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A bibliometric analysis of research on arsenic in drinking water during the 1992–2012 period: An outlook to treatment alternatives for arsenic removal

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ABSTRACT

A bibliometric analysis based on the Web of Science database was carried out to identify the global research related to arsenic in the drinking water field from 1992 to 2012 and to improve the understanding of the research trends in the same period. The results from the analysis reveal a linearly increasing number of annual publications and a high effort to find effective technical solutions to the problems caused by the presence of arsenic in water. The most relevant research aspects of the four main technologies applied to arsenic removal from drinking water (coagulation, flocculation and precipitation followed by filtration; adsorption and ion exchange; membrane-based processes and biological treatments) were summarized in this paper, with adsorption appearing to be the alternative that has received most attention according to the research trends during the studied period.

1. Introduction

Arsenic (As) is a metalloid that is very abundant and can be easily found in soils, water and biota (specifically in marine organisms), being the twentieth most common element in the earth's crust, the fourteenth one in the seawater and the twelfth one in the human body [299]. It can be considered as a dangerous environmental pollutant, especially in the aquatic medium. The two common oxidation states of arsenic in water are As^{+3} and As^{+5} as part of the dissolved oxyanions arsenite ($AsO_3{}^{-3}$) and arsenate ($AsO_4{}^{-3}$), respectively [54,151]. Both species can be present in different protonated forms as a function of the pH: the As^{+3} system includes H_3AsO_3 and the corresponding deprotonated derivatives $H_2AsO_3{}^{-}$, $HAsO_3{}^{-2}$ and $AsO_3{}^{-3}$ with dissociation constant values of 9.2, 12.7 and 13.4 for pK_{a1}, pK_{a2} and pK_{a3} [228], while the As^{+5} system includes H_3AsO_4 and the corresponding protolitic derivatives $H_2AsO_4{}^{-}$, $HAsO_4{}^{-2}$ and $AsO_4{}^{-3}$ with dissociation constant values of 2.3, 6.8 and 11.6 for pK_{a1}, pK_{a2} and pK_{a3} [3]. According to these data, the prevalent species around neutral conditions (typical pH range for natural surface waters and groundwaters from 6.5 to 8.5) are H_3AsO_3 for As^{+3} and $H_2AsO_4{}^{-}$ and $HAsO_4{}^{-2}$ for As^{+5}. This fact implies that arsenate remains as anion while arsenite appears as a non-charged molecule and it only converts to anion when the pH value is higher than 9.2.

Besides pH, the redox potential plays an important role in the control of the mobility of the arsenic species. A complete speciation diagram for arsenic in aqueous systems as a function of pH and redox potential can be found in bibliography [32]. On one hand, under oxidant conditions, the As^{+5} state becomes clearly dominant over the As^{+3} one but, on the other hand, the As^{+3} species are prevalent under reducing conditions. Therefore, the arsenate system is thermodynamically more stable for surface waters. The case of groundwaters is a bit more complex as both oxidation states are going to coexist and the incidence of each one depends on the arsenic input to the system, the chemical conditions and the biological activity [35].

The presence of arsenic in the environment can be mainly justified by the natural sources because of weathering of rocks and sediments, mineral ores formation processes with hydrothermal origin, volcanic eruptions and geothermal activity [97]. However, anthropogenic activities account for a widespread arsenic contamination as a result of some industrial activities such as mineral ores processing, combustion of arsenic-enriched coals, and manufacturing of semiconductors, glass and some pharmaceuticals [47,172,145].

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Although arsenic is suspected to be an essential trace element in living organisms, where as far as 15 mg of As can be present in an adult human body and it could be related to the synthesis of some aminoacids and the metabolism of zinc [246,171], this metalloid must nevertheless be considered as a high priority toxic pollutant. Arsenic can cause acute, chronic and lethal intoxications. The lethal dose of inorganic arsenic for an adult human is estimated in the range 1–3 mg As/kg [74]. The probability of acute intoxication by ingestion of arsenic-enriched water is minimal and most cases of acute arsenic poisoning occur from accidental ingestion of arsenic compounds or less commonly from attempted suicide [225]. Long-term exposure to arsenic induces chronic intoxication called arsenicosis [65]. It has been reported in people living in endemic zones suffering high levels of arsenic in drinking water [126,21] or burning arsenic-enriched coals indoors [318,153]. It is additionally necessary to mention the health consequences attributable to the genotoxic, mutagenetic and carcinogenic properties of arsenic, since it is classified as a Group I carcinogen, that is, human carcinogen [122].

The presence of arsenic in drinking water is one of the most alarming challenges to be solved in order to assure the human right to safe access to potable water [277]. The contamination of drinking water by arsenic has been reported in more than 70 countries where above 150 million habitants are under high health risk. More than 100 million of these persons live in Southeastern Asian countries like Bangladesh [114], Cambodia [9], China [110], India [257], Laos [53], Myanmar [22], Nepal [305], Pakistan [30], Taiwan [158] and Vietnam [217]. In Latin America, especially in Argentina [229], Bolivia [88], Chile [263] and Peru [76], the same problem has also been reported since several decades ago, affecting mainly urban and rural poor populations. High arsenic concentrations in natural groundwaters or surface waters polluted from industrial sources and the resulting arsenic poisoning episodes have been reported in other countries all over the world [259]. To avoid these toxic conditions, standards for arsenic in drinking water have become stringent. World Health Organization (WHO) produces international norms on water quality and human health in the form of guidelines that are used as the basis for regulation and standard setting [297]. For the particular case of arsenic, the standard limit is fixed at 10 ppb (10 μg/L).

The amount of published papers about arsenic and drinking water that can be found in bibliography is huge, so basic managing tools are very useful to handle all this information. Bibliometrics refers to the research methodology employed in library and information sciences that applies quantitative analysis and statistics methods to describe the distribution patterns of publications according to some given categories such as topic, field, source, author or country. The term bibliometrics was first introduced by Pritchard, who explained that the term "deals with relationships among numbers of scientific papers, numbers of patents, amounts of exports and other quantities" [220]. It is a powerful tool that helps to explore, organize and analyze large amounts of information in a quantitative manner [72] and it has became a common method to analyze the trends in research, including multiple knowledge areas: medicine [93,190], social sciences [121,139], computer sciences [219,193], psychology [41,288], economics [58,237], agricultural sciences [28,59], environmental sciences [85], mathematics, physics and chemistry [216,179,147], arts and humanities [69,45] or engineering [77,176]

The purpose of this study is to bibliometrically analyze the literature published in Web of Science from 1992 to 2012 related to the research on arsenic removal from drinking water. These documents were analyzed and evaluated according to several criteria and were employed to determine the quantitative characteristics of the research on arsenic in drinking water worldwide and find the most relevant present and future trends related to this topic.

2. Data sources and methodology

The data source was Web of Science, the scientific citation indexing service maintained by Thomson Reuters. This searchable platform of publications gives access to several databases and other sources of technical information that can be relevant for the diffusion and evaluation of the scientific research. After the disappearance of Web of Knowledge in 2014, Web of Science has remained as a unique noun and a new denomination for the global database has been created: Web of Science Core Collection. The citation databases provide authoritative, multidisciplinary coverage from more than 12,000 high impact research journals worldwide, including Open Access journals. Cover-to-cover indexing of content is provided by Science Citation Index Expanded, Social Sciences Citation Index Expanded, Conference Proceedings Citation Index, and Arts & Humanities Citation Index [276].

For this bibliometric analysis, the online search within Web of Science was carried out by the insertion of "drinking water" and "arsenic" as keywords in the topic field of the search-engine in order to compile a complete bibliography with all the articles related to the research on arsenic in drinking water published during the period from 1992 to 2012. The final number of articles that were found was 4143.

All the compiled articles were assessed with the following aspects: publication year, document type and language of publications, distribution of output in subject categories and journals and publication outputs of countries and institutes.

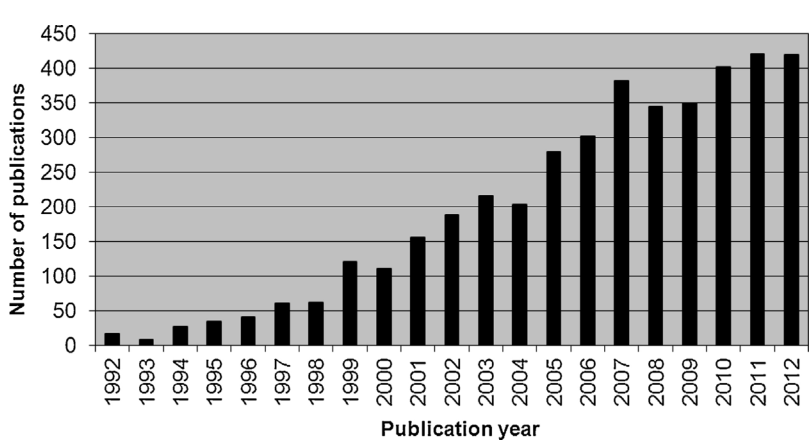

Fig. 1. Annual publication output.

Table 1
The top 10 languages employed by the publications.

Ranking	Languages	Documents	Percentage (%)
1	English	4076	98.38
2	Spanish	26	0.63
3	German	11	0.27
4	French	7	0.17
5	Chinese	5	0.12
6	Polish	5	0.12
7	Czech	3	0.07
8	Japanese	3	0.07
9	Turkish	3	0.07
10	Serbo–Croatian	2	0.05

3. Results and discussion

3.1. Bibliometric analysis of research trends on arsenic in drinking water (1992–2012)

3.1.1. Publication year, document type and language of publications

The distribution of annual publication output identified by Web of Science is shown in Fig. 1. It is obvious that there is a continuously increasing general trend in the number of publications that appears each year, although some exceptions can be found, as for example the year 2007, which was especially productive and has a value higher than the 2008 and 2009 years. As a consequence, the references published for the last six years (from 2007 to 2012) account for more than the half [56%] of the total found publication. Nevertheless, this rise can be considered as a linear evolution for the complete temporal range and it was decided to apply a linear regression to the data. The result was a very good fitting, with a R^2 value of 0.965. The assessed slope was 23.5 publications per year, a figure that gives an idea about the publication rate evolution.

The distribution of document types was analyzed. Nine different document types were found in the total 4143 publications during the studied period. These document types are non-exclusive, so it is possible that some documents can be classified in more than one category simultaneously. Therefore, the sum of the number of documents in each category is above the total number of documents, and a similar result can be found when percentages are analyzed, with a result above 100%. Article (3416) was the most frequently used document type, comprising 82.5% of total production, followed by proceedings paper (482; 11.6%), and review (258; 6.2%). These percentages and specifically the clear supremacy of articles over other types of publication are very concordant with the figures obtained by other authors when analyzing the trends on the research about other contaminants in water [116,311]. The other less significant categories include meeting abstract (119), editorial material (48), letter (21), news item (20), book chapter (16) and correction (5).

An overwhelming majority [98.4%] of all the publications were published in English. Several other languages completed the top 10, Spanish and German appearing as the second and third languages, respectively. The other top 10 languages can be consulted in Table 1. The publications written in the languages that do not appear in the table contribute less than 0.05% to the full bibliography.

It is without doubt that English is the main language of scientific research [92] and it has became the best option for wider communication in many other fields [262,201]. As a consequence, most of the journals listed in Web of Science are published in English [52] and it is common to find bibliometric analyses that refer to the water research area where nearly all the relevant documents are written in English [55,40].

3.1.2. Publication distribution of countries, institutes and authors

The existence of a small number of countries that dominate the publication of scientific documents was expected, as this pattern occurs in most scientific fields [174], including water research [86]. The top 25 countries ranked by the number of total publications are shown in Table 2. Once again, the categories are non-exclusive and a document can be related to more than one country as a consequence of international collaborations. In this case, USA is the most productive country, with more than 1500 documents, which implies a percentage of 38.0%. This leading country is followed by a group of five Asian countries (India, China, Bangladesh, Japan and Taiwan) that jointly produce a higher percentage than the one corresponding to USA [40.5%]. The first two European territories are England and Germany, in the 9 and 10th positions of the ranking, respectively. The high relevance of the Asian countries is not a common fact in other bibliometric analysis about water research issues [266,319]. Besides, the appearing of Bangladesh in the 4th position of the ranking despite its limited scientific contribution to other research fields gives a preliminary idea about the relative importance of the presence of arsenic in drinking water in this part of the world [46], so the South-East Asian countries have to be considered as a main agent in this research topic.

Table 2
The top 25 most productive countries.

Ranking	Country	Documents	Percentage (%)
1	USA	1572	38.0
2	India	513	12.4
3	China	340	8.2
4	Bangladesh	304	7.4
5	Japan	264	6.4
6	Taiwan	252	6.1
7	Canada	177	4.3
8	England	177	4.3
9	Germany	158	3.8
10	Mexico	150	3.6
11	Sweden	136	3.3
12	Australia	120	2.9
13	Spain	104	2.5
14	France	88	2.1
15	Chile	83	2.0
16	South Korea	79	1.9
17	Argentina	71	1.7
18	Turkey	63	1.5
19	Italy	56	1.4
20	Switzerland	55	1.3
21	Scotland	46	1.1
22	Pakistan	44	1.1
23	Greece	42	1.0
24	Brazil	35	0.8
25	Austria	32	0.8

The top 10 institutions were ranked by the number of articles (Table 3). Among the top 10 institutions, 4 were in USA, 2 in Taiwan and one each in China, India, Bangladesh and Sweden. All these countries appeared in the top 7 ranking, except Sweden which holds the 11th position in the corresponding country ranking. However, the Swedish Karolinska Institute is one of Europe's largest and most prestigious medical universities. According to the 2011 Academic Ranking of World Universities, the Karolinska Institute is ranked 9th in the world in the field of clinical medicine and pharmacology and among the first 20 universities in life sciences.

The leading organization was the Environmental Protection Agency of United States (US EPA) with 147 publications, followed by another American institution, the Columbia University (118 documents). The National Taiwan University closes this top 3 exclusive group (107 documents), but again an American institution appeared in the 4th position, in this case, the University of California Berkeley (90 publications).

The 10 most prolific authors in this research topic are shown in Table 4. The top 3 authors have published more than 70 documents each about arsenic in drinking water and the rest of the authors are above 49 articles. The current affiliations of this select group

Table 3
The top 10 most productive institutions.

Ranking	Institutes	Country	Documents	Percentage (%)
1	US EPA	USA	147	3.5
2	Columbia University	USA	118	2.9
3	National Taiwan University	Taiwan	107	2.6
4	University of California Berkeley	USA	90	2.2
5	Chinese Academy of Sciences	China	79	1.9
6	Karolinska Institute	Sweden	76	1.8
7	Jadavpur University	India	74	1.8
8	University of Arizona	USA	71	1.7
9	University of Dhaka	Bangladesh	68	1.6
10	National Cheng Kung University	Taiwan	63	1.5

Table 4
The top 10 most prolific authors.

Ranking	Author	Current affiliation	Documents
1	Allan H. Smith	School of Public Health, University of California (USA)	77
2	Mahfuzar Rahman	International Centre for Diarrhoeal Disease Research (Bangladesh)	75
3	Marie Vahter	Institute of Environmental Medicine, Karolinska Institute (Sweden)	74
4	Habibul Ahsan	Department of Epidemiology, Mailman School of Public Health, Columbia University (USA)	70
5	Chien-Jen Chen	Graduate Institute of Epidemiology, College of Public Health, National Taiwan University (Taiwan)	65
6	Alexander van Geen	Lamont-Doherty Earth Observatory, Columbia University (USA)	57
7	Faruque Párvez	Department of Environmental Health Sciences, Mailman School of Public Health, Columbia University (USA)	54
8	Dipankar Chakraborti	School of Environmental Studies, Jadavpur University (India)	51
9	Yu Chen	Department of Population Health, New York University School of Medicine (USA)	51
10	Joseph H. Graziano	Department of Environmental Health Sciences, Mailman School of Public Health, Columbia University (USA)	49

of researchers are in agreement with the most productive institutions (for example four of these authors work in the University of Columbia). It is worth mentioning the presence of Dr. Mahfuzar Rahman, a leading epidemiologist who is nowadays working for the International Centre for Diarrhoeal Disease Research in Bangladesh, the country probably more affected by the presence of arsenic in drinking water [224].

3.1.3. Distribution of output in research areas and journals

The distributions of research areas are shown in Table 5, where the 10 most popular categories are compiled, taking into consideration that some publications are included in more than one category. The ranking indicates that Environmental Sciences & Ecology, Engineering, Toxicology, and Public, Environmental & Occupational Health are the top 4 most common research areas. The results are in agreement with those ones obtained by a bibliometric analysis applied to the research tends on lead in drinking water [116], but in that case the “Engineering” category was not so relevant and Environmental Sciences & Ecology, Toxicology, and Public, Environmental & Occupational Health were the top 3 subjects in this same order. This fact gives an idea about the significant efforts applied to the search of effective technical solutions to the problems caused by the presence of arsenic in drinking water.

The distribution of outputs in journals is shown in Table 6. The corresponding Impact Factors (IF) of the top 20 journals were also included. Environmental Health Perspectives, Toxicology and Applied Pharmacology and Environmental Science & Technology are the top 3 journals with the highest number of publications (more than 100 documents each) about research on arsenic in drinking water field. The most relevant journal according to the IF among the top 20 journal is also the one with most documents as the IF of Environmental Health Perspectives is 7.260.

3.1.4. Most frequently cited papers

The 10 most cited papers of the found bibliography are presented in Table 7. According to this list, environmental and toxicological aspects of arsenic in water have mainly captured the attention of the researchers along the last twenty years. The two leading documents are reviews about the source, behavior, distribution, metabolism and toxicity of arsenic in water [259,165].

Most of the other documents are related to health and ecology, but the article in third position refers to a treatment alternative to remove arsenic from water since it is a very complete review of adsorbents that can be applied to this purpose [182].

3.2. Trending topics of the research on arsenic in drinking water (2012)

A global approach to the results obtained for the research areas and journals complemented with data about the most cited documents and the affiliations of the most productive authors is useful to evaluate the importance of the engineering component in the global research framework. Although Engineering appears as the second most relevant subject category, none of the most productive authors is working in an engineering-related department and only one of the most cited articles is clearly related to engineering. Besides, the leading journals with most published papers reflecting the research trends on arsenic in drinking water are dealing with different environmental and health aspects.

For this reason, the author keywords analysis was not carried out for the full bibliography taken from Web of Science. Instead, it was decided to analyze the keywords of the articles published only during 2012 by the employment of the Engineering Village database. This database is a service of scientific and technological information that provides integrated access to several important databases covering reference documents all over the world in wide fields of science and engineering. In this case, Engineering Village was used for consulting three different sources of information: INSPEC, COMPENDEX and NTIS. The final number of articles published during 2012 found by Engineering Village when using (“drinking water” and “arsenic”) as references in the Subject/Title/Abstract field of the search-engine was 197. The limitation about the year of publication was imposed in order to consider only the current state of the art. This way, the focus on the current trends and the future directions can be more easily appointed, giving as a result a review that is less biased by previous references.

Table 5
The top 10 most popular subject categories.

Ranking	Researech areas	Documents	Percentage (%)
1	Environmental sciences/Ecology	1579	38.1
2	Engineering	938	22.6
3	Toxicology	863	20.8
4	Public, Environmental & Occupational Health	802	19.4
5	Water Resources	610	14.7
6	Chemistry	472	11.4
7	Pharmacology/Pharmacy	307	7.4
8	Biochemistry/Molecular Biology	161	3.9
9	Geochemistry/Geophysics	161	3.9
10	Geology	147	3.5

Table 6
The top 20 most productive journals.

Ranking	Journal	IF	Documents	Percentage (%)
1	Environmental Health Perspectives	7.260	162	3.9
2	Toxicology and Applied Pharmacology	3.975	138	3.3
3	Environmental Science & Technology	5.257	113	2.7
4	Science of the Total Environment	3.258	109	2.6
5	Journal of Hazardous Materials	3.925	93	2.2
6	Water Research	4.655	86	2.1
7	Applied Geochemistry	1.708	79	1.9
8	Journal of Environmental Science and Health (Part A)	1.252	77	1.9
9	Environmental Geochemistry and Health	2.076	62	1.5
10	Epidemiology	5.738	62	1.5
11	Toxicological Sciences	4.328	59	1.4
12	Toxicology Letters	3.145	59	1.4
13	Desalination	3.041	55	1.3
14	American Journal of Epidemiology	4.780	46	1.1
15	Environmental Research	3.238	43	1.0
16	Journal American Water Works Association	0.627	43	1.0
17	Toxicology	4.017	37	0.9
18	Chemosphere	3.137	35	0.8
19	Journal of Health Population and Nutrition	1.121	34	0.8
20	Chemical Research in Toxicology	3.667	32	0.8

3.2.1. Distribution analysis of author keywords

Fig. 2 lists the 20 most often used keywords appearing in the 197 considered documents. It is clear that "arsenic" is the most relevant keywords as it appears as keyword in 180 papers (91.4% of the total amount of articles). The following five positions of the ranking belongs to keywords that are related to water such as "groundwater", "water supply", "aquifers", "potable water" and "pollution control". All these keywords are used more than 30 times and just with this number of times appears the first mentioned technology employed as keyword: "adsorption". However, within the top 20 most frequent keywords, it does not appear any other mentioned technology.

A look to the results is enough to find three other chemicals in addition to arsenic in the ranking: "manganese", "iron oxides" and "lead". Manganese and iron oxides could again enforce the idea that adsorption is the leading technology, as it is well know that manganese and iron oxides can act as effective adsorbents, specifically for arsenic removal [187,182]. Besides, manganese and iron ions are able to induce the oxidation of As^{+3} species to As^{+5} ones, which are more easily removed by precipitation, adsorption, ion exchange or membrane technologies [119,25,255,250]. Moreover, the interest on low cost reactive filtration media for groundwater treatment has implied the analysis of the potential of green sands for this purpose [143,144], including the removal of arsenic [264,289,275]. These green sands are industrial byproducts of the gray iron foundry industry. They are composed of fine silica, clay binder, organic carbon, and residual iron particles with hydrous manganese oxide and usually mixed with hydrous manganese oxide to form manganese green sands, which can be applied to water treatment. Finally, another reason that justifies the presence of manganese and iron in the ranking is the coexistence of these metals with arsenic in several groundwaters [230,128,188,129,34,154,156]. This same explanation can be applied to lead, the resting chemical mentioned as keyword [223,84,227,304,36,111,203].

Although a further comment on the current research trending topics will be made in the next section, the facts show that keyword analysis can be applied to reveal research trends in the research about arsenic removal from drinking water.

3.3. Current treatment alternatives for arsenic removal from drinking water and future research directions

The list of existing and emerging technological options for arsenic removal from drinking water is quite long but all the alternatives can be included in one of the following categories [7,152,242]:

- Coagulation–flocculation–precipitation followed by filtration.
- Adsorption and ion exchange
- Membrane-based processes
- Biological treatments

A brief summary about the main topics investigated by the found references in each category is shown in Table 8 and a further analysis of each category is carried out in the next sections on the basis of the most relevant references.

3.3.1. Coagulation–flocculation–precipitation followed by filtration

The use of ferric chloride Fe_3Cl as coagulant seems to be the preferred option [117,89,314], since it is widely used in water treatments due to its low price, availability and low risk usabil-

Table 7
The top 10 most cited papers.

Ranking	Articles	Times cited
1	Title: A review of the source, behaviour and distribution of arsenic in natural waters Author(s): Smedley, P.L.; Kinniburgh, D.G. Source: Applied Geochemistry Published: 2002	2057
2	Title: Arsenic round the world: a review Author(s): Mandal, B.K.; Suzuki, K.T. Source: Talanta Published: 2002	825
3	Title: Arsenic removal from water/wastewater using adsorbents – A critical review Author(s): Mohan, D.; Pittman, C.U., Source: Journal Of Hazardous Materials Published: 2007	673
4	Title: Hazards of heavy metal contamination Author(s): Jarup, L. Source: British Medical Bulletin Published: 2003	630
5	Title: Contamination of drinking-water by arsenic in Bangladesh: a public health emergency Author(s): Smith, A.H.; Lingas, E.O.; Rahman, M. Source: Bulletin Of The World Health Organization Published: 2000	608
6	Title: Cancer risks from arsenic in drinking-water Author(s): Smith, A.H.; Hopenhaynrich, C.; Bates, M.N.; et al. Conference: International Symposium On the Role Of The Alveolar Macrophage In The Clearance Of Inhaled Particles Published: 1992	551
7	Title: The ecology of arsenic Author(s): Oremland, R.S.; Stolz, J.F. Source: Science Published: 2003	476
8	Title: Comparative toxicity of trivalent and pentavalent inorganic and methylated arsenicals in rat and human cells Author(s): Styblo, M.; Del Razo, L.M.; Vega, L.; et al. Source: Archives Of Toxicology Published: 2000	462
9	Title: Cancer potential in liver, lung, bladder and kidney due to ingested inorganic arsenic in drinking-water Author(s): Chen, C.J.; Chen, C.W.; Wu, M.M.; et al. Source: British Journal Of Cancer Published: 1992	452
10	Title: Arsenic contamination of groundwater and drinking water in Vietnam: A human health threat Author(s): Berg, M.; Tran, H.C.; Nguyen, T.C.; et al. Source: Environmental Science & Technology Published: 2001	440

ity, including the removal of arsenic [112]. As^{+3} and As^{+5} can form surface complexes with iron hydroxide sites in co-precipitated and adsorbed solids [177]. Nevertheless, the employment of a different iron salt (ferrous sulphate $FeSO_4 \cdot 7H_2O$) can be found [124]. In this case, the remediation approach was based on the pre-oxidation using Fenton's reagent and the subsequent removal of arsenic, which is well referred in bibliography [136,120]. Besides, the filtration was carried out by a slow sand filter instead of microfiltration membranes, which is the most common alternative [298,260,61].

Table 8
Main topics defining current trends about technological solutions for arsenic removal.

	Main topics
Coagulation Flocculation Precipitation Filtration	Iron salts as coagulants Electrocoagulation
Adsorption Ion exchange	Iron-based sorbents Aluminum-based-sorbents Modified solid wastes as sorbents Titanium dioxide Other metallic oxides and hydroxides as sorbents
Membranes	Reverse osmosis Supported liquid membranes Donnan dialysis
Biological treatments	Bacterial oxidation, coprecipitation and sorption Constructed wetlands

Electrocoagulation is based on applying an electric current to a sacrificial anode to generate metallic ions. These ions generated during the process polymerize rapidly, forming precipitates in situ with a high arsenic affinity. Iron and aluminum are the most usual metals [185,283], but others, such as titanium, have been tested [137]. Electrocoagulation is a promising arsenic removal approach that is receiving much more attention in recent years [98,19,169,132,140,82].

In an opposite condition, classical aluminum coagulants (alum, polyaluminum chloride or aluminum sulphate) are not covered by the most relevant identified research trends and seem to have lost the attention they acquired some years ago to remove arsenic from drinking water [101]. Nonetheless, some recent attempts to advance the knowledge of the main mechanisms and the most influencing factors to improve the applicability of aluminum-based coagulants can be found [24,49].

3.3.2. Adsorption and ion exchange

This treatment category is the most mentioned one as confirmed by the conclusions obtained after the distribution analysis of author keywords. A very complete review of the adsorbents that have been tested for arsenic removal can be found among the 10 most cited papers listed in Table 7 but other significant reviews can be mentioned [64,67,91]. A brief analysis of the trends on adsorption shows

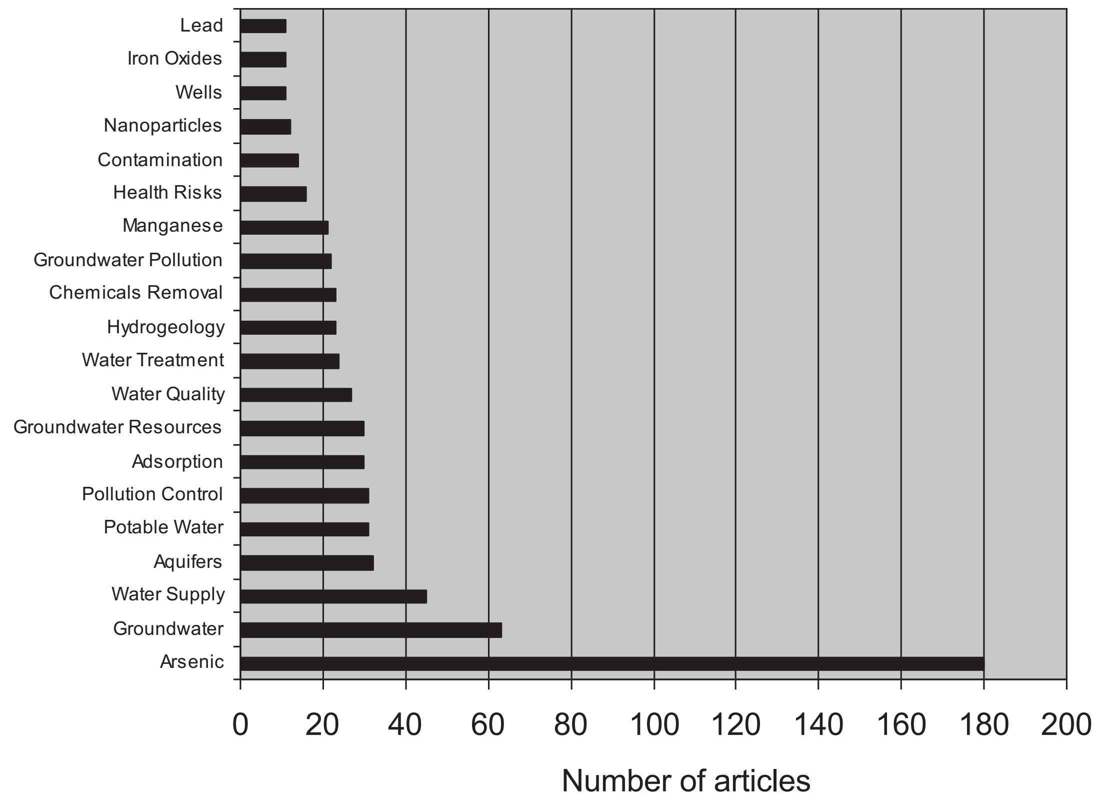

Fig. 2. The top 20 most frequently used keywords.

that the main part of the found papers studies the use of metal-based sorbents, but a great diversity of metals, states and forms is cited.

Iron is by far the most recurrent metal present in the found sorbents. Iron oxides and oxyhydroxides are well reported as efficient sorbents for arsenic removal [29]. Some prepared iron oxide based sorbents are commercially available and have been successfully employed for arsenic removal [202]. More advanced and sophisticated options resort to nanotechnology for the production of nanocomposites [285]. These structures have already been investigated as promising adsorbents for various metallic and organic pollutants [206,231,291,292,221,235,306]. Another quite common approach takes advantage of the magnetic properties of adsorbents based on some iron compounds such as magnetite [96,256,196,1], maghemite [208,261] or jacobsite [209]. They are employed in coordination with graphene and other substances to produce different types of nanocomposites [159,191,252]. The removal of arsenic by zero-valent iron (ZVI) is a well-known process based on the adsorptive properties of the compounds generated by the corrosion of ZVI in water [141,148,37,310,75].

The removal of arsenic from water by zeolites is well-reported [73,253,181] but more recent efforts have been focused on modifying their surface properties. Among all the possible alternatives for these modifications, the use of surfactants has gained relevance [146,56,313,175,270]. The use of other aluminum-based adsorbents has been tested, including aluminum embedded in polymeric matrixes [163,200,218,232,268] and Bauxsol, [94,95,44,108].

Titanium dioxide based arsenic removal methods developed to date have been focused on the photocatalytic oxidation of arsenite to arsenate and the subsequent adsorption of arsenate [211,215] The application of titanium dioxide in arsenic removal can be divided into two aspects: it functions as both photocatalyst and adsorbent when UV light or sunlight irradiation are present but it works only as adsorbent in the absence of irradiation. Many efforts have been taken to improve its performance by combing it with adsorbents that exhibit good adsorption properties to obtain multifunctional systems, like activated coal [307,39,308,302] or polymeric materials [194,81,13]. Titanium dioxide based materials are relatively well established for arsenic photocatalytic oxidation and adsorption but some future needs for this technology have been proposed, including the reduction of the treatment cost, the decrease in the operational complexity of the technology or the disposal of the arsenic-bearing treatment wastes [102].

Investigations using zirconium loaded materials such as activated charcoal [212,245], porous resin [269], chelating resin [18], nanoparticles [300] or waste biomass such as orange waste [26] have shown promising results for arsenic removal, so it is not unusual to see new efforts to apply nanotechnology for the production of advanced zirconia-based sorbents [60,197]. Other adsorbents based on different metal have been investigated for arsenic removal from water, for instance, copper [38,214,315] and cerium [247,71,107,303]. Once again, the promising obtained results have been improved by innovative approaches to obtain nanostructured adsorbents [170,48,99,226,267].

The use of commercially available ion exchange resins is not covered by any of the found references. Papers regarding anion exchange resins to remove arsenic from water are more common during the first years of the last decade [133,281,131,149,189], but some recent references indicate that this field is still worthy of some attention [51,16]. The potential of novel ligand and chelating resins [10,123,183,11,2,15,134,184], metal-loaded ion exchange resins [248,186,254,160,109] or low-cost ion exchangers from waste biomass [296,12,280,79,258] are more current topics. However, the contribution of the nanotechnology to enhance the properties of ion exchangers is the real trigger that has revolutionized this field [68,100,239,284,115,150,27,63,241,279,17].

It is clear that the application of nanotechnological solutions to improve the performance of adsorbents and ion exchangers shows great prospects in advancing drinking water treatment to increase arsenic removal efficiency as well as to improve water supply by a safer use of unconventional water sources.

3.3.3. Membrane-based processes

The oxidation from As^{+3} to As^{+5} is crucial for the performance of the nanofiltration and reverse osmosis membrane technologies. As^{+3} is present at typical pH values of most natural waters as the non-ionic form of arsenious acid H_3AsO_3, which is very poorly retained by the membranes. Further information about pre-oxidation of trivalent arsenic can be found in the bibliography [164]. More recently, one reference introduces a study that explains the experimental results obtained by application of different reverse osmosis membranes to the treatment of arsenic-enriched well waters in Texas, taking into account the employment of different oxidant chemicals (chlorine and hydrogen peroxide) to carry out an oxidation pretreatment previous to the membrane process [272].

Besides the oxidation state of arsenic, pressure driven membrane treatments have to deal with other important drawbacks, as another reference reviews [173]. The reduction of the degree of arsenic removal with the decrease of arsenic concentration in the feed solution has been demonstrated [290], a fact that has to be taken into great consideration because initial arsenic levels in natural waters are mainly in the sub-ppm range. The applied pressure only slightly affects the efficiency of arsenic removal by nanofiltration and reverse osmosis membranes, so the advantages of operation at high pressures to obtain higher permeate production are not reflected in better permeate quality [198,244,233].

Other relevant papers use advanced membrane systems like supported liquid membranes [162] and Donnan dialysis, an electrochemical potential driven membrane technology [317].

Three different stages and phases are involved in supported liquid membranes: solute extraction from the feed phase, diffusion of solute through the extractant-containing phase and re-extraction of solute to the acceptor phase. For the particular case of arsenic removal, both feed and acceptor phases are aqueous while the extractant is maintained in an organic phase [207,103]. Hollow fiber membrane contactors are excellent supports for the design of supported liquid membranes [90,243,31]. Several extractants have been identified for the removal of arsenic: organophosphorus extractants such as the Cyanex® trade family [168,104], Aliquat 336 [23], citric acid [180] or ascorbic acid [210]. Some new bio-based chemicals are nowadays under investigation for arsenic extraction, such as the application of mucilage, a pectic polysaccharide extract for a cactus [83]. This potential extractant is especially attractive since it is renewable, biodegradable and can be sustainably managed, contributing this way to establish more accessible technological solutions.

In a typical Donnan dialysis process, neither high applied pressures nor electric fields are required since the separation is based on an ion exchange membrane. This ion exchange membrane separates two solutions (feed and stripping solutions) and excludes co-ions (ions with the same electrical charge as the ion exchange membrane) from permeating the membrane. Therefore, the flux of a counter-ion (ions with the opposite electrical charge to the ion exchange membrane) through the membrane caused by a concentration difference is always coupled with the transport of identical numbers of counterions in the opposite direction to maintain the electroneutrality in both solutions [240]. Anion-exchange membranes are required to remove arsenic from the feed solution and an anionic counterion (most of the times chloride) has to be present in the stripping solution. Several Donnan-dialysis based arsenic removal processes have been tested and they have demonstrated the feasibility and ease for implementation [316,287].

Furthermore, another innovative technology based on membranes has been investigated: membrane distillation. This type of process, based on a non-conventional technology, can be a feasible alternative to high arsenic content waters. The process operates in the temperature range of 40-80 °C and can be powered by waste process heat or low grade renewable heat sources [309]. The arsenic removal rates, taking into consideration that minimal measured retentions over 99.7% have been reported [222,167,205], were much better than for other reported membrane filtration techniques like nanofiltration and ultrafiltration [322]. Besides, membrane distillation operation has shown another important advantage: it allows total arsenic removal from water without the use of oxidant agents which can result in lower environmental impact [161]. Therefore, these promising results should promote a higher number of studies to analyze the real-scale application of this technology.

It is clear that the amount of papers regarding membrane technologies can not be compared with the huge production about arsenic adsorption. Anyway, the noteworthy contribution about membrane research during the first decade of this century has to be taken into consideration [125,195,282,199,286,135,301,204,192] and more recent examples can also be found [5,80,106,213,249,6,234,4,57,274,312].

3.3.4. Biological treatments

The application of biological processes to the removal of arsenic from water is an emergent field that shows a great potential mainly because it has to be seen as the most environmentally compatible treatment option [42]. Biological activity can remove arsenic by various different mechanisms such as sorption, biomethylation–demethylation, complexation, co-precipitation or oxidation–reduction processes. Besides, a diversity of organisms shows potential for its application for arsenic removal, including bacteria [87,127,320], fungi [271], yeasts [251], algae [236,105,138,265], lichens [238], plants [142,118,50,8,70] and complex biological communities as constructed wetlands [33,155,14,157] or even waste biomass [273,62,166].

The analysis of bacterial systems is the most common practice among the papers identified. However, the references make use of the microorganisms' activity in different ways. Anaerobic reactors that contain denitrifying and sulphate reducing bacteria can reduce the arsenic content of polluted waters [278]. Under these conditions, the arsenic removal relies on the generation of iron precipitates (oxy-hydroxides and sulphides), which are very effective in sequestering arsenic by co-precipitation [78]. However, the contributions of adsorption and complexation mechanisms can not be neglected. Adsorption is the main mechanism of the other references. A method for bacterial production of a bio-sorbent for arsenic removal was developed [113]. *Leptothrix* spp. bacteria were used to obtain biological iron precipitates that can be employed as bio-sorbents [43,130]. The characterization of these precipitates determined the presence of iron oxides and oxy-hydroxides in association with bacterial flocs to form multilayer structures. The use of arsenite-oxidizing bacteria as an effective pre-oxidation step previous to the adsorption onto zero-valent iron-coated sand has also been investigated [294]. The bacteria *Thiomonas arsenivorans* was inoculated to a fixed-bed reactor filled with the adsorbent to convert As^{+3} to As^{+5}, a much more easily adsorbed specie. Although many arsenite-oxidising bacteria have already been isolated from different environments and described in the literature [178], *T. arsenivorans* is able to use As^{+3} as an energy source for growth [20], so it is of particular interest for the development of arsenic removal bioprocesses [66,293].

Constructed wetlands have been employed extensively around the world during the last decades for improving water quality, including the removal of heavy metals and emerging contaminants. The main removal pathways of As in constructed wetlands are biologically-mediated precipitation, co-precipitation and sorption, but the relative role importance of the plants and the microorganisms present in the medium has to be assessed [321]. Another topic still pending is the evaluation of additional plant species for As removal in order to establish the widest possible geographic and climatic ranges for this biotechnological treatment.

Biological methods appear as efficient ways to treat arsenic contaminated waters but, until now, most biotechnological treatments for arsenic removal have only been investigated in laboratory studies under well-defined conditions. Further full-scale demonstrations are required to demonstrate their technical viability in more complex real systems and the corresponding cost-effectiveness analysis need to be conducted to assess their applicability [295].

4. Conclusions

An overview of the research on arsenic in drinking water was presented with the information related to annual publications, document types, languages, countries, institutions, authors, categories, journals and research emphases and tendencies. The number of publications about this subject increased linearly during the 1992–2012 period. Many studies in the categories of environmental sciences and ecology, engineering, and toxicology have been investigating the environmental and health consequences of arsenic in water and looking for technical solutions to overcome the problems this pollutant causes when present in drinking water. The USA appears as the leading country in total number of publications, but it is followed by a group of Asian countries (India, China, Bangladesh, Japan, and Taiwan) which are very concerned about the aresenic problem. The U.S. Environmental Protection Agency had the highest productivity among the institutions.

The proposed analysis by words in title, abstract or subject within the Engineering Village database provided the clues for the identification of hot technological issues. Adsorption was by far the most investigated treatment alternative. Several metallic compounds (mainly oxides and hydroxides), including iron, aluminum, titanium, zirconium, cerium or copper, are the most relevant sorbents under study, but some attempts to modify solid wastes to transform them into effective adsorbents can also be found in the bibliography. Commercially available ion exchange resins have moved out of the major research focus to be replaced by novel ligand and chelating resins, metal-loaded resins, and low-cost ion exchangers from waste biomass. The application of nanotechnology to improve the performance of adsorbents and ion exchangers shows a very promising potential. Classical membrane technologies such as nanofiltration and reverse osmosis with a long history can still be considered as compelling solutions, but more advanced options like liquid membranes or Donnan dialysis have emerged to demonstrate their viability and improved performance. Another classical option such as iron salts as coagulants is still a common research focus, but the more sophisticated electrocoagulation appears as a more promising alternative. The most deeply investigated biological treatments (that is, constructed wetlands and bio-assisted oxidation, co-precipitation, and sorption processes employing bacteria) have proved its usefulness to treat arsenic contaminated waters but, until now, most biotechnological treatments for arsenic removal have only been studied in laboratory tests, so further investigation becomes necessary to demonstrate their technical and economic viability in real systems.

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