

## ORGANIC SULFUR TRANSFORMATIONS AND SULFUR POOL SIZES IN SOIL AND LITTER FROM A SOUTHERN APPALACHIAN HARDWOOD FOREST<sup>1</sup>

TIMOTHY C. STRICKLAND,<sup>2,3</sup> JOHN W. FITZGERALD,<sup>2</sup> JARU T. ASH,<sup>2</sup> AND WAYNE T. SWANK<sup>4</sup>

**We assayed litter and soil samples from a mixed mature hardwood forest for the capacity to incorporate sulfate-sulfur into organic matter and for the capacity to subsequently mobilize the organic sulfur formed. The levels of total S, carbon-bonded S, ester sulfate, and free and adsorbed S were also determined. We found seasonal variation in sulfur pool sizes, transformation rates between inorganic sulfate and organic sulfur, and in transformation rates among the organic sulfur pools. Higher incorporation rates were observed with samples having higher contents of carbon-bonded S. Amounts of this form of sulfur in a given horizon may reflect levels of amino acid S and sulfolipid, thus reflecting the level of readily available carbon and energy. During sampling dates when ester sulfate contents were low, organic S mobilization rates were elevated and free and adsorbed S pools increased. These observations support the possibility that ester sulfate in soil may serve as an important supply of sulfur for biological uptake.**

The apparent accumulation of inorganic sulfate ( $\text{SO}_4^{2-}$ ) by forested watersheds, as indicated by input-output differences, has been observed at the Coweeta Hydrologic Laboratory in the southern Appalachian mountains (Swank and

Douglass 1977). A portion of this accumulation was attributed to microbial incorporation of sulfate-S into soil and forest floor organic matter (Fitzgerald et al. 1983; Swank et al. 1984). Results of additional work (Strickland et al. 1984) suggest that this process is not unidirectional in these watersheds. Sulfur that has been incorporated into organic matter via covalent bond formation can be mobilized, either as low-molecular-weight organic S components or as inorganic  $\text{SO}_4^{2-}$ . The term *mobilization* (Strickland et al. 1984) is used here to describe the solubilization of a previously nonsalt-extractable sulfur pool that may or may not involve S mineralization. The differences between incorporation and mobilization rates for A1 horizon soil samples from watershed 18 was considered (Fitzgerald et al. 1987) to be responsible for up to 20% of the potential annual S accumulation at Coweeta. Fitzgerald and coworkers (1987) noted further that the O1, O2, and A1 horizons of this watershed contained 1.5, 1.3, and 0.3 mg S  $\text{g}^{-1}$ , respectively, and that >90% of this sulfur was present as organic S in each horizon. Because microbial biomass generally contains <5% of the total S in soil (Saggar et al. 1981; Strick and Nakas 1984), the majority of sulfur in soil from Coweeta is present outside of living cells as organic S.

Although some attention has been given to potential capacities for organic S formation, the forms of sulfur comprising this organic pool have not been characterized for soil and forest floor components from this watershed. In this study we report levels of organic ester sulfate, carbon-bonded S, as well as amounts of free and adsorbed S, in samples collected roughly quarterly for one year. These results are compared with laboratory-derived potentials for  $\text{SO}_4^{2-}$ -S incorporation (organic S formation) and organic S mobilization.

### MATERIALS AND METHODS

Samples were collected between July 1983 and August 1984 from a mixed mature hardwood

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<sup>2</sup> Dept. of Microbiology, Univ. of Georgia, Athens, Ga. 30602.

<sup>3</sup> Present address: Dept. of Forest Science, Oregon State Univ. Corvallis, Oreg. 97331.

<sup>4</sup> Southeastern Forest Exp. Stn., Coweeta Hydrologic Laboratory, Otto, N.C. 28763.

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forest (watershed 18) at the Coweeta Hydrologic Laboratory near Franklin, North Carolina. The predominant soil type is a sandy loam Ashe, a member of the Typic Dystrachrepts. The slope of this catchment is about 50% over a parent material of gneiss. Random samples were collected in triplicate from the O1 (0.5-1 cm thickness), O2 (2-4 cm thickness), and A1 (10 cm) horizons at each of 10 (0.01-ha) plots established along a transect of the watershed (Swank et al. 1984). The average pH of the samples was 5.0, 5.2, and 6.5 for each of the respective horizons. Samples were sieved (<1 cm) to remove large roots and gravel, and triplicates were pooled by plot and horizon. After mixing, the samples were stored at about 4°C in sealed polyethylene bags until analyzed (usually 1-2 wk after collection).

#### *S incorporation and mobilization*

A detailed description of procedures (summarized in Fig. 1) can be found in Strickland et al. (1984). To allow incorporation of sulfate-S into organic matter, samples were incubated at 20°C for 48 h with  $3 \times 10^4$  Bq of  $^{35}\text{S}$ -labeled sodium sulfate (approximately  $4 \times 10^{10}$  Bq  $\text{mmol}^{-1}$ ; Amersham). The final concentration of added sulfate was brought to  $0.72 \mu\text{g g}^{-1}$  with unlabeled anion. Samples were then extracted with acidic phosphate (pH 4.2) to remove non-metabolized sulfate. The loss of microflora, which might take place during this extraction, was minimized by the addition of an aqueous extract of untreated parent material, and the samples were reincubated at 20°C for 24 h. This second incubation was terminated by extraction with salt, followed by alkaline oxidation and acid hydrolysis. The percentage of  $^{35}\text{S}$  present in each extract was determined by liquid scintillation counting. Sulfur incorporated into organic matter was determined as  $^{35}\text{S}$  recovered in extracts A plus B (Fig. 1). The amount of  $^{35}\text{S}$  mobilized from the newly formed organic S was determined as the amount of  $^{35}\text{S}$  in extract A divided by the sum of the amounts of  $^{35}\text{S}$  in extracts A and B, and expressed as a percentage. This assay for organic S mobilization was carried out, by necessity, under conditions of  $\text{SO}_4^{2-}$  depletion. The validity of this approach is justified in view of long-standing observations that most of the sulfur in agricultural as well as forest soils is found in the organic rather than inorganic form (see for example, Fitzgerald 1976 and Tables 1-3).

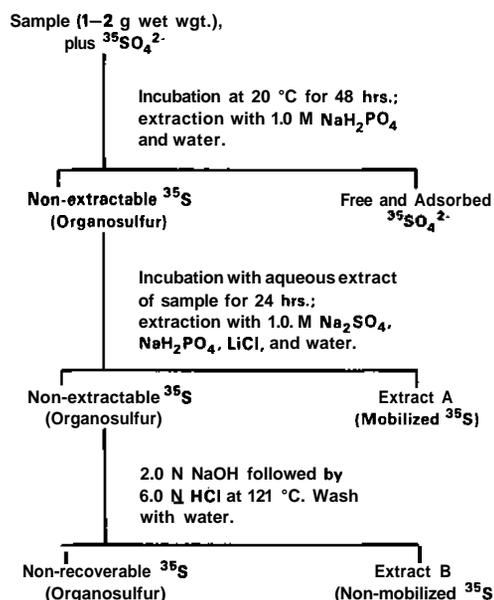


FIG. 1. Procedure for measuring organic sulfur formation and mobilization in litter and soil.

Although indirect, this method for determining organic S formation and mobilization has been verified directly by field incubation for organic S formation (Strickland et al., 1986a) and by isolation and characterization of the organic S formed from  $\text{SO}_4^{2-}$  (Fitzgerald et al. 1985b). Moreover, the direct addition of the isolated organic S to all three horizons resulted in mobilization (solubilization and S mineralization) of this fraction after laboratory incubation (Strickland and Fitzgerald 1984) and field incubation (Strickland et al. 1986b).

#### *Characterization of organic S in situ*

Ester sulfate ( $\text{C-O-SO}_3^-$ ) was determined with boiling hydriodic acid (HI) by the method of Johnson and Nishita (1952) as modified by Landers et al. (1983). Total S was determined by hypobromite oxidation followed by HI reduction (Tabatabai and Bremner 1970a). Because HI will not reduce the C-S (amino acid) or C-SO<sub>3</sub><sup>-</sup> (sulfonate) linkage, carbon-bonded S (amino acid and sulfonate S) was considered to be that fraction of the total S that was not reduced by HI. The amount in this form was determined by difference. For each sampling date, determinations were made individually for samples from each of the 10 plots in the transect, and a watershed mean ( $n = 10$ ) was calculated for

levels of S incorporation and mobilization and for the amounts of total, carbon-bonded, insoluble ester sulfate, and free (soluble) and adsorbed S (sulfate and ester sulfate) present in situ. This latter fraction was obtained by sequential water and phosphate extraction, as described previously (Strickland et al. 1986). The amount of sulfur in the combined extracts was determined by HI reduction.

For sulfur pool sizes expressed as kg S ha<sup>-1</sup> (A1 horizon, Table 3), the quantity of substrate was estimated as 1.19 x 10<sup>6</sup> kg ha<sup>-1</sup>. Because substrate quantities in the O1 and O2 layers (approximately 3 X 10<sup>3</sup> kg ha<sup>-1</sup> each) vary substantially during the year and were not measured at each sampling period, sulfur pool sizes and transformations are reported on a µg g<sup>-1</sup> basis for these horizons (Tables 1-2).

#### RESULTS AND DISCUSSION

With A1 horizon samples, significant differences (Table 3) were observed between all pools. Absolute levels of carbon-bonded S followed the same pattern of fluctuation as did levels of total S during the sampling period, increasing from November to February and again from May to August (Table 3). Although variations in free

and adsorbed S may be too great to attach any significance to the small standard errors and relatively large differences in means observed between the lows in November and May and the highs in July and August suggest that there may well be an increase in inorganic SO<sub>4</sub><sup>2-</sup> levels in the summer months. In contrast to the levels of free and adsorbed S, the percentage of total S as insoluble ester sulfate was highest in February and lowest in July and August.

Because the mineral horizons serve as a much larger reservoir of sulfur than do the organic horizons (Table 4), pool fluctuations in the former horizon should be more representative of overall transformation processes that occur on the watershed. Observations made with organic layers do, however, serve a purpose in estimating effects on S transformations at the process level. There were significant differences in the percentages of total S contributed by individual sulfur species in the O1 and O2 horizons (Tables 1 and 2). Seasonal fluctuation in pool size as a percentage of total S indicates changes in transformation rates among pools. Carbon-bonded S was the dominant form of sulfur present in all three horizons at any time during the year (except in the A1 horizon during November 1983, Table 3). By horizon, the distribution of this

TABLE 1  
*Seasonal fluctuation in sulfur pool sizes and transformations in the O1 horizon*

| Determination  | Mean (± SE) <sup>a</sup> |              |              |              |                |
|--|--------------------------|--------------|--------------|--------------|----------------|
|  | July 1983                | Nov. 1983    | Feb. 1984    | May 1984     | Aug. 1984      |
| Total S,<br>µg g <sup>-1</sup>                         | ‡1035.4(95.2)            | t604.7(55.6) | t696.5(64.2) | t539.5(41.0) | £1020.2(110.6) |
| C-bonded S,<br>µg g <sup>-1</sup>                      | ‡816.4(69.9)             | t492.3(64.0) | t613.1(64.5) | t459.1(35.3) | £838.2(101.6)  |
| % of Total S <sup>b</sup>                              | 79.7(2.5)                | 79.0(4.2)    | 87.4(1.3)    | 84.8(1.4)    | 81.7(2.4)      |
| Ester sulfate,<br>µg g <sup>-1</sup>                   | ‡187.6(36.9)             | t80.3(16.8)  | ‡39.6(7.5)   | t80.4(9.5)   | £159.0(26.9)   |
| % of Total S   | ‡17.2(2.5)               | ‡‡15.3(3.9)  | £6.0(1.2)    | ‡‡15.2(1.4)  | t15.9(2.2)     |
| Free + adsorbed S, <sup>c</sup><br>µg g <sup>-1</sup>  | £t31.3(4.1)              | £t32.1(5.9)  | ‡43.8(8.3)   | 0            | £22.9(3.6)     |
| % of Total S   | ‡3.1(0.4)                | t5.7(1.2)    | t6.7(1.3)    | 0            | £2.4(0.4)      |
| SO <sub>4</sub> -S incorporated,<br>µg g <sup>-1</sup> | ‡‡0.45(0.04)             | t0.35(0.04)  | t0.32(0.04)  | £t0.44(0.05) | £0.54(0.07)    |
| Organic S mobilized,<br>µg g <sup>-1</sup>             | 0.14(0.01)               | t0.07(0.01)  | t0.08(0.02)  | t0.11(0.01)  | 0.24(0.02)     |
| % 24 h <sup>-1</sup>                                   | ‡16.4(1.0)               | £9.4(0.9)    | ‡‡12.5(1.7)  | £t12.1(0.5)  | 23.9(1.7)      |

<sup>a</sup> Values in any row preceded by the same symbol are not significantly different (Tukey's HSD test, *P* < 0.05); *n* = 10 for each determination and sampling date.

<sup>b</sup> No significant differences.

<sup>c</sup> Water-soluble and adsorbed S includes inorganic sulfate and ester sulfate.

TABLE 2  
Seasonal fluctuation in sulfur pool sizes and transformations in the O2 horizon

| Determination  | Mean ( $\pm$ SE) <sup>a</sup> |               |               |               |              |
|--|-------------------------------|---------------|---------------|---------------|--------------|
|  | July 1983                     | Nov. 1983     | Feb. 1984     | May 1984      | Aug. 1984    |
| Total S,<br>Mg g <sup>-1b</sup>                        | 1344.4(165.1)                 | 1038.6(95.4)  | 1105.0(94.4)  | 1198.3(119.0) | 1320.8(94.1) |
| C-bonded S,<br>Mg g <sup>-1b</sup>                     | 936.8(140.3)                  | 702.3(87.2)   | 789.3(83.0)   | 948.4(111.1)  | 955.8(86.7)  |
| % of Total S   | ††68.0(2.8)                   | †65.7(3.7)    | †70.3(2.1)    | †77.9(2.1)    | †72.1(2.9)   |
| Ester sulfate,<br>μg g <sup>-1</sup>                   | †119.7(13.8)                  | \$246.5(28.8) | \$315.7(18.9) | \$234.7(22.2) | 1189.9(38.2) |
| % of Total S   | \$10.2(1.8)                   | †24.8(3.1)    | †29.7(2.1)    | †20.8(2.1)    | \$14.1(2.4)  |
| Free + adsorbed S, <sup>c</sup><br>μg g <sup>-1</sup>  | 275.5(32.0)                   | 89.8(7.2)     | 0             | 15.3(2.9)     | 175.2(28.9)  |
| % of Total S   | 21.2(2.4)                     | 9.4(1.2)      | 0             | 1.3(0.2)      | 13.8(2.2)    |
| SO <sub>4</sub> -S incorporated,<br>μg g <sup>-1</sup> | †0.15(0.01)                   | \$0.24(0.05)  | †0.15(0.03)   | \$0.28(0.05)  | \$0.25(0.02) |
| Organic S mobilized,<br>μg g <sup>-1</sup>             | ††0.15(0.02)                  | †0.09(0.01)   | †0.09(0.01)   | †0.15(0.02)   | \$0.20(0.02) |
| % 24 h <sup>-1</sup>                                   | †47.1(3.3)                    | \$21.6(1.8)   | ††32.7(3.8)   | ††29.7(1.6)   | †41.0(1.9)   |

<sup>a</sup> See footnotes, Table 1.

TABLE 3  
Seasonal fluctuations in sulfur pool sizes and transformations in the A1 horizon

| Determination  | Mean ( $\pm$ SE) <sup>a</sup> |               |              |                |              |
|--|-------------------------------|---------------|--------------|----------------|--------------|
|  | July 1983                     | Nov. 1983     | Feb. 1984    | May 1984       | Aug. 1984    |
| Total S,<br>kg ha <sup>-1</sup>                          | †385.8(29.8)                  | \$315.9(24.6) | †370.9(48.3) | \$307.9(43.6)  | †372.5(38.9) |
| C-bonded S,<br>kg ha <sup>-1</sup>                       | ††161.3(23.2)                 | \$127.4(19.4) | †187.8(36.9) | \$1178.0(31.4) | 242.7(34.5)  |
| % of Total S   | \$40.5(3.3)                   | \$38.6(3.8)   | ††47.1(4.4)  | ††55.6(3.5)    | †62.2(4.0)   |
| Ester sulfate,<br>kg ha <sup>-1</sup>                    | 1128.3(9.6)                   | 1120.1(10.1)  | 159.0(18.2)  | 1107.7(14.5)   | 46.7(4.5)    |
| % of Total S   | \$34.3(2.7)                   | \$38.8(2.8)   | †45.1(3.5)   | \$37.3(4.3)    | 13.3(1.7)    |
| Free + adsorbed S, <sup>c</sup><br>kg ha <sup>-1</sup>   | †96.9(9.4)                    | †67.9(7.0)    | \$24.0(3.7)  | \$22.2(7.5)    | †83.0(6.1)   |
| % of Total S   | †25.2(1.9)                    | †22.4(2.8)    | \$7.8(1.5)   | \$7.1(2.1)     | †24.5(3.5)   |
| SO <sub>4</sub> -S incorporated,<br>kg ha <sup>-1d</sup> | †3.02(0.18)                   | ††3.83(0.50)  | ††4.00(0.42) | \$5.41(0.64)   | †3.23(0.20)  |
| μg g <sup>-1</sup> 48h <sup>-1b</sup>                    | 0.06(0.003)                   | 0.07(0.009)   | 0.07(0.008)  | 0.10(0.012)    | 0.06(0.004)  |
| Organic S mobi-<br>lized,<br>kg ha <sup>-1d</sup>        | †3.34(0.29)                   | \$2.33(0.26)  | \$2.90(0.26) | 4.08(0.44)     | †3.48(0.29)  |
| % 24 h <sup>-1</sup>                                     | †54.8(1.8)                    | 30.9(0.8)     | \$37.3(2.0)  | \$38.4(1.4)    | †54.4(1.4)   |
| S accumulated, <sup>e</sup><br>kg ha <sup>-1</sup>       | -0.32                         | 1.50          | 1.10         | 1.33           | -0.25        |

<sup>a</sup> See footnotes, Table 1.

<sup>d</sup> Values are extrapolated to a quarterly basis.

<sup>e</sup> SO<sub>4</sub>-S incorporated minus organic S mobilized (kg ha<sup>-1</sup> qtr<sup>-1</sup>); negative values indicate that mobilization exceeds incorporation.

pool as a percentage of total S was 01 > 02 > A1. Compared with levels of carbon-bonded S, ester sulfate constituted a lower percentage of the total S in all horizons. With the exception

of July and August, the distribution of this form of sulfur exhibited a pattern opposite to that of the carbon-bonded S (i.e., A1 > 02 > 01, Tables 1-3). Because the free and adsorbed S fractions

may also include organic S as ester sulfate, conversion of substantial amounts of insoluble ester sulfate to a soluble form may account for this discrepancy during these months. **Hydriodic** acid reduction will not distinguish this organic linkage from  $\text{SO}_4^{2-}$ , and ion **chromatographic** determinations indicate that up to 27% of the total S in the A1 horizon can be composed of soluble and adsorbed ester sulfate (unpublished results, this laboratory).

Seasonal fluctuations in  $\text{SO}_4^{2-}$ -S incorporation and organic S mobilization potentials were also observed (Tables 1-3). Further, a relationship may also exist between these rates and sulfur pool size. As mentioned above, fluctuations occurred in the O2 and A1 horizons in the percentages of total S contributed by individual pools of sulfur. Of particular interest was the percentage of ester sulfate in the O2 and A1 horizons (Tables 2 and 3). In both horizons, the ester sulfate content (percentage of total S) was lowest during July and August. This decrease in ester content occurred **concomitantly** with maximum percentages of free and adsorbed S and with peak organic S mobilization rates (percentage  $24 \text{ h}^{-1}$ ). Taken together these results suggest a net capacity for loss of sulfur from the system during these summer months. The method used here to estimate organic S mobilization (i.e., phosphate extraction to remove  $\text{SO}_4^{2-}$ ) by necessity created an  $\text{SO}_4^{2-}$  deficiency in the samples. Under these conditions, the only source of sulfur for growth is the organic S pool. This

technique may, therefore, be considered to give an estimate of the capacity of these soils to supply  $\text{SO}_4^{2-}$  by drawing on the organic S pools in the event of  $\text{SO}_4^{2-}$  depletion. It is interesting to note that the amount of carbon-bonded S (percentage of total S, Tables 2 and 3) did not change significantly between July and November or between May and August, as did ester sulfate. Thus, it would appear that the ester sulfate pool may serve as an immediate reservoir of sulfur that can be mineralized in response to biological demand. The same observation was made previously for agricultural soils (Bettany et al. 1980) and may reflect the ready availability of sulfatase enzymes in soil capable of **hydrolyzing** these esters (Tabatabai and Bremner 1970b; Fitzgerald et al., 1985). Extrapolation of 48-h incubation values indicates that in the absence of vegetation, a capacity to mobilize approximately  $12.9 \text{ kg S ha}^{-1} \text{ yr}^{-1}$  (or 3.7% of the total organic S, Table 4) exists in the A1 horizon in response to microbiological demand.

It was noted previously (Strickland and Fitzgerald 1984; Swank et al. 1984) that the mineral horizons incorporate much more  $\text{SO}_4^{2-}$ -S  $\text{ha}^{-1}$  than do the litter layers because of the larger amount of substrate present. Those results were substantiated in this study (Table 4). However, the rate ( $\mu\text{g g}^{-1} 48 \text{ h}^{-1}$ ) of  $\text{SO}_4^{2-}$ -S incorporation into organic matter was highest in the O1 layer and decreased in the O2 and A1 horizons. Moreover, while the O1 layer exhibited the highest rate of  $\text{SO}_4^{2-}$ -S incorporation, this horizon also

TABLE 4  
Mean amounts of organic S formed and mobilized ( $\pm 1 \text{ SE}$ ) in relation to in situ levels of sulfur<sup>a</sup>

| Determination                        | O1                 | O2          | A1            |
|--------------------------------------|--------------------|-------------|---------------|
| S incorporated,                      |                    |             |               |
| $\text{kg ha}^{-1} \text{ yr}^{-1}$  | 0.23(0.01)         | 0.12(0.01)  | 15.61(0.89)   |
| $\mu\text{g g}^{-1} 48\text{h}^{-1}$ | 0.42(0.02)         | 0.22(0.02)  | 0.07(0.004)   |
| Organic S mobilized,                 |                    |             |               |
| % $24\text{h}^{-1}$                  | 14.82(0.91)        | 33.87(1.68) | 42.89(1.54)   |
| $\text{kg ha}^{-1} \text{ yr}^{-1}$  | *0.07(0.01)        | *0.07(0.01) | 12.89(0.64)   |
| <b>Total S,</b>                      |                    |             |               |
| $\text{kg ha}^{-1}$                  | <b>*2.32(0.13)</b> | *3.59(0.15) | 349.87(17.08) |
| Carbon-bonded S,                     |                    |             |               |
| % <sup>b</sup>                       | 82.56(1.21)        | 70.93(1.34) | 48.98(2.09)   |
| Ester sulfate S,                     |                    |             |               |
| % <sup>b</sup>                       | 13.84(1.20)        | 20.33(1.44) | 33.75(2.07)   |
| Free and adsorbed S,                 |                    |             |               |
| % <sup>b</sup>                       | 3.60(0.51)         | 8.63(1.29)  | 17.24(1.58)   |

<sup>a</sup>  $n = 50$ . Values in any row preceded by an asterisk are not significantly different (Tukey's HSD test,  $P < 0.05$ ).

<sup>b</sup> Percentage of total S.

had the lowest rate of organic S mobilization. The opposite relationship was true for the A1 horizon. Further, the horizon with the highest proportion of carbon bonded S (O1 layer) exhibited the greatest rate of S incorporation, while the horizon with the highest proportion of ester sulfate (A1 horizon) had the highest capacity for organic S mobilization. We suggest that the high levels of carbon-bonded S in the O1 and O2 horizons reflect high levels of amino acid S and plant sulfolipid. Plant protein provides a readily available source of carbon and energy for soil **microflora**, as do the glucose, glycerol, and fatty **acyl** units comprising the plant sulfolipid (Harwood 1980). The higher levels of incorporation in the litter layers than in the A1 horizon may reflect higher microbial metabolic rates due to increased availability of carbon-bonded S. Stimulation of  $\text{SO}_4^{2-}$ -S incorporation by carbon and energy sources is well documented (Freney et al. 1971; Fitzgerald et al. 1983; Strickland and Fitzgerald 1985).

## REFERENCES

- Bettany, J. R., S. Sagggar, and J. W. B. Stewart. 1980. Comparison of the amounts and forms of sulfur in soil organic matter fractions after 65 years of cultivation. *Soil Sci. Soc. Am. J.* 44:70-75.
- Fitzgerald, J. W. 1976. Sulfate ester formation and hydrolysis: A potentially important yet often ignored aspect of the sulfur cycle of aerobic soils. *Bacteriol. Rev.* 40:698-721.
- Fitzgerald, J. W., J. T. Ash, T. C. Strickland, and W. T. Swank. 1983. Formation of organic sulfur in forest soils: A biologically mediated process. *Can. J. For. Res.* 13:1077-1082.
- Fitzgerald, J. W., T. C. Strickland, and J. T. Ash. 1985a. Isolation and partial characterization of forest floor and soil organic sulfur. *Biogeochemistry* 1:155-167.
- Fitzgerald, J. W., M. E. Watwood, and F. A. Rose. 1985b. Forest floor and soil **arylsulphatase**: Hydrolysis of **tyrosine** sulphate, an environmentally relevant substrate for the enzyme. *Soil Biol. Biochem.* 17:885-887.
- Fitzgerald, J. W., W. T. Swank, T. C. Strickland, J. T. Ash, D. D. Hale, T. L. Andrew, and M. E. Watwood. 1987. Sulfur pools and transformations in litter and surface soil of a hardwood forest. *In* Forest hydrology and ecology at Coweeta. W. T. Swank and D. A. Crossley (eds.). Springer-Verlag, New York (*forthcoming*).
- Freney, J. R., G. E. Melville, and C. H. Williams. 1971. Organic sulphur fractions labelled by the addition of  $^{35}\text{S}$ -sulphate to soil. *Soil Biol. Biochem.* 3:133-141.
- Harwood, J. L. 1980. Sulfolipids. *In* The biochemistry of plants, vol. 4. P. K. Stumpf and E. E. Conn (eds.) Academic, New York, pp. 301-320.
- Johnson, C. M., and H. Nishita. 1952. Microestimation of sulfur in plant materials, soils, and irrigation waters. *Anal. Chem.* 24:736-742.
- Landers, D. H., M. B. David, and M. J. Mitchell. 1983. Analysis of organic and inorganic sulfur constituents in sediments, soils and water. *Int. J. Environ. Anal. Chem.* 14:245-256.
- Sagggar, S., J. R. Bettany, and J. W. B. Stewart. 1981. Measurement of microbial sulfur in soil. *Soil Biol. Biochem.* 13:493-498.
- Strick, J. E., and J. P. Nakas. 1984. Calibration of a microbial sulfur technique for use in forest soils. *Soil Biol. Biochem.* 16:289-291.
- Strickland, T. C., and J. W. Fitzgerald. 1984. Formation and mineralization of organic sulfur in forest soils. *Biogeochemistry* 1:79-95.
- Strickland, T. C., J. W. Fitzgerald, and W. T. Swank. 1984. Mobilization of recently formed forest soil organic sulfur. *Can. J. For. Res.* 14:63-67.
- Strickland, T. C., and J. W. Fitzgerald. 1985. Incorporation of sulphate-sulphur into organic matter extracts of litter and soil: Involvement of ATP **sulphurylase**. *Soil Biol. Biochem.* 17:779-784.
- Strickland, T. C., J. W. Fitzgerald, and W. T. Swank. 1986a. *In situ* measurements of sulfate incorporation into forest floor and soil organic matter. *Can. J. For. Res.* 16:549-553.
- Strickland, T. C., J. W. Fitzgerald, and W. T. Swank. 1986b. *In situ* mobilization of  $^{35}\text{S}$ -labelled organic sulphur in litter and soil from a hardwood forest. *Soil Biol. Biochem.* 18:463-468.
- Swank, W. T., and J. E. Douglass. 1977. Nutrient budgets for undisturbed and manipulated hardwood forest ecosystems in the mountains of North Carolina. *In* Watershed research in eastern North America, vol. 1. D. L. Correll (ed.). Smithsonian **Inst.**, Washington, D. C. pp. 343-363.
- Swank, W. T., J. W. Fitzgerald, and J. T. Ash. 1984. Microbial transformations of sulfate in forest soils. *Science* 223:182-184.
- Tabatabai, M. A., and J. M. Bremner. 1970a. An alkaline oxidation method for determination of total sulfur in soils. *Soil Sci. Soc. Am. Proc.* 34:62-65.
- Tabatabai, M. A., and J. M. Bremner. 1970b. **Arylsulphatase** activity of soils. *Soil Sci. Soc. Am. Proc.* 34:225-229.