

Agronomic Effectiveness of Granular Nitrogen/Phosphorus Fertilizers Containing Elemental Sulfur with and without Ammonium Sulfate: A Review

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ABSTRACT

Deficiency of S in soils has become a soil fertility issue worldwide because of a decrease in S deposition from air to soil due to legislation and increased crop removal. Continuous use of high-analysis nitrogen/phosphorus (NP) fertilizers lacking in S further exacerbates the S deficiency for crop production. Several newly developed granular NP fertilizers such as monoammonium phosphate (MAP), diammonium phosphate (DAP), and triple superphosphate (TSP) containing micronized elemental sulfur (ES) with/without ammonium sulfate (AS) have been marketed to farmers. It is claimed that these products can provide available $\text{SO}_4\text{-S}$ through AS and ES oxidation during the growing season. The objective of this review was to carefully examine the literature that addresses the agronomic effectiveness of the granular NP-ES or NP-(ES+AS) fertilizer products. The review shows that oxidation of ES particles in granular NP fertilizers is generally nil or inadequate to provide available S to seasonal (or first) crops in greenhouse studies. This is due to the negative locality effect on granular ES oxidation. In contrast, available S can be obtained from the associated AS component of the granular (ES+AS). Under field conditions, limited studies showed these granular (ES+AS) were as effective as $\text{SO}_4\text{-based}$ sources at a high single S rate, but lack of data at multiple S rates. The detailed evaluation of available data so far often shows that the granular NP fertilizers containing ES or (ES+AS) cannot provide available S as compared with traditional $\text{SO}_4\text{-based}$ S sources for season-long or first field crops.

Core Ideas

- Sulfur nutrient is important to crop growth to produce maximum crop yield.
- Several new S fertilizers are marketed to farmers often without scientific data to support the fertilizer producers' claims.
- This review article examines the available data to check the claims; the results often cannot validate the claims.

THE WIDESPREAD USE of high-analysis, granular S-free NP fertilizers such as urea, MAP, DAP, and TSP, together with more intensified cropping with higher yields, has resulted in increasing occurrences of soil S-deficiency in many countries (Chien et al., 2009). Furthermore, a reduction in SO_2 emissions to the atmosphere from industry, as required by environmental laws, has a significant local effect on S deposition from the atmosphere to soils. For example, Sawyer et al. (2011a, 2011b) reported significant maize (*Zea mays* L.) and alfalfa (*Medicago sativa* L.) yield increases in response to S applications at field sites in northeastern Iowa. Grant (2013) reported significant canola (*Brassica napus* L.) response to S fertilizers in Canada.

Because pure ES is 100% S, incorporation of ES into high-analysis granular NP fertilizers will not significantly reduce N and P contents of the NP fertilizers and minimize increase in transportation costs as compared with the inclusion of AS. However, AS is considered readily plant available, whereas ES is not, unless it is oxidized to $\text{SO}_4\text{-S}$ by soil microbes. The oxidation rate of soil incorporated ES particles increases with decreasing particle size of ES (Boswell and Friesen, 1993). For this reason, some fertilizer companies have been developing processes in which the very fine micronized ES particles (<150 μm) are incorporated into granular NP fertilizers. Their idea assumes that once the fertilizer granules dissolve and ES particles are released back to the original micronized size, these very fine ES particles will result in rapid oxidation of the ES in soils. For granular NP-ES products, it has been claimed that ES oxidation provides adequate available S during the cropping season. In an effort to make up for potentially slow ES oxidation of granular NP fertilizers and to provide initial available $\text{SO}_4\text{-S}$, products containing a mixture of (ES + AS) with various S ratios have also been developed. It has been claimed that these products provide initial available S from AS while ES provides available S at later stages up to maturity in the growing season.

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Abbreviations: AS, ammonium sulfate; DAP, diammonium phosphate; ES, elemental sulfur; MAP, monoammonium phosphate; NP, nitrogen/phosphorus; SEF, sulfur-enhanced fertilizer; SSP, single superphosphate; TSP, triple superphosphate.

While several reviews of powdered ES fertilizers and their use on crops and pastures have been reported (Hagstrom, 1986; Johnson, 1975; Palmer et al., 1983), including a comprehensive one by Boswell and Friesen (1993), there has not been a published review of the agronomic effectiveness of granular NP fertilizers containing ES with/without AS in laboratory, greenhouse, and field evaluations. The objective of this review is to summarize and discuss published studies of potential positive and negative effects for granular NP-ES or NP- (ES+AS) products as S sources for crop production. To our knowledge, most of the reported field trials were presented by researchers as non-peer reviewed proceedings papers at regional or international conferences. Also, in most field trials only a single S rate, instead of multiple S rates, was used to compare the agronomic effectiveness of these granular NP fertilizers containing ES or (ES + AS) with standard S sources such as AS and gypsum.

Although most greenhouse studies do not include leaching of $\text{SO}_4\text{-S}$ during plant growth, these studies can evaluate the degree of granular ES oxidation during the plant growth since ES is not plant available. However, decrease in soil pH induced by ES oxidation may slow down and complicate the actual ES oxidation (Barrow, 1971). Under field conditions, available S to plants from the granular ES with/without AS will have a confounding interaction with ES oxidation and leaching of $\text{SO}_4\text{-S}$ from applied AS or SO_4 produced after ES oxidation in soils. This review will present and discuss the results based on the studies of (i) laboratory soil incubation, (ii) greenhouse experiments, and (iii) agronomic field trials.

SOURCES OF GRANULAR NITROGEN/ PHOSPHORUS FERTILIZERS CONTAINING SULFUR NUTRIENT

Currently there are several commercial-grade granular NP fertilizers available containing micronized ES particles with/without AS. The ratio of ES-S/AS-S among different products varies widely. Some examples of the compositions of the granular products are labeled by the producers and are listed in Table 1. In

Table 1. Some commercial-grade granular monoammonium phosphate (MAP) and triple superphosphate (TSP) fertilizers containing elemental sulfur (ES) with or without ammonium sulfate (AS).

Fertilizer	Chemical composition	Total S	%	
			ES-S	AS-S
MAP-10S	MAP + ES + AS	10	5	5
MAP-15S	MAP + ES + AS	15	7.5	7.5
TSP-12S	TSP + ES	12	12	0
TSP-12S	TSP + ES + AS	12	8	4

general, the products of MAP or DAP containing ES with AS are produced by ammonization of mixed $\text{H}_3\text{PO}_4 + \text{H}_2\text{SO}_4$ and molten ES before the granulation step (IFDC/UNIDO, 1998).

LABORATORY SOIL INCUBATION STUDIES

Chien et al. (2009, 2011) showed that no oxidation of bentonite-granulated ES (90% ES) to $\text{SO}_4\text{-S}$ occurred in an acid sandy soil during incubation up to 10 wk whereas powdered ES particles oxidized rapidly in the soil (Fig. 1). The lack of adequate S oxidation of ES granules can be explained by the concept of the negative “locality effect” on ES oxidation as discussed by Chien et al. (2009, 2011). This concept implies that when the fertilizer granule disintegrates and releases micronized ES particles to the soil, the very fine ES particles still cluster and localize around the applied granule site. This clustering results in decreased contact between localized ES particles and soil, which in turn decreases the colonization of soil S-oxidizing bacteria on the surface of the ES particles (Janzen and Bétrany, 1987). Also ES is hydrophobic and the released micronized ES particles can coalesce to form larger aggregates that further decrease ES oxidation (Friesen, 1996). Another factor that may play a role was localized pH effects developed on oxidation of ES in the ES particles (Barrow, 1971). The lack of increase in powdered ES oxidation from 6 to 10 wk in the sandy soil (Fig. 1) was probably due to build-up of acidity effect on ES oxidation as described by Barrow (1971).

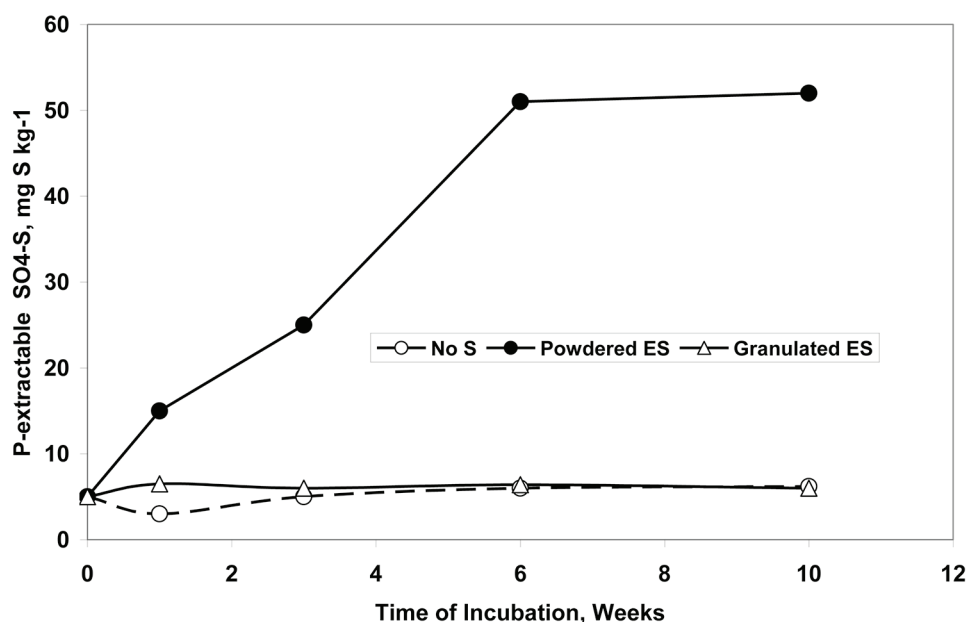


Fig. 1. Phosphate-extractable $\text{SO}_4\text{-S}$ from powdered elemental sulfur (ES) and granulated ES with bentonite in an acid sandy soil during incubation (Chien et al., 2009).

In a recently published paper by McLaughlin et al. (2015) using a soil leaching column technique to estimate the degree of ES oxidation of the granular MAP-(ES+AS) as compared with that of powdered ES, about 70% of powdered ES was oxidized in 6 wk when incorporated whereas only about 20% of incorporated granular MAP-(ES+AS) containing micronized ES particles was oxidized. The study also showed that about 90% of powdered ES was oxidized in 20 wk whereas it took 56 wk to oxidize 90% of the micronized ES particles in the granular MAP (ES+AS). In other words, it was significantly much less in ES oxidation rate of micronized ES particles as compared with powdered ES particles when the micronized ES particles were in the granular form due to the negative locality effect on ES oxidation, unless the micronized ES particles released from the granules on disintegration were dispersed by mixing in the soil, for example, plowing the soil after first crop.

GREENHOUSE EXPERIMENTS

In a study by Friesen (1996) with an S-deficient sandy Lakeland soil (thermic, coated Typic Quartzipsamment), powdered ES (<150 μm) was co-granulated with TSP, DAP, urea, or bentonite. The soil nutrients other than S were balanced at adequate levels so that S was the only limiting nutrient on plant growth. All granules had the same size (1.68–3.36 mm diam.) and contained fine ES particles homogeneously mixed throughout each granule. The results showed that even though all of the disintegrated granules were mixed with the soil, they were less effective than granulated gypsum with the same granule size when dry matter yield of maize was measured (Fig. 2). Furthermore, based on the statistical significance of the linear slope of each ES source, P carriers (TSP, DAP) were significantly better for enhancing maize yield than a non-P carrier (urea), which was better than an inert carrier (bentonite), presumably attributable to the effect of P and N nutrients on the activity of S oxidizing microorganisms (Boswell and Friesen, 1993). Although ES–TSP and ES–DAP eventually produced

Table 2. Total dry matter yield and S uptake of maize (Blair, 2010a).

Treatment†	Dry matter yield of maize‡ g pot ⁻¹	S uptake‡ mg S pot ⁻¹
TSP (no S)	4.12d	3.2e
Gypsum	4.79bc	11.6a
TSP, 10% ES	4.58c	3.9d
ES, 106 μm	5.07b	5.9c
ES, 53 μm	5.43a	7.2b

† TSP = triple superphosphate; ES = elemental sulfur.

‡ Values with the same small letter within each column within each row are not significantly different ($p < 0.05$).

the same maximum dry matter yield of maize as gypsum, a rate of 135 mg S kg^{-1} of ES–TSP and ES–DAP was required to be equivalent to gypsum applied at a rate of 15 mg S kg^{-1} (Fig. 2).

When maize was grown in an S-deficient soil treated with different S sources including granular TSP–10% ES, gypsum, and two different particle sizes of powdered ES, there was a significant S response (Table 2) (Blair, 2010a). An increase in total dry matter yield of maize with all S sources indicated ES oxidation of granular TSP–ES did occur, as shown by Blair (2010b) in a soil incubation study. However, he did not report % ES oxidation of total ES applied over an 87-d period. It is interesting to note that ES (53 μm) produced a significantly higher dry matter yield of maize (5.43 g pot⁻¹) than gypsum (4.79 g pot⁻¹), but significantly much lower S uptake for the ES (7.2 mg S pot⁻¹) than for gypsum (11.6 mg S pot⁻¹) (Table 2). Although TSP (10% ES) and gypsum produced about the same total dry matter yield, S uptake from gypsum was much greater than that from TSP–10% ES. Since only a single S rate (equivalent to 15 kg S ha^{-1}) was used, it is not clear whether a lower S rate might have enabled gypsum to attain a higher dry matter yield of maize than granular TSP–ES. Thus it is uncertain whether TSP–ES was actually as effective as gypsum. Furthermore, the two powdered ES sources (106 and 53 μm sizes) incorporated in the soil were more effective than

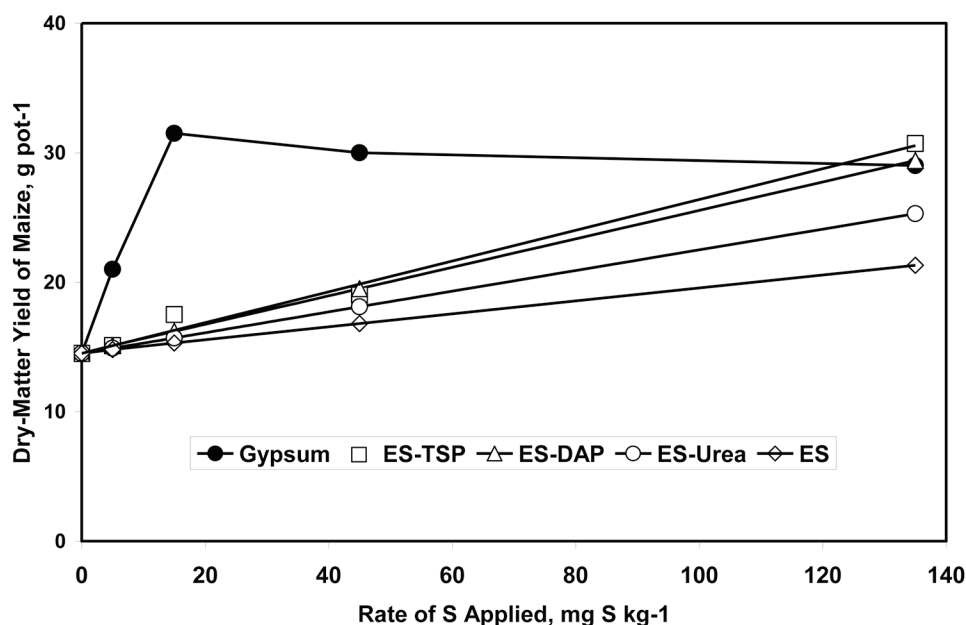


Fig. 2. Dry matter yield of maize obtained with different NP granular fertilizers (TSP-triple superphosphate, DAP-diammonium phosphate and urea) containing elemental sulfur (ES) compared with gypsum, and bentonite mixed with ES (Friesen, 1996).

Table 3. Dry matter yield (DMY) and S uptake by two successive canola crops (6 wk each) obtained with ammonium sulfate (AS) and two granular monoammonium phosphate (MAP)–(AS+ elemental sulfur [ES]) products (Degryse et al., 2016).

Treatment	First crop†			Second crop†			
	AS	ES	Total S	DMY	S Uptake	DMY	S Uptake
	mg S kg ⁻¹			g pot ⁻¹	mg pot ⁻¹	g pot ⁻¹	mg pot ⁻¹
MAP‡	3.2	0	3.2	1.9b	2.1e	1.5cd	1.5c
MAP+AS	6.7	0	6.7	2.3ab	3.1d	1.7bc	1.7bc
MAP+AS	10	0	10	2.6a	4.9c	1.5cd	1.7bc
MAP+AS	20	0	20	2.5a	13.7a	1.9ab	2.3b
MAP-(4%AS-S+8%ES)	6.7	13.3	20	2.3ab	4.4c	2.0a	3.5a
MAP-(5%AS-S+5%ES)	10	10	20	2.5a	5.7b	2.0a	3.5a

† Means within a column not followed by the same letter are significant difference ($p \leq 0.05$, Duncan's multiple range test).

‡ The MAP contained impurity of AS.

powdered ES inside the incorporated TSP granules in dry matter yield and S uptake suggesting (i) possible locality effect of ES particles on reducing the ES oxidation of granular TSP–ES and (ii) less accessible of the $\text{SO}_4\text{-S}$ to plant roots derived from the ES oxidation of granular TSP–ES.

Cumulative dry matter yield of ryegrass (*Lolium multiflorum* L.) was significantly greater with granular AS than with granular MAP (5% ES–S+5% AS–S) at 10 mg S kg⁻¹ shown in Fig. 3 (Chien et al., 2013). When applied at rates of 25 mg S kg⁻¹ or higher, both products had an equal effect on maximum dry matter yield (Fig. 3). Therefore, a comparison of the agronomic effectiveness of NP fertilizers containing (ES + AS) with AS should not be based on the use of a single relatively high S rate. This use of a single higher S rate may lead to the conclusion that the S sources were equal.

Another example showing the importance of using multiple S rates to compare various S sources is demonstrated in a recently published study by Degryse et al. (2016) for two successive canola crops (6 wk each) on comparing AS with two MAP–(ES+AS) products as shown in Table 3. Both MAP–(AS+ES) products were statistically as effective as AS

in dry matter yield based on a single rate at 20 mg total S kg⁻¹ for the first canola crop. However, the maximum dry matter yield was attained at ≥ 6.7 mg S kg⁻¹ of AS–S alone. Both MAP–(AS+ES) products provided ≥ 6.7 mg S kg⁻¹ of AS–S at a total S rate of 20 mg S kg⁻¹. Thus the MAP–(AS+ES) products would be as effective as AS alone at 20 mg total S kg⁻¹. In contrast, S uptake by the first crop from AS was much higher than that from the MAP–(ES+AS) products (Table 3).

For the second crop, AS and MAP–(ES+AS) products were no significant differences in dry matter yield, but S uptake from AS was lower than that from MAP–(ES+AS) products at 20 mg S kg⁻¹ initially applied (Table 3). The authors explained that the low S availability from AS to the second crop was due to a high depletion of $\text{SO}_4\text{-S}$ in the first crop. The higher S uptake from MAP–(ES+AS) products was due to further oxidation of ES during the second crop, resulting in a continuing supply of $\text{SO}_4\text{-S}$. It should be pointed out, however, after the first harvest, the soil from each pot was removed, mixed, and placed back to the pot followed by open incubation for 38 d before the second crop was planted. By mixing the soil would disperse the ES particles in the soil that would significantly increase ES oxidation

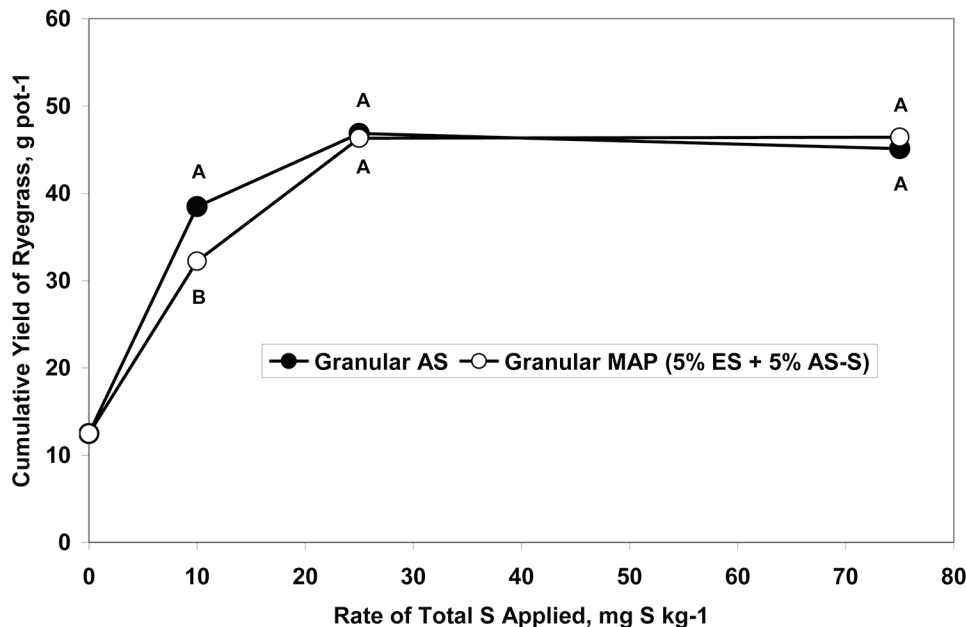


Fig. 3. Cumulative ryegrass yield as influenced by rates of total S applied from granular ammonium sulfate (AS) and granular monoammonium phosphate (MAP) containing 5% elemental sulfur (ES) +5% AS–S. Values with the same letter at each total S rate are not significantly different based on pair-wise contrast test ($p < 0.05$) (Chien et al., 2013).

Table 4. Means of S uptake by two successive (first + second) canola (45 d each) after emergence in two soils treated with 41 mg S pot⁻¹ with monoammonium phosphate (MAP) with elemental sulfur (ES), ammonium sulfate (AS), or (ES + AS) (Kroeker, 2005).

S Source	S Uptake by canola†			Total S recovery %
	First	Second	(First + second)	
	mg S pot ⁻¹			
MAP (no S)	4.7a	2.7a	7.4a	–
MAP+ES	4.7a	3.3a	8.0a	1
MAP+AS	20.0c	4.8b	24.8c	42
MAP (7.5%ES+ 7.5% AS-S)	10.0b	5.5c	15.5b	20

† Values with the same letter within each column are not significantly different according to Tukey's test ($p < 0.05$).

and resulted in enhancing S uptake by the second crop. This procedure, unlike Chien et al. (2014) and Prochnow et al. (2007) who did not disturb the soil pots between first and second maize crop, would not mimic field conditions under which the granular MAP-(ES+AS) products would not be disturbed and dispersed during the first canola growth, especially in no-till system.

In a study similar to the procedure used by Degryse et al. (2016), Kroeker (2005) conducted a growth chamber study to compare a homogenous granular NPS (15%S) fertilizer containing (7.5% ES + 7.5% AS-S) and MAP+ES (particle size of ES < 150 μ m) with MAP+AS for two successive canola crops (45 d each) in two soils that were treated with 41 mg S pot⁻¹ (Table 4). The apparent sum of S recovery by the two canola crops from the NPS fertilizer was approximately 50% of that from MAP+AS, whereas MAP+ES was ineffective in increasing S uptake as compared with MAP (no S) (Table 4). The SO₄-S (i.e., AS-S) portion of NPS fertilizer appeared to be available to both crops, with no measurable amount of ES oxidation of the ES portion as concluded by Kroeker (2005). The discrepancy in the results between Degryse et al. (2016) and Kroeker (2005) for the ES oxidation to provide available S to the second canola crop could be due to (i) characteristics of the same fertilizers but produced at different times, (ii) types

of soil, (iii) biological activity of the soils, and (iv) soil temperature, etc. Kroeker (2005) stated that in an early field trial, after application of the same granular NPS product for canola grown to maturity, there was no significant ES oxidation to provide available S to the canola crop in the first year. Grant (2013) also concluded that conversion of ES to SO₄-S in the NP fertilizers containing ES with/without AS is not rapid enough to supply available S in the year of application.

Both rape straw and seed yields at maturity were higher for granular AS than granular MAP (5% ES-S+5% AS-S) (Table 5) indicating inadequate ES oxidation of the MAP-(ES+AS) granules (Chien et al., 2013). An application rate of 25 mg S kg⁻¹ as AS-S was required to attain the maximum straw and seed yields of rape at maturity. Since MAP-(ES+AS) provided only 5 and 12.5 mg AS-S kg⁻¹ of the total 10 and 25 mg S kg⁻¹, respectively, MAP-(ES+AS) was inferior to AS. At the 75 mg S kg⁻¹ rate, MAP-(ES+AS) provided 37.5 mg AS-S kg⁻¹, which was more than 25 mg AS-S kg⁻¹ required to attain the maximum straw and seed yield. Thus MAP-(ES+AS) was effective as AS at this high S rate for rape straw and seed yields.

To quantify the degree of ES oxidation from the granular NP fertilizers containing (AS + ES) fertilizers to provide available SO₄-S in greenhouse experiments, a simple test can be used

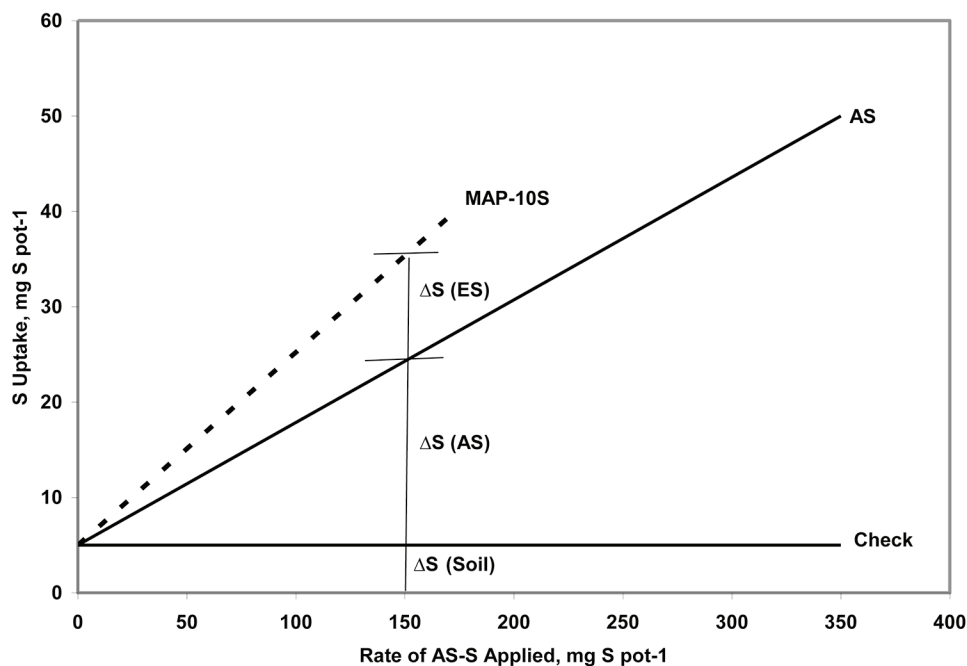


Fig. 4. Hypothetical S uptake from ammonium sulfate (AS) and granular monoammonium phosphate (MAP)-10S containing (5% AS-S + 5% elemental sulfur [ES]-S) as influenced by the AS-S rate applied. ΔS (ES), ΔS (AS) and ΔS (soil) represent S uptake by crop from ES, AS, and soil, respectively.

Table 5. Straw and seed yields of rape harvested at maturity with granular ammonium sulfate (AS) and granular monoammonium phosphate (MAP) containing 5% elemental sulfur (ES) + 5% AS-S (MAP-10S) in a sandy soil (Chien et al., 2013).

S Rate†	Rape straw yield‡		Rape seed yield‡	
	AS	MAP-10% S	AS	MAP-10S
mg S kg ⁻¹	g pot ⁻¹			
10	55.6aA	37.0aB	0aA	0aA
25	92.2bA	77.4bB	16.1bA	0.2aB
75	95.2bA	96.8cA	17.7bA	21.0bA

† Check (no S) for rape straw yield = 3.4 g pot⁻¹ and rapeseed yield = 0 g pot⁻¹.

‡ Values with the same lowercase within each column and uppercase within each row for straw and seed are not significantly different ($p < 0.05$).

without using expensive labeled isotopic technique. Figure 4 shows two hypothetical linear relationships (applicable to non-linear as well) between S uptake from AS alone and granular MAP-10S (5% AS-S + 5% ES-S) and AS-S (not total S) rate applied to a sandy soil. In this case, AS-S rate based on total S applied from MAP-10S is 50% of that from AS alone since MAP-10S contains 50% each of total S as AS-S and ES-S. If ES oxidation of the granular MAP-10S provides available SO₄⁻S, that is, ΔS (ES) which represents S uptake by crop from ES in the soil, the S uptake line of MAP-10S should be above the line of that from AS alone, that is, ΔS (AS) which represents S uptake from AS (Fig. 4). The ratio of ΔS (ES)/ΔS (AS) can be used to express percent ES oxidation that provides available S from the granular MAP-10S in the soil. For example, if the ratios are 0, 0.4, and 1.0, it suggests that 0, 40, and 100% for plant available S from ES oxidation for the granular MAP-10S, respectively. In case ΔS (ES) = 0% which suggests that no contribution of available S from ES oxidation in the granular MAP-10S, S uptake from both MAP-10S and AS alone should follow the same line as a function of AS-S rate applied.

Table 6. Means of dry matter yield and S uptake of two successive maize crops (6 wk each) grown on a S-deficient soil in Brazil. The soil was not mixed between the two crops (Chien et al., 2014).

S Source†	First maize‡		Second maize‡	
	Dry matter yield	S Uptake	Dry matter yield	S Uptake
	g pot ⁻¹	mg S pot ⁻¹	g pot ⁻¹	mg S pot ⁻¹
No S	7.9c	4.8c	7.3c	5.3c
AS	13.3a	8.6a	25.5a	10.6a
MAP-15S	11.8b	7.5b	19.6b	6.7b

† AS = ammonium sulfate, MAP-15S = monoammonium phosphate containing 15%S (7.5% AS-S + 7.5% elemental sulfur [ES]-S).

‡ Mean values (across 10, 25, 50, and 75 mg S kg⁻¹) of AS and MAP-15S in each column followed by the same letter are not significantly different according to Tukey's test ($p < 0.05$).

A greenhouse study was conducted in Brazil with two successive maize crops grown in 5-kg soil pots to compare the S availability of granular AS with granular MAP (7.5% ES-S+7.5% AS-S) or MAP-15S (Chien et al., 2014) at S rates of 0, 50, 100, 250 and to 375 mg S pot⁻¹. After harvest of the first maize crop grown for 6 wk, the second maize was seeded without disturbing the soil pots to measure the residual S effect for another 6 wk. Since source × rate interaction was not significant ($p < 0.05$) as revealed by ANOVA, the means of dry matter yield and S uptake for the first and second maize crop were combined and calculated across all S rates (Table 6). Both dry matter yield and S uptake of the first and second maize crops were significantly ($p < 0.05$) higher with granular AS than granular MAP-15S. When S uptake by both the first and second maize was plotted against AS-S rate applied from AS and MAP-15S, both granular AS and MAP-15S followed the same S uptake regression line (Fig. 5). This demonstrates that there was no significant ES oxidation of the granular MAP-S to contribute available S during the growth of maize. Otherwise, all the data points of granular MAP-15S would be

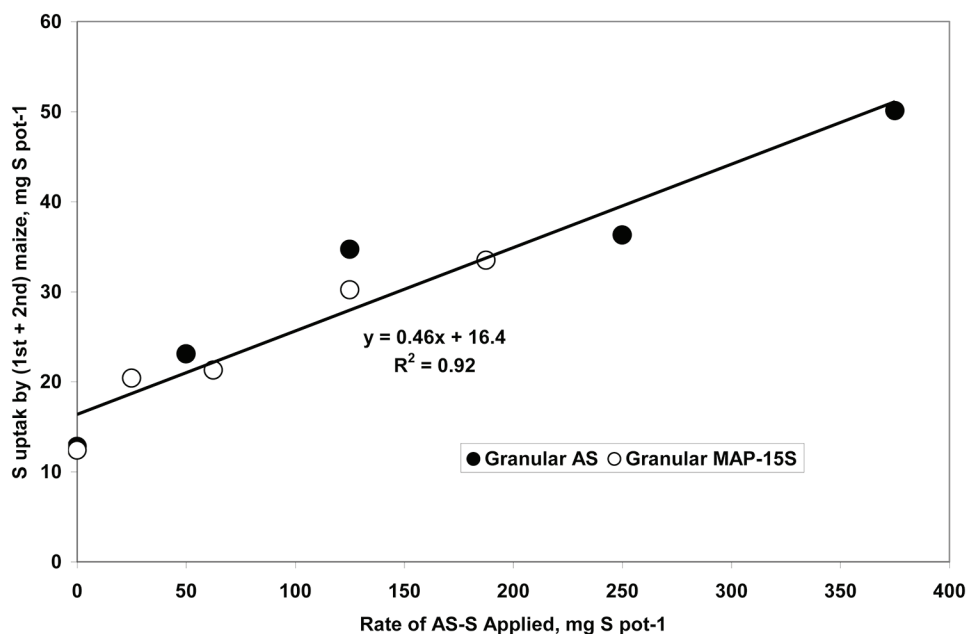


Fig. 5. Sulfur uptake by two successive (first + second) maize (6 wk each) grown in 5-kg soil pots in Brazil obtained with granular ammonium sulfate (AS) and granular monoammonium phosphate (MAP) containing 7.5% elemental sulfur (ES)+7.5% AS-S (MAP-15S) in relating to rate of AS-S applied (Chien et al., 2014).

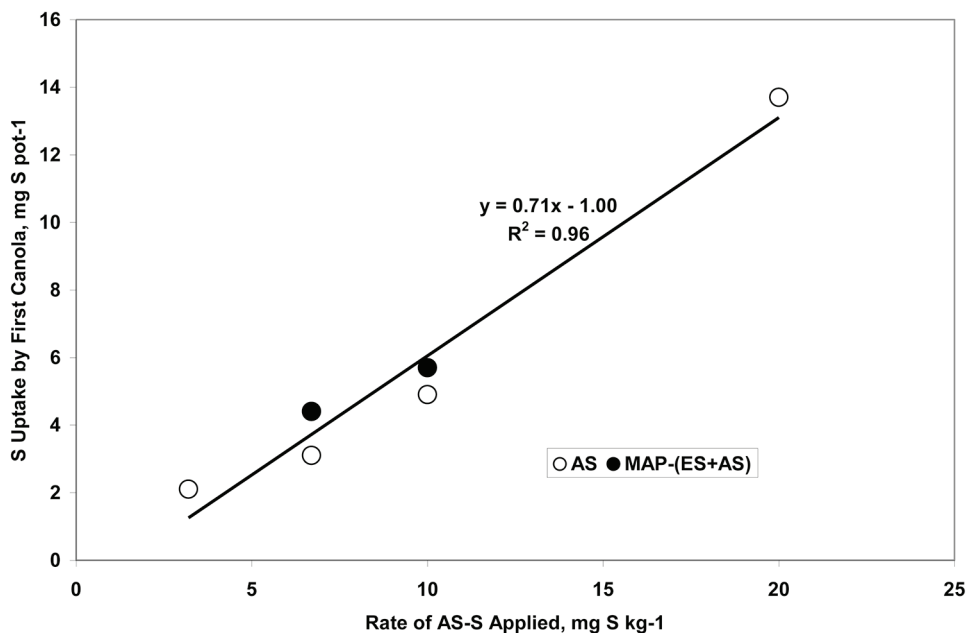


Fig. 6. Sulfur uptake by first canola crop (6 wk) obtained with granular ammonium sulfate (AS) and two granular monoammonium phosphate (MAP)–(elemental sulfur [ES]+AS) products containing (4% AS–S+8%ES) and (5%AS–S+5%ES), respectively as related to AS–S rate applied (adapted from Degryse et al., 2016).

significantly above the regression line due to additional $\text{SO}_4\text{-S}$ from the ES oxidation.

Similarly, a plot of S uptake vs. AS-S rate applied for the first canola crop (Table 3) shows that the two data points of the granular MAP–(AS+ES) products followed the regression line of AS alone (Fig. 6). This suggests a no significant ES oxidation from the granular MAP–(AS+ES) products to contribute available S to the first canola crop. Otherwise, the two data points of MAP–(ES+AS) products should be above the regression line of AS.

The aboveground biomass (straw + grain) of wheat (*Triticum aestivum* L.) grown in 4-kg soil pots for 120 d from seeding to maturity was higher for AS than granular MAP–15S (7.5%

AS–S + 7.5% ES–S) (Fig. 7) (Chien et al., 2014). Based on the segmented linear function (Black, 1993), MAP–15S produced maximum yield at total S rate of $135 \text{ mg S pot}^{-1}$. Since granular MAP–15S contains 50% of total S as AS–S, it provided 68 mg S pot^{-1} of AS–S which was more than 40 mg S pot^{-1} required for AS to produce the maximum yield (Fig. 7). Therefore, granular MAP–15S was less effective than granular AS at the lowest S rate. Sulfur uptake by wheat biomass was 30% of total S applied from AS while it was 15% from MAP–15S as shown by the coefficients (slopes) of the regression lines (Fig. 8a). This indicates that MAP–15S was 50% as effective as AS to provide available S to wheat. Since MAP–15S contained 50% AS–S

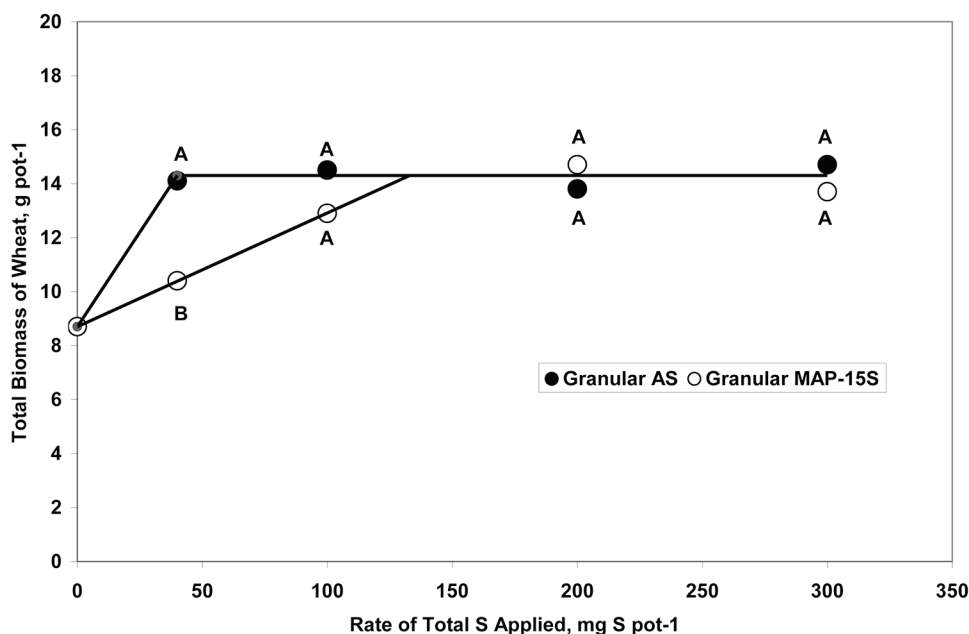


Fig. 7. Aboveground biomass (straw + grain) of wheat at maturity in 5-kg soil pots obtained with granular ammonium sulfate (AS) and granular monoammonium phosphate (MAP) containing 7.5% ES–S + 7.5% AS–S (MAP–15S) in Brazil. Values with the same letter at each total S rate is not significantly different based on pair-wise contrast test ($p < 0.05$) (Chien et al., 2014).

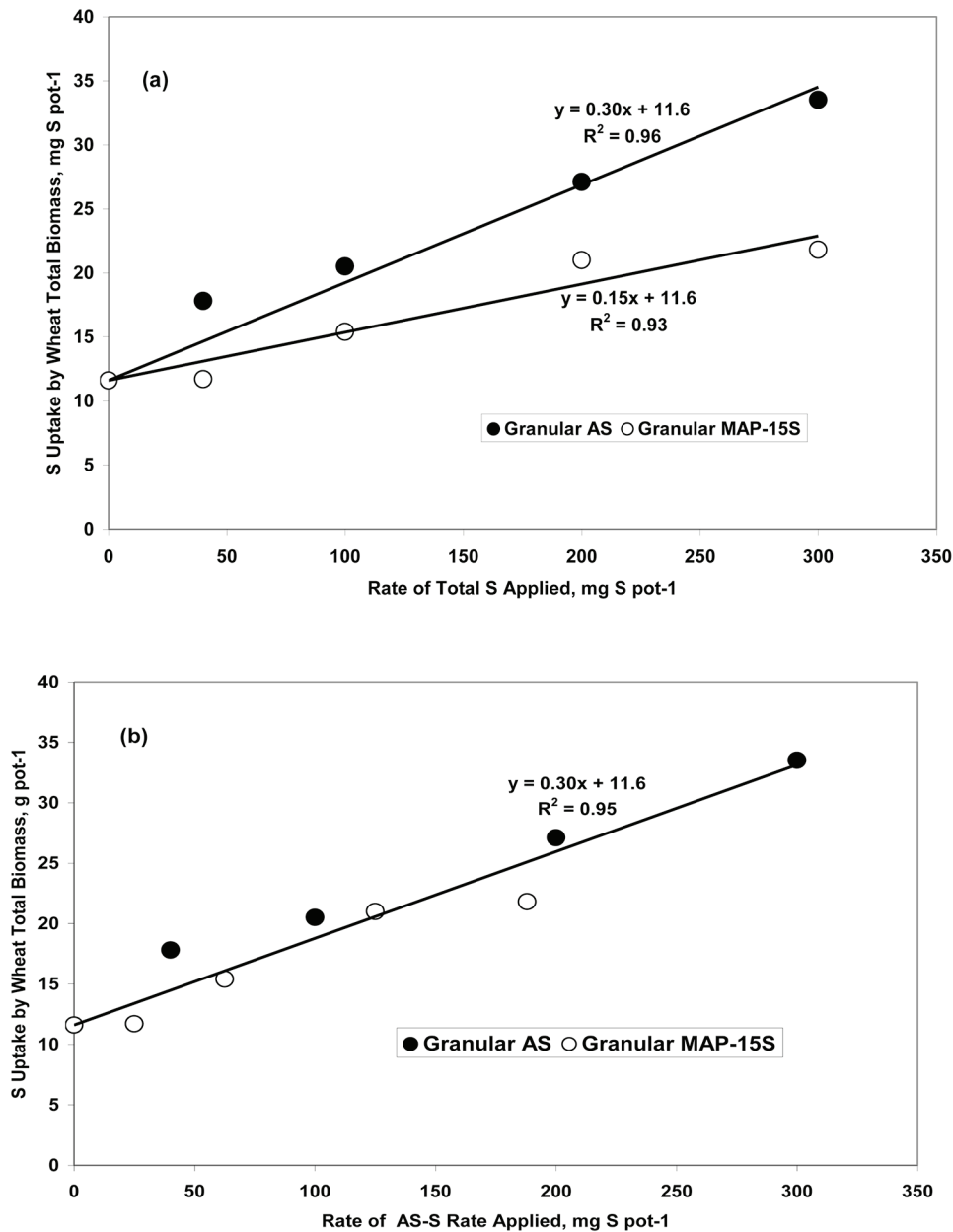


Fig. 8. Sulfur uptake by wheat total biomass (straw + grain) in 5-kg soil pots from granular ammonium sulfate (AS) and granular monoammonium phosphate (MAP)-15S based on (a) total S and (b) AS-S rate (Chien et al., 2015).

and 50% ES-S of total S, it suggests that there was little contribution of available S due to ES oxidation of granular MAP-15S. Indeed, data points of MAP-15S followed the regression line of AS when S uptake was plotted against AS-S rate applied for both S sources (Fig. 8b), supporting the explanation.

AGRONOMIC FIELD TRIALS

The agronomic effectiveness of granular NP-(ES+AS) products under field conditions depends on (i) rate of ES oxidation (not too fast and not too slow), (ii) leaching of AS-S on rainfall, and (iii) other soil and environmental conditions. To our knowledge, unlike greenhouse experiments, there are no results from the agronomic field trials on the granular NP-ES or NP-(AS+ES) products ever published in peer-reviewed scientific journals. The available articles in literature on this issue have been published in the regional or international conferences' proceedings. Therefore, examination and discussion of the data

from the agronomic field trials of granular NP-ES or NP-(AS+AS) products will be limited in details.

Blair (2010a) reported a field trial with a cropping system of soybean (*Glycine max* L.)/wheat/soybean/wheat rotation designed to measure initial and residual S effects (Table 7). The S rate was not mentioned in the report. There was a significant S response in wheat grain yield as shown by comparing gypsum with no S (MAP). There was no significant ES oxidation of granular TSP (10% ES); and thus no S response of TSP (10% ES) was observed compared to the control. Therefore, granular TSP (10% ES) was not effective for the wheat crop. In the following soybean crop, ES oxidation of the residual TSP (10% ES) was sufficient to provide available S to increase soybean grain yield at the same level as the residual gypsum. The relative yield of gypsum for the soybean crop (114%) compared with the control was lower than the relative yield from the previous wheat crop (138%) suggesting a decrease in the S availability

Table 7. Response of wheat and soybean to monoammonium phosphate (MAP), gypsum, and triple superphosphate (TSP) (10% elemental sulfur, ES) in a field trial with an Oxisol in Brazil (Blair, 2010a).

Initial fertilizer applied	Relative wheat grain yield† %	Residual fertilizer (no S Applied)	Relative soybean grain yield† %
MAP (no S)	100a	MAP (no S)	100a
Gypsum	138b	MAP (no S)	114b
TSP (10% ES)	103a	MAP (no S)	115b

† Values with the same letter within each column are not significantly different ($p < 0.05$).

of the residual S effect of gypsum, probably due to leaching and depletion of gypsum-SO₄ in the soil by the previous wheat crop.

In a complicated field trial with a soybean-wheat rotation in a Brazilian Oxisol with a no-till system, Santos et al. (2010) found no significant S response in terms of grain yield for soybean fertilized with various S sources except gypsum at a rate of 20 kg S ha⁻¹ in terms of S uptake (Table 8). For the second wheat crop, gypsum, MAP-S (8%ES + 4% AS-S) and TSP (9% ES) were freshly applied to the three control plots which had been treated with MAP (no S) in the previous soybean crop. Gypsum produced significantly higher wheat grain yield and S uptake when compared to the TSP (9% ES), which was not significantly different from the control (Table 8). Although gypsum and MAP-(8% ES+4% AS-S) produced statistically no different wheat grain yield, S uptake by wheat from gypsum was significantly higher than MAP-(8% ES+4%AS-S). These results suggest that little or inadequate ES oxidation took place for TSP (9% ES) or MAP-(8% ES + 4% AS-S) during the wheat cropping season.

Use of granular ES for surface application may perform as well as or better than 100% SO₄-S when a combination of conditions occurs such as (i) shallow rooting system of pastures, (ii) break-up of ES granules and dispersion of ES particles by heavy rainfall or animal trampling on the soil surface, and (iii) leaching of AS-S by heavy rainfall (Chien et al., 2009). Under these circumstances, slow ES oxidation of granular NP fertilizers containing ES with/without AS can indeed be as good as or better than single superphosphate (SSP) which contains 12% S as gypsum or AS.

Flavel et al. (2010) showed that granular MAP-12%S (8% ES+4% AS-S) was more effective than SSP in cumulative yield of clover (*Trifolium repens* L.) after four harvests in a coarse-textured soil in Australia (Table 9), presumably due to heavy rainfall that caused substantial leaching of SO₄-S after initial

Table 9. Cumulative clover yield after four harvests, S uptake, and percent fertilizer S recovery from single superphosphate (SSP-S), monoammonium phosphate (MAP), and granular MAP-12% S) in a field trial in Australia (adapted from Flavel et al., 2010).

S Source	Clover yield† Mg ha ⁻¹	S Uptake† kg S ha ⁻¹	Apparent fertilizer S recovery %
MAP (no S)	1.45a	3.65a	–
SSP-S	2.25b	5.05b	16a
MAP (12% S)‡	2.85c	6.38c	32b

† Values with the same letter within each column are not significantly different (< 0.05).

‡ MAP-(8% elemental sulfur [ES] + 4% ammonium sulfate [AS]-S).

fertilizer application. Granular MAP (8% ES+4% AS-S) surpassed SSP in increasing clover yield only beginning at the third harvest and especially at the last harvest, apparently after significant oxidation of residual ES over time. At the first harvest, SSP still performed better than MAP (8% ES+4% AS-S) indicating initial ES oxidation was inadequate to provide available S. Furthermore, the AS-S component in the granular MAP (8% ES-S+4% AS-S) would be subject to leaching. Thus, the MAP (8% ES-S+4% AS-S) was inadequate in providing initial available SO₄-S.

In a recent study, Degryse et al. (2015) reported that 80% of the applied SO₄-S had leached below 91 cm of depth when fall applied compared to 40% spring applied under field conditions. They concluded the benefit of ES as a slow release S fertilizer in high rainfall environments. While this is true due to longer time frame from fall applied than spring, AS-S leaching from granular (ES + AS) products follows the same pattern as AS alone. Under severe leaching conditions after application, the granular (ES + AS) products may depend on the slow ES oxidation that may provide inadequate available S to the plants. Furthermore, if severe rainfall occurs after ES oxidation and before plant S uptake, the resulted SO₄-S can also be subject to leaching (C.A. Grant, personal communication, 2014). All these suggest that the agronomic effectiveness of granular (ES + AS) products greatly depends on the rate of ES oxidation and the dynamic rainfall conditions.

The results of 84 field trials conducted in China (72), Brazil (8), Argentina (3), and Australia (1) compared the agronomic effectiveness of SSP with granular NP fertilizers (mainly TSP) containing up to 15% ES as micronized ES particles (called sulfur-enhanced fertilizers or SEF) were summarized by Blair (2009). Of these yield trials, SEF > SSP at 28 sites, SEF = SSP at 50 sites, and SEF < SSP only at 6 sites. Similarly, SEF trials in China reported by Blair

Table 8. Effect of various S sources on soybean and wheat yield and S uptake in an Oxisol under field conditions in Brazil (Santos et al., 2010).

First crop fertilizer treatment	Crop 1 (Soybean)‡		Second crop fertilizer treatment	Crop 2 (Wheat)‡	
	Grain yield Mg ha ⁻¹	S Uptake kg ha ⁻¹		Grain yield Mg ha ⁻¹	S Uptake kg ha ⁻¹
Control (-PS)	3.06ab	8.2bc	Control(-PS)	2.20c	2.5b
MAP (no S)	3.05ab	7.9bc	MAP-S†	2.58abc	3.9b
MAP (no S)	2.96ab	7.0c	Gypsum	3.04a	5.5a
MAP (12% elemental sulfur, ES)	2.92b	6.3c	MAP (no S)	2.72abc	3.5b
MAP-S†	3.13ab	8.9bc	MAP (no S)	2.49abc	3.7b
Gypsum	3.37a	10.8ab	MAP (no S)	2.63ab	3.3b
MAP (no S)	3.01ab	7.9bc	TSP (9%ES)	2.27bc	3.4b

† MAP-S = (8% ES-S+4% AS-S)

‡ Values with the same within each column are not significantly different ($p < 0.05$).

(2010b) showed SEF > SSP at 25 sites, SEF = SSP at 59 sites, and SEF < SSP only at 5 sites. Performance of SEF ≤ SSP was in a very high rate of 95% in the reported trials, presumably due to efficient ES oxidation and leaching of SO₄-S in the soils. However, the reports provided only limited information regarding experimental protocols including variables such as S rate(s) used, exact % ES (or % AS) of the products, soil physical and chemical properties, climate, initial or residual S effect, fertilizer management (till, no-till), experimental design, etc.

The good performance of granular TSP-ES as compared with SSP for various crops (Blair, 2009, 2010a, 2010b) including flooded rice (*Oryza sativa* L.) may deserve further evaluation. In general, flooded rice soils under reduced conditions would be expected to decrease ES oxidation. In a greenhouse study of flooded rice (Chien et al., 1987), the relative agronomic effectiveness of powdered ES was 96% of gypsum in rice grain yield when surface broadcast and 87% when incorporated. The effectiveness of prilled-urea containing ES was significantly reduced to 58% when broadcast and 61% was incorporated as compared with gypsum. It may be argued that P of TSP-ES enhanced ES oxidation as compared with urea-ES (Boswell and Friesen, 1993). However, granular P products containing ES were much in promoting maize growth worse than gypsum in a greenhouse study under no leaching conditions suggesting poor ES oxidation (Fig. 2). Granular bentonite-ES containing 90% ES or 95% ES was not as effective S source as reported in an S review chapter by Franzen and Grant (2008). For example, average canola seed yields of three field experiments for the zero-S control, granular ES-90 (90% S), ES-95 (95% S), and granular SO₄-S on severely S deficient soils were 29, 28, 35, and 1135 kg ha⁻¹, respectively, indicating little ES oxidation of granular ES-90% and ES-95%. Seed yields of canola improved considerably when ES fertilizers were surface broadcast or broadcast and incorporated into the soils as finely ground powder form. Seed yields of canola with powder ES were comparable to SO₄-S fertilizer (Franzen and Grant, 2008).

In a field trial conducted with labeled S isotope in a tropical Brazilian Oxisol with annual soybean/maize rotation for 2 yr (Degryes et al., 2014), 4.2% of plant was derived from ES and 1.4% from SO₄-S in the first crop from applied granular MAP containing 7% ES + 2% AS-S. Over the four crops 16% of fertilizer ES and 9% of fertilizer SO₄-S was recovered in the crop. Based on a developed model, it was estimated 70% oxidation of applied ES at the end of the second growth season (2 yr). All these above-mentioned results differed considerably from those reported by Blair (2009, 2010 a, 2010b) and warrant further studies to resolve the issue.

Under certain conditions ES may deliver available S in the first year. Sousa et al. (2014) applied different S sources to an S-deficient Brazilian Oxisol under no-till in a soybean-maize-soybean rotation. Before the first cropping, millet (*Pennisetum glaucum* L.) as a cover crop was planted to deplete soil S. It was then cut and left on the soil as a mulch. They found that TSP containing ES or a mixture of ES (two-thirds of total S) and SO₄-S (one-third of total S) produced the first soybean grain yields similar to that of a mixture of TSP and gypsum (Table 10). However, that only happened when the fertilizers were broadcast over the soil after sowing, whereas the ES-containing TSP placed in the seed furrow was not as efficient gypsum. For gypsum, the method of fertilizer placement did not affect the S-response. The authors attributed the response to ES in the first year to the combination

Table 10. Grain yields of first soybean obtained with triple superphosphate (TSP), gypsum, elemental sulfur (ES) and ammonium sulfate (AS) were broadcast in a Cerrado Oxisol in Brazil under no-till management (Sousa et al., 2014).

S Source	S Rate	Soybean grain yield†
	kg S kg ⁻¹	Mg ha ⁻¹
TSP (No S)	0	3.19b
TSP + Gypsum	15	3.90a
TSP-(10% ES)	15	4.00a
TSP-(8% ES + 4% AS-S)	15	3.81a

† Values with the same letter within the column are not significantly different (< 0.05).

of spreading the fertilizer granules on the soil surface and the millet straw cover provided by the no-till system, which increased moisture retention and microbial activity, making the oxidation of ES easier for micronized ES in the granular TSP.

Comparing TSP-gypsum with TSP- (10% ES) and TSP- (8% ES + 4% AS-S) only at a single rate (15 kg S ha⁻¹) could not be interpreted that the granular ES or (ES+AS) products therefore were as effective as gypsum (Sousa et al., 2014). Without testing S sources at multiple S rates ranging from low to high, it is difficult to compare the effectiveness of various S sources. This is especially true for S response in which S rates as low as 10 kg S kg⁻¹ often already produced maximum crop yield with AS or gypsum (Fig. 2, Table 3). It is unknown if a lower S rate of TSP-gypsum than the 15 kg S ha⁻¹ rate that was used by Sousa et al. (2014) (Table 10) might have produced maximum grain yield of the first soybean crop while TSP-10% ES and TSP-(8% ES + 4% AS-S) might not. For example, had only a single total S rate at 25 mg S kg⁻¹ been used for ryegrass (Fig. 3), it would have been concluded that granular MAP-(5% ES + 5% AS-S) was as effective as granular AS. However, at a lower rate of 10 mg S kg⁻¹, MAP-(5% ES + 5% AS-S) was clearly less effective than AS (Fig. 3) Therefore, accurate assessment of effectiveness of various S fertilizer sources should be based on multiple rates of S nutrient.

SUMMARY AND CONCLUSION

Several granular NP fertilizers that contain micronized ES particles with/without AS have been marketed to farmers. For granular NP-ES products, it has been claimed that ES oxidation of the granular products can provide available S during the cropping season. For granular NP- (ES+AS) products, it has been claimed that AS provides initial available S and ES oxidation provides available S at later stages of crop growth to maturity during the annual or first crop growth cycle. In this review, it was concluded that the granular form of ES, in most cases, may not benefit crops planted after its application due to the negative locality effect on ES oxidation that supplies available S to the season-long or first field crops. Additionally, the disintegrated but localized ES particles around the applied granule sites after incorporation in soils may result in a decrease for the plant roots to access SO₄-S after ES oxidation as compared with incorporation of powdered ES particles in the soils. Consequently, for the first crop or in the year after fertilizer application, available S often comes only from the AS component of the products and the associated micronized ES particles contribute an inadequate amount of available S. Furthermore, these granular NP products containing ES with/without AS often do not perform well when compared with traditional SO₄-based sources like SSP, AS, and gypsum.

Although many reports on the factors affecting the oxidation of powdered ES in soils are available in literature, only limited information has been published on the oxidation of granular ES. Additionally, limited information is available on the $\text{SO}_4\text{-S}$ leaching from the associated AS component of the granular (ES+AS) products as compared with $\text{SO}_4\text{-based}$ fertilizers at the same total S rate of these two types of S fertilizer products during crop growth. Similarly, research should be focused on the leaching of $\text{SO}_4\text{-S}$ after ES oxidation but before plant uptake under severe rainfall conditions.

More future research should be conducted to identify the soil, crop, fertilizer application method, and climate conditions in which ES oxidation of granular ES products may have a potential long-term (>3 yr) residual S effect on crop production. It is also hoped that more agronomic trials of these S products as compared with traditional $\text{SO}_4\text{-based}$ products be conducted on multiple S rates. The results should be published in peer-reviewed scientific journals for public interest.

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