Transformations of Elemental Sulfur in Soil

Subjects: Agronomy

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Sulfur is an essential element in determining the productivity and quality of agricultural products. It is also an element associated with tolerance to biotic and abiotic stress in plants. In agricultural practice, sulfur has broad use in the form of sulfate fertilizers and, to a lesser extent, as sulfite biostimulants.

sulfate sulfite nutraceuticals polysulfanes polysulfides soil microbiome

1. Introduction

Sulfur is one of the most abundant elements on Earth and is an essential element for living beings, of which constitutes on average 1% of dry weight. In plants, S content varies strongly between species, ranging from 0.1 to 6% of dry weight (0.03 to 2 mmol g^{-1} dry weight) [1]. S belongs to the VIA group of the periodic system, where it is found together with O, Se, Te, and Po; naturally, S is a mixture of four isotopes, ^{32}S, ^{33}S, ^{34}S, and ^{35}S. The natural abundance of each is 95.1%, 0.74%, 4.2%, and 0.016%, respectively. Sulfur exists in oxidation states ranging from +6 to −2 (**Table 1**), with the most oxidized state in the form of sulfate (SO_4^{2-}), which is the chemical form that plants absorb from the soil to feed themselves with S [2].

Table 1. Representative sulfur compounds and their oxidation states.

Oxidation State	Representative Compound and Formula	Oxidation State	Representative Compound and Formula
+6	Sulfate, SO_4^{2-}	0	S^0, elemental sulfur. Sulfoxide (R-S(-O)-R such as dimethyl sulfoxide (DMSO). Oxidized derivatives of sulfide and sulfenic acid (RSOH).
+6 and −2	Thiosulfate, $S_2O_3^{2-}$	−1	Disulfide (R-S-S-R) is a persulfide found in the linkages between two cysteine residues in proteins. RSSH denotes persulfides (or hydrosulfides) obtained by the action of H_2S on cysteine residues (R-SH). Thioethers and thiols can be oxidized to disulfides. Major products of decomposition of persulfides are polysulfanes. Thiyl-radical RS*.
+5 and −2	Polythionates ($^{-}O_3S\text{-}S_n\text{-}SO_3^{-}$): Dithionate, $S_2O_6^{2-}$; Trithionate, $S_3O_6^{2-}$; Tetrathionate, $S_4O_6^{2-}$	−2	Sulfide, S^{2-}, polysulfides, S_2^{2-}, S_3^{2-}, S_5^{2-}; carbon disulfide (CS_2); FeS_2; NaHS and Na_2S are sources of S^{2-} and of its conjugated acids SH^{-} and H_2S. Polysulfides (with Sn > 2) contain S^0 atoms, which allows a diversity of oxidation states.
+4	Sulfur dioxide, SO_2; Sulfite, SO_3^{2-}; Disulfite, $S_2O_5^{2-}$; Sulfone, OS(S) the oxidation product of sulfoxides	−2	Hydrogen sulfide (H_2S), disulfane (H_2S_2), and polysulfanes (RSS_nSR, $n > 2$). Polysulfanes contain S^0 atoms, which allows a diversity of oxidation states.
+3	Dithionite, $S_2O_4^{2-}$	−2	Thioethers (C-S-C) such as dimethyl sulfide (DMS), $CH_3\text{-}S\text{-}CH_3$ and dimethyl disulfide (DMDS), $CH_3\text{-}S\text{-}S\text{-}CH_3$.
+2	Carbonyl sulfide (COS), OCS	−2	Thiols (R-SH) such as glutathione (GSH) and methyl mercaptan, $CH_3\text{-}SH$. Thiols are derived from the sulfhydryl group -SH of cysteine, which enables multiple oxidation states (−2 to +6). Thiolates are derivatives of thiols in which a metal or other cation replaces H.

Oxidation State	Representative Compound and Formula	Oxidation State	Representative Compound and Formula
0	Elementary sulfur (S^0), mainly S_8 (cycloocta-S)	−2	Carbon disulfide, CS_2.

ced states 0, −1, and −2. For example, it is known that approximately 40% of enzymes depend for their catalytic activity on the presence of sulfhydryl groups (-SH). These -SH groups participate in redox reactions, provide binding sites for toxic or physiologically important metals, and are related to the detoxification of various xenobiotics. It is also known that the tertiary and quaternary structure of many proteins is the result of the presence of disulfane bonds (-S-S-) formed by the oxidation of -SH groups of cysteine, a sulfur amino acid that, together with methionine, is a key factor in determining the nutritional value of plants, as well as a central element in the metabolism of S in all organisms [2].

For the above reasons, a close relationship between nitrogen and sulfur nutritional status has been found in plants [3][4]. Approximately 80% of nitrogen and sulfur incorporated in organic compounds of plants is found in proteins when both elements are in adequate proportions. The S/N balance of a plant, described by the organic S/N ratio, is in the range of 0.025 (legumes) to 0.032 (grasses) and is relatively constant from one species to another. Therefore, the amount of S required by a plant is strongly dependent on its N nutrition. The consequence is that the availability of S below the needs of the crops does not allow the adequate use of applied N [5].

Compounds as important as β-lactam antibiotics (penicillins, cephalosporins, and cephamycins) have an S atom derived from cysteine. The sulfur compound S-adenosyl-l-methionine (SAM) is the most crucial methylating agent known in all organisms; SAM-mediated transmethylation reactions are essential in the regulation of gene expression, the activity of various enzymes, the synthesis of compounds such as the osmolyte DMSP (dimethyl sulfoniopropionate) and DMS (dimethyl sulfide) gas, as well as in the production of antibiotics [2].

The Earth's S stores are located in the lithosphere, hydrosphere, atmosphere, and biosphere. Human activities result in the extraction of S from the lithosphere (burning of fossil fuels, mining of elemental S and metals) and biosphere (oxidation of organic matter from the soil and burning of biomass). Anthropogenic S is incorporated into the global cycle mainly in the form of SO_2 emitted into the atmosphere [6].

Between the terrestrial and marine masses, there is a constant flow of S via the atmosphere through the gaseous forms of the element (SO_2, COS, H_2S, DMS, and CS_2) [6] and aerosols (mainly SO_4^{2-} from the oxidation of sulfur gases, and <10% of organosulfates) [7], or by runoff from terrestrial to oceanic regions (**Figure 1**). The constant mobilization of S causes changes in the sulfur species that move from one terrestrial compartment to another. Under oxic conditions, the predominant inorganic form of S is SO_4^{2-}, resulting from atmospheric deposition or oxidation of reduced forms of S. In the soil, continuous land tillage that oxidizes soil organic matter and repeated extractions for crops cause the decrease of S stores; for this reason, the regular application of S with the fertilizers is recommended [8].

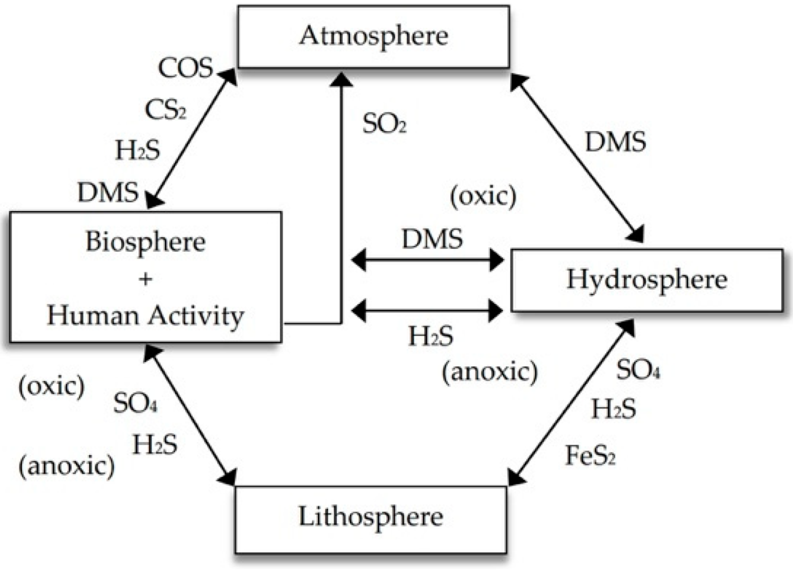

Figure 1. Simplified biogeochemical sulfur cycle. Human activities, fauna, vegetation, and soil microorganisms can be visualized as an interface (as source and sink) to accelerate the transfer of sulfur species between the lithosphere,

atmosphere, and hydrosphere.

In soils, most S is found in organic forms; the inorganic forms are elemental sulfur (S^0) or SO_4^{2-}, the latter can be found as gypsum or be adsorbed in the inorganic exchange matrix. The SO_4^{2-} adsorbed in the soil is in dynamic equilibrium with the soil solution, and the adsorption/desorption quotient inversely depends on the pH value of the soil and the cations present in the exchange matrix, showing higher affinity for $Al^{3+} > Ca^{2+} > K^+$ [9][10].

In soil, SO_4^{2-} is subject to dissimilatory and assimilatory reduction. Dissimilatory reduction occurs when SO_4^{2-} is used as a final acceptor of electrons in the anaerobic metabolism of microorganisms, producing H_2S that is reoxidized in the presence of O_2 or volatilized into the atmosphere. Assimilatory reduction is used by prokaryotes, algae, plants, and fungi for the biosynthesis of organic compounds, e.g., amino acids. Animals and protists cannot perform assimilatory reduction of SO_4^{2-}; therefore, they depend on the organic sulfur compounds synthesized by other organisms [6]. In many crop species, sulfur is an element associated with nutritional quality and density of mineral nutrients, tolerance to stress, and the management of certain pests and pathogens [11][12][13].

In agricultural soils, SO_4^{2-} used by crop plants comes mainly from the contribution of fertilizers with sulfates, such as ammonium sulfate, gypsum, potassium sulfate, magnesium sulfate, single superphosphate, ammonium phosphate sulfate, potassium magnesium sulfate, and sulfates of micronutrients [8][14], as well as the oxidation of S^0, and of S^{2-} contained in organic fertilizers. Another part of the S of crops is obtained from SO_4^{2-} and aerosols coming from precipitation, as well as the absorption by soil and plants of aerosols and gases such as H_2S, COS, and DMS. When S is added to the soil in the form of SO_4^{2-}, plants and aerobic prokaryotes absorb it and incorporate it into a reductive metabolism that produces sulfide (S^{2-}). On the other hand, when S is supplied as S^0 or in the form of organic fertilizers (S^{2-}), it must be oxidized to SO_4^{2-} by the action of soil prokaryotes to be available to plants [2][6][8].

2. Transformations of Elemental Sulfur in Soil

The S available for plants in agricultural ecosystems is in dynamic storage (**Figure 2**). It comes from gaseous forms and aerosols of S from the atmosphere, from dissolved S (mostly SO_4^{2-}) in rain and snow precipitation, and from SO_4^{2-}, which is obtained from the oxidation of S of soil organic matter and S^0. Sulfates can be fixed in the soil exchange matrix or leached to the subsoil [10]. In arid regions, SO_4^{2-} can be stored in large quantities as gypsum in the subsoil, but in areas with higher water availability, leached SO_4^{2-} is mobilized to lower horizons and to the subsoil [15].

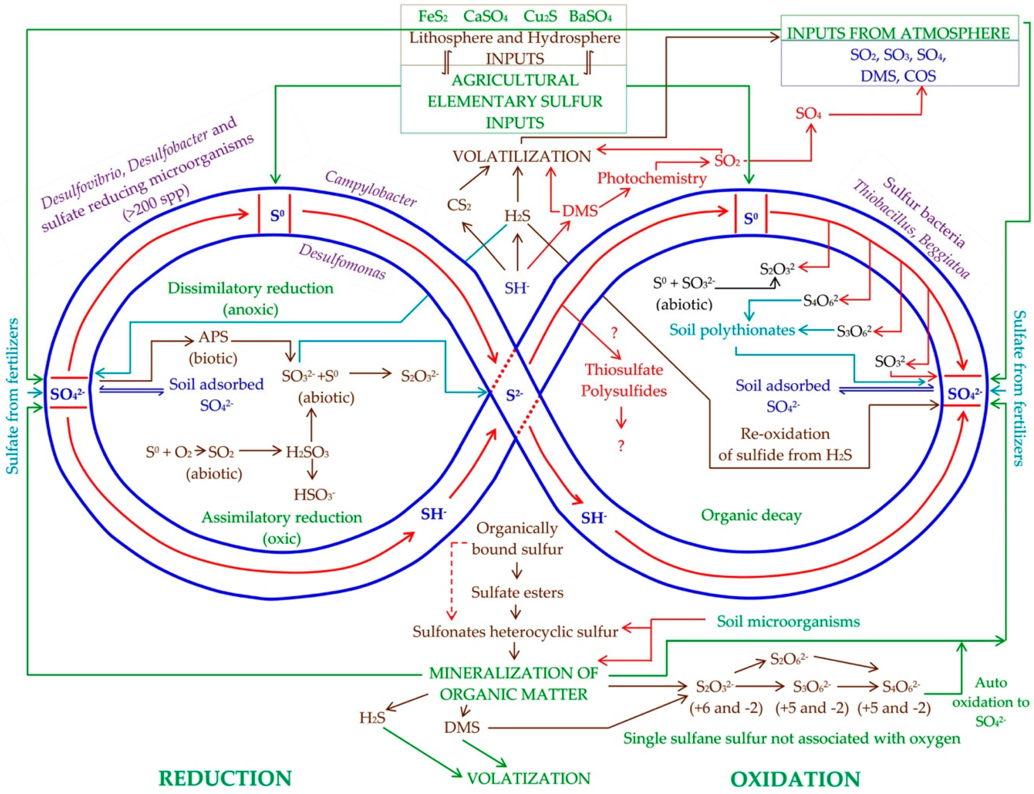

Figure 2. Schematic representation of the flow of sulfur in soil. APS = adenosine 5′-phosphosulfate. Oxidation states of sulfur in the different molecules are: SO_4^{2-} (+6); $S_2O_6^{2-}$ (+5 and −2); $S_4O_6^{2-}$ (+5 and −2); $S_3O_6^{2-}$ (+5 and −2); SO_3^{2-} (+4); SO_2

(+4); $S_2O_3^{2-}$ (+6 and −2); COS (+2); S^0 (0); SH^- (−2); S^{2-} (−2); DMS (−2); CS_2 (−2).

In the anoxic zones of the soil, S^0 and SO_4^{2-} are transformed to H_2S that is volatilized or is reoxidized to S^0 y and sulfate in the oxic zone. Plants and microorganisms take the SO_4^{2-} and reduce it to S^{2-} to incorporate it into a huge variety of organic compounds. Subsequently, these same plants and microorganisms transform a part of the sulfur to H_2S, DMS, and CS_2 [8][16]. The above volatile molecules have been associated with detoxification metabolism, stress tolerance, and signaling in plants and prokaryotes [17][18]. As with iodine [19], soil organic matter can transform the S to volatile forms by means of abiotic reactions, but the rate of transformation is very low in comparison with biotic metabolism of S [16].

Since there are several access ways by which S can enter the agricultural ecosystem, it is not possible to mark a specific starting point. Therefore, arbitrarily, the assumption of an application of S^0 to the soil is taken, and the transformations that this material experiences up to SO_4^{2-} are described. Once in the form of SO_4^{2-}, it is assimilated into plant cells in the form of myriad organic compounds. The final part of the flow of S from soil to plants ends with the production of volatile compounds by plant cells, or in the transformation of the S contained in plant waste (**Figure 2**).

S atoms tend to avoid double bonds, therefore, in the S^0, instead of forming molecules of S_2 (S=S) the S atoms are grouped in the form of cyclic allotropes (cyclosulfur) or as long chains Sn (catena sulfur) [20]. The S^0 used to apply to soil consists mainly of molecules of S_8 (cycloocta-S) that are grouped, forming polymers of variable size; S_8 is the most stable form from a thermodynamic point of view. S_8 is a very electrophilic Lewis acid, so it reacts with nucleophilic anions or Lewis bases such as OH^-, sulfides (S^{2-}), thiols (R-SH), thiolates (RS^-), I^-, CN^-, and SO_3^{2-} [21][22].

S^0 is applied to the soil or substrate in quantities ranging from 20 to 250 kg ha^{-1} yr^{-1}, the last figure being equivalent to 200 mg S^0 kg^{-1} soil. Once in the soil or substrate, S^0 begins to transform into other chemical forms, mainly through biotic processes, and, to a lesser extent, by abiotic processes. The transformation rate is inversely proportional to the particle size and directly proportional to the temperature (Q_{10} = 4.0), humidity availability, and abundance of edaphic microorganisms [8][23][24].

Any factor that decreases bacterial activity, such as temperatures <10 °C or >40 °C and lack of humidity in the soil, will reduce the transformation of S. Flooded or compact soils will have anoxic conditions that induce high rates of conversion of S^0 and SO_4^{2-} into gaseous forms of sulfur [8][24]. The metabolism of S in soils can modify other processes, as in rice paddies, where the use of gypsum amendment has been shown to decrease greenhouse methane emissions [25]. In alkaline soils, it has been observed that the use of S^0 induces acidification (by H_2SO_4), which increases the bioavailability of elements such as P [26].

When it is desired that S^0 produces SO_4^{2-} rapidly available for crops, an S^0 source with a small particle size (<150 µm or 100 mesh) should be chosen. Contrarily, if a long-term impact (two or more consecutive crops) is sought, it is desirable to use S^0 sources with a larger particle diameter, or even granular forms such as S^0 prills or S^0-fortified N-P-K and DAP fertilizers [27][28]. At a temperature of 14 °C, it was found that, in 51 weeks, 51% of S^0 with particle diameter 41 µm (300 mesh) was oxidized, compared to 18% of S^0 with 125 µm (120 mesh). In soils with low temperatures, S^0 sized 41 µm will oxidize at a rate equivalent to S^0 sized 125 µm in soils with higher temperatures [23]. In another experiment, applying 50 kg ha^{-1} of S^0, it was found that 80–90% of S^0 with particles <150 µm was oxidized over a period of 340 days [29].

On the other hand, it has been found that repeated applications of S^0 to soil increase the population and the activity of oxidizing bacteria of S^0 [24]. Accompanying the increase in S^0 oxidant bacteria was a reduction in the number of fungi and protists, while bacterial and actinomycete populations remained stable [30]. Other authors reported a decrease in biomass and bacterial metabolism by applying S^0 annually for five years [31].

When S^0 is in micronized form (<177 µm, <80 mesh) it is used for the control of mites and some fungi [32][33]. The reactivity of micronized S^0 is a consequence of the high quotient surface/volume of the particles, estimated to be 1300 to 1940 cm^2 g^{-1} for S^0 of 125 and 41 µm, respectively [23]. Micronized S^0 can be applied through the foliar route or even by using pressurized irrigation systems to incorporate it into the soil [34][35]. When applied by irrigation system, the problems associated with the application of micronized S^0 (because it is a flammable and irritant material) by dusting machines are reduced [24].

The use of S nanoparticles for the control of pathogens in plants has also been described [36][37]. Taking into account the high value of the surface/volume ratio of S nanoparticles, furthermore being a source of S for rapid assimilation by plants and microorganisms, it is possible that they function as biostimulants [38], and that they provide highly reactive S^0 that works as a tolerance-inducing factor against pathogenic fungi [32][39].

To be available for plants, S^0 applied to the soil or substrate must be oxidized to SO_4^{2-}. The change in the oxidation state of sulfur from 0 to +6 allows reduction equivalents to be obtained ($8H^+ + 6e^-$). The oxidation is carried out by most soil microorganisms, highlighting *Thiobacillus*, *Beggiatoa*, *Desulfomicrobium*, and *Desulfovibrio*, as well as other heterotrophic aerobics S-oxidizing bacteria such as *Bacillus*, *Pseudomonas*, and *Arthrobacter* [2][40]. Two metabolic pathways have been described that allow the oxidation of inorganic S to SO_4^{2-}: the Kelly–Friedrich pathway, which does not involve the production of intermediates such as polythionates, and the Kelly–Trudinger pathway, which includes as an intermediate output tetrathionate ($S_4O_6^{2-}$) and other polythionates [16]. The existence of two different routes and the large number of taxa that carry out the oxidation of S^0 allow a high redundancy, and capacity to tolerate extensive changes in pH and salinity in soils [41][42].

In **Figure 2**, the oxidation activity from S^0 to SO_4^{2-} is presented on the right side, and shows the Kelly–Trudinger pathway with the production of polythionates such as $S_4O_6^{2-}$ and $S_3O_6^{2-}$ (as well as $S_2O_3^{2-}$ and SO_3^{2-}), which serve as a source of reducing potential and possibly act as inducers of stress tolerance in plants, perhaps by containing a single sulfane sulfur not associated with oxygen [16].

Under anoxic conditions, S^0 is produced as part of the dissimilatory reduction of SO_4^{2-}. Later, the S^0 can be assimilated into S^{2-} that will be part of the biomolecules, or it will be volatilized in the case of excess S (see the central section of **Figure 2**). At the left and right ends of **Figure 2**, in the central part, the SO_4^{2-} from fertilizers, precipitation, and mineralization of organic matter is represented. A portion of this SO_4^{2-} forms a soil adsorbed sulfate storage, which will be in dynamic equilibrium with SO_4^{2-} dissolved in the soil solution. Under oxic conditions, SO_4^{2-} is assimilated in S^{2-} by assimilatory reduction and then transformed back into SO_4^{2-} during the organic decay and mineralization of organic matter [42].

As part of the processes of organic decay, mineralization of organic matter, and sulfate reduction, both the soil, through abiotic reactions, and micro-organisms and plants can be source or sink of volatile forms of S, such as H_2S, DMS, COS, CS_2, and SO_2 (**Figure 2**). Generally, under anoxic conditions, the oxidized forms of S are reduced by the soil microbiome to H_2S, CS_2, COS, DMDS, methyl mercaptan, and COS [8]. These gaseous molecules are believed to be part of a mechanism of dissipation of excess S, although participation in other processes is not ruled out [16][43].

In terms of reductive and oxidative microbial reactions, the most abundant forms of sulfur in the soil and the edaphic microbiome are S^{2-}, R-SH, RSSH, polysulfides (RS_n^{2-}), $S_2O_3^{2-}$, SO_3^{2-}, SO_4^{2-} and polythionates [16]. SO_4^{2-} applied as fertilizer or obtained through the processes described above is the form of S that plants assimilate through their roots [40].

At best growth conditions, a plant's sulfur requirement ranges from 2 to 10 μmol g^{-1} plant fresh weight day^{-1} [1]. As the flow of S is a dynamic process where the ecosystem receives S from the atmosphere, precipitation, subsoil water, and fertilizers, and loses S through the process of volatilization of S by soil and plants and by leaching, it is difficult to estimate the actual amount of S that a plant surface absorbs, assimilates, leaches, and volatilizes. As an exercise, let us suppose a single sampling point for a field of maize, for example before the harvest. In one hectare, there may be 78,000 kg of fresh weight ha^{-1}, which would be equivalent to 25 kg of sulfur contained in the plants. However, the 25 kg ha^{-1} accumulated in the plant tissues at that specific sampling time does not include the S volatilized by the plant itself, or that leached, assimilated, or volatilized in the soil and by the microorganisms.

The point to highlight with the data of the previous paragraph is that the S of the soil is in constant exchange and extraction by the crops, atmosphere, and soil water. Therefore, a continuous supply of S is required, which is recommended to be applied in the form of S^0 (40–60 kg ha^{-1}) every one or two years, to maintain the edaphic store.

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