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A mini review on phytoremediation of fluoride-contaminated waters: a bibliometric analysis

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Traditional techniques for removing fluoride (F⁻) from water are costly and complex, whereas phytoremediation is a less expensive method that can be adopted as an ecotechnological tool to control not only fluorine, but also other pollutants in urban and agricultural watersheds. Phytoremediation as a strategy for defluoridation is little used and still has a long way to go. Therefore, it is important to get a picture that describes scientific achievements and trends around this topic and support decision-making processes to move forward. Hence, the aim of this study was to collect bibliometric data on the use of phytoremediation to remove F⁻ from Scopus and Web of Science databases. Data collected considered the scenario of related publications regarding countries and researchers that publish the most, the chronological evolution of the publications, the co-occurrence of terms to point out overall focus and interests of researches and collaboration initiatives around the subject. India has the highest number of publications on F⁻ phytoremediation, which can be explained by the high incidence of fluorosis in that country. The number of publications per year, although still small, has grown over the last 5 years. A cluster map showing the co-occurrence of terms indicated research focused on the effects of F⁻ on plant metabolism, and the correlation of this contaminant when in the presence of heavy metals and with issues of sanitation. Different studies also seek species of accumulator plants, and investigate physical and chemical parameters of the environment that influence the absorption of the F⁻ anion. New approaches to optimize the process and phytoremediation overall are on the rise like the use of phytohormones, growth-promoting bacteria, sorption processes and genetic manipulation.

KEYWORDS

bibliometrics, fluorine, defluoridation, fluorosis, accumulator plants

1 Introduction

Water quality is a reflection of the environmental conditions and human activity in the watershed. Fluorine, e.g., is a recurrent element in nature, and its presence in water is one of the challenges to be overcome by society. Dividing opinions in the scientific community, fluorine, albeit beneficial to health in small concentrations, at high levels becomes a public health problem, culminating in a high incidence of fluorosis and other ailments. Fluorine

enters the water via natural or anthropogenic pathways, in the form of the fluoride ion (F^-) and the World Health Organization recommends a maximum value of 1.5 mg L^{-1} of F^- in the drinking water (WHO, 2017).

While there are traditional methods of defluoridation for when fluoride levels are inappropriate, these methods are costly and complex (Solanki et al., 2021). It is therefore necessary to find less expensive solutions to adjust the levels of fluoride in water when it becomes a problem for supplying the population.

One simple alternative is phytoremediation, a method on the rise for many pollutants. However, when it comes to fluoride, phytoremediation remains a little-used technique, with room for improvement. Previous reviews carefully covered the subject (Weerasooriyagedara et al., 2020), but bringing up bibliometric assessments to the discussion would certainly improve the comprehension of the state of art and support decision-making processes to move forward. Therefore, this study aimed to carry out a mini-review and a bibliometric analysis on phytoremediation as a strategy for the treatment of fluoride-contaminated water, presenting the findings of science so far, and relating them to aspects of public health and the environment.

2 Materials and methods

Searches were carried using two databases: Web of Science (Clarivate Analytics) and Scopus (Elsevier). These databases provide a lot of information, such as the number of publications related to the search terms by year, country, area, type of document, journal, language and affiliation.

The terms selected to search the databases were fluoride, fluorine and phytoremediation. Other terms related to the word phytoremediation were tested in the searches, such as phytoextraction, phytodegradation, phytostabilisation, phytovolatilisation, phytofiltration, and rhizofiltration. However, including these terms did not increase the results, and only the initial three terms were employed, with the search configured to return documents containing these words in the title, abstract or keywords. No restrictions were placed on the period of publication when searching for the initial year, and 2021 was chosen as the final year, to close the decade. The Boolean operators AND and OR were used to organise and filter the search results, with the search carried out using the expression “(fluoride OR fluorine) AND phytoremediation,” so that the search returned results that would include the term phytoremediation together with either fluoride or fluorine. The results from each database were used to evaluate the documents in relation to the year of publication, country of origin and affiliation, authors with the highest number of publications, research area (employing the categories used by the databases), and language.

The database that returned the largest number of files was selected to create bibliometric maps using the VosViewer software. The results for terms in the parent databases are downloaded in .csv format (Excel) and input to the software to create clusters maps. These maps present such information as the frequency and co-occurrence of the terms, co-authorship, and citations, among others, showing the relation strengths between the items.

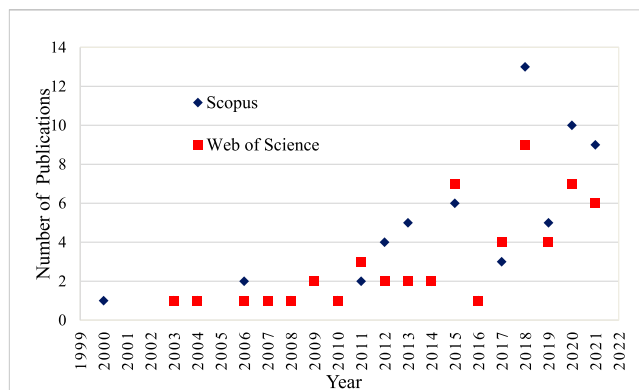


FIGURE 1

Number of publications per year (up to 2021) from Scopus and Web of Science databases after searching the expression “(fluoride OR fluorine) AND phytoremediation.”

The co-occurrence map was produced considering all the keywords in the documents, but was limited to words that occurred at least three times. All words that met this criterion were filtered to exclude repeated terms or terms that added nothing to the analysis, for example, article, priority journal, and review, among others. The co-authorship map was produced without a minimum number of documents per author, but was limited to those with at least three citations in published works.

To generate the maps, the strong association method of normalisation was adopted, a measure of probabilistic similarity for normalising co-occurrence data recommended by the VosViewer developers. This method considers the ratio between the observed number of co-occurrences of two objects and the expected number of co-occurrences when it is assumed that these objects are statistically independent. The distance-based maps supported by the software may present only a subset of labels depending on the amount and relationship between items. More details can be found at Van Eck and Waltman (2009).

3 Results

The results of the searches for the terms “(fluoride OR fluorine) AND phytoremediation” generated a collection of 67 documents in the Scopus database and 55 in the WoS, which can be considered a limited number of publications on the topic. Within these databases, 82% (55 items) and 89% (49 items), respectively, represent original papers, with the remainder being review articles, book chapters, editorials or conference works. Only three documents are not published in English (one in Chinese, one in Portuguese, and the other Russian). The main area of published papers in both databases is the Environmental Sciences. Also among the most relevant areas are Agricultural Sciences, Water Resources, Chemistry, Toxicology and Botany.

India, by itself, is the country with the highest number of publications relating to fluoride phytoremediation in both databases, followed by China and USA. The first six authors with the highest number of papers, also in both databases, are either Indian or are affiliated with Indian institutions. Suphiya Khan and Vinay Sharma appear with the most publications indexed in Scopus and WoS.

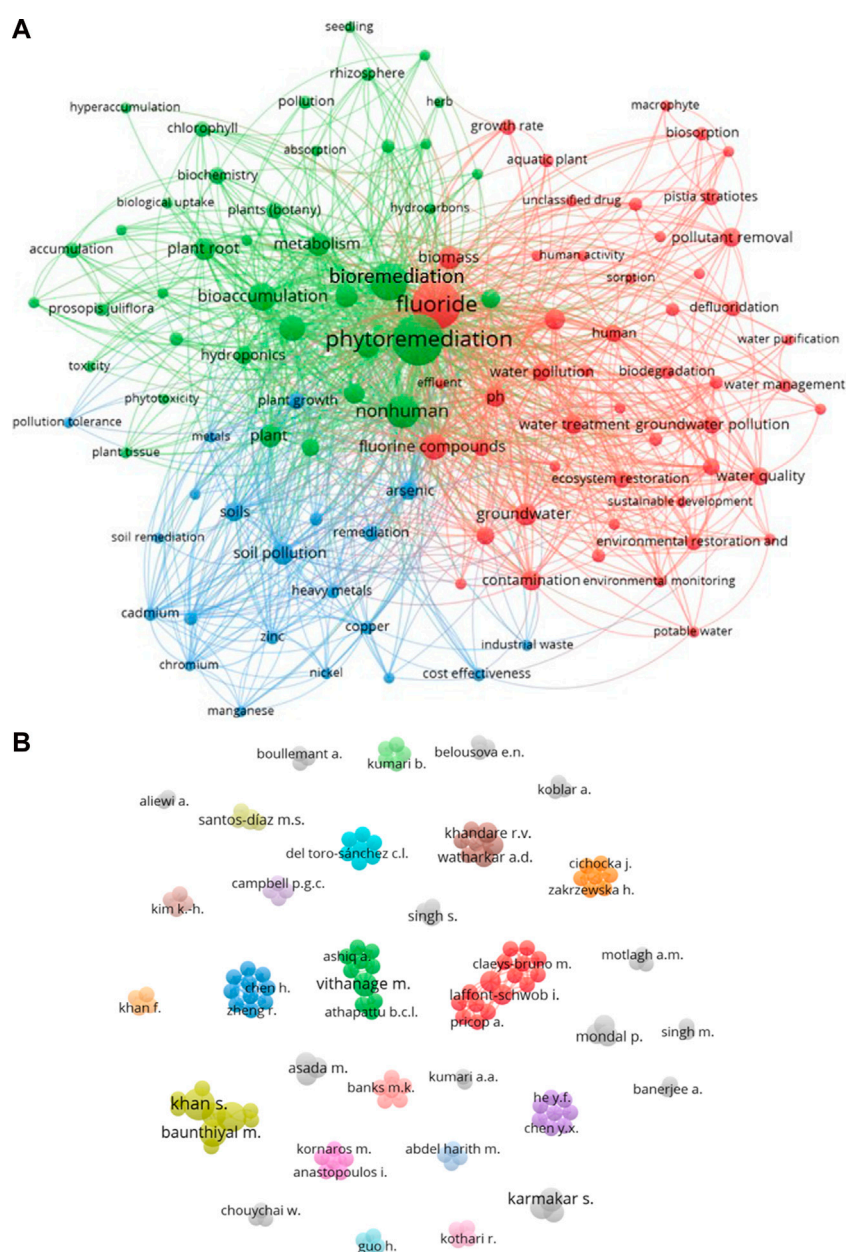


FIGURE 2 Cluster maps showing (A) the co-occurrence of terms in the titles, abstracts and keywords of the documents obtained and (B) the co-authorship of the documents gathered from the Scopus database.

The oldest publications on fluoride phytoremediation in the databases and included here date from 2000 to 2003 for Scopus and WoS, respectively, with 2018 having the highest number of publications in both databases (Figure 1). The data also show that from 2015 to 2021 the number of studies on the subject has increased. These are indications that the strategy of using plants to remediate media contaminated with fluoride is a recent approach.

While there are files in common, some documents only appear in one database or the other. As there is a limitation with VosViewer on combining one database acquired from various different databases to generate the grouping maps (clusters), the database

with the highest number of documents returned in the search was selected.

When analyzing the co-occurrence of terms, 130 met the criterion of appearing at least 3 times within the database. After excluding repeated and irrelevant terms, this number was reduced to 110 terms that were distributed over five clusters. However, there was a certain affinity between the terms that made up the clusters and, as such, the map was reduced to three groups by manipulating the resolution tool of the program that carries out the grouping (Figure 2A).

The terms, “fluoride” and “phytoremediation” occupied the centre of the map, being very close to each other, despite being grouped in different clusters. They showed a greater frequency and

relation strength with the remaining terms, which is expected since they were the keywords used in the search to obtain the database. The term “fluorine,” also used in the search, appeared less frequently and in the same grouping as “phytoremediation.” F atoms basically exist in the anionic form of fluoride (F^-), the free form being practically non-existent (Howe-Grant, 1995). As a result, the studies give preference to the term “fluoride” instead of “fluorine.” Disregarding the search terms, the three terms with the highest number of occurrences were “bioremediation,” “nonhuman” and “bioaccumulation” in that order, all of them grouped with the term “phytoremediation.”

The grouping presented in green shows that this cluster is limited to studies focused on plant performance and the effects caused by fluoride on their health and metabolism. In the blue-coloured grouping, the terms refer to metals, metalloids and soils. This grouping, therefore, indicates research in which fluoride phytoremediation, in both water and soil, was studied together with the presence of other contaminants, especially metals. As for the red group, it includes a set of terms related to sanitation and environmental and human health.

For the co-authorship map, generated for authors with at least three citations in their work regardless of the number of works per author, 155 of the 245 authors met the minimum criteria. These were distributed over 31 isolated groupings, i.e., having no relationship with each other, which contained between 2 and 16 authors (Figure 2B).

Although there is some collaboration between researchers allocated to the same cluster for one or more studies, such collaboration does not occur outside of these clusters, indicating little or no collaboration between research groups. When evaluating the institutions and countries of the authors that make up the same cluster analysis, it is clear that they are, either wholly or in the most part, affiliated with the same institution; or they are from the same country, and may come from more than one institution; or the authors are from the same subarea of study.

4 Discussion

The small but growing number of publications found in both databases for the period assessed is a sign that the use of plants for fluoride removal is a new and on the rise approach. Phytoremediation as a process has now been studied for several decades, although it is also considered recent, and there are other, often established ways, of removing fluoride from water, although these are complex and expensive, and therefore quite often inaccessible (Sinha et al., 2003; Boukhris et al., 2015).

It is natural for India to be strongly involved in research on the decontamination of water with high concentrations of fluoride, since the country faces a serious public health problem due to the high incidence of fluorosis in the population (Khairnar et al., 2015). The region is located in one of the areas known as the fluoride belts. These belts delimit regions where the rocks are rich in fluoride and, therefore, active weathering processes as well as mineral exploitation end up releasing large amounts of the element into the soil and bodies of water (Chowdhury et al., 2019).

For the co-occurrence map (Figure 2A), studies grouped in the green cluster show that when investigating tolerant and accumulator species of a contaminant, it is important to evaluate how it enters the plant, how it behaves in the plant tissue. Furthermore, the effects

fluoride presence causes on plants, whether morphological, anatomical or enzymatic, in which organs it is accumulated, how the plant reacts to the stress it causes, and if the plant eliminates it, how it does so, are also critical points to assess (Sinha et al., 2000; Saini et al., 2012; Saini et al., 2013; Singh and Verma, 2013; Koblar et al., 2015; Zhao et al., 2015; Karmakar et al., 2016; Karmakar et al., 2018; Rodrigues et al., 2020; Siahouei et al., 2020; Zhang and Chen, 2020; Calabrese and Agathokleous, 2021; Khandare et al., 2021). Plant species who show a high rate of assimilation of a given contaminant without suffering severe damage are considered accumulator species, and are ideal for the phytoremediation of that contaminant (Khandare et al., 2017; Gadi et al., 2020).

The fluoride ion is absorbed passively through the roots by the processes of diffusion, from where it is distributed both via symplastic and apoplastic pathways to other parts of the plant, or via the xylem to the leaves (Singh et al., 1995; Hong et al., 2016). Fluorine uptake by plants is related to its speciation, which in turn is dependent on the pH and the activity of the fluoride ion, though some findings may raise controversy (Stevens et al., 1998; Karmakar et al., 2018; Braga et al., 2021; Vaz et al., 2023). The presence of other anions or cations (e.g., metals) in the solution and redox potential can also interfere with this process in a way yet to be fully understood, thus observations are inconclusive (Ruan et al., 2004; Singh et al., 2018; Banerjee and Roychoudhury, 2019). Sinha et al. (2003) also found that an increase in temperature favoured the removal of F^- by *Eichhornia crassipes*.

Mondal (2017) found reductions of up to 19% in the germination rate of *Orisa sativa* when exposed to fluoride concentrations of up to 20 mg L^{-1} . As fluoride interferes with amylase activity, and seed germination is highly dependent on the energy from carbohydrate synthesis, the process is impaired, and reflects in reduced plant propagation (Ram et al., 2014; Mondal, 2017). Sinha et al. (2000) saw reductions in the chlorophyll and protein content of *Hydrilla verticillata* in aquatic environments with up to 25 mg L^{-1} fluoride. The chlorophyll and carotenoid content of *Pistia stratiotes*, *Spirodela polyrrhyza* and *Eichhornia crassipes* were not affected following exposure to fluoride concentrations of between 3 and 20 mg L^{-1} ; as such, the stress was considered negligible for the three species (Karmakar et al., 2016).

Most plants show signs of phytotoxicity at low fluorine concentrations, with some species being extremely sensitive to the presence of this element in the medium (Jha et al., 2009). On the other hand, despite excess fluoride having a negative effect on most plants, some species, being accumulators, are tolerant to media rich in the element (Baunthiyal and Sharma, 2012). In Mexico, Santos-Díaz and Zamora-Pedraza (2010) investigated 17 species tolerant to F and HF (hydrogen fluoride), and concluded that only *Camellia japonica*, *Pittosporum tobira* and *Saccharum officinarum* were fairly efficient in removing up to 40% of the fluoride from the medium.

Karmakar et al. (2018), using *Pistia stratiotes*, demonstrated that aquatic plants with accumulating potential resist oscillations in pH without undergoing large variations in assimilating a contaminant such as fluorine. This was observed by Braga et al. (2021) as well, where, despite obtaining greater fluoride removal by *Landoltia punctata* under alkaline conditions (up to 21%), high levels of removal were also achieved at a pH of less than seven.

On the other hand, there are also certain disadvantages to the method, which is subject to local and seasonal climate conditions.

Good plant performance also depends on their tolerance to the target contaminants, i.e., the toxicity and concentration of the chemical species present in the medium to be remedied greatly influence the health of the plants and their removal ability. Furthermore, the reuse of the biomass produced is a challenge once it is not suitable for animal feeding, similar to heavy metals and any other non-degradable contaminant. Some strategies have been investigated such as the production of fiber and paper, depending on plant species, and polymers, biofuels, and adsorbent materials as well, though regarding fluoride not a lot has been published (Ajithram et al., 2020).

In regards to the group in blue (Figure 2A), fluoride was studied alongside the presence of other contaminants, mostly metals, a very common situation for wastewater from industrial activity (Gadi et al., 2020; Ahila et al., 2021; Calabrese and Agathokleous, 2021; Thakur et al., 2021). Finally, the group in red indicates that the presence of fluoride in the drinking water, regardless of the source, is related not only to severe scenarios involving fluorosis, but also to other adverse health effects (Ghaderpoori et al., 2019). The situation is even more worrying when certain populations, especially rural or poorer populations, depend exclusively on springs, or surface or underground sources, where the water is rich in this element, which often occurs together with other contaminants (Maity et al., 2021). It is therefore essential to find techniques of defluoridation that can also serve communities that are far from large centres, or that lack financial resources and access to healthcare (Sundaravadivel and Vigneswaran, 2010; Maity et al., 2021).

Co-authorship map clearly shows that cooperation between countries, subareas and even institutions is still rare, which may be another indication that the topic is still new, with much that still needs to be studied. Among the scientists with the most publications, citations, and collaborations, these from Indian universities stand out.

Lastly, phytoremediation is a technique that shows great application potential, although, like any other technique, there are disadvantages to be considered and processes to be improved. A successful search for suitable plant species, an understanding of the main physicochemical factors, as well as innovative operational approaches aimed at improving the process or adapting it to different pollutants, have paved the way for optimisation. For instance, the research of new materials for use as a support medium for plants, the application of phytohormones, manipulation of growth-promoting bacteria, genetic engineering of accumulator plants, and combined or conjugated systems, among others, have been increasingly reported (Sinha et al., 2003; Kim and Owens, 2010; Bulak et al., 2014; Li et al., 2014; Athapattu et al., 2017; Sarwar et al., 2017; Kaur et al., 2019; Barbosa et al., 2023; Vaz et al., 2023).

5 Conclusion

Bibliometric analysis is a tool that details and enhances comprehension around a topic. It corroborates what previous reviews found and sometimes bring up to surface what such publications could not. There is still a lot to be investigated with regard to fluoride phytoremediation given the small number of publications found in the databases searched herein, although this number has been growing over the last years and considering that other databases may index more studies. India is very concerned about the high concentrations of fluoride

present in its water sources, which is reflected in the high incidence of fluorosis in the population. It is therefore natural that the country should publish the most on the subject.

The bibliometric map showing the co-occurrence of terms (cluster map) was structured into three large groupings, from which it could be seen that the use of plants to remove fluoride from the environment is being investigated both from the perspective of plant health and to find tolerant and accumulator species, as well as from the point of view of sanitation including public and environmental health. Furthermore, some macrophytes have already been identified as accumulators, although there is still no consensus on certain physicochemical variables in the environment that influence the process of fluoride assimilation by these plants, including the effect of heavy metals.

Although the technique of phytoremediation is relatively new, especially when applied to fluoride removal, strategies to improve the process have been emerging, such as the use of phytohormones, growth-promoting bacteria, enrichment of the medium with sorptive materials (e.g., biochar), and genetic manipulation. Combining the technique with filter media instead of just hydroponics, as is the case of constructed wetlands, has also been reported in the literature, with good results in terms of removal efficiency. Improvements also need to aim towards the reuse of biomass in a safe and sustainable manner.

Author contributions

LV: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Writing—original draft. JC: Data curation, Supervision, Writing—review and editing. AB: Conceptualization, Data curation, Methodology, Project administration, Supervision, Writing—review and editing.

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Conflict of interest

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