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ENVIRONMENT

Zeolites are Emerging Soil Amendments for Improving Soil Physical and Chemical Properties in Agriculture: A Review

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ABSTRACT

Soil water and soil nutrient contents are the most important factors for crop productivity and agricultural sustainability. Water has become very scarce in rainfed agriculture and the cost of chemical fertilizers is increasing day by day. Nutrients availability and their translocation also reduce under limited water availability. Therefore, it is very important to manage both water and nutrient resources and to improve their use efficiency through environment friendly strategies. In the recent time, the use of zeolites has been emphasized in improving the physical and chemical properties of soils due to their high CEC, moisture holding capacity and etc. Zeolites are naturally occurring aluminosilicates with voids and channels in their inner structure, where water and nutrients are stored and exchanged. This characteristic of zeolites can be capitalized to enhance the use efficiency of fertilizers and water and to reduce the risk of environmental pollution occurring due to nitrate leaching and emissions of nitrous oxides and NH_3. It is with this concern; the present review is focused on harnessing the potential of zeolites, for improving the water and nutrient use efficiency with reduced carbon foot prints.

Highlights

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Keywords: Fertilizers, Nutrient use efficiency, Water use efficiency, Zeolite.

Indian soils are getting depleted in their fertility with the increase in nutrient mining. Nutrient mining refers to excess nutrient removal by crop than nutrient additions. There is a net gap of 8-10 Mt per annum of N, P_2O_5 and K_2O (Tandon 2004) and this may increase further with increased food production to meet increasing population in future. However, nutrient depleted soils cannot meet the high agricultural production targets unless it is properly addressed. The quantum of this nutrient depletion is found to be more in arid and rainfed areas. Since, 1975 to 2002, the extent of phosphorus (P) and potassium (K) depletion was 7.7% and 13.4% respectively in arid soils of India (Singh *et al.* 2007). And the nutrient deficiency in Indian soils increased from one nutrient element (N) to nine nutrient elements (N, P, K, S, B, Cu, Fe, Mn, and Zn) from 1950 to 2005–2006 (Bhattacharyya *et al.* 2015). In the recent years, a declining trend of total factor productivity and low nutrient use efficiency has been primarily observed. In addition, due to frequent dry spells, moisture has become a major constraint for crop production in arid, semi-arid and sub-humid regions of India. Among all the sectors, agriculture is the largest consumer of water and this consumption could be 1072 billion cubic meters by 2050 years, if trends in current practices of crop production continue. Moreover, maximum importance is being given towards production of high water requiring crops than low water requirement crops. Thus, there is an imminent need to improve the water use efficiency or more importantly, the water productivity. Research showed that nutrient uptake also declines with decline in soil moisture. Further, the most difficult and challengeable task is to improve water productivity under water scarce condition. Hence, recent research is focused on strategies that enhance both nutrient and water use efficiency.

Zeolite as nutrient additives helps in enhancing nutrient use efficiency (Kavvadias *et al.* 2019; Bernardi *et al.* 2016; Ramesh and Reddy 2011) and water use efficiency (Ozbahce *et al.* 2014) by improving the physical and chemical properties of soil. Its use in agriculture is called as rock farming. Zeolites are crystalline, hydrated aluminosilicates of alkali and alkaline earth cations with three dimensional networks of AlO_4^{5-} and SiO_4^{4-} tetrahedra, linked by sharing of all oxygen atoms (Fig. 1).

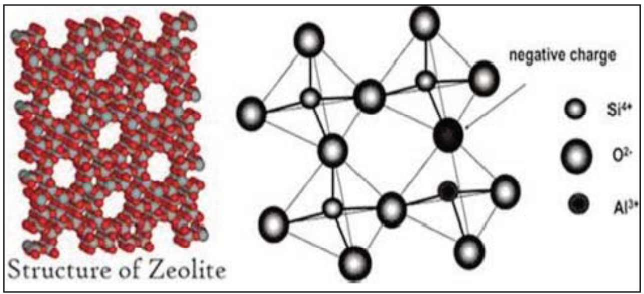

Fig. 1: Inner structure of zeolites with voids and channels

Source: *Yuan et al. (1999) .*

They are characterized by an ability to lose and gain water reversibly and to exchange their constituent elements without a major change in structure (Leggo *et al.* 2006; Ok *et al.* 2003). They occur naturally in soils and also been synthesized due to many uses such as water treatment, aquaculture, animal nutrition, human medicine, manure management, as catalysts in hydrocarbon conversion and in a wide variety of agricultural processes. Their unique properties include high cation exchange capacity ranging from 2.0- 5.6 meq g^{-1} zeolite, high adsorption properties and structure stability over the long term. The name 'zeolite', means "boiling stones" (from Greek words), as Alex Cronstedt, in late 1750's. (a Swedish mineralogist) found that if rapidly heated, stilbite crystals would release steam. The zeolites are porous and crystalline in structure with numerous voids that are interconnected with channels in their framework. Moreover, the internal structure of these frameworks is hydrophilic. To this date, about 1000 different types of zeolite minerals have been reported, predominantly from sedimented rocks of volcanic origin. Zeolite deposits exist in all over the world, huge natural zeolite reserves are found in Japan, USA, Russia, Hungary, Croatia, Serbia and Turkey (Sarioglu 2005). Japan started using zeolite in agriculture way back since 1960s. They mainly use it for increasing the pH of acidic soils and to maintain moisture content in soils. In India, Ca-rich zeolitic soils are found to occupy an area of ~2.8 m ha (Bhattacharyya *et al.* 2015). These zeolites have several species and each of the species has its own crystal although all these structures have SiO_4 and AlO_4 tetraherdal linked together in a simple geometric form and hence, vary in morphology, physical and chemical composition. For studying their morphology, X-ray diffraction, scanning electron microscopy, X-ray fluorescence spectrometer and N_2 adsorption–desorption techniques were found promising. Huge literature is available with respect to crystal structure of zeolite. (Zhang *et al.* 2011) reported that mordenite zeolite had fiber-like, rod-like, prism-like and needle-like structures. (Mansouri *et al.* 2013) reported that clinoptilolite consists of lamellar texture and its grains have latent fine-crystalline structure with average crystal sizes of 50 × 300 × 700 nm (Sprynskyy *et al.* 2010). Its Si/Al ratio generally ranges from 4 to 5.5 (Tsitsishvili *et al.* 1992; Ramesh *et al.* 2015). Erionite is a natural fibrous zeolite with molecular formula (Na_2, K_2, Ca, Mg) 4.5 Al_9 $Si_{27}O_{72}$•$27H_2O$) (International Agency for Research on Cancer (IARC) 1987). Its fibres resemble amphibole asbestos fibres morphologically (Dogan and Dogan 2008). The fibers' size generally ranges from less than 0.2 to 10 μm in diameter and have lengths that range from 2 μm to over 200 μm (Lowers *et al.* 2010). Also, occur as simple penetrating twins of pseudocubic rhombohedra and lens shaped and they are characterized as chabazite and phacolite respectively. Ramesh *et al.* (2015) studied the crystal morphology of clinoptilolite fractions and reported that it is having particle size grains of 396 nm (fine fraction, 125 μ sieves), 488 nm (medium fraction, 125–250 μ sieve) and 541 nm (coarse fraction, (250 μ sieve) with layered structure.

Water use efficiency

Water use in a sustainable way is the immediate need in Indian agriculture to ensure food security with available water resources; hence, technologies that enhance the water use efficiency are being

Table 1: Effect of zeolite application on different soil physical properties

Sl. No.	Soil type	SFC (%)	WP (%)	BD (g cm^{-3})	PD (g cm^{-3})	P (%)	HC (cm h^{-1})
1	N1	43.31 (41.01)	26.72 (22.65)	1.45 (1.37)	2.69 (2.69)	46.04 (49.07)	0.95 (0.41)
2	N2	44.31 (41.63)	28.13 (25.70)	1.43 (1.49)	2.64 (2.63)	46.05 (43.35)	1.6 (1.75)
3	N3	26.81 (23.06)	16.21 (13.44)	1.31 (1.19)	2.61 (2.65)	49.68 (55.09)	7.58 (18.50)
4	N4	30.46 (26.73)	16.77 (14.82)	1.45 (1.38)	2.62 (2.70)	44.75 (48.89)	1.37 (1.23)
5	N5	29.98 (27.25)	16.9 (15.43)	1.44 (1.47)	2.67 (2.68)	46.26 (45.15)	1.19 (0.98)
6	N6	36.34 (33.92)	21.65 (21.07)	1.47 (1.54)	2.57 (2.69)	42.67 (42.75)	0.94 (0.82)
7	N7	12.88 (10.56)	5.95 (5.33)	1.51 (1.50)	2.63 (2.62)	42.59 (42.75)	4.08 (5.31)
8	N8	28.7 (27.11)	18.61 (15.21)	1.29 (1.27)	2.6 (2.58)	50.31 (50.78)	4.17 (5.88)

Note: *The values in the parenthesis are initial values and values not in the parenthesis are the values after zeolite application.*
SFC: Soil Field Capacity; WP: Wilting Point; BD: Bulk Density; PD: Particle Density; P: Porosity; HC: Hydraulic Conductivity.
Source: *Boyraz and Nalbant (2015).*

widespread. Zeolites started gaining considerable attention towards soil application to enhance water use efficiency. Zeolite application in eight different soil types (N1 to N8) with thin (heavy) textured -N1, N2, N4 and N6; medium-thin textured -N3 and N5; and medium coarse (light) textured -N7 and N8 has increased the field capacity that ranged from 5.6 to 21.1% in these soils (Table 1).

Application of zeolite @ 10g per kg soil could maintain maximum percentage (8.4%) of field capacity and delay in PWP in sandy clay loam soils of Iran (Torkashvand and Shadparvar 2013). Important properties for managing irrigation, drainage, solute movement, soil temperature and soil aeration are soil hydraulic properties. (Al-Busaidi *et al.* 2008) reported that as the fine particles and micro pores of zeolite slowed the percolation of water in the soil, the infiltration rate of soil was negatively affected by zeolite and the reduced soil infiltration increased the content of water in the soil. In fine-grained calcareous loess soil, soil treated with zeolite could increase infiltration by 7–30% on gentle slope land and more than 50% on steep slope land. Even in the extreme drought condition, the zeolite treated soil could increase soil moisture by 0.4–1.8% as compared to increase in soil moisture by 5–15% in general situation (Xiubin and Zhanbin 2001). Even, zeolite and selenium applications can reduce the water deficit stress damages in crops like rapeseed (Valadabadi *et al.* 2010). (Gholizadeh-Sarabi and Sepaskhah 2013) reported that saturated hydraulic conductivity was increased at salinity levels of 0.5-1.5 dS m^{-1} in clay loam and loam with 8 and 4 g zeolite kg^{-1} soil, respectively, and at salinity levels of 3.0–5.0 dS m^{-1} with 16 g zeolite kg^{-1} soil. Highest water productivity of 0.258 kg m^{-3} was observed in rice field with 8 t ha^{-1} zeolite application (Sepaskhah and Barzegar 2010). (Colombani *et al.* 2014) quantified changes in flow and transport parameters induced by the addition of zeolites in a silty-clay soil and reported that NH_4^+-enriched zeolites increase the water retention capacity even in silty-clay soils, thus limiting water and solute losses. (Gholamhoseini *et al.* 2013) reported that under limited irrigation and nutrient management with zeolite (urea+ 21 % (w/w) zeolite) has shown maximum irrigation water productivity (0.81 kg m^{-3}), while the minimum value was found for the full irrigation and urea application (0.48 kg m^{-3}). Zeolite enhanced soil moisture content at each measured soil suction of 0, 5, 7.5, 10, 33, 100, 500, and 1500 kPa in sandy and clay loam soil of Iran (Haghshenas and Beigi 2010). (Ghazari 2015) found that for 10 mm of rainfall intensity, time of runoff beginning was 15 min in untreated soil and 30 min in zeolite (20%) treated soil, and zeolite treated soil could maintain higher soil water content compared to untreated soil.

Due to incorporation of the 1-3 mm zeolite tuff (@30,45,60 t ha^{-1} to a depth of 20-25 cm in apple orchard, increased the water-stable structural aggregates in the upper 20 cm layer by 24% after four years of incorporation (Jakab and Jakab 2010). Zeolite particle size has role in altering saturated hydraulic conductivity, hence it was reported that particle sizes >0.25 and 0.25-0.047 mm would be desirable in humid regions and arid and semiarid regions respectively (Huang and petrovic 1994). Soil aggregates are one of the important factors that influence the uptake of water as well as nutrients. (Aminiyan *et al.* 2015) reported that the mean weight diameter of water stable aggregates increased by 0.735 mm due to application of 30% nanozeolite and 0.685 mm due to 30% zeolite along with 5% alfa alfa straw application in both the cases. As the zeolite has the ability to enhance the soil aggregrate stability (Al-Busaide *et al.* 2007; Andry *et al.* 2007; 2008; Yamada *et al.* 2007), it can be used as soil amendment to reclaim sodic soils. (Moritani *et al.* 2010) found that incorporation of 10% artificial zeolite in sodic soils has resulted in improved wet aggregate stability which ranged 22.4% and 59.4% in two different soils. Application of zeolite along with chemical fertilizers or organic manure (Zeolite @ 7.5 t ha^{-1} + sugarcane filter cake @ 22.5 t ha^{-1}) has improved the soil properties with average assessment ranking 'good' and 'excellent' in terms of water stable aggregates and degree of soil aggregation in Vertisols (Cairo *et al.* 2017).

Nutrient use efficiency

Nutrient use efficiency can be improved by adequate agronomic nutrient management. Presently, more emphasis is on nitrogenous fertilizers for improving their nutrient use efficiency to safeguard soil and environmental quality. Nearly 30-50% of nitrogen from nitrogenous fertilizers is available to crops after their application and rest is subjected to various losses- leaching, mineralization, erosion and denitrification processes. These processes are also responsible in adding GHG to atmosphere. Zeolites (aluminosilicates) application can put a check on these processes. Zeolite has permanent negative charge on their surface, due to which they have high adsorption capacity for cations such as the ions of potassium, ammonia, calcium, sodium and magnesium and many other (Sprynskyy *et al.* 2005; Zabochnicka-Swiatek 2007). The effective diffusion coefficient was around 4–5×10−12 $m^2 s^{-1}$ for ammonium and sodium ions in clinoptilolite. Several researchers found an improvement in soil chemical properties due zeolite application (Latifah *et al.* 2017; Afrous and Goudarzi 2015; Troma *et al.* 2014; Gholamhoseini *et al.* 2013; Vilcek *et al.* 2013; Mohanraj 2013). The adsorption capacity of these ions onto zeolite was determined by isotherms and kinetics by several researchers and this property is used for various purposes such as waste water treatment etc. There are number of factors such as mass, particle size, initial concentration of ammonium ions of model solution, contact time, temperature and pH that affect adsorption efficiency of these zeolites (Mazloomi and Jalali 2016; Gorre *et al.* 2016). The removal efficiency of ammonium ions increases with increasing pH from 2 to 7 and then it decreases gradually from pH 8 to 10 with the maximum value being achieved at pH 7. There will be less competition between hydrogen and ammonium ions for adsorption/exchanging sites onto zeolite particles due to decrease of hydrogen ions in solution, with the increase in pH up to the maximum point (pH 7), hence, there will be an increase in adsorption of ammonium (Ugurlu and Karaog 2011).

However, due to reduction of ion-exchange potential, above pH 7, ammonium adsorption decreases (Wang and Zhu 2006). It is reported that by modifying zeolite surface with strong acids, sorption capacity for ammonium nitrate can be increased in zeolite (Shaikh and Chendaku 2016). The modification of natural zeolite includes pretreatment by grinding and sieving, modifying by sodium salt and finally, calcinations can cause a change in the pore size and surface area of zeolite, thereby increasing the ammonium ion uptake (Liang and Ni 2009). These parameters also help in understanding the role of zeolite towards enhancing nutrient use efficiency when applied to soil. Soil application of zeolite in combination with chemical fertilizers reduces nitrogen leaching (Aghaalikhani *et al.* 2012; Vilcek *et al.* 2013; Omar *et al.* 2015); slows down the mineralization process and release nutrients slowly (Li *et al.* 2013); reduces volatilization losses (Haruna Ahmed *et al.* 2008; Yore *et al.* 2013; Rech *et al.* 2017) and reduces GHG emissions to atmosphere (Čepanko and Baltrėnas 2011; Zaman *et al.* 2012). Also, it is reported to reduce soil run off (Behzadfar *et al.* 2017). The researchers had clearly seen the difference in the ammonia loss with chemical fertilizers and chemical fertilizers + zeolite in their incubation study (Ahmed *et al.* 2006; Palanivell *et al.* 2016) and reported that zeolite application reduces the ammonia loss when applied along with fertilizers (Table 2).

The application rates of zeolite vary with soil texture, nitrogen dosage and crop type. (Vilcek *et al.* 2013) reported that ammonia nitrogen in the top soil was 14-20% less in zeolite treated field

Table 2: Effect of zeolite application on reduction in NH_3 volatilisation losses

Sl. No.	Comparative treatments	Days of Incubation	% NH_3 loss Reduction	Soil/manure type or soil order	References
1	250 g of soil + 2.60 g granular urea vs 250 g of soil + 2.60 g granular urea + 12 g of zeolite	12	11.4	Sandy clay loam	Bundan *et al.* 2011
2	250 g of soil + 2.425 g urea vs 250 g of soil + compound fertilizer (4.85 g urea) + 27.04 g of zeolite	15	55.5	Sandy loam	Rabai *et al.* 2012
3	250 g of soil + complete fertilization vs 250 g of soil + complete fertilization + 60 g of zeolite	33	26.3	Typic Paleudults	Palanivell *et al.*, 2015
4	200 g manure vs 200g manure + 5% zeolite	5	81.8	Feedyard manure	Todd, 2014
5	Manure vs manure + 20 % zeolite	8	80.0	Broiler litter	Sheng *et al.* 2015

as compared to without zeolite after a month of fertilizer application. Slowly after three months, the content increased and reached 24-59% higher values in zeolite treated soils and reported that nitrification process in the soil became less intense due to zeolite application in Eutric cambisols. The zeolite reduces nitrification process by physically protecting the NH_4^+ ions by holding them in small molecular size of the open-ringed structure. The apparent nitrogen recovery has increased to 65% due to zeolite application compared to 40% in sole nitrogen fertilizer application in rice crop (Kavoosi 2007). The nitrogen use recovery efficiency was 84.8% due to application of zeolite @ 8t ha^{-1} and nitrogen @ 80 kg ha^{-1} in rice (Sepaskhah and Barzegar 2010).

Importance of organic manure in agriculture has increased and its application along with chemical fertilizers is highly encouraged for maintaining the soil quality and to sustain the crop. Composting is a way to convert agricultural farm waste into valuable organic amendments. The process of manure management and composting leads to N losses. Application of zeolite controls N loss and increases the manorial quality. So, apart from gypsum and SSP, zeolite can also be used to arrest the N loss from manure. For effectiveness, 14-21% zeolite should be added to fresh manure. The extent of reduction in total nitrogen and even phosphorus losses can be enhanced by combined application of zeolite and PAC or alum (Murnane *et al.* 2015) to manure. The inner chemistry behind zeolite use in enhancing nitrogen use efficiency is due to its high specific selectivity for ammonium (NH_4^+) that helps in holding NH_4^+ when it comes in contact with nitrogenous fertilizers, in this way it reduces the loss. Moreover, their small internal channels protect ammonium ions (NH_4^+) from excessive nitrification by microbes (Latifah *et al.* 2016). Thus, the nitrogen use efficiency of N fertilizers can be enhanced by using impregranation of N fertilizers with zeolite as it will act as slowly releasing carrier of fertilizers. The zeolite application not only helps in reducing NO_3^- leaching but also reduces P leaching; however, it helps in reducing NO_3^- leaching greater than P leaching (Gholamhoseini *et al.* 2013; Moharami and Jalali 2014; Shokouhi *et al.* 2015). Moraetis *et al.* 2016) reported that there was 18-fold increase in bioavailable K through kinetic experiment with zeolite addition to soil-compost mixture suggesting a high potassium affinity for soil-compost-zeolite mixture. (Antoniadis *et al.* 2012) reported an increase in N and P recovery efficiency of 30 and 4.02% due to zeolite application in acidic soil. Recently, it was that combined application of natural zeolite (Clinoptilolite) and compost had positive impact on soil properties as well on crop yield parameters (Kavvadias *et al.* 2019).

The application of natural zeolites has been reported to diminish nutrient leaching and increase crop water use efficiency (Gholamhoseini *et al.* 2013; Coltorti *et al.* 2012; Githinji *et al.* 2011; Saadata *et al.* 2012). Micronutrient use efficiency also increases due to zeolite application (Ozbahce *et al.* 2015; Najafi-Ghiri and Rahimi 2016). (Sheta *et al.* 2003) reported the ability of five natural zeolites and bentonite minerals to adsorb and release zinc and iron. (Islander *et al.* 2011) found that about 74.7% was extractable by DTPA after three successive extractions and rest was found retained by zeolite in case of Zn, while it was found that only 15.37% Mn was retained by zeolite and rest was extractable by DTPA. (Sheta *et al.* 2003) reported that natural zeolite has high affinity for Zn and Fe. Although recent and past studies reported that zeolite application enhanced the availability of primary, secondary and micronutrients in soils, their application is found effective in arid and semi arid regions where water and nutrient scarcity is high. (Morkou *et al.* 2015) reported that waste water nutrients can be recycled and used for microalgal and cynobacterial biomass production by using zeolite as a medium.

The zeourea and and nano-zeourea contained 18.5 and 28% of N and capable of releasing N up to 34 and 48 days, respectively, while the N release from conventional urea takes just 4 days (Manikandan and Subramanian 2014). Channels of 0.4 to 1 nm pore diameter in natural and nanoporous zeolite would be used for loading N and K (Bansiwal *et al.* 2006). (Sepaskhah and Barzegar 2010) reported that zeolite can be used as soil conditioner in lowland rice production system. Zeolites can decrease heavy metals mobility in soils hence can be used as amendment for heavy metal remediation apart from acting as excellent regulators of chemical fertilizers. They also enhance some of the biological properties in soils.

The treatment with 30% zeolite + 70% urea resulted in a positive effect on microbiological community in spring barley, soyabean and maize (Susana 2015). (Andronikashvili *et al.* 2008) reported that introduction of clinoptilolite containing tuffs into soils has positive effect on bacteria, fungi and actinomycetes population. There are reports stating that zeolite when mixed with soil, increased the arbuscular mycorrhizal (AM) fungi colonization. Arbuscular Mycorrhiza with zeolite promoted reforestation of post-opencast coal mine field as it increased the growth of tree species (Wulandari *et al.* 2016). The soil microbial biomass is a sensitive indicator of changing soil conditions. And addition of zeolite increased microbial biomass carbon compared to untreated soil (Usman *et al.* 2004). After 21 d of incubation, microbial biomass C was 17.9 mg g^{-1} C org^{-1} (with addition of zeolite). Zeolite also helps in carbon sequestration. According to (Aminiyan *et al.* 2015), application of zeolite (30%) and crop residues (5%) to wheat could maintain highest amount of organic carbon in light and heavy fractions. Light fraction of SOM is not only sensitive to changes in management practices, but also correlates well with the rate of N mineralization.

Specific channel size enables zeolites to act as molecular gas sieves where it can also be used for mitigating climate change. Periodical measurements of N_2O and N_2 emissions in fields due to application of cow urine or potassium nitrate (KNO_3) each applied at 200 kg N ha^{-1} rate with and without addition of lime or zeolite (clinoptilonite) showed that zeolite significantly reduced total N_2O emissions by 11% from urine treated soils; while no such effect was observed on N_2O emission in KNO_3 treated soils. (Wang *et al.* 2012) recommended straw or zeolite as amendment to reduce GHG emissions from duck manure as they found reduction of GHG emissions upto 27% (CO-equivalents) with reed straw or zeolite use in manure management. (Filcheva and Chakalov 2002) reported that zeolite (may be natural or modified or organozeolite) can improve the soil capacity for carbon sequestration.

CONCLUSION

Zeolites aid in water retention and improving moisture levels in soils. Their presence in soil improves soil physical properties and reduces runoff by increasing infiltration rate. Zeolite application along with fertilizers to soil will help in retention of nutrient and aid in controlled release which otherwise will get mineralized quickly and subjected to several losses. The most important nutrient- Nitrogen, mostly found deficit in all the soils, can be efficiently managed with zeolites. It also helps in improving manorial quality by holding the nitrogen content in organic manures. Like gypsum and SSP, zeolite can be used to arrest N losses from manure. It also serves as heavy metal remediating agent in soil. It also improves biological properties and reduces environmental pollution by reduced N_2O emissions. Zeolite enhances crop yields by modifying physical, chemical and biological properties, thereby, more residues gets added to soil which helps in carbon sequestration. Thus, zeolites addition can improve soil nutrient and water use efficiency.

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