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ORIGINAL PAPER

BIG DATA RESEARCH ON AGRICULTURAL SOIL CONTAMINATION BY ZEOLITE APPLICATION*

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Abstract

Food scarcity has become a global issue. The large amount of solid waste produced by cities every day generates an incalculable amount of toxic substances, including mercury, lead, iron and other heavy metals and harmful chemicals, as well as radionuclide ions, which can cause great harm to people's health and safety when they are transferred to the soil, water, plants and air. Excessive industrial development has led to the infiltration of high polymers and heavy metals into agricultural soil, endangering the health of our food. One other major factor impacting soil quality is the waste produced from construction sites; this waste is mostly backfilled directly into the local soil, causing varying degrees of secondary contamination. Scientists have had to adopt various methods to adjust soil structure, change soil pH, as well as reduce the composition and content of heavy metals in the soil. Among these different approaches, scientists find natural zeolites can improve soil quality and combat soil pollution. Zeolite has excellent ion exchange and adsorption capacity, and is currently widely used as an environmentally friendly soil conditioner in many soil restoration and improvement fields such as ecological organic agriculture, urban soil pollution improvement, and artificial soil construction, which has very important social and economic values for improving soil quality and agricultural production. Adding zeolite to the soil to be restored and improved in the city can effectively adsorb heavy metals and other substances. Based on current research, we collected and obtained data from a large number of previous papers to evaluate the extent of zeolite's effect on soil as well as plants, as scientists attempted to use zeolite to mitigate soil contamination.

Keywords: soil contamination, zeolitization, heavy metal, soil pH, pollution degradation.

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As highlighted in the Global Food Crisis Report 2021, due to ongoing conflicts, pre-existing and COVID-19 related economic shocks, and extreme weather impacts, food security has become one of the prevalent issues affecting sustainable human development. However, the production of major crops such as rice (Oryza sativa L.) and maize (Zea mays L.), which account for more than half of the world's crop production, is plagued by soil contamination. Soil contamination adversely affects food security in two ways: i) it may reduce crop yields due to the toxicity levels of the contaminants; and ii) crops grown in contaminated soils are unsafe for animal and human consumption. For example, in China and India, two major rice-producing regions, the excessive use of mineral nitrogen fertilizers has increased soil acidity, resulting in significantly lower rice yields. Simultaneously, industrial activities such as mining and calcination have led to the contamination of rice fields with heavy metals (e.g., zinc, copper, or cadmium), making rice a bioaccumulator of carcinogenic trace elements, which raises concerns about the health risks of long-term ingestion.

Soil is a critical component of the environment and is the foundation for society to obtain resources, and therefore it has a great significance in the development and progress of humanity. Currently, with the continuous prosperity of the world industrial economy, heavy metal pollution in soil is becoming more and more serious. In addition, soil heavy metal pollution is somewhat hidden and irreversible, which has a great impact on the ecological environment and human health. This problem requires scientists to strengthen the analysis of the current situation of soil heavy metal pollution and figure out the causes of its pollution, so as to study effective treatment measures and improve soil pollution levels. For example, in Japan, a soil heavy metal poisoning disease called "Itai disease" emerged in the 1970s, which was caused by mining contamination of soil in Toyama Prefecture. The contamination resulted in cadmium poisoning (Kaji, 2012). A high concentration of cadmium in soil eventually destroys the soil quality and further leads to an excess of cadmium in rice grown in the soil. As a result of the contamination, the mitochondria of kidney cells are destroyed, leading to severe pain felt in the spine and joints (Inaba et al. 2005).

Zeolite, or natural zeolite, is a group of silicate minerals with a wide distribution. It consists of a three-dimensional lattice of silica-oxygen tetrahedra and aluminum-oxygen tetrahedra linked by shared oxygen (Rollmann et al. 1995). The excess negative charge resulting from the replacement of tetravalent silicon by trivalent aluminum is balanced by monovalent or divalent metal cations, usually alkali or alkaline earth metal cations. Zeolites have adsorption and ion-transformation functions, and because each zeolite has its own specific homogeneous pore size (0.3 to 1 nm), only molecules of the corresponding size can pass through. This allows for zeolites to be widely used as catalysts or carriers, desiccants, feed additives, soil conditioners, sewage purifiers, fillers for plastics and paper, and potassium extraction from seawater.

Over the past few decades, scientists have conducted various studies and experiments on soil remediation. The application of zeolite has been reported to be effective, reasonably priced, and widely used to improve soil acidity and crop yields. Zeolite use can improve pH and reduce heavy metal solubility in soil by increasing precipitation and adsorption efficiency. Additionally, some evidence suggests that zeolite can reduce heavy metal concentrations in plants by decreasing the phytoeffectiveness of soil metals. However, the effects of zeolite on crop growth and soil metal concentrations may vary depending on environmental factors such as crop type, initial soil pH, and soil organic carbon (SOC) content. The heterogeneity of studies, environmental conditions, and crop types makes it challenging to determine global attitudes toward zeolite use from the observations of different researchers. This obstacle hinders the global dissemination of zeolite treatments and a comprehensive assessment of their long-term impact on global food security.

This challenge can be addressed by a quantitative approach that integrates the results of multiple studies. Therefore, a large meta-analysis was conducted to summarize the effects of zeolite application on soil remediation and plant growth in the context of heavy metal contamination with the goal to identify important factors affecting the efficiency of zeolite application on global crop production.

MATERIAL AND METHODS

Material

To collect data, we used Scopus (SCOPUS.com) to retrieve peer-reviewed articles published before January 2022 for zeolite "heavy metal" soils or zeolitization "heavy metal" in plant soils for article titles, abstracts, and keywords. The following criteria was used to exclude irrelevant literature. First, the study must have a blank or unqualified group as a control group. Second, the other treatments needed to have the same variables in each experiment. Third, the paper must include either soil pH or heavy metal concentration data. Fourth, the replication time of each experiment needed to be recorded in the paper.

In this study, we collected 175 pairs of observations from 56 relevant papers (Table 1). The information was processed manually from the content of these papers. Considering the non-intuitive graphical data, we used Get-Data software to extract useful information, mean and standard deviation. If the value is used as standard error, we will use this formula to calculate the standard error back to the standard deviation. For this study, all papers

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		ARH (%)	48.00	79.80	73.90	18.00	78.00	68.60	75.00	75.00	74.80	80.80	57.70	58.00	70.00	70.00	73.00	73.00	68.50	77.80	81.00	56.00	64.20	71.00	78.00	74.00	74.00	75.00	83.00	00.74	
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Table 1

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Reference	Country	City	Continent	Latitude	Longitude	Elevation (m)	MAP (mm)	MAT (°C)	ARH (%)	MAS (h)
Li et al. (2018)	China	Wuhan	Asia	114° 16'E	30° 34'N	133.00	1316.00	17.10	75.00	1865.50
Licina et al. (2007)	Serbia	Belgrade	Europe	20°26'E	44°47'N	170.00	694.00	13.10	68.00	2111.90
Lin et al. (1998)	China	Taipei City	Asia	121° 31'E	25° 02'N	219.00	2368.50	23.50	74.80	1373.80
Mahabadi et al. (2007)	Iran	Gilan	Asia	50° 9'E	37° 11'N	337.00	1293.00	13.00	69.40	2044.00
Mineyev et al. (1990)	Russia	Moscow	Europe	37° 36'E	55° 45'N	170.00	713.00	6.30	78.00	1731.00
Mohamed et al. (2017)	Canada	Vancouver	North America	123°7'W	49°16'N	32.00	1189.00	10.40	70.30	1937.50
Moutsatsou et al. (2004)	Greece	Lavrion	Europe	24° 3'E	37° 42'N	11.00	255.00	25.10	61.00	4343.50
Moutsatsou et al. (2006)	Greece	Athens	Europe	23° 43'E	37° 59'N	70.00	366.50	19.00	60.40	2773.40
Oste et al. (2002)	Netherlands	Wageningen	Europe	5° 40'E	51° 58'N	12.00	848.00	14.00	75.00	1332.00
Panuccio et al. (2009)	Italy	Reggio Calabria	Europe	15°39'E	38°06'N	31.00	546.80	18.30	76.00	2470.00
Pedron et al. (2009)	Italy	Pisa	Europe	10° 24'E	43° 43'N	1.00	893.60	14.50	72.00	2274.50
Peter et al. (2012)	Romania	Baia Mare	Europe	23° 34'E	47° 39'N	228.00	720.00	14.70	70.00	3467.50
Prasad et al. (1999)	Portugal	Coimbra	Europe	8°24'W	40°12'N	40.00	1014.00	16.00	72.80	2480.00
Puschenreiter et al. (2003)	Austria	Vienna	Europe	16° 21'E	48° 12'N	172.00	651.00	10.40	61.50	1930.00
Puschenreiter et al. (2005)	Austria	Vienna	Europe	16° 22'E	48° 13'N	172.00	651.00	10.40	61.50	1931.00
Querol et al. (2006).	Spain	Alicante	Europe	0°28'W	38°20'N	3.00	311.00	18.30	66.00	2851.00
Radziemska et al. (2013).	Poland	Warsaw	Europe	21° 1'E	52° 14'N	218.00	481.70	9.00	77.80	1998.10
Simon et al. (2005)	Hungary	Gyöngyösoroszi	Europe	19° 52'E	47° 51'N	249.00	582.00	15.00	74.00	2130.00
Sun et al. (2014)	China	Guangzhou	Asia	113°15'E	23°7'N	18.00	1736.10	22.40	78.00	1628.00
Sun et al. (2017)	China	Zhengzhou	Asia	113°39'E	34°45'N	110.00	640.90	14.70	65.00	2181.90
Turan et al. (2019)	Turkey	Bingöl	Europe	40° 29'E	38° 53'N	1711.00	935.10	12.30	51.20	2254.90
Usman et al. (2005)	Germany	Stuttgart	Europe	9°12′E	48°48'N	249.00	718.70	9.40	79.00	1807.20
Usman et al. (2006)	Germany	Stuttgart	Europe	9°11'E	48°47'N	249.00	718.70	9.40	79.00	1807.20
Van-Herwijnen et al. (2007)	UK	Surrey	Europe	0° 30'W	51° 23'N	76.00	2599.00	12.00	70.00	2402.40
Vara et al. (2003)	India	Hyderabad	Asia	78° 29'E	17° 23'N	528.00	854.60	26.80	46.00	2698.30
Zhou et al. (2014)	China	Changsha	Asia	112° 56'E	28° 13'N	84.00	1428.10	17.40	79.00	1544.70
Zorpas et al. (1999)	Greece	Athens	Europe	23° 44'E	37° 60'N	70.00	366.50	19.00	60.40	2773.40
Zorpas et al. (2003)	Greece	Athens	Europe	23° 43'E	37° 59'N	70.00	366.50	19.00	60.40	2773.40



collected data on zeolite dosage (kg ha⁻¹), experiment duration (years), percentage of clay in the soil, soil organic carbon value (g kg⁻¹) and total nitrogen (g kg⁻¹). For laboratory data, we collected data on soil pH, and soil metal concentrations (mg kg⁻¹) of copper, lead, nickel, arsenic, cadmium, zinc, and iron (mg kg⁻¹) before and after zeolitization.

Methods

We have drawn a global statistical map (Figure 1) of the experimental sites through the R language. This allows better visualization of the distribution from which the data originated.

We collected the mean and standard deviation of soil pH and heavy metal concentrations. These data were used to calculate coefficients of variation for the control and experimental groups. For all control and experimental variables, we calculated the data as the natural logarithm of the response ratio for Meta-analysis (Liao et al. 2021) by the equation:

$$\ln \mathrm{RR} = \ln \left(\frac{Xt}{Xc}\right) \tag{1}$$

where, in this equation, X represents the mean, pH, or heavy metal concentration of the data for the treatment and control groups. In RR is calculated as the inverse of the variance. Unrecorded variances were filled by averaging hold list data (Liao et al. 2021, Qian et al. 2020).

For the significance analysis, the variance of ln RR was calculated using the following equation:

variance
$$= \frac{\mathrm{SD}^2 t}{N t X^2 t} + \frac{\mathrm{SD}_c^2}{N_c x_c^2}$$
(2)

where SD denotes the standard deviation of the treatment and control groups, N denotes replicates of each study, and x denotes the arithmetic mean of pH values and different heavy metal concentrations.

We used subgroup analysis (Figures 2-11) to average zeolite rate data, time data during the experiment, clay percentage, and soil organic carbon concentration into low, medium, and high. For the resampling test, we used 4999 as the number of iterations. We standardized the units of zeolite dosage to kg ha⁻¹, duration to years, clay amount to percentage, soil organic carbon to g kg⁻¹, and total nitrogen to g kg⁻¹. Summary analysis was used in Metawin software to assess the significance of each subgroup of data by entering the effective size and variance of ln RR. We calculated Prob (chi-square test), Prob (Rand test), mean effect size, 95% confidence interval, Bootstrap confidence interval and deviation confidence interval to demonstrate whether these data are highly significant.

RESULTS

After analyzing all the data, it was found that zeolite application was very effective in controlling soil contamination. The average amount of zeolite used in the experiment was 14,851.95 kg ha⁻¹. The average duration of the experiment was 1.3 years. The average condition of the soil was 17.9% clay compared to other components of the soil, such as sand and chalk. The average soil organic carbon concentration was 16.8 g kg⁻¹ and the total nitrogen value was 373.9 g kg⁻¹.

Zeolite application decreased the As content in the soil by 19.1 mg kg⁻¹ and the proportion by 13.6%. The Cr content in the soil decreased by 13.2 mg kg⁻¹, with a proportional decrease of 11.1%. The Cu content in the soil decreased by 29.1 mg kg⁻¹ and the proportion decreased by 17.4%. The nickel concentration in the soil decreased by 10.5 mg kg⁻¹, into a proportional decrease of 10.9%. The iron concentration in the soil decreased by 9.5 mg kg⁻¹, a proportional decrease of 13.4%. Lead concentration in the soil decreased by 32.7 mg kg⁻¹, a proportional decrease of 18.2%. The Zn content in the soil decreased by 14.6 mg kg⁻¹, a proportional decrease (Figures 2, 3).

The results of the subgroups are shown below, after the calculation of the Excel code, and all background data were aliquoted. The category of zeolite usage below 2300 kg ha⁻¹ will be considered as "low", above 2301 kg ha⁻¹ but below 8000 kg ha⁻¹ will be considered as "medium", and above 8001 kg ha⁻¹ will be considered as "medium", and above 8001 kg ha⁻¹ will be considered as "high". Experimental time less than 0.17 years will be considered as "low", more than 0.18 years but less than 0.68 years will be



Fig. 2. Experimental results of highly toxic heavy metals: Cr, As, Pb and Ni



Fig. 3. Experimental results of heavy metals: Fe, Cu, Zn and pH value

considered as "medium", and more than 0.68 years will be considered as "high". Soil organic carbon concentration less than 5 g kg⁻¹ is considered "low", more than 5.1 g kg⁻¹ less than 19.6 g kg⁻¹ is "medium", and greater than 19.7 g kg⁻¹ is "high". The category of total soil nitrogen concentration less than 0.41 g kg⁻¹ will be considered "low", higher than 0.42 g kg⁻¹ but less than 1.5 g kg⁻¹ will be considered "medium", and higher than 1.51 g kg⁻¹ will be considered "high".

The data indicates that zeolite application has a positive effect on raising soil pH and reducing heavy metal concentrations. To further explore the correlation between these changes and environmental factors and their significance, we need to summarize the analysis of the different subgroups we presented in the results section.

DISCUSSION

During the initial analysis of the data, the overall homogeneity test was performed, and the result of the homogeneity test Prob (chi-square test) is less than 0.05, which proves the existence of heterogeneity, using a fixed effects model, not significant, using a random effects model. In the second step, inter-subgroup effects were tested. First, different subgroups were tested for heterogeneity, high and medium, if less than 0.05, this proved the existence of heterogeneity and the need for further grouping, if not significant, this proved the absence of heterogeneity within the group. Comparisons between the groups (if significant) can demonstrate that the effects are significantly different between the high and medium dose groups. The effect values and 95 confidence intervals E+ are given for each subgroup. Finally, the overall heterogeneity test and E++ implementation are given. Sub-groups in the following figures from up to down are: clay percentage (%), duration of application (years), zeolitization rate (kg ha⁻¹), soil organic carbon (g kg⁻¹), and total nitrogen (g kg⁻¹).

The effect of zeolite on soil arsenic content under different soil properties

From the Figure 4, it can be seen that the effect of different clay content on soil arsenic content is basically the same (p=0.996); the mid clay percentage group had a significant negative effect on soil arsenic content, zeolite



Fig. 4. Heterogeneity test results of As

treatment can significantly reduce soil arsenic content, confidence interval are less than 0.

The effect of different duration (years) of zeolite application on soil arsenic content had a significant difference; zeolite treatment with lower application duration (years) can significantly reduce soil arsenic Zeolite treatments with lower application time significantly reduced soil arsenic content.

The effect of zeolite on soil chromium content under different soil properties

It can be seen from Figure 5 that the effects of zeolite on soil chromium content were consistent for different groups of clay percentage, duration of application (yr), zeolite dose (kg ha⁻¹), SOC (g kg⁻¹), total N (g kg⁻¹), p>0.05.



Fig. 5. Heterogeneity test results of Cr

The effect of zeolite on soil copper content under different soil properties

It can be seen from Figure 6 that there was a significant difference in the effect of zeolite on the soil copper content in the different zeolite dose



(kg ha⁻¹) groups (p=0.019). There was no significant effect of zeolite on the soil copper content in the low zeolite dose group, while there was a significant negative effect in the medium high zeolite dose groups. In all groups, zeolite reduced soil copper content.

The effect of different total nitrogen amount (g kg⁻¹) on the soil copper content was significantly different (p=0.001). In the low, medium, and high

total nitrogen groups, zeolite significantly reduced the soil copper content, but the effect observed in the low total nitrogen group was substantially greater than that of the medium and high total nitrogen groups.

The effect of zeolite on soil nickel content under different soil properties

From Figure 7, it can be seen that the effect of different clay percentage groups and zeolite on the soil nickel content differed in significance



Fig. 7. Heterogeneity test results of Ni

(p=0.005). In the low and mid clay percentage groups, zeolite had a significant effect on reducing soil nickel content; however, the effect of the high clay percentage group is not significant.

There was a significant difference between different zeolite dose (kg ha⁻¹) groups on the soil nickel content (p<0.01). In the high dose group, zeolite

could significantly reduce the soil nickel content. In the mid and low dose groups, zeolite had no significant effect on the soil nickel content.

The effect of zeolite on the soil nickel content was significantly different for different SOC (g kg⁻¹) groups. Zeolite in low and high SOC groups could significantly lower soil nickel content, and zeolite in the mid SOC group had no significant effect on soil nickel content.

The effect of different total nitrogen (g kg⁻¹) groups on soil nickel content was significantly different (p=0.029). In the low and mid total nitrogen groups, zeolite significantly reduced the soil nickel content, but in the high total nitrogen group, zeolite had no significant effect on the soil nickel content.

The effect of zeolite on soil iron content under different soil properties

From Figure 8, it can be seen that the effect of zeolite on the soil iron content in different duration of application (years) groups differed in signifi-



Fig. 8. Heterogeneity test results of Fe



Fig. 9. Heterogeneity test results of Pb

cance (p=0.002). In the low and mid duration groups, zeolite had no significant effect on the soil iron content, while the high duration group zeolite demonstrated a significant reduction of the soil iron content.

There was a significant difference of the effect of zeolite on the soil iron content in the different zeolite dose (kg ha⁻¹) groups (p=0.027). The mid dose group significantly reduced the soil iron content, while in the high and low dose groups, zeolite had no significant effect on the soil iron content.

The effect of zeolite on the soil iron content was significantly different for different SOC (g kg⁻¹) groups. Within the mid SOC group, zeolite was able to significantly reduce the soil iron content, but in the high and low SOC groups, zeolite had no significant effect on the soil iron content.

The effect of zeolite in combination with different total nitrogen (g kg⁻¹) groups was significantly different (p<0.001). In the mid total nitrogen group, zeolite was shown to significantly reduce the soil iron content. However,



Fig. 10. Heterogeneity test results of Zn

in the high and low total nitrogen groups, zeolite had no significant effect on soil iron content.

The effect of zeolite on soil lead content under different soil properties

From Figure 9, it can be seen that different clay percentages resulted in significant differences among the effects of zeolite on the soil lead content (p=0.001). Within the low and mid clay percentage groups, zeolite significantly reduced soil lead content, while the effect of zeolite with the high percentage group is not significant.

The effect of different total nitrogen (g kg⁻¹) concentration on zeolite's ability to affect the lead content was significantly different (p=0.005). Zeolite in low, mid, and high total nitrogen groups was able to significantly reduce

soil lead content; the effect in the low total nitrogen group was the largest and the effect of mid group was the smallest.

The effect of zeolite on soil zinc content under different soil properties

Within all the different analyzed soil groups, there was no significant difference observed. Zeolite did not have a statistically significant effect on the zinc content as the *p*-value of all data sets was above 0.05 (Figure 10).

The effect of zeolite on soil pH under different soil properties

From Figure 11, it can be seen that the effect of zeolite on soil pH in different duration of application groups differed significantly (p=0.016). Zeo-



Fig. 11. Heterogeneity test results of Zn

lite in the low, mid, and high duration groups significantly increased soil pH; the effect in the low duration group was the largest and the effect in high duration group is the smallest.

Soil As, Cr, Cu, Fe, Ni, Pb, Zn, pH total effect

From Figure 12, it can be seen that zeolite has a significantly negative effect on soil copper, iron, nickel, lead and zinc, demonstrating that zeolite can significantly reduce the content of these metals in soil; meanwhile, there is a difference between studies regarding the effect of zeolite on soil As, Cr, Fe, Ni and Zn content, p<0.05.



Fig. 12. Heterogeneity test results of pH

CONCLUSIONS

We have found that natural zeolites have a significant remedial effect on contaminated soils. The application of zeolite led to a significant improvement in the acidification of the soil ponds. The heavy metal content of the soil decreased to varying degrees, especially for copper, lead and zinc. We also discussed the issue of data heterogeneity and concluded that the data and analysis were reliable.

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