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SULPHATE SORPTION–DESORPTION CHARACTERISTICS OF LATERITIC SOILS OF WEST BENGAL, INDIA

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ABSTRACT: Laboratory study was undertaken in order to get valuable information about the sulphate sorption - desorption characteristics of four red and lateritic soils viz. Sriniketan (Typic ochraqualf), Suri (Haplustalfs), Sainthia (Typic ochraqualfs) and Illambazar (Vertic ochraqualfs) of Birbhum district of West Bengal, India. The sulphate sorption capacity varied widely among the soils and increases with the increase in S concentration of equilibrium solution. However, the adsorption tends to reach a maximum limit with increase in the ambient S concentration. The mean sorbed sulphate sulphur was highest in Suri (84.7 %) followed by that in Sriniketan (83.5%), Illambazar (78.4%) and Sainthia (77.2%). Sulphate sorption data were fitted to the Freundlich and Langmuir adsorption isotherms. The sorption parameters viz. Langmuir Adsorption maximum (b), binding energy (K), maximum buffering capacity were determined from the Langmuir adsorption isothems and Freundlich constants (K and 1/n) were determined from the Freundlich adsorption isotherms, indicate larger variations in the SO_4 sorption behaviour of these soils. The values of Freundlich constant 1/n are less than unity (0.43 to 0.56) indicates that the amount of sulphate adsorbed increased more rapidly than its concentration in soil solution. The sulphate sorption constants derived from Freundlich equations showed that the K values, which is analogous to stability constant for complexes varied from 1.37 to 1.62 mg kg^{-1} in soils. During desorption, the amount of sorbed SO_4 at a given equilibrium concentration was always higher than that during sorption indicating the low desorbability of the sorbed sulphate. The mean recovery of sorbed SO_4-S that was desorbed varied between 52.7 to 66.4 % in soils. The mean desorability pattern of SO_4-S in soils studied followed the order: Illambazar > Sainthia > Suri > Sriniketan. The extent of hysteresis effects involved in the given SO_4 sorption-desorptoin process could well mean that the sorbed SO_4-S has undergone a transformation that imparts to it a greater affinity for the surface.

Key words: Sulphate sorption-desorption characteristics, red and lateritic soils, Freundlich and Langmuir adsorption isotherms, sulphate sorption-desorption parameters

INTRODUCTION

Sulphur deficiencies in India are widespread and scattered throughout 120 districts out of 400 districts [**1**]. Intensification of agriculture with high yielding crop varieties and multiple cropping, coupled with use of high analysis S -free fertilizers and restricted use of organic manures, has accelerated the depletion of soil reserves [**2**]. In India, red and lateritic soils cover an area of about 91 million hectares. These soils are derived from granite, gneiss, schist, and sand stone, shale parent rocks on gently to undulating geomorphic surfaces. Basically these soils are well drained and acidic with lower cation exchange capacity and organic matter content and have mixed or kaolinitic clay mineralogy enriched with sesquioxides [**3**].

Red and Lateritic soils occupy an area of about 28,000 sq km in West Bengal, which is about 28 per cent of the total geographical area of the state [**4**]. Of these, red soil occur in the districts of Birbhum, Bankura, Burdwan, Midnapore and some other parts of Malda and West Dinajpur and gravelly soils are found in the districts of Purulia, part of Midnapore, Bankura and Birbhum [**5**]. Some parts of Birbhum, Burdwan, Bankura, and Midnapore have also laterites and lateritic soils [**6**]. In fact, the red, laterite and associated soils of Eastern India are acidic in soil reaction, light textured, low in organic matter and P and are often deficient in S [**2,6**].

The increasing demand for and escalating cost of sulphur fertilizers during the last decade have stimulated increased interest in the development of technology for more efficient S fertilizer use. Adsorption-desorption characteristics are useful for describing, studying and managing the sulphur status of soils. The adsorption process, which refers to surface S accumulation on soil components, may, in some cases, be accompanied by penetration of the adsorbed S by diffusion into the adsorbent body, leading to further adsorption of the adsorbed species. The general term sorption sometimes used to denote both of these processes taking place simultaneously. Both adsorption and desorption studies have indicated the fertility status of soil, particularly under long-term fertilizer trials dealing with both organic and inorganic materials. The nature of sulphate adsorption in a system can be known by fitting adsorption data to isotherm equations.

The main motivations for describing adsorption curves are: (1) To identify the soil constituents involved in adsorption [**7**,**8**], (2) To predict the amount of fertilizer needs of soil to meet the demand of plant uptake for an optimum yield [**9**], and (3) To study the nature of the adsorption processes to learn more about the mechanism of the process [**10**]. A variety of isotherm shapes are possible, depending upon the affinity of the adsorbent for the adsorbate. In general, for soil sulphur and phosphorus, Langmuir and Freundlich adsorption equations have been used extensively by different workers [**11**,**12**]. Several factors affect the sulphate sorption by soil. Among these, pH and presence of complexing anions [**13**], clay content, extractable Fe and Al [**14**, **15**], soil horizon type [**16**, **12**], organic carbon [**17**], $CaCO_3$ content [**18**] and native extractable sulphate [**19**] are the most important.

The process of desorption refers to the reversible release of adsorbed sulphur into the soil solution phase. Desorption of once sorbed sulphur from soil and clays often has been shown to be irreversible leading to a large hysteresis effect [**12**].

Thus, the concentration of sulphate in soil solution as predicted by sulphate sorption – desorption curves provides a valuable information on sulphur availability to crop plants. It indicates the immediate concentration at which sulphate should be available to plants and concentration of sulphate in water that drains from a particular horizon. Sorption of sulphate can result in the release of OH^- ions to the bulk soil solution and can increase the cation exchange capacity [**26**]. This aspect of SO_4 sorption could be valuable in acidic red and lateritic soils of tropics due to their low cation exchange capacities. Information on the sulphate sorption characteristics of acid soils of India is meager. Hence the present investigation pertaining to study the sulphate sorption–desorption characteristics has special bearing in acid red and lateritic soils of the region.

MATERIALS AND METHODS

In order to study the sorption-desorption characteristics in red and lateritic soils of Birbhum district of West Bengal, India four acidic soil samples (0-0.15m depth) in bulk quantities were collected from Sriniketan (Typic ochraqualf, $23^{0}03^{/}$N and $87^{0}04^{/}$ E), Suri (Typic Haplustalfs, $23^{0}55^{/}$ N and $87^{0}32^{/}$E), Sainthia (Typic ochraqualfs, $23^{0}95^{/}$ N and $87^{0}.68^{/}$ E) and Illambazar (Vertic ochraqualfs, $23^{o}53^{/}$N and 88^{0} $05^{/}$ E) during July 2006. Air-dried soil samples after processing (<2mm) were analyzed for pH [**20**], organic carbon [**21**], CEC [**20**], Free Fe ad Al oxides in a-citrate-bicarbonate-dithionite extracts of soil [**20**], available S [**22**], total S [**23**] and mechanical analysis by Bouyocous hydrometer method [**24**].

Sulphate Sorption Study

Three grams soil was shaken for 24 hrs with 15 ml solution of K_2SO_4 varying in SO_4 concentrations from 0 to 120 mg S L^{-1}. The sulphate sorption studies were carried out in triplicate. The suspension was filtered through Whatman No. 42 filter paper and the amount of S remaining in the solution was determined turbidimetrically [**25**]. The amount of SO_4 sorbed was estimated as the difference between equilibrium SO_4 concentration and initial SO_4 added. The sorption data was fitted to the Freundlich and Langmuir equations as given below.

Freundlich equation:

$$\log x/m = \log k + (1/n) \log C \ (n > 1.0)$$

Langmuir equation:

$$C/ x/m = (1/Kb) + (C/b)$$

Where,

C = equilibrium concentration of SO_4-S in soil solution (mgL^{-1})
x/m = the amount of SO_4-S sorbed (mg kg^{-1})
b = SO_4-S sorption maximum (mg kg^{-1})
K = constant related to bonding energy (L mg^{-1})

n and k are empirical constants in the Freundlich equation depending on the given adsorbate and the temperature (characteristics of the system).

The Freundlich k, Langmuir sorption maxima and the binding energy for the soils under considerations are presented in Table 3.

Sulphate desorption Study

In the desorption experiment, soils were allowed to sorb sulphate as in the sorption studies and this sorbed sulphate was extracted by shaking for 24 hrs with 15 ml KH_2PO_4 solution containing 500 mg P L^{-1}. Sulphate desorption studies were conducted in triplicate. The amount of sulphate desorbed was calculated with respect to the sorbed SO_4.

RESULTS AND DISCUSSION

The sorption-desorption behaviour of four red and lateritic soil samples viz. Sriniketan (Typic ochraqualf), Suri (Typic Haplustalfs), Sainthia (Typic ochraqualfs) and Illambazar (Vertic ochraqualfs) have been presented through tables and figures. Some properties of these soils are presented in Table 1.

Table 1. Some properties of the soils studied for sorption-desorption characteristics of sulphur

Location	Taxonomic classification	pH (1:2)	Org.C (g/kg)	Sand (%)	Silt (%)	Clay (%)	Available S (mg/kg)	Total S (mg/kg)	Free Fe_2O_3 (%)	Free Al_2O_3 (%)
Sriniketan	Typic Ochraqualf	6.2	5.1	77.4	5.6	17	13.5	329.2	2.04	1.95
Suri	Halpuslalf	6.3	3.2	67.5	11.5	21	12.8	344.9	1.63	1.21
Sainthia	Typic Ochraqualf	5.5	5.2	60.2	20.8	19	25.3	401.3	3.60	2.65
Illambazar	Vertic Ochraqualf	5.3	5.3	71	9.2	18.8	9.5	195.4	4.45	4.75

Sulphate Sorption Behaviour

The data related to the SO_4 sorption pattern of soils studied containing various amounts of SO_4 (75 to 600 mg S L^{-1}as K_2SO_4) for 24 hours is shown in Table 2. The sorption of SO_4 in soils increased with increasing levels of the added sulphur.

The mean sorbed SO_4 sulphur in soil was highest in Suri (84.7 %) followed by that in Sriniketan (83.5%), Illambazar (78.4%) and Sainthia (77.2%). The percentage of sorbed SO_4-S in soils varied between 71.4 and 94.8 per cent in Suri, 72.0 and 95.6 per cent in Sriniketan, 68.2 and 93.2 per cent in Illambazar and 66.9 and 89.5 per cent in Sainthia. It is interesting to note that the adsorption though gradually reduced but not in proportion following the trend of more adsorption with increase in SO_4 concentration of equilibrium solution. The hyper-bolic shapes of curve (Figure 3) indicate that adsorption tends to reach a maximum limit with increase in the ambient SO_4 concentration. Therefore, continuous application of S may lead to considerable build up of residual S whatever sulphate would have left following the fate of fixation or adsorption.

Data obtained on SO_4 sorbed by unit weight of soil (X) and equilibrium SO_4 concentration (C) were fitted to the Langmuir and Freundlich adsorption isotherm equation (Figure 1 and Figure 2). The Freuindlich equation gave a better fit of the equilibrium S sorption than did the Langmuir equation.

Table 2. Adsorbed and desorbed sulphate (µg/g soil) in soils treated with different levels of sulphur

Soil		SO_4-S applied (µg/g soil)					
		75	150	300	450	600	Mean
Sriniketan	Adsorbed	71.7(95.6)*	139.9(93.3)	245.9(81.9)	324.2(72.0)	450.1(75.0)	246.4(83.5)
	Desorbed	42.0 (58.5)*	68.3(48.8)	126.3(51.3)	187.5(57.8)	225.5(50.0)	129.9(53.3)
Suri	Adsorbed	71.1 (94.8)	138.9(92.6)	246.0(82.0)	371.3(82.5)	428.7(71.4)	251.2(84.7)
	Desorbed	53.5(75.2)	60.3(43.4)	133.1(54.1)	195.3(52.5)	255.6(59.6)	139.6(55.5)
Sainthia	Adsorbed	67.1(89.5)	123.5(82.3)	234.3(78.1)	312.1(69.3)	401.5(66.9)	227.7(77.2)
	Desorbed	58.3(86.8)	93.3(75.5)	160.3(68.8)	172.0(55.1)	272.1(67.7)	151.2(66.4)
Illambazar	Adsorbed	69.9(93.2)	127.3(84.9)	214.8(71.6)	307.1(68.2)	445.2(74.2)	232.9(78.4)
	Desorbed	50.5(72.2)	77.7(61.0)	161.3(75.0)	225.5(73.4)	250.7(56.3)	153.1(65.7)

*Data in parentheses indicates per cent sorption and desorption at different levels of SO_4-S, respectively.

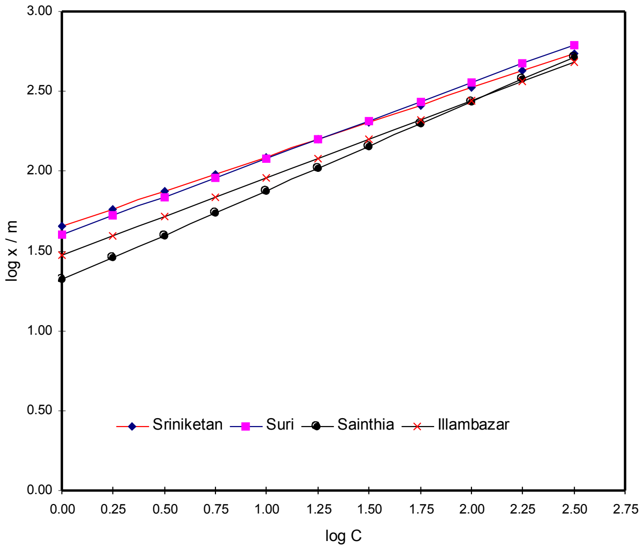

Fig. 1 Freundlich adsorption isotherms for soils

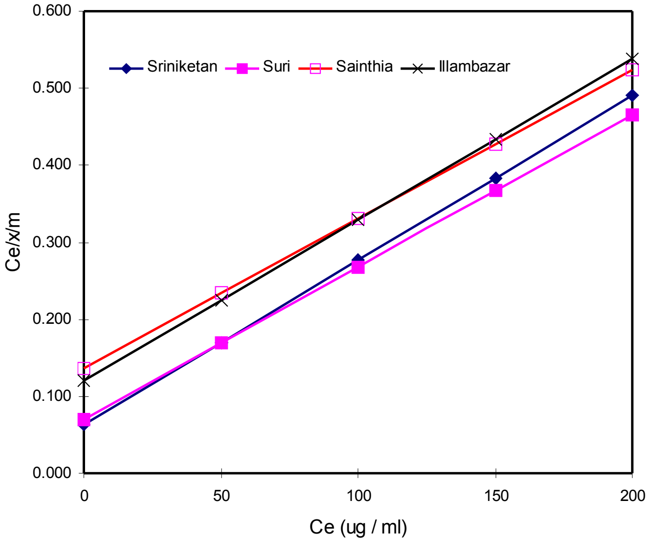

Fig. 2 Langmuir adsorption isotherm for soils studied

The Langmuir sulphate sorption maxima (b), and the constant relating to binding energy (K), calculated from the Langmuir equation (Figure 1), are presented in Table 3 indicating larger variations in the SO_4 sorption behaviour of these soils. Results clearly indicate that Sriniketan soil had the highest SO_4 adsorption capacity (3.70 mg kg^{-1}) followed by Suri (3.03 mg kg^{-1}), Sainthia (2.00 mg kg^{-1}) and Illambazar (1.75 mg kg^{-1}) soils of Birbhum district of West Bengal, India. The Langmuir sorption maxima (b) will decrease with the increasing levels of fertilizer sulphur to soil. This may be due to the fact that at higher dose of sulphur application the availability of exchange sites on clay micelle may be low for adsorption of added sulphur in soil. Results (Table 3) also indicate the variations in bonding energy (K) or affinity constant for the soils studied. The value of bonding energy parameter (K) usually decreased with increasing levels of S in soils. The maximum buffering capacity (MBC) was found to be highest in the soil of Suri (28.54 L kg^{-1}) followed by Sriniketan (26.97 L kg^{-1}), Illambazar (16.62 L kg^{-1}) and Sainthia (12.50 L kg^{-1}). The results suggest that for supplying the same amount sulphur to soil, the Suri soil will require higher amount of sulphur as fertilizer followed by Sriniketan, Illambazar and Sainthia. Higher values of MBC of soil suggest a concomitant greater degree of resistance to changes in concentration of sulphur in soil solution.

Table 3. Langmuir and Freundlich constants of sulphate sorption in acid red and lateritic soils

Soils	Langmuir constants			Freundlich constants	
	Sulphate sorption maximum (b) mg kg^{-1}	Binding energy (K) L mg^{-1}	Maximum buffering capacity (b x K) L kg^{-1}	k (mg kg^{-1})	1/n
Sriniketan	3.70	7.29	26.97	1.62	0.43
Suri	3.03	9.42	28.54	1.57	0.47
Sainthia	2.00	6.25	12.50	1.37	0.56
Illambazar	1.75	9.50	16.625	1.58	0.48

The sulphate sorption constants derived from the Freundlich equation showed that the k values varied from 1.37 to 1.62 (mg kg^{-1}) in soils under consideration (Table 3). The amount of k, which is analogous to stability constant for complexes, was the highest in Sriniketan and the lowest in Sainthia. The values of 1/n varied from 0.43 to 0.55. The values of 1/n are less than unity indicating that the amount of sulphate adsorbed increased more rapidly than its concentration in soil solution.

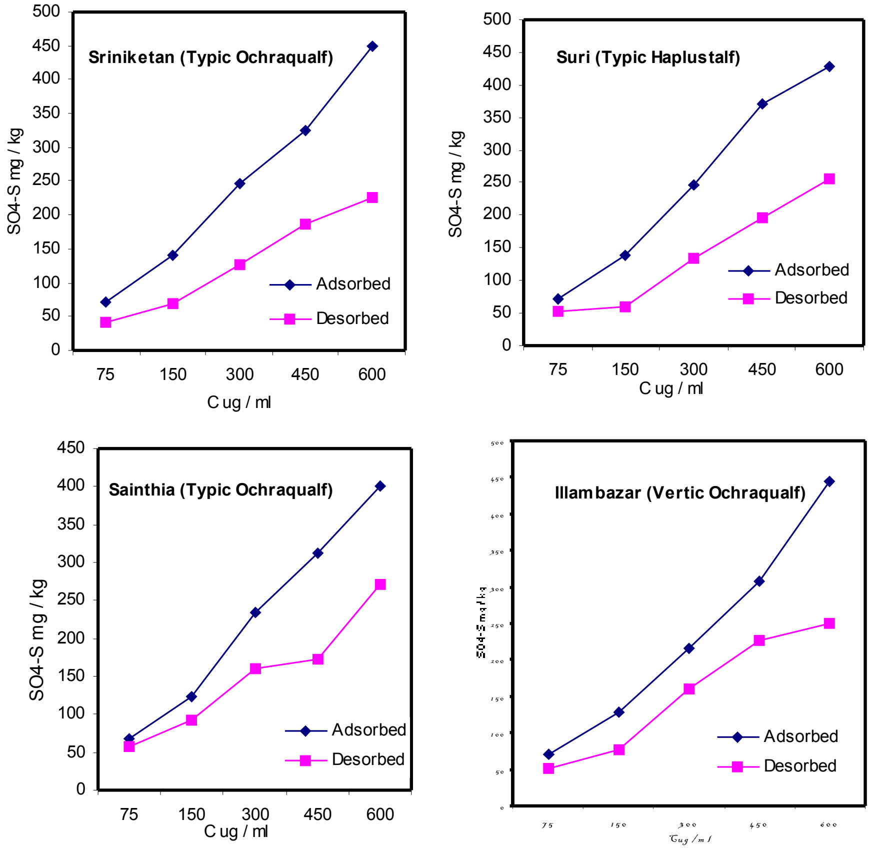

Fig.3. Relationship between sulphate sorbed or desorbed and equilibrium S concentrations in soil solution

The exact mechanisms of sulphate retention by soils remain an enigma in spite of recent advances in soil science. In general, the mechanisms of anion adsorption in soils can be divided into two types: non-specific, where the anion is held electrostatistically as a counter-ion in the diffuse double layer next to a positively charged colloid surface [**27**] and specific or chemisorptions or ligand exchange, in which a bidentate complex is formed where $SO_4^=$ bridges across two Al or Fe atoms to give a six-membered ring following the displacement of two ligands (mainly OH- or H_2O) [**28** ,**29**]. Both mechanisms occur simultaneously in a given soil horizon. Sulphate adsorption is extremely rapid and the initial charge of the adsorption site could be either positive or neutral [**30**]. However, unlike phosphate, $SO_4^=$ adsorption does not appear to disrupt hydrous oxide polymers, break Fe-OH or Al-OH bonds, or displace structural silicates [**31**]. Curtin and Syres [**32**] support an alternative adsorption hypothesis postulated by Uehara and Gillman [**33**] in which SO_4 actually enters the Stern or compact layer, thereby inducing further positive charge on the surface through OH^- release, rather than chemisorptions, in which the adsorbed ion effectively becomes part of the surface.

The amount of $SO_4^=$ adsorbed on the soil surface is dependent not only on the number and type of adsorption sites, but the quantity of adsorbed $SO_4^=$ and soil solution concentration can usually be described adequately by mathematical functions called adsorption isotherms: Freundlich, Langmuir, Tempkin [**27**].

The amount of sulphate adsorbed by soils mainly depends upon free Fe and Al fractions, surface area of these adsorbent and the pH of the system. In the SO_4 adsorption mechanism on iron oxides, two surface hydroxyl groups are replaced by one sulphate ion and two oxygen atoms of the sulphate ion are coordinated each to a different Fe^{3+} ion, resulting in the binuclear bridging surface complex Fe-O-S (O_2)-O-Fe. This complex is formed on the surfaces of the goethite, hematite, lepidocrocite, akageneite and amorphous ferric hydroxides. Similar species may be formed when sulphate is adsorbed on hydrous aluminium oxides [**16**]. Reddy et al. [**12**] also reported the similar sulphate sorption behaviour in some Indian acid soils of Karnataka, Orissa and Madhya Pradesh. The amount of sulphate adsorbed by soils mainly depends upon free Fe and Al fractions, surface area of these adsorbents and the pH of the system [**12**]. The variations in sulphate sorption in soils might be due to variations in soil pH, clay content, and free Fe and Al oxides.

In the present study, the highest sulphate sorption was observed in Suri (Typic Haplustalf) followed by that in Sriniketan (Typic Ochraqualf), Illambazar (Vertic Ochraqualf) and Sainthia (Typic Ochraqualf). It is interesting to note that the adsorption though gradually reduced but not in proportion following the trend of more adsorption with increase in SO_4 concentration of equilibrium solution. The hyper-bolic shapes of curve (Figure 3) indicate that adsorption tends to reach a maximum limit with increase in the ambient SO_4 concentration. Therefore, continuous application of S may lead to considerable build up of residual S whatever sulphate would have left following the fate of fixation or adsorption.

The percentage of sorbed SO_4-S in soils varied between 71.4 and 94.8 per cent in Suri, 72.0 and 95.6 per cent in Sriniketan, 68.2 and 93.2 per cent in Illambazar and 66.9 and 89.5 per cent in Sainthia. In general, Freuindlich equation gave a better fit of the equilibrium S sorption than did the Langmuir equation, especially at higher equilibrium SO_4 concentrations in solutions [**34**]. This may be because of the Freundlich equation, although originally empirical, implies that the affinity for sorption decreases exponentially with increasing saturation in the surface, which is closer to reality than the assumption of constant binding energy inherent in the Langmuir equation [**34**, **35**].

Sulphate Desorption Behaviour

During desorption, the amount of SO_4 desorbed at a given equilibrium SO_4 concentration in solution was always lower than that sorbed during sorption (Table 2). It was also evident from the corresponding higher values of Freundlich k presented in Table 3. The amount of sulphate desorbed by KH_2PO_4 varied considerably among the soils and the desorbed amounts increased in proportion to amounts of sulphate sorbed. The mean desorption in soils was highest in Illambazar (153.1mg kg^{-1}) followed by Sainthia (151.2 mg kg^{-1}), Suri (139.6 mg kg^{-1}), and Sriniketan (129.9 mg kg^{-1}). The percentage of sorbed SO_4-S that was desorbed varied between 48.8 and 58.5 per cent in Sriniketan, 43.4 and 75.2 per cent in Suri, 55.1 and 86.8 per cent in Sainthia and 56.3 and 75.0 per cent in Illambazar soil. The mean recovery was highest in soils of Sainthia (66.4 %) followed by Illambazar (65.7%), Suri (55.5 %) and Sriniketan (52.7%). In earlier studies, Dolui and Jana [**26**] reported the desorption of 71.5 to 80.3 per cent of the sorbed sulphate in Inceptisols and Reddy *et al.* [**12**] also reported the desorption of 73.13 to 80.16 per cent of the sorbed sulphate in four Indian soils collected from Bangalore and Uchagaon in Karnataka, Bhubaneswar (Orissa) and Raipur (Madhya Pradesh).

Depletion of solution sulphate concentration by plant uptake or leaching promotes desorption of sulphate adsorbed on soil mineral surfaces. In this context, adsorbed sulphate in the plant root zone provides an immediate, plant available reserve of sulphur in soil. Most adsorbed sulphate can be readily desorbed as soil solution concentration are depleted. The amount of readily desorbed sulphate tends to decrease with time, particularly at higher temperatures because of either conversion to a more tightly held form or as microbially immobilized forms. Aylmore et al. [**36**] showed that sulphate sorption was essentially reversible on kaolinites but almost irreversible on Fe and Al oxides. Curtin and Syers [**32**], while investigating the release of adsorbed $SO_4^=$, were able to quantitatively remove $SO_4^=$ by extraction with an indifferent electrolyte (1M NaCl). Adsorbed $SO_4^=$, through desorption, is available to plants and microorganisms and in many soils provides the major source of plant available S [**37**]. In other more strongly weathered tropical soils, S deficiency has been observed in crops, despite the soil containing several thousand kg / ha of $SO_4^=$ -S within the root zone [**38**]. The low availability of adsorbed $SO_4^=$ is associated with high Fe and Al oxide content and / or low soil pH. Soils with a low Fe and Al oxide content adsorb and release $SO4^=$ into soil solution more rapidly than do soils with high Fe and Al oxide content [**14**, **15**].

In our present investigation, it was noticed (Table 1) that the soils with relatively low Fe and Al oxide content (Suri and Sriniketan soils) desorbed low amount of $SO_4^=$ than do soils (Illambazar and Sainthia) with relatively high Fe and Al oxide content which is in accordance with the above statement.

Management practices such as liming and phosphate additions are known to release some of the adsorbed $SO_4^=$ [**39**]. The mechanism of $SO_4^=$ release is through ligand exchange and decreased positive charge on the soil surface. This released $SO_4^=$ may be used by plants and microorganisms, or may be leached, depending upon the rainfall regime [**10**].

Desorption of once adsorbed sulphur from soil and clays often has been shown to be irreversible leading to a large hysteresis effect [**12**]. During desorption, the amount of sorbed SO_4 at a given equilibrium concentration was always higher than that during desorption indicating the low desorbability of the sorbed sulphate. In the present investigation, the mean recovery of sorbed SO_4-S that was desorbed varied between 52.7 to 66.4 % in soils. The adsorption isotherm was thus displaced to the left of the sorption isotherm. The extent of hysteresis effect involved in the given SO_4 sorption-desorptoin process is illustrated in Figure 3. Similar observations has also been reported by Reddy et al. [**12**] in studies related to sulphate sorption-desorption characteristics in some acid soils and incase of PO_4 sorption-desorption by Barrow [**40**] and Bolan *et al.* [**41**].

The extent of hysteresis effects involved in the given SO_4 sorption-desorptoin process could well mean that the sorbed SO_4-S has undergone a transformation. Such hysteresis effect leads to an over estimation of the replacing ability of soil to an over estimation of the replacing ability of soil to supply sulphur or phosphorus to the solution, when sulphur or phosphorus solution isotherms are used for the purpose [**42**]. More over it has also been suggested that because adsorption equilibrium was slow, and apparent readsorption during the desorption step is possible [**40**]. The diffusive migration of initially adsorbed sulphur beneath the adsorbing sulphate may also be cited as a probable reason for apparent reversibility of sulphur desorption as has been mentioned by Barrow [**42**] in case of phosphate desorption.

CONCLUSION AND RECOMMENDATION

The overall results of the present study indicate that the sulphate sorption capacity of the red and lateritic soils studied was at a higher degree of magnitude. During desorption, the amount of sulphate desorbed at a given equilibrium S concentration in solution was always lower than the amount of sulphate sorbed during sorption, however, the desorbed amounts increased in proportion to amounts of sulphate sorbed.

The hysteresis effects involved in sulphate sorption-desorption process could well mean that the sorbed S has undergone a transformation that imparts to it a greater affinity for the surface.
To maintain the optimum soil solution sulphur to meet the crop requirements, addition of more S over recommended doses have special bearing in red and lateritic soils.

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