A Review: Nanofertilizers for the Biofortification and Sustainable Agriculture

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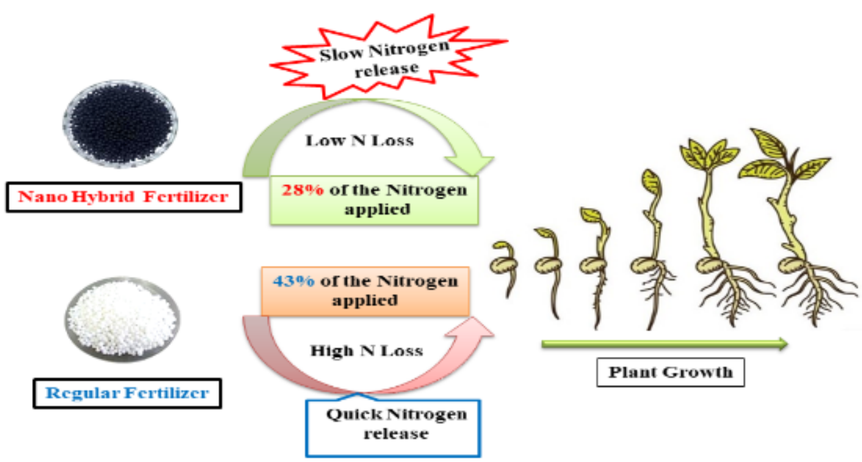
Abstract

Improving nutritional efficiency (NUE) by endangering environmental quality has been a central issue for agricultural food production (FPS) programs in order to sustain the growing population. Nanotechnology with nanoscale inputs for the production of nano agri-inputs (NAIPs) has already emerged as an important solution to address the problem of low or moderate nutrient utilization with minimal environmental impact. Recently, a few new hybrid nanofertilizer (HNF) formulations have been developed for biofortification and sustainable agriculture. Urea-modified hydroxyapatite was reported, which is a rich source of nitrogen, calcium, and phosphate. Gehan Amaratunga of the University of Cambridge has stated, "such fertilizers could reduce runoff and lead to harmful algal blooms in water bodies". The NAE (N Agronomic use Efficiency) for the urea-HA nanohybrids is 48%, while the NAE for pure urea is 18%, at the field level. However, these fertilizers are expensive and have not been shown to be commercially viable up to date. Recently, nanoparticles such as copper, iron, and zinc were incorporated into urea-modified hydroxyapatite to further increase the efficiency of the proposed fertilizer.



Keywords: Nanofertilizers, Biosafety, Sustainable Agriculture, Nutritional Efficacy, Biofortification

Introduction

Nanotechnology is a promising strategy with enormous potential to solve agriculture-related problems like the decline in land quality, low crop productivity, nutrient deficiency, and leaching losses (He et al., 2019). Agriculture as a source of food, feed, fodder, and fiber has always been very important in a world of declining resources and a growing global population (Brennan 2012). Ensuring food security in a nation with a small natural history has led to an ongoing search for new and less expensive solutions by the scientific community. Globally, sustainable and balanced food production systems are now needed in view of climate change. Nitrogen fertilizers are still a predominant input, which is important in increasing sustainable crop production. Its contribution to the use of the full crop yield is well documented. The efficiency of the nitrogenous fertilizer currently used is very low. The current efficiency of fertilizer is as low as 40-50% in N and 2-5% in micronutrients such as iron (Fe), manganese (Mn), zinc (Zn), copper (Cu), and boron (B). It provides a great opportunity to improve nutrient use efficiency (NUE). Excessive use of fertilizers increases the cost of the product while reducing the economic status of the farmers. In this regard, the overuse of fertilizers and their loss in the root zone is one of the major causes of soil, water, and air pollution. To address these problems, nanotechnology offers a template to improve the agricultural sector with the potential to increase food security, global food production, crop protection, plant and animal disease control, monitor crop growth, and reduce waste leading to "sustainable growth" (Gruère et al., 2011) (Frewer et al., 2011) (Pérez-de-Luque and Hermosín, 2013) (Prasad et al., 2014) (Ditta, 2012, Sonkaria et al., 2012). In this regard, a few steps have been taken worldwide. However, only focusing on research and development in advanced fertilizer is not sufficient to address the problem at an economically viable scale.

Recently, nanotechnology has emerged as an effective solution to address plant nutritional deficiencies through improved nutrient bioavailability and moderate environmental losses (Agrawal and Rathore, 2014). According to data from farm field trials, a 50% reduction in urea consumption allows yield to be maintained at ~7.9 tonnes/ha using nanohybrids, which is significantly higher than the yields for urea-only rice yields (7.3 tonnes/ha) using recommended urea levels (Kottegoda et al., 2017). Nanoscale materials can improve the efficiency of the fertilizer, while foliar applications could meet the nutritional needs of plants effectively according to their requirements. Therefore, the application of fertilizer to the crop instead of the soil saves farming systems from natural challenges caused by reduced nutrient utilization.

Fertilizer Application Status; Denial of Nutrition and Increasing Malnutrition

From a sustainable agricultural perspective, the use of nanotechnology in agriculture is considered one of the most important ways to improve crop production and feed the world's fastest-growing population (Lal, 2008). In India, grain production has registered impressive growth from 522 kg ha-1 in 1950/51 to 2,235 kg ha-1 in 2017/18 but declining fertilizer production relative to grain production shows a decrease in nutrient depletion in the soil and a decrease in NUE.

This has led to a reduction in plant reactions to plant nutrients from 15 kg of grain kg-1 NPK during the fifth phase (1974/79) to <6 kg of grain kg-1 NPK over the 11th period (2007/12) (Prasad et al., 2013) which also reduces 2.7 kg of grain kg-1 NPK in irrigated planting systems (Table 1) (Chaudhari et al., 2015).

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Table 1: Increasing food grain production *vis-a-vis* decreasing fertilizer response ratio

Year	Fertilizer Application (Kgha^{-1})	Yield(qha^{-1})	Fertilizer Response Ratio (Kg grain Kg^{-1} NPK)
1970	54	18	13.4
1975	67	19	11
1980	102	18	8.4
1985	140	20	7.0
1990	175	21	5.3
1995	178	21	4.8
2000	202	21	4.1
2005	218	21	3.7
2010	281	22	3.2
2015	254	22	2.7

Alternatively, fertilizer use efficiency (FUE), which is dependent on several factors, including nutrient uptake efficiency and soil health, determines our agricultural and environmental sustainability. The innovative fertilizer can successfully achieve [4R] targets to address the decline of FUE. Fertilizer use in India is uneven and deviated from urea-N. As a result, the NPK usage ratio has grown from 4:3.2:1 in 2009/10 to 7:2.8:1 in 2019/20 (Figure 1).

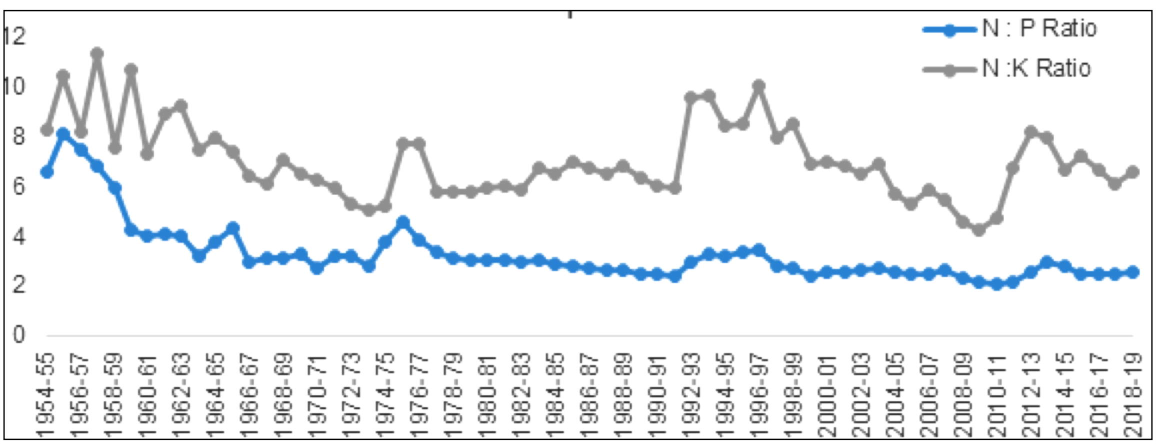

Figure 1. Fertilizer consumption ratio.

Nitrogen application should be measured at high application areas and increased at low application areas. With respect to secondary and micronutrients, widespread deficiency of sulfur (S), Zn and B were recorded in India (Table 2) (Raliya, 2019). At the state level, these differences are alarming. Analysis of regional level data, district level of soil samples (46,180) second and micronutrients by Tamil Nadu Agricultural University (TNAU), Coimbatore showed Zn deficiency at 42.0% followed by B at 19.9% and Cu at 16.7%. This suggests that the national micronutrient deficiencies should be improved in order to produce better crops and benefit farmers. Biofortification of micronutrients in plants will address the lack of microbes in humans and animals.

Table 2: Micronutrient deficient soil samples (%)

Micronutrient	Percentage
Zinc (Zn)	49
Boron (B)	33
Iron (Fe)	12
Molybdenum (Mo)	11
Manganese (Mn)	5
Copper (Cu)	3

Imprudent Application of Urea – Matter of a Growing Concern

The most common use of nitrogen fertilizer (N) and phosphorus (P) has become a major anthropogenic factor leading to global eutrophication problems in freshwater and coastal ecosystems (Correll, 1998, Conley et al., 2009). Farmers have been urged to reduce their use of urea by at least 25% in order to achieve better environmental quality and profitability. This problem needs to be addressed in terms of the sustainability of all agricultural production systems and biogeochemical cycles. Urea makes up 82% of the nitrogen fertilizer used in most Indian plants. About 33 million tons (Mt) of urea is used in various crops every year. Its use is expected to reach 37 Mt during the 2020/21 period (Table 3) (Raliya et al., 2015).

Table 3. Production, import, and consumption of urea during the last 5.

Year	Production (Mt)	Import (Mt)	Urea consumption (Mt)		
			Kharif	*Rabi*	Total
2016-17	24.20	4.97	14.36	15.26	29.62
2017-18	24.02	6.01	14.83	15.06	29.89
2018-19	23.90	7.56	15.45	16.57	32.02
2019-20	24.46	9.12	15.37	18.33	33.70
2020-21	15.15	6.61	17.78	2.43	20.21

There has been a steady increase in the production, importation, and use of urea in successive years. The importation of urea has increased over the past few years, reaching 9.12 Mt during 2019-2020, and is expected to be around 7.2 by April 2021 to January 2022 (Table 3). The subsidy burden for the importation of urea comprises 26% of the total urea grant paid annually by the Government of India (Table 4) (Raliya et al., 2016). To curb the growing use of urea in its prudent use, R&D efforts, the introduction of new products, and current policy measures are needed.

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Table 4. The subsidy paid by the Government of India for urea during the last 3 years (Rs. Crore).

Year	Indigenous urea	Imported Urea	Total
2016-17	40,000	11,257	51,257
2017-18	36,974	9,980	46,954
2018-19	32,190	17,155	49,345

Revolutionary Fertilizers - Key to Sustainability

Nanotechnology has great potential in the agricultural revolution, high efficiency, better bio-availability, bioactivity, and more (Gutiérrez et al., 2011). Newborn and new fertilizers without the need for enhanced nutrient intake offer benefits in terms of reducing natural traits. The fertilizer industry has emerged and introduced advanced fertilizers (EEFs) that cater to the niche market only. In India's heavily funded fertilizer market, new economically viable high-tech fertilizers can also be a real solution. Nanotechnology can be used to develop agricultural solutions, which can increase food production per unit of materials and resources. Nano fertilizers based on nano technology due to the magnitude of their benefits and the controlled production process using chemical, physical, and biological methods, have emerged as an effective option to fill this gap in the conventional and youth fertilizer market.

Nanofertilizers

Nanofertilizers, by definition, say "An integrated or modified form of traditional fertilizer, organic fertilizers or extracts of different plant, microbial or animal origin produced by chemical, physical, mechanical or biological methods with the help of nanotechnology with particles size range (1-100 nm) but not limited to that". Nanofertilizers or nano-encapsulated nutrients have properties to release nutrients effectively on demand that regulate plant growth and enhance target activity (DeRosa et al., 2010, Nair et al., 2010).

There are many reports where the use of nano fertilizer produced a positive effect leading to improved crop yields and reduced environmental pollution. Urea-modified hydroxyapatite nanoparticle encapsulated Gliricidia sepium nanocomposite showed slow and sustained release of nitrogen over time at three different pH values (Kottegoda et al., 2017). Manikandan and Subramanian (2014) reported that the nanoporous zeolite used in N fertilizer might be used as another strategy to improve the efficiency of N in the plant production system. Soils modified with metallic Cu – nanoparticles significantly increased the growth of 15-day lettuce seedlings from 40% to 91% (Shah and Belozerova, 2009). Nano and sub nano compounds control the release of nutrients in the fertilizer capsule made of natural kaoline and abandoned foam plastics (Liu et al., 2006). According to a recent study, nanotechnology has the potential to modify agricultural systems (Manjunatha et al., 2016) that, allow for the gradual release of nutrients for the benefit of crops, and ultimately increase the rate of crop production by lowering the environmental impact (Scott and Chen, 2013). The paradigm shift from traditional methods of crop production to nano and advanced technologies can increase agricultural productivity with improved nutrient use efficiency, the efficient and effective use of resources that ensure nutrient protection, increase productivity, boost farmers' economy, generate agricultural value, and decrease environmental pollution is a burning need (Subramanian and Tarafdar, 2011).

Nanofertilizers, because of their properties, play a major role in sustainable agriculture (El-Ramady, 2014). On the nanoscale, the physical and chemical properties of nano-fertilizer are flexible and different from their macro counterparts. Due to their high surface area to volume ratio, they have high availability and absorption. The particle size of nano fertilizers is in the range of 1-100 nm at least in one dimension, which facilitates better absorption onto soil or leaves, resulting in the longer accumulation of nutrients and production of more photosynthate and biomass needed in healthy plants.

Nanofertilizers have advantages such as low demand and minimal transport and installation costs which result in less accumulation compared to conventional soil fertilizers. The actual use of nano fertilizer delivery systems has recently been introduced to agriculture (Kuzma and VerHage, 2006, Roco, 2011, Scott and Chen, 2013). Foliar-added nanofertilizers increase NUE and the nutritional value of plants through bio-fortification. Replacing traditional fertilizer with nano fertilizer is beneficial as it releases nutrients into the soil gradually and in a controlled manner, thus preventing water and air pollution (Naderi and Danesh-Shahraki, 2013). Nano fertilizers can be applied into the soil (to be taken by plant roots), or used as a foliar spray (to be taken by the leaves)(O'Neill et al., 2014), or both (Yan et al., 2018).

Nano Nitrogen Fertilizers

Previously it was reported that urea loaded with zeolite chips (Millan et al., 2008) and N-containing nanocomposites (Jinghua, 2004) were used to induce slow N release and thus increase plant N intake. It was reported that hybrid nano fertilizer has the potential for a slow release of Ca2+, PO43−, NO2−, NO3−, Cu2+, Fe2+, and Zn2+ nutrients. This nanofertilizer was applied on Abelmoschus esculentus and showed maximum nutrient use efficiency and higher yields (Tarafder et al., 2020).

Other materials used for the same purpose include sources of nutrients coated with thin polymer films and nutrients embedded in nanoporous materials (Rai et al., 2012). Nano nitrogen based on the principles of nanotechnology, provides another new way to move farmers away from urea. Hydroxyapatite nanoparticle is a promising and top research area to obtain N and P slow-release properties when combined with urea. Also, it has been used for macro-micro plant nutrient delivery. Therefore, it is worth mentioning a few recent pieces of research conducted in this field (Tarafder et al., 2020). The nanoscale benefits of nitrogen particles should be utilized to effectively optimize the nitrogen demand in plants. Accuracy and targeted use of nitrogen by infusing nano nitrogen with leaves reduce urea loss; increase the efficiency of nutrient uptake; and deal with environmental issues of soil, air, and water pollution. It leads to better crop yields with less nitrogen application in each area, thus, leading to a better farm economy.

Spraying nano nitrogen at a rate of 2-4 mL per litre of water at critical stages of crop growth triggers crop response, fulfils its nutritional needs, and improves nutrient uptake in the rhizosphere (Liu and Lal, 2015) (Preetha and Balakrishnan, 2017) (Kumar et al., 2021, Lahari et al., 2021). When nanofertilizer is sprayed onto leaves, N penetrates easily through the stomata due to its nano size (<100 nm) (Wang et al., 2013). It is distributed to other parts of the plant through phloem translocation and is absorbed into the body, such as proteins, amino acids, etc., according to the need of the plant.

Nano nitrogen particles with a small size (20 nm) can easily penetrate through the cell wall and reach the plasma membrane. Particles with a large size (20 - 50 nm) can enter through the cavities of the abdomen. They are also transported by phloem cells via plasmodesmata (up to 40 nm in diameter) to other plant parts. They can bind company proteins using aquaporin, ion channels, and endocytosis and are digested inside the plant cell. Therefore, the use of nanoscale particles, such as nanoparticles containing nitrogen through foliar, leads to better absorption and entry of nitrogen into the plant system. It improves

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metabolic processes and promotes meristematic activities that lead to higher apical growth and photosynthetic space of the leaves. The combined effect of all these activities ultimately leads to higher yields and reduced nitrogen deficiency within the plant systems.

Nano Zinc and Nano Copper as micronutrients

Zinc is a micronutrient essential for plants, animals, and humans to grow well and develop. It was reported that HNF (hybrid nanofertilizer) of urea-modified hydroxyapatite increases Cu2+, Fe2+, and Zn2+ nutrient uptake efficiency more than the commercial fertilizer like single superphosphate (SSP), triple superphosphate (TSP), urea, nitrogen−phosphorous−potassium (NPK), monoammonium phosphate (MAP), and diammonium phosphate (DAP) within a few days (Tarafdar et al., 2020). Similarly, Cu is also one of the eight most important plant nutrients needed for plant operations and healthy seed production. Copper deficiency can lead to increased susceptibility to fungal and bacterial pathogenic diseases, which can lead to significant yield losses. Current fertilizer options do not perform well due to their complexation in soil. Long-term use disturbs the pH of the soil and also reduces microbial activities, which are naturally good for plants.

Recently, Ekanayake et al. reported an alginate-based hydrogel on ZnO and CuO nanoparticles (Ekanayake and Godakumbura, 2021). The main purpose of nano zinc and nano copper is to replace their common analogs of fertilizers using between 2-5%, increase plant production, and improve their quality with agronomic intensification. In addition, nano zinc also helps the plant absorb more P, leading to better physical growth and bringing similarity to the shape and size of the fruit. Similarly, nano copper builds up plants' natural defences against harmful pathogens of fungi and bacteria that affect their overall growth and development. If nano zinc and nano copper are sprayed on the leaves due to their small size, they can be easily absorbed by the plant directly or through the stomach holes. Upon entering the leaves, these are then distributed to plant parts by phloem transfer and metabolic according to the plant's needs.

For best results, nano zinc or nano copper is sprayed twice in the critical stages of plant growth, first in the first growth stages and second in the pre-flowering phase and at a rate of 2-4 mL per plant water. Nano zinc and nano copper can be mixed together during spraying if needed; if not, they can be used separately.

Multi-location- Multi Crop 'On Station' and 'On Farm' Trials of Nano fertilizers

Experimental tests performed during rabies in Zaid 2019-20 on various crops such as paddy, wheat, mustard, maize, tomatoes, cabbage, cucumber, capsicum, onions, and regions have recorded encouraging results. A summary of the State Agricultural University and Krishi Vigyan Kendra Knowledge Network experiments shows that nano nutrients can improve crop yields without making significant use of subsidized fertilizer (Raliya et al., 2015).

Tests conducted by South Asian University Research Centers show that a 50% reduction in urea is possible through the application of nanoparticles. All traits contributing to growth and yield were maximal and significantly better in the treatment receiving 2 nanonitrogen sprays or alternative combined nanonitrogen, nano zinc and nano copper sprays with 50% reduction in nitrogen and zinc wherever recommended. The ICAR-Indian Agricultural Research Institute, New Delhi states that nano fertilizer (alone or in combination) when used in fixed doses of fertilizer, can lead to a 50% reduction in nitrogen fertilizer usage. A reduction of 25% of nitrogen fertilizer in wheat and 50% of nitrogen fertilizer in the nostrils is possible with two nano nitrogen sprays. Similar or better results have been communicated to other research institutes and SAUs. Increased numbers of active tillers, high growth, and biomass yields as well as grain and grass yields, are recorded in the treatment of nano fertilizers.

Multi-Location – Multi Crop Farmer Field Trials (FFTs) of Nano fertilizers Farmer's field tests have confirmed that nano nitrogen leads to a reduction in urea consumption and a better economy for farmers. 8719 successful Farmer Field Trials - FFTs on 94 plants conducted and closely monitored by

ICAR- KVKs in all 28 States / UTs and recorded (Raliya et al., 2017), and an average yield of 7-8% was recorded at 50% less than the inclusion of urea.

Table 5: Effect of nano fertilizers on crops (Table derived from (Kumar et al., 2020)

Crop (Data in parenthesis are the number of trials)	Parameters	Farmer fertilizer practice (FFP)	FFP - 50% N +2 sprays of nano nitrogen	FFP + 2 sprays of nano zinc	FFP + 2 sprays of nano copper	FFP(-50% N)+ 1 spray of nano nitrogen + 1 spray of nano zinc + 1 spray of nano copper
Wheat (480)	Lowest yield (kg ha^{-1})	2250	2400	2370	2370	2380
	Highest yield (kg ha^{-1})	6410	6760	6610	6580	6875
	Mean yield (kg ha^{-1})	4330	4580	4490	4475	4628
	Response over FFP (kg ha^{-1})	-	250	160	145	297.5
	Per cent increase over FFP	-	5.77	3.7	3.35	6.87
	Net return over FFP (Rs. ha^{-1})	-	4813	3080	2791	5727
Barley (9) L owest yield (kg ha^{-1})))	Lowest yield (kg ha^{-1})	3200	3380	3300	3250	3350
	Highest yield (kg ha^{-1})	5260	5620	5730	5790	5900
	Mean yield (kg ha^{-1})	4230	4500	4515	4520	4625
	Response over FFP (kg ha^{-1})	-	270	285	290	395
	Per cent increase over FFP	-	6.38	6.74	6.86	9.34
	Net return over FFP (Rs. ha^{-1})	-	4118	4346	4423	6024
Maize (4) L owest yield (kg ha^{-1})	Lowest yield (kg ha^{-1})	4100	4300	4400	4100	4500
	Highest yield (kg ha^{1})	5500	6000	5700	5550	6000

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	Mean yield (kg ha^{-1})	4800	5150	5050	4825	5250
	Response over FFP (kg ha^{-1})	-	350	250	25	450
	Per cent increase over FFP	-	7.29	5.21	0.52	9.38
	Net return over FFP (Rs. ha^{-1})	-	6160	4400	440	7920
Chickpea (27)	Lowest yield (kg ha^{-1})	1437	1566	1498	1466	1677
	Highest yield (kg ha^{-1})	2500	2700	2650	2600	2650
	Mean yield (kg ha^{-1})	1969	2133	2074	2033	2164
	Response over FFP (kg ha^{-1})	-	165	106	65	195
	Per cent increase over FFP	-	8.36	5.36	3.28	9.91
	Net return over FFP (Rs. ha^{-1})	-	8019	5143	3144	9506
Urdbean (3)	Lowest yield (kg ha^{-1})	1650	1850	1925	1750	1975
	Highest yield (kg ha^{-1})	1700	1850	2000	1800	2150
Lowest yield (kg ha^{-1})	Mean yield (kg ha^{-1})	1675	1850	1963	1775	2063
	Response over FFP (kg ha^{-1})	-	175	288	100	388
	Per cent increase over FFP	-	10.45	17.16	5.97	23.13
	Net return over FFP (Rs. ha^{-1})	-	9975	16388	5700	22088
Mustard (70)	Lowest yield (kg ha^{-1})	1100	1200	1170	1120	1180
	Highest yield (kg ha^{-1})	4200	4300	4500	4200	4600
Lowest yield (kg ha^{-1})	Mean yield (kg ha^{-1})	2650	2750	2835	2660	2890
	Response over FFP (kg ha^{-1})	-	100	185	10	240
	Per cent increase over FFP	-	3.77	6.98	0.38	9.06
	Net return over FFP (Rs. ha^{-1})	-	4425	8186	443	10620
Potato (187)	Lowest yield (kg ha^{-1})	13250	15000	14000	14000	16000
	Highest yield (kg ha^{-1})	61200	64300	61800	61800	62700
Lowest yield	Mean yield (kg ha^{-1})	32298	35414	33568	33824	34798
	Response over FFP (kg ha^{-1})	-	3117	1270	1526	2500
	Per cent increase over FFP	-	9.65	3.93	4.72	7.74
	Net return over FFP (Rs. ha^{-1})	-	31165	12702	15259	24997
Lentil (5)	Lowest yield (kg ha^{-1})	625	680	665	660	650
	Highest yield (kg ha^{-1})	2019	2056	2032	2038	2024
L	Mean yield (kg ha^{-1})	1677	1715	1696	1696	1689

owest yield (kg ha^{-1})	Response over FFP (kg ha^{-1})	-	37	19	19	12
	Per cent increase over FFP	-	2.23	1.11	1.13	0.72
	Net return over FFP (Rs. ha^{-1})	-	1795	893	912	576

The results of 600 experimental farm trials of 8 crops conducted during the winter of 2019-20 in various regions of Rajasthan showed that the amount of urea used by farmers to supply nitrogen to crops could be effectively reduced to half (Table 5) (Kumar et al., 2020). Yields achieved by 50% less nitrogen and 2 nano nitrogen sprays on stagnant plants provided higher yields than those applied to most of the 8 plants tested in these experiments. Apart from this, the effect of nano-zinc and nano-copper was also tested. Since the deficiency of these micronutrients is not as universal as nitrogen, the critical responses to these nanofertilizers depend on the magnitude of the deficiency of certain micronutrients and the nature of the plant.

The results of 730 field demonstrations held in various districts of Uttar Pradesh on 12 farmers' plantations of wheat, chickpeas, urad beans, maize, barley etc. proved that with the use of nano nitrogen, the amount of urea used by farmers to supply nitrogen to their crops could be effectively reduced (Table 5). Yields obtained by 50% less nitrogen compared to N used under farmer's fertilizer practice (FFP) and the application of 2 nano nitrogen sprays to standing plants provided higher yields than FFP in most plants tested in these demonstrations.

These results clearly confirm that with the use of nano-fertilizer, NUE can be significantly improved as it has been shown to have 50% savings in urea with 2 sprays of nano nitrogen. Nanofertilizers are considered a new way to conserve nutrients, especially nitrogen, and to protect the environment.

Conclusion

Nanotechnology-based solutions are even more important in many countries with increasing populations. Increasing consumer awareness about food tracking and the friendliness of farm work requires novel and innovative solutions such as nanofertilizers.

Nano fertilizers should be considered entirely an option to address the challenges facing modern agriculture. It is time for nano-fertilizer to be introduced as an 'informed choice' to address the ongoing barriers to sustainable agriculture and agricultural profitability.

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