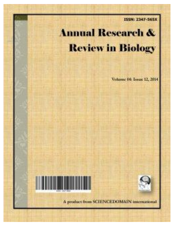

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Advanced Forms of Sulphur Formulations for Improving Use Efficiency in Crop Species

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Authors' contributions

This work was carried out in collaboration between all authors. Authors SS and DS designed the study and wrote the first draft of the manuscript. Authors M and MB managed the literature searches. Authors SR and RKS explored the importance of improved sulphur fertilizers. Authors SK and AR promulgated the idea and checked the whole manuscript. All authors read and approved the final manuscript.

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ABSTRACT

Widespread incidences of sulphur (S) deficiency in agricultural crops and soils have been reported globally. To meet this gap, various new forms of S fertilizers are available in the market, and they are in the process of testing for their validations in varied type of crop species and soils. A current global trend of using these advanced S formulations for plant production has emerged to overcome the challenges of S nutrition in crops. This paper highlights the potential benefits of improved S

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fertilizers in agriculture as compared to conventional S fertilizers. However, there is also a need for increasing awareness among the growers to recognize the importance of S (fourth primary plant nutrient) and simultaneously exploring the options for elevating the efficiency of S.

Keywords: Sulphur; sulphur deficiency; sulphur nutrition; improved sulphur fertilizers; conventional S fertilizers.

1. INTRODUCTION

Sulphur (S) balances in agricultural soils have become a significant concern for the agriculturists all over the world because they are mostly negative. Such situation, i.e., declining S levels in soils have been attributed to strict environmental rules on industrial S emissions [1,2], use of high analysis S-free fertilizers and high yielding varieties, intensive cropping [3,4], and limited or no use of organic manures [5] and S-containing pesticides [6,7]. Tripathi [8] claimed that at least 57 million ha out of 142 million ha arable land in India is deficient in S. Several researchers have reported S deficiency in various states of India, viz., Uttar Pradesh [9], Uttarakhand [10], Odisha [11], Madhya Pradesh, Maharashtra, Jharkhand, West Bengal, Andhra Pradesh, and Karnataka [12]. Similarly, the deficiency has been observed in many regions of the world due to continuous depletion of native S reserves. Thus, the importance of S was recognized very quickly, and S-containing fertilizer products were introduced into the markets.

Sulphur is the fourth primary plant nutrient required for the normal growth of plants, and it plays an important role in many plant processes; which indicate that plant metabolism is dependent upon S and its deficiency will cause primary metabolic impairment. Plant S concentrations are found to be lower than nitrogen (N) [13], but quite similar to that of phosphorus (P) [14]. It is essential for the synthesis of amino acids (cysteine and methionine) which are the basic structural units of protein molecules and constituent of several enzymes, chlorophyll, oils, and vitamins [15]. It regulates the activity of nitrate reductase in plants [15], and also helps in microbial fixation of atmospheric N [16]. The behaviour and reactions of S in the soil are very similar to those of N which are mainly dominated by the organic or microbial fractions of soil [17]. Its deficiency often becomes a major hindrance for the sustainable growth and productivity of field crops [18]. Successful crop production of not only of oilseeds, pulses, vegetables, and forages but also of many cereals are dependent on S nutrition.

Generally, the use efficiency of S fertilizers is very low (8-10%) [19]. Until now traditional S fertilizers, viz., ammonium sulphate, single super phosphate (SSP), gypsum, etc. were common in use, while S fertilizers like elemental S (S°), bentonite S, micronised S, etc. and their advanced formulations are becoming popular nowadays. The inorganic fertilizers containing S as sulphate (SO_4^{2-}) and S° fall in the category of conventional and advanced S fertilizers, respectively. Elemental S came into the demand because of high concentrations of S (70-100%), negligible leaching and run-off losses, continued residual effects on the S nutrition of the subsequent crop, and low transport and application costs (for it is 100% S) [1,20]. Sulphate fertilizers provide S to plants quickly, but they are susceptible to leaching losses [21]. Plant takes S in SO_4^{2-} form, so the S° fertilizers must be converted into that form through the process of oxidation which is mediated the by S-oxidizing microbes [22]. A similar process is also required for bentonite and micronised S fertilizers for solubilisation of S. These fertilizers have been termed as slow-release fertilizers, and they have the advantage of the long-term supply of S to crops.

This literature review provides insights of the potentials of improved S fertilizers in the management of S nutrition in agricultural crops for enhancing their use efficiencies and qualitative yields.

2. SULPHUR CHEMISTRY IN SOIL

Sulphur is found as sulphides in rocks (igneous and metamorphic), but in the soil, it occurs in combination with organic matter, and it is also present in industrial wastes, oceans, and as gas in atmosphere. Hence, it exists in soil in both inorganic and organic forms. However, the proportion of inorganic to organic S is dependent upon soil texture, pH, calcium carbonate, organic matter, and other soil characteristics [23]. Out of the several inorganic forms like sulphide (S^{2-}), elemental sulphur (S°), sulphate (SO_4^{2-}), etc., plants absorb SO_4^{2-} for their growth and development. Organically bound S is more common in most agricultural soils than inorganic S [24]. In Indian soils, about 30% of total S is found with organic combination in alluvial soils, whereas it reaches to 70% in the Mollisols of *tarai* region [9]. As these organic compounds are unavailable to plants, they are converted to inorganic SO_4^{2-} by biochemical or microbiological mineralization before plant uptake [25]. Highly weathered soils (Oxisols and Ultisols) are generally deficient in S [26]. Precipitation of S is found in the form calcium, magnesium or sodium sulphate. Accumulation of pyrite (FeS_2) is seen in marshy lands with large amounts of sulphide metals where the S compounds are oxidised to SO_4^{2-} with decrease in pH after drainage of water. Soil solution SO_4^{2-} is in equilibrium with the solid phase forms [17], and often adsorbed to clay minerals and sesquioxides like phosphate, but the binding strength for SO_4^{2-} is less strong [27]. Overall S balance in the soil-plant system is dependent upon plant uptake, leaching, mineralisation/immobilisation, and volatilisation.

3. THE REQUIREMENT OF SULPHUR FOR CROP NUTRITION

Sulphur plays an important role in improvement of yield and quality of crops [28-30]. It is linked with N metabolism, and its application increases the uptake of N by plants [31,32]. Besides N, it also enhances the uptake of other beneficial nutrients like phosphorus, potassium, and zinc, and checks the uptake of toxic elements like sodium and chlorine [14,33]. Plants deficient in S have less resistance to biotic and abiotic stresses [34,2]. Moreover, the primary and secondary metabolism involving amino acids, carbohydrates, glucosinolates, and biosynthesis of many other secondary compounds are moderated in plants during S stress [13,2]. Visual symptoms of S deficiency include chlorosis on younger leaves and reduced plant growth (premature defoliation, thin and woody stem, reduced leaf size, stunted growth, etc.); and its toxicity symptoms include chlorosis, interveinal necrosis, mottling in young leaves, inhibition of apical growth, bluish green appearance of older leaves, busy appearance of lateral branches, and ultimately reduction in growth [35].

The requirement of S nutrition varies with the type of crops (Table 1). It is generally in the order of *Cruciferae* > *Leguminosae* > *Gramineae* [16]. Oilseed crops are known to deplete the S content of soil as their uptake for production of seed is very high [36]. Sulphur required to produce one ton of seed is about 3-4 kg for cereals, 8 kg for pulses, and 12 kg for oilseeds [37,15]. Walker and Booth [38] estimated crop removal of S for oilseed rape is 20-30 kg ha^{-1}, but for cereals, it is 10-15 kg ha^{-1}. Plant S concentrations varies between 0.1 and 0.5% [17]. The content is generally high during vegetative growth stages compared to maturity [39]. The N:S ratio is also an important factor which influence the S requirement of plants since both N and S are closely linked in synthesis of protein, addition of N must be considered in scheduling S fertilization [40]. Application of 30 and 60 kg S ha^{-1} in spring oats produced 17.4 and 8.2 kg forage per kg S, respectively, when applied without N, but yielded 31.7 and 15.7 kg forage per kg S, respectively, when applied with N [41]. Generally, it is established that one part of S is required for every 15 parts of N, and their ratio lies in the narrow range of 15:1 [42]. However, fertilizer recommendation also depends on climatic conditions, locations, soil types, and cultivars.

The production of oilseed crops and their quality is significantly influenced by supply of proper S nutrition [43,33]. Quality attributes of oilseeds like oil content [44,45], glucosinolate concentrations [46,47], protein concentrations [48,47], etc. were found to increase with appropriate S application. Similarly, the growth and development of many crop species are affected; and are responsive to S supply. Sulphur stress diminish the baking or breadmaking quality of wheat [49,50]. Salvagiotti et al. [51] observed high nitrogen use efficiency in wheat as S addition enhanced the N uptake. Biological N_2 fixation and consequently higher dry matter accumulation in legumes is influenced by S fertilization because of increased nodulation and better root growth [52]. Tripathi et al. [53] reported that application of 45 kg S ha^{-1} recorded the highest grain and straw yield, yield attributing characters (number of pod, pod length, seed per pod, and 1000 seed weight) and protein content of mungbean. Sulphur compounds in onion and related alliums (garlic, leek, shallot, and chives) are responsible for the flavor profile or pungency of these crops [54].

Table 1. Crop demand for S in different agro-climatic conditions as evidenced from different field experiments

Sl. No.	Crop	Sulphur requirement	Agro-climatic conditions and location	Soil	Reference
A.	**Cereals**				
1.	Aromatic rice	60 kg ha^{-1}	Hot semi-arid (New Delhi)	Sandy clay loam	[55]
2.	Rice	40 kg ha^{-1}	Humid subtropical (Patna)	Silty clay loam	[56]
3.	Wheat	50 kg ha^{-1}	Hot semi-arid (Sheikhupura, Pakistan)	Saline sodic (Clay loam)	[14]
4.	Wheat	30 kg ha^{-1}	Subhumid subtropical (Udham Singh Nagar)	Sandy clay loam	[7]
5.	Maize	60 kg ha^{-1}	Tropical (Udaipur)	Clay loam	[57]
6.	Maize	60 kg ha^{-1}	Tropical wet and dry (Hyderabad)	Sandy loam	[58]
7.	Sorghum	30 kg ha^{-1}	Hot moist semi-arid subregion of Andhra Pradesh (Mahabubnagar, Nalgonda, and Kurnool)	Alfisols	[59]
8.	Barley	30 kg ha^{-1}	Semi-arid (Jobner)	Sandy loam	[60]
B.	**Oilseeds**				
1.	Mustard	40 kg ha^{-1}	Tropical (Budaun)	Sandy loam	[45]
2.	Indian mustard	45 kg ha^{-1}	Subtropical (Research Farm, BCKV, Nadia)	Clay loam	[61]
3.	Indian mustard	40 kg ha^{-1}	Humid subtropical (Varanasi)	Clay loam	[62]
4.	Groundnut	45 kg ha^{-1}	Tropical wet and dry (Seethampeta)	Sandy clay loam	[63]
5.	Sesame	45 kg ha^{-1}	Tropical wet and dry (Karaikal)	Sandy clay loam	[64]
6.	Sesame	45 kg ha^{-1}	Humid subtropical (Rawalpindi, Pakistan)	Sandy clay loam	[33]
7.	Sunflower	20 kg ha^{-1}	Humid subtropical (Rawalpindi, Pakistan)	Sandy clay loam	[65]
8.	Canola	40 kg ha^{-1}	Continental (Nowshera, Pakistan)	Sandy loam	[47]
C.	**Pulses**				
1.	Mungbean	45 kg ha^{-1}	Humid subtropical (Varanasi)	Sandy clay loam (Inceptisol)	[53]
2.	Black gram	40 kg ha^{-1}	Humid subtropical (Kumarganj, Faizabad)	Sandy loam	[66]
3.	Soybean	40 kg ha^{-1}	Humid subtropical (Ambikapur)	Sandy loam	[67]

Table 1. *Cont.*

4.	Soybean	20 kg ha^{-1}	Tropical wet and dry (Dharwad)	Vertisol	[68]
5.	Soybean	40 kg ha^{-1}	Tropical wet and dry (Hyderabad)	Sandy loam	[69]
6.	Pigeonpea	20 kg ha^{-1}	Subtropical (Nagpur)	Dark clay	[70]
7.	Sunnhemp	40 kg ha^{-1}	Subtropical (Pratapgarh, UP)	Sandy loam (Inceptisol)	[71]
D.	**Sugar crops**				
1.	Sugarcane	80 kg ha^{-1}	Subtropical (Shahjahanpur)	Sandy loam	[72]
2.	Sugarcane	60 kg ha^{-1}	Humid subtropical (Lucknow)	Sandy loam	[73]
E.	**Tuber crops**				
1.	Potato	40 kg ha^{-1}	Subtropical (Etawah)	Alluvial	[30]
F.	**Vegetables**				
1.	Head cabbage	40 kg ha^{-1}	Temperate (North-East Poland)	Loamy Sand	[74]
2.	Onion	40 kg ha^{-1}	Temperate (North-East Poland)	Loamy Sand	[74]
3.	Onion	40 kg ha^{-1}	Humid subtropical (Ranchi)	Acid Alfisol	[75]
4.	Onion	40 kg ha^{-1}	Subtropical (Old Brahmaputra Flood Plain, Mymensingh, Bangladesh)	Silty loam	[76]
5.	Garlic	40 kg ha^{-1}	Humid subtropical (Ranchi)	Acid Alfisol	[75]

4. SULPHUR DEFICIT ACROSS DIFFERENT REGIONS

Analysis of 135,000 soil samples of India revealed that about 42.3% samples are deficient in available S, and currently, 300 districts are suffering from S deficiency as compared to 70 districts estimated in 1991 [77]. Severe condition have been noticed in the soils of semi-arid tropical regions [78,79] and eastern Gangetic plains of the country [9,80]. Singh and Kumar [80] reported that pulse-growing upland red soils are comparatively higher in S deficiency (92.0%) than alluvial (48.0%) and black soils (37.0%) soils of eastern Uttar Pradesh (UP). Recently, Pandey et al. [30] found about 62% deficiency of S in potato soils of Etawah district of the state (UP). Widespread deficiency of S has also been reported in the UK and other European countries [46,1,81,6]. The Sulphur Institute (TSI) estimated the world deficit of S as a plant nutrient to reach 16.74 million tons (Mt) per year in 2015 as compared to 13.3 Mt per year in 2005 (Table 2). Regional S deficit is highest in Asia with leading countries like China and India [82]. Crop removal of S in India is estimated to be about 1.26 Mt, but its recovery through fertilizers is only about 0.76 Mt [83].

5. NEW SULPHUR FERTILIZERS

The properties of S° led the manufacturers to make new S formulations from it, which include not only fertilizers but also various pesticides. Bentonite S generally contains 90% S° and 10% bentonite clay. Micronised S also contains a high amount of S° (usually > 80%). These S° fertilizers are granular in nature, and often consist of prills or pastilles. The bentonite clay is used as a binder. Besides these degradable solid products, liquid suspensions (colloidal S) and anhydrous ammonia S formulations are also available in the market as S° fertilizers [85]. As they are insoluble in water, they must be oxidised by the microorganisms in soil to SO_4^{2-} form for plant absorption. The rate of oxidation depends on certain factors such as aeration, moisture, temperature, and microbial population known as the soil environmental factors, and also on the properties of fertilizer like particle size of S° [86,1,20], dispersion of the S° particles in soil, and composition of S°-containing fertilizers [85].

Table 2. Emergence of global deficit for S as a plant nutrient as estimated by The Sulphur Institute (TSI)

Sl. No.	Region	Sulphur deficit (million tons per year)			Reference
		2005	2010	2015	
1.	Asia	5.4	5.8	6.7	[84,82]
2.	China	2.0	2.4	2.6	
3.	India	1.5	1.8	2.1	
4.	Africa	1.3	1.5	1.6	
5.	North America	1.3	1.5	1.5	
6.	Latin America	0.7	0.9	1	
7.	Western Europe	0.7	0.5	0.8	
8.	Eastern Europe	0.4	0.5	0.44	

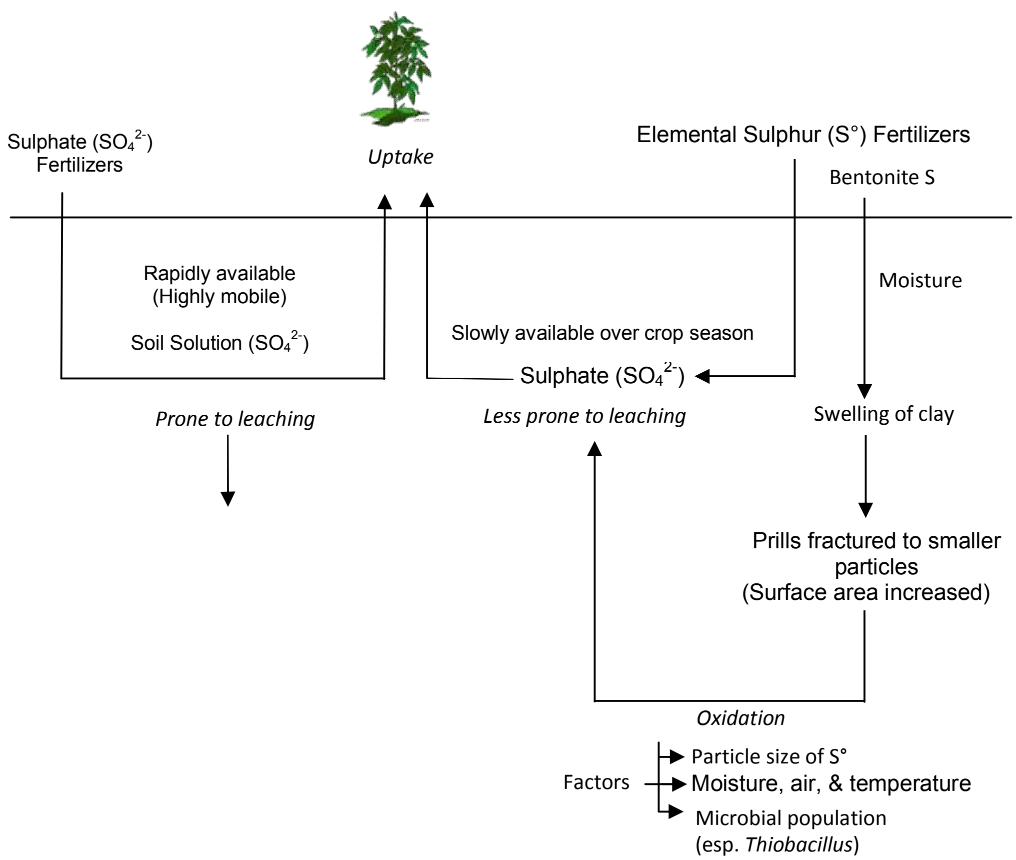

Fig. 1. Mechanisms involved in release of plant-available S from S° fertilizers as compared to SO_4^{2-} fertilizers

Reduction in particle size, i.e., finely ground S° increases the microbial colonisation due to increase in surface area [17]. Microbial S oxidizers include autotrophic chemolithotrophs, obligate chemolithotrophs, and heterotrophs; bacteria of the genera *Thiobacillus*, *Thiomicrospira*, *Thiosphaera*, etc.; and fungi of the genera *Alternaria*, *Penicillium*, *Aspergillus*, etc. [87]. When the S° fertilizers come in contact with soil water, the clay (bentonite) swells by absorbing water, which causes the disintegration of prills and release of small S° particles [1,4; Fig. 1]. After their division into fine fragments, the microbial oxidation starts; the process being rapid in warm, moist, and well-aerated soils [40]. Janzen and Bettany [88] evaluated the release pattern of plant-available S from two soluble sources (thiosulphate and sulphate) and four elemental S forms (S powder, S-coated urea, S suspension, and S-impregnated urea) in a growth chamber by using five successive harvests of rapeseed. They found thiosulphate to

Table 3. Effect of improved S fertilizers on yield/yield attributes, S uptake/use efficiency, and quality parameters of different crop species

Sl. No.	Improved sulphur fertilizer	Rate of sulphur application	Crop	Yield/yield attributes	Sulphur uptake/use efficiency	Oil/protein content	Experiment condition	Reference
1.	Micronised S° – 80% finely divided S° particles (5-8 µm) + 20% inert agent coating to aid dispersal	240 mg pot^{-1}	Spring wheat (cv. Canon)	Average increase in grain yield by 36% over control	Significantly higher (on average 164%) than control	-	Pot	[1]
2.	Nitrosulf – liquid formulation (33% S and 12% N)	0.2% (three times spraying)	Rapeseed (cv. B-9)	17.6 and 9.2% (two years pooled) increase in seed yield over 30 and 60 kg elemental S ha^{-1}, and further pooled results show increase in plant height (19.94%), number of siliqua $plant^{-1}$ (34.75%), number of seeds $siliqua^{-1}$ (42.87%), and thousand seed weight (18.84%) over control	65.2 and 34.1% (two years pooled) higher uptake than that of 30 and 60 kg elemental S ha^{-1}	25.9 and 14.2% increase in oil yield, and 29.5 and 14.9% increase in protein yield over 30 and 60 kg elemental S ha^{-1}, respectively	Field	[22]
3.	Gromor bentonite S pastille	60 kg ha^{-1}	Hybrid rice (cv. Rajyalaxmi)	Highest significant grain yield (5.8 t ha^{-1}) over gypsum applied @ 60 kg ha^{-1}	Grain uptake was significantly higher over control, but not in the case of straw uptake	-	Field	[93]

Table 3. *Cont.*

4.	Bentonite S – 90% (Pellets)	-	Aromatic rice [var. Pusa Improved Basmati (Pusa 1460)]	Effective tillers, grains panicle^{-1}, 1000 grain weight were significantly higher than gypsum, SSP, and elemental S	Grain, straw, and total uptake were higher than other sources, but at par with SSP	Higher crude protein content	Field	[4]
5.	Granular elemental sulphur (ES)-fortified ammonium phosphate	200 mg kg^{-1}	Canola	Highest yield (in the second crop) over ES pastilles	Highest uptake (in the second crop) over ES pastilles	-	Pot	[20]
6.	Bentonite S	30 kg ha^{-1}	Indian mustard [cv. Binoy (B9)]	-	Higher uptake and better use efficiency than SSP	-	Field	[92]
7.	Micronized-sulfur with bentonite (MSB) (86%) + Fe (2%) + Zn (2%)	500 kg ha^{-1} (applied five months before transplanting)	Tomato	Total fruit yield increased by 27% over control	-	-	Greenhouse (360 m^2)	[94]

be rapidly oxidized, and its rate of SO_4^{2-} release could be compared with the sulphate fertilizers on a short-term basis, but the oxidation of S° forms showed a different pattern (finely divided S suspension > coarse S powder > S-impregnated urea > S-impregnated urea). Among the S° forms, S-coated urea was concluded not to be a viable S source as it failed to produce required amounts of SO_4^{2-} over the five harvests. Boswell et al. [89] observed that increasing the proportion of sodium bentonite (expanding clay) in the prills from 5-40% increased the availability of S for uptake by the pasture plants, and at least 10% content of bentonite was essential to maintain a satisfactory rate of supply. Elemental S is a good source to overcome the deficiency of S in soils of humid tropics and irrigated subtropics as well as enhance the available status of P in arable soils [90]. It is also effective in alkaline soils because it reduces the soil pH, and thereby creates a suitable condition for the uptake of nutrients (phosphorus, iron, manganese, zinc, etc.) by the plants. This is generally noted after biochemical oxidation of S° to H_2SO_4 [91].

The potentials of improved fertilizers in crops have been evaluated by several researchers (Table 3). Riley et al. [1] found micronised S° to be as effective as SO_4^{2-} fertilizer as both of them resulted in similar increases in grain yield (on average 36%) and S uptake (on average 164%) of wheat over the control. Shivay et al. [4] evaluated the performance of bentonite S in *Basmati* rice and concluded it to be as good as SSP as a source of S to rice. Higher uptake and better use efficiency of S by the application of bentonite S in mustard as compared to SSP is also reported [92].

6. CONCLUSION

Sulphur is an essential nutrient required for crop growth and development, but with intensified agricultural practices and cleaner air programs, the deficiency of S corresponding to the ascending demand is increasing at a higher pace which needs to be fulfilled with the use of S fertilizers, advanced S formulations like bentonite S, and liquid S formulations. These new formulations must be taken into consideration for sustainable agricultural development and alongside greater emphasis has to be laid on the site-specific requirements as well as the demand of crops to S nutrition. Thus, we can fulfil our target of crop production and enhance the quality of food as well.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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