A bibliometric analysis and assessment of priorities for heavy metal bioavailability research and risk management in contaminated land

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Abstract

Risk assessment has been recognized as an important tool for evaluating heavy metal pollution and providing risk-based information for decision makers. In order to accurately assess the risk of heavy metals in contaminated soil to human health, it is necessary to conduct bioavailability studies on heavy metals in soil. Bioavailability of heavy metals in soils and the implications for risk assessment and land management/remediation – has evolved over the decades and now has considerable practical and economic implications internationally. This article aims to explore it's evolution by undertaking a bibliometric analysis of the research fields which have addressed heavy metal bioavailability in soils, with a focus on the risk assessment of contaminated land and human exposure to soil-borne metals. Bibliometric analysis techniques are applied to monitor and assess the changing research literature on the bioavailability of heavy metals in contaminated soils. Over 5000 articles were found for the period 1979-2020. The purpose was not to perform an exhaustive literature review, but to draw out trends and patterns in the literature, and to make observations on past and current priorities. Key words were extracted from the analysis and the roles of different countries in driving the research literature identified. Three phases in literature/subject development were identified. Between 1979-2000 (initial phase, 213 articles), studies used extraction procedures and solubility studies to investigate the roles of soil properties on metal form/speciation and focused on bioavailability to (crop) plants in agricultural soils. Between 2001-2010 (slow development phase, 1,105 articles) attention switched to metals introduced in soil amendments and wastes, metal impacts on soil microbial processes, and incorporating bioavailability in risk assessment. More rigorous techniques were being used, such as the Diffusive Gradients in Thin Films (DGT) technique to better understand kinetic and metal speciation in soils and the quantitative relationship to bioavailability. By 2011-2020 (rapid development phase, 3,137 articles), research was being conducted in many countries (site specific, often industrially contaminated and urban sites), with a focus shift to health risk assessment, remediation, and bioavailability to various ecological receptors (e.g. humans and animals), with the development of many methods of bioavailability (e.g. simulated gastro-intestinal tract enzymolysis methods). Some priorities for research on soil heavy metal bioavailability are identified.

Key words: bibliometric, heavy metal, bioavailability, contaminated soil, phase development

1. Introduction

With the development of global industrialization, urbanization and modernization over recent decades, soil heavy metal pollution has become a focus for environmental pollution management worldwide (Nriagu, 1996; Järup, 2003). Risk assessment has been recognized as an important tool for evaluating heavy metal pollution and providing risk-based information for decision makers (Fairbrother et al., 2007). Currently, most risk assessments are based on the total concentration of heavy metals in the soil, but these metals are not absorbed with 100% efficiency by biological systems, so the risks of heavy metals in soil are often over-estimated. This leads to higher land management and remediation costs. How to determine the fraction that can be effectively absorbed by the human body (or other receptors, such as grazing animals or crops) is a key part of contaminated land risk assessment. In order to accurately assess the risk of heavy metals in contaminated soil to human health, it is necessary to conduct bioavailability studies on heavy metals in soil – or use methods which can mimic or estimate the available fraction. Although the definition of bioavailability can be different in different research fields (Harmsen, 2007; Meyer, 2002), in the context of soil heavy metal contamination of current study, it refers to the fraction of pollutants in the soil that can transfer into biological systems. For example, for humans this means entering the human body through the oral cavity, skin, breathing and other exposure pathways, passing through the gastrointestinal tract and being absorbed into the human body (Kelley et al., 2002). Many countries have incorporated bioavailability measurement methods into risk assessment techniques. For example, in 2007, the United States Environmental Protection Agency (USEPA) issued guidelines for evaluating the bioavailability of soil heavy metals in human health risk assessment (USEPA, 2007). Although the logic of assessing the bioavailable fraction, rather than using total metal concentrations, has been appreciated by experts in the field for many years, there is still a lack of consensus about the method(s) to determine the bioavailability of soil heavy metals. The fraction of heavy metal which is bioavailable in soils varies between elements and is influenced by soil types, heavy metal forms, exposure pathways, soil physical and chemical properties, and environmental factors. The focus for research on soil heavy metal bioavailability has differed over time and in different regions of the world, as knowledge and priorities change driven by politics, technologies, improved scientific understanding, economic development etc. (Sun et al., 2019).

Determination of bioavailability, or at least methods to estimate it, can be *in vitro* and *in vivo* procedures (Juhasz et al., 2010; Brattin et al., 2013). As examples, an *in vitro* measure of bioavailability from soil to crop plants may use a chemical extractant to mimic the 'bioavailable fraction'. *In vivo* bioassay methods can use test plant species, earthworms or micro-organisms. For higher organisms, different animal models have been used. The fraction of heavy metal absorbed by the animal fed known amounts of a prescribed diet is determined by comparing what is ingested by the animal with what is excreted. Obviously this can vary between species and with diet/soil type, because the chemical conditions, residence time, microbiology etc varies in the gastro-intestinal (GI) tract of the different test species. The animal models commonly used for heavy metal determination are primates, pigs, rabbits and mice, but they are limited by ethical considerations, cost and differences in physiology, which limit the use of primates, pigs and rabbits for example (Juhasz et al., 2010; Brattin et al., 2013). Although previous studies have shown that *in vivo* experiments in mouse models have considerable advantages in determining the bioavailability of heavy metals (Bradham et al., 2011; Bradham et al., 2018; USEPA, 2021), they are not suitable for risk assessment of heavy metal contaminated sites at a large scale for ethical reasons and cost issues (Juhasz et al., 2010). Therefore, for sites that require a large number of bioavailability measurements, it is necessary to develop *in vitro* simulated model systems to replace them. The GI simulation method is currently a widely used *in vitro* simulation method (Juhasz et al., 2009; Denys et al., 2012; Li et al., 2020a). It measures the dissolution rate of soil pollutants in the simulated human GI tract. There are now more than 10 GI simulation methods used worldwide. In addition to GI simulation methods, there are also simpler techniques, which determine the soluble or extractable or labile fractions of heavy metals in soils (Harper et al., 2000). This is where the study of heavy metals in soils began, several decades ago, with a focus on the influences of soil chemistry, and the bioavailability/uptake by plants. For example, Alloway & Morgan (1986) carried out a long-term study concerned with the forms of soil pollutants including Cd, Pb and Ni from various sources and the extent to which these metals were taken up and translocated into the edible parts of different food crops. Baker et al. (1994) assessed the performance of a range of plant species (hyperaccumulator species and non-accumulating species) to extract metals under crop conditions.

As this Introduction has briefly shown, this general research topic – bioavailability of heavy metals in soils and the implications for risk assessment and land management/remediation – has evolved over the decades and now has considerable practical and economic implications internationally. In this article we explore it's evolution by undertaking a bibliometric analysis of the research fields which have addressed heavy metal bioavailability in soils, with a focus on the risk assessment of contaminated land and human exposure to soil-borne metals. More specifically, the analysis explores the evolution of the field, its changing focus on different topics, tools and techniques. It identifies those countries where most research and assessment has been undertaken and the trends in literature over past decades. We then use the analysis to consider current research activity, and highlight recent research trends and upcoming priorities.

2 Data acquisition and analysis methods

2.1 Data acquisition

This study selected the Web of Science (WOS) core database of the Institute for Scientific Information (ISI) as the data source. WOS is a comprehensive database, widely used in various fields of bibliometric analysis. It can obtain relevant documents of global scientific research, covering the most comprehensive academic information resources in the world over most disciplines, including natural sciences, biomedicine, and engineering technology. The database holds information from >8,700 core academic journals which have the most influence in various research fields, and it is a universal international scientific research analysis tool. Therefore, the literature selected the WOS Core Collections sub-database by searching the topics (TS). In the process of advanced database searches, the use of keywords is crucial to the accuracy of the search. The search formula is: TS = (soil AND (heavy metal OR heavy-metal) AND (bioavailability OR bio-availability OR bio availability)). The key search time span is 1979-2020, the document type is selected as "Article". After screening and manual removing the less relevant documents which are lack of authors/keywords information and irrelevant to the topic, a total of 5,117 papers were obtained. After removing any replicated articles, there were 4,455 articles used in the co-occurrence and cluster analyses of keywords and country.

2.2 Analysis methods

This study firstly evaluates the current research status and influencing factors in the field of global heavy metal bioavailability from three perspectives: 1) the time trend characteristics of the number of papers, 2) their countries of origin, and 3) the current research hotspots. In addition, CiteSpace software was used to perform a cluster analysis of the relevance and hot trends in the literature, to identify soil heavy metal bioavailability studies in different countries and regions. The largest cluster in the map is the cluster generated with "soil heavy metal bioavailability" as the theme. Modularity Q and Mean

Silhouette are two important indicators for evaluating clustering. Modularity Q is an indicator for evaluating the effectiveness of clustering. Modularity Q >0.3 indicates that clustering is effective; Mean Silhouette is an indicator for clustering homogeneity, the larger the value indicates that the homogeneity of the cluster is better, and Mean Silhouette >0.7 indicates that the cluster is reliable.

3 Results and Discussion

3.1 Yearly publication records

The first article in the field of bioavailability of soil heavy metals identified by the database search was published in 1979 (Bingham, 1979), which explored the relationship between the bioavailability of 21 food crops and the heavy metal content of sludge-amended soils. In total, there are 4,455 article records used in this analysis from 1979 to 2020. Figure 1 clearly shows that the number of records for each year and cumulative records have increased strongly with time. Based on the cumulative records of publications, research relating to bioavailability of soil heavy metals over the past ~41 years can be divided into three broad phases - an initial phase, a slow development phase and a rapid development phase. Although the precise time points of these phases are somewhat imprecise, we take these to be as follows - an initial phase (213 records) between 1979-2000, a slow development phase (1,105 records) between 2001-2010 and the rapid development phase (3,137 records) between 2011-2020 (and likely still continuing). We present an analysis of these phases to highlight changes in topic focus, country of origin etc., as explained below.

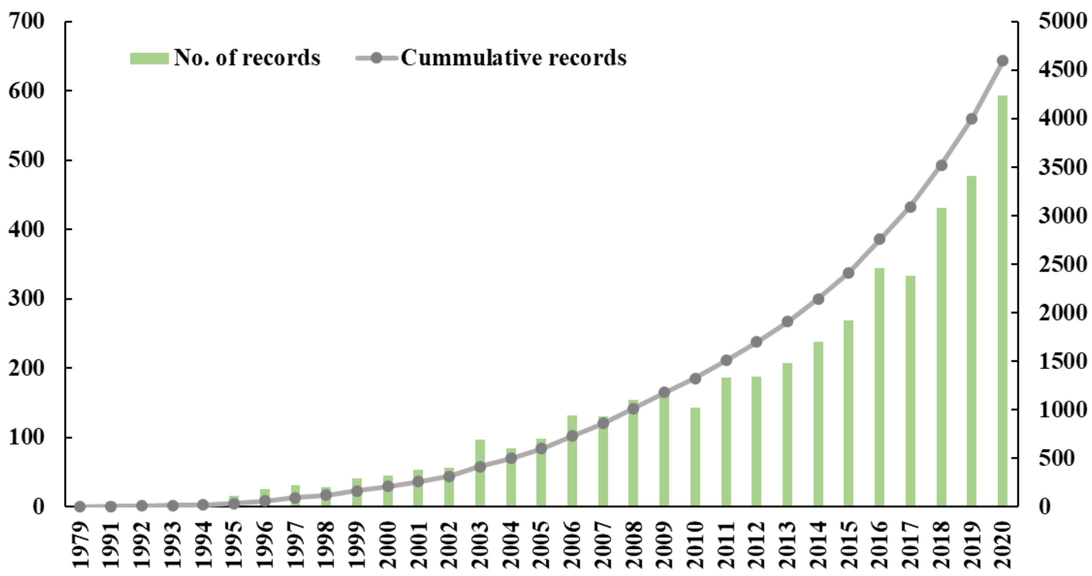

Figure 1: Publication records relating to bioavailability of soil heavy metals from 1979 to 2020 in the Web of Science database.

Table 1 shows the top 14 most productive countries (frequency ≥ 100) with publications on the bioavailability of soil heavy metals during 1979-2020. Over the whole time period the sequence is as follows: China (1,336), USA (484), Spain (263), Australia (184), France (178), Italy (163), Pakistan (163), Poland (163), India (141), South Korea (129), Iran (125), Brazil (123), UK (113) and Germany (101). China has focused on soil heavy metal bioavailability because of the fast development of urbanization, with the need for evaluation and potential re-development of inner city/industrialised areas. Although the highest frequency among all countries occurred in China, the highest centrality was found in USA (1.06). The greater the centrality, the more research will be carried out through this point, and the centrality exceeds 0.1, which is considered to have a higher influence. Thus, the USA has had the highest influence in the field of soil heavy metal bioavailability so far.

Table 1 Frequency (Frequency of different countries ≥ 100) and centrality (centrality of keywords ≥ 0.1) of articles relating to soil heavy metal bioavailability in different countries between 1979 and 2020.

Country	**Frequency**	**Centrality**	**Keywords**	**Frequency**	**Centrality**
China	1336	0.16	Contamination	499	0.49
USA	484	1.06	Availability	404	0.41
Spain	263	0.11	Cu	842	0.36
Australia	184	0.05	Element	121	0.34
France	178	0.09	Remediation	310	0.25
Italy	163	0.05	Adsorption	288	0.25
Pakistan	163	0.03	Phytoremediation	227	0.23
Poland	163	0.03	Heavy metal	2,891	0.22
India	141	0.01	Sediment	351	0.17
South Korea	129	0.03	Speciation	699	0.13
Iran	125	0	Amendment	258	0.13
Brazil	123	0.01	Accumulation	681	0.11
UK	113	0.03	Toxicity	412	0.11
Germany	101	0.09	Mobility	249	0.1

3.2 Keyword frequency analysis

Keywords are the core of a bibliometric analysis and indicate the research hotspots in the subject area. The software analysis tools identified different keywords and different numbers of keywords in the different time periods. This reflects the dominance/strength of different topics, the research hotspots of the research field, and the frontiers and trends of research. The data of 4,455 articles was imported into the CiteSpace software to generate a keyword co-occurrence map of the bioavailability of heavy metals in soil. The keywords with a frequency of greater than or equal to 100 are shown in Figure 2, while Table 1 & Table S1 gives detailed information of frequency and centrality. The keywords “soil”, “heavy metal”, and “bioavailability” appeared 1,149, 2,891 and 1,734 times, respectively. From Figure 2 and Table 1, research content revolves around three aspects of soil heavy metal bioavailability, namely: migration and transformation, risk assessment, remediation. There are 2,891 articles relating to heavy metals, with a focus on Cd, Zn, Pb and Cu. There are 1,734 articles relating to bioavailability, mainly focused on plant availability. There are 1,149 articles relating to soil, with the main focus on agricultural soil. There are 680 articles relating to contaminated soil.

In addition to the frequency of keywords, centrality is also a major indicator to determine research hotspots. Centrality refers to the mediating effect of a node in a certain field and its influencing factors. The greater the centrality, the more research will be carried out through this point; if the centrality > 0.1, it is considered to have a higher influence. In the keyword co-occurrence map, keywords with high centrality are usually displayed with a purple aperture (Figure 2). The results indicate that heavy metal, Cu, accumulation, speciation, contaminated soil, sediment, remediation, adsorption, amendment, mobility, phytoremediation, and element have the greatest influence (Table 1).

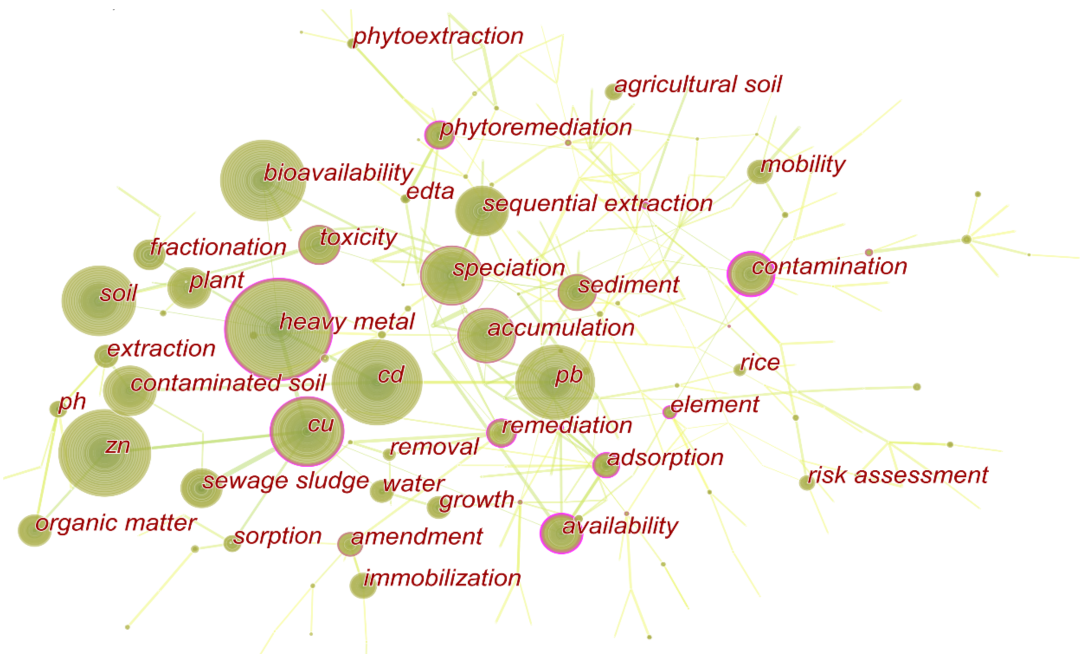

Figure 2 Co-occurrence map of key research keywords (frequency ≥ 100) for the bioavailability of soil heavy metals in 1979-2020

3.3 Changes over time in the research hotspots and frontiers relating to soil heavy metal bioavailability

By analyzing temporal changes in clustering keywords, trends in the field of soil heavy metal bioavailability research focus are identified which provides a reference for considering future research directions. Keyword clustering can focus on the research field, to help decipher and understand the knowledge structure basis of the field, and aggregate closely related words to highlight relatively independent research fields.

3.3.1 Main research keywords in the initial phase (1979-2000)

Figure 3 shows the research keywords of soil heavy metal bioavailability focused on six aspects between 1979-2000 – namely #0 sewage sludge, #1 heavy metal, #2 sequential extraction procedure, #3 community DNA fingerprint, #4 desorption kinetics and #5 heavy metal geochemistry. In this initial development phase, research on soil heavy metal bioavailability mainly focused on the speciation (#2, #4) and geochemistry (#5), with a focus on heavy metals entering agricultural soils due to the addition of sewage sludge (#0) as a soil amendment. This was a hot topic in Europe and North America at this time, with countries discussing whether addition of sludge to land needed to be regulated/controlled, because of the presence of heavy metals (Towers & Paterson, 1997). Analysis of the 213 research articles published in the 1979-2000 timeframe identified four burst keywords (Figure 4A) - Zn, Cu, sewage sludge and pH.

Studies during this phase often focused on specific sites, soil types and chemical extraction methods to assess heavy metal release and forms in soils (Chlopecka, 1996; Cieśliński et al., 1996; Krishnamurti et al., 1995; Rieuwerts & Farago, 1995). For example, Krishnamurti et al (1995) explored the speciation of soil particulate-bound Cd by using a sequential chemical extraction procedure. Chlopecka (1996) evaluated the forms of Cd, Zn and Pb in contaminated calcareous and gleyed soils. Cieśliński et al (1996) showed soil types affected on Cd bioaccumulation and distribution within the plants. Researchers realized that the bioavailability of heavy metals might change with their speciation. Focus turned to bioavailability rather than the total concentration of heavy metals (Alexander, 2000). During this period, chemical methods were developed to measure the speciation of heavy metals - single extraction and sequential extraction methods. Researchers found that speciation of heavy metals varied with soil properties and chemical extractants. The principle of sequential extraction methods is to use a series of reagents to extract heavy metals step-by-step, with weaker then stronger extractants. This approach can reveal the binding status of metals and soil solid phase components. It can help with understanding of their adsorption-desorption, migration and transformation processes in the soil, and their soil plant nutrient (e.g. Cu, Zn) chemistry and environmental chemistry. During this period, the Tessier five/seven-step sequential extraction method (Tessier et al., 1979), the BCR four-step sequential extraction method (Whalley & Grant, 1994; Rauret et al., 1999) and the Maiz three-step sequential extraction method (Maiz et al., 1997) were proposed. Although the Tessier method has been studied and tested for a relatively long time, it still has some shortcomings, such as multiple extraction steps, possible re-sorption and re-distribution during the extraction process, and weak reproducibility of experimental results (Pueyo et al, 2004). The BCR method was based on the Tessier methods, adapted by the European Community Bureau of Reference, which generated the first standard reference material for geochemical speciation studies (BCR601). This method has relatively simple steps and accurate results; it has been widely used to determine the speciation of heavy metals in soils and sediments (Davidson et al., 1994).

Figure 3 Cluster map of research keywords on the bioavailability of soil heavy metals during 1979-2000. Based on the log-likelihood analysis, Modularity Q is 0.7007, Weighted Mean Silhouette S=0.9262. Each module in the keyword clustering map is a cluster. The larger the module, the greater the number of keywords in the cluster. The text on the module is the name of the cluster, and the largest cluster is marked with #0.

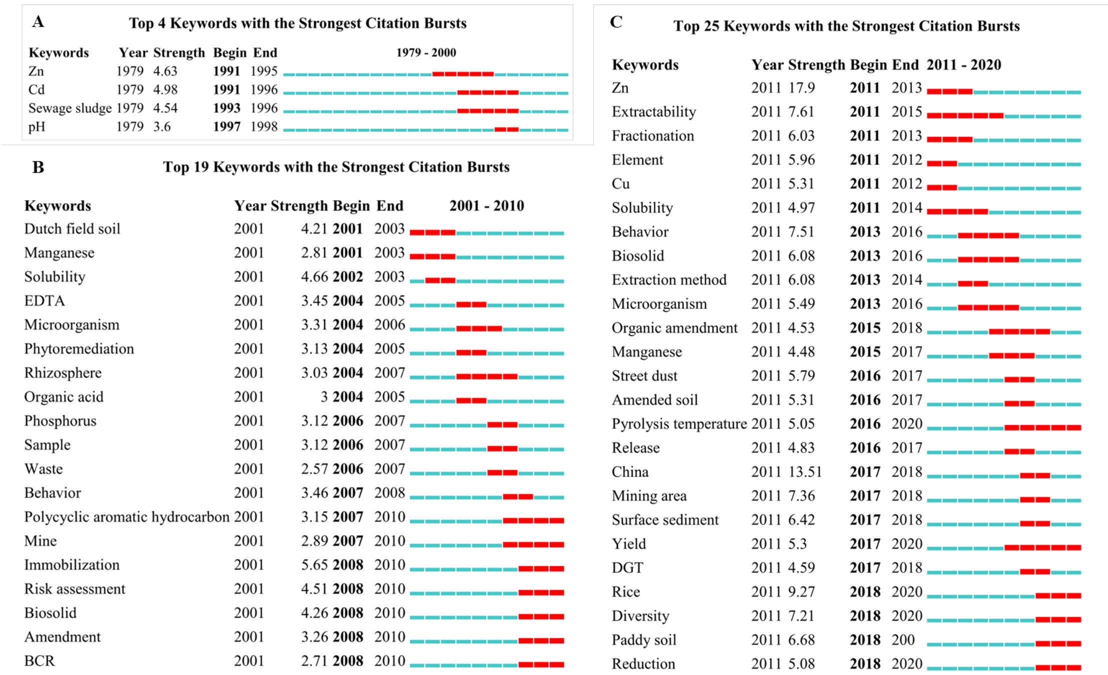

Figure 4 Strongest citation bursts of top keywords relating to soil heavy metal bioavailability during 1979-2000 (A), 2001-2010 (B) and 2011-2020 (C).

3.3.2 Main research keywords in the slow development phase (2001-2010)

Figure 5 shows the research hotspots on soil heavy metal bioavailability between 2001-2010. During the slow development phase, research continued to focus on the speciation (#5, #8, #7) of heavy metals, and sewage sludge (#2). Studies on the influence of soil types emerged (e.g. agricultural soil (#4) and calcareous soil (#6)). New research keywords showed studies exploring the influence of soil amendments (#1), studies on soil-plant interactions (test plant species (#3)) and the impacts of heavy metals on soil microbial processes (basal respiration (#9)).

Analyzing 1,105 article records, 19 burst keywords were obtained (Figure 4B) which included: Dutch field soil, manganese, solubility, EDTA, microorganism, phytoremediation, rhizosphere, organic acid, phosphorus, sample, waste, behavior, mine, immobilization, risk assessment, biosolid, amendment. These highlight that studies were focusing on the solubility, extractability and interactions of metals from soils, the effects of various soil treatments and amendments on heavy metals in soils, and risk assessment and remediation of metal contaminated soils.

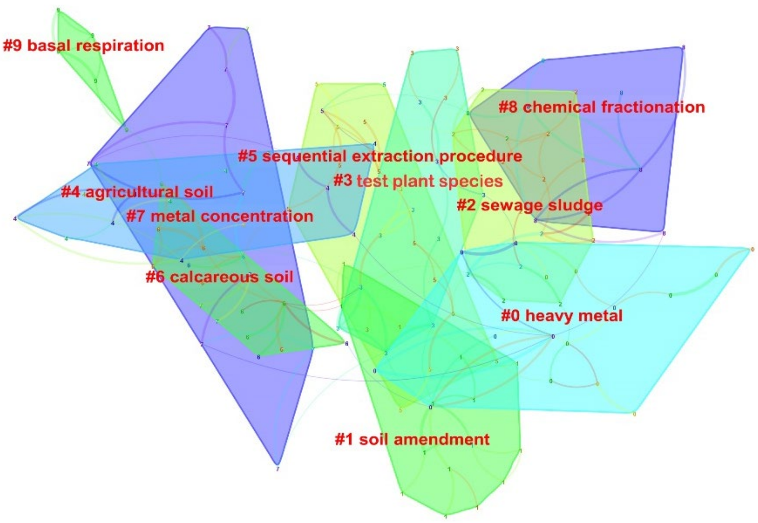

Figure 5 Cluster map of research keywords of the bioavailability of soil heavy metals during 2001-2010. Based on the log-likelihood analysis, Modularity Q is 0.7704, Weighted Mean Silhouette S=0.9216. Each module in the keyword clustering map is a cluster. The larger the module, the greater the number of keywords in the cluster. The text on the module is the name of the cluster, and the largest cluster is marked with #0.

(1) Speciation (#5, #8, #7) of heavy metals:

During this period, speciation of heavy metals in soils measured by sequential extraction methods still was a hot topic, with a focus on methods to evaluate metal availability in studies around the world (e.g. in China and Canada). For example, Lu et al (2003) analyzed the mobility and bioavailability of Cu, Zn, Pb and Cr based on the ~150 soil samples from 20 urban and 3 non-urban soil profiles in Nanjing, China. Li et al (2006) applied the short sequential extraction procedure for the determination of available fraction of Zn, Cu and Cd in Harbin, China. Preciado & Li (2006) evaluated Cu, Fe, Pb, Mn and Zn of roadside sediments using sequential extraction method in British Columbia, Canada. China had the highest frequency of publications on bioavailability at this time (Table S2), as urbanization and re-development of contaminated sites became an important environmental issue in the country (Sun et al., 2019, 2022).

(2) Heavy metals in sewage sludge-amended soils (#2):

During the 2001-2010 period, applications of sludge to land remained an important topic, while the production of sewage sludge in urban areas of developing countries increased in line with greater urbanization and industrialization. Landfill, farmland utilization and incineration accounted for a large proportion of sludge disposal in developed countries. The EU’s policy on waste treatment was to produce no waste, minimize the amount of waste produced, recycle, incinerate, and landfill. Agricultural use and landfilling were the two most important methods of sludge disposal in most countries. The carbon and nutrient value of sludge provided benefits to soils, while the heavy metal content could be a concern. This is why the content of heavy metals in the sludge should meet standards for the control of sludge applications to land and why the bioavailability of metals to crops and the potential impacts on soil processes and organisms has been investigated.

(3) Phytoremediation (Studies on the influence of soil types emerged (e.g. agricultural soil (#4) and calcareous soil (#6)), Studies on soil-plant interactions (Brassica juncea (#3)), The impacts of heavy metals on soil microbial processes (basal respiration (#9)) and Studies exploring the influence of soil amendments (#1)):

During this period, the area of soil contaminated by heavy metals is expanding rapidly. Research on the remediation of contaminated soils was developing rapidly and became a popular research field. In-depth development of the occurrence and development processes of contaminated soil, contaminated soil remediation research and applications became important in this phase. Research on the remediation of contaminated agricultural soil has become a current hot scientific issue and frontier field. Conventional physical and chemical treatment technologies, such as soil replacement, washing, heat treatment, solidification, vitrification were being explored and were coupled with studies that investigated changes in metal form, availability and effects. Phytoremediation was an attractive concept, because it was presented as a 'green technology' (Raskin & Ensley, 2000), but it has many practical limitations (Sarwar et al, 2017). Literature explored how plant stabilization may be a way to reduce the bioavailability of polluting elements *in situ* (Yan et al., 2020), rather than a permanent method to remove pollutants from the soil (Adeoye et al., 2022). For example, phytostabilization is one of widely used techniques of phytoremediation, which seeks to physically stabilize the contaminated site with vegetation cover (Adeoye et al., 2022). In this technique, some plants with high metallic-tolerant ability have been applied to reduce the bioavailability of heavy metals in soils, further to immobilize these metals belowground and reduce the possibility of these metals from entering to the food chain. For example, a variety of *Miscanthus* × *giganteus* has been reported that can reduce metal accumulation in above-ground biomass (Zgorelec et al., 2020).

3.3.3 Main research keywords in the fast development phase (2011-2020)

Figure 6 shows the research hotspots of soil heavy metal bioavailability during 2011-2020. During this fast development phase, more diverse research on soil heavy metal bioavailability has taken place than the initial and slow development phases (#1, #0, #11, #12). Research has developed on health risk assessment (#8) and remediation (#2, #14, #3) of soil heavy metal in industrial soils (#13). In addition, research interests have started to focus on contamination factors (#15) and mechanisms of action (#4, #9, #10, # 6) of soil heavy metals.

Analyzing 3,137 article records, 25 burst keywords were obtained (Figure 4C). These are: Zn, extractability, fractionation, element, Cu, solubility, behavior, biosolid, extraction method, microorganism, organic amendment, manganese, street dust, amended soil, pyrolysis temperature, release, China, mining area, surface sediment, yield, DGT, rice, diversity, paddy soil and reduction.

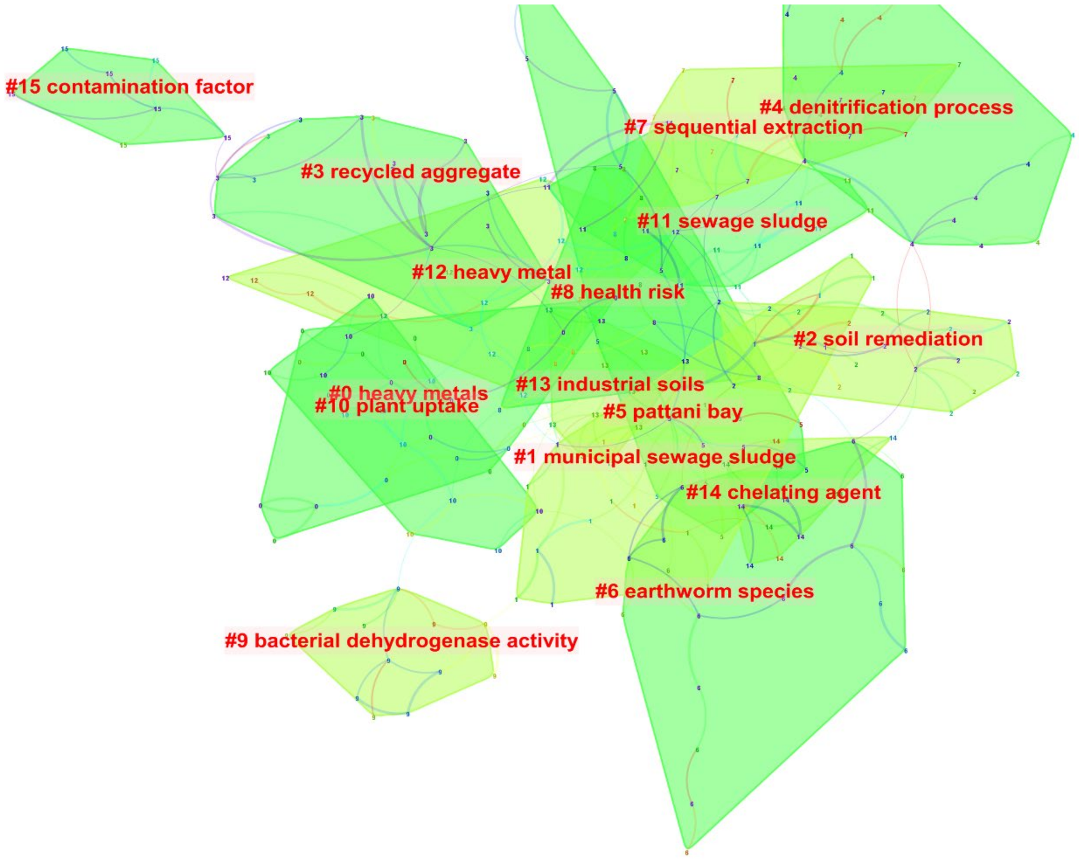

Figure 6 Cluster map of research keywords of the bioavailability of soil heavy metals during 2011-2020. Based on the log-likelihood analysis, Modularity Q is 0.7768, Weighted Mean Silhouette S=0.9113. Each module in the keyword clustering map is a cluster. The larger the module, the greater the number of keywords in the cluster. The text on the module is the name of the cluster, and the largest cluster is marked with #0.

(1) Health risk assessment (#8) of soil heavy metal:

The World Health Organization's definition of a comprehensive risk assessment is "a scientific method for estimating the risks of humans, biological populations and natural resources". It was formulated in 2001 with the assistance of the US Environmental Protection Agency and the World Organization for Economic Cooperation and Development. A comprehensive evaluation framework for health and ecology was established, and suggestions and methods for comprehensive evaluation of health and ecology risks were put forward. The European Union has also formulated technical guidelines for comprehensive health and ecological risk assessment, recommending and guiding EU member states to adopt a new comprehensive assessment system to carry out environmental risk assessment. Since the 1990s, scientists realized the limitations of artificially separating health risks and ecological risks for evaluation, and have begun to explore comprehensive health and ecological risk assessment schemes. This requires an assessment of the ability of heavy metals to transfer from soils to different receptors.

In recent years, there has been a wider realization of the complexity of contaminated sites and the physical and chemical properties of pollutants in site management practices, and the potential risks to human health and the environment are very different. In order to reasonably allocate the limited resources, human, material, and financial resources among the large number of contaminated sites, countries have shifted from the original application of common site cleaning standards, including soil standards for contaminated site evaluation and remediation to risk-based methods and management methods (USEPA, 2007). Studies on bioavailability and the implications of metals in contaminated land have therefore been developed in this period (Li et al., 2016; Zhu et al., 2019; Li et al., 2020b). For example, Li et al (2016) explored the bioavailability prediction of arsenic by using different animal models and found mouse liver or kidney can be used as the target organs of arsenic. Further, Zhu et al (2019) showed the unified bioaccessibility model (UBM) assay as a robust method can predict the relative bioavailability of As, Cd and Pb in contaminated soils by comparing the correlation between mouse models and the UBM gastric phase assay.

(2) Remediation (#2, #14, #3) of soil heavy metal:

There are many types of remediation technologies for contaminated sites, and new remediation technologies appear from time to time (Gong et al., 2018). According to classification methods, remediation technologies can be divided into different two types (Ifon et al., 2019). For example, according to the location of the remediation treatment project, it can be divided into *in-situ* or *ex-situ* remediation approaches. Commonly used *in-situ* remediation techniques include bioremediation and immobilization and stabilization. Commonly used ex-situ remediation techniques are immobilization and stabilization, incineration, thermal desorption, and bioremediation. Bioremediation technology is currently the most commonly used innovative technology. Although many contaminated site remediation technologies have been developed, they all have certain limitations on their scope of application, especially potentially high cost. Therefore, besides of developing the new type of remediation technologies, methods that can accurately evaluate the remediation target value are essential. Currently, the studies of bioavailability of heavy metals in soils have been a hot topic for evaluating the soil remediation target value (Bisht, 2018).

(3) Mechanisms of action (#4, #9, #10, # 6) of soil heavy metals:

During this period, many studies of soil heavy metal bioavailability have been carried out on soil-plant action mechanisms. Low-molecular-weight organic acids play an important role in the morphological changes and bioavailability of heavy metal elements, and have attracted the attention of researchers. The low-molecular-weight organic acids secreted by plant roots participate in soil formation and promote mineral dissolution; change the physical and chemical properties of rhizosphere soil and promote the absorption of nutrients by plants; relieve symptoms of hypoxia and reduce metal and other toxicities. Elemental toxicity to plants, etc., can also form complexes or chelates with metal ions, thereby changing the mobility of metals in the soil (Riaz et al., 2021).

Studies have shown that the root exudates can change the physical and chemical properties of the rhizosphere environment such as pH, Eh and other physical and chemical properties. Root exudates provide nutrition and energy for rhizosphere microorganisms. These microorganisms can convert macromolecular exudates into various low-molecular-weight compounds such as organic acids. In-depth study of the effectiveness of heavy metals in rhizosphere soil is helpful for understanding the environmental behavior of heavy metals in soil plant systems and evaluating the health risks of contaminated soils. In addition, plant roots can also cause the release of metals in different forms (Huang et al., 2014; Zhalnina et al., 2018).

4. Current research status and a forward look

This study is a systematic bibliometric analysis of heavy metal bioavailability research and risk management in contaminated land during 1979-2020. In these 40+ years, the total number of article records of soil heavy metal bioavailability around the world shows an almost exponential upward trend. This indicates that researchers are paying increasing attention to the field. Among 75 studied countries, China is the most productive, which suggests it is the most active in this research area. This might because of the fast development of urbanization after 2000, with the need for evaluation and potential re-development of inner city/industrialized areas. Especially during 2011-2020, the total number of article records from China is at least 5 times that of other countries. According to this development trend and the needs of the current national conditions, it is expected that China will continue to pay attention to research on the bioavailability of soil heavy metals for a long time. It is worth noting that although the number of publications in the US is not the highest, it has the highest centrality, showing that the US has been the most influential country in this field of study. The early urbanization and industrialization development has enabled the US to have a relatively complete management system in soil heavy metal contaminated sites, especially in soil pollution risk assessment and remediation technologies. These experiences can be of benefit to other countries, such as China, where they could be used as a reference.

Through keyword clustering analysis on soil heavy metal bioavailability, the main research hotspots during 2011-2020 were more than before 2011. The main keywords more recently were “health risk assessment”, “remediation”, “mechanisms of action” of soil heavy metals, indicating these issues have become more important to the re-development of contaminated land in recent years. In addition, “rice”, “paddy soil” and “mining area” have the strongest citation bursts, which suggests that agricultural land and industrial land is still a research subject. Therefore, future research will continue to be conducted in agricultural and industrial land, but the challenge is still how to determine accurate estimation of soil heavy metal bioavailability of large-scale land areas.

The keyword clustering analysis of three stages also indicated that studies on the bioavailability of heavy metals in soils are reliant on the development and use of appropriate validated methodologies. In the initial phase (1979-2000), researchers mainly focused on extraction bioavailability measurement methods. Examples include use of single or sequential extractants, EDTA complexing etc. Subsequent approaches, in the slow development phase (2001-2010) and more recently (2011-2020) have seen much more rigorously researched and scientifically based procedures being tested, validated and used widely.

Probably one of the best examples is the Diffusive Gradients in Thin Films (DGT) passive sampler, for which the theory, research applications and practical applications for measuring speciation and bioavailability have been widely explored (Davison and Zhang, 1994; Zhang et al., 1995; Zhang et al., 1998; Harper et al., 2000; Muhammad et al., 2012; Senila et al., 2012). This approach has been most widely tested for soil-plant bioavailability work. More recently, the development of animal based, enzymolysis and simulated GI tract methods have been developed. During the development of science and techniques, many methods of bioavailability of heavy metals in soils have been invented, and they have their own advantages. For example, gastrointestinal simulation assays are commonly used in the risk assessment of contaminated land currently, with more than 10 different gastrointestinal simulation assays so far. However, they can give inconsistent results between methods, and can involve complex procedures. In the current phase of the field's development, these tools and methodologies have gained more acceptance and are being routinely deployed as part of the site evaluation, risk assessment and remediation projects in several countries. They may not yet be part of formal 'approved' methodologies by government agencies, but this may be an outcome in the coming years.

Author contribution

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Yiming Sun. The first draft of the manuscript was written by Yiming Sun, Kevin Jones and Qingbao Gu, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data availability

Data available in supplementary information.

Declarations

The authors declare that they have no conflict of interest.

Ethical statement

This is a review study on the bibliometric analysis of soil heavy metal bioavailability. All data of the research was obtained from Web of Science. The research does not involve any experiments relating to human or animal subjects.

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