Review Article

Zeolites: A potential source of soil amendments to improve soil properties

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Abstract

Zeolites are naturally occurring volcanogenic, sedimentary aluminosilicate minerals with voids and channels in their inner structure. Physical as well as chemical properties of soil are the major factor of plant growth and its development as it affects the soil environment. Therefore, it is very crucial to manage soil chemical and physical properties for improving nutrient use efficiency with the adoption of environment friendly strategies. Recent studies show that use of zeolites is beneficial as it helps in improving the physical as well as chemical properties of soils. It is mainly due to its higher cation exchange capacity (CEC), higher specific surface area, internal void structure, higher moisture holding capacity *etc.* Thus, these properties of zeolites can help in enhancing the water and nutrient use efficiency. Also, it reduces the risk of environmental pollution occurring due to nitrate leaching, emissions of nitrous oxides and NH_3. Therefore, zeolite application is beneficial for providing balanced NPK fertilization, improving soil physical and chemical properties which helps in improving soil quality as well as soil health and it also minimizes environmental pollution for economic and environmental considerations.

Keywords: Aluminosilicates, Cation exchange capacity, Specific surface area, soil quality, Zeolites

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Introduction

Zeolite is a naturally occurring volcanogenic sedimentary mineral primarily composed of aluminosilicates [1]. It is made up of 3-dimensional crystal lattice, having loosely bound cations and it is capable of hydrating and dehydrating without alteration of the crystal structure [2]. Existence of about 40 natural zeolites and 100 synthetic zeolites are reported till date [3]. In 1756, Alex Fredrik Cronstedt who was first to identify the zeolite as rapidly heating the material earlier known as “stilbite”. It produced large amounts of steam from water which was absorbed by the stilbite. Based on this, he re-named this material as “Zeolite”, made up of Greek word meaning “boiling stones”. Zeolites represent a group of soft, white aluminosilicate minerals of tectosilicate type, *i.e.* a 3-D framework of interconnected tetrahedral containing of aluminum, silicon and oxygen atoms. They made up of crystalline structure having $[AlO_4]^{5-}$ and $[SiO_4]^{4-}$which is bonded together in such a way that all four oxygen atoms located at corners of each tetrahedron are shared with adjacent tetrahedral crystals. The general formula of a zeolite is $Me_2/n\ O.\ Al_2O_3.\ xSiO_2.\ yH_2O$ where, Me is any alkali or alkaline earth atom, n is the charge on that atom, x is the number of Si tetrahedron varying from 2 to 10, and y is the number of water molecules varying from 2 to 7. In addition to this, they possess microscopic pores of small size as of molecular dimension sand. Hence, they are also known as the “molecular sieves” which facilitates cation exchange in process of adsorption. Based on these attributes of zeolites, it consists applications in separation and filtration processes.

Most of the countries failed to report production of natural zeolites or estimated production is reported late with 2 to 3 year of time lag. Globally, China ranks first in production of natural zeolites, which accounts for approximately 30% of estimated worldwide production (**Table 1**). In India, about 2.8 Mha area was reported with zeolitic soils which is nearly 1% of the total geographical area of the country [4]. In India, about 86% area of the zeolitic soils covers under arid and semi-arid climates. More than 60% area of zeolitic soils contributed from the states Maharashtra (43.01%), Madhya Pradesh (15.40%) and Andhra Pradesh (43.01%) *etc*. Ca-rich zeolites were first reported in Indian soils in the region of Western Ghats. The utility of Ca-rich zeolites in Indian soils has been reported for the amelioration of harmful effects of high pH, sodium, magnesium and poor drainage [5]. These zeolites have been described as the natural saviour for soils to maintain its quality.

Table 1 Estimated world mine production of zeolites [4]

Country	Mine production (tonnes)	Reserves
United States	79,000	World reserve data are unavailable but are estimated to be large
China	3,00,000	
Cuba	55,000	
Jordan	13,000	
Korea	2,00,000	
New Zealand	80,000	
Turkey	60,000	
Other countries	3,50,000	
Total (approx.)	1,137,000	

Effect of zeolites on physical properties of soil

Effect of zeolites on bulk density

A significant difference in apparent density of zeolite treated soils observed by Hassan and Radi [6]. They reported bulk density of 0.84 Mg m^{-3} and 0.75 Mg m^{-3} in sandy soil and 0.70 and 0.66 Mg m^{-3} for the loamy soil when application of zeolite in soil was done at the rate of 0.8 and 1%, respectively. Zeolite dose of 0.2 to 0.4, 0.6, 0.8 and 1% to sandy soil resulted decrease in bulk densities (1.28 to 1.15, 1.08, 0.84 and 0.75, respectively) Mg m^{-3}. Similarly, bulk density of 1.12, 0.92, 0.87, 0.70 and 0.66 Mg m^{-3}, respectively was recorded in loamy soils for dose of zeolite. This gradual decrease in bulk density was reported due to the increase in the applied dose of porous zeolite which have a positive impact on the physical properties of soil particularly on total porosity resulting in reduced virtual density values is consistent [7]. It may be attributed due to the difference in the nature of structure of the soil and the proportion of micro pores which increases the density reduction of sandy soil.

Effect of zeolites on soil porosity

Hassan and Radi [6] observed that soil porosity increased in sandy soil by 61.52% and 63.96% and in loamy soil by 56.83% and 59.82% with the application of zeolite 0.8% and 1%, respectively as compared to 0% zeolite application (38.83% sandy soil and 46.86% loamy soil). The main reason of increased porosity due to the zeolite improved porous soil system, improve soil construction and decrease the soil density. Also may be due to high porosity of zeolite metal [8]. and thus improved soil composition and physical properties of the soil in terms of porosity and retention of water [7].

Effect of zeolite on saturated hydraulic conductivity

The saturated hydraulic conductivity of sandy soils decreases in zeolite treated soil reported by Hassan and Radi [6]. It was found to be decreased from 0.317 cm min^{-1} to 0.117 cm min^{-1} with the application of 0% and 1%, respectively. In the loamy soils with the increase of zeolite doses saturated conductivity found to be increased. Saturated conductivities of 0.016 cm min^{-1}, 0.029 cm min^{-1} and 0.030 cm min^{-1} was observed with the application of 0%, 0.2%, 0.4%, respectively zeolite doses. The significant decrease of hydraulic conductivity was due to the increased level of zeolite in soil that reflects the metal's ability to improve hydraulic properties of the soil, increase its water capacity and reduce the rate of sandy soil hydraulic conductivity [9, 10]. Hydraulic conductivity mainly depends on the geometry of the pores and moisture content. The added metal results in the production of different porous voids due to changes in soil characteristics. These results corporate with the findings of Hassan and Mahmoud [7]. They reported the role of increase in soil moisture content, porosity, reduced bulk density and the horizontal spread of water to sandy soil with the application of zeolite. However, the extent of the increase in values of water conductivity of the sandy soil is higher than the loamy soils due to different texture soil, the size of soil pores and rough texture compared to medium texture soil.

Effect of zeolites on soil water content

The infiltration rate and soil water content which was found increased significantly in zeolite treated soil as compared to the untreated soil studied by Ghazavi [9]. Whereas significant decrease in runoff volume, drained water volume and sediment amount was reported in zeolite treated soil ($P<0.01$). In both treated and control soil samples, soil moisture decreased with time, but soil water content in the treated soil was found higher than untreated soil during the whole period (**Figure 1**). Soil water content measured at one-day interval was reported significantly different in the zeolite treated soil samples as compared to untreated soil. Higher water holding capacity, and higher adsorption capacity of natural zeolite was reported in many research studies. Also the impact of zeolite was found on physical soil improvement by increasing the availability of Ca^{2+} and K^+ and reducing the leaching losses of exchangeable cations, especially K^+ [11].

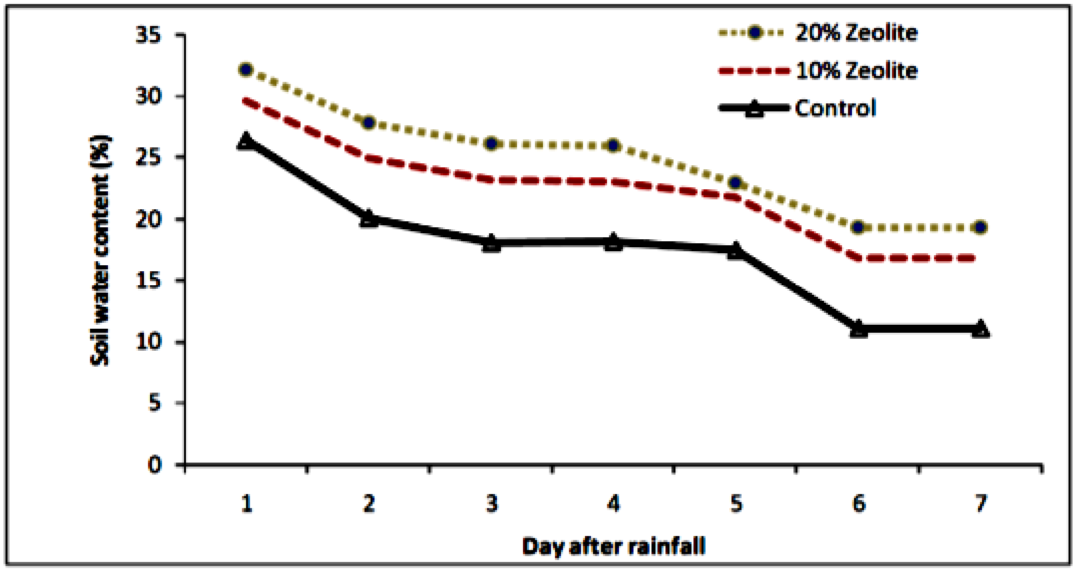

Figure 1 Effect of zeolites on soil water content in alkaline soil

Effect of zeolites on infiltration rate and soil water releasing processes in soil

The infiltration rate at different intensity of rainfall reported by Xiubin and Zhanbin [12] and observed that during low intensity rainfall resulted higher infiltration rate due to increased ability of adsorption (with surface area of about1150.5 m^2/g). Soil treated with zeolite can increase 7–30% and 35% infiltration rate as compared to the normal soil on gentle slope land and steep slope land, respectively. The zeolite treated soil can also increase soil moisture by 0.4–1.8% in the drought condition while 5–15% in general situation. Consequently, it can reduce overland flow (surface runoff) and protect soil from erosion. It shows the water releasing process followed the same trend with the time (**Figure 2**), but water content in the zeolite-treated soil was higher than that of the normal soil during the whole period. Also, the zeolite-treated drain out water more rapidly than the normal soil due to large amount of free water in the structural channels [8].

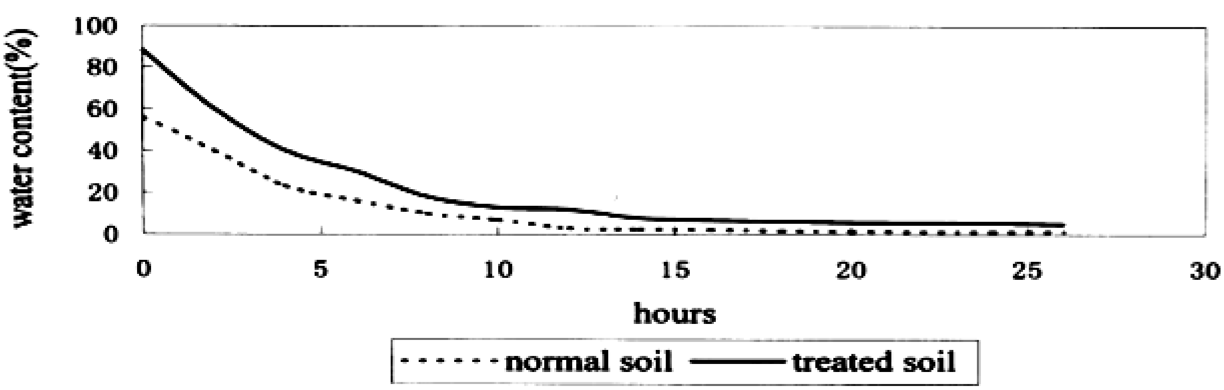

Figure 2 Effect of zeolites on soil water releasing processes in calcareous soil

Effect of zeolites on mean weight diameter (MWD) of soil

Aminiyan [13] studied that MWDw and OC fractions in the soil as affected by nanozeolite, zeolite, alfalfa, and wheat straws percentages applied to soil. MWDw increased with the addition of higher percentages of nanozeolite and zeolite (10 and 30 %) and 5 % plant residues into the soil. The mean weight diameter was observed to be increased by 0.735 and 0.685 mm in the soil treated with 30 % nanozeolite + 5 % alfalfa straw (compound A), and 30 % zeolite + 5% alfalfa straw (compound B), respectively as compared to control. Although MWDw is also known to be related wit soil aggregate stability as the results of recent research studies indicated that the SOC is closely related in the formation as well as stability of soil aggregates (**Figure 3**). Thus, the soil amendments with the use of organic residues can improve soil structure and also increases aggregate stability [14]. Soil management practices that can leave more plant residues on the soil surface generally results higher improvements in soil aggregation and aggregate stability. Aggregate stability and SOC have also been shown to be positively correlated with polyphenol concentration [15], which has been proved to be an important precursor of humic substances that can stabilize aggregates. Nanozeolite and zeolite are clay minerals, and they have considerable calcium (Ca) content. In addition to its role as a binding agent its polyvalent cations also enhances the formation of cationic bridges between organic matter and clay [16]. It is attributed to high saturation of clay particles with these cations that helps the organic–mineral complex to remain more flocculated and condensed as well as helps in reducing the efficiency of microbial and enzymatic attacks.

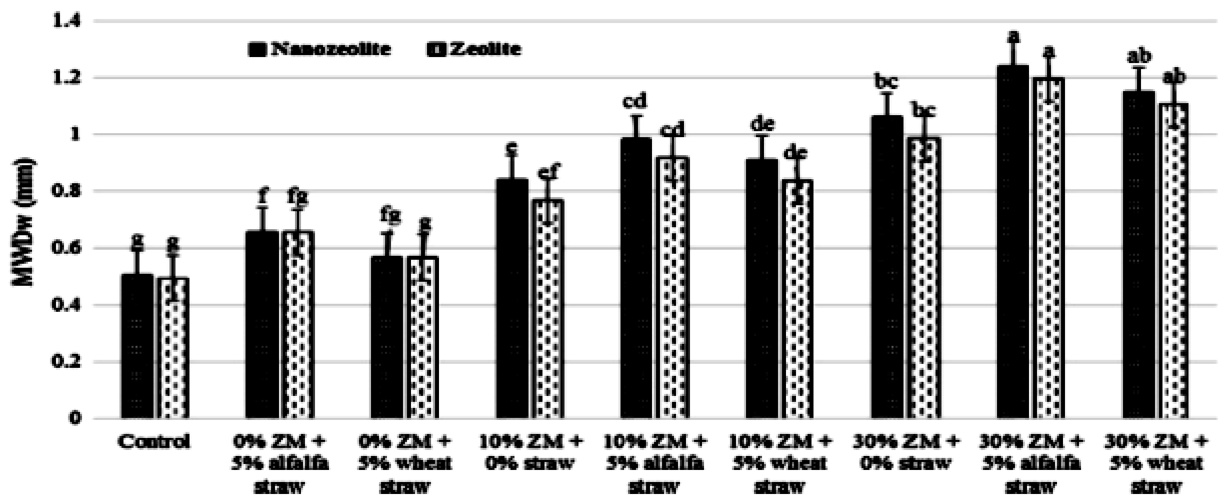

Figure 3 Effect of zeolite on mean weight diameter (MWD) of soil

Effect of zeolites on chemical properties of soil

Effect of natural zeolites and organic fertilizers on soil chemical properties

Glisic [17] presented the results of two-year study on the effect of natural zeolites, organic fertilizer-cattle manure and mineral NPK fertilizer (15:15:15) on soil properties of shallow eroded vertisol (**Table 2**). The results revealed that the acidity considerably decreased from 5.07 units (prior to trial establishment) to 6.01 (after the two-year study period). The humus content was found to be increased from 2.92 to 3.50% with increase in soil depth. Soil chemical properties are observed to be improved with the application of combination natural zeolite 'Agrozel' (1 kg m^2) and Manure (1 kg m^2) treatment which mainly results in decrease of soil acidity by 0.94-unit whereas humus content increased by 0.58%. Although total nitrogen content was found non-significant with variable soil depth but phosphorus content was found to be increased. Therefore, it was concluded that the agrozel type of natural zeolite, when combined with organic fertilizer (manure) can be successfully utilized as amendments for problematic soils having unfavorable chemical properties as well as for enhancing the practice of cultivation of certain fruit crops on such soils. The two-year cattle manure treatment was found to increase the humus content from 2.92 to 3.25% which may be attributed to average organic matter content (25%) present in well matured cattle manure. The two-year applications of the Agrozel-manure combination resulted significant changes in soil chemical attributes. Significant reduction of 0.94 units was recorded for pH. In addition to increased humus content, it also resulted in soil acidity reduction because significant proportion of H_2CO_3 reacts with the soil adsorptive complex. This resulted in the formation of $NaHCO_3$ which dissolves into H_2CO_3 (weak non-dissociating) & NaOH (strong dissociating) in the aqueous solution and also it produces several OH^- ions in the soil that resulted in soil acidity reduction. The exchangeable cation sites of Agrozel are occupied by exchangeable divalent metal Ca^{2+} ions [18] which is being substituted by other ions present in the soil solution. So, in soil solution zeolites helps in indirect inducing the decrease in acidity. This is for similar reasons that resulted in increase of potassium content (>400 mg kg^{-1}). Agrozel is dominated by the K^+ ion containing mineral clinoptilolite. The ion-exchange property is one of the important characteristics of zeolite having the CEC of about 216 mmol/100g soil [18, 19]. The main reason of increasing soil potassium content can be related to its highest ion-exchange capacity that resulted in rapid release of potassium from the zeolite lattice into the soil solution.

Table 2 Effect of natural zeolites and organic fertilizers on chemical properties of acid soil

Soil properties	Control	NPK + Manure	Agrozel + Manure	LSD (p<0.05)
pH	5.07	5.10	6.01*	0.74
Humus (%)	2.92	3.25	3.50	0.31
Total N (%)	0.14	0.15	0.18	0.04
P_2O_5 (mg kg^{-1})	68.0	70.5	148.5**	23.0
K_2O (mg kg^{-1})	263.0	300.0	>400**	51.0

Reduction in NH_3 volatilization loss by zeolite application

An incubation experiment was conducted by Rabai [20] to compare the effect of different ratios of compound fertilizer amended with clinoptilolite zeolite on NH_3 volatilization, soil exchangeable NH_4 and available NO_3 contents with surface-applied urea without additives. Treatments with zeolite significantly reduced NH_3 loss over the urea treatment without addictives. There is a potential for compound fertilizer when applied in combination with zeolite to improve nitrogen use efficiency by lowering ammonia volatilization and increasing accumulation of exchangeable NH_4 and NO_3. Their combination can also improve the retention of exchangeable NH_4 and NO_3 and this was possible because zeolite favoured the formation of ammonium and nitrate over ammonia. Ammonia volatilization generally increases with NH_4–N rate and decreases in the following pattern for different N sources: $CO (NH_2)_2$ > $(NH_4)_2SO_4$ >NH_4NO_3. Clinoptilolite zeolite (CZ), a porous mineral with high cation exchange capacity and with great affinity for NH_4^+ has been used for reduction of NH_3 emission as well as to eliminate NH_3 toxicity in plants. The NH_4–N concentration was recorded to be increased in the first week of incubation as a result of NH_4–N release from urea hydrolysis, and then decreased rapidly [21].

Comparison of natural zeolite and synthetic zeolite (K-MesoLite) on NH_4^+ ion retention

The use of natural zeolites, modified and synthetic zeolites for reducing the NH_4^+ leaching as well as for improving the NUE demonstrated by Zwingmann [22]. MesoLite is an artificial zeolite, which is produced by a caustic treatment of kaolin at 80-95 °C. It has a very high CEC (~500 cmol (+) kg^{-1}) and moderately low surface area (9-12 m^2 g^{-1}) [23]. The study was conducted to investigate the effectiveness of the K^+ MesoLite and NH_4^+ MesoLite to compare with natural clinoptilolite for retaining and releasing NH_4^+ in columns of amended sandy soils. The additions of 8 g kg^{-1} natural zeolite and K-MesoLite resulted in decrease of NH_4^+ leaching whereas K-MesoLite amended soil exhibited a much greater retention efficiency of NH_4^+. Soil amendment with K-MesoLite retained higher NH_4^+ of about 90%, while only 30 and 40% retention was found in clinoptilolite treatment for amended and unamended soil, respectively. The effectiveness of K-MesoLite was reported about 11.5 times greater than clinoptilolite zeolite. The higher efficiency of K-MesoLite might be explained by the higher CEC of K-MesoLite which is about five times in comparison to clinoptilolite zeolite [24].

Zeolite as slow release phosphorus fertilizer in sunflower

An experiment conducted by Pickering [25] which greatly enhanced the P plant uptake from RP when applied in combination with NH_4^+ zeolite. Though the P uptake was quite lower in comparison to soluble P source. Within the applied area of NH_4^+ zeolite/RP band, root proliferation was found to be prominently increased which may be due to the increased exchange-induced dissolution. The K^+ zeolite system failed to produce significantly higher yield. This may be probably because basal application of adequate K^+ supply resulted in reduction of uptake within the zeolite/RP band and hence reduced the extent of exchange-induced dissolution. The zeolite/RP system offers the considerable unique advantage of P release in synchronization with plant demand. Solubility of rock phosphate (RP) have been reported to be increased with use of aluminosilicates minerals named zeolites when saturated with mono-valent nutrient cations, such as NH_4^+ and K^+. The main mechanism which is proposed to account for the solubilisation of RP is ‘Exchange-induced dissolution’. It was proposed that with the plant uptake of NH_4^+ and K^+ from exchange sites results in substitute of Ca^{2+} which lowers the Ca^{2+} concentration of soil solution and also induces further dissolution of RP [26].

Mechanism of P release

The exchange-induced dissolution model of P release can be summarized for a NH_4^+ saturated zeolite as:

$$RP + NH_4^+ \text{ zeolite} == Ca^{2+} \text{ zeolite} + NH_4^+ + PO_4^{3-}$$

In this model system, the NH_4^+ released to the soil solution was taken up by plant roots that further induced the release of NH_4^+ from the exchange complex. The exchange sites vacated by NH_4^+ are substituted by Ca^{2+} that induced dissolution of RP and then release P into solution. Thus, the release of P will occur at a rate which is determined by the NH_4^+ uptake rate of plant which helps in plant growth. As the release of P from the zeolite/RP system depends upon the plant nutrient uptake rate, root proliferation within the zeolite/RP application band. In the NH_4^+ saturated zeolite/RP system which shows that the plant roots extensively ramified within the fertiliser band. This response can also be observed in the K-saturated zeolite/RP system but to a lesser extent. Extent can be higher also when applied with higher basal treatment of K mixed with the potting substrate [27].

Nanozeolite nutrient release pattern

The effect of natural zeolite (clinoptilolite) fortified with Zn by loading zinc sulphate ($ZnSO_4$) on soil and crop studied by Yuvaraj and Subramanian [28]. The results showed that Zn released from the nano-zeolite substrate was found for prolonged period of 1176 hrs while the Zn released from the ordinary $ZnSO_4$ ceased to exist within 216 hrs (**Figure 4**). The data suggested that the nano-sized zeolite is capable of retaining Zn and slowly release into the soil solution which may be act as a slow release Zn fertilizer and improves the nutrient use efficiency by crops. Zeolites are also known for extensive surface area and capable of regulating the adsorption and desorption of nutrients that eventually increases the crop yield [29]. Utilization of zeolites in agriculture is possible because of their special cation exchange properties, molecular sieving and adsorption. Zeolite can also hold nutrients in the rhizosphere which leads to improved use nutrient efficiency. The large honey comb crystal structure of zeolite provides large surface area. Cationic plant nutrient ions *i.e.* Potassium (K^+) and zinc (Zn^{2+}) and water molecules are also stored in the zeolite crystal and are readily available to the plants [30].

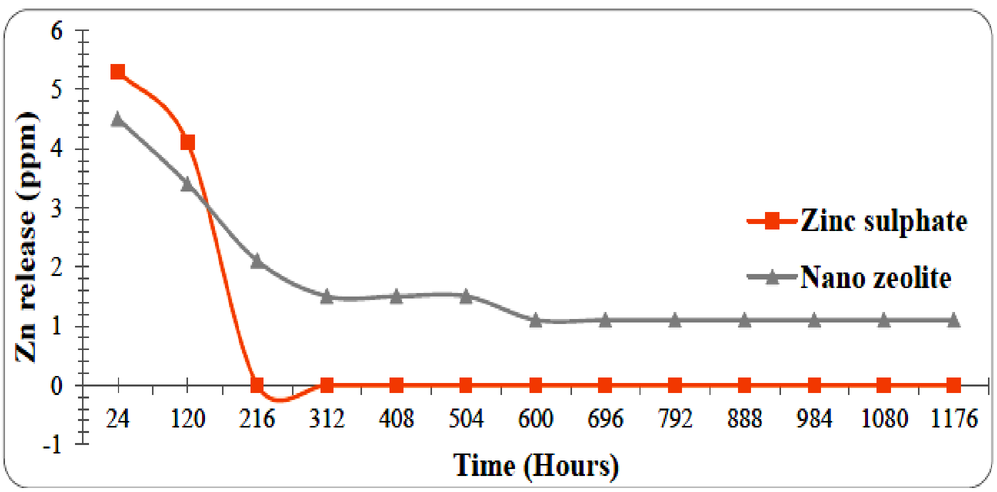

Figure 4 Nutrient release pattern of nanozeolite in black soil

Effect of synthetic nanozeolite and nanohydroxyapatite on soil P content in calcareous soil

A greenhouse experiment conducted by Mikhak [31] to assess the fertilizing effect of synthetic nano-zeolite (nCp)/nano-hydroxyapatite (nHA) on phosphorus uptake on chamomile plant. showed Maximum phosphorous content was reported in Cp-NH_4^+ + nHA treatment whereas all other traits were significantly differing. Minimum phosphorous content was observed in control treatment which had no significant difference with Cp-NH_4^+, PR, and Cp-NH_4^+ + PR treatments. The combination of nCp-$(NH_4)_2SO_4$ and nHA showed better results in all measured traits in comparison to other treatments. Later nCp-$(NH_4)_2SO_4$ along with triple super phosphate (TSP) was found to enhanced the growth of chamomile. The use of nitrogen causing-acid fertilizers with phosphorus fertilizers created an acidic environment around the plants roots and rhizosphere which helped in better adsorption of phosphate. In addition to pH reduction, the application of nCp-$(NH_4)_2SO_4$ improved physical properties, improved phosphorus adsorption, porosity and better moisture provision in the rhizosphere. With increase in the proportion of nCp-$(NH_4)_2SO_4$ to nHA, phosphorus uptake and soil physical properties improved. TSP is an acid- forming fertilizer; its acidic property is nearly negligible as compared to other nitrogen fertilizers and P fertilizers which has slight impact on soils with high pH. In the above study, nCp-$(NH_4)_2SO_4$ /nHA compound fertilizer was found more effective in decreasing pH than TSP [32].

Effect of zeolite amended urea on ammonium and nitrate leaching under maize

Latifah [33] showed that the mixtures amended with (0.192 kg/ha zeolite) clinoptilolite zeolite [U1Z (130 kg /ha urea), U2Z (97 kg /ha urea and U3Z (65 kg /ha urea)] significantly reduced NH_4^+ leaching from urea as compared to soil alone (T0) and urea alone (U1). Because the clinoptilolite zeolite has high surface area for NH_4^+ adsorption (**Figure 5**). The lower amounts of NO_3^- leached from the treatment without clinoptilolite zeolite (U1) may be due to the loss of NH_4^+. The NH_4^+ retention in the treatments amended with clinoptilolite zeolite was possibly because of the specific selectivity of clinoptilolite zeolite for NH_4^+ adsorption [34]

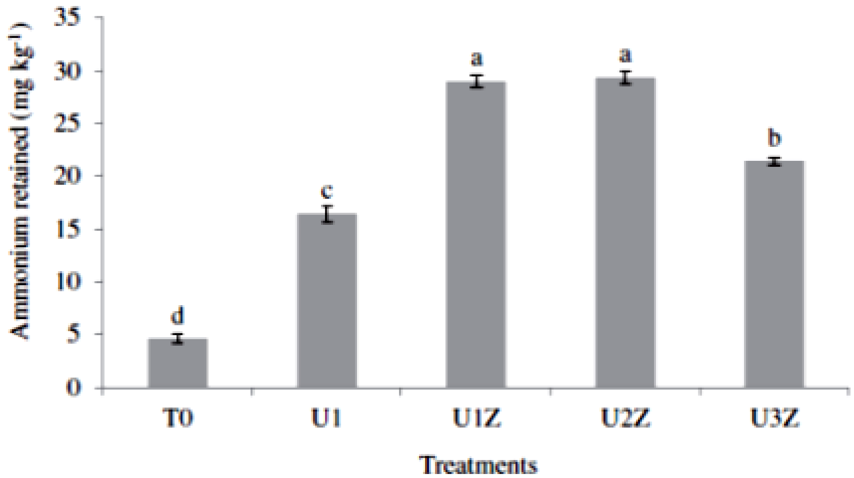

Figure 5 Effect of zeolite amended urea on ammonium retention in black soil

Constraints

There are various beneficial effects of zeolites, but due to the excess sodium content of commercial zeolites which negatively influence plant growth and yield. Application of zeolites is suitable under coarse-textured, highly leached and acidic soil pH conditions. Some zeolite like, erionite is reported to be harmful to human health. Limited availability of resources leads to increased price of such type of fertilizers.

Future prospects

Availability of zeolite deposits has to be characterized in each country. Analyzed the physical stability of zeolites in a different soil environmental conditions. Valuation of long-term influence of zeolites on soil biological functions. Innovation of new protocol for cost effective organo-zeolitic fertilizers and also nutrient release pattern from organo-zeolites.

Conclusion

Zeolite amendment is an effective way to improve physical properties *i.e.* bulk density, porosity, mean weight diameter, hydraulic conductivity, infiltration, water content in soil. Zeolites are able to reduce ammonia volatilization losses and also improve the ammonium ion retention in soil. Zeolites when mixed with major nutrients or micronutrients acts as a slow released fertilizer. Zeolite could partly affect microbial dynamics in soil. Zeolites are effective as soil ameliorants and in remediation of heavy metal contaminated soils.

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