

## Article

# Worldwide Research Analysis on Natural Zeolites as Environmental Remediation Materials

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**Abstract:** Society faces a significant problem in regards to the spread of harmful products in the environment, primarily caused by accelerated growth and resource consumption. Consequently, there is a need for materials to be processed in less harmful ways and to remedy the contaminated sources they generate. Microporous materials have been studied for a long time and are used in waste treatment alternatives. Natural zeolites, on which this study is based, are attractive to the scientific and technological communities, due to their numerous applications as decontaminants and adsorption properties. This study analyzes the intellectual structures of publications related to natural zeolites in environmental remediation, using bibliometric methods to determine their volumes and trends. The methodology comprises of an analysis based on 1582 articles, using VOSviewer software, with data from 1974 to 2020, via the Scopus database. Results reflect a notable increase in publications from the end of the 1990s; the greatest contribution in the area comes from Eurasian countries. The study considers that development in this line of research will continue to increase and serve as a great contribution to preserve the environment in coming years, with themes that focus on water treatment (e.g., drinking water, wastewater, greywater), removal of heavy metals, ammonium, ammonia, and construction.

**Keywords:** natural zeolites; adsorbent; environmental remediation; bibliometric analysis; knowledge mapping



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## 1. Introduction

Soil and water pollution have increased in recent decades [1] due to human activities (i.e., demand and consumption to meet growing population needs). The discharge of harmful pollutants into the environment, such as mercury (Hg), lead (Pb), arsenic (As), and cobalt (Co), can (directly or indirectly) cause numerous diseases and disorders [2]. Prolonged exposure or bioaccumulation of these agents [3] threaten food sources, nature, and health [4]. Urban, industrial, and mining waste contaminate soil and water [5]; moreover, an estimated 2000 million cubic meters of water are contaminated daily [3].

In order to preserve our environment, researchers seek materials and methods that counteract adverse environmental effects. Natural zeolites were discovered more than 200 years ago [6], and are studied due to their peculiar characteristics; for example, more than 20,000 articles and 2000 patents have been published on these materials to date [7].

The discovery of abundant deposits of natural zeolites (NZ) worldwide has led to more studies being conducted on these minerals, to understand their applications and benefits.

Natural zeolites can be described as “crystalline aluminosilicates with a three-dimensional structure based on repeating units of silicon-oxygen ( $\text{SiO}_4$ ) and aluminium-oxygen ( $\text{AlO}_4$ ) tetrahedra” [8]. They are also attributed as “molecular sieves” [9–11] due to their ability to accept and reject molecules based on their size [12]. On the other hand, regarding their origins, the geological environments of formation can vary: alteration of volcanic materials in fresh, brackish, and marine waters; hydrologically closed or open systems; volcanic materials in alkaline soils, and sediments metamorphosed by hydrothermal alteration [6,8,13].

Among the characteristics that make zeolites unique materials compared to other crystalline inorganic oxides are: their tetrahedral structure, which give them different properties on the external and internal crystalline surface (high area); the exposure of the metal–oxygen tetrahedra on the internal surface makes them accessible to modifications (cation exchange, replacement of Si and Al, the addition of metal particles); the ability to form diverse zeolite compositions and structures [14]. The main characteristics of the natural zeolites described in [15] are:

- A high degree of hydration;
- A high degree of crystallinity;
- Low density and the large volume of free spaces (dehydrated);
- Shape selectivity;
- Ion exchange characteristics;
- Catalytic characteristics;
- Sorption of molecules and ions;
- Electric conductivity.

One topic of interest is heavy metal contamination in soil and water since it constitutes a significant risk to nature and society [16,17]. Despite this, methods, such as ion exchange, chemical precipitation, reverse osmosis, carbon adsorption, solvent extraction, and electrodialysis [18–20], allow the removal of heavy metals, of which ion exchange is characterized by its simplicity of application [21]. However, the use of specific treatment methods may require high investment costs. Therefore, there is an increase in the search for alternative, low-cost materials (such as natural zeolites) to remove or stabilize heavy metals [8,22–24].

The study of zeolites is vast, as previously described by the authors in [6,7]. Nevertheless, it is estimated that only “10% of natural zeolites have destined for wastewater treatment, animal feed, horticulture, odor control and other diverse applications, being 90% for the construction industry” [25,26]. The studies mainly focus on water treatment, wastewater management, soil decontamination, and nuclear waste. Zeolite applications in environmental remediation are mainly due to its ion exchange properties [27], where, depending on the type of zeolite, its pore size, the shape of its internal channels, and the type of exchange cation—it can interact with cations from the environment, with which it can have high selectivity [28–31]. In a medium where there are many cations, these can enter the internal structure of the zeolite by cation exchange, saturating it in a short time, and reducing its effectiveness.

Hundreds of investigations have been devoted toward studying water purification methods [32] and wastewater treatment (urban, industrial, mining, and agricultural). Case studies carried out on these topics include the removal of Pb (II) in wastewater, determining the affinity of mordenite with the cation [33]; as well as studies on the use of heulandite-clinoptilolite membranes in desalination and degreasing of water [34]; implementation of sorption columns filled with zeolites for wastewater treatment [35]; research on the adsorption capacity of clinoptilolite to remove pesticides from waters [36]; and heavy metals from acid mine drainage (AMD) [37].

On the other hand, other studies have been carried out, e.g., applying NZ as antibacterial agents based on their ion-exchange properties [38]. Likewise, research was conducted in the field of organic compounds, reporting the potential of NZ to stabilize

mixed pollutants in sediments, including hydrophobic organic contaminants (HOCs), such as polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers (PBDEs), and pyrethroids [39]; the adsorption of non-ionic organic contaminants (NOC), such as benzene, toluene, and phenol in wastewater [40], and even the use of NZ in the pharmaceutical industry [41].

Zeolites are also used in radioactive materials, reflecting a great interest in their applications [42,43]. One example is their use to reduce the consequences of nuclear accidents, such as Chernobyl [44]. Moreover, they have a higher selectivity to certain radionuclides, such as  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{45}\text{Ca}$ ,  $^{51}\text{Cr}$ , which, compared to organoresins, are cheaper and more resistant to nuclear degradation [6]. Similarly, NZ applications for soil remediation and protection are described [45,46].

Applying these microporous materials for the remediation of the environment is the basis of this study. Scientific knowledge has increased in this area throughout the years, but not via large-scale applications. It is necessary to understand the journals, countries, documents, associated topics, and representative experts that have contributed to the development of this area, which will serve as a guide to obtain new data and develop research. It is feasible to consider the scientific publications on natural zeolites applied to environmental remediation in order to find patterns of techniques and applications registered in the Scopus database, to determine the main trends in relationships. The aim was to analyze the publications related to natural zeolites as environmental remediation materials in the Scopus database using bibliometric methods, applying VOSviewer to determine their intellectual structures and trends.

## 2. Materials and Methods

In scientific research, literature reviews are relevant to researchers, allowing them to know a particular area of knowledge under eligible criteria, and to minimize potential errors and biases when collecting information [47]. Bibliometric analyses exhibit similar procedures, by presenting formal and rigorous structure that allow the information displayed to be of quality [48,49].

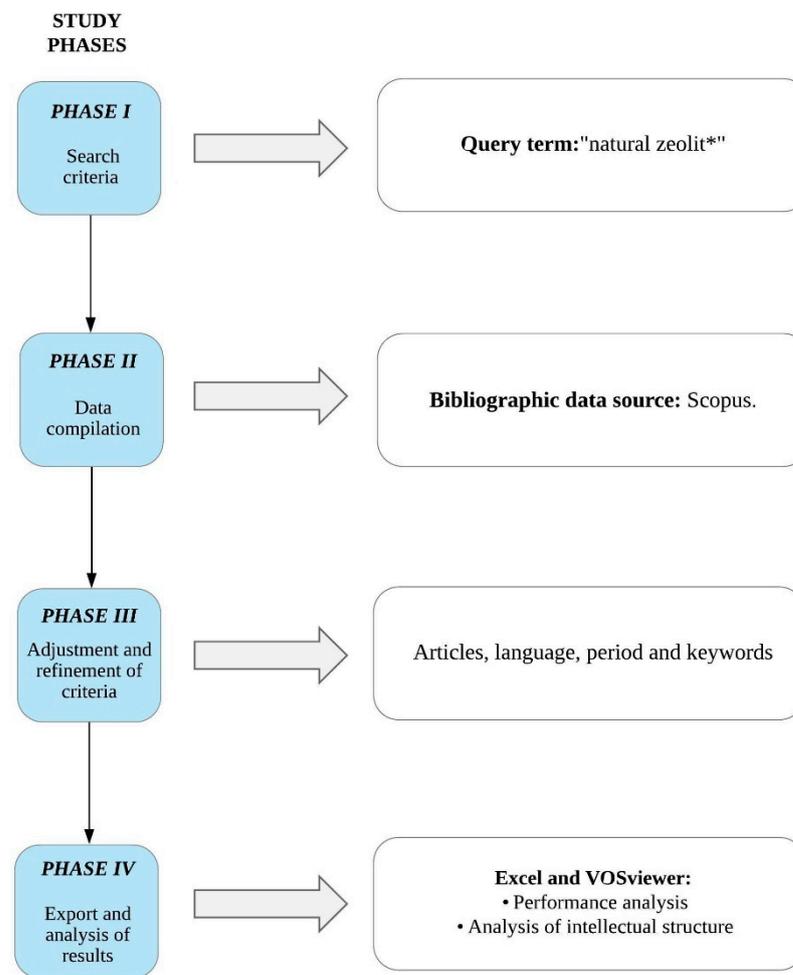
A bibliometric analysis provides researchers with tools (bibliometric methods) that guide them in their quests to collect essential information in a particular scientific field or discipline. Moreover, structure and development are recognized through thorough and precise processes [50,51]. Information can be seen through bibliometric maps, highlighting the most influential words (documents, countries, sources, authors, organizations) on a subject to carry out the comprehensive analysis [52]. This bibliometric analysis was structured in four stages, as shown in Figure 1, by collecting data to construct the information base that allowed the comprehensive evaluation of the field under study. Next, we explain the procedure.

### 2.1. Defining the Appropriate Search Terms

Some terms referred to natural zeolites in the academic literature, such as natural zeolite, natural zeolitic, and natural zeolitic material [53–55]. The common query term “natural zeolit\*” was selected, which facilitated a search for information and included the words already mentioned.

### 2.2. Initial Search Results

Bibliometric studies require the use of a database that has comprehensive coverage, with consistent and accurate data, to obtain a reliable base that provides quality information [56]. The Scopus database was selected because: (i) it is considered one of the largest databases with comprehensive coverage of documents in most academic disciplines [57,58]; (ii) it provides more excellent (in-time) coverage compared to other databases [59]; (iii) it provides excellent coverage of journals related to earth sciences [60]; and (iv) it provides tools that allow data visualization and analysis [61].



**Figure 1.** Methodological scheme for bibliometric analysis. The "\*" allows the word to be completed in both its singular and plural form.

The initial search of the data in this database was carried out in early August 2020. The search fields "article title, abstract and keyword" were applied, resulting in 3788 documents.

### 2.3. Refinement of Search Results

The initial data refinement was carried out using the information inclusion and exclusion criteria, by applying the type of document (article), language (English), and dates until 2020. The following search equation was utilized: TITLE-ABS-KEY ("natural zeolit\*") AND (LIMIT-TO (DOCTYPE, "ar")) AND (LIMIT-TO (LANGUAGE, "English")).

Additionally, by seeking a more rigorous refinement that allowed obtaining a set of data that located natural zeolites as environmental remediation materials, the research was limited itself with exact words, grouped below for the ease of the reader: (Group A), Heavy Metal\*, Metals, Metal ions, Ammonia, Ammonium, Soil Pollution, Water Pollution, Water Pollutant\*, Wastewater, Waste Water, Industrial Waste; (Group B), Gas Adsorption, Pollutant Removal, Removal, Purification, Water Purification, Water Management, Water Treatment, Wastewater Treatment, Waster Water Management, Waste Component Removal, Waste Treatment and (Group C), Controlled Study, Experiment Study, and Groundwater. The selection of these words was made based on the high selectivity of NZ towards heavy metals and ammonium [8,29,31], used mainly in the treatment, stabilization, and removal of said elements in drinking water, wastewater, and soil contamination. As a result of this initial refinement, a total of 1585 articles were obtained.

## 2.4. Data Extraction and Analysis

Search results (1585) were downloaded in CSV (comma-separated values) format, and included the information in the following fields: citation information, bibliographic information, abstract, and keywords, and as additional information—references. For data analysis, two types of software were used:

- (i) Microsoft Excel—in which the data were normalized to eliminate possible errors [62], a process in which three duplicate articles were found, and a total of 1582 articles were obtained. Additionally, the software allows analyzing scientific production in this field of study [63].
- (ii) VOSviewer—a tool that allows the construction, exploration, and visualization of two-dimensional bibliometric networks, exposing a specific field's intellectual structure [52]. Moreover, it is handy since it allows the visualization of bibliometric maps in great detail through graphical representations of straightforward interpretations [64,65]. Science maps allow observing relationships between various variables that allow information on this structure obtained at a micro-level (co-occurrence of author keywords), meso-level (co-citation of cited authors), and macro level (co-citation of cited journals) [66,67]. VOSviewer is a friendly platform used to carry out these analyses; its creators provided documentation (manual and research articles) that allowed us to understand its functionality and use [52,64,68]. This program is used to study various scientific disciplines related to geology, for example, geotourism [69], fracking [70], and geological risks [71], as well as other areas of knowledge management [72], occupational health [73], education [74], etc.

## 3. Results

### 3.1. Performance Analysis

#### 3.1.1. Scientific Production Analysis

Figure 2 presents a graph of scientific production, featuring the evolution of publications concerning the application of natural zeolites as environmental remedial materials, from 1973 to 2020, totaling 44,891 citations belonging to 1582 publications. One can observe more significant development in this area from the year 1997.

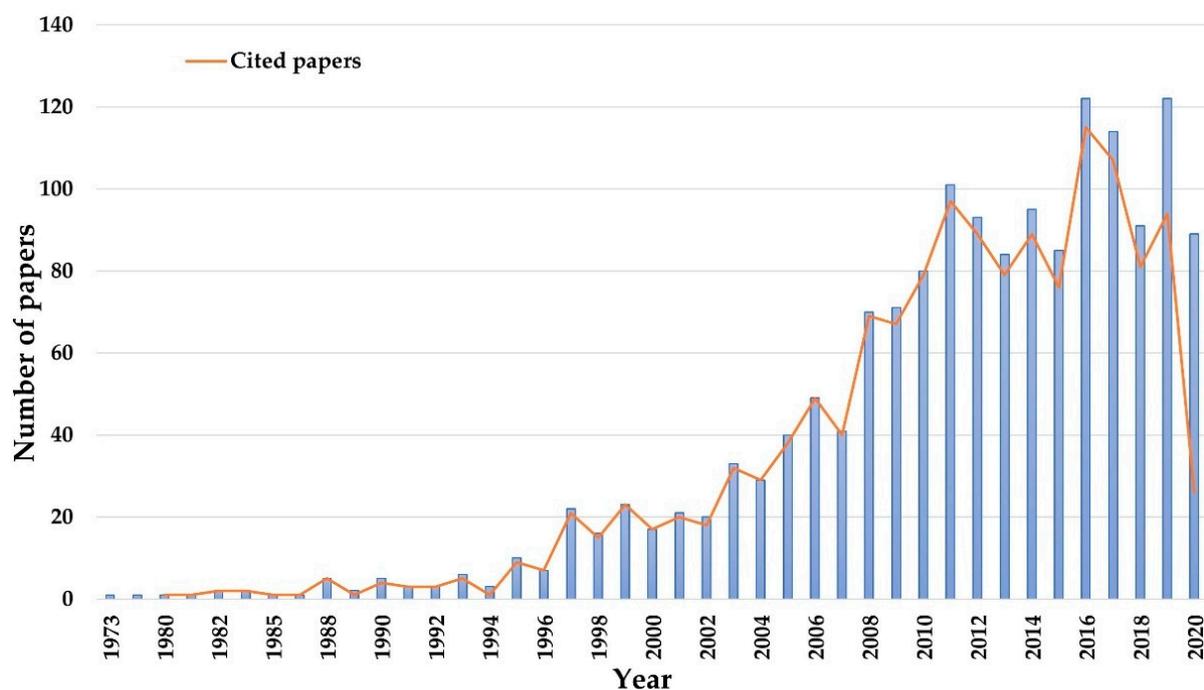


Figure 2. The growth trajectory of the literature on natural zeolites as environmental remedies, 1973–2020 (n = 1582).

Publications on natural zeolites as environmental remedies began in the 1970s, representing 0.12% of papers; only two articles added eight citations, related to radioactive cobalt sorption [75] and the treatment of wastewater using clinoptilolite [76]. In the 1980s, there was a slight increase (15 publications). For example, [21] with 883 citations, and [77] with 108 citations—the authors dealt with the usefulness of NZ as ion exchangers for the removal of ammonium ions from drinking water, and conducted a study on the influence of pretreatment on the capacity and selectivity of clinoptilolite to metal ions, respectively. In the 1990s, a notable increase was observed, with a total of 98 publications (6.15%), among them, Mumpton (1999) stands out, with the highest number of citations at the time (477); his article referred to the various uses of natural zeolites. Moreover, [78], with 285 citations, focused on removing lead and cadmium from wastewater. The most cited years were 1997 and 1999, accumulating 1378 and 1709 citations, respectively. Later, in the 2000s, there was a significant increase (24.65%), with a total of 391 papers (21,000/44,891 citations). There were 2522 citations obtained from 2007, dealing with issues such as ammonium ion removal efficiency using a natural Turkish zeolite (for environmental quality) [79]; NZ as adsorbents to remove malachite green and  $Pb^{2+}$  ions from an aqueous solution [80]; and lead retention by a Mexican tuff rich in clinoptilolite [81]. Likewise, there were 2694 citations obtained from 2009, with topics focusing on heavy metal adsorption from mining acid drains [82]; the remediation of lead-contaminated soil using NZ [83], and the adsorption of uranium (VI) on manganese oxide-coated zeolite (MOCZ) of an aqueous solution [84].

Finally, from 2010, the most significant contributions in this area are presented, covering 68.14% of articles (1076 out of 1582) with a total of 17,805/44,891 citations. The topics highlighted benefits of natural zeolites due to their adsorption properties [85], ion exchange [30], and porosity [86], which could be applied in different settings.

### 3.1.2. Country Contribution

Table 1 lists 10 out of 81 countries (using natural zeolites for environmental remediation) that have the highest number of publications and citations on environmental remediation. Major countries that top the list include Turkey, with a total of 195 publications and the highest number of citations (6940), followed by China, with 161 publications and 5618 citations, and Iran, with 155 publications and 3866 citations.

**Table 1.** Top 10 countries with the most publications in the area.

Rank	Country	Documents	Citations Count
1	Turkey	195	6940
2	China	161	5618
3	Iran	155	3866
4	United States	90	3970
5	Italy	81	1733
6	Australia	62	3056
7	Indonesia	62	384
8	Mexico	62	1621
9	Greece	60	2162
10	Spain	60	1751

Table 1 shows that most of these countries belong to Europe or Asia, followed by America and Oceania. However, variations of this list exist among the most cited countries (Table S1). Turkey and China top the list, but the United States ranks third, with 3970 citations (90 documents), Iran fourth, followed by Australia (3056 citations).

Greece moves up to sixth place, with 2162 citations, and Croatia (1758 citations) joins the top 10, followed by Spain (1751 citations), Italy (1733 citations), and Canada (1642 citations). A deeper analysis revealed that, among the countries published in the study area, the majority belong to the European continent, representing 40% of all countries, followed by the Asian continent, with 36%. Surprisingly, only Eurasia represents 76% of

publications, followed by America, which accounts for 14%; the most significant contributions correspond mainly to Canada, the United States, and Mexico. Next, Africa makes up 9% of the countries, with Egypt (17 documents and 398 citations) and South Africa (13 documents and 166 citations) as the main contributors. Finally, Oceania represents 2%; its contributions come from Australia (62 publications and 3056 citations) and New Zealand (nine publications and 478 citations).

### 3.1.3. Journal's Performance

Table 2 shows the top 10 journals published in this field of study, out of a set of 564 journals, where the number of articles (AT), contribution percentage (%), H-Index (HI), SCImago Journal Rank (SJR), and the journal quartile was considered. The journals shown have a total of 349 publications, representing 22.06% of the 1582 articles obtained, receiving 19,187 (42.74%) citations of the total citations (44,891). The first three journals on the list are: *Microporous and Mesoporous Materials*, with 69 publications in total, corresponding to 4.36% of all articles collected, with HI of 151, SJR = 1 and Q1. In second place is the *Journal of Hazardous Materials* (4.11%, 63 publications, HI: 260, SJR: 1, Q1), and in third, *Construction and Building Materials* (2.40%, 38 publications, HI: 147, SJR: 1.49, Q1).

**Table 2.** Top 10 journals with the highest number of publications.

Rank	Journal	AT <sup>1</sup>	% <sup>2</sup>	HI <sup>3</sup>	SJR <sup>4</sup>	Quartile
1	<i>Microporous and Mesoporous Materials</i>	69	4.36	151	1	Q1
2	<i>Journal of Hazardous Materials</i>	65	4.11	260	2.01	Q1
3	<i>Construction and Building Materials</i>	38	2.40	147	1.49	Q1
4	<i>Desalination and Water Treatment</i>	32	2.02	51	0.33	Q2
5	<i>Chemical Engineering Journal</i>	27	1.71	198	2.32	Q1
6	<i>Water Research</i>	26	1.64	285	2.93	Q1
7	<i>Water Science and Technology</i>	24	1.52	131	0.47	Q2
8	<i>Desalination</i>	24	1.52	169	1.81	Q1
9	<i>Bioresource Technology</i>	22	1.39	273	2.43	Q1
10	<i>Journal of Environmental Science and Health—Part A Toxic/Hazardous Substances and Environmental Engineering</i>	22	1.39	67	0.48	Q2

<sup>1</sup> AT = articles; <sup>2</sup> % = contribution percentage; <sup>3</sup> HI = H-Index; <sup>4</sup> SJR = SCImago journal rank.

### 3.1.4. Author Contribution

Table 3 presents the rank (R) of the 10 principal authors who contributed significantly to the use of NZ as valuable materials for environmental remediation. The number of publications, citations, and H-index in this area was analyzed to identify the top 10 out of 4436 authors.

Karapinar Nazan from Pamukkale Üniversitesi (Turkey) stands out for the number of citations accumulated, with 1172 citations in the area, which places him in first place on the list.

These citations belong to two publications; the first is a 2004 publication regarding the removal of heavy metal cations (1061 citations) in the *Journal of Colloid and Interface Science* [87]; and the second concerns phosphorus and ammonium removal from a 2009 publication, in the *Journal of Hazardous Materials*, with 111 citations. The first publication involved a collaboration with two authors from this list, Erdem Emin and Donat Ramazan, from the same institute, with 1061 citations each, but corresponding to the same publication.

In second place on the top 10 list is Wang Shaobin, from the University of Adelaide, Australia, with 1151 citations and the highest H-index (111). He has 12 publications on the subject, the most cited publication concerns removing basic dyes from aqueous solutions (279 citations).

The author with the highest number of publications on this list is Shekarchi Maryam, with 15 publications and 814 citations. Likewise, three authors on the list share the same publication that deals with removing heavy metals in water, with 883 citations each.

**Table 3.** Top 10 authors in NZ studies related to environmental remediation.

R <sup>1</sup>	Authors	Country	Institute/University	Citations Count	Publications	HI <sup>2</sup>
1	Karapinar N.	Turkey	Pamukkale Üniversitesi	1172	2	6
2	Wang S.	Australia	The University of Adelaide	1151	12	111
3	Erdem E.	Turkey	Pamukkale Üniversitesi	1061	1	11
4	Donat R.	Turkey	Pamukkale Üniversitesi	1061	1	12
5	Maunaye M.	France	ENSCR Ecole Nationale Supérieure de Chimie de Rennes	883	1	10
6	Martin G.	France	ENSCR Ecole Nationale Supérieure de Chimie de Rennes	883	1	17
7	Blanchard G.	France	ENSCR Ecole Nationale Supérieure de Chimie de Rennes	883	1	1
8	Shekarchi M.	Iran	Iranian Ministry of Health and Medical Education	814	15	13
9	Lin J.	Taiwan	National Chiao Tung University Taiwan	758	10	35
10	Trgo M.	Croatia	Sveučilište u Splitu	703	13	10

<sup>1</sup> R = Rank; <sup>2</sup> HI = H-Index.

### 3.1.5. Frequently Cited Documents

Table 4 shows the 10 most cited articles on the subject, which, together, add up to a total of 5022/44,891 citations. Additionally, the number of citations received for published articles (TC) and the Annual Citation Index (ACI) are included.

**Table 4.** Top 10 most cited articles.

Rank	Article Title	Authors	Type of Document	TC <sup>1</sup>	Year	ACI <sup>2</sup>
1	The removal of heavy metal cations by natural zeolites	[87]	Article	1061	2004	62.4
2	Removal of heavy metals from waters by means of natural zeolites	[21]	Article	883	1984	23.9
3	La roca magica: Uses of natural zeolites in agriculture and industry	[6]	Review	477	1999	21.7
4	Adsorption of basic dyes on granular activated carbon and natural zeolite	[88]	Article	467	2001	23.4
5	Arsenic—A review. Part II: Oxidation of arsenic and its removal in water treatment	[89]	Review	398	2003	22.1
6	Removal of zinc, copper and lead by natural zeolite—A comparison of adsorption isotherms	[90]	Article	394	2004	23.2
7	Purification of metal electroplating waste waters using zeolites	[91]	Article	370	2003	20.6
8	Adsorption characteristics of natural zeolites as solid adsorbents for phenol removal from aqueous solutions: Kinetics, mechanism, and thermodynamics studies	[85]	Article	337	2011	33.7
9	Study of equilibrium, kinetic and thermodynamic parameters about methylene blue adsorption onto natural zeolite	[92]	Article	319	2009	26.6
10	Adsorption of heavy metals from acid mine drainage by natural zeolite	[82]	Article	316	2009	26.3

<sup>1</sup> TC = the total number of citations received for published articles; <sup>2</sup> ACI = Annual Citation Index.

The three most cited documents are described below:

The first document was published in the *Journal of Colloid and Interface Science*; in this research, the authors propose using NZ as alternatives due to their low cost and availability. Furthermore, they emphasize the effectiveness of natural zeolites properties (clinoptilolite), specifically “adsorption”, to eradicate metal cations, such as Co<sup>2+</sup>, Cu<sup>2+</sup>, Zn<sup>2+</sup>, and Mn<sup>2+</sup> from wastewater.

Second, published in the journal *Water Research*, and like the previous document, it uses clinoptilolite (in drinking water) with the “ion exchange” method, through the property of selectivity of the material to eliminate ammonium ions.

Occupying third place, and published in the journal *Proceedings of the National Academy of Sciences of the United States of America*, this article compiles the essential aspects of the properties of these microporous materials in various areas (agriculture, construction, medicine, water treatment, etc.), in which they promise significant contributions.

It is necessary to consider that, some of the most cited articles do not necessarily have a more significant number of citations per year. For this reason, the Annual Citation Index (ACI) was proposed, which establishes the relationship between the total number of citations and the age of the document. Table 4 shows this index; the document of 2004 [87] has the highest index, showing an average of 62.4 annual citations. Afterward, a more recent article (2011) [85] with an ACI = 33.7, and a third, from the year 2009, with an ACI (26.6).

### 3.2. Intellectual Structure Analysis

#### 3.2.1. Author’s Keyword Co-Occurrence Network

Author keywords were used through a content analysis technique that allows the construction of semantic visual maps, showing the study area’s cognitive structure and most relevant topics [50,71,93]. For this purpose, the VOSviewer software was used to construct the bibliometric co-occurrence map, using the mentioned words; 3323 keywords were recorded, obtained from 1582 publications. Only 638 words matched at least twice, 84 words at least five times, and 42 (ten times).

Keywords are presented in Figure 3, employing nodes, which vary in size according to the number of times the keyword appears in documents. Similarly, keywords are related to each other through links, where a higher degree of relationship is presented with a greater width of the link, showing a better relationship between the words. According to node colors, keywords are grouped into 11 groups, 84 words with 698 links, and a total strength of 2448.

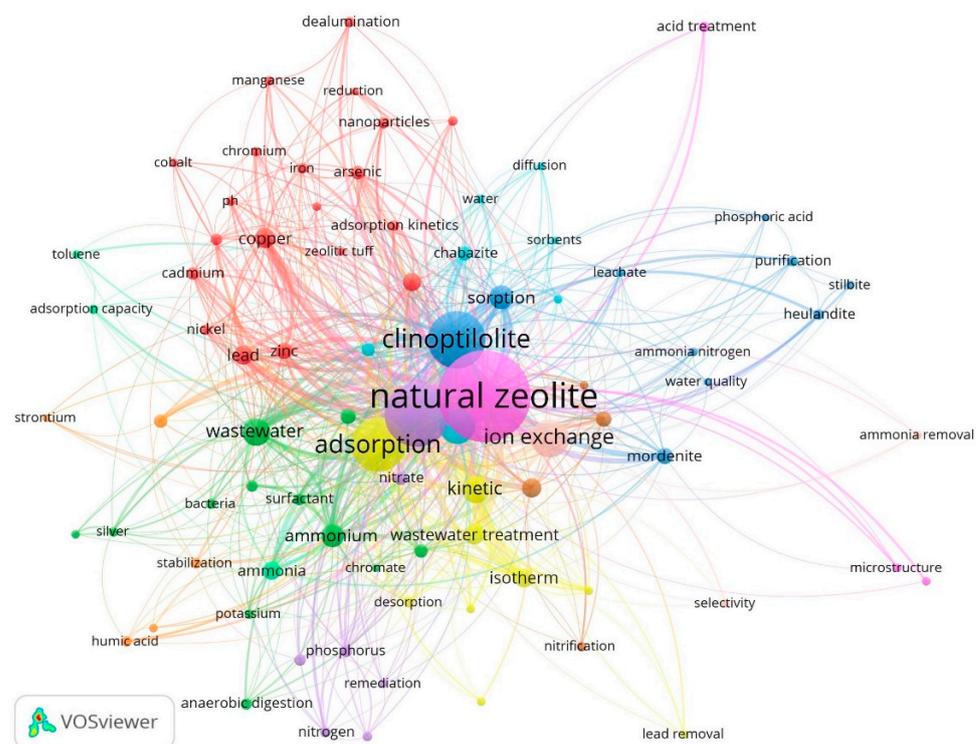


Figure 3. Co-occurrence bibliometric map of author keywords.

The author keyword network analysis was based on the number of co-occurrences represented in the nodes (circles) of colors, starting from those with the most significant numbers to those with the least numbers. The description of each cluster was made based on the main nodes shown in Figure 3 and Table S2. In addition, the 20 most used keywords based on the number of co-occurrences are in Table 5. The period comprises the years 1973–2020, divided into four sub-periods that correspond to decades to facilitate the analysis. The Table 5 initially shows seven terms that comprise this structure’s origin, consolidated into 20 main topics over the years. The topics (terms) are highlighted: natural zeolite (667 articles), zeolite (329), and clinoptilolite (259).

**Table 5.** Top 20 keywords (1973–2020).

Keyword	Occurrences	1973–1989	1990–1999	2000–2009	2010–2020
natural zeolite	667	-	39	159	469
zeolite	329	1	11	84	233
clinoptilolite	259	3	10	97	149
adsorption	237	-	2	52	183
ion exchange	119	3	9	53	54
heavy metal	99	-	3	31	65
wastewater	61	1	4	18	38
kinetic	60	-	-	15	45
sorption	48	-	2	15	31
ammonium	43	-	2	10	31
copper	34	-	1	14	19
wastewater treatment	32	-	1	8	23
isotherm	31	-	-	7	24
water treatment	31	-	1	11	19
lead	30	-	3	12	15
adsorption isotherm	28	-	1	4	23
ammonia	26	1	2	10	13
mordenite	22	1	4	6	11
zinc	22	-	-	11	11
ammonium removal	20	1	2	4	13

Cluster 9 (pink), called “Acid Water (AW) treatment with NZ” (see Table S1), is composed of four nodes with 690 co-occurrences, and studies the capacity of natural zeolites in the treatment of acidic waters [94], taking advantage of the adsorption properties [95], ion exchange [96] to facilitate the removal of heavy metals, and reduce pollution damage due to acid mine waters [97]. This cluster shows the main keywords, “natural zeolite”, “acid treatment”, and “microstructure”. The first of the terms mentioned, “natural zeolite”, is the nucleus of this structure, with 667 co-occurrences.

Cluster 4 (yellow), “Kinetic of adsorption”, consists of nine nodes with 393 co-occurrences, where simulations [98] and various models [99] are applied in kinetic studies [100] to study the adsorption characteristics of NZ to various elements [85]. This cluster presents some keywords that stand out for their occurrence, such as “adsorption”, “kinetic”, and “isotherm”.

Cluster 5 (purple), “NZ soil remediation” has seven nodes (392 co-occurrences), studies that test the effectiveness of mineral amendments with natural zeolites [101], to remediate soils with potentially toxic elements (PTE) [102] extracting or immobilizing metals [103], in order to limit environmental impacts and protect the quality of soils [104]. It has keywords like “zeolite”, “remediation”, and “nitrogen”.

Cluster 3 (blue), “NZ water purification”, is formed by 10 nodes with 379 co-occurrences. Various topics on water purification are dealt with, presenting methods such as desalination and de-oiling of water with natural zeolite membranes [34], treatments for nitrate removal [105], treatments are evaluated in waters contaminated with endocrine disruptors [106] and even the purification of groundwater [107], with keywords “purification”, “water quality”, and “clinoptilolite”.

Cluster 1 (red) called “Metal removal” with 20 nodes and 258 co-occurrences, with topics that study the equilibrium [108] and adsorption forces [109], the kinetics of removal [110] and affinity towards heavy metal ions [111], has keywords such as “copper”, “lead”, and “zinc”.

Cluster 2 (green), “Urban WasteWater (WW) treatment”, conformed by 12 nodes with 209 co-occurrences, where characteristics of the NZ are taken advantage of, such as their low cost, chemical composition, and properties [112] in recovery processes of heavy metals [113] and other substances in municipal wastewater treatment plants [114], even considering these waters as new sources for other activities [115], with keywords “wastewater”, “removal”, and “ammonium”.

Cluster 6 (light blue) “NZ HM adsorption” has seven nodes (154 co-occurrences); they evaluate the adsorption characteristics of zeolites under various parameters [98] and physical–mechanical properties [116] to solve problems, such as water contaminated with lead and cadmium [117], making use of preferential adsorption for HM classification [109], with keywords “heavy metal”, “chabazite”, and “phillipsite”.

Cluster 10 (light pink), entitled “Ammonia selectivity by Ion Exchange (IE)” (three nodes, 132 co-occurrences), studies the ion exchange process in NZ [118] for the suppression of ammonia in surface waters [119], and comparisons with other processes [120], with keywords “ion exchange”, “selectivity”, “ammonia removal”.

Other small clusters are cluster 8 (brown) “NZ use in Drinking Water (DW)”, four nodes with 66 co-occurrences, which show studies concerning the efficiency of the removal of  $\text{NH}_4^+$  [121] and other pollutants in surface waters, taking advantage of the adsorption capacity of these minerals in water for human consumption [121], with keywords “water treatment”, “drinking water”, and “ammonium removal”. This is followed by cluster 7 (orange) “NZ use in Soil Degradation (SD)” with five nodes and 45 co-occurrences. It is applied as a soil amendment method with natural zeolites to reduce and control the dynamics of nitrate and ammoniacal nitrogen [122], reducing the losses of gaseous N and favoring crop yield [123], promoting mitigation for, especially, agricultural soils [124], with keywords “degradation”, “cation exchange”, and “stabilization”. Finally, in cluster 11 (turquoise), “Ammonia Adsorption Capacity (AC) by NZ” (three nodes, 40 co-occurrences), based on studies to determine the adsorption capacity of ammonia using natural zeolites [125], which project great potential and efficacy as microporous materials [126], with keywords “ammonia” and “adsorption capacity”.

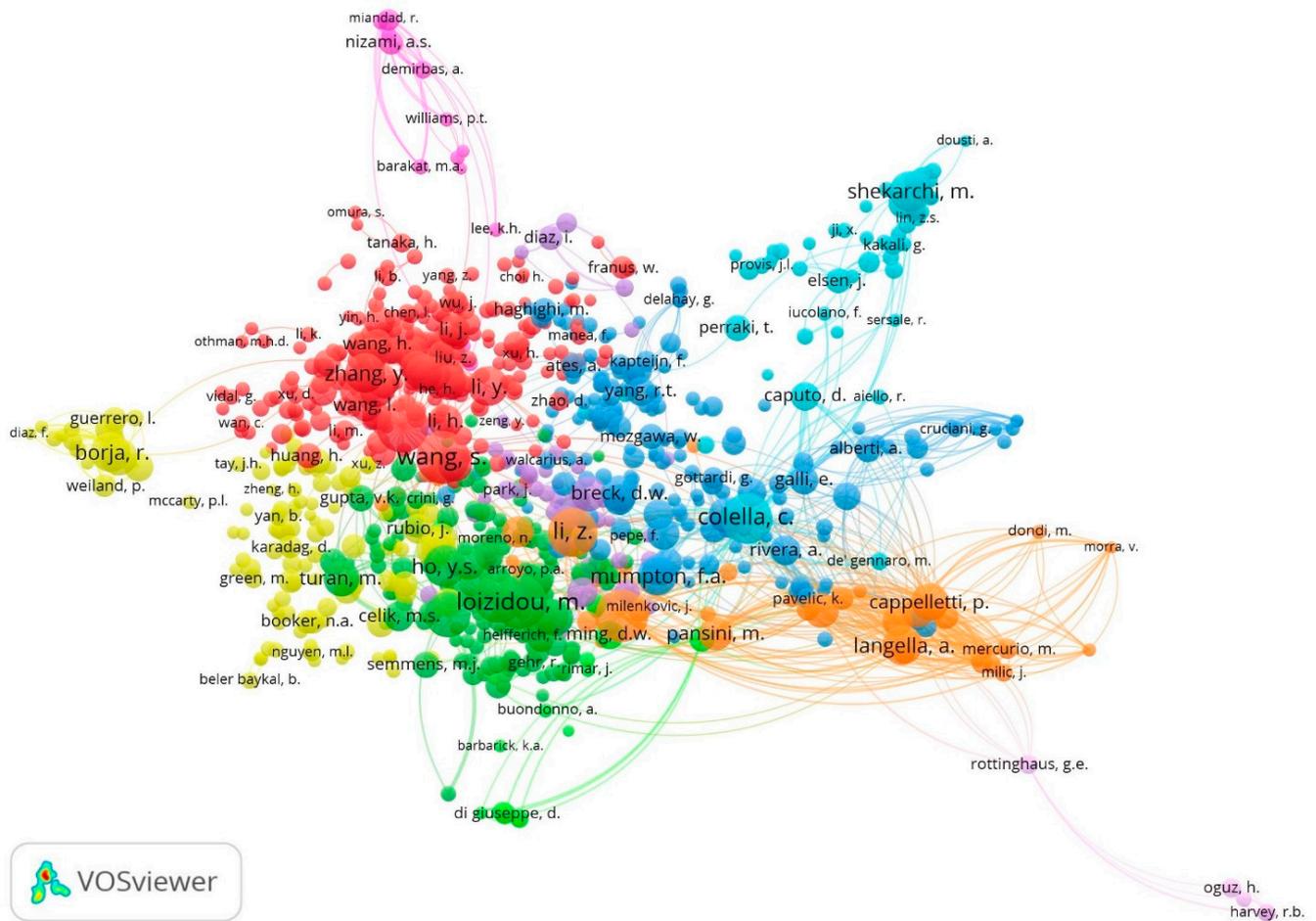
### 3.2.2. Cited Authors Co-Citation Network

VOSviewer software was used to construct the cited authors’ co-citation map, shown in Figure 4. The intellectual structure of interest was studied (through the authors of the reference works) [69,127]; 947 authors had at least 20 citations per author, appearing in 10 clusters of different colors, with a total force of 928,935.

The present analysis was carried out based on the number of cited author co-citations in descending order. A representative name was established for each cluster based on the similarity of the NZ topics, among the principal authors with the highest number of citations, obtaining groups that linked scientific fields, summarized in Table S3.

The following describes a clusters group with more than 5000 citations. Cluster 1 (red) is named “Ion removal in aqueous media”, with 262 authors and 11,447 citations. It studies the effectiveness of natural zeolites in the removal of ions for different purposes, such as wastewater, and removal of basic dyes, among others; the leading researchers most cited are Wang, S., Wang, Y. and Li, Y. Cluster 2 (green) “Ion exchange of clinoptilolite” with 183 authors and 8402 citations. It consists of investigations based on clinoptilolite zeolite’s beneficial properties; the authors include Loizidou, M., Inglezakis, V.J., and Ho, Y.S. Cluster 3 (blue) “Properties and applications of the NZ”, with 177 authors and 7015 citations. The studies explore the variety of beneficial applications for different areas, including the environment, featuring prominent authors, such as Mumpton, F.A., Breck, D.W., and Misaelides, P. Cluster 4 (yellow) “NZ livestock applications”, with 125 authors

and 5289 citations. The NZ used for piggery waste treatment; authors include Borja, R., Sanchez, E., and Buszewski B.



**Figure 4.** Co-citation bibliometric map of cited authors.

Other clusters with less than 5000 citations: Cluster 7 (orange) “Sorption kinetics of the NZ”, with 44 authors and 2835 citations. This cluster uses the sorption property of NZ in nonpolar organic compounds, inorganic cations, and inorganic oxyanions. Authors include Bowman, R.S., Li, Z., and Langella, A. Cluster 6 (light blue), with 58 authors and 2525 citations, is based on natural zeolite sustainable applications in the construction area, defined as “NZ applications in construction”. The authors include Colella, C., Shekarchi, M., and Ahmadi, B.

Cluster 5 (purple) “Sorption processes with NZ”, with 59 authors and 2167 citations. The cluster focuses on studies concerning zeolite-rich tuffs for sorption processes; authors include Olguin, M.T., Hrenovic, J., Solache-Rios, M. Cluster 8 (green) “Agricultural uses of the NZ”, with 19 nodes and 748 citations. In this cluster, the studies focus on soil remediation, authors include Ming, D.W., Passaglia, E., Rehakova, M. Cluster 9 (pink) “NZ applied to energy recovery”, with 14 nodes and 463 citations. In this cluster, the studies look for economic and viable alternatives, such as NZ, to develop technologies applied to the energy area. Authors include Nizami, A.S., Rehan, M., and Barakat, M.A. Finally, Cluster 10 (light pink), with 6 authors and 222 citations, deals with topics such as the adsorption of organic compounds with NZ, represented by authors such as Rottinghaus, G.E., Oguz, H., and Harvey, R.B.



(purple) presents 15 nodes and 2274 citations, where the journal with the highest number of co-citations is *Applied Clay Science* (603 co-citations; H-index: 119), followed by *Construction and Building Materials*. Lastly, cluster 4 (yellow), called “Energy and waste management”, is made up of 22 nodes and 1437 citations, with representative journals such as *Industrial & Engineering Chemistry Research* (H-index 211; 294 co-citations) and *Waste Management* (H-index 145; 198 co-citations).

#### 4. Discussion

An analysis of future trends in this line of research reflected that the studies of most significant interest, in the period 2000–2010, were those referring to the removal of heavy metal cations [87]; dye adsorption [92]; wastewater purification [91]; and ammonium and ammonia removal [79,130]. Likewise, in the 2010–2020 period, there was interest in removing ammonium from water, heavy metals, and purification of aqueous solutions, with natural or modified zeolites. Another topic of industrial interest was the improvement of construction materials, mainly for pozzolanic cement and mortars. Further developments in this study are expected in coming years.

The more significant numbers of scientific contributions from certain countries in this area, at the beginning, could have been attributed, in part, to discovering new potential commercial deposits of NZ (in the late 1950s). Moreover, since several countries in Europe, Asia, and North America were the leading suppliers in the NZ market, according to Roskill (a British firm specializing in market studies), in 1988, promoted research with natural zeolites in different areas [7]. However, currently, approximately 30 countries are leading producers, worldwide, of natural zeolites, including: China, Jordan, the Republic of Korea, Turkey, Japan, Slovakia, the United States, and Cuba [13]. China tops the list as the primary producer; however, it ranks second among the countries with the most publications and citations (see Table 1). Moreover, it has a close relationship with countries, such as Turkey, Iran, Australia, and the United States.

The analysis of author contributions reflected that the most cited authors did not necessarily have a large amount of research in this area. Most of the top 10 presented in Table 3 vary, with a range of 1 to 15 publications. The interesting thing about this is reflected in the number of citations that a single investigation can have, reflecting the scientific community’s interest in specific topics. Surprisingly, China does not appear, being the most cited authors are from Turkey, Australia, and France.

At first glance, the most cited topics were those related to eliminating heavy metals in water for human consumption and wastewater [21,87]. Moreover, it is not surprising, given the accumulation of heavy metals in water and soils that seriously threaten health, food sources, and the environment, based on the need to apply practical, affordable, and, if possible, low-cost treatments [6].

Another aspect considered in Table 4 is TC, which specifies the number of total citations per article, and which differs from the ACI index (that calculates the number of citations of the article for its years of seniority). Some articles do not have a considerable TC due to the few years of publication, which puts them at a disadvantage compared to those with a higher TC. Table 4 shows that some articles that have a low TC do not necessarily have a lower ACI, as is the case with Yousef et al. (2011), with a TC = 337 (occupies the eighth place in the table), becoming the second most cited article per year, with an ACI of 33.7. On the other hand, with more in-depth analysis, it can be seen (in Table 2) that four thematic groups make up the top 10 journals: *Microporous Materials*, *Water Research*, *Treatments*, and *Technologies*. *Microporous and Mesoporous Materials*, the *Journal of Hazardous Materials*, and *Construction and Building Materials* are highlighted. Despite being the most cited journals, some of the principal authors with the highest number of citations were not published in such journals. Only Lin J. has publications in *Microporous and Mesoporous Materials*; in the *Journal of Hazardous Materials* (Karapinar N., Erdem E., Donat R., Wang S., and Lin J.), and *Construction and Building Materials* (Shekarchi). Publications by the top 10 authors (Table 3) were also featured in other significant journals, such as *Bioresource*

*Technology and Water Research*. However, curiously, only four of the most cited documents were published in one of the top 10 journals, the journal *Water Research*, where the document with the highest number of citations belongs to the *Journal of Colloid and Interface Science*.

The analyses of the intellectual structures of the bibliometric maps showed the networks and relationships present. Keyword co-occurrence mapping (see Figure 3) reveals that the term “natural zeolite” is the center of the structure, with 667 occurrences, followed by the term “zeolite” with 329 occurrences (Table 5), which have a stronger relationship with other terms, such as clinoptilolite, adsorption, ion exchange, and heavy metal. These agree, since the studies are based on properties, such as adsorption and ion exchange. The appearance of the term “clinoptilolite” among the principal relationships suggests that it is one of the most studied types of natural zeolites since it is considered one of the most abundant in nature [131], in addition to its properties, which make it stand out among other natural zeolites. Likewise, there are similar relationships between the cluster groups presented. Clusters 5 and 7 deal with soil care issues; clusters 1 and 6—thematic on heavy metals, and clusters 2, 3, and 8 focus on aqueous fluids. These close relationships between clusters give rise to topics focused on using natural zeolite properties to eliminate, retain, or reduce different pollutants.

After the previous analysis, in the author co-citation mapping (see Figure 4), the relationships between all clusters presented an intellectual structure focused on processes and applications in a great variety of areas, including energy. It was determined that the principal authors of this network with the highest number of co-citations are: Loizidou, M. from cluster 2 with 346 co-citations, with more robust links with author Inglezakis V. J.; Wang S. (cluster 1) with 339 co-citations—closely related to author Wang Y.; cluster 7 lead author Bowman R. S. (335 co-citations), showing the most robust relationship with Li Z.

Finally, the bibliometric mapping of journal co-citation (see Figure 5) exposed the thematic areas in which NZ research applied toward preserving the environment. The prominent journals with the highest number of co-citations are the *Journal of Hazardous Materials* (2367 co-citations, cluster 6), *Water Research* (1792 co-citations, cluster 2), and *Microporous and Mesoporous Materials* (1444 co-citations, cluster 1). The two leading journals have a strong relationship; moreover, they have the *Chemical Engineering Journal* in common, which is in the fifth position of the most cited (see Table 2). The journal *Microporous and Mesoporous Materials* has strong relationships with the *Journal of Hazardous Materials*, *Water Research*, and the *Chemical Engineering Journal*.

## 5. Conclusions

This article summarizes the investigations that applied natural zeolites as environmental remediation materials from 1973 to 2020.

Natural zeolites contain impurities, unlike synthetic zeolites, which are pure, and make NZ less efficient, but that, due to the cation exchange capacity, can be modified (i.e., by cation exchange—by adding certain cations to increase their effectiveness in certain specific uses). For example, they can be made highly bactericidal, i.e., by injecting the zeolite with  $\text{Ag}^+$  cations through a prepared  $\text{AgNO}_3$  solution.

The results observed in the scientific production indicate a latent interest on the part of the literature, which is consistent, given the need for advances in the development of materials and methods that serve as environmental remedies in a world experiencing serious pollution problems. Moreover, a bibliometric analysis was carried out to understand the intellectual structure of this area, by evaluating scientific production, geographical distribution, and the contributions of frequently cited journals, authors, and documents. A total of 1582 publications, 81 countries, 564 journals, and 4436 authors were analyzed, reliably showing that natural zeolites have a wide range of application for protection, remediation, and environmental control issues.

Research in this study area has enhanced since the year 1997. The authors of these publications are mainly in Europe or Asia, followed by America and Oceania. The Eurasian

continent provided the greatest scientific contributions in this area, with 76% of the publications. Countries such as Turkey, China, and Iran topped the list.

The authors with the highest number of citations in the area are Karapinar N. and Erdem E. from the Pamukkale Üniversitesi institute (Turkey), and Wang S., the University of Adelaide in Australia. According to the number of publications, the journals that lead these contributions in the study area are *Microporous and Mesoporous Materials*, *Construction and Building Materials*, and the *Journal of Hazardous Materials*. The main processes investigated and linked to these journals include important research of NZ used as nanomaterials.

The cognitive structure, visualized by the bibliometric analysis, is presented in the co-occurrence maps of author keywords and the co-citation of authors and journals, revealing the most relevant topics of the study area, focusing on the use, processes, and applications of natural zeolites to preserve the environment. We consider that, presenting a study of natural zeolites with a bibliometric approach, contributes to the scientific community by analyzing the intellectual structure, allowing us to extract experiences from researchers in this field of study. Experiences are transmitted concisely and briefly about countries, institutions, and authors involved, as well as the topics and research themes. This study is a contribution to the young researchers in this area of study.

However, the research has certain limitations: (i) it considers only one type of document (articles); (ii) the preference for only one language (English); (iii) it does not include several databases, it uses only one (Scopus); and (iv) it applies a single approach when examining a particular field of study.

Finally, according to the analyses carried out, further developments in this line of research will continue to grow, i.e., in regards to water treatment (drinking water, wastewater, and greywater), removal of heavy metals, ammonium, ammonia, and construction.

**Supplementary Materials:** The following are available online at <https://www.mdpi.com/article/10.3390/su13116378/s1>, Table S1: Top 10 most cited countries in the study field, Table S2: Detail of the Keywords with the highest Co-occurrence, Table S3: Detail of the authors with the highest Co-citation, Table S4: Detail of the journals with the highest number of Citation.

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## Abbreviations

The following abbreviations were used in this manuscript:

AC	Adsorption Capacity
ACI	Annual Citation Index
AT	Number of Articles
AMD	Acid Mine Drainage
AW	Acid Water
CSV	Comma-Separated Values
DW	Drinking Water
HI	H-Index
HM	Heavy Metals
HOCs	Hydrophobic Organic Pollutants
IE	Ion Exchange
MOCZ	Manganese oxide-coated zeolite
NOC	Non-ionic Organic Pollutants
NZ	Natural Zeolites
PAHs	Polycyclic Aromatic Hydrocarbons
PBDEs	Polybrominated Diphenyl Ethers
PTE	Potentially toxic elements
R	Rank
SD	Soil Degradation
SJR	SCImago Journal Rank
TC	Total number of citations
WW	WasteWater

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