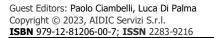


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Systematic Review and Meta-analysis on the Use of Metal Nanoparticles in the Remediation of As and Pb Contaminated Soils

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The use of metallic nanoparticles for environmental applications has been of great interest due to their physical and chemical properties, allowing them to act as immobilizing agents in soils contaminated with heavy metals. The objective of this research was to evaluate the efficiency of metal nanoparticles in the remediation of soils contaminated with arsenic (As) and lead (Pb) by means of a systematic review and meta-analysis. For the systematic review, selected research was collected from Scopus, Web of Science and ScienceDirect databases published between January 2010 and September 2020. For the meta-analysis of the included studies, Review Manager 5.4 software (RevMan) was used and the statistical data were interpreted with the value ranges of Higgins & Green (2011), at a 95% confidence interval. The results showed that metal nanoparticles are good adsorbents and reductants that have higher efficiency at sizes smaller than 100 nm. The studies included for the meta-analysis showed a heterogeneity of 60% and 0% for As and Pb, respectively. Finally, it was concluded that metallic nanoparticles are efficient for the remediation of As and Pb contaminated soils, showing removal percentages higher than 80%.

1. Introduction

Globally, soil contamination is a major concern, as it alters its biodiversity, decreases its organic matter and filtering capacity (Cabello-Torres et al. 2021; Espejo H. et al. 2021; Valdiviezo G. et al. 2023). This is generated by various industrial, agricultural and domestic activities, among others, where the most frequent contaminants found in the soil are persistent organic pollutants, emerging pollutants and heavy metals, which also affect health and food production. It is estimated that 95% of food resources are generated directly and indirectly from soil (FAO, 2018). Soils contaminated by heavy metals are generally caused by urban, industrial and agricultural activities, which could affect food security, human health and ecosystem sustainability (C. Li et al. 2020). These pollutants such as chromium, arsenic, cadmium, copper, mercury, lead, selenium, zinc, nickel and others has increased considerably in the last three decades, estimating that globally more than 10 million sites have been contaminated, representing more than 50% of the degree of heavy metal contamination caused by industrialization and urbanization (Khalid et al. 2017). There are different techniques, so whether it is necessary to implement two or more techniques for decontamination will depend on the needs of the soil, either by chemical depuration or bioremediation where own stimuli intervene (Domènech & Peral 2006). The metallic nanoparticles used in the environment to decontaminate the soil through the remediation technique, possess characteristics such as adsorbent, reductant, oxidant and catalyst to remove various heavy metals, organic pollutants and inorganic pollutants (Liu et al. 2020). The use of nanoparticles has emerged as an effective remediation technology, providing chemical reduction conditions in the contaminated area, thus leading to nano-remediation (Saravanan et al. 2021). Therefore, the present research through a systematic review aimed to evaluate the efficiency of metal nanoparticles in the remediation of soils contaminated with arsenic (As) and lead (Pb).

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2. Materials and methods

2.1 Type of study

This research is applied because it focuses on obtaining data collected and statistical analysis extracted from research articles that lead to answering the established questions and the objectives that were set.

2.2 Sources of information and search strategy

The systematic review of the present research was developed based on the PRISMA methodology and the data found were processed in a meta-analysis (Aquije M. et al., 2021; Acruta P. et al., 2022). The search for information in the databases (Scopus and Web of Science) was carried out systematically according to keywords such as "nanoparticles", "soils", "remediation", "removal" and "arsenic", using the Boolean operators "AND" and "OR", and in the English language. In addition, all included studies were verified to be no older than 10 years as of 2020.

2.3 Inclusion and exclusion criteria for scientific investigations

Initially, research that did not contain the keywords in the title, abstract and keywords were excluded. Duplicate investigations were eliminated according to the title. Subsequently, all research was downloaded for review according to the inclusion criteria. Finally, all selected researches were evaluated by reading according to the inclusion and exclusion criteria described as follows:

- 1. Studies showing the efficacy of using metallic NPs or combined NPs using application doses were included.
- 2. Studies that worked with soils contaminated with heavy metals such as As and Pb were included.
- 3. Studies that worked on the removal of As and Pb in other media (water, plants, etc.) were excluded.
- 4. Studies with insufficient data were excluded.

2.4 Data meta-analysis

Data analysis was performed using the Review Manager program (RevMan 5.4). This statistical software allows an adequate review of the included studies. In addition, the Microsoft Excel program was used to generate tables with the included research.

The meta-analysis was performed using dichotomous data, which were presented and compared with the odds ratio, with the 95% confidence interval. For the interpretation of the statistical data, the following ranges of values were used (Higgins & Green 2011).

3. Results and discussion

3.1 Search and selection of the studies

27 studies were included and 1356 were excluded, out of a total of 1383 investigations. In the first phase, studies were excluded if they did not include the search keywords in the title, abstract and keywords of the paper, i.e. they were not related to the topic under study. In the second phase, duplicate studies were excluded. In the last exclusion phase, the following factors were taken into account: use of nanoparticles in water, non-metallic nanoparticles and insufficient data.

3.2. Description of the studies

Table 1 shows the main characteristics of the 27 studies included for remediation of Arsenic (As) and Lead (Pb), published between January 2010 and September 2020. Two investigations (Cao et al. 2020 and Z. Li et al. 2020) used metal nanoparticles to remediate both metals (As and Pb).

3.3 Meta-analysis

For the meta-analysis, all the researches that worked with a minimum of two doses of metallic nanoparticles for the remediation of As and Pb contaminated soils were selected. In some studies, the application doses had to be converted to percentages, and in others more than one type of NPs were considered. The investigations included are shown in Table 2 and Table 3.

From Table 2, the 2 studies that had the highest percentage of remediation were from Baragaño et al. (2020b) (99.80%) and Azari & Bostani (2017), using nGeothite and Fe (nZVI), NPs, respectively. However, not all had the highest percentage expected, the study by Z. Li et al. (2020) had the lowest value (31%), using Z-nZVI NPs. Figure 1 and Figure 2 show the studies included in the meta-analysis on the remediation percentages of As and Pb contaminated soils in each test.

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No.	Type of pollutant	Type of NPs	Authors		
1	As	Magnetite	(Baragaño et al. 2020a)		
2	٨٥	nZVI	— (Baragaño et al. 2020b)		
	As —	nGeothite			
3	As and Pb	Nano-silica (RNS-SFe)	(Cao et al. 2020)		
4	As	nZVI	(Fan et al. 2020)		
5	As and Pb	Z-nZVI	(Z. Li et al. 2020)		
6	As	Iron oxide	(Su et al. 2020)		
7	As	nZVI	(Gil-Díaz et al. 2019)		
8	As	starch-stabilized Fe/Cu	(Babaee, Mulligan & Rahaman 2018)		
9	As	Fe (nZVI)	(Azari & Bostani 2017)		
		nZVI - RNIP	(Gil-Díaz et al. 2017)		
10	As	nZVI - RNIP-D			
		Nano hierro (25S)			
11	As	Hematite	(Mansouri, Golchin & Neyestani 2017)		
12	As	Fe (nZVI)	(Gil-Díaz et al. 2014)		
13	As	Magnetite (Fe3O4)	(Liang & Zhao 2014)		
14	As	stabilized Fe-Mn oxide	(An & Zhao 2012)		
45		Magnetite	(Shipley, Engates & Guettner		
15		Hematite	2011)		
		ZVI			
16	As	FeS	(Zhang et al. 2010)		
		Fe3O4			
17	Pb	NANOFER 25S iron (nZVI)	(Fajardo et al. 2020)		
18	Pb	Fe3O4@C-COOH	(Ma et al. 2020)		
19	Pb	ZnO	(Seleiman et al. 2020)		
20	Pb	nZVI	(Vasarevičius, Danila & Januševičius 2020)		
21	Pb —	nZVI Fe3O4	(Fajardo et al. 2019)		
22	Pb	nZVI	(Zhou et al. 2019)		
23	Pb	Maghemite	(Hughes et al. 2018)		
24	Pb	nZVI	(Okuo et al. 2018)		
25	Pb	nZVI	(Mar Gil-Díaz et al. 2014)		
		MnO _{1.26} amorphous	(
26	Pb	γ-Fe2O3	(Michálková et al. 2014)		
		Fe3O4			
27	Pb	nZVI	(Wang et al. 2014)		

Table 1: Characteristics of selected studies in the systematic review on the use of metal nanoparticles in the remediation of arsenic and lead-contaminated soils

Table 2: Research included for the meta-analysis of metal nanoparticle concentrations in the remediation of arsenic contaminated soils.

Arsenic (As)								
No.	Type of NPs	Dose of	NPs (%)	Removal of As (%)		Authors		
NO.		Dose 1	Dose 2	1	2	Autions		
1	Magnetite	2	5	70	92.3	(Baragaño et al. 2020a)		
2	nZVI	5	10	96.2	97.6	(Baragaño et al. 2020b)		
3	nGeothite	2	5	99,7	99.8	(Baragaño et al. 2020b)		
4	Nano-sílica (RNS-SFe)	3	6	80.1	89.2	(Cao et al. 2020)		
5	Z-nZVI	1	3	14.3	31	(Z. Li et al. 2020)		
6	starch-stabilized Fe/Cu	0.01	0.04	80.4	92.2	(Babaee et al. 2018)		
7	Fe (nZVI)	0.5	2.5	98.9	98.9	(Azari & Bostani 2017)		
8	Nano Iron (25S)	5	10	87.79	92.38	(Gil-Díaz et al. 2017)		
9	nZVI - RNIP	5	10	86.23	93.94	(Gil-Díaz et al. 2017)		
10	nZVI - RNIP-D	5	10	89.35	93.94	(Gil-Díaz et al. 2017)		
11	Hematite	0.1	0.2	51.83	65.31	(Mansouri et al. 2017)		
12	nZVI	1	10	49	85	(Gil-Díaz et al. 2014)		

	Dose	1	Dose 2		Odds Ratio		Odds Ratio
Study or Subgroup	Events	Total	Events	Total	Weight	M-H, Fixed, 95% CI	M-H, Fixed, 95% Cl
Azari & Bostani, 2017	99	198	99	198	8.9%	1.00 [0.67, 1.48]	
Babaee et al., 2018	80	172	92	172	8.9%	0.76 [0.49, 1.16]	
Baragaño et al., 2020a	70	162	92	162	9.4%	0.58 [0.37, 0.90]	
Baragaño et al., 2020b-1	96	194	98	194	8.9%	0.96 [0.64, 1.43]	-+-
Baragaño et al., 2020b-2	100	200	100	200	9.0%	1.00 [0.68, 1.48]	- + -
Cao et al., 2020	80	170	90	170	8.6%	0.79 [0.52, 1.21]	
Gil-Díaz et al., 2014	49	134	85	134	9.7%	0.33 [0.20, 0.55]	
Gil-Díaz et al., 2017-1	88	180	92	180	8.5%	0.91 [0.61, 1.38]	
Gil-Díaz et al., 2017-2	86	180	94	180	8.9%	0.84 [0.55, 1.27]	
Gil-Díaz et al., 2017-3	89	183	94	183	8.7%	0.90 [0.59, 1.35]	
Mansouri et al., 2017	52	117	65	117	6.5%	0.64 [0.38, 1.07]	
Z. Li et al., 2020	14	45	31	45	3.9%	0.20 [0.08, 0.50]	
Total (95% CI)		1935		1935	100.0%	0.77 [0.68, 0.87]	•
Total events	903		1032				
Heterogeneity: Chi ² = 27.55, df = 11 (P = 0.004); I ² = 60%							
Test for overall effect: Z = 4.13 (P < 0.0001)							0.01 0.1 1 10 100 Dose 1 Dose 2
	•						Dose i Dose z

Figure 1: Meta-analysis of metal nanoparticle concentrations in the remediation of arsenic-contaminated soils

According to the Odds Ratio effect measure in Figure 1, a value of 0.77 was obtained, which represents an increase in the percentage of remediation of As-contaminated soils. Dose 2 is favored with the Odds Ratio effect measure unlike dose 1. The included investigations showed moderate heterogeneity ($I^2 = 60\%$), which represents that the results of the investigations and the effects are moderately homogeneous.

Table 3: Research included for the meta-analysis of metal nanoparticle concentrations in the remediation of lead contaminated soils.

Lead (Pb)							
No.	Type of NPs	Dose of	NPs (%)	Removal of Pb (%)		Authors	
		Dose 1	Dose 2	1	2	Authors	
1	Nano-silica (RNS-SFe)	3	6	97.1	99.6	(Cao et al. 2020)	
2	Z-nZVI	1	3	92.20	96.8	(Z. Li et al. 2020)	
3	nZVI	0.7	1.05	52.3	63.6	(Vasarevičius et al. 2020)	
4	nZVI	2.5	5	93.8	97.3	(Fajardo et al. 2019)	
5	nZVI	5	10	90.8	95.6	(Zhou et al. 2019)	
6	MnO _{1.26} amorphous	1	2	100	100	(Michálková et al. 2014)	
7	γ-Fe2O3	1	2	38.9	27.8	(Michálková et al. 2014)	
8	Fe3O4	1	2	44.4	47.2	(Michálková et al. 2014)	
9	nZVI	0.2	0.3	82	81	(Wang et al. 2014)	

Table 3 showed that, the 2 studies that had the highest percentage of remediation were from Michálková et al. (2014) (100%) and Cao et al. (2020) (99.60%), using $MnO_{1.26}$ amorphous and Nano-silica (RNS-SFe) NPs, respectively. However, not all had the highest percentage expected, the study by Michálková et al. (2014) had the lowest value (27.8%), using γ -Fe2O3.

	Dose 1 Dose 2		2	Odds Ratio		Odds Ratio	
Study or Subgroup	Events	Total	Events	Total	Weight	M-H, Fixed, 95% Cl	M-H, Fixed, 95% Cl
Cao et al., 2020	97	197	100	197	14.1%	0.94 [0.63, 1.40]	
Fajardo et al., 2019	94	191	97	191	13.7%	0.94 [0.63, 1.40]	-+-
G. Wang et al., 2014	82	163	81	163	11.2%	1.02 [0.66, 1.58]	+
Michálková et al., 2014-1	100	200	100	200	13.9%	1.00 [0.68, 1.48]	+
Michálková et al., 2014-2	39	67	28	67	3.2%	1.94 [0.98, 3.85]	
Michálková et al., 2014-3	44	91	47	91	6.7%	0.88 [0.49, 1.57]	
Vasarevicius et al., 2020	52	116	64	116	9.8%	0.66 [0.39, 1.11]	
Z. Li et al., 2020	92	189	97	189	13.8%	0.90 [0.60, 1.35]	
Zhou et al., 2019	91	187	96	187	13.7%	0.90 [0.60, 1.35]	-
Total (95% CI)		1401		1401	100.0%	0.95 [0.82, 1.10]	•
Total events	691		710				
Heterogeneity: Chi ² = 6.46, df = 8 (P = 0.60); l ² = 0%							
Test for overall effect: Z = 0.72 (P = 0.47)							0.01 0.1 1 10 100 Dose 1 Dose 2

Figure 2: Meta-analysis of metal nanoparticle concentrations in the remediation of lead-contaminated soils

According to the Odds Ratio effect measure in Figure 2, a value of 0.95 was obtained, which represents an increase in the percentage of remediation of Pb contaminated soils. Dose 2 is favored with the Odds Ratio effect measure unlike dose 1. The included investigations showed no heterogeneity ($I^2 = 0\%$), which represents that the results of the investigations and the effects are significantly homogeneous.

4. Conclusions

The systematic review and meta-analysis on the use of metal nanoparticles in the remediation of As and Pb contaminated soils included 27 investigations, 14 for As, 11 for Pb and 2 for both metals. The values of the average maximum removal efficiencies of As and Pb were 84.4% and 81.2%, respectively. The results showed that the application dose of the nanoparticles in As and Pb contaminated soils depends on the initial concentration of the contaminant in relation to the contact time. In addition, it is worth noting the type of nanoparticles to use.

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