Koc. J. Sci. Eng., 5(1): (2022) 42-53 https://doi.org/10.34088/kojose.955299

Kocaeli University

Kocaeli Journal of Science and Engineering

http://dergipark.org.tr/kojose

Arsenic Removal Technologies: Mapping Global Research Activities (1970-2019)

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Article Info

Research paper

Received : *June 21, 2021*
Accepted : *November 21, 2021*

Keywords

Arsenic
Bibliometric
Contamination
Water
Removal
Research Trends

Abstract

Arsenic contamination in drinking water poses worldwide threat to public health and requires emergency actions in some parts of the world. Several technologies have been used to overcome arsenic contamination issues and to meet the arsenic concentration limitations for public health. In this study, research tendencies on arsenic removal technologies were evaluated. A total of 4083 publications, published between 1970 and 2019, on arsenic removal from drinking water, groundwater and wastewater were retrieved from Web of Science (WoS) database. A bibliometric analysis was carried out and word frequency along with visualization map analysis were used to provide a quantitative analysis, and an overview on the current research trends and research prospects. The results showed that annual output of the "arsenic removal" subject increased significantly after the year 2000. "Article" was the most preferable publication type, and "Journal of Hazardous Materials" had the highest publication number. The most productive country in terms of number of total articles on arsenic removal was China. Also, the South-East Asian countries highly contributed to the literature. "Adsorption" was found to be the most frequently researched arsenic removal technology and nanotechnology plays a significant role in the adsorption development.

1. Introduction

Arsenic (As) is a prevalent element which can be found in air, soil, water, and living organisms. It is the 18th most common element in the universe, the 20th in the earth's crust, the 14th in the seawater, and the 12th in the human body [1]. This abundance is mostly caused by soil erosion, mineral ore formation, volcanic, geothermal, and hydrothermal activity [2]. It has several oxidation states (-III, 0, +III, +V), among those forms +III and +V states are mostly found in natural systems [3] as a part of dissolved oxyanions such as, arsenite (AsO_3^{-3}) and arsenate (AsO_4^{-3}) [4]. Inorganic As compounds are classified as "Group 1" carcinogens by International Agency for Research on Cancer [5] and recognized as toxic by many authorities such as World Health Organization (WHO) and United States Environmental Protection Agency (USEPA). Toxicity of As depends on chemical forms and oxidation states, and is due to reversible reactions with thiol groups and phosphorus substitution [6]. In addition, physical state, particle size, absorption rate and chemical structure of arsenic compounds have important role on toxicity as well [7]. Inorganic arsenic is highly associated with skin, lung, urinary system, liver cancers, and other serious cancer types [8]. 200 million people are exposed to As concentration of higher than 50 µg L^{-1} globally,[9]. In South and South-East Asia, 100 million people face risk of arsenic contamination. As contamination is critical in countries like Bangladesh [10], Chile [11], Taiwan [12], Vietnam [13], Argentina [14] due to geological characteristics. Arsenic crisis has been affecting millions

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of people in nearly 70 countries and causing adverse health effects [15]. On the ground of high toxicity and serious health effects induced by As, WHO lowered As limit in drinking water to 10 μg L^{-1} in 1993 [16]. European Union (EU) passed council directive that accepted the same limit for bottled water in 1998 [17]. USEPA accepted the same limit for drinking water in 2001 [18]. There are numerous As removal technologies, such as oxidation, coagulation, ion exchange, membrane filtration and adsorption technologies [19,20]. Researchers have often used nanomaterials for As removal as nanomaterials provide exceptionally high surface areas, catalytic activity and versatility via surface-functionalization [21,22]. However, residual contamination levels are higher than the water quality standard [19]. Coagulation process uses positively charged coagulants to stabilize negatively charged suspended solids. Usually, addition of metal salts, such as alum salt, starts the flocculation process and can be removed from the bottom of the tank. Oxidation processes uses different oxidants (chloride and ozone) to oxidize and remove pollutants from water. Ion exchange is a physical-chemical process that exchanges ions within solid resin. There are two types of resins: anion and cation. Membrane filtration is a barrier between two phases. These phases could be layers, film, or different structures. Usually, arsenic removal technologies are successful at removing highly contaminated water to certain levels.

Bibliometric analysis uses scientific data within science by quantitative analysis [23]. Basically, it is a methodology that conducts quantitative analysis and statistics to illustrate patterns, global trends, and research productivity of certain constituents, such as key word, topic or title. In this study, bibliometric analysis was used to evaluate literature in Web of Science (WoS) database from 1970 to 2019 regarding As removal from drinking water, groundwater and wastewater. This study aims to describe the publication pattern, to evaluate national and organizational contributions and to provide insights regarding most relevant technologies and future trends on As removal.

2. Materials and Methods

A bibliometric analysis was carried out using the Web of Science (WoS) database. The topic search that traces related information in the abstract, keywords and title was utilized to obtain publications from WoS database. Different combinations of words related to arsenic removal was used to retrieve publications. The key terms and the combinations of key terms are arsenic + removal + ("drinking water"/ "waste water"/ "ground water"/ groundwater / wastewater). "arsenic" and "removal" were always used in the search and one of the given keywords were added to retrieve publications. Different combinations of key terms used in the study are given in Table 1.

Table 1. Different combination of key terms

Keywords	Number of Publications
Arsenic + removal + "drinking water"	2144
Arsenic + removal + "waste water"	463
Arsenic + removal + "ground water"	100
Arsenic + removal + groundwater	1011
Arsenic + removal + wastewater	365
Total	**4083**

As, water, groundwater, drinking water, wastewater and removal were not included in the frequency analysis as they were used in the database search. Different forms of words such as As (V), arsenic (V), As-V and arsenate; As (III), arsenic (III), As-III and arsenite; iron, ZVI, nZVI, zerovalent, zero-valent; nanoparticle, nanoparticles; membrane and membranes, ion exchange, ionexchange and ion-exchange were merged into one category in the frequency analysis. Publications from 1970 to 2019 were downloaded from WoS online database. Data retrieved from WoS database were pre-processed, each publication was evaluated, and duplications were removed. A total of 4739 publications were reduced to 4083 after removing duplications. Reference type of publications, distribution of major journals, contribution of countries were analyzed and visualization map analysis was conducted. Contribution of countries and institutions were identified by the country of the affiliation of at least one author of the publication [24]. Bibexcel that is freely available versatile bibliometric toolbox [25] was utilized for frequency analysis in keywords, titles, and abstracts. Network diagram of cooperation between countries was visualized by VOSviewer [26]. Retrieved publications were also evaluated in three separate terms for determination of research tendencies, hotspots and prospects on As removal studies

3. Results and Discussion

3.1. Publication Outputs

The overview of As removal research retrieved from WoS database is given in Fig. 1. A total of 4083 publications related to As removal from various water resources including wastewater were found in the WoS database. The number of publications demonstrate a growing trend since 1990s, although there are some exceptions such as the year 2006 and 2015. The first publication was in 1973 and there were only 3 publications in 1993. A total of 85 publications were obtained in WoS database in the 1970-1999 period while 928 publications were obtained in the 2000-2009 period, and 3070 publications were obtained in the 2010-2019 period. The most productive year was 2019 with 449 publications. Time periods were chosen according to publication numbers and legislations regarding As limitations. Growing trend since 1990s is consistent with As legislations introduced by WHO in 1993, EU in 1998, and USEPA in 2001. The number of publications on arsenic removal has increased significantly after USEPA's legislation in 2001. This correlation might suggest USEPA's drinking standard had the highest impact on the science community. However, synergistic effect of EU and WHO should be considered. In addition, advancing technologies that enable researchers to detect arsenic more accurately and social impacts of arsenic poisonings cannot be overlooked. Publication types were analyzed, and various types were found in the total of 4083 publications. Type of a publication is non-exclusive hence any given publication can be classified in more than one category. Thus, the number of publications in a category is more than total publication number. Article was the most favorable way of contribution to WoS, followed by proceedings papers and reviews. The most favorable research areas are Engineering, Environmental Science/Ecology and Chemistry, which are compatible with literature [15,27].

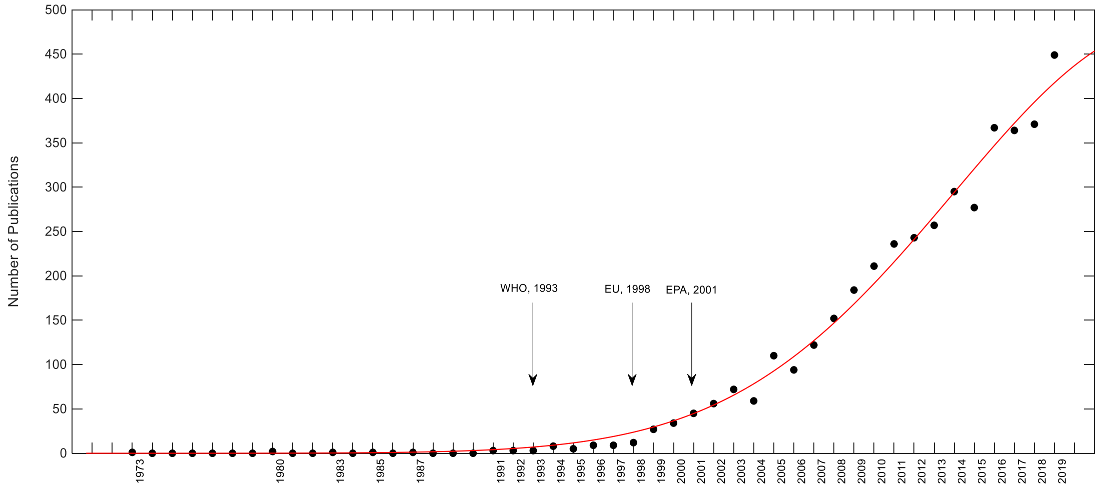

Figure 1. Worldwide overall publications on As removal

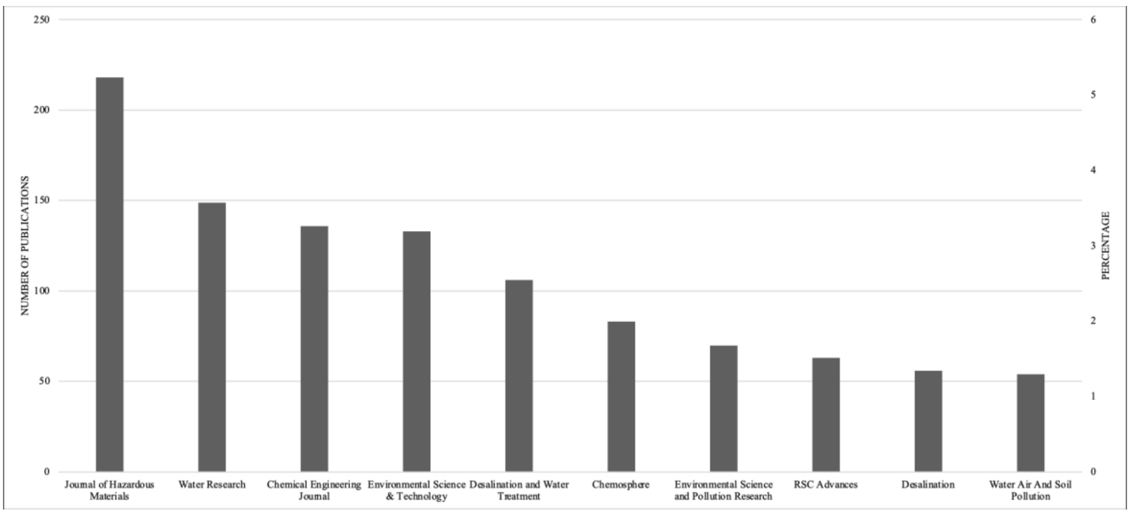

Figure 2. The most productive journals

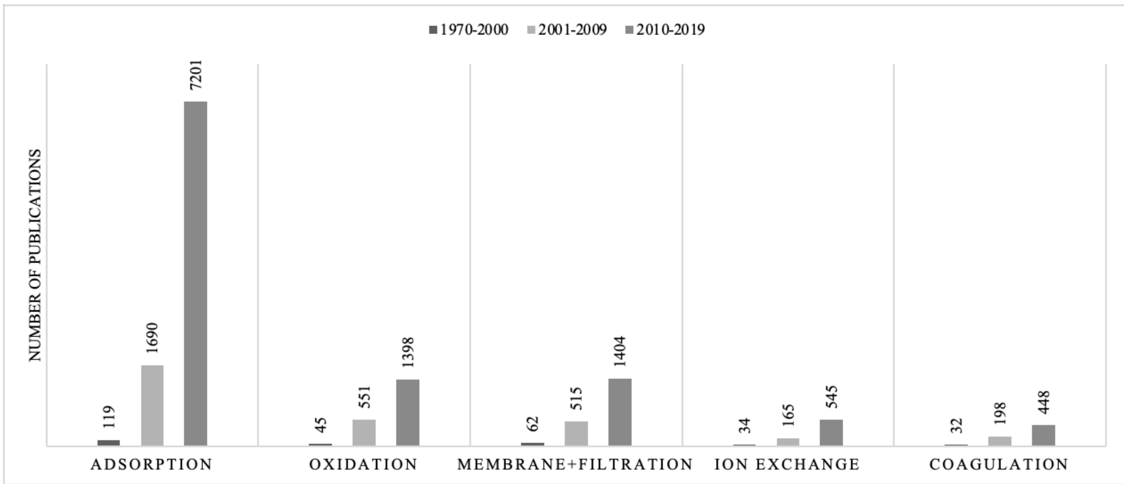

Figure 3. Evolution of As removal technologies since 1970

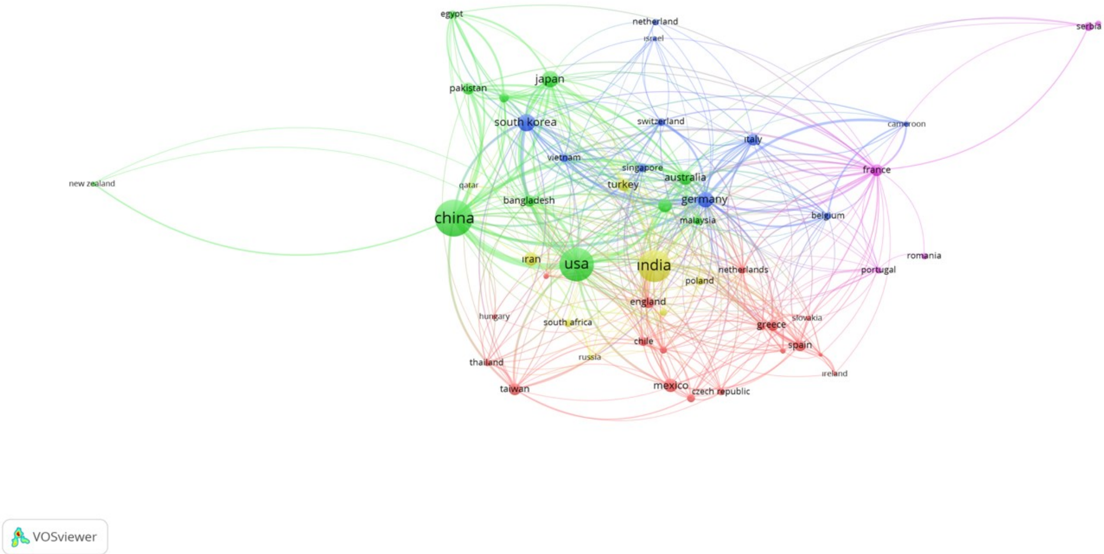

Figure 4. Visualization map analysis of collaboration of countries; minimum number of occurrences of 10

Table 2. The top 10 most cited articles on arsenic removal

Ranking	Articles	Times Cited
1	Arsenic removal from water/wastewater using adsorbents – A critical review Author(s): Mohan, Dinesh; Pittman, Charles U., Jr. Source: Journal of Hazardous Materials Year: 2007	2163
2	Water-dispersible magnetite-reduced graphene oxide composites for arsenic removal Author(s): Chandra, Vimlesh; Park, Jaesung; Chun, Young; et al. Source: ACS Nano Year: 2010	1382
3	Physico-chemical treatment techniques for wastewater laden with heavy metals Author(s): Kurniawan, TA; Chan, GYS; Lo, WH; et al. Source: Chemical Engineering Journal Year: 2006	1045

Table 2. (Cont.) The top 10 most cited articles on arsenic removal

Ranking	Articles	Times Cited
4	Applications of nanotechnology in water and wastewater treatment Author(s): Qu, Xiaolei; Alvarez, Pedro J.J; Li, Qilin Source: Water Research Year: 2013	985
5	Low-field magnetic separation of monodisperse Fe_3O_4 nanocrytals Author(s): Yavuz, Cafer T.; Mayo, J. T.; Yu, William, W; et al. Source: Science Year: 2006	918
6	Remediation technologies for metal-contaminated soils and groundwater: an evaluation Author(s): Mulligan, CN; Yong, RN; Gibbs, BF Source: Engineering Geology Year: 2001	861
7	Advances in water treatment by adsorption technology Author(s): Ali, Imran; Gupta, V.K. Source: Nature Protocols Year: 2006	826
8	Removal of arsenic (III) from groundwater by nanoscale zero-valent iron Author(s): Kanel, S.R; Manning, B; Charlet, L; et al. Source: Environmental Science & Technology Year: 2005	745
9	Arsenic contamination of groundwater and drinking water in Vietnam: A human health threat Author(s): Berg, M; Tran, HC; Nguyen, TC; et al. Source: Environmental Science & Technology Year: 2001	730
10	A review on nanomaterials for environmental remediation Author(s): Khin, Mya Mya; Nair, A. Sreekumaran; Babu V. Jagadeesh; et al. Source: Energy & Environmental Science Year: 2012	616

3.2. Distribution of Major Journals

The top ten productive journals with number of publications are given in Fig. 2. Journal of Hazardous Materials is the leading journal with 218 articles while Water Research is second with 149 articles, and Chemical Engineering Journal is third with 136 articles in the field of As removal. There are only 1068 articles within the top ten productive journals. Desalination and Water Treatment is in Quartile 4 (Q4), Water Air and Soil Pollution is in Q3, RSC Advances and Environmental Science and Pollution Research is in Q2 and remaning six of the top 10 journals are in Q1 category.

The top ten most cited papers present fundamental understanding of any given field [28,29]. Table 2 illustrates publications that have highest citations on As removal from water. The most cited article is a detailed review about types of adsorbents that have been used for As removal and was published in 2007. Two studies are related to environmental remediation of arsenic, one study is on the health and toxicity aspects of As in Vietnam, six of the top ten publications are related to nanotechnology enabled arsenic treatment from water. The list points out that adsorption which uses nanomaterials leads the field of arsenic removal water. Additionally, iron based nano adsorbents are the most frequently used nanomaterial in the top 10 cited articles.

3.3. Contribution of Countries

Distribution and collaboration of top productive countries which have more than 10 publications are illustrated in Fig. 4. A publication can be classified within more than one country, document type and research areas.

China is the most productive country with 880 publications, and it is followed by USA (734), India (649), South Korea (193) and Japan (182). China, USA and India are well ahead on arsenic removal from water. Leading countries, except USA, are Asian countries with a noticeable momentum in recent years. In addition to these countries, contribution of Bangladesh (89) and Taiwan (87) should not be overlooked. High scientific contribution of these countries in academia is not common [37]. This attention is due to high As contamination in drinking water in South-East Asian countries [38]. Hence, Bangladesh and Taiwan contribute on arsenic related research more than other research areas. WHO considered As contamination in Bangladesh to be the largest mass population poisoning in the history [39]. WHO's limit for As in drinking water puts millions of people in the world and in Asia at risk for high As contamination in drinking water [40]. Figure 3 illustrates network of collaborations and links between international scientific communities. The size of the node represents the number of articles published by named country [41]. Different color grouping indicates frequent collaborations between different countries. The minimum document number of a country was set to 10 publications and countries that have met the threshold were shown in the figure below.

The thickness of lines illustrates the strength of collaboration whereas the size of nodes represents the number of publications. The distance between countries indicates the collaboration frequency [26]. Countries that are in the same cluster and closer to each other on the map have more frequent research collaboration. There are 52 countries that have met the 10-publication threshold and 5 different clusters with distinct colors. Leading countries China and USA are in the same cluster and have strong collaboration. Countries that suffer the most from As contamination such as; Bangladesh, Vietnam, Taiwan, Malaysia and Chile have strong links that suggest frequent collaborations with many different countries. Red cluster consists of 18 countries; green cluster consists of 11 countries; blue cluster consists of 10 countries; yellow cluster consists of 8 countries and purple cluster consists of 5 countries. A series of 118 articles for given keywords has been published between 1970 and 2019 from Turkey. Top 5 contributing universities are: Gebze Technical University, Ege University, Yildiz Technical University, Selcuk University and Yalova University. Highest number of publications were submitted to Desalination and Water Treatment, Journal of Hazardous Materials, Desalination, Separation Science and Technology and Chemical Engineering Journal. Approximately 30% of publications from Turkey were published in the top 10 journal list. The publication trend in Turkey correlates with global publication trend.

3.4. Research Tendencies and Hotspots

Nanotechnology provides effective and sustainable solutions for As removal technologies. It has enabled researchers to synthesize, tailor and modify very specific functionalities in nanoparticles [42]. Various nanomaterials which have unique and efficient properties were developed later to remove As from water. For instance, TiO_2 based nanomaterials increase their efficiency more than 200-fold under UV light, zirconium (Zr) based nanomaterials have the highest surface area, magnesium (Mg) based nanomaterials have the highest adsorption capacities and so on [22]. Iron based nanoparticles, mostly iron oxides, have super-paramagnetic properties [43] and it is the most favorable adsorbent in As removal studies. It is cheap and available, easy to produce, have high surface area and easy to separate in water due to magnetic properties [22]. Iron oxide nanoparticles (maghemite or magnetite) has gained momentum in biomedical applications due to their biocompatibility in the early 1990s and later found niche in engineering applications [42].

The top 5 As removal technologies are listed in Fig. 3 Frequency analysis of As removal technologies does not provide information about on-site applications, rather presents scientific approach for As removal. As a result, adsorption is the most used As removal technology with a total of 9010 appearances. It is followed by oxidation (1994), membrane and filtration technologies (1981), ion exchange (744), and coagulation (678). It is crucial to understand that usually these technologies do not operate as a single unit. For example, membrane technologies can require oxidation, As^{+3} to As^{+5}, to enhance removal efficiency. Similarly, adsorption can be used with filtration and in some cases ion exchange. This study only focuses on frequency of these words and does not include combination of these technologies. The evolution of research on As removal shows that until 2000, there wasn't a significant gap in the number of publications between various As removal technologies. However, since 2000, significant work has been done on developing adsorption-based treatment for As removal, making it favorable and dominant research topic for As removal. Majority (80%) of adsorption studies have been published after 2010 while other technologies are between 66 and 73 percent. This finding indicates that adsorption is not only the most studied As technology, it is also gaining more popularity in recent years.

Adsorption has gained popularity because it is efficient, easy to use, cost effective and does not produce toxic byproducts [44]. Nano adsorbents have improved upon limited efficiency of conventional adsorbents, such as the lack of selectivity and active sites [20]. The particle size and pH value also have great importance on arsenic adsorption. Higher adsorption capacity can be achieved for As (III) and As(V) by simply reducing size of nanoparticles from 300 nm to 12 nm [45]. In addition to pH control, arsenic speciation, As (V) and As (III), for As removal is important. As has -III, 0, +III and +V oxidation states and the most common forms of As in natural waters are: arsenite and arsenate, referred to as As (III) and As (V) [46]. Arsenate species (AsO_4^{3-}, $HAsO_4^{2-}$, $H_2AsO_4^-$) are dominant and stable in aerobic environments while arsenite species dominate anaerobic environments like groundwater [19]. Bacteria can also reduce arsenate to arsenite in anoxic environment [6]. Redox potential and pH are driving forces in controlling As speciation [47]. When oxidizing conditions occur, $H_2AsO_4^-$ is predominant at pH lower than 7. $HAsO_4^{2-}$ becomes predominant at higher pH. The uncharged arsenite species predominate under reducing conditions at pH lower than 9.2 [48]. Also, pH_{pzc} values of iron oxide are between 6.8 and 7.5 depending on the type and production parameters of oxides. When pH value is greater than pH_{pzc}, the particle surface becomes negatively charged. Therefore, negatively charged As species lose electrostatic attraction to particle surface area. The optimum operating pH value ranges from around 7–9 for all the iron oxides except for commercial ones which can perform better in acidic conditions [49]. Nanomaterials perform better and usually can be found as powder which is not convenient for column applications due to low hydraulic conductivity and there is a problem of separation and regeneration of certain adsorbents [50]. Oxidation, photochemical, photocatalytic and biological reactions are mainly used to oxidize As (III) to As (V), followed by removal via precipitation. Ion exchange technology in drinking water treatment is mostly used for softening and As removal, but also requires pre-oxidation to enhance As removal performance [40]. These results suggest that when studying As removal from water, As speciation and pH control are fundamental steps towards efficient removal. Different membrane technologies exist for As removal such as nanofiltration (NF) and reverse osmosis (RO). Nanofiltration require sensitive pH adjustment [51] and pre-oxidation [52]. Integrated membrane technologies, such as RO followed by membrane distillation, remove total As from water and produce less toxic by-products than oxidation followed RO. Nevertheless, high energy consumption, with addition of membrane fouling, is limiting worldwide application of membrane technologies [20,53]. Coagulation and flocculation, using a coagulant to form a floc, is a typical conventional method which produces excessive amount of sludge with high concentration of As [40].

3.5. Visualization Map Analysis

The network visualization map of keywords identifies tendencies in a research field of interest; and typically title, abstract and keywords are being used [53,54]. It was carried out within 4083 publications. Fig. 5 illustrates a network of the co-occurrence of keywords. The size of the node represents the number of repeating words. Different color grouping indicates co-occurrence links between keywords [55]. The minimum document number for keywords was set to 10 publications; and 27 keywords that have met the threshold were shown in the figure below. The result of visualization map analysis of keywords is coherent with frequency analysis of As removal technologies. Adsorption is the leading As removal technology with 264 occurrences and oxidation is the second with 100 occurrences. Water, arsenic, removal, and groundwater have high occurrence because they were used in keyword research. Adsorption, oxide, iron, nanoparticle, and bacterium keywords stand out in keyword analysis. Fig. 6illustrates a network of the co-occurrence of words in abstract. The minimum document number for words in abstract was set to 100 publications and 222 words that have met the threshold were shown in the figure below. There are three clusters with 87, 81 and 54 words respectively. The top 5 most repetitive words are arsenic, removal, water, concentration, and adsorption.

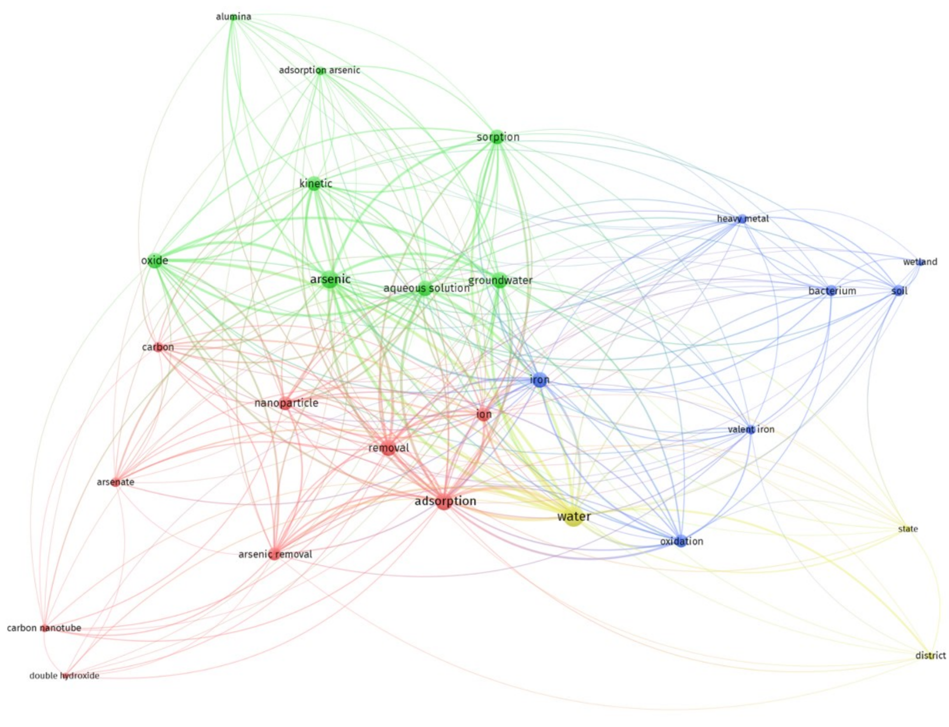

Figure 5. Visualization map analysis of all keywords; minimum number of occurrences of 10

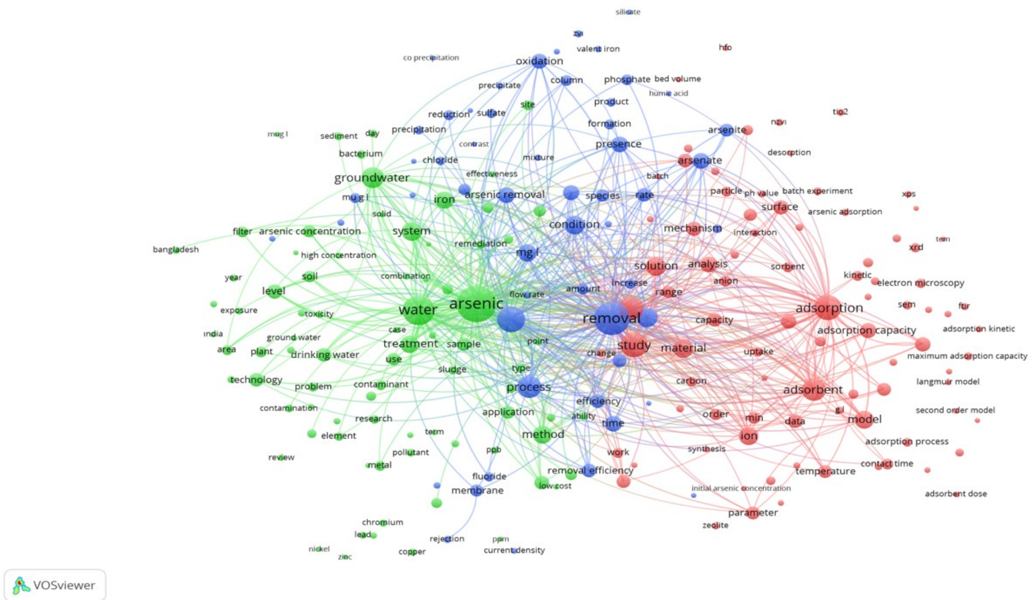

Figure 6. Visualization map analysis of all words in abstracts; minimum number of occurrences of 100

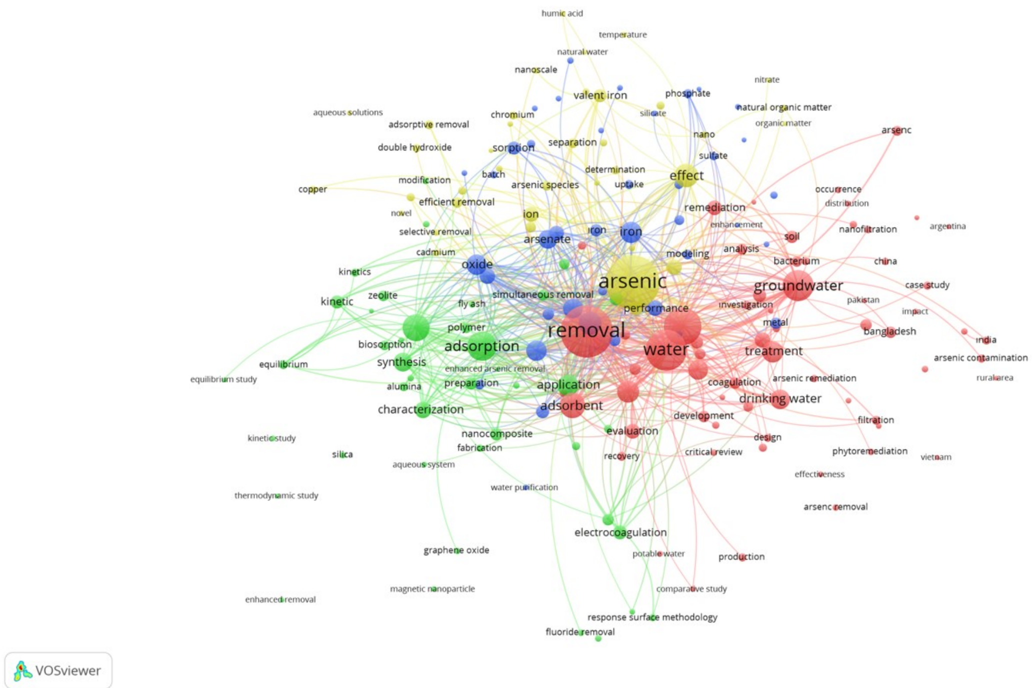

Figure 7. Visualization map analysis of all words in title; minimum number of occurrences of 10

abstract and keywords being used. Using clustering techniques with visualization map analysis points out technologies and methods that are more related to each other.

Fig. 7. illustrates a network of the co-occurrence of words in the title. The minimum document number for words in abstract was set to 10 publications and 186 words that have met the threshold were shown Fig. 7. There are four clusters with 65, 43, 40 and 38 words respectively. Adsorption, iron, and nanoparticle stand out in the title search. The highest number of occurrences and strong links in keyword, title and abstract search are the same keywords in Table 1. Since these words were used in topic search, they have higher occurrence numbers than other keywords as expected. It is crucial to investigate other high occurrence words and clusters when analyzing the visualization map. Taking everything into consideration, visualization map analysis illustrates that adsorption combined with nanotechnology is getting more attention from science community and the data is coherent with frequency analysis and top cited articles.

4. Conclusions

This study presents an overall evaluation of As removal technologies. Publications, journals, countries, and keywords were investigated, and research tendencies and prospects were discussed. A total of 4083 publications related to As removal technologies from 1970 to 2019 were evaluated using bibliometric analysis. The followings are the remarks and conclusions of this study:

- As removal research has a significantly increasing trend after 2000. It is assumed that is a cumulative effect of legislations enacted by WHO, EU and USEPA.
- Article is the most favorable publication type as expected and engineering is the most studied research area. Journal of Hazardous Materials has the highest number of publications with 218 articles.
- China is the leading country in the number of As related publication. Asian countries especially South-East Asian countries draw more attention on As removal studies and their contribution to literature should not be overlooked. Because this points out the obvious fact that these countries suffer from contaminated As water sources.
- It is safe to assume that adsorption techniques using nanoparticles would be the most likely leading As removal technology in that nanotechnology gives major advantages to adsorption compared with other technologies. However, pH sensitivity, cost, regeneration, separation, field applicability will be deciding factors over the years.
- The difficulties regarding As removal technologies are important but temporary, including high cost and limitations. Advancing nanotechnology can provide sustainable solutions to these issues and improve technological ability.

Conflict of Interests

No conflict of interest was stated by the authors.

Acknowledgement

The authors would like to acknowledge the support from The Scientific Research Projects Coordination Unit, Akdeniz University (Project No: FDK-2018-3844). Authors also would like to thank Dr. Hassan Javed, Rice University researcher for insightful contribution.

Declaration of Ethical Standards

The authors of this article declares that the materials and methods used in this study do not require ethical committee permission and legal-special permission.

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