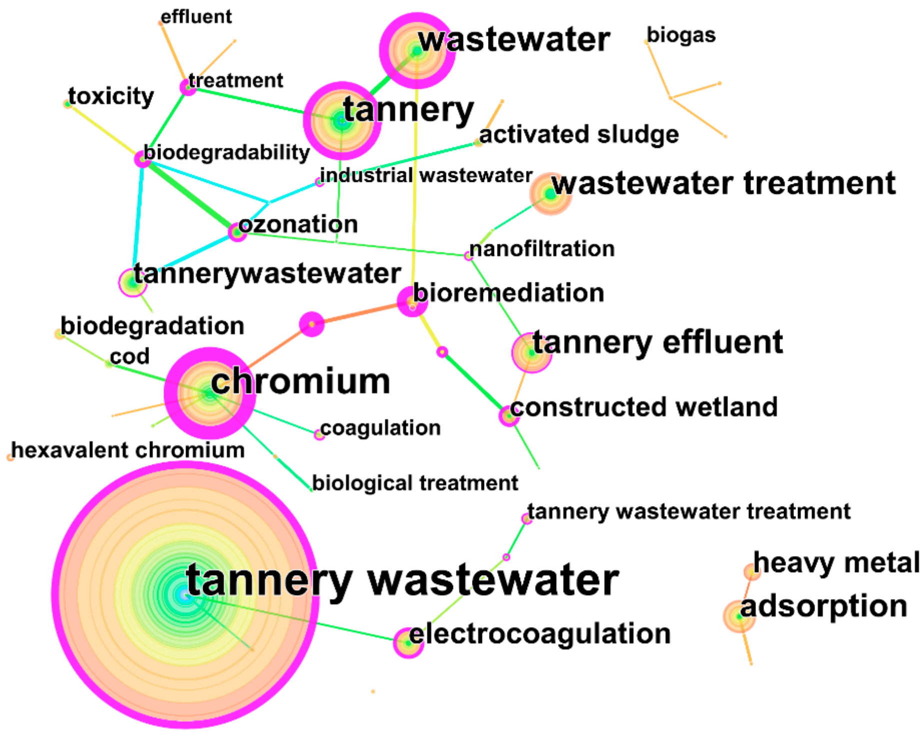

Figure 6. Keyword co-occurrence map of tannery wastewater research. It demonstrates that "tannery wastewater", "chromium" and "tannery" are often used terms in tannery wastewater studies (228, 77 and 67). Because "tannery wastewater" is the topic of this study, it is expected that it ranks #1 in frequency. There are other keywords such as "wastewater treatment", "adsorption" and "electrocoagulation" that are popular topics in tannery wastewater studies.

In this study, the top 20 keywords with the highest frequency are listed in Table 5. For frequency, chromium had the highest frequency, reaching 77. For centrality, chromium still ranked first with a value of 1.24. It shows that chromium removal is a hot topic in the field of TWW processing [30], and the research in this field mainly focuses on chromium.

In addition, the most frequently used words were "tannery" (67), "wastewater" (61), "wastewater treatment" (48), "tannery Effluent" (44), "adsorption" (40), "electrocoagulation (26)", "bioremediation" (18), etc. From the word frequency, it can be seen that most of the research fields with regard to TWW treatment focus on the characteristics and treatment methods of TWW, among which bioremediation, electroflocculation and adsorption are currently hot topics [31–34]. The main characteristics of TWW are high organic loads and specific pollutants such as chromium [35]. Bioremediation and electroflocculation are environmentally friendly and by-product-free treatment technologies, which can effectively remove specific pollutants from TWW. In addition, the removal rate of chromium from TWW, such as via a cation-exchange resin and the adsorption method, can reach more than 95% [36]. Therefore, the adsorption method has a good application potential in removing chromium from TWW. For the half-life period, activated sludge and heavy metal are the highest, reaching 21 and 22, indicating that these two keywords have a far-reaching influence. In addition, there is still a lot of space for future research in biological treatment and chromium removal, which is worthy of in-depth exploration by scholars.

Table 5. Top 20 keywords in terms of frequency in tannery wastewater research. The greater the size of the node, the more frequently the keyword it represents appears in search results. The color of the node changes from chilly to warm as it moves from the center to the outside. The specifics of the top 20 high-frequency keywords in tannery wastewater studies are described in greater depth.

Serial Number	Frequency	Centrality	Keyword	Year	Half-Value Period
1	228	0.7	tannery wastewater	1996	20
2	77	1.24	chromium	1997	18
3	67	0.71	tannery	1995	20
4	61	0.81	wastewater	2001	17
5	48	0.05	wastewater treatment	2001	16
6	44	0.11	tannery effluent	2004	13
7	40	0.03	adsorption	2003	15
8	29	0.13	tannerywastewater	1997	15
9	28	0.1	heavy metal	1997	22
10	26	0.25	electrocoagulation	2007	9
11	21	0.26	constructed wetland	2006	8
12	18	0.97	bioremediation	2015	3
13	16	0.25	ozonation	1997	14
14	16	0.07	biodegradation	2004	12
15	16	0.07	activated sludge	1992	21
16	14	0.05	toxicity	2004	4
17	12	0.13	tannery wastewater treatment	2006	13
18	12	0.1	COD	2009	4
19	12	0	hexavalent chromium	2018	1
20	11	0.13	nanofiltration	2011	3

3.2.2. Analysis of Keyword Clustering

The keywords were clustered by CiteSpace (Figure 7), and the analysis results reflect the research themes of TWW treatment in the last 30 years; the information of each cluster is shown in Table S1. The cluster numbers were the 11 clustered themes resulting from the clustering of keywords by the LLR algorithm, which can be generally classified into biological, chemical and physical methods. The Q-value and S-value are indicators to describe the network structure and clustering. When the cluster analysis parameters' Q-value is greater than 0.3 and S-value is greater than 0.7, the clustering results are convincing [37]. The Q-value and S-value of this study are 0.7905 and 0.8034, respectively, which meet the criteria of the study. Therefore, the CiteSpace clustering results are more reasonable and can be followed up for analysis. The biological, chemical and physical methods of treating TWW are analyzed, in turn, below.

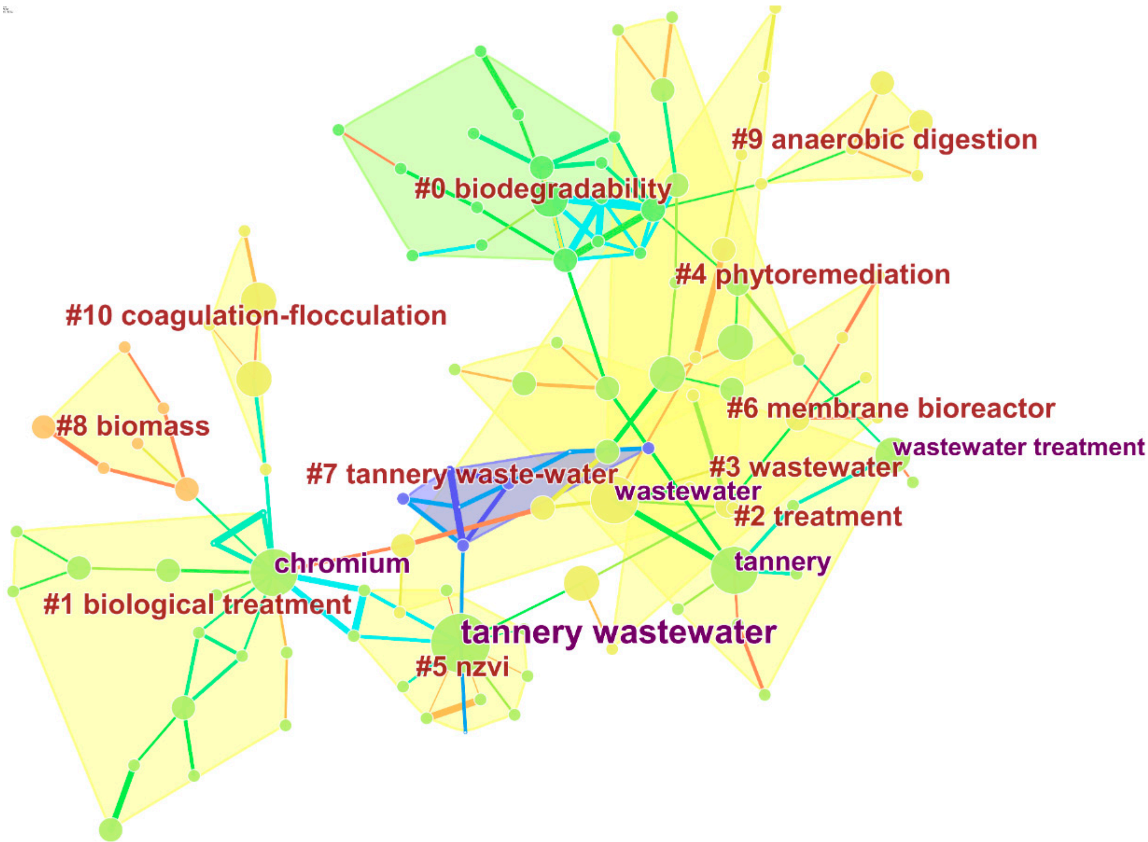

Figure 7. Co-occurrence clusters of the keyword map of tannery wastewater research. It shows that "#0 biodegradability", "#1 biological treatment" and "#10 coagulation-flocculation" are frequently used terms in tannery wastewater research. In addition, cluster keywords such as "#4 phytoremediation", "#6 membrane bioreactor" and "#9 anaerobic digestion" were also hotspots of tannery wastewater research.

The biological method was mainly composed of the keywords of #0 biodegradability, #1 biological treatment, #2 treatment, #4 phytoremediation, #6 membrane bioreactor, #8 biomass and #9 anaerobic digestion, indicating that the biological method is one of the main means of TWW treatment. In addition, the biological method is an environmentally friendly method for treating industrial wastewater, where the main process is the decomposition of waste into harmless and stable inorganic solids through aerobic or anaerobic processes. Additionally, the most commonly used methods for the biological treatment of TWW are an activated sludge process, upflow anaerobic sludge blanket, phytoremediation, membrane bioreactor and anaerobic treatment [38–42]. The activated sludge process is a traditional biological treatment method widely used as a process for TWW treatment. It allows sufficient contact and mixing with water, good sludge settling performance, and good biological denitrification [43,44]. Another traditional biological treatment method is the upflow anaerobic sludge blanket, which is characterized by a short hydraulic residence time, high volumetric load, simple construction and easy operation [39]. The application of the anaerobic treatment in water treatment has changed the monolithic nature of TWW treatment, which is characterized by low energy consumption and low sludge production. The anaerobic filters, consisting of upflow anaerobic filters and anaerobic filters, are mainly used to treat TWW [42,45]. Phytoremediation is widely used as a biological treatment for TWW treatment, and is most represented by constructed wetlands, which can effectively remove pollutants from highly polluted industrial and municipal wastewater [46]. Constructed wetlands are small ecosystems composed of some plants, microorganisms and aquatic animals that serve to purify pollutants through physical, chemical and biological purification processes [30,40,47–49]. In addition, membrane bioreactors, which are a combination of membrane processes such as microfiltration or ultrafiltration and bioreactors, are also gradually applied in TWW treatment [50,51]. The sludge settling performance is not affected by filamentous swelling when eliminating sludge from the settling tank [45,52,53]. In recent years, some researchers have also introduced microbial fuel cells to treat TWW as a new method [54].

The chemical method was composed of the keywords of #10 coagulation-flocculation, #5 nZVI (nano zero-valent iron), and #7 tannery waste-water, indicating that the chemical method occupies an important position in TWW treatment. The chemical method has the advantages of the easy operation of equipment, easy automatic detection and control, and easy recycling [55], and can be used as a pretreatment process for TWW. In addition, the most commonly used chemical methods to treat TWW are coagulation and flocculation, the advanced oxidation process, the Fenton oxidation process, and electrochemical treatment technology [56–59]. Flocculation and coagulation are the most basic chemical treatment methods and are usually used as premethods or postmethods. Traditional inorganic flocculants or coagulants such as polymeric aluminum chloride (PAC) [60], ferric chloride [60], alum [61], polymeric silicate [62] and polymeric aluminum chloride ferric (PAFC) [63] have been used to study the coagulation/flocculation process of TWW [64] and to reduce the concentration of COD, TSS, chromaticity and chromium before further TWW treatment [56,61,65,66]. Advanced oxidation processes are a collection of chemical process treatments. They refer to the method of oxidation and degradation of pollutants using strong oxidants (O_3, H_2O_2) and catalysts (Fe, Mn, TiO_2), while sometimes also using the electrical current and high energy radiation from UV lamps [45,57,67–70]. This method can efficiently oxidize most of the recalcitrant organic pollutants (e.g., benzoquinone, benzene, phenols, chlorophenols, dyes and formaldehyde) [1,45]. The advanced oxidation processes include processes such as Fenton oxidation, photo-oxidation, ozonation and photocatalysis, which are widely used to treat TWW [1,45,70–72]. Advanced oxidation processes can reduce the pollution load and toxicity during the treatment of TWW, allowing the treated TWW to be reintroduced into the receiving water or reused in the process. In addition, another type of advanced oxidation is the electrochemical treatment, a process in which an electric current is passed through an aqueous solution containing metals. It is characterized by its environmental friendliness, good versatility, and high efficiency, and is generally applicable to the treatment of TWW [72,73]. The electrochemical treatment includes processes such as electrodeposition, electrocoagulation, electrodisinfection, electro-oxidation, electroreduction, electropermeation, and electroflotation [2,33,72].

The physical method consisted of the keywords of ceramic membrane, microfiltration, etc. in #3 wastewater clustering. Researchers have combined nanofiltration and reverse osmosis processes to recover water from membrane systems of TWW successfully [74]. In addition, membrane filtration has been a hot area for TWW treatment, especially through the application of ceramic membranes in the TWW treatment [75,76].

3.3. Analysis of Research Trends

3.3.1. Research Frontiers

The frontier and focus of development trends in a certain field can be expressed by emergent keywords [21]. Kleinberg proposed a burst detection algorithm for keywords in 2002. Burst keywords are defined as keywords with a sudden increase in their relative growth rate during a specific period of time [28]. CiteSpace software has a burst detection function, which can find content that does not reach the frequency threshold but has important informatic significance in the development of a research field. By detecting sudden changes in keywords, keywords representing research frontiers and development trends can be more truly and scientifically displayed. To grasp the topic development of a research field [77]. CiteSpace software has a burst detection function, which can find content that does not reach the frequency threshold but has important informatics significance in the development of a research field. By detecting sudden changes in keywords, keywords representing research frontiers and development trends can be more truly and scientifically displayed. Additionally, we can then grasp the topic development of a certain research field [77].

Through the detection algorithm of the CiteSpace software, the top 25 emergence intensity keywords in the field of TWW were formed. The specific emergence intensities and hotspot durations are shown in Table 6. The outbreak intensity of the tannery industry was the strongest from 1991 to 2022, which was 3.77 from 2009 to 2014. However, the bursting strength of "biological treatment" and "tannery waste-water treatment" was 3.62 and 3.29, respectively. From 2020 to 2022, chromium recovery and water treatment emerged as new research frontiers. Chromium is a major metal contaminant in TWW, which is a topic that needs urgent attention. Notably, biological treatments have excellent effects on chromium recovery and removal because they can accumulate and transport the metal [78]. In terms of biological treatment, especially chromium treatment in constructed wetlands, plant selection is a problem. Future studies focusing on the biological treatment of tanning wastewater, chromium transfer, potential sources and toxicity mechanisms are needed. In conclusion, the CiteSpace software has a burst detection function, which can find content that does not reach the frequency threshold, but has important informatic significance for the development of a research field. By detecting sudden changes in keywords, keywords representing research frontiers and development trends can be more truly and scientifically displayed.

Table 6. The top 25 keywords with the highest number of citation bursts are listed below (red bars indicate keywords that have been referenced frequently; blue bars show keywords that have been cited infrequently). There is a clear indication of the time interval and burst strength of the keywords. "Year" refers to the time of the first occurrence of the mutation in the retrieval record, "Strength" refers to the intensity of the mutation, "Begin" refers to the time of the beginning of the mutation and "End" refers to the time of the ending of the mutation. "Strength" refers to the intensity of the mutation. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article).

Keywords	Strength	Begin	End	1991–2022
tannery waste-water	2.74	1991	1996	
activated sludge	2.19	1997	2005	
heavy metal	1.77	1997	2002	
wastewater treatment	2.74	2000	2008	
biological treatment	3.62	2003	2008	
tannery waste-water treatment	3.29	2003	2011	
reverse osmosis	2.2	2003	2017	
constructed wetland	2.92	2006	2014	
sludge production	2.34	2006	2011	
COD fractionation	1.94	2006	2008	
tannery industry	3.77	2009	2014	
leather industry	2.88	2009	2017	
chromium removal	1.7	2009	2014	
membrane bioreactor	2.81	2012	2014	
tannery sludge	2.53	2015	2020	
electrochemical oxidation	2.42	2015	2020	
chemical oxygen demand	1.97	2015	2020	
ceramic membrane	1.82	2015	2020	
soak liquor	1.77	2015	2017	
azo dye	1.67	2015	2017	
activated carbon	1.56	2015	2020	
hexavalent chromium	2.89	2018	2020	
water treatment	2.04	2018	2022	
chromium recovery	1.93	2018	2022	
oxidative stress	1.57	2018	2020	

3.3.2. Research Trends

The time-zone was chosen as the analysis node to generate a keyword time-series evolution graph for the TWW treatment domain on WOS. The specific metrics and thresholds were set as follows: The years per slice was 3, the threshold was chosen as g-index = 5 and the keywords with smaller nodes were hidden, resulting in 116 nodes, 137 connected lines and a density of 0.0205 (Figure 8).

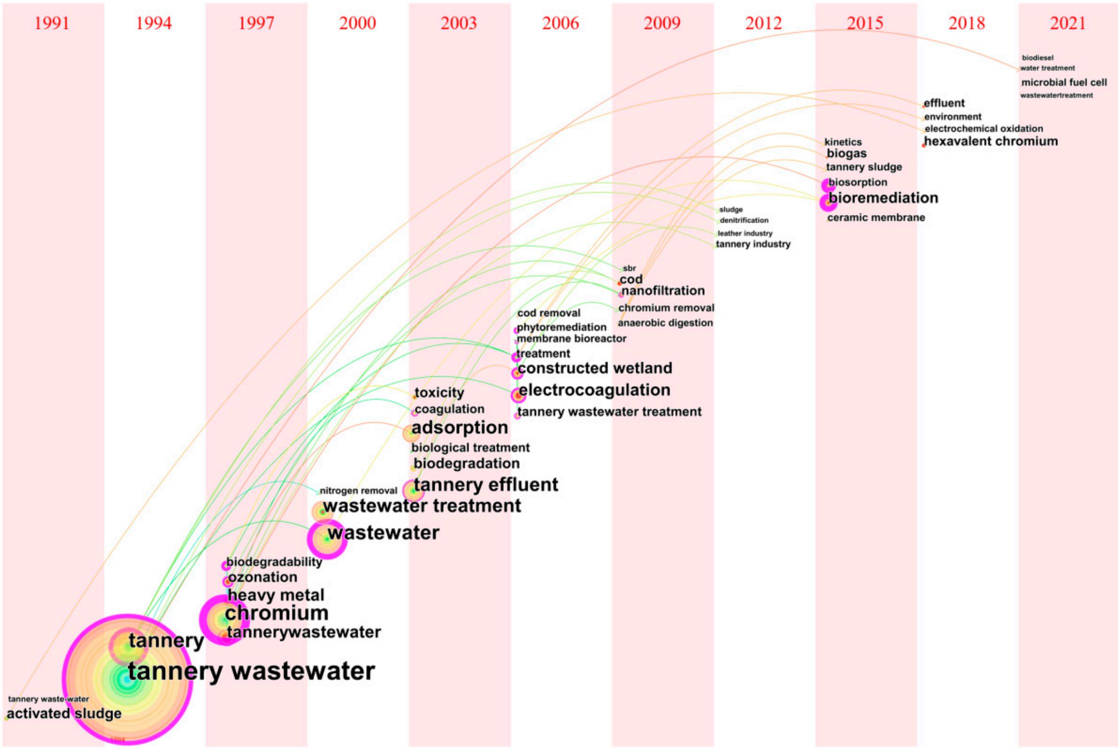

Figure 8. Time-zone view of keywords in tannery wastewater research. It shows that the keyword relationship in the field of tannery wastewater from 1991 to 2022 is quite close, whereas the highest occurrence of keywords is concentrated in the period from 1997 to 2003. Time-zone map is an evolutionary relationship of keywords. Each circle in the figure represents a keyword, and the corresponding year indicates the year that the keyword first appeared in the data analyzed.

To further elucidate the evolution of research themes over time, the timeline map of the co-citation network was plotted to elucidate the time period of development of each research theme (cluster) and the time nodes of the significant literature (Figure 8). Further analysis of these clustering labels categorized the research into three areas: (1) Research on TWW treatment methods, including “tannery wastewater”, “tannery” and “wastewater “, which mainly focuses on the treatment of TWW by biochemical methods. (2) The removal of chromium from TWW, which is reflected in the graphs of “chromium”, “ozonation” and “tannery wastewater”, which mainly focus on the removal of chromium from TWW by ozonation. (3) TWW is used in new energy applications, as reflected in the clusters of “microbial fuel cell” and “biodiesel”, which mainly focus on TWW as a reaction medium for biofuel cells and biodiesel.

In terms of time, the overall evolution can be divided into four stages: the start-up stage, the initial development phase, the rapid development phase and the stable development phase. In the initial stage (1991–1995), the main research topics were leather wastewater, activated sludge, ammonia removal, tanning and electrochemical treatment, indicating that TWW was initially treated by activated sludge, ammonia nitrogen removal, electrochemical treatment and other technologies [79–83]. In 1991, activated sludge clustering appeared on the time distribution map for the first time, and the study showed that chromium promoted the biomass of activated sludge cultivation to increase significantly [80]. Subsequently, the effects of other treatment methods on wastewater treatment were discussed. Removal and electrochemical treatments were the starting point of the research, followed by the exploration of the role of wastewater treatment. Thereafter, the research entered the initial development stages (1996–2005). The research topics in the initial development phase were tannery, tannery wastewater, chromium, ozonation, wastewater, wastewater treatment, biodegradation, toxicity, etc. This indicates that the field of TWW treatment during this period mainly started to revolve around chromium-centered approaches and technologies for wastewater treatment, and was usually attempted using biodegradation or ozone oxidation [79,84]. Over time, the research entered a rapid development phase (2006–2014). The research themes in the rapid development phase were mainly constructed wetlands, tannery wastewater treatment, electrocoagulation, treatment, COD, chromium removal and nanofiltration. This indicates that the phase concentrates on the removal of chromium through constructed wetlands and electroflocculation [85–87], and wastewater purification by nanofiltration and other means [74,88]. Constructed wetlands, as an emerging economic and environmentally friendly phytoremediation technology, is considered as an alternative to traditional biological systems. For example, plants such as Phragmites australis and Typhimus broadleaved are used to remove pollutants from TWW [89]. After 2015, the research entered a stable development phase, with research topics covering bioremediation, biogas, ceramic membranes, chromium, microbial fuel cells, effluents, etc. At the same time, research on the use of bioremediation for TWW treatment and the application of new energy sources (microbial fuel cells, biodiesel) also started during this period [54,90–93]. In addition, alkali precipitation, wet air oxidation, bioleaching, solar evaporation and ion exchange have also been reported for TWW treatment [94–98].

4. Conclusions

In this study, a scientometric approach was used to summarize the articles published in the last 30 years (1990–2022) on the progress of TWW processing research and to analyze the trend of articles in this field. The following conclusions were obtained: (1) In the temporal dimension, the research progress of TWW treatment was roughly divided into three stages: the initial research stage (1991–2003), the stable development stage (2004–2012) and the rapid development stage (2013-present). In terms of research power, India was the leader in the field of TWW treatment research. The Cent Leather Res Inst has become the core institution in the field of TWW treatment and promoted the development and progress of TWW treatment technology. Gutterres M, Sekaran G and Munz G were the top published scholars in the field of TWW treatment. The top three journals in the field of TWW treatment research in terms of the number of articles published were *Desalination and Water Treatment*, *Water Science & Technology* and *Journal of the American Leather Chemists Association*. (2) In terms of cluster analysis, the main methods such as coagulation and flocculation, adsorption, biological treatment, membrane filtration, and advanced oxidation processes were briefly discussed. Because of the rapid and efficient performance of AOPs, which can oxidize most of the recalcitrant organic pollutants, they have received more and more attention from researchers in recent years, and thus, they are introduced in more detail. (3) Through the frontier analysis, it was found that the hotspots mainly focus on the research of TWW treatment methods; the removal of chromium in TWW; and the application of TWW in new energy. The application of new energy indicates that achieving multifunctionality, recycling and intensification should be the direction of future developments of TWW treatment.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/catal12111317/s1, Table S1: Keywords information of tannery wastewater research.

Author Contributions: Conceptualization, M.L. and M.S.; methodology, X.J. and J.W. (Junhao Wu); software, Y.W. (Yang Wang) and J.W. (Jingrui Wang); writing—original draft preparation, M.L. and M.S.; writing—review and editing, H.X.; visualization, Y.W. (Ying Wang) and Y.C.; supervision, H.X.; funding acquisition, M.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Department of Science and Technology of Jilin Province, grant number 20190303092SF and 20200403032SF.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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