Selenium Removal from Water and Wastewater by Different Technologies: A Systematic Review

*Abdolmajid Fadaei [1], Abdollah Mohammadian-Hafshejani [2]

1. *Department of Environmental Health Engineering, School of Health, Shahrekord University of Medical Sciences, Shahrekord, Iran*
2. *Department of Epidemiology and Biostatistics, School of Health, Shahrekord University of Medical Sciences, Shahrekord, Iran*

*Corresponding Author: Email: ali2fadae@yahoo.com

Abstract

Background: Selenium (Se) is an essential element playing a vital role in the metabolism of organisms. Se can generally be discharged in the potable water through natural and anthropogenic activities. Both excess and shortage of Se can cause significant adverse health effects in humans. Excess values of se may **stimulate** toxicity, leading to selenosis and alkali disease in humans and grazing animals, respectively.

Methods: A review search was systematically carried out from the databases Embase, PubMed/MEDLINE, Scopus, PubMed Central (PMC), Google Scholar, as well as medRxiv by using the following keywords: "waste water", "bioremediation", "selenium removal", "adsorption", and "drinking water". This study provides a review of the recent literature covering the period between 2011 and 2021. After screening the full text of the articles, 27 papers were enrolled. This study reviews the reported techniques for Se removal from water and wastewater, including adsorption, biological treatment, microbial reduction, bioreactors, fungal bioreactor, algal treatment, phytoremediation, and photocatalysis.

Results: Biological and bioremediation techniques, such as microbial reduction, biotransformation, and fluidized bed reactor have removal efficiency about 100%. The highest Se concentration of 15-7600 μg/L was achieved in ground waters in Ethiopia and the lowest level of 0.07 μg /L in Finland.

Conclusion: The combination of biological treatment with chemical or physical technologies is envisaged to optimize se elimination and to ensure ecological protection and human health safety.

Keywords: Adsorption; Bioremediation; Selenium (Se) removal; Water and wastewater

Introduction

Release of municipal and industrial wastewater containing hazardous materials and heavy metals into the environment can pose serious threats to humans, animals, plants and urban ecosystems (1). Selenium (Se) is an essential micro-nutrient element at low levels, but toxic when an excessive amount is consumed. The micronutrient benefit of se for human physiological functions is be-

tween the concentration of 63 and 135 μg/L, above this limit, Se is considered to be toxic (2). Symptoms of se toxicity were noted to have occurred at a daily intake of more than 800 μg /d, while chronic toxicity results in selenosis. Typical dietary intake of se in the United States is between 80–120 μg/d but varies significantly across other world regions (3). However, the National

Academy of Sciences Institute of Medicine has set 400 μg/d of Se as the tolerable upper safe limit(4). Se generally exists in different oxidation states i.e. Se^{6+} (selenate), Se^{4+} (selenite), Se^0 (elemental Se), Se^{2-} (selenide), volatile methylated selenides (dimethyl selenide and dimethyl diselenide), and organic forms (selenocysteine and selenomethionine) (5). In water and wastewater, se generally exists as soluble Se oxyanions, selenite ($SeO3^{2-}$) and selenate ($SeO4^{2-}$), corresponding to Se (IV) and Se(VI), respectively. World consumption of Se included electronics (9.1%), pigments (10.3%), chemicals (15.2%), agriculture and other uses (15.7%), metallurgy (23.9%), and glass manufacturing (25.7%) (6).

The major sources of Se are oil refineries, mining of phosphates and wastewater sludge (1). The adverse effects of Se could not be ignored. Se effects on human health appear as dysfunction of the endocrine system, lack of mental alertness, gastrointestinal disturbances, impairment of natural killer cells activity, cardiovascular diseases, selenosis, deformation and loss of nails, and liver damage. Se effects on plants included alteration of protein properties, and reduction of plant biomass. Se also adversely affects animals. Alkali disease and blind staggers are two main syndromes caused by excessive intake of Se in animals (7-9). Therefore, it is very vital to choose a suitable and efficient method for Se removal from water and wastewater.

There are several treatment techniques that can be employed to remove Se from contaminated water and wastewater, including adsorption, bioremediation, photocatalysis, electrocoagulation, advanced oxidation processes (AOPs), reverse osmosis (RO), electrochemical methods, ion-exchange, centrifugation, chemical precipitation, and coagulation (10). Recently, bioremediation through adsorption has been widely investigated as a cost effective alternative because of the possibility of sustainable implementation, including the regeneration and reuse of the adsorbing media (11).

The present study focuses on the evaluation of Se removal by applying several treatment techniques to water and wastewater with emphasis on bioremediation.

Methods

The principal focus of this review is on the Se removal methods and processes. Databases including Google Scholar, Science Direct, PubMed/ MEDLINE, Cochrane, Scopus, medRxiv, and Web of Science were searched to retrieve several papers on the topic. Keywords like "waste water", "bioremediation", "selenium removal", "adsorption", "drinking water", "water treatment", and "ground water" were added to the above mentioned methods to retrieve appropriate papers. As shown in Fig. 1, 91 peer reviewed publications were accessed based on the relevance of titles to the research. These articles were further screened to 56 after reading through their abstracts. Following full text screening of the articles, 27 of them were used for this review, excluding the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) reference (12).

Google search
Search items: waste water, bioremediation, Se removal, adsorption, drinking water
Relevant papers screened from titles, N=91
Papers screened from abstract, papers excluded at this level, N=35
Papers included after screening from abstract level, N=56
Papers screened from full text, papers excluded at this level, N=19
Papers included in this study after screening from full text, N=27

Fig. 1: Chart presentation of the review process

This study provides a review of the recent literature covering the period between 2011 and 2021. The inclusion criteria were access to the original article, English language, and using the type of aqueous solution investigated in this study such as surface water, sea water, ground water, domestic wastewater, mining wastewater, and industrial wastewater were investigated in this study. The exclusion criteria were **unavailability of full text** of the article, review studies, book reviews, guidelines, protocols, letters-to-editors, articles submitted to conferences, theses, white papers, etc.

Statistical analysis

The effect size of the relationship between bioremediation/biological treatment and Se elimination was reported with the 95% confidence interval (CI) for odds ratios (OR). A random-effects meta-analysis model was used to calculate the overall summary estimates. Graphically, the illustration of the individual OR and summary estimates was done in the form of forest plots. Heterogeneity among studies was tested by Cochran's Q test (reported with a χ^2 value and P-value, with $P<0.1$ considered as the significance level) and the I2 statistics. I2 with values of 25%, 50%, and 75% demonstrated low, moderate, and high levels of heterogeneity, respectively. Publication bias was assessed using Begg's tests. All statistical analyses were performed using Stata 14.0 software (Stata LLC, College Station, TX, USA). $P<0.05$ was considered as the level of significance.

Results and Discussion

The reviewed articles had used various methods, including adsorption (12), biological and bioremidation (11) technique (i.e. biological treatment, microbial reduction, bioreactors, ABMet@ system, fungal bioreactor, algal treatment, phytoremediation), and photocatalysis process (4).

Adsorption

Adsorption is one of the techniques recommended for the elimination of hazardous elements existing in water and wastewater.

The highest capacity of bimetallic diatom composite adsorbent for Se removal was 227 mg/g, and the lowest removal capacity was 0.009 mg/g for activated alumina adsorbent (Fig.2 and Table 1).
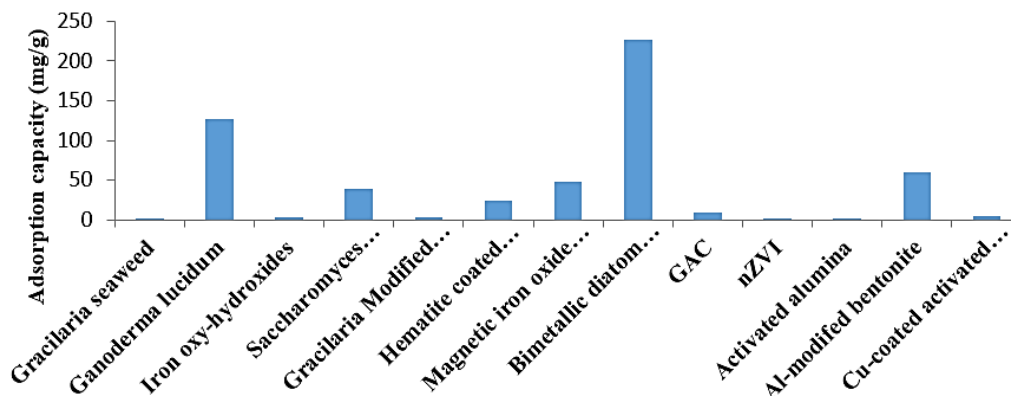

Fig. 2: Comparison of the adsorption capacities of various adsorbents

Table 1: Adsorption method used for Se removal from aqueous phase

Type of adsorbent	Type of media	Reaction time	Species of Se	pH	Removal efficiency	Ref
Gracilaria seaweed	Wastewater	4 h	Se(VI)	2.5 to 8	2.72 mg/g	(13)
Ganoderma lucidum	Artificial water	90 min	Se(VI)	5	126.99 mg/ g	(14)
Iron oxy-hydroxides	Natural waters	-	Se(VI) and Se(IV)	2to9	10 μg Se(VI)/g)and 4.3 mg Se(IV)/g.	(15)
Saccharomyces cerevisiae	Artificial water	30 min	Se(IV)	5	39.0 mg /g (96.1%)	(1)
Gracilaria Modified Biochar	Wastewater	-	Se(VI)	-	3.8 mg/g (98%)	(16)
Hematite coated magnetic nanoparticle	Water	120 min	Se(IV)	4 to 9	25.0 mg/g (97%)	(17)
Magnetic iron oxide nanoparticles	Contaminated water	24 h	Se(IV)	3 and 5	48 mg/g(>99%)	(18)
Bimetallic diatom composite	Artificial water	24 h	Se(IV)	8	227 mg/g	(19)
GAC and nZVI	Artificial wastewater	3 h	Organic Se (Selenomethionine and selenocysteine)	4 to9	GAC: selenocysteine10 mg/g (96.1%) and Selenomethionine 18.9 mg/g (86.7%) nZVI: selenocysteine0.59 mg/g (39.4%) and Selenomethionine < 1.1%	(20)
Activated alumina	Drinking water	70-120 min	Se(VI) and Se(IV)	4 to 9	9.02 μg/ g(80%) and 5.38 μg/ g(72%)	(21)
Al-modified bentonite	Artificial water	-	Se(IV)	3	60.1 mg/ g	(22)
Cu-coated activated carbon	Artificial water	4 h	Se(VI)	6	4.48 mg/g(88%)	(23)

The maximum adsorption capacity of fly ash zeolites for Se (IV) removal from an aqueous solution reported to be 3.93-4.16 mg/g at pH 2, with adsorbent content of 5.0 g/L, and initial concentration of 10 mg/L (24). The adsorption capacity of graphene oxide nanocomposite for Se(IV) removal from contaminated water was 1.62 mg/g at 25°C and pH 4-8 (25). The Se(IV) and Se(VI) removal efficiency by nanocrystalline aluminum oxide(nanocrystalline aluminum oxide) was 97% and 92%, respectively at pH 6.5-7.3, with initial concentration of 10 mg/L(26). Tang et al. reported the Se(VI) removal efficiency by zero-valent iron to be nearly 100% within 16 h, with an addition of 0.2 mM of Fe^{+2} (27). The efficiency of nanoscale zerovalent iron for Se removal from water was 90% at pH 3, with100 mg of the adsorbent (28). The removal efficiency of Se from wastewater by GAC was 78% at pH 7, reaction time 4 h, and 2.5 g /L of the adsorbent (29). Selenite removal efficiency reached above 90% after 6 h adsorption for the initial Se level of 2 mg/L and equilibrium was attained after 48 h(30). The removal efficiency of Se was found from water by oxide/hydroxide nanoparticles to be 95%-98% at pH 4, and 0.01 equilibrium concentration (31). The removal efficiency of Se from aqueous solution was 41%-78% at pH 5 using Al/Si and Fe/Si co-precipitates (32). The removal efficiency of Se from aqueous solution by iron oxide was 100% within 6 h, with an initial concentration of 1 mg/L at pH 6, adsorbent content of 25 mg, and mixing speed of 150 rpm (33). The adsorption capacity of natural goethite for Se(IV) removal from water was found to be 7.740 mg/g within 72 h with an initial level of 150 μg/L at pH 4, and adsorbent content of 0.333 g/L (34).

Biological and bioremediation technique

Biological treatment has emerged over recent years as a leading technology for Se removal from water and wastewater.

Table 2 shows bioremediation and biological treatment for elimination of Se from water and wastewater.

In biological systems, anaerobic and aerobic bacteria, algae, fungi, and plants are capable of cata-lyzing the Se removal under environmental conditions (35). Bioremediation combines the Se treatment with the potential to remove, recover, and reuse Se in the form of Se biominerals. Microbial reduction is a proven technique for converting soluble Se oxyanions (($SeO4^{2-}$ and $SeO3^{2-}$) to insoluble (Se^0) forms to eliminate the pollution from the water (9). The removal efficiency of Se from aqueous solution by using bio-reduction was 88%, at pH 6 to 7, with an initial level of 11.6 mM at 30 °C (36). A novel solution was investigated by using a novel bacterial aerobic selenite reductase (*Alishewanella sp.*) for Se (IV) removal (37). *Chlorella vulgaris* was used for treatment (by biological uptake) of Se(IV) and Se(VI). The high elimination efficiency of 89% was observed for the initial Se level of 1580 μg/L(38). Another study was carried out on the use of *C. vulgaris* for Se elimination (by biological uptake) under varying conditions of initial Se level, algal density, temperature and pH. This study has also reported a removal efficiency of about 90% for Se (38). The alga Chlorella vulgaris removed 96% of Se supplied as Se oxyanions (1.58 mg/L) from the wetland water column within 72 h (39). The removal efficiency of se by bioremediation (algal-bacterial biofilm and biofertilizer) was about 65% within 6 d (11). The removal efficiency of Se from soil leachate by plants, such as Lemna minor and Egeria densa was 77% and 60%, respectively at pH 7, initial level of 74 μg/L within7d(40). The removal efficiency of Se (IV) and Se(VI) from environment by fungi(i.e. *Acremonium strictum, Alternaria alternate, Paraconiothyrium sporulosum, Plectosphaerella cucumerina, Pyrenochaeta* sp*, and Stagonospora* sp) was 85%–93% and 20%–30%, respectively, with initial concentration of 0.01 mM, within 10 d (41). The removal efficiency of Se from aqueous solutions by biotransformation was 40.40% after 21 d, with initial level 140-1400 μg/L (42). The adsorption capacity of Se(IV) and Se(VI) from aqueous solution by *Lactobacillus plantarum* was reported to be 700 μg/g, at pH 3-9 and 20–50º C (43). The adsorption capacity of Se(IV) from aqueous solution by *Cladophora hutchinsiae* was 74.9 mg/g, at pH 5 and 20º C (44). The adsorption capacity of Se(IV) from

aqueous solution by *Aspergillus* sp. *J2* was 5.67 mg/g, at pH 4.0-10.7 and temperature 38° C (45). One study used bacterium *Pseudomonas stutzeri NT-I* for bioremediation of selenium-containing refinery wastewater in two different bioreactors (46). The bio-reduction of selenate was reported to elemental Se using aerobic methanotrophs (47). Kieliszek et al. reported the Se removal from environment by yeast cells (*Saccharomyces cerevisiae, Candida utilis,* and *Yarrowia lipolytica*)(48). The removal of selenate was reported from aqueous solution using ethanol-fed fluid-

ized bed reactor via redox potential at ORP-360 mV and pH of 4.0 (49). The efficiency of bioelectrochemical barrier for selenate removal from artificial wastewater was reported to be about 99.5%-99.8% with an initial level of 70 mg/L and voltage of 1.4 V. The selenite removal was reported from activated sludge by bioremediation (*Citrobacter Providencia* sp*.,* and *Citrobacter* sp)(50). The Se removal was investigated from contaminated waters and wastewaters by biomineralization (9).

Table 2: Bioremediation and biological treatment for elimination of Se from water and wastewater

Type of organism	Selenium species	Temperature	pH	Removal efficiency	Ref
Macroalgae (*Oedogonium* sp)	Se(VI)	20 ° C	4	84.14%	(51)
Volatilization(*Pseudomonas stutzeri*)	Se(VI)	38° C	9	82%	(52)
Phytoremediation(Cattail (CT; Typha angustifolia L and muskgrass)	Se(VI) and Se(IV)	-	-	75% and 74%	(53)
Bioreduction (anaerobic granular sludge)	Se(IV)	30°C	7.3	above 92%	
Burkholderia strains	Se(IV)	27 °C	7	75%	(54)
Biotransformation(*Stenotrophomonas maltophilia* SeITE0)	Se(IV)	27 °C	8	100%	(54)
Rhodocyclaceae (*Azospira oryzae* and *Rhizobium* s)	Se(VI)	-	7	99%	(55)
Fungal-pellet bioreactor (*Phanerochaete chrysosporium*)	Se(VI)	30 °C	4.5	70%	(56)
Inverse fluidized bed bioreactor	Se(IV)	30 °C	7	98%	(57)
Fluidized bed reactor	Se(IV) and Se(VI)	-	4.0 -5.0	100%	(58)
Upflow anaerobic sludge blanket (UASB)	Se(IV)	55 °C	7.3	94.4%	(59)
Biological(Cronobacter)	Se(IV)	25 °C	7.5	100%	(60)

Relationship of bioremediation/biological treatment and Se elimination

Considering the adjusted OR from each study in the meta-analysis, the OR of Se elimination with low levels is equal to 66.95% (95% CI=54.29–79.62, $P\leq0.001$) as compared with the group with

high levels of bioremediation/biological treatment, (Fig. 3). There was significant heterogeneity identified in the results during the meta-analysis (chi^2 = 64.94, df = 11, $P\leq0.001$, I2=83.1%).
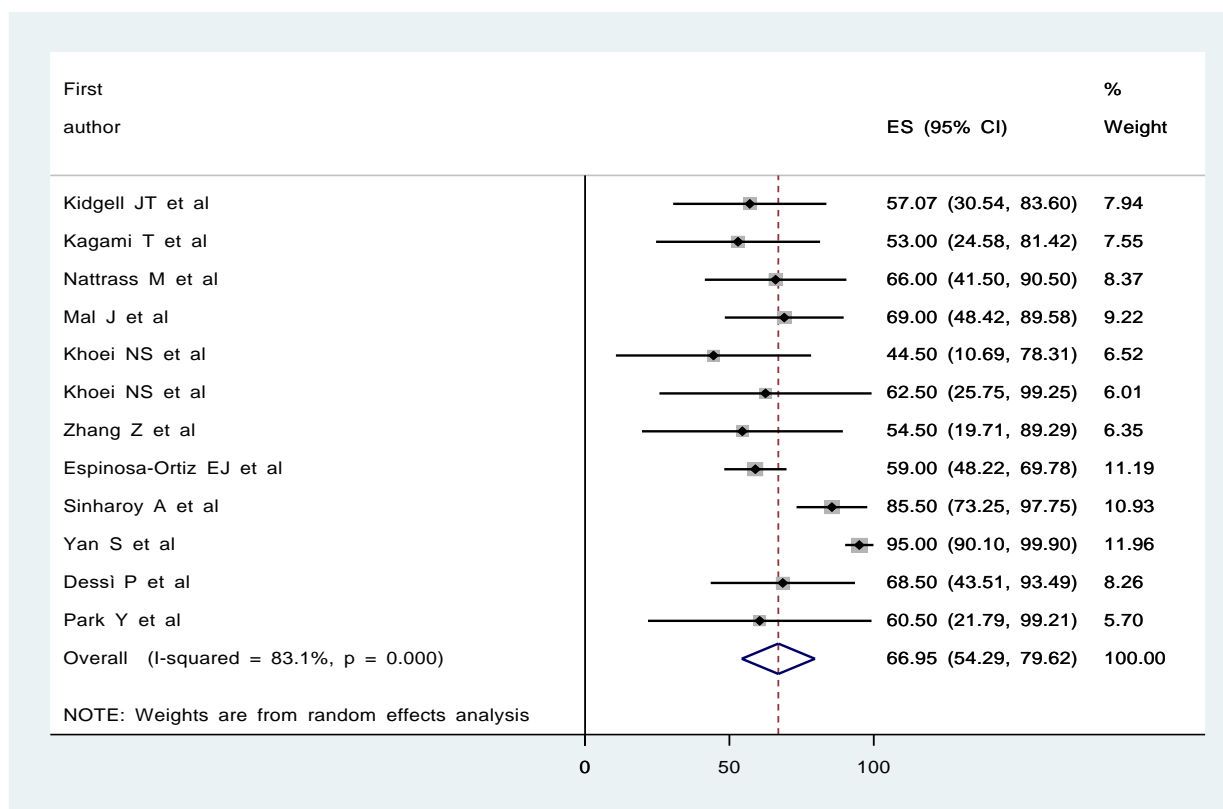
Fig. 3: The forest plot displays the elimination of Se (%) by bioremediation /biological treatment based on the random effects model (CI = 95%) of the average effect size

Evaluation of publication bias related to the relationship of bioremediation/biological treatment and elimination of Se

Evidence of publication bias was suspected upon examining the reported association between the relationship of bioremediation/biological treatment and elimination of Se. Therefore, the statistical tests were undertaken to evaluate potential publication bias in the reported studies. Begg's test ($P=0.631$) produced no statistically significant results. Therefore, published studies on the relationship of bioremediation/biological treatment and Se elimination are not significantly associated with publication bias, which cannot affect the final results of the meta-analysis.

Photocatalysis process

Recently, photocatalysis has gained wide attention in water and wastewater treatment due to its energy effectiveness and strong redox ability. In addition, it is also advantageous for the recovery of heavy metals from contaminated environments (61). Table 3 shows photocatalytic process for elimination of Se from aqueous phase.

A study demonstrated the photocatalysis (UV_A/TiO_2) for selenate removal from water with an initial level of 0.06 mM, at pH<5 in the presence of 300 mg/L formic acid (62). The removal efficiency of Se from mine water by photocatalysis was reported to be 99.6%, with TiO_2 dose of 0.2 g/L, at pH 3, UV intensity 11.03 mW/cm² (UV/TiO_2)(63). The use of photocatalytic reduction for Se(IV) removal was reported from an aqueous solution, at pH 3.5, with TiO_2 dose of 1.5 g/L, initial level of 80 mg/L in the presence of 300 mg/L formic acid (64). The use of photocatalytic reduction for Se(IV) and Se(VI) was demonstrated from aqueous solution, at pH 3.5, with TiO_2 dose of 1.1 g/L, initial level of 20 mg/L in the presence of 300 mg/L formic acid

within 120 min (65). The removal efficiency of total Se from mine water by using photocatalysis (UV/TiO$_2$) was reported 95.3%, at pH 7, with TiO$_2$ dose of 1.0 g/L, 15 W UV lamp, within 360 min (66).

Table 3: Photocatalytic process for elimination of Se from aqueous phase

Type of technique	Environment	Removal efficiency	comment	Ref
Photocatalytic	Synthetic wastewater	86.7%	Se(VI) and Se(IV) 10 mg/L, pH 4, natural log model	(61)
Solar photocatalytic	Aqueous environment	67%	Se(VI) Se(IV) and selenocyanate10 to 20 mg/L, pH 4, TiO2 =0.25 g/L, at 2 h	(67)
Photocatalytic	Aqueous phase	98%	Se(VI) and Se(IV) 20 mg/L, pH 4,reaction time 3 h, DO= 7 mg/L,T= 26∘C	(68)
Photocatalytic	Wastewater	About 100%	Se(VI) 0.5 mg/L, TiO2 =0.2g/L, reaction time1.5 h, HCOOH 2.0 mM	(69)

Relationship between photocatalysis process and Se elimination

Considering the adjusted OR from each study in the meta-analysis, compared to the group with high levels of photocatalysis process, the OR of Se elimination with low levels is equal to 72.21% (95% CI=49.34-95.08, $P \leq 0.001$) (Fig. 4). A significant heterogeneity was identified in the results during the meta-analysis (chi^2=9.54, df=3, $P \leq 0.001$, I2=68.6%).
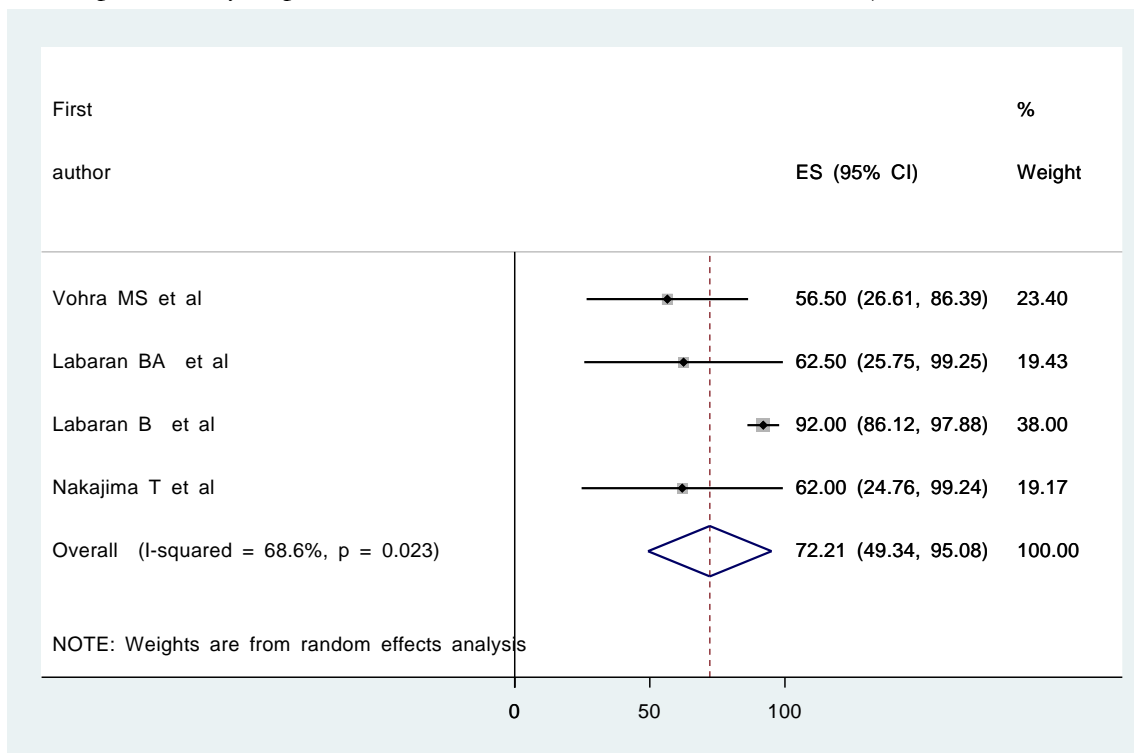

Fig. 4: The forest plot displaying the elimination of Se (%) by photocatalytic process based on the random effects model (CI = 95%) of the average effect size

Evaluation of publication bias related to the relationship of the photocatalysis process and elimination of Se

Evidence of publication bias was suspected upon examining the reported association between the relationship of the photocatalysis process and Se elimination. For this purpose, the statistical tests were undertaken to evaluate potential publication bias in the reported studies. Begg's test (*P*-value=0.734) produced no statistically significant results. Therefore, published studies on the relationship of the photocatalysis process and Se elimination are not significantly associated with publication bias, which cannot affect the final results of the meta-analysis.

Se from global perspective

Regarding ground waters, the highest Se concentration of 15to7600 µg/L was achieved in Ethiopia and the lowest concentration of 0.07 µg/L was found in Finland (Table 4), while in surface waters, the highest Se concentration of 165 to 311µg/L was found in Canada, and the lowest concentration of 0.21µg/L in Brazil (Table 4).

The WHO and EPA recommend that Se in potable water should not exceed 40 and 50 µg/L, respectively (Table 5).

Table 4: Selenium concentration in surface and ground waters from several countries

Regions/countries	Water sources	Selenium concentration	Ref
India (Chandigarh)	Groundwater	0.9 µg/L	(70)
Nigeria	Groundwater	7.33 - 46.3 µg/L	(71)
India(Chennai)	Groundwater	0.15–0.43 µg/L	(70)
Jordan	Groundwater	0.09 - 0.74 µg/L	(72)
Ethiopia	Groundwater	15–7600 µg/l	(73)
Finland	groundwater	0.07 µg/L	(74)
China(Enshi)	groundwater	0.17 µg/L	(74)
Canada(Saskatchewan River)	Surface water	165–311 µg/L	(75)
Australia (Lake Macquarie)	Surface water	50-300 µg/L	(35)
Belgium(Scheldt River)	Surface water	0.23–1.78 µg/L	(76)
Brazil(Amazon River)	Surface water	0.21 µg/L	(76)

Table 5: Summary of guidelines for Se concentration in drinking water

Country/Organization	Concentration (µg/L)	Ref
European Union	10	(77)
World Health Organization	40	(78)
United State Environmental Protection Agency	50	(79)
Canada	10	(6)
Australian	10	(80)
United States: Oregon	120	(81)
South Africa	20	(82)
New Zealand	10	(74)

Generally, Se concentration in normal water is less than 1 µg/L, while in seawater its content usually ranges from 0.1 to 0.35 µg/L. Global concentration of Se in river water has been reported to be 0.02 to 0.5 µg/L with an average value of 0.07 µg/L. The Se concentration in sur-

face and ground waters has been reported to be up to 0.2, and above 500 μg/L, respectively (83, 84). On Se concentration in natural waters ranges from 0.1 to 400 μg/L and sometimes reaches up to 6000 μg/L(71). In 51 locations in Colorado, New Mexico, and Utah, many of the groundwater samples were found saline with Se concentration of 1000 μg/L(85).

Conclusion

A systematic review of various treatment techniques developed over the last decade for Se (selenite and selenate) removal from water and wastewater was conducted. This study was mainly focused on biological and bioremediation technologies. In spite of the problems with these methods, biological treatment and bioremediation have appeared as the foremost treatment practices for Se removal from wastewater due to their advantages, such as low price, flexibility, lack of chemical waste formation and ability to eliminate Se in a recoverable form. Biological treatment is competently capable of reducing the total effluent Se while allowing for lower operation costs and easier system operation. Techniques, such as microbial reduction, biotransformation, and fluidized bed reactor have removal efficiency about 100%. Commonly, most of the techniques were not effective enough for selenate Se (VI) removal compared to selenite Se (IV). The Se removal efficacy can be enhanced by: 1) Combining biological treatment with chemical or physical methods to optimize Se removal and to ensure environmental protection and human health safety. 2) Removing Se (IV) and Se (VI) from polluted waters and wastewaters in both experimental and real settings.

Journalism Ethics considerations

Ethical issues (Including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc.) have been completely observed by the authors.

Conflict of interest

The author declares that there is no conflict of interest.

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