

# **BIOMONITORING FORESTS NEAR TWO SOUR GAS PROCESSING PLANTS**

*D.G. Maynard*

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## ABSTRACT

A biomonitoring study was carried out in 1981 and 1985 to assess the impact of gaseous and particulate sulfur (S) pollution from two sour gas processing plants on lodgepole pine (*Pinus contorta* var. *latifolia* Engelm.) forests in west-central Alberta. Past and present levels of S gas emissions have had little or no measurable effect on soils and vegetation in areas not contaminated with elemental S ( $S^0$ ), other than a possible elevation of S concentrations in the foliage at two sites within 6 km of the gas plants. At the sites heavily contaminated by  $S^0$ , it is possible that sulfur dioxide may have had some impact; however, the overwhelming influence of  $S^0$  at these sites precluded any measurements. Significant impact of  $S^0$  was limited to areas contaminated with  $>5000 \text{ mg kg}^{-1}$  of  $S^0$  (approximately  $450 \text{ kg ha}^{-1}$  of  $S^0$ ) in the LFH horizon, generally within 1 km of the  $S^0$  storage or handling facilities. Only the under-story vegetation was affected in 1981; however, in 1985, at the most heavily contaminated site the pine exhibited considerably more needle loss than at any other site and little or no current-year growth. There was approximately 5 to 10 years from the time of initial deposition and oxidation of  $S^0$  until changes in the soil chemistry were observed followed by the manifestation of symptoms in pine. Damage to the vegetation as a result of  $S^0$  contamination occurred first in the moss, followed by the herb and shrub, and finally the tree layer.

## RESUME

Une étude de biosurveillance a été effectuée en 1981 et 1985 pour évaluer l'impact de la pollution par le soufre (S) à l'état gazeux et particulaire dégagé par deux usines de gaz sulfureux sur les forêts de pins tordus latifoliés (*Pinus contorta* var. *latifolia* Engelm.) du centre-ouest de l'Alberta. Les niveaux antérieurs et actuels d'émissions de gaz sulfureux ont eu très peu d'effet mesurable, sinon aucun, sur les sols et la végétation dans les régions non contaminées par du soufre élémentaire ( $S^0$ ), à part une augmentation possible des teneurs en S du feuillage des arbres de deux stations situées à moins de 6 km des usines de gaz. Dans les stations fortement contaminées par le  $S^0$ , il est possible que l'anhydride sulfureux puisse avoir eu certaines incidences; l'influence prépondérante du  $S^0$  dans ces stations a toutefois empêché la prise de telles mesures. Le  $S^0$  avait un impact significatif dans les endroits contaminés à plus de  $5000 \text{ mg kg}^{-1}$  (environ  $450 \text{ kg ha}^{-1}$  de  $S^0$ ) dans les sous-horizons L, F et H, généralement à moins de 1 km des installations de stockage ou de manutention du  $S^0$ . Seule la végétation du sous-étage était affectée en 1981; toutefois, en 1985, la chute des aiguilles des pins de la station la plus fortement contaminée était beaucoup plus importante que partout ailleurs, la croissance de l'année en cours étant nulle ou presque. De 5 à 10 ans se sont écoulés entre le dépôt initial et l'oxydation du  $S^0$  et le moment où des modifications de la chimie du sol ont été observées suivies de l'apparition de symptômes chez les pins. Les dommages causés à la végétation par la contamination par le  $S^0$  se sont d'abord manifestés dans la strate muscinale, puis dans la strate herbacée et arbustive et, enfin, dans la strate arborée.

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## NOTE

*The exclusion of certain manufactured products does not necessarily imply disapproval nor does the mention of other products necessarily imply endorsement by Forestry Canada.*



## INTRODUCTION

Many studies have been carried out on the pattern and impact of pollutant deposition in Alberta. The majority of these studies have been centered around point sources such as the oil sands operations, coal-fired power plants, and sour gas processing plants (Legge et al. 1986; Addison et al. 1984; Lore 1984). Unlike in Europe and eastern North America, the potential in Alberta for regional air pollutant effects on forests does not appear to be an immediate concern (Fraser et al. 1985; Addison et al. 1986). The sour gas processing industry is the largest sulfur (S) producer in Alberta (Colley and Picard 1987), with sulfur dioxide (SO<sub>2</sub>) and dust fall consisting of elemental S (S<sup>0</sup>) being the two main pollutants. Considerable controversy exists on the potential for acidification of Alberta soils and on the most probable pollutant involved.

Nyborg et al. (1977) suggested that, as a result of SO<sub>2</sub> deposition downwind of a sour gas processing plant, soil pH would drop between 0.5 and 1.0 pH units in 10 to 20 years. Lavery and Carson (1977) also suggested that acidification of agricultural soils could be attributed to SO<sub>2</sub> emissions from such plants. In contrast, Lore (1984) found no acidification trend in pH downwind of a plant in southern Alberta. Similarly, in the initial soil survey of the Strachan<sup>1</sup> and Ram River<sup>2</sup> plant areas (Addison et al. 1984) there were no reductions in soil pH that could be attributed to SO<sub>2</sub>. A depression in pH at the two sites within 200 m of the processing plants was the result of S<sup>0</sup> contamination and its subsequent oxidation (Addison et al. 1984). Legge et al. (1986) also found significant decreases in the pH of soils within 2.8 km of a plant in West Whitecourt. Initial studies on these soils (e.g., Legge et al. 1981; Bewley and Parkinson 1984) suggested that SO<sub>2</sub> was the major pollutant; however, no measure of S<sup>0</sup> concentration of the soil was determined. Subsequently, Legge and co-workers (Legge et al. 1986; Laishley and Bryant 1987; Legge and Crowther 1987) have indicated that S<sup>0</sup> was involved, at least since 1979. Contamination with S<sup>0</sup> prior to this date is virtually

certain, since approximately  $9 \times 10^6$  tonnes of S<sup>0</sup> were shipped from the site, much of it in a crushed bulk form<sup>3</sup>. At sites heavily contaminated by S<sup>0</sup> it is impossible to determine if exposure to S gases influenced the soil and vegetation chemistry because of the overwhelming influence of S<sup>0</sup> (Addison et al. 1984; Maynard and Addison 1987).

A 5-year study was initiated in 1981 to study the effects of sour gas processing on forest ecosystems in west-central Alberta. A biomonitoring network of 26 sites was established in a 30 × 30 km area surrounding the Strachan and Ram River sour gas processing plants. Results from the initial survey and characterization of the sites are synthesized in an earlier report (Addison et al. 1984). Three sites within 1 km of the gas plants had significantly higher S concentrations in the LFH horizon than the other sites. In addition, at the most heavily impacted site, site 15, elevated S concentrations were found at 22-34 cm depth. Lower, but not significant concentrations of manganese (Mn), potassium (K), magnesium (Mg), and aluminum (Al) in the LFH were also observed at site 15. Further, the plant response to S<sup>0</sup> concentrations >14 000 mg kg<sup>-1</sup> was limited to the understory vegetation in 1981; moss and herb cover were particularly sensitive. A growth or reproductive response in trees was not detected other than that there were elevated S concentrations in foliage. Three sites within 6 km of the gas plants had no measurable S<sup>0</sup> but had elevated S concentrations in foliage. Addison et al. (1984) were not able to determine whether higher S concentrations at these sites were pollutant related (i.e., either due to S<sup>0</sup> or exposure to S gases).

These same biomonitoring sites were reexamined in 1985 to determine if additional impact to the forest ecosystem had occurred in the vicinity of the sour gas processing plants in the 4 years since the initial survey. Reexamination of plots also provided additional information for assessing potential pollutant effects on the forest ecosystem.

<sup>1</sup> Gulf Canada Resources Limited owns and operates the Strachan plant.

<sup>2</sup> Husky Oil Operations Ltd. (formerly Canterra Energy Ltd.) owns and operates the Ram River plant.

<sup>3</sup> Personal communication from David McCoy of Husky Oil Operations Ltd. (formerly Canterra Energy Ltd.), Calgary, Alberta, June 1988.



## MATERIALS AND METHODS

### Site Establishment

Twenty-five of the original sites established in 1981 in an area  $30 \times 30$  km centered on the two sour gas processing plants (Fig. 1) were resampled in 1985. Detailed descriptions of the site establishment, soils, and vegetation were presented earlier (Addison et al. 1984). Site 15, the most heavily impacted site in 1981, was not resampled because it was cleared following an  $S^0$  block fire at the Ram River gas plant in 1984.

The sites were originally grouped into three categories: (1)  $S^0$  contaminated and with elevated sulfation, (2) elevated sulfation, (7-year sulfation rate  $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ ), and (3) background sites ( $<0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ ), (Addison et al. 1984)<sup>4</sup>. In 1985 the mean sulfation rates for 1981-85 (Fig. 2) showed that only the  $S^0$  contaminated sites (1, 2, 5, and 12) were in areas with a 5-year (1981-85) mean sulfation rate  $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ . Seven sites (3, 6, 10, 11, 22, 23, and 24) were within areas containing approximately  $0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  (based on an average from 1981 to 1985). The remaining sites had average sulfation rates of  $<0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ . The sites, therefore, were divided into three groups in 1985: (1)  $S^0$  contaminated (excluding site 3), (2) elevated sulfation ( $\geq 0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ ), and (3) background sites ( $<0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ )<sup>5</sup>.

### Elemental S Deposition

Ten replicates of a  $132\text{-cm}^2$  ( $12 \times 11$  cm) plastic container with terry towel fabric covers were placed at locations adjacent to sites 1 and 2 to determine the rate of  $S^0$  deposition. The containers were sunk into the soil with the toweling flush with the soil surface. The toweling was changed every 2 weeks, and the  $S^0$  deposits were determined using the method of Maynard and Addison (1985). The toweling was used to provide a rough surface to prevent the  $S^0$  from moving and also to provide a material that, when extracted, did not produce any interferences to the colorimetric determination of  $S^0$ .

### Soil and Plant Analysis

Soil and plant chemical analyses were completed on soil and plant material from the 25 sites that were still in place in 1985. Soil samples were collected in the same way as outlined by Addison et al. (1984). The five soil pits were extended, and the LFH and top three mineral horizons were sampled from the face of the pit. Samples were placed in plastic bags and stored in a frozen state.

Total soil analyses were done on air-dried subsamples ground to pass through a 60-mesh sieve. The samples were prepared in the same manner as used in 1981: digestion of soils using  $\text{HNO}_3\text{-HClO}_4\text{-HF-HCl}$  followed by analysis by inductively coupled plasma-atomic emission spectrometry (ICP-AES) (Addison et al. 1984; McQuaker et al. 1979).

Elemental S and pH were determined on field-moist samples, using the same method as in 1981. Extractable concentrations (cations and S) were also determined on field-moist LFH samples in 1985 (1.0 mol  $\text{NH}_4\text{Cl}$ , 1:20 moist soil to solution ratio). Since extractable S and cation concentrations in the LFH horizon followed a similar pattern to the total concentrations, only the totals are reported.

Two additional soil analyses, electrical conductivity (EC) and phosphate extractable sulfate, were done in 1985. Electrical conductivity was determined on all horizons (field-moist samples) using a 2:1 distilled water to soil ratio (volume/oven-dry weight basis) after the soil was brought to a saturated paste with distilled water (Maynard and Addison 1987). Mineral soils were extracted moist (1:20 moist soil to solution ratio) with  $500 \text{ mg L}^{-1}$  of P as  $\text{Ca}(\text{H}_2\text{PO}_4)_2$  to test for adsorbed  $\text{SO}_4^{2-}\text{-S}$ . Sulfate is not adsorbed by organic horizons; therefore, this analysis was not done on the LFH horizons (Maynard et al. 1987).

Ten replicate samples from lodgepole pine (*Pinus contorta* Dougl.), spruce (either *Picea glauca* (Moench) Voss or *P. mariana* (Mill.) B.S.P.), twin-flower (*Linnaea borealis* L.), and feather moss (*Pleurozium schreberi*) were collected in 1985. The

<sup>4</sup> Elemental S contaminated sites had measurable ( $>100 \text{ mg kg}^{-1}$ )  $S^0$  present. The sulfation isopleth of  $0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  is arbitrary and is 10% of the provincial exposure guideline.

<sup>5</sup> Elemental S contaminated sites had measurable ( $>100 \text{ mg kg}^{-1}$ )  $S^0$  present. The sulfation isopleth of  $0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  is arbitrary and is  $<10\%$  of the provincial exposure guideline.

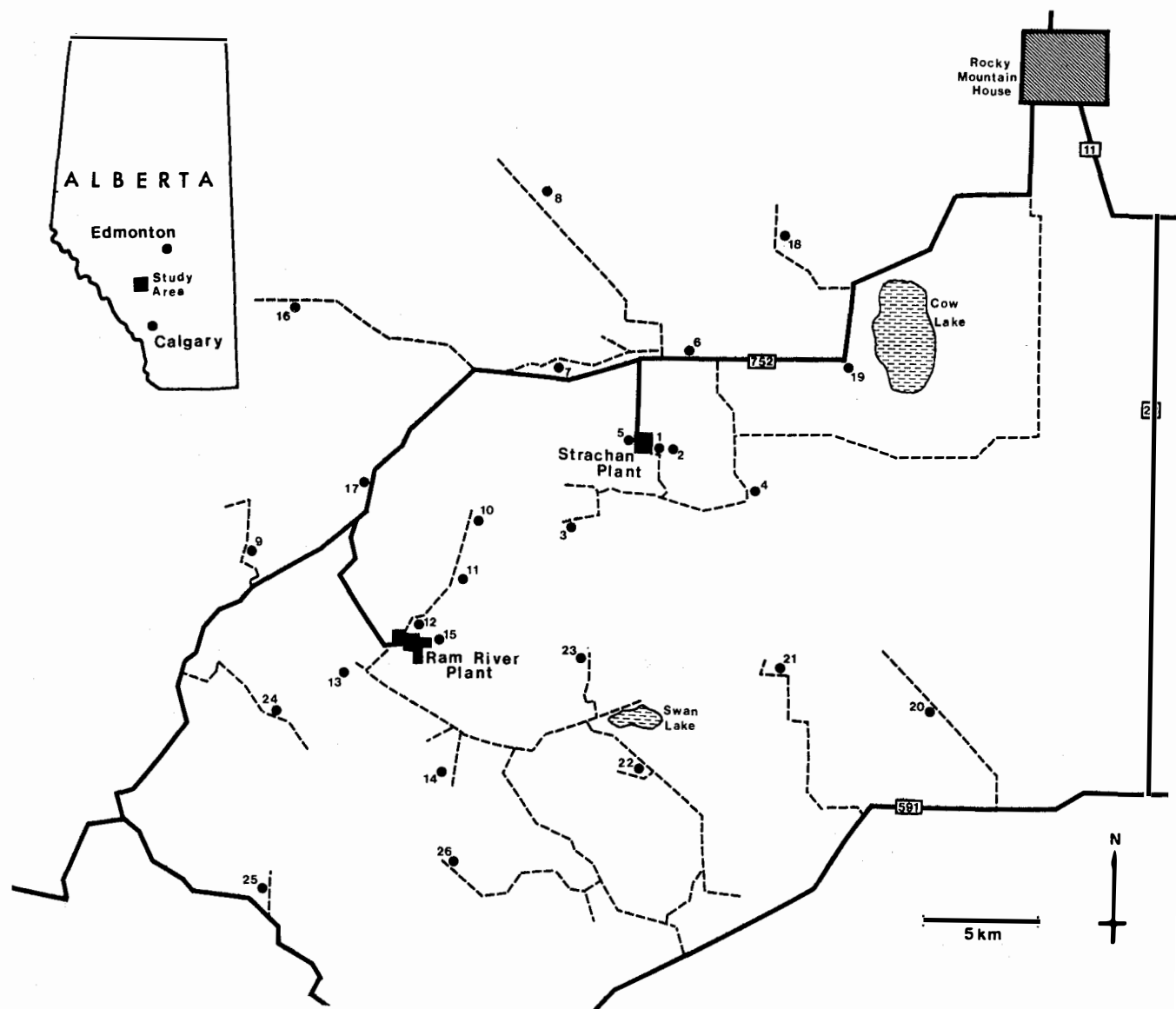
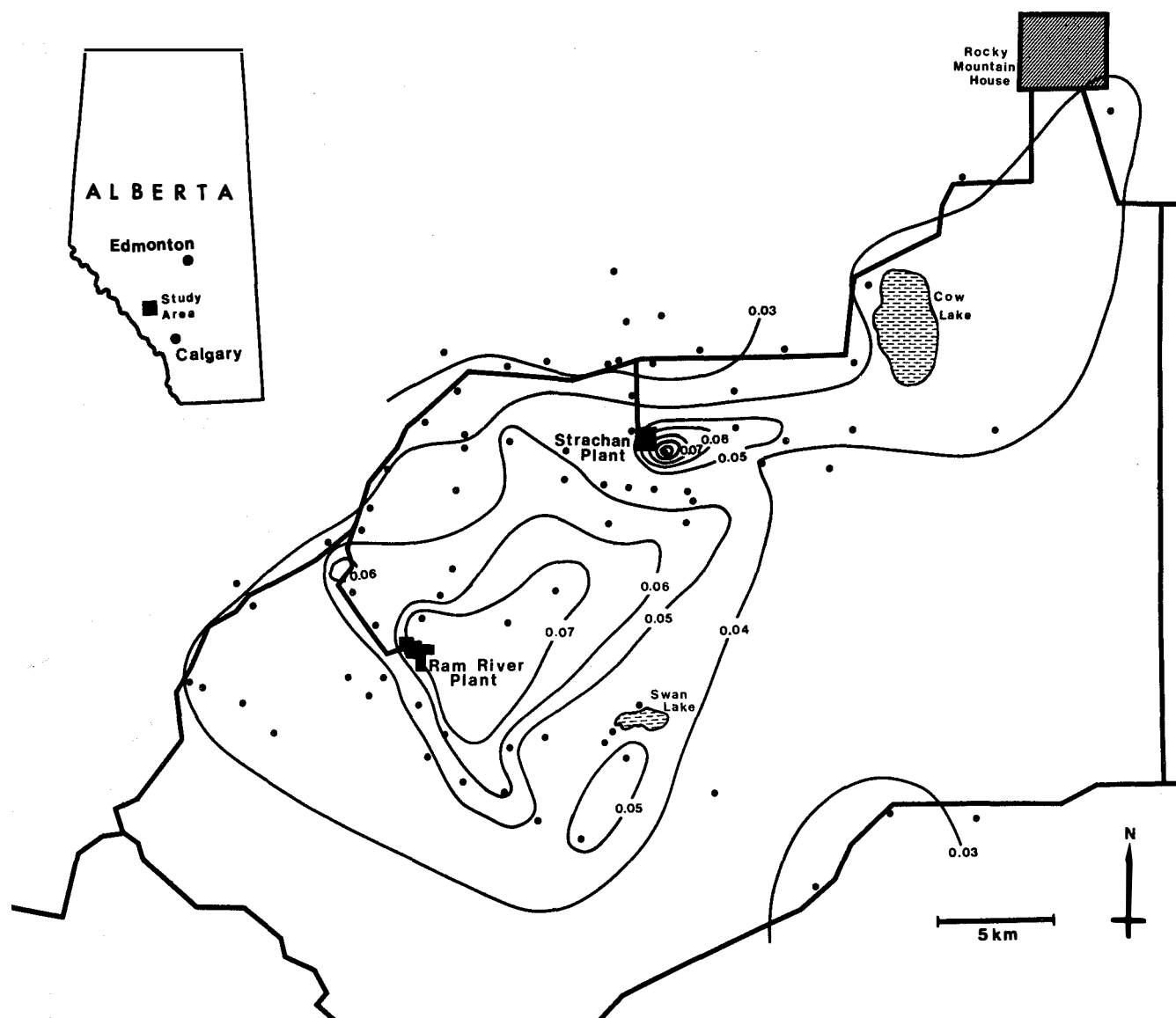


Figure 1. Location of biomonitoring plots and sour gas processing plants in west-central Alberta. (Source: Addison et al. 1984.)



**Figure 2. Isopleths of average sulfation rates in the vicinity of sour gas processing plants in west-central Alberta, 1975-81. (Source: Addison et al. 1984.)**

lodgepole pine samples were 1-year-old needles taken from the lower one-third of the crown taken at the edges of each plot. In 1981, the foliage was collected from the leaders of trees harvested for growth and reproduction measurements. Spruce foliage was collected from permanently marked trees, using the same method as in 1981. Twin-flower and feather moss were sampled using the same technique as in 1981, except sampling occurred in June rather than August.

Plant samples were analyzed as they were in 1981. The samples were oven-dried at 70°C, ground in a Wiley mill to pass through a 2-mm sieve, digested with  $\text{HNO}_3\text{-HClO}_4\text{-HCl}$ , and analyzed using ICP-AES analysis.

### **Vegetation Cover and Species Richness<sup>6</sup>**

Twenty randomly distributed  $1 \times 1$  m permanent quadrats were established within each of the  $20 \times 20$  m plots of all the sites in 1981. Plant cover estimates were remeasured at 23 sites in 1985. Repeat measurements of sites 3, 15, and 19 were not possible. Visual estimates of the percentage cover and enumeration of plant species were repeated by the same observer in both 1981 and 1985. Plant cover estimates were corrected for measurement errors (Kennedy and Addison 1987).

### **Tree Growth Measurements**

In 1985 only needle number, needle weight, stem length, and stem weight were determined. Other parameters sampled in 1981 (Addison et al. 1984) were not included because they were highly variable and showed no significant difference among sites.

### **Growth Chamber Incubation**

A growth chamber study was set up to determine the buffering capacity of the LFH with respect to  $\text{S}^0$  oxidation. Elemental S was applied at 1000, 10 000, and 50 000  $\text{mg kg}^{-1}$  of  $\text{S}^0$  to approximate  $\text{S}^0$  concentrations measured in LFH horizons at sites 5, 2, and 1, respectively, in 1981. The LFH horizon samples were incubated for 28 weeks. The samples were maintained at a constant temperature of 20°C and at a constant moisture content (80% of field capacity)<sup>7</sup>. Samples were taken weekly and  $\text{S}^0$  and pH were determined on moist subsamples as previously outlined.

### **Statistics**

The 1985 soils and plant chemistry data were analyzed using a one-way analysis of variance (ANOVA). Scheffé multiple contrasts for pairwise comparisons were calculated when a probability of significance at the 0.05 level existed. Comparisons between the 1981 and 1985 soil chemistry data were done using a *t*-test of the differences between two means, which is equivalent to an ANOVA for two groups (Sokal and Rohlf 1981).

Data were tested for homogeneity of variances by the Bartlett test for equality of variances prior to the one-way ANOVA (Neter and Wasserman 1974). Where necessary, data were mathematically transformed prior to analysis. For the data that could not be normalized by transformations, a one-way ANOVA was applied to the ranks followed by Scheffé multiple contrasts ( $p < 0.05$ ).

<sup>6</sup> Species richness is the number of species in a given area.

<sup>7</sup> Field capacity is the percentage of water remaining in the soil 2 or 3 days after the soil has been saturated and free drainage has practically ceased.

## RESULTS AND DISCUSSION

### Pollution Distribution

#### Sulfation

Average amounts of sulfur gases ( $\text{H}_2\text{S}$  and  $\text{SO}_2$ ) as measured by Husky Oil Operations Ltd. (formerly Canterra Energy Ltd.) and Gulf Canada Resources Limited sulfation candles from 1975 to 1981 (Fig. 3; Addison et al. 1984) were compared to 1981-85 (Fig. 2), and showed very similar patterns. The isopleths were not directly comparable in all cases, particularly in the areas with  $<0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  because of fewer sites. In general, the average sulfation in areas of elevated sulfation rates ( $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ ) was about 30% ( $0.02 \text{ mg dm}^{-2} \text{ d}^{-1}$ ) lower between 1981 and 1985 compared to the 7-year average prior to 1981. This corresponded to a 30% drop in  $\text{SO}_2$  emissions from the two sour gas plants over the same time period (Fig. 4). In 1981, the four  $\text{S}^0$  contaminated sites (1, 2, 5, and 15) and six additional sites (3, 11, 12, 13, 14, and 23) were considered to have elevated sulfation rates ( $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$ , based on the 7-year average from 1975 to 1981; Fig. 3, Addison et al. 1984). Using the same criteria (from 1981 to 1985,  $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$ ), only the  $\text{S}^0$  contaminated sites (sites 1, 2, 5, and 12) fell within areas of elevated sulfation rates. Of the six sites considered to be in areas of an elevated sulfation rate in 1981, three (sites 3, 11, and 23) were within areas with an average sulfation rate of approximately  $0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  for 1981-85. The other three sites were within areas with an average sulfation rate  $<0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  for 1981-85 (Fig. 2). The 0.06 isopleth surrounding the Ram River gas plant in 1981 decreased in area, and the 1985 averages indicate that the 0.05 isopleth is in approximately the same location as the 0.06 isopleth was in 1981. The area of the 0.05 isopleth east of the Strachan sour gas plant extended further east during the 1981-85 period, compared to the average sulfation data prior to 1981 (1975-81) but did not encompass any of the 25 sites.

In areas outside those with elevated sulfation rates, sulfation data for the two time periods were similar. For example, the average sulfation at the gas companies' sulfation candles near sites 6 and 7 (north

of Highway 752, Fig. 1) was approximately  $0.03 \text{ mg dm}^{-2} \text{ d}^{-1}$  for both time periods.

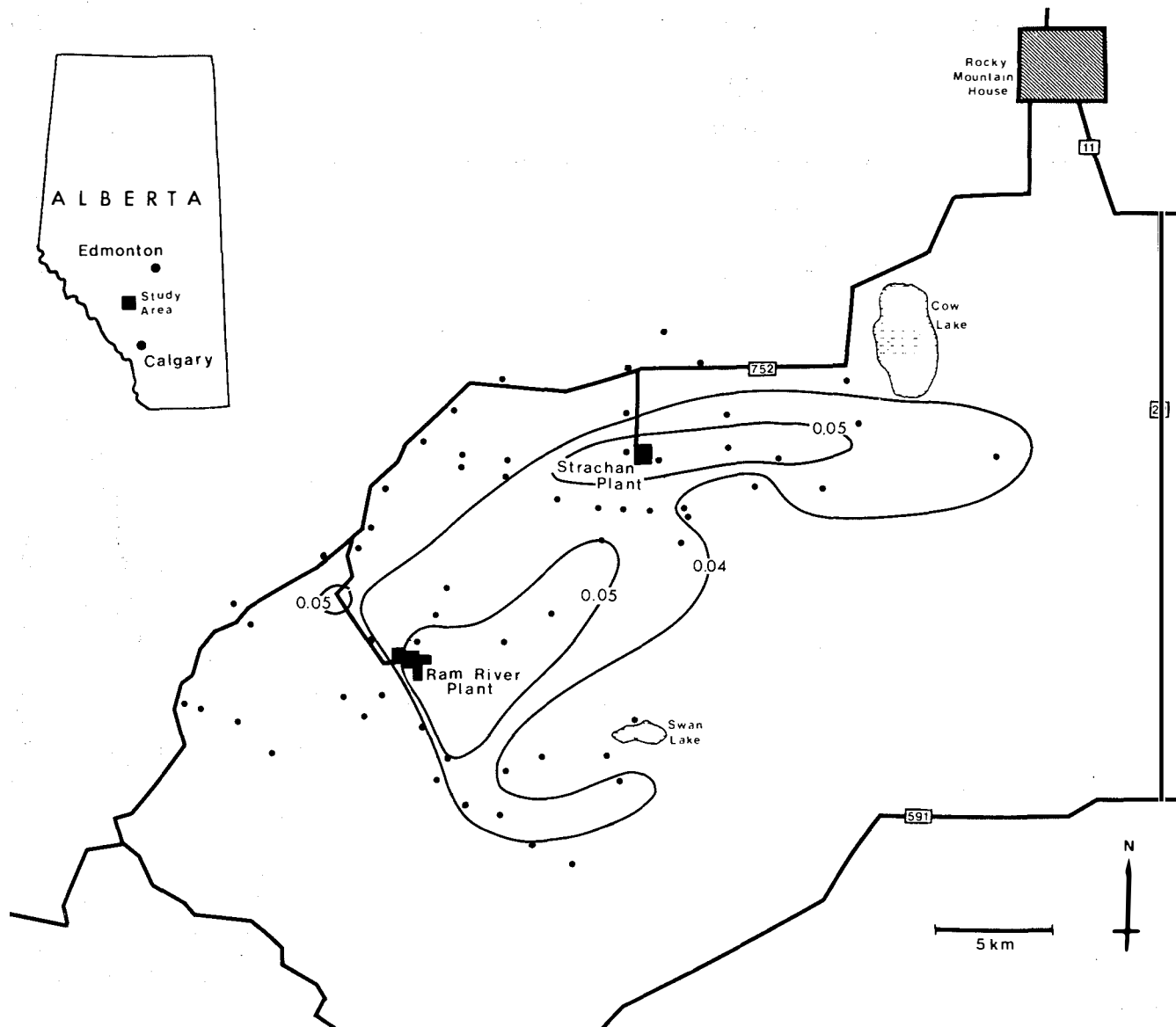
#### Elemental S Particulate

There appeared to be a greater measurable dispersion of  $\text{S}^0$  in 1985 compared to in the initial sampling in 1981. Detectable  $\text{S}^0$  concentrations in the LFH were observed at three sites that had measurable  $\text{S}^0$  concentrations in 1981 (sites 1, 2, and 5) and at two additional sites (4 and 12) that had no detectable  $\text{S}^0$  in 1981 (Table 1). Site 15, the most heavily impacted site at the time of the initial survey in 1981, was not included in 1985 because the site had been cleared following an  $\text{S}^0$  block fire at Ram River in May 1984. In 1981 elevated S concentrations were observed in the foliar analyses, but no measurable  $\text{S}^0$  was found in the LFH at site 12, which is within 500 m of the Ram River plant.

Site 4 had detectable  $\text{S}^0$  concentrations (detection limit is  $100 \text{ mg kg}^{-1}$ , which is approximately  $9 \text{ kg ha}^{-1}$  of  $\text{S}^0$ )<sup>8</sup> in the LFH horizon in 1985. Elemental S blown from trains transporting  $\text{S}^0$  from both gas plants was the  $\text{S}^0$  source at site 4, which is located  $>2 \text{ km}$  from the plants but is within 300 m of the train tracks. Site 3 also had detectable  $\text{S}^0$  concentrations in the LFH horizon in 1985 ( $128 \pm 50 \text{ mg kg}^{-1}$ ); however, there was no obvious mechanism for transport of  $\text{S}^0$  to this site. Site 3 was not considered as an  $\text{S}^0$  contaminated site in the Results and Discussion section of this report because there were no measurable effects on the other S parameters and the  $\text{S}^0$  concentration was at the detection limit.

Results of the 1985 soil sampling programs indicated that the amount of  $\text{S}^0$  deposited outside the companies' leases was similar to previous years but the distribution pattern had changed (Hardy Associates (1978) Ltd. 1986; Pedology Consultants Ltd. 1986). Results of the soil sampling program around the Ram River plant indicated that increased  $\text{S}^0$  deposition had occurred around the  $\text{S}^0$  handling and prilling tower (Hardy Associates (1978) Ltd. 1986). No major changes were observed in the total soil S concentrations in other areas. A shift in the  $\text{S}^0$

<sup>8</sup> The conversion of S concentrations (milligrams per kilogram) in the LFH horizon to a kilogram per hectare basis was calculated assuming a bulk density of  $0.15 \text{ g cm}^{-3}$  and a depth of 6.0 cm. Bulk density and depth values were the average of 50 samples from a non- $\text{S}^0$  contaminated site within the study area.



**Figure 3. Isopleths of average sulfation rates in the vicinity of sour gas processing plants in west-central Alberta, 1981-85.**

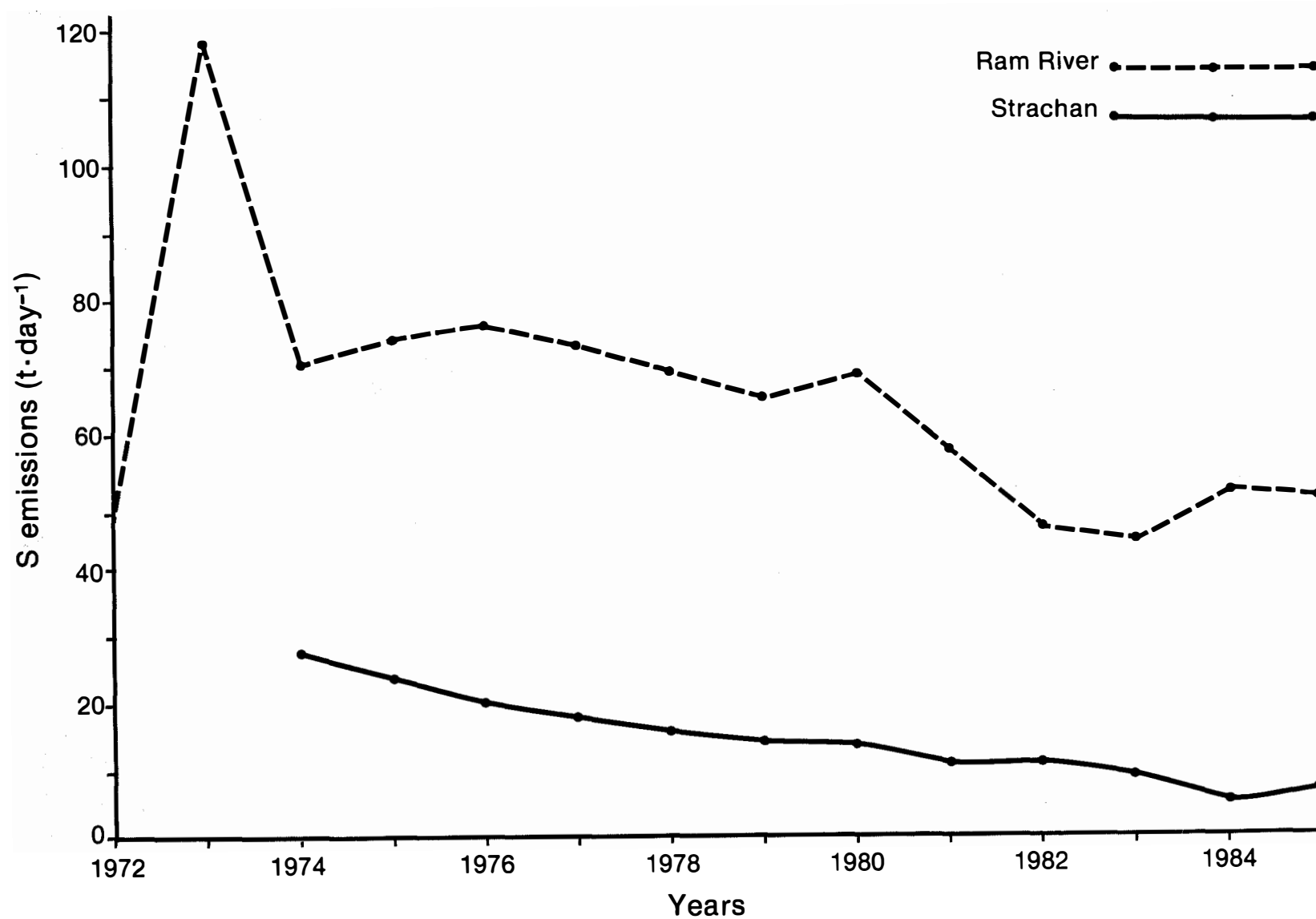


Figure 4. Sulfur dioxide emissions from incinerator stacks of Ram River and Strachan sour gas processing plants (1972-85).

**Table 1. Elemental sulfur content of the LFH horizon at selected sites in 1981 and 1985.** Values are means  $\pm$  95% confidence limits.

Site <sup>a</sup>	Elemental sulfur (mg kg <sup>-1</sup> )	
	1981	1985
1	43 500 $\pm$ 12 700	51 300 $\pm$ 19 100
2	14 600 $\pm$ 14 300	7 860 $\pm$ 7 440
5	581 $\pm$ 452	3 360 $\pm$ 1 350
12	B.D. <sup>b</sup>	361 $\pm$ 55
3	B.D.	128 $\pm$ 50
4	B.D.	155 $\pm$ 35

<sup>a</sup> All other sites had elemental sulfur concentrations below the detection limit of the method (100 mg kg<sup>-1</sup> soil).

<sup>b</sup> B.D. = below detection limit of the method. For litter material the detection limit of S<sup>0</sup> is 100 mg kg<sup>-1</sup> soil.

deposition pattern at the Strachan plant could also have resulted in increased S<sup>0</sup> deposition occurring west of the plant (Pedology Consultants Ltd. 1986). Significant increases in the S<sup>0</sup> concentration of the LFH horizon at site 5 from 1981 to 1985 support this (Table 1).

The heaviest S<sup>0</sup> contamination was observed at sites 1 and 2, with the majority of this S<sup>0</sup> deposited during 1979 and 1980 (the Strachan plant began shipping S<sup>0</sup> in 1979; Fig. 5). The S<sup>0</sup> deposited between June and October 1982 and April and October 1983 accounted for approximately 10% of the total S<sup>0</sup> present in the LFH (Table 2). Elemental S deposition was, therefore, still occurring, but at a much lower rate than previously recorded.

The heavy deposition in the 2 years prior to 1981 coincided with mechanical breakup of the S<sup>0</sup> blocks. Since that time S<sup>0</sup> has been removed by other processes such as Ellithorpe remelters (in addition to mechanical breakup) that produce less S<sup>0</sup> dust. Amounts of S<sup>0</sup> shipped were lower in 1982 and 1983 compared to 1981 (Fig. 5). The lower rate of S<sup>0</sup> deposition observed in 1982 and 1983, therefore, was probably a combination of the improved S<sup>0</sup> handling procedures and a reduction in the amount of S<sup>0</sup> shipped. Based on these data, S<sup>0</sup> deposition rates should not be extrapolated beyond the time of observation because S<sup>0</sup> deposition depends upon many factors, particularly on the amount of S<sup>0</sup> being shipped and block recovery techniques.

### Liming

Sites 1, 2, and 5 were limed (CaCO<sub>3</sub> added) in 1979 and 1980 prior to the initial sampling (Addison et al. 1984). From 1981 to 1984 these areas were excluded from the regular liming programs carried out by the companies for the purposes of this study. In June 1984 and the spring of 1985, however, all or portions of sites 1, 2, and 5 as well as site 12 were limed. Site 12 was limed in the spring of 1985 with 5 t ha<sup>-1</sup> (Hardy Associates (1978) Ltd. 1984). Sites 1 and 2 were limed with between 1600 and 2100 kg ha<sup>-1</sup> (0.9-1.2 tons per acre; Pedology Consultants 1983). Site 5 was limed with between 500 and 1100 kg ha<sup>-1</sup> (0.3-0.6 tons per acre; Pedology Consultants 1983).

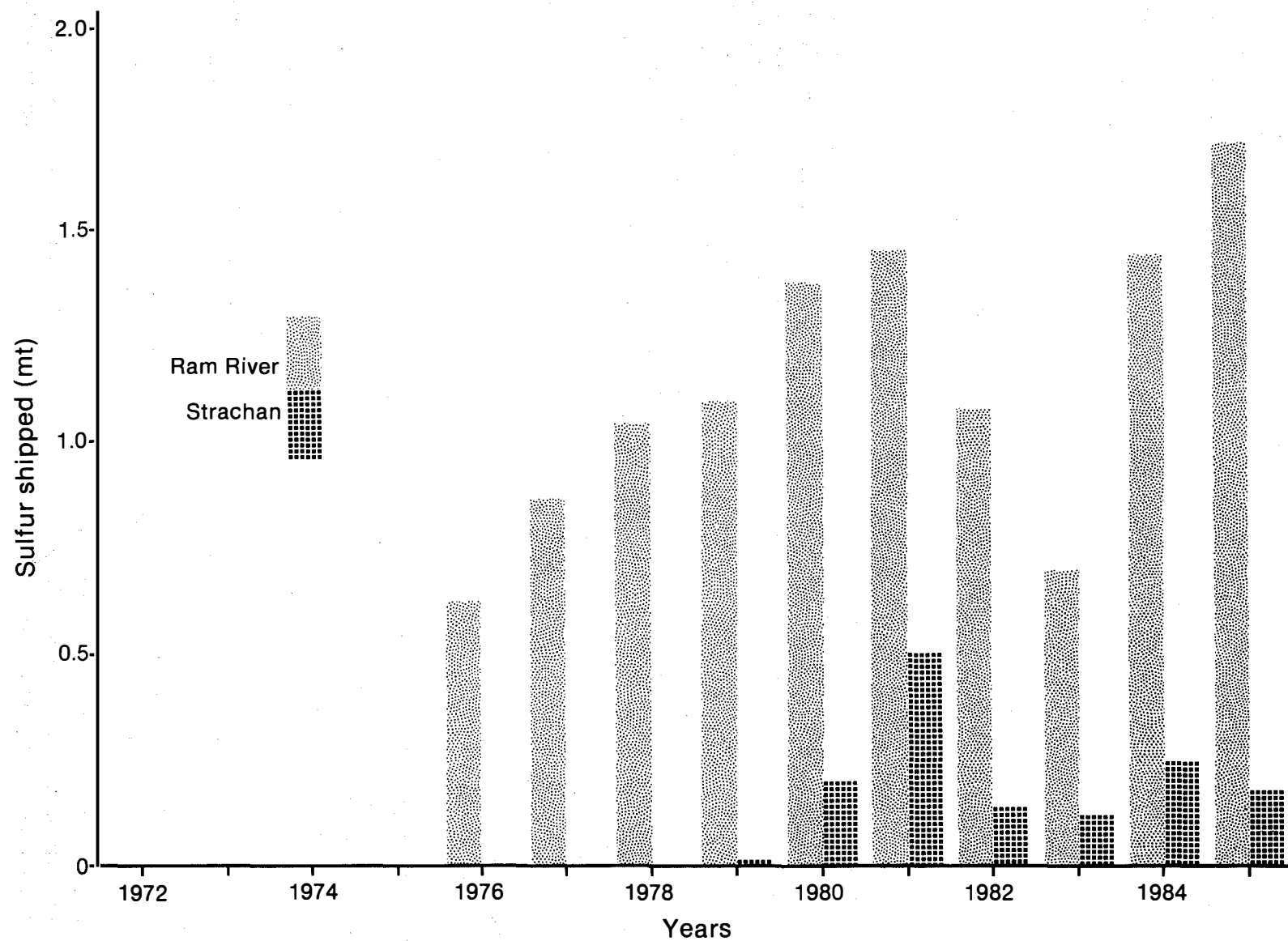
### Soil Chemical Analyses

#### LFH Horizon

Total S in the LFH horizon at S<sup>0</sup> contaminated sites (Table 3) was similar to that recorded in 1981: site 1 is greater than site 2, which is greater than site 5, which is the same as site 12 (significant at  $p < 0.05$ ), and all but site 12 had S concentrations significantly higher ( $p < 0.05$ ) than the non-S<sup>0</sup> contaminated sites. These four sites are within 500 m of the gas plants. Site 4 had measurable S<sup>0</sup> concentrations in 1985 in the LFH (Table 1); however, it was not reflected in the total S concentrations (Table 4).

Sites 1 and 2 were the most heavily S<sup>0</sup> contaminated and impacted sites in 1985 (Tables 1 and 3).





**Figure 5. Formed sulfur shipped from Ram River and Strachan sour gas processing plants (1976-85).**

**Table 2. Deposition of elemental sulfur ( $S^0$ ) and leaching of sulfur (S) adjacent to sites 1 and 2 in relation to the total S,  $S^0$ , and  $SO_4^{2-}$ -S concentrations of the LFH horizon from 1982 to 1984 ( $kg\ ha^{-1}$ )**

Dates	Site 1					Site 2				
	Deposition of $S^0$	Leaching <sup>a</sup> of S	S concentrations <sup>b</sup>			Deposition of $S^0$	Leaching of S	S concentrations <sup>c</sup>		
			Total	$S^0$	$SO_4^{2-}$ -S			Total	$S^0$	$SO_4^{2-}$ -S
08/81	ND <sup>d</sup>	ND	3160 ± 562	2350 ± 552	643 ± 323	ND	ND	887 ± 517	809 ± 639	115 ± 52
15/06/82 to 14/10/82	123	427	2860 ± 2170	2720 ± 2110	80 ± 60	26	168	787 ± 480	760 ± 517	27 ± 9
25/11/82 to 19/04/83	ND	105	3660 ± 1250	3490 ± 1320	44 ± 32	ND	17	565 ± 252	386 ± 210	29 ± 21
19/04/83 to 06/10/83	263	35	2770 ± 1860	2820 ± 2050	71 ± 45	70	24	665 ± 222	437 ± 158	66 ± 63
06/10/83 to 08/05/84	ND	48	3480 ± 1110	3350 ± 815	41 ± 15	ND	11	465 ± 317	444 ± 326	24 ± 13
08/05/84 to 07/08/84	ND	13	1750 ± 653	2160 ± 886	57 ± 17	ND	8	434 ± 207	330 ± 195	40 ± 26
07/85	ND	ND	2592 ± 538	2770 ± 831	234 ± 41	ND	ND	610 ± 257	436 ± 332	64 ± 12

<sup>a</sup> Total S in the leachate from zero tension lysimeters below the LFH horizon. There were five lysimeters per site, and the bottles were changed every 2 weeks.

<sup>b</sup> Sulfur concentrations at site 1 in kilograms per hectare (mean ± standard deviation) are calculated using the S values in milligrams per kilogram ( $n = 10$ ), a bulk density of  $0.135\ g\ cm^{-3}$ , and a depth of 4.0 cm. Bulk density and depth were determined on 50 samples collected at site 1.

<sup>c</sup> Sulfur concentrations at site 2 in kilograms per hectare (mean ± standard deviation) are calculated using the S values in milligrams per kilogram ( $n = 10$ ), a bulk density of  $0.126\ g\ cm^{-3}$ , and a depth of 4.4 cm. Bulk density and depth were determined on 50 samples collected at site 2.

<sup>d</sup> ND = not determined.

**Table 3. Total sulfur concentrations (mg kg<sup>-1</sup>) of the LFH soil horizon at selected sites in 1981 and 1985. Values are means  $\pm$  95% confidence limits (C.L.).**

Site	Total sulfur concentration (mg kg <sup>-1</sup> )		Between years <sup>a</sup>
	1981	1985	
S <sup>0</sup> contaminated			
1	58 600. ± 12 900	48 000 ± 12 400	NS
2	16 000 ± 11 600	11 000 ± 5 750	NS
4	659 ± 285	643 ± 214	NS
5	2 340 ± 624	3 810 ± 1 200	*
12	1 010 ± 366	1 440 ± 243	*
Elevated sulfation rates <sup>b</sup>			
3	1 150 ± 240	945 ± 195	NS
6	790 ± 237	643 ± 266	NS
10	1 190 ± 124	929 ± 191	*
11	1 000 ± 133	1 150 ± 353	NS
13	919 ± 503	1 040 ± 174	NS
14	1 080 ± 390	1 040 ± 184	NS
22	1 160 ± 168	876 ± 323	NS
23	1 270 ± 238	952 ± 112	*
24	1 220 ± 134	1 080 ± 114	NS
All non-S <sup>0</sup> contaminated sites <sup>c</sup>			
Mean	1 060	915	
± 95% C.L.	± 68	± 67	
Range	659 - 1 260	643 - 1 150	

<sup>a</sup> Data analyzed by *t*-test for paired comparisons between years. NS = not significant, and \* = *p* < 0.05.

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rates >0.04 mg dm<sup>-2</sup> h<sup>-1</sup> of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

There were no significant differences (*p*  $\geq$  0.05) in S<sup>0</sup> and total S concentrations of the LFH horizon at sites 1 and 2 between 1981 and 1985. The lack of significant changes in S concentrations between years at sites 1 and 2 was attributed primarily to the large variability in the S concentrations.

Given the large variability and overwhelming influence of the S<sup>0</sup> it was only possible to estimate changes in S<sup>0</sup> deposition and oxidation rates and losses of S from the LFH horizon at sites 1 and 2 (Table 2). In 1982 leaching losses greatly exceeded deposition. In 1983 the opposite was true. Deposition would also have occurred throughout the year, whereas leaching losses are largely limited to the growing season months. These estimates do, however, indicate that the deposition of S<sup>0</sup> that occurred in 1982 and 1983 was at a rate that would not have resulted in a continual buildup of S in the LFH horizons. This is supported by the total S and S<sup>0</sup>

concentration data from sites 1 and 2. At site 2 it appeared that losses from the LFH horizon may have exceeded S<sup>0</sup> deposition between 1981 and 1985.

Site 5, on the opposite side of the Strachan plant, had a significant increase in both S<sup>0</sup> and total S concentrations in 1985 compared to 1981. In 1985, the S<sup>0</sup> concentration had increased to 3 360 mg kg<sup>-1</sup> (approximately 300 kg ha<sup>-1</sup>), with a significant increase in the total S concentration. The contrary pattern of S<sup>0</sup> deposition between sites 1 and 2 and site 5 indicates the variable distribution of S<sup>0</sup> deposition.

Detectable amounts of S<sup>0</sup> were observed in the LFH at site 12 in 1985. No detectable S<sup>0</sup> in the LFH was found in 1981. The S<sup>0</sup> concentrations in 1985 were reflected in increased total S concentrations (significant at *p* < 0.05). Elevated S concentrations in the vegetation were observed in 1981, but it was

not possible to determine the source of S at that time (Addison et al. 1984). The presence of S<sup>0</sup> at this site in 1985 indicates that S<sup>0</sup> was probably the main pollutant.

The total S concentrations in the LFH at sites other than the S<sup>0</sup> contaminated sites did not change significantly ( $p \geq 0.05$ ) between years with few exceptions (Table 3). At the sites where S concentrations did change significantly, the S content decreased, but it did not fall below the lower range of S found in either 1981 or 1985. The changes observed at these sites are probably related to the temporal and spatial variability of S concentrations in the LFH. The

mean and range of total S concentrations in the LFH horizons of the non-S<sup>0</sup> contaminated sites were similar in 1981 and 1985.

In 1981, only the LFH at sites 1 and 15 had decreases in pH relative to all other sites as a result of S<sup>0</sup> contamination (Addison et al. 1984). The pH at the S<sup>0</sup> contaminated sites did not change significantly ( $p \geq 0.05$ ) between 1981 and 1985 (Table 4). The pH of the LFH at site 1 remained the lowest and was significantly lower ( $p < 0.05$ ) than all but four sites (including site 2). The addition of lime (CaCO<sub>3</sub>) in 1984 did not increase the pH of the LFH at sites 1, 2, or 5; however, the lime may have neutralized any

**Table 4. The pH in 1981 and 1985 and the electrical conductivity (EC) in 1985 of the LFH soil horizon at selected sites.** Values are means  $\pm$  95% confidence limits.

Site	pH				Between years <sup>a</sup>	EC (dS m <sup>-1</sup> )
	1981		1985			
	Mean	Range	Mean	Range		
S <sup>0</sup> contaminated						
1	2.5	2.4-2.7	2.5	2.2-3.1	NS	4.68 ± 1.74
2	4.3	2.9-5.1	3.9	2.8-5.6	NS	1.71 ± 0.61
4	4.7	4.4-5.0	5.2	5.0-5.8	*	0.38 ± 0.22
5	4.9	4.4-6.5	5.2	3.8-6.2	NS	0.75 ± 0.16
12	4.5	3.8-5.3	5.4	5.0-5.8	*	0.59 ± 0.24
Elevated sulfation rates <sup>b</sup>						
3	4.6	3.9-5.1	4.5	4.2-4.8	NS	0.49 ± 0.15
6	5.5	5.3-5.8	5.0	4.2-5.6	NS	0.47 ± 0.14
10	4.2	3.9-4.4	3.9	3.7-4.1	*	0.35 ± 0.13
11	4.4	4.1-4.5	4.4	4.0-4.8	NS	0.44 ± 0.11
13	4.5	4.2-4.9	4.4	4.1-4.7	NS	0.42 ± 0.07
14	4.4	4.1-4.7	4.6	4.3-4.8	NS	0.57 ± 0.13
22	4.4	4.0-4.8	4.3	4.0-4.9	NS	0.58 ± 0.24
23	4.2	4.0-4.4	4.2	4.0-4.5	NS	0.59 ± 0.19
24	4.2	4.0-4.4	4.0	3.9-4.1	*	0.66 ± 0.28
All non-S <sup>0</sup> contaminated sites <sup>c</sup>						
Range		4.2-5.5		3.9-5.2		0.34 - 0.66
Background site						
9	4.5	4.3-4.7	4.1	4.0-4.2	**	0.38 ± 0.18

<sup>a</sup> Data analyzed by *t*-test for paired comparisons between years. NS = not significant, \* =  $p < 0.05$ , and \*\* =  $p < 0.01$ .

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rates  $> 0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

acid produced by the oxidation of  $S^0$ . Liming was not done at these sites between 1980 and 1984 for the purposes of this study. Lime significantly increased ( $p < 0.05$ ) the pH of the LFH at site 12 in 1985, although the pH was not significantly higher ( $p \geq 0.05$ ) than at any other site.

The buffering capacity of the LFH could also have neutralized a portion of the acid produced from  $S^0$  oxidation. A growth chamber study was set up to determine the ability of the LFH to buffer acid produced by  $S^0$  oxidation. The LFH samples containing  $1\,000\text{ mg kg}^{-1}$  of  $S^0$  oxidized all the  $S^0$  within 12 weeks with no change in pH (pH 4.1). Samples containing  $10\,000\text{ mg kg}^{-1}$  of  $S^0$  decreased in pH from 4.1 to 2.0, with approximately 90% of the  $S^0$  oxidized after 28 weeks. At  $50\,000\text{ mg kg}^{-1}$  of  $S^0$ ,  $20\,000\text{ mg kg}^{-1}$  of  $S^0$  remained after 28 weeks, and the pH was reduced to 1.4. In relating the incubation to field conditions, deposition and oxidation of  $S^0$  would have to exceed  $1000\text{ mg kg}^{-1}$  before a reduction in LFH pH would be expected. Deposition of  $10\,000\text{ mg kg}^{-1}$  of  $S^0$  may or may not reduce LFH pH to 2.0 under field conditions because the growth chamber study was done under optimum moisture conditions (optimizing oxidation rates) and this would not be true for field conditions. At site 12, therefore, no decrease in pH would have been expected in 1985 even if the site had not been limed. It is more difficult to assess what may have happened at sites 2 and 5 because  $S^0$  has been present in the LFH horizons since at least 1981. Had these sites, particularly site 2, not been limed in 1979 and again in 1984, the pH would probably have decreased, given the amount of  $S^0$  in the LFH and the length of time since initial deposition.

The pH of the LFH horizons did not change significantly ( $p \geq 0.05$ ) between 1981 and 1985 at 17 of the remaining 21 sites (Table 4). At four sites (4, 9, 10, and 24) the pH did change significantly ( $p < 0.05$ ) but by  $<0.5$  pH unit and was related to natural soil variability. Palmer et al. (1985) found that for pH differences must be greater than 0.5 of a unit to be considered above the natural spatial and temporal variability found in forest soils.

Sites 1 and 2 had significantly higher EC than at any other site ( $p < 0.05$ , Table 4). The EC in the LFH at site 1 was above the salinity range known to restrict the yields of many agricultural crops (Mengel and Kirkby 1979). At site 2 the EC would only have a negative effect on the most saline sensitive plants. The EC of the remaining sites was  $<0.75\text{ dS m}^{-1}$  and

would not affect plant growth. The EC at sites 1 and 2 was a function of the low pH ( $\text{H}_3\text{O}^+$  and  $\text{SO}_4^{2-}\text{-S}$ ) as a result of  $S^0$  oxidation and increased salt (mainly Ca) concentrations through increased solubility at low pH and input to the system by liming.

Total cations in the LFH are given in Table 5. In 1985, Ca concentrations in the LFH were the highest at the limed sites 1, 2, 5, and 12. Many of the differences among sites and between years were not significant ( $p \geq 0.05$ ) because of the large variability in Ca associated with aerial liming. At site 1, the Ca added by liming could have replaced Ca lost by leaching due to the acidification of the LFH horizon. An intensive sampling program was implemented adjacent to sites 1 and 2 between April 1982 and July 1984. Between April 1982 and May 1984 the total Ca concentration adjacent to site 1 decreased significantly ( $p < 0.05$ ) from  $3660$  to  $1120\text{ mg kg}^{-1}$ . In July 1984, after the lime application, the total Ca concentration had increased to  $5910\text{ mg kg}^{-1}$ . The concentrations are not directly comparable to the site 1 concentrations since the sampling occurred adjacent to the site. They do, however, indicate a significant decrease in Ca concentrations in the LFH between 1982 and prior to the lime application in 1984. Adjacent to site 2, there were no significant decreases in the Ca concentrations over the same time period. Therefore, leaching of Ca from the LFH as a result of acidification of this horizon appeared to be limited to site 1.

There were no differences in the total Mg concentration of the LFH among sites in 1985, and there were very few changes in the total Mg concentrations of the LFH between years at any site (Table 5). At the intensive sampling location adjacent to site 1 total Mg concentrations did not decrease between 1982 and 1984. There was, however, an increase in the total Mg concentration (similar to Ca) between May 1984 and July 1984, suggesting the lime source used in 1984 at site 1 contained some Mg. No increases were observed in the total Mg concentrations of the LFH at any other limed site.

Total K concentrations in the LFH horizon at sites 1 and 2 were considerably lower than at any other sites (Table 5). In addition, sites 1 and 2 were the only sites that had significant decreases ( $p < 0.05$ ) in total K between 1981 and 1985. There was no significant decrease ( $p \geq 0.05$ ) of K at either sites 5 or 12 between 1981 and 1985.

The effect of soil acidification on the loss of bases (e.g., Ca, Mg, and K) is well-documented

**Table 5. Total concentrations (mg kg<sup>-1</sup>) of the basic cations (Ca, Mg, and K) in the LFH horizon at selected sites in 1981 and 1985. Values are means  $\pm$  95% confidence limits (C.L.).**

Site	Ca			Mg			K		
	1981	1985	Between years <sup>a</sup>	1981	1985	Between years	1981	1985	Between years
<b>S<sup>0</sup> contaminated</b>									
1	11 700 $\pm$ 6 350	9 340 $\pm$ 9 900	NS	735 $\pm$ 443	1 190 $\pm$ 1 090	NS	1 880 $\pm$ 624	973 $\pm$ 329	**
2	12 900 $\pm$ 14 900	16 800 $\pm$ 26 600	NS	857 $\pm$ 435	1 450 $\pm$ 1 940	NS	1 880 $\pm$ 596	1 010 $\pm$ 567	*
4	3 720 $\pm$ 1 200	4 710 $\pm$ 2 810	NS	1 290 $\pm$ 410	1 630 $\pm$ 435	NS	2 050 $\pm$ 545	2 260 $\pm$ 496	NS
5	12 700 $\pm$ 12 900	17 800 $\pm$ 10 200	NS	958 $\pm$ 468	948 $\pm$ 451	NS	1 700 $\pm$ 628	1 580 $\pm$ 641	NS
12	4 460 $\pm$ 2 410	34 000 $\pm$ 17 800	***	1 110 $\pm$ 524	1 140 $\pm$ 1 050	NS	2 090 $\pm$ 925	1 870 $\pm$ 1 130	NS
<b>Elevated sulfation rates<sup>b</sup></b>									
3	5 040 $\pm$ 2 000	4 390 $\pm$ 1 290	NS	1 250 $\pm$ 367	1 150 $\pm$ 214	NS	2 030 $\pm$ 475	2 000 $\pm$ 226	NS
6	6 970 $\pm$ 2 660	5 560 $\pm$ 1 460	NS	2 030 $\pm$ 287	1 530 $\pm$ 618	NS	2 700 $\pm$ 233	2 140 $\pm$ 729	NS
10	3 120 $\pm$ 813	3 040 $\pm$ 505	NS	754 $\pm$ 74	634 $\pm$ 272	NS	1 670 $\pm$ 372	1 510 $\pm$ 606	NS
11	4 000 $\pm$ 1 470	3 370 $\pm$ 1 580	NS	1 070 $\pm$ 116	1 110 $\pm$ 297	NS	2 070 $\pm$ 172	2 030 $\pm$ 162	NS
13	4 640 $\pm$ 1 870	4 840 $\pm$ 1 570	NS	918 $\pm$ 140	1 380 $\pm$ 670	NS	1 700 $\pm$ 365	2 130 $\pm$ 818	NS
14	4 840 $\pm$ 1 480	4 400 $\pm$ 1 770	NS	1 100 $\pm$ 643	1 120 $\pm$ 503	NS	1 800 $\pm$ 874	1 570 $\pm$ 426	NS
22	4 360 $\pm$ 2 160	3 800 $\pm$ 1 790	NS	950 $\pm$ 290	1 200 $\pm$ 358	NS	1 800 $\pm$ 613	1 910 $\pm$ 443	NS
23	3 150 $\pm$ 964	2 810 $\pm$ 702	NS	798 $\pm$ 297	866 $\pm$ 266	NS	1 830 $\pm$ 737	1 540 $\pm$ 562	NS
24	3 700 $\pm$ 1 040	3 330 $\pm$ 523	NS	824 $\pm$ 132	663 $\pm$ 41	**	2 100 $\pm$ 441	1 840 $\pm$ 621	NS
<b>All non-S<sup>0</sup> contaminated sites<sup>c</sup></b>									
Mean	5 050	4 540		1 190	1 160		2 160	1 950	
95% C.L.	$\pm$ 661	$\pm$ 582		$\pm$ 142	$\pm$ 153		$\pm$ 155	$\pm$ 177	
Range	3 120 - 8 370	2 810 - 7 060		754 - 2 030	634 - 1 940		1 670 - 3 050	1 380 - 3 080	

<sup>a</sup> Data analyzed by *t*-test between years. NS = not significant, \* = *p* < 0.05, \*\* = *p* < 0.01, and \*\*\* = *p* < 0.001.

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rate > 0.04 mg dm<sup>-2</sup> d<sup>-1</sup> of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

(Johnson and Cole 1977; McCoy and Webster 1977; Ulrich 1980; Mollitor and Raynal 1982; Haynes and Swift 1986). With increased acidification, exchangeable basic cations are replaced on the exchange sites by  $\text{H}_3\text{O}^+$  and solubilized Al species. If the associated anions are mobile, the basic cations are leached as counterions (Johnson et al. 1983). Studies of soils under temperate deciduous forests have found atmospheric acid input increases the annual net losses of cations, but in no case would the losses be considered extreme when compared to the total soil exchangeable cations (Johnson et al. 1983). McCoy and Webster (1977) indicated that a 10-20% decrease in base cations resulted from a pH decline of about 0.5-0.8 units due to fertilizer applications in an Alberta luvisol. The amounts of acidity in all of those studies were not as extreme as were observed at sites 1 and 2. The addition of lime made it difficult to interpret the changes in the Ca and Mg (at site 1) concentrations at these sites. The changes in the K concentrations, however, indicate that considerable leaching losses have occurred at sites 1 and 2 in the LFH horizon. The observations from the heavily  $\text{S}^0$  contaminated sites suggest that leaching losses of Ca, Mg, and K, in particular, could be of concern with respect to the long-term productivity of these sites.

The Ca, Mg, and K concentrations in the LFH of non- $\text{S}^0$  contaminated sites were very stable. Significant changes were observed in the concentrations at only a few sites and, in all cases, the concentrations remained within the range of the other non- $\text{S}^0$  contaminated sites.

Total phosphorus and manganese concentrations in the LFH horizon are given in Table 6. These were the only other elements measured that appeared to be affected by the  $\text{S}^0$  contamination at sites 1 and 2 (Table 6). Total P concentrations decreased by >50% at these sites between 1981 and 1985 (significant at  $p < 0.05$ ) and were the lowest P concentrations of any sites in 1985. Organic matter influences anion adsorption negatively (Johnson et al. 1979; Singh 1980) and there was very little anion adsorption by these LFH horizons (Maynard et al. 1987). Therefore, extremely high  $\text{SO}_4^{2-}$ -S concentrations in the LFH could have displaced P (organic or inorganic phosphate) from the exchange sites, making P more susceptible to leaching. There are no reports

in Morrison's (1984) literature indicating soil acidification affects changes in P availability.

Total Mn concentrations in the LFH at sites 1 and 2 were considerably lower than at any other site (Table 6) in 1985. The differences, although not significant ( $p \geq 0.05$ ), were very striking, particularly at site 1. A large decrease in Mn concentration between 1981 and 1985 was also observed at site 12, although the variability within the site for Mn in 1981 was large ( $1700 \pm 1690$ ) and the Mn concentration in 1985 remained within the range of the non- $\text{S}^0$  contaminated sites.

Total Al and Fe concentrations showed few differences between years or among sites. Some significant changes ( $p < 0.05$ ) between years were observed at a few non- $\text{S}^0$  contaminated sites; however, the Al and Fe concentrations remained within the range of the other sites.

### **Mineral Horizons**

The total S concentrations and pH of the three mineral horizons in 1981 and 1985 are given in Tables 7 and 8, respectively. In 1981, differences in soil chemical properties were observed mainly in the LFH horizon. Higher S concentrations in the Ael horizon were observed at sites 1 and 15 and in the Bm horizon of site 15 in 1981, but no other effects of  $\text{S}^0$  were measurable. Increased S concentrations (Table 7) and a reduction in pH (Table 8) of the three mineral horizons at sites 1 and 2 (particularly 1) in 1985, indicated S had leached at least to the 30 cm depth (Ael horizon) at sites 1 and 2<sup>9</sup>. There were no measurable effects of  $\text{S}^0$  deposition on the mineral horizons of sites 5 and 12 in 1985. A similar pattern was found for extractable S concentrations; therefore, these data are not presented.

The total S concentrations of the mineral horizons at virtually all the non- $\text{S}^0$  contaminated sites and site 12 were lower in 1985 compared to 1981. Three possible hypotheses are:

- i) differences due to the S analysis,
- ii) reduced S emissions from the two major sour gas plants, and

<sup>9</sup> The soil monitoring program carried out by Husky Oil Operations Ltd. (formerly Canterra Energy Ltd.) indicated, in areas heavily contaminated by  $\text{S}^0$ , that S had leached to at least a 60 cm depth by 1985.

**Table 6. Total P and Mn concentrations (mg kg<sup>-1</sup>) of the LFH horizon at selected sites in 1981 and 1985. Values are means  $\pm$  95% confidence limits (C.L.).**

Site	Total P			Total Mn		
	1981	1985	Between years <sup>a</sup>	1981	1985	Between years
<b>S<sup>0</sup> contaminated</b>						
1	1220 $\pm$ 343	492 $\pm$ 64	***	660 $\pm$ 986	45 $\pm$ 17	NS
2	1150 $\pm$ 165	586 $\pm$ 148	***	858 $\pm$ 884	202 $\pm$ 325	NS
4	717 $\pm$ 320	792 $\pm$ 243	NS	1530 $\pm$ 437	1090 $\pm$ 460	NS
5	1080 $\pm$ 298	840 $\pm$ 237	NS	614 $\pm$ 601	718 $\pm$ 1150	NS
12	1220 $\pm$ 508	950 $\pm$ 152	NS	1700 $\pm$ 1690	492 $\pm$ 201	NS
<b>Elevated sulfation rates<sup>b</sup></b>						
3	1090 $\pm$ 197	943 $\pm$ 173	NS	1660 $\pm$ 1670	936 $\pm$ 891	NS
6	796 $\pm$ 153	664 $\pm$ 103	NS	1070 $\pm$ 185	627 $\pm$ 156	**
10	1160 $\pm$ 288	894 $\pm$ 339	NS	588 $\pm$ 261	596 $\pm$ 230	NS
11	1370 $\pm$ 292	1360 $\pm$ 343	NS	1290 $\pm$ 928	887 $\pm$ 628	NS
13	969 $\pm$ 202	1060 $\pm$ 209	NS	458 $\pm$ 325	390 $\pm$ 165	NS
14	820 $\pm$ 174	781 $\pm$ 111	NS	893 $\pm$ 537	700 $\pm$ 642	NS
22	1130 $\pm$ 309	740 $\pm$ 624	NS	980 $\pm$ 727	730 $\pm$ 385	NS
23	820 $\pm$ 174	979 $\pm$ 275	NS	1130 $\pm$ 1140	1180 $\pm$ 926	NS
24	1190 $\pm$ 174	1110 $\pm$ 222	NS	662 $\pm$ 391	628 $\pm$ 397	NS
<b>All non-S<sup>0</sup> contaminated sites<sup>c</sup></b>						
Mean $\pm$ 95% C.L.	1080 $\pm$ 92	941 $\pm$ 95		1160 $\pm$ 280	811 $\pm$ 136	
Range	717 – 1590	664 – 1360		335 – 3140	390 – 1510	

<sup>a</sup> Data analyzed by *t*-test between years. NS = not significant, \*\* =  $p < 0.01$ , and \*\*\* =  $p < 0.001$ .

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rate  $> 0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.



**Table 7. Total sulfur concentrations (mg kg<sup>-1</sup>) of the three mineral horizons at selected sites in 1981 and 1985. Values are means  $\pm$  95% confidence limits.**

Site	Ael			Bm			Aell		
	1981	1985	Between years <sup>a</sup>	1981	1985	Between years	1981	1985	Between years
<b>S<sup>0</sup> contaminated</b>									
1	450 $\pm$ 111	495 $\pm$ 107	NS	517 $\pm$ 104	1550 $\pm$ 610	**	208 $\pm$ 65	328 $\pm$ 261	NS
2	229 $\pm$ 22	171 $\pm$ 40	**	295 $\pm$ 78	748 $\pm$ 490	NS	171 $\pm$ 58	170 $\pm$ 101	NS
4	218 $\pm$ 76	79 $\pm$ 44	**	221 $\pm$ 72	96 $\pm$ 17	**	158 $\pm$ 21	30 $\pm$ 8	***
5	234 $\pm$ 62	68 $\pm$ 57	***	272 $\pm$ 34	165 $\pm$ 62	**	175 $\pm$ 16	70 $\pm$ 34	***
12	296 $\pm$ 79	144 $\pm$ 37	**	270 $\pm$ 52	180 $\pm$ 36	*	168 $\pm$ 38	90 $\pm$ 16	***
<b>Elevated sulfation rates<sup>b</sup></b>									
3	192 $\pm$ 42	83 $\pm$ 31	***	174 $\pm$ 17	87 $\pm$ 24	***	155 $\pm$ 36	38 $\pm$ 14	***
6	291 $\pm$ 79	96 $\pm$ 88	**	229 $\pm$ 41	100 $\pm$ 57	***	166 $\pm$ 10	29 $\pm$ 6	***
10	209 $\pm$ 30	56 $\pm$ 27	***	306 $\pm$ 93	154 $\pm$ 63	**	173 $\pm$ 63	60 $\pm$ 14	**
11	280 $\pm$ 37	119 $\pm$ 62	***	343 $\pm$ 68	192 $\pm$ 40	***	200 $\pm$ 73	104 $\pm$ 34	*
13	79 $\pm$ 87	65 $\pm$ 22	NS	74 $\pm$ 82	128 $\pm$ 99	NS	41 $\pm$ 99	70 $\pm$ 11	NS
14	216 $\pm$ 110	53 $\pm$ 31	*	159 $\pm$ 103	26 $\pm$ 20	*	179 $\pm$ 27	4 $\pm$ 4	***
22	188 $\pm$ 29	19 $\pm$ 21	***	219 $\pm$ 42	97 $\pm$ 50	***	159 $\pm$ 12	31 $\pm$ 17	***
23	220 $\pm$ 32	41 $\pm$ 42	***	257 $\pm$ 77	89 $\pm$ 52	**	187 $\pm$ 106	21 $\pm$ 14	*
24	230 $\pm$ 40	88 $\pm$ 67	***	444 $\pm$ 112	404 $\pm$ 93	NS	456 $\pm$ 154	438 $\pm$ 182	NS
<b>All non-S<sup>0</sup> contaminated sites<sup>c</sup></b>									
Mean	225 $\pm$ 41	78 $\pm$ 14		245 $\pm$ 61	113 $\pm$ 36		200 $\pm$ 34	79 $\pm$ 45	
Range	79 - 561	41 - 148		74 - 732	26 - 404		41 - 456	3.6 - 438	

<sup>a</sup> Data analyzed by *t*-test between years. NS = not significant, \* =  $p < 0.05$ , \*\* =  $p < 0.01$ , and \*\*\* =  $p < 0.001$ .

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rate  $>0.04$  mg dm<sup>-2</sup> d<sup>-1</sup> of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

**Table 8. The pH of the three mineral horizons at selected sites in 1981 and 1985.** Values are means and pH ranges.

Site	AeI			Bm			AeII		
	1981	1985	Between years <sup>a</sup>	1981	1985	Between years	1981	1985	Between years
<b>S<sup>0</sup> contaminated</b>									
1	4.1 ± 0.6	3.3 ± 0.1	*	5.3 ± 0.4	4.2 ± 0.0	**	5.1 ± 0.4	4.2 ± 0.2	***
2	4.7 ± 0.7	3.9 ± 0.6	*	5.7 ± 0.6	4.8 ± 0.5	*	5.4 ± 0.3	5.0 ± 0.2	*
4	4.9 ± 0.3	4.9 ± 0.2	NS	5.5 ± 0.6	5.4 ± 0.4	NS	5.4 ± 0.3	5.7 ± 0.2	NS
5	4.6 ± 0.3	4.5 ± 0.3	NS	5.6 ± 0.2	5.4 ± 0.5	NS	5.5 ± 0.3	5.4 ± 0.5	NS
12	5.1 ± 0.4	4.8 ± 0.5	NS	5.6 ± 0.2	5.3 ± 0.4	NS	5.5 ± 0.1	5.4 ± 0.2	NS
<b>Elevated sulfation rates<sup>b</sup></b>									
3	4.8 ± 0.8	4.7 ± 0.5	NS	5.4 ± 0.7	5.3 ± 0.5	NS	5.4 ± 0.4	5.6 ± 0.3	NS
6	5.3 ± 0.6	5.0 ± 0.4	NS	5.5 ± 0.4	5.5 ± 0.2	NS	5.7 ± 0.2	5.6 ± 0.2	NS
10	4.1 ± 0.2	4.1 ± 0.2	NS	5.4 ± 0.4	5.0 ± 0.3	NS	5.7 ± 0.4	5.7 ± 0.2	NS
11	4.6 ± 0.3	4.6 ± 0.5	NS	5.1 ± 0.4	4.8 ± 0.5	NS	5.4 ± 0.2	5.2 ± 0.2	*
13	4.5 ± 0.2	4.1 ± 0.2	**	5.0 ± 0.2	4.3 ± 0.1	***	5.0 ± 0.2	4.4 ± 0.1	***
14	4.8 ± 0.3	4.4 ± 0.3	NS	5.6 ± 0.4	4.8 ± 0.3	**	5.7 ± 0.4	5.1 ± 0.2	**
22	4.2 ± 0.3	4.1 ± 0.2	NS	5.3 ± 0.6	5.0 ± 0.2	NS	5.4 ± 0.4	5.4 ± 0.3	NS
23	4.1 ± 0.4	4.1 ± 0.4	NS	4.9 ± 0.4	4.7 ± 0.8	NS	5.2 ± 0.5	5.2 ± 0.5	NS
24	4.0 ± 0.2	4.2 ± 0.3	NS	5.2 ± 0.7	5.1 ± 0.5	NS	5.0 ± 0.3	5.1 ± 0.2	NS
<b>Background site</b>									
16	4.8 ± 0.3	4.2 ± 0.3	*	5.0 ± 0.2	4.5 ± 0.2	**	4.9 ± 0.0	4.6 ± 0.1	***
<b>All non-S<sup>0</sup> contaminated sites<sup>c</sup></b>									
Range	4.0 - 5.5	4.1 - 5.2		4.9 - 5.6	4.3 - 5.9		4.9 - 5.6	4.4 - 6.2	

<sup>a</sup> Data analyzed by *t*-test between years. NS = not significant, \* = *p* < 0.05, \*\* = *p* < 0.01, and \*\*\* = *p* < 0.001.

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rate >0.04 mg dm<sup>-2</sup> d<sup>-1</sup> of SO<sub>3</sub>.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

- iii) natural variability owing to environmental changes other than pollutants (e.g., precipitation and leaching).

The S methodology used in 1981 and 1985 was the same. The National Bureau of Standards (NBS) reference materials (pine needles, citrus leaves, and river sediment) and standard laboratory samples analyzed in 1985 produced S values within 5% of what was determined in 1981 and within 2% of the NBS certified concentrations. Therefore, it is very unlikely that the differences in the S concentrations between 1981 and 1985 were related to either the S digestion methodology or the S analysis.

The second hypothesis relates to actual S emissions. Amounts of gaseous S emissions were approximately 30% lower between 1981 and 1985 than in the 7 years previous to 1981. The sulfation data (Figs. 2 and 3) reflects the decrease in gaseous S emissions that were considered to be in areas of elevated sulfation rates in 1981 ( $>0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ ; Addison et al. 1984). In areas with sulfation rates  $<0.05 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$  there was little change in the sulfation rates. Reductions in S concentrations of the mineral horizons, however, were found for all sites, excluding the  $\text{S}^0$  contaminated sites 1, 2, and 5. The decrease in S at 15 sites cannot be attributed to a reduction in S emissions since these sites are located in areas where the sulfation rate did not change. In addition, there was no strong pattern of decreased S concentrations in the LFH horizons. The LFH would be expected to reflect any changes in concentrations due to changes to S gas exposure, before the mineral horizons. Factors other than S deposition, therefore, must have affected the S levels in the mineral soils.

There is considerable evidence that significant differences in various soil chemical parameters measured over time are the result of the natural temporal and spatial variability of the soil (Arp and Krause 1984; Palmer et al. 1985; Courtin et al. 1983; Quesnel and Lavkulich 1980). Differences in S concentrations in mineral horizons probably resulted from temporal changes owing to environmental variations other than S pollutants. The leaching data at locations adjacent to sites 1 and 2 indicated that movement of S out of the LFH horizon was lower in 1983 and 1984 compared to 1982 (Table 2). If reduced movement of S into the mineral horizons was consistent over the region (the amount of S would be considerably lower than at the  $\text{S}^0$  contaminated sites), it is possible that a net loss of S from the mineral horizons occurred.

An additional extraction, phosphate extractable S ( $500 \text{ mg L}^{-1}$  of P as  $\text{Ca}(\text{H}_2\text{PO}_4)_2$ ), was carried out in 1985 to determine the amount of adsorbed sulfate in the three mineral horizons. Determination of both total extractable S and  $\text{SO}_4^{2-}\text{-S}$  in the phosphate extracts indicated the majority of the S was in a sulfate form at sites other than sites 1 and 2. Acetate solutions are not as effective as phosphate solutions in extracting adsorbed  $\text{SO}_4$  and contain only a portion of the adsorbed  $\text{SO}_4^{2-}\text{-S}$  (Ensminger and Freney 1966). The acetate extractant removed a similar amount of S in the Ael and Aell horizons compared to the phosphate extractant. An additional 18% S was removed by the phosphate extractant in the Bm horizon, indicating additional adsorbed sulfate not removed by the acetate extractant was present in the Bm horizon. This coincided with the higher S concentrations in the Bm horizon compared to either the Ael or Aell horizons at sites 1 and 2 in 1985 (Table 7). Similarly, work by Johnson et al. (1981) found higher levels of sulfate adsorption in B horizon forest soils compared to A horizons.

In general, adsorbed  $\text{SO}_4^{2-}\text{-S}$  occurs in mineral soils that contain appreciable amounts of hydrous oxides of Fe and Al or clay colloids, kaolinite, or allophane with Al or Fe as major components (Metson 1979). The B horizon is the illuvial (accumulating) horizon and would contain the components most responsible for anion ( $\text{SO}_4^{2-}\text{-S}$ ) adsorption (Bohn et al. 1986). The Ae horizons (eluviated), however, are characterized by the destruction and removal of clay, and Fe and Al oxides, and would have a lower capacity to adsorb  $\text{SO}_4^{2-}\text{-S}$ . This is an important reason for sampling mineral soil by horizon rather than by depth. Large variations in analysis could occur in soils with variable horizon depths when sampling by depth because of the specific characteristics of the individual horizons.

The pH in the three mineral horizons at site 1 were the lowest of any site. The pH at site 1 decreased significantly ( $p < 0.05$ ) between 1981 and 1985 (Table 8). The lower pH coincided with an increase in the S content of the mineral horizons. The pH of the Ael horizon at site 2 also showed a significant decrease ( $p < 0.05$ ) similar to the decreases found at site 1 (Table 8). Changes in the pH of the Bm and Aell horizons at site 2 were not as obvious. Although the pH in the Bm horizon decreased by 0.9 pH unit ( $p < 0.05$ ) between 1981 and 1985, the pH was still within the range of the non- $\text{S}^0$  contaminated sites. The decrease in pH of the Aell horizon was  $<0.5$  pH unit and the pH remained within the range of pH found

for the Aell horizons at the non-S<sup>0</sup> contaminated sites.

The acidification of mineral horizons at site 1 resulted from leaching of H<sup>3</sup>O<sup>+</sup> and SO<sub>4</sub><sup>2-</sup>-S produced in the LFH horizon by the oxidation of S<sup>0</sup>. The S concentrations in the mineral horizons of site 2 indicated that the S had been leached at least to the Aell horizon (20-30 cm depth). The degree of acidification at site 2, however, appeared to be less than at site 1, probably due to the lower amount of S<sup>0</sup> deposition at site 2. There was no indication that acid produced from S<sup>0</sup> oxidation at sites 5 and 12 had leached below the LFH horizon.

Few significant changes ( $p < 0.05$ ) in the pH of the mineral horizons between 1981 and 1985 were found at any other sites (including 5 and 12, Table 8). Significant decreases ( $p < 0.05$ ) were found in all three mineral horizons of sites 13 and 16. Lower pH in the Bm and Aell horizons of sites 13 and 16 were found only at site 1. Site 13 had roads built on two sides of it, which resulted in a change in drainage. Plant cover was reduced (see Vegetation Section), possibly because of a reduction in the moisture regime of the soil. This change in drainage could also have resulted in the decrease in the mineral soil pH at site 13. The reasons for the lower pH in the mineral horizons at site 16 are unknown. The reduction in the mineral soil pH at sites 13 and 16 did not appear to be related to S pollutants because there was no significant increase in the S concentrations of the LFH at either site between 1981 and 1985.

Higher EC values were observed in all three mineral horizons for sites 1 and 2 in 1985. The EC did not exceed 0.90 dS m<sup>-1</sup> in any case. The highest EC observed was in the Ael of site 1 (0.83 dS m<sup>-1</sup>). The results are consistent with the pH and S concentration data and support the conclusion that at the most heavily S<sup>0</sup> contaminated sites (1 and 2), by-products of the oxidation of S<sup>0</sup> have leached at least to the Aell horizon (20-30 cm).

The increased SO<sub>4</sub><sup>2-</sup>-S concentrations and decreased pH in the mineral horizons had only a minimal effect on other soil chemical properties measured at site 1 and possibly site 2. Decreases in concentrations of Ca, Mg, and K of mineral horizons were observed at site 1 between 1981 and 1985. The decreases were significant ( $p < 0.05$ ) for extractable Ca and K only. Concentrations were still within the range of the concentrations found in the mineral horizons of the non-S<sup>0</sup> contaminated sites. Adsorp-

tion or accumulation of S in the mineral horizons (particularly the B horizon) could limit the leaching of cations (Erik von Freiesleben 1988).

Total P concentrations increased significantly ( $p < 0.05$ ) in the Ael and Bm horizons of site 1 between 1981 and 1985 and the P concentrations for these two horizons were the highest of any other site. This coincided with significant decreases ( $p < 0.05$ ) in P concentrations of the LFH (Table 6). Phosphorus was leached from the LFH and had accumulated in the upper two mineral horizons of site 1. This is consistent with the increased S concentrations at depth and the adsorption properties of P in relation to S. Phosphorus is adsorbed preferentially and more strongly than S (Metson 1979), thus it would be expected that S would be more readily leached than P in soils capable of anion adsorption. There were no significant changes in the total P concentrations for any other horizon or any other site.

### Chemical Composition of the Vegetation

Total S concentrations of vegetation are given in Tables 9 and 10. The highest S concentrations of all four species in 1985 were observed at sites 1, 2, and 5 (moss was absent at site 1). Site 12 had higher S concentrations than all remaining sites and for all species except spruce. This pattern was similar to that observed in 1981 except the magnitude of the differences between the S<sup>0</sup> contaminated sites and the non-S<sup>0</sup> contaminated sites was considerably less, particularly for the understory species (twin-flower and feather moss). This is consistent with the reduced deposition rate of S<sup>0</sup> between 1981 and 1985.

Sulfur concentrations in pine and spruce foliage were significantly lower ( $p < 0.05$ ) in 1985 compared to 1981 at S<sup>0</sup> contaminated sites (except pine foliage at sites 2 and 5), although not to the same extent as in twin-flower and feather moss. The decrease in S<sup>0</sup> deposition at these sites could have affected the S concentrations in the tree foliage; however, the decrease in the S concentrations of the spruce foliage between 1981 and 1985 was observed for all sites (i.e., a 30% decrease in S). This would suggest that environmental factors other than S<sup>0</sup> are influencing the S concentration in the spruce foliage.

Sulfur concentrations found in pine and twin-flower foliage also decreased between 1981 and 1985 at virtually all the non-S<sup>0</sup> contaminated sites (Tables 9 and 10). Decreases were consistent and significant

**Table 9. Total sulfur concentrations ( $\text{mg kg}^{-1}$ ) in the current needles of lodgepole pine and spruce at selected sites. Values are means  $\pm$  95% confidence limits (C.L.).**

Site	Lodgepole pine		Between years <sup>a</sup>	Spruce		Between years
	1981	1985		1981	1985	
<b>S<sup>0</sup> contaminated</b>						
1	2460 ± 355	2070 ± 228	*	2890 ± 235	2000 ± 310	***
2	2030 ± 229	1770 ± 223	NS	1770 ± 203	1200 ± 170	***
4	1220 ± 71	875 ± 74	***	925 ± 63	508 ± 45	***
5	1460 ± 143	1460 ± 144	NS	1280 ± 108	918 ± 103	***
12	1510 ± 247	1160 ± 81	*	1040 ± 89	712 ± 57	***
<b>Elevated sulfation rates<sup>b</sup></b>						
3	1320 ± 149	839 ± 87	***	1160 ± 53	619 ± 41	***
6	986 ± 136	1060 ± 107	NS	779 ± 85	499 ± 20	***
10	1080 ± 110	860 ± 63	**	1160 ± 91	582 ± 34	***
11	1300 ± 147	977 ± 81	***	886 ± 78	653 ± 70	***
13	1380 ± 119	882 ± 91	***	857 ± 61	652 ± 75	***
14	1400 ± 167	1080 ± 81	**	1180 ± 109	704 ± 84	***
22	1060 ± 97	883 ± 50	**	978 ± 80	584 ± 106	***
23	1040 ± 106	972 ± 93	NS	993 ± 110	693 ± 84	***
24	1050 ± 87	842 ± 101	**	859 ± 77	509 ± 40	***
<b>All non-S<sup>0</sup> contaminated sites<sup>c</sup></b>						
Mean	1190	910		953	580	
95% C.L.	± 65	± 41		± 65	± 46	
Range	985 - 1400	730 - 1080		702 - 1210	448 - 789	

<sup>a</sup> Data analyzed by *t*-test for paired comparisons between years. NS = not significant, \* =  $p < 0.05$ , \*\* =  $p < 0.01$ , \*\*\* =  $p < 0.001$ .

<sup>b</sup> Sites that have 5-year (1981-85) sulfation rates  $> 0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ .

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites.

Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

( $p < 0.05$ ) for most sites. Significant decreases ( $p < 0.05$ ) in S concentration of feather moss were observed at several sites including sites 11 and 23 (S gas exposed sites in 1981 and 1985; Table 10); however, these differences were approximately 100  $\text{mg kg}^{-1}$  and S concentrations were within the range of the S concentrations of feather moss at other sites. Mean S concentrations of moss at all non-S<sup>0</sup> contaminated sites in 1981 and 1985 were similar ( $1170 \pm 96$  and  $1070 \pm 70$ , respectively; Table 10).

Decreases in S concentrations of vascular plants were similar to decreases observed in mineral soils and are probably not pollutant-related. If the decreases in S concentrations were pollutant-related, a gradient response would have been expected in

foliage S concentrations. Larger decreases should also have been observed in the vegetation at the sites with the greatest decrease in exposure to S gases, and no changes at sites considered background. The decreases in the S concentrations of the vegetation, however, were similar at all sites. Furthermore, if the decreases in the S concentrations of the vegetation were attributed to a reduction in S gas emissions it would have been most evident in the feather moss, because this species obtains virtually all of its nutrients from the atmosphere.

In 1981, sites 3, 12, and 14 were among the six sites with the highest total S levels for every species sampled, and for twin-flower these three sites had significantly higher ( $p < 0.05$ ) S concentrations

(Addison et al. 1984). Elevated S concentrations in the foliage at these sites suggested some S contamination either by gaseous S emissions or low levels of  $S^0$  (below detection limit). The 1985 data indicates that  $S^0$  was the probable pollutant at site 12. The S concentrations of the pine and twin-flower foliage at site 14 were the highest of any of the non- $S^0$  contaminated sites, although the differences were not significant ( $p \geq 0.05$ ). For the other two species at site 14, and for all four species sampled at site 3, the S concentration of the foliage was within the range of the non- $S^0$  contaminated sites. It is, therefore, very difficult to determine if the slightly higher S concentrations at site 14 were due to S pollutants. Sulfur concentrations in vegetation at site 14 were similar

to those observed for similar species in different locations (Beaton et al. 1965; Morrison 1974; Legge et al. 1986) and followed the same pattern as the S concentrations at the non- $S^0$  contaminated sites, decreasing between 1981 and 1985.

There was very little evidence of changes occurring in the foliage concentrations (other than S) with the exception of feather moss at site 2 that could be attributed to S deposition. The feather moss at site 2 had the lowest concentrations of Mg and Mn and had significantly reduced concentrations of Ca and K. The concentrations, however, were not as low as were observed for the moss at sites 1 and 15 in 1981 prior to the disappearance of the feather moss at these two sites. Plant cover estimates (Table 11) and

**Table 10. Total sulfur concentrations ( $\text{mg kg}^{-1}$ ) in the plant tissue of twin-flower and feather moss at selected sites. Values are means  $\pm$  95% confidence limits (C.L.).**

Site	Twin-flower		Between years <sup>a</sup>	Feather moss		Between years
	1981	1985		1981	1985	
S <sup>0</sup> contaminated						
1	16 400 ± 3 460	5 220 ± 1 280	***	79 100 ± 29 900	N.D. <sup>b</sup>	
2	6 400 ± 1 100	2 670 ± 162	***	20 700 ± 5 040	4 690 ± 712	***
4	1 350 ± 58	895 ± 58	***	1 270 ± 72	1 140 ± 62	**
5	3 310 ± 330	1 710 ± 104	***	9 880 ± 1 070	2 110 ± 138	***
12	1 670 ± 81	1 490 ± 91	**	2 040 ± 82	1 800 ± 53	***
Elevated sulfation rates <sup>c</sup>						
3	1 450 ± 67	788 ± 55	***	1 260 ± 94	1 170 ± 65	NS
6	1 250 ± 85	749 ± 66	***	1 240 ± 37	1 250 ± 55	NS
10	1 120 ± 54	820 ± 51	***	1 100 ± 48	1 030 ± 38	**
11	1 080 ± 52	858 ± 33	***	1 250 ± 55	1 170 ± 56	*
13	1 030 ± 50	734 ± 30	***	1 110 ± 67	1 160 ± 54	NS
14	1 550 ± 115	900 ± 69	***	1 200 ± 108	1 230 ± 72	NS
22	1 170 ± 60	597 ± 51	***	1 140 ± 74	1 030 ± 37	**
23	1 150 ± 39	689 ± 45	***	1 250 ± 44	1 160 ± 30	***
24	1 110 ± 47	792 ± 54	***	1 150 ± 57	723 ± 35	***
All non-S <sup>0</sup> contaminated sites <sup>d</sup>						
Mean	1 190	769		1 170	1 070	
95% C.L.	± 78	± 35		± 96	± 70	
Range	922 - 1 550	597 - 900		902 - 1 270	723-1 290	

<sup>a</sup> Data analyzed by *t*-test for paired comparisons between years. NS = not significant, \* =  $p < 0.05$ , \*\* =  $p < 0.01$ , \*\*\* =  $p < 0.001$ .

<sup>b</sup> N.D. = not determined. There was no sample in 1985 since all the moss had died.

<sup>c</sup> Sites that have 5-year (1981-85) sulfation rates  $>0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ .

<sup>d</sup> In 1981 the non- $S^0$  contaminated sites included site 12. In 1985, site 12 was excluded from the non- $S^0$  contaminated sites.

Sites 3 and 4 are still included even though  $S^0$  was detected at these sites.

**Table 11. Total plant cover and species richness of each site in 1981 and 1985**

Site	Plant cover (%)		No. species	
	1981	1985	1981	1985
<b>S<sup>0</sup> contaminated</b>				
1	18	0.5	24	6
2	29	8	24	17
4	42	56	38	36
5	67	76	36	34
12	34	42	27	26
<b>Elevated sulfation rates<sup>a</sup></b>				
3	ND <sup>b</sup>	ND	ND	ND
6	57	50	33	29
10	48	53	19	19
11	42	68	25	25
13	55	38	29	30
14	34	64	28	28
22	50	75	22	24
23	36	56	23	22
24	22	47	21	18
<b>Non-S<sup>0</sup> contaminated sites<sup>c</sup></b>				
4	42	56	38	36
7	44	61	33	29
8	43	55	38	35
9	31	34	25	24
16	57	81	32	40
17	43	43	20	19
18	40	64	32	35
20	40	41	24	27
21	36	49	27	30
25	23	60	21	19
26	40	28	17	19

<sup>a</sup> Sites that have 5-year (1981-85) sulfation rates  $>0.04 \text{ mg dm}^{-2} \text{ d}^{-1}$  of  $\text{SO}_3$ .

<sup>b</sup> ND = not determined.

<sup>c</sup> In 1981 the non-S<sup>0</sup> contaminated sites included site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

visual observations indicated that the moss at site 2 had deteriorated but had not completely disappeared. The chemical composition of the moss along with the S<sup>0</sup> concentration in the LFH and the plant cover estimates would suggest that site 2 was not as severely impacted as either sites 1 or 15 in 1981.

It is difficult to assess other significant changes observed in the chemical composition of the vegetation at the heavily contaminated sites. Several signifi-

cant increases and decreases in concentrations were observed between 1981 and 1985 for some elements in the four species sampled. Similar significant changes, however, were observed at many of the non-S<sup>0</sup> contaminated sites. In addition, most of the concentrations observed at the S<sup>0</sup> contaminated sites were within the range of the concentrations observed at the non-S<sup>0</sup> contaminated sites and were similar to those reported elsewhere for similar species (Beaton et al. 1965; Morrison 1974; Legge et al. 1986).

## Vegetation Response

The understory vegetation of sites 1 and 2 died between 1981 and 1985. Plant cover decreased 97% in site 1 and 72% in site 2, and species richness decreased 75% and 29%, respectively (Table 11). The decreases in plant cover at sites 1 and 2 could not be accounted for by the uncertainty in the estimation of the species cover since the decreases were much larger than the measurement uncertainty of the technique (10%; Kennedy and Addison 1987). In addition, differences were greater than the year-to-year variability at site 1 (10%; Kennedy and Addison 1987). The changes in the vegetation at sites 1 and 2, therefore, appeared to be related to the deposition of  $S^0$  and its subsequent oxidation, although gaseous S emissions cannot be completely ruled out.

Changes in vegetation at site 1 from 1981 to 1985 resulted in conditions comparable to site 15 in 1981. The moss cover was 1% in 1981, leaves were chlorotic, and apical leaves were white. By 1982, all the moss was dead and the herb cover decreased 54% (Kennedy et al. 1988). This indicated a pattern in the decline of vegetation in areas of  $S^0$  deposition: mosses affected first, followed by herbs and then shrubs. Changes at site 2 were similar but less severe than at site 1. Temporal differences between sites were attributed to the duration of exposure to  $S^0$  and its breakdown products (Addison et al. 1984).

There were no discernible changes in the vegetation that could be attributed to either gaseous S emissions or  $S^0$  at any other site (except at sites 1 and 2; Table 11). Cover estimates and species richness were stable (within the measurement uncertainty of the technique (10%); Kennedy and Addison 1987) and there were no visible symptoms of injury.

Variations in species response and soil chemical changes make it difficult to link specific soil factors with plant response. Direct effects of  $H_3O^+$  on agricultural crops have been observed in many studies (Clark 1984); however, the sensitivities of native plant species in this study are unknown. Soil acidity could also have affected the plants indirectly. With increased acidification there were increased

conductivities (Table 4), nutrient losses (Tables 5 and 6) associated with increased solubilization and leaching from the LFH horizon, and changes in microbial populations (Maynard et al. 1986).

All the plant species did not respond simultaneously to soil changes at site 1. Some species such as bishop's-cap *Mitella nuda*) died quickly while others, for example, Canadareedgrass (*Calamagrostis canadensis*), declined over time before they died, and a few persisted, for example, prickly rose (*Rosa acicularis*) (Table 11). Some possible explanations for the differences in species response include rooting depth, carbohydrate root reserves, root anatomy, or species physiology, although specific mechanisms were not determined.

## Pine Growth

Lodgepole pine needle production and retention were the only growth measurements taken in 1985 (Table 12). Needle weights between 1981 and 1985 were not directly comparable because in 1981 the samples were taken from the top of the trees and in 1985 (because no trees were felled) samples were taken from the bottom one-third of the crown. In addition, environmental conditions such as available moisture can affect mean needle weight.

As in 1981 (Addison et al. 1984), there were no differences among sites for needle production or needle retention, with one exception. The weight of needles retained at site 1 in 1985 was <50% of the weight of the needles retained in 1984. For all other sites the percentage of needles retained in 1985 compared to 1984 was 76-140%. In addition, visual observations at site 1 indicated many of the branches had little or no current growth (1985). Site 1 has been impacted by  $S^0$  and S gas emissions for more than 4 years yet pine growth effects were only observed in 1985. This time lag, combined with the significant changes in the soil chemistry found at site 1 in 1985, indicates that the main pathway for pollutant effects on pine growth may be through the soil rather than the direct effect of  $S^0$  or S gas to the foliage.



**Table 12. Lodgepole pine needle production and retention at selected sites.** Values are means  $\pm$  95% confidence limits (C.L.).

Site	Needles produced in 1984	1984 weight per needle	Weight of needles retained (% of 1984)						
	(g cm <sup>-1</sup> of stem)	(mg)	1985	1984	1983	1982	1981	1980	1979
S <sup>0</sup> contaminated									
1	0.35 ± 0.08	13.7 ± 3.2	44	100	122	97	117	80	38
2	0.25 ± 0.10	15.2 ± 2.8	110	100	121	131	142	73	36
4	0.40 ± 0.12	17.3 ± 3.1	108	100	103	57	8	9	0
5	0.28 ± 0.08	13.2 ± 1.4	102	100	110	76	31	17	1
12	0.49 ± 0.16	17.2 ± 2.8	96	100	112	100	69	67	68
Elevated sulfation rates <sup>a</sup>									
3	0.42 ± 0.12	16.9 ± 3.2	120	100	116	126	18	18	0
6	0.35 ± 0.07	13.4 ± 1.6	88	100	126	85	33	11	3
10	0.68 ± 0.19	18.5 ± 3.1	93	100	87	89	14	10	1
11	0.56 ± 0.09	18.0 ± 1.9	108	100	138	120	57	40	52
13	0.45 ± 0.09	15.7 ± 3.3	91	100	118	107	28	68	46
14	0.40 ± 0.12	14.3 ± 2.1	95	100	115	111	38	28	28
22	0.52 ± 0.08	17.8 ± 1.9	96	100	123	109	47	44	19
23	0.49 ± 0.13	15.5 ± 3.2	105	100	125	111	32	40	35
24	0.64 ± 0.23	21.2 ± 1.9	131	100	108	86	61	39	35
All non-S <sup>0</sup> contaminated sites <sup>b</sup>									
Mean	0.49	17.1	101	100	113	94	29	24	16
95% C.L.	±0.04	±1.2	±8		±8	±9	±8	±8	±8
Range	0.35 - 0.68	12.7 - 22.3							

<sup>a</sup> Sites that have 5-year (1981-85) sulfation rate  $>0.04$  mg dm<sup>-2</sup> d<sup>-1</sup> of SO<sub>3</sub>.

<sup>b</sup> In 1981 the non-S<sup>0</sup> contaminated sites include site 12. In 1985, site 12 was excluded from the non-S<sup>0</sup> contaminated sites. Sites 3 and 4 are still included even though S<sup>0</sup> was detected at these sites.

## CONCLUSIONS

Results of the biomonitoring study in the forests surrounding the two sour gas processing plants indicate that, at past and present S gas emission rates, there was no measurable impact to the soils and vegetation that could be attributed to S gas emissions alone other than a possible elevation of S concentrations in the foliage of twin-flower and lodgepole pine at two sites. Measurable impact of S pollutants was limited to within a 2-3 km radius of the plants and was primarily the result of  $S^0$  contamination. At sites heavily contaminated by  $S^0$ , it is possible that  $SO_2$  may have had some impact; however, the overwhelming influence of  $S^0$  at these sites precluded measurement.

Changes in S concentrations and other soil chemical properties measured in 1985 that were pollutant-related were limited to the four  $S^0$  contaminated sites (1, 2, 5, and 12). This contradicts earlier work (Nyborg et al. 1977; Chaudhry et al. 1982) that suggested direct deposition of  $SO_2$  to moist soils downwind of these sour gas processing plants would average  $50 \text{ kg ha}^{-1} \text{ yr}^{-1}$  of S and would result in a drop of the pH level from 0.5 to 1.5 units over a period of 10-20 years (assumed constant deposition). That study was conducted in 1973-74; the results of the monitoring in 1981 and 1985 found no evidence of increased S concentrations nor reduced pH in areas of S gas exposure in the vicinity of two large sour gas plants, except in areas contaminated with  $S^0$ . Nyborg et al. (1977) had indicated, however, that their predicted acidification of soils was subject to errors due to the imprecision of the method of measurement of the total soil S. In addition, the apparent decrease in pH may not have been real, as the measured changes were less than natural temporal and spatial variability.

The decline of vegetation in the understory of the lodgepole pine forest at site 1 was probably caused by changes in soil chemistry, primarily acidification (soil pH 2.5), due to the oxidation of  $S^0$ . Increased acidity and decreased nutrient levels in the LFH horizon were observed in 1981 and further depletion of nutrient elements has occurred since then. There was no measured or noticeable affect on pine growth until 1985, when a loss of needles was observed in lodgepole pine at site 1. Damage to the vegetation followed the sequence of feather moss, herbs, shrubs, and finally trees.

Specific mechanisms leading to vegetation decline at sites 1 and 2 could not be ascertained;

however, a number of plant and soil factors may be involved: depth of rooting, carbohydrate reserves, physiological sensitivities to  $H_3O^+$ , high conductivities, and nutrient deficiencies. It was apparent that, as in most forest situations, plants responded to the complex rather than the individual stresses.

The following conclusions and recommendations result from the 5-year biomonitoring study in the vicinity of the two sour gas processing plants.

1. Past and present levels of S gas emissions have had little or no measurable effect on soils and vegetation in the area.
2. Significant impacts of  $S^0$  from sour gas processing plants on forests were limited to areas contaminated with  $>5000 \text{ mg kg}^{-1}$  of  $S^0$  (approximately  $450 \text{ kg ha}^{-1}$  of  $S^0$ ) after some 5-10 years of exposure in the LFH horizon, generally within 1 km of a S block or S handling facility.
3. The level of  $S^0$  deposition varies with the method of  $S^0$  handling and  $S^0$  block removal, and the amounts of  $S^0$  shipped. Deposition rates, therefore, cannot be extrapolated beyond the measurement period. Deposition of  $S^0$  into the forest adjacent to the two sour gas processing plants decreased between 1981 and 1985 as a result of reduced activity (i.e., shipping of  $S^0$ ) and improved methods of  $S^0$  removal from the blocks. The breakup and removal of the base pads could increase  $S^0$  deposition.
4. Significantly lower S concentrations in the mineral horizons and the foliage, particularly spruce and twin-flower, were found in 1985 compared to 1981. These decreases were probably not pollutant-related but a response to other natural environmental conditions.
5. Some heavily  $S^0$ -contaminated sites should be reexamined in 1990 to measure the potential amelioration of the liming programs that are being conducted regularly around the sour plants gas. The addition of lime may or may not be effective in mitigating or preventing impacts associated with soil acidification.
6. Often because of spatial and temporal variability there is difficulty in determining if differences in measured parameters between sites are a pollu-

tant response or a response to other natural environmental conditions (e.g., precipitation and temperature). The variability of a given technique or measurement parameter should be evaluated

for the soils and vegetation of an area prior to the establishment of any long-term biomonitoring studies.

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