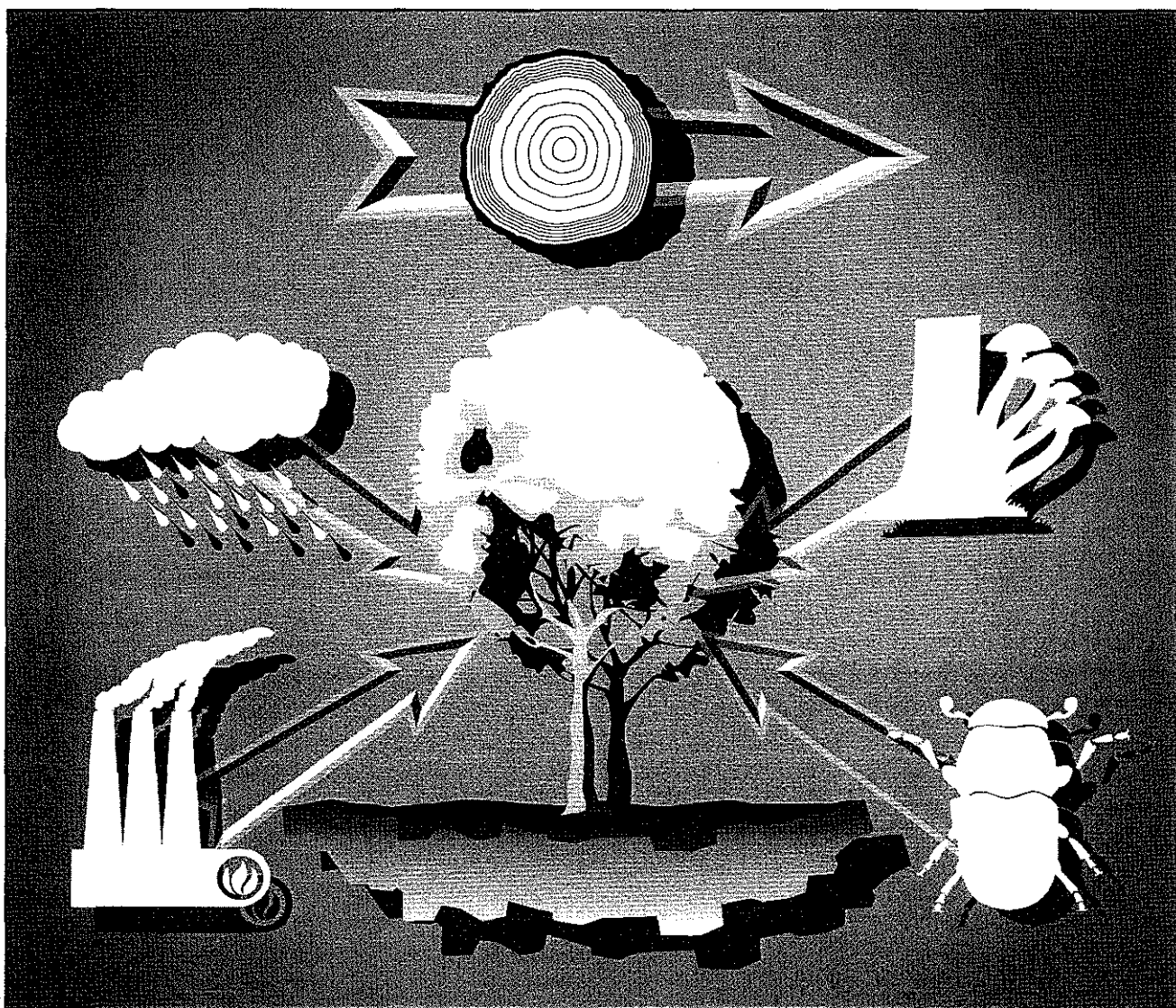




## Sulfur impacts on forest health in west-central Alberta

D.G. Maynard, J.J. Stadt, K.I. Mallett, W.J.A. Volney  
Northwest Region • Information Report NOR-X-334



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# **SULFUR IMPACTS ON FOREST HEALTH IN WEST-CENTRAL ALBERTA**

*D.G. Maynard, J.J. Stadt, K.I. Mallett, and W.J.A. Volney*

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## ABSTRACT

The processing of sour natural gas results in the emission of sulfur dioxide (SO<sub>2</sub>) and elemental sulfur (S<sup>0</sup>). An assessment was carried out in 1991 to compare soil and foliar chemical properties originally measured in 1981 and 1985 in forests near two sour gas processing plants in west-central Alberta. This study was expanded to evaluate forest health and tree growth in relation to sulfur (S) deposition in mature and immature lodgepole pine (*Pinus contorta* Dougl. var. *latifolia* Engelm.) and mature trembling aspen (*Populus tremuloides* Michx.). Ten (50 x 2 m) plots were established in 30 mature lodgepole pine sites, in 6 young lodgepole pine sites, and in 3 trembling aspen-dominated sites. The mature pine sites were classified into 3 deposition classes: high, medium, and low. The distribution of measurable S<sup>0</sup> deposition increased from 2 km to 4–6 km from the S<sup>0</sup> load-out areas; however, S<sup>0</sup> concentration in the LFH near the gas plants has decreased since 1981. The medium deposition class sites had S concentrations in the LFH and foliage 6–20% higher than the low deposition class sites. Calcium (Ca) and magnesium (Mg) concentrations and pH were about 10% less in the medium deposition class sites than in the low deposition class sites. It was, however, not possible to determine a cause and effect relationship between higher S in these sites and lower pH, Ca and Mg. A greater proportion of recently dead trees was found in high deposition class sites, but no difference was seen between the other two deposition classes. There was no increase in insect- and disease-caused damage with increased S deposition in the pine stands. Trembling aspen appeared to be affected by S<sup>0</sup> deposition with increased proportions of dead and declining trees and increased incidences of Armillaria root rot and Hypoxylon canker in S<sup>0</sup>-dusted areas. There were no differences among deposition classes on radial increments at dbh. Annual volume increments were depressed in high S<sup>0</sup> deposition sites; however, specific volume increment was depressed in both the medium and high deposition classes indicating a possible physiological impact historically. Elevated S concentrations in the LFH and foliage and a depression in the annual volume increments were the only indicators of S impacts in lodgepole pine forests near the two sour gas processing plants. There was no apparent widespread forest decline outside of areas dusted with S<sup>0</sup>.

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## RÉSUMÉ

Le traitement du gaz naturel sulfureux provoque l'émission de dioxyde de soufre (SO<sub>2</sub>) et de soufre élémentaire (S<sup>0</sup>). Une évaluation a été effectuée en 1991 afin de comparer les propriétés chimiques du feuillage et du sol mesurées en 1981 et 1985 dans des forêts situées près de deux usines de gaz sulfureux dans le centre-ouest de l'Alberta. La portée de cette étude a été élargie pour inclure l'évaluation de la santé des forêts et de la croissance des arbres, en fonction des dépôts de soufre (S), dans des forêts mûres et jeunes de pins tordus latifoliés (*Pinus contorta* Dougl. var. *latifolia* Engelm.) et dans des forêts mûres de peupliers faux-trembles (*Populus tremuloides* Michx.) 10 parcelles (50 m sur 2 m) ont été établies

dans 30 stations occupées par des pins tordus latifoliés mûrs, dans 6 stations de jeunes pins tordus et dans 3 stations dominées par des peupliers faux-trembles. Dans les stations de pins mûrs, 3 classes de dépôt ont été établies, soit élevé, moyen et faible. La distribution des dépôts mesurables de  $S^0$  s'est étendue, passant de 2 à 4 à 6 km du point d'émission; toutefois, la concentration de  $S^0$  dans les sous-horizons LFH a diminué près des usines depuis 1981. Les stations à dépôt moyen avait des concentrations de S dans le feuillage et les sous-horizons LFH 6 à 20 % plus élevées que les stations à dépôts faible. Les teneurs en calcium (Ca) et en magnésium (Mg) et le pH étaient plus basses d'environ 10 % dans les stations de la catégorie moyenne que dans les stations à dépôt faible. Nous n'avons toutefois pu établir de relation de cause à effet entre les concentrations plus élevées de S dans ces stations et les teneurs plus faibles en Ca et Mg et le pH plus bas. Un pourcentage plus élevé d'arbres morts récemment a été constaté dans les stations à dépôt élevé, mais aucune différence n'a été relevée entre les deux autres classes. Les dégâts causés par les insectes et les maladies n'ont pas augmenté avec l'accroissement des dépôts de S dans les peuplements de pins. Le peuplier faux-tremble semblait être affecté par les retombées de  $S^0$ ; en effet, nous avons constaté un pourcentage plus élevé d'arbres morts et moribonds et des taux d'infection accrus par le pourridié-agaric et le chancre hypoxylonien dans les secteurs recevant des retombées de  $S^0$ . L'accroissement radial à hauteur de poitrine ne présentait aucune différence d'une classe de dépôt à l'autre. L'accroissement annuel en volume était moindre dans les stations à dépôt élevé; l'accroissement spécifique en volume était toutefois moindre dans les classes de dépôt moyen et élevé, indiquant un impact physiologique possible dans le temps. Des teneurs plus élevées en S dans les sous-horizons LFH et dans le feuillage et une diminution de l'accroissement annuel en volume sont les seuls signes indiquant que les retombées de S pourraient avoir des incidences sur les forêts de pins tordus latifoliés situées près des deux usines de gaz sulfureux. Il n'y avait aucun dépérissement apparent à grande échelle des forêts à l'extérieur des secteurs recevant des retombées de  $S^0$ .

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## CONTENTS

INTRODUCTION . . . . .	1
MATERIALS AND METHODS . . . . .	3
Study Sites . . . . .	3
Climate . . . . .	5
Soil and Foliar Chemical Analysis . . . . .	5
1981, 1985, and 1991 Comparison . . . . .	5
1991 Mature Pine Sites . . . . .	8
1992 Young Pine and Mature Aspen Sites . . . . .	8
Forest Health Survey . . . . .	8
Pine Growth . . . . .	9
Measures of Tree Productivity . . . . .	9
Field and Laboratory Procedures . . . . .	10
Analysis . . . . .	10
Statistics . . . . .	10
RESULTS AND DISCUSSION . . . . .	11
Pollution Distribution . . . . .	11
Sulfation . . . . .	11
Elemental Sulfur Particulate . . . . .	12
Liming . . . . .	13
Classification of Stands . . . . .	13
Description and Composition of Stands . . . . .	14
Mature Stand Survey . . . . .	14
Aspen Stand Survey . . . . .	15
Young Stand Survey . . . . .	15
Soil and Foliar Chemical Analysis . . . . .	16
Sulfur . . . . .	16
pH, Calcium, and Magnesium . . . . .	22
Other Elements . . . . .	28
1992 Young Pine Sites . . . . .	31
1992 Aspen Sites . . . . .	33
Forest Health Survey . . . . .	35
Health Conditions . . . . .	35
Tree Part Damage . . . . .	39
Damaging Agents . . . . .	42
Pine Growth . . . . .	44
Analysis of Increment Cores . . . . .	44
Analysis of Annual Volume Increments . . . . .	44
Analysis of Specific Volume Increments . . . . .	45
Comparison of Early Growth in Young and Old Stands . . . . .	47
SUMMARY . . . . .	48
ACKNOWLEDGMENTS . . . . .	49
LITERATURE CITED . . . . .	50

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## FIGURES

1. Location of biomonitoring plots and sour gas processing plants in west-central Alberta . . . . .	4
2. Climate diagram for the Prairie Creek Ranger Station . . . . .	5
3. May to August precipitation and average temperature at the Baseline Mountain Lookout between 1968 and 1992 . . . . .	6
4. Sulfur dioxide emissions from the incinerator stacks of the Ram River and Strachan sour gas processing plants . . . . .	11
5. Isopleths of average sulfation rates near sour gas processing plants in west-central Alberta, 1975–86 . . . . .	12
6. Formed sulfur shipped from Ram River and Strachan sour gas processing plants . . . . .	13
7. Classification of mature lodgepole pine study sites on the basis of sulfation and total sulfur in the LFH . . . . .	14
8. The pH of the surface organic horizon at selected sites in 1981, 1985, and 1991 . . . . .	23
9. The pH of the IAe horizon at selected sites in 1981, 1985, and 1991 . . . . .	24
10. The pH of the Bm horizon at selected sites in 1981, 1985, and 1991 . . . . .	25
11. The pH of the IAe horizon at selected sites in 1981, 1985, and 1991 . . . . .	25
12. Proportion of all trees from the 31 mature lodgepole pine study sites in the four health classes . . . . .	36
13. Proportion of lodgepole pine trees from the 31 mature lodgepole pine study sites in the four health classes . . . . .	36
14. Size class distribution of lodgepole pine in the seven mature high deposition class sites . . . . .	37
15. Size class distribution of lodgepole pine in the 13 mature medium deposition class sites . . . . .	37
16. Size class distribution of lodgepole pine in the 11 mature low deposition class sites . . . . .	38
17. Proportion of trembling aspen trees from the 31 mature lodgepole pine study sites in the four health classes . . . . .	38
18. Proportion of trembling aspen trees from the three trembling aspen study sites in the four health classes . . . . .	38



19. Proportion of all trees from the two low S deposition young lodgepole pine sites in the three health classes . . . . .	40
20. Proportion of all trees from the three medium S deposition young lodgepole pine sites in the three health classes . . . . .	40
21. Proportion of all trees from the high S deposition young lodgepole pine site in the three health classes . . . . .	40
22. Proportion of all living trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class . . . . .	40
23. Proportion of living lodgepole pine trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class . . . . .	41
24. Proportion of living white spruce trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class . . . . .	41
25. Proportion of living black spruce trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class . . . . .	41
26. Proportion of living trembling aspen trees in the three trembling aspen sites with symptoms vs. no symptoms by S deposition class . . . . .	41
27. Proportion of dead trembling aspen trees in the three trembling aspen sites with symptoms vs. no symptoms by S deposition class . . . . .	42
28. Proportion of total observed symptoms and damage on each tree part of lodgepole pine trees in the mature lodgepole pine sites by S deposition class . . . . .	42
29. Proportion of stem damage to non-stem damage on lodgepole pine trees in the mature lodgepole pine study sites by S deposition class . . . . .	42

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**TABLES**

1. Number and proportion of trees by major tree species in the three deposition classes of the mature pine stands, in the young pine stands, and in the mature aspen stands . . . . .	15
2. Elemental sulfur concentrations of the LFH horizon at selected mature pine sites in 1981, 1985, and 1991 . . . . .	16
3. Total sulfur concentrations of the LFH horizon at selected mature pine sites in 1981, 1985, and 1991 . . . . .	17
4. Extractable sulfur concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991 . . . . .	18
5. Extractable sulfur concentrations of the mineral horizons at sites 1 and 2 in 1981, 1985, and 1991 . . . . .	18

6. Total and extractable or exchangeable concentrations of the LFH in 1991 by deposition class . . . . .	19
7. Total sulfur concentrations in 1-year-old pine needles at selected sites in 1981, 1985, and 1991 . . . . .	20
8. Total sulfur concentrations in 1-year-old spruce needles at selected sites in 1981, 1985, and 1991 . . . . .	20
9. Total sulfur concentrations in feather moss at selected sites in 1981, 1985, and 1991 . . . . .	21
10. Total sulfur concentrations in twin-flower at selected sites in 1981, 1985, and 1991 . . . . .	21
11. Total sulfur concentrations in alder at selected sites in 1981 and 1991 . . . . .	22
12. Total sulfur concentrations in 1991 foliage by deposition class . . . . .	23
13. The exchangeable calcium concentrations of the LFH at selected sites in 1981, 1985, and 1991 . . . . .	26
14. The exchangeable magnesium concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991 . . . . .	26
15. Total calcium concentrations in 1991 foliage by deposition class . . . . .	27
16. Total magnesium concentrations in 1991 foliage by deposition class . . . . .	27
17. Exchangeable potassium concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991 . . . . .	28
18. Total potassium concentrations in 1991 foliage by deposition class . . . . .	29
19. Exchangeable aluminum concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991 . . . . .	29
20. Exchangeable aluminum concentrations of the mineral horizons of sites 1 and 2 in 1981, 1985, and 1991 . . . . .	30
21. Exchangeable manganese concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991 . . . . .	31
22. Total and extractable sulfur in the quadrat LFH of the young lodgepole pine sites . . . . .	32
23. Total sulfur concentrations in the current and 1-year-old needles of the young lodgepole pine . . . . .	32
24. Total sulfur concentrations in the foliage of understory species of the young lodgepole pine stands . . . . .	32
25. The pH, total, and extractable or exchangeable concentrations in the LFH of the trembling aspen stands . . . . .	33

26. The pH and extractable or exchangeable concentrations in the mineral horizons of the trembling aspen stands . . . . .	34
27. Total sulfur and calcium concentrations of selected species from the three trembling aspen stands . . . . .	35
28. Incidence of damaging agents in the three sulfur classes of the 31 mature pine stands . . . . .	43
29. Incidence of major pests in the young lodgepole pine stands in the three deposition classes . . . . .	44
30. Analysis of variance of aggregated standardized radial increments for the decades after 1960 . . . . .	45
31. Analysis of variance of annual volume increment index for decades after 1960 . . . . .	45
32. Least square means of specific volume and annual volume increment indexes for decades after 1960 and deposition classes . . . . .	46
33. Specific volume increment index for decades after 1960 . . . . .	46
34. Analysis of variance of cumulative volume increment to age 11 years for trees germinating before and after the sour gas plants opened . . . . .	46
35. Least squares means of cumulative volume to age 11 years for periods before and after the opening of the sour gas plants and deposition class . . . . .	47

**Note**

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## INTRODUCTION

The potential impact of sulfur (S) emissions from sour gas processing in Alberta continues to be a concern. Detailed studies near sour gas processing facilities (Addison et al. 1984; Lore 1984; Legge et al. 1988) and tar sands processing plants (Addison et al. 1986) have found impacts near the pollutant source, but little or no measurable impact on a regional basis. Elemental S ( $S^0$ ) has been measured up to 6 km from the  $S^0$  sources (Maynard 1990); however, major impacts to soils and understory vegetation have been limited to <1 km from the sour gas processing facilities. The accumulated amounts of  $S^0$  within the heavily impacted areas were in the  $t\ ha^{-1}$  range compared to the  $kg\ ha^{-1}$  range ( $S^0$  and  $SO_2$ ) in areas >1 km from the plants with little or no measurable impact. Changes in forest conditions that might be directly associated with gaseous S emissions (primarily sulfur dioxide [ $SO_2$ ]) were not found.

The effects of low level gaseous  $SO_2$  deposition is difficult to isolate from effects of  $S^0$  deposition. Sulfur dioxide is absorbed by the soil and vegetation and is rapidly converted into S compounds that exist naturally. Elemental S, however, does not occur naturally in surface forest soils and thus, its presence would indicate deposition (Maynard and Addison 1985). Sulfur dioxide emissions from Husky's Ram River and Gulf's Strachan sour gas processing plants are greater than  $S^0$  deposition, however,  $SO_2$  is distributed over a wider area resulting in much lower rates of S deposition on  $kg\ ha^{-1}$  (area) basis (Concord Scientific Corporation 1989). Estimated cumulative S deposition from  $SO_2$  in the study area during the lifetime of the Ram River plant (22 years) ranged from 28 to 93  $kg\ S\ ha^{-1}$  (David McCoy, personal communication, FAX request, February 25, 1994). Estimates of  $S^0$  deposition during the 6 months between April and October 1983 were 263 and 70  $kg\ S\ ha^{-1}$  at sites 50 and 250 m east of the Strachan plant, respectively (Maynard 1990). At these sites, cumulative  $SO_2$  deposition during the lifetime of the Strachan plant was about 90  $kg\ S\ ha^{-1}$  (based on the average sulfation rate between 1975 and 1986); however, the overwhelming influence of  $S^0$  precluded measurement of any  $SO_2$  effect. At sites outside the zone of heaviest  $S^0$  deposition, up to 6–8 km from the gas plants, increased S concentrations in the vegetation and soil were observed (Addison et al. 1984; Maynard 1990). It has not been possible to determine if the source of increased S in the LFH and foliage was  $SO_2$  or  $S^0$ .

In the past decade public attention was drawn to so-called forest declines and diebacks, and in particular the Waldsterben (the German term for forest decline) and the maple decline in North America. These declines have been blamed, in whole or in part, on anthropogenic airborne pollutants; however, to date there has been no common consensus on a definition of forest decline (Manion and Lachance 1992), let alone the cause of declines or diebacks. Several models have been proposed to explain forest decline (Manion and Lachance 1992). They all describe the decline as an effect on the host tree species brought about by the interactions of the host tree species, a climatic or abiotic event, and other organisms (pathogens or insects). These interactions often take place over a prolonged period of time.

All forest stands, whether they be natural, planted, exposed or unexposed to pollutants will have constituent trees that can be classified into various health condition categories: healthy, declining, and dead. Health of trees, as with any long-lived organisms, will be strongly affected by the past and current environmental conditions. These conditions, such as climate, edaphic factors, and air pollutants can directly and indirectly influence tree health. Forest trees, like any other population of living organisms, are subject to disease and predation, which are, in turn, influenced by environmental factors.

Tree productivity is closely related to tree health. These two characteristics are not identical, however, because a healthy tree may exhibit very slow growth. The height as well as the radial and volume growth rates of trees in forest stands have predictable patterns as trees age. These patterns may be used to assess the impact of environmental influences on the productivity of trees and consequently the health of stands. A major concern in this process, however, is that tree growth and tree health are affected by many environmental factors simultaneously. As a result, the unequivocal implication of a single factor affecting tree health or growth requires considerable replication and objective measures of the intensity of that factor. By comparing the tree health and growth in areas affected by different levels of a factor, an epidemiological statement can be developed to identify the association between changes in tree growth and the single environmental agent. This process does not establish

cause and effect. Rather it leads to the formulation of plausible hypotheses, which may be tested by experiment.

The most obvious factor affecting the health and growth of trees is associated with the conditions of competition among trees in a single stand. Characteristically the annual growth of an individual tree increases at the time of tree establishment to a maximum at a time when competition from neighboring trees checks the unimpeded growth of the individual tree. Following this phase, growth rates decline over the remaining life of the tree. The tree may show spectacular changes in this pattern if environmental conditions change. Thus if neighboring trees are removed from the stand, surviving trees respond with increased growth. Thus stand density, as an index of the intensity of competition, is related to the productivity of individual trees.

Site conditions, as reflected by moisture available to the root system and soil fertility, also affect growth. Excessive moisture and drought both reduce growth. Changes in the drainage conditions of the site can have dramatic effects on tree growth. The physical and chemical properties of the soil also contribute to variation in tree growth. Similarly, these edaphic conditions can directly or indirectly influence tree health. Factors that change these properties of the site will ultimately affect the productivity and health of trees growing on the site.

Biotic conditions have a major influence on the health and productivity of individual trees. Insects and diseases affect trees by reducing productivity as a result of their activities. Each of these agents have their own pattern of impact that can be isolated from the effects of other factors provided a history of the stand is known. Some biotic associations, such as mycorrhizal fungi, improve tree and stand productivity.

Climatic factors, such as temperature and moisture, can have a direct effect on tree health and growth, for example, winter desiccation. Climatic factors can also indirectly influence tree health, and therefore ultimately growth, through interactions with insect predators and pathogens of trees. Specific environmental conditions must be present in order for an insect or disease outbreak to occur. The local climate and the weather in a particular year set the general level and the degree of annual variation in observed tree growth. The effects of

weather and microclimate can be quite local, affecting trees only in an individual stand. Some episodic events, such as hail damage, may depress tree growth for several years during which the tree is recovering. The potential for tree growth is determined in one year by the growing conditions that year, but the expression of that potential depends on conditions in the following year. Weather has a great influence on tree health and growth.

The genetic characteristics of the species and the individual tree also determine its reaction to various extrinsic environmental agents. Lodgepole pine (*Pinus contorta* Dougl. var. *latifolia* Engelm.) as a species is extremely tolerant of a wide range of environmental conditions. Its range extends from Mexico to the Arctic, from coastal locations to montane habitats at 3000 m elevation along the same latitude, and from sandy, well-drained sites to water-saturated soils in the same region. Individual trees of this species are thus able to withstand a wide variety of conditions. The species may only show symptoms of decline under the most extreme conditions.

Air pollutants, such as SO<sub>2</sub> or ozone, can cause direct injury to tree species (Malhotra and Blauel 1980, Manion 1991). In some cases, they are also thought to affect plants indirectly by predisposing the plant to other deleterious agents such as insect and disease-causing organisms (Huttunen 1984). The indirect effects of acid deposition on tree health and growth are most often associated with soil acidity resulting in 1) increased leaching of base cations, 2) increased solubility of aluminum (Al) to toxic levels, 3) nutrient imbalances, and 4) impacts on microorganisms (Foster 1989; Binkley et al. 1989). The effects of sulfur deposition have to be evaluated with respect to all factors known to influence the reaction of lodgepole pine trees. The long-term pattern in tree growth and health in affected and unaffected plots before and after the initiation of sour gas processing can be evaluated to assess the impact on stand productivity. The detection of episodic events, such as S<sup>0</sup> block fires or acute fumigations, which are of short duration, is unlikely using this process.

Two previous studies in west-central Alberta (Addison et al. 1984; Maynard 1990) assessed the chemistry of soils and vegetation as well as species richness and plant cover of the understory vegetation. Tree health was estimated by measuring tree basal area, needle length, weights and needle retention. The evaluation of tree health among sites was

inconclusive because of the large variation within sites and a paucity of information on the overall forest health in the region (i.e., insect and disease data). Increased needle loss of mature lodgepole pine at site 1 in 1985 suggested that tree health was being negatively impacted at sites contaminated by  $S^0$  (Maynard 1990).

In 1991, a 10-year assessment was carried out to remeasure the soil and foliar chemical properties originally measured in 1981 and again in 1985. The 1991 study was expanded in recognition of the complexity of environmental factors, both natural and anthropogenic, affecting forest health. Different methodology was used to reflect changes in the scientific approach to biomonitoring for acid deposition. A detailed forest health survey was initiated in 1991 as a result of evidence of indirect relationships between acid deposition and tree health. An intensive analysis of tree growth was initiated to address data gaps in the original study and to provide an historical perspective of tree health and pollution impacts.

The overall objectives of the 1991 study were to

- 1) assess changes in soil and foliar chemical properties measured in 1981, 1985, and 1991 at mature lodgepole pine sites and evaluate soil and foliar chemical properties of young stands of lodgepole pine and trembling aspen sites in relation to S deposition,
- 2) evaluate forest health conditions in relation to S deposition,
- 3) develop estimates of lodgepole pine tree growth and evaluate the variation in growth rates with respect to sulfur deposition in stands near two sour gas processing plants, and
- 4) compare the growth of young lodgepole pine (<25 years old) growing since the start-up of the sour gas processing facilities to the corresponding early growth period of currently mature lodgepole pine (80–120 years old).

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## Study Sites

The study included 31 sites established in mature, even-aged lodgepole pine dominated stands, 6 sites established in young lodgepole pine stands, and 3 sites established in mature trembling aspen (*Populus tremuloides* Michx.) stands. All the sites were located in an area 30×30 km with the two sour gas processing plants at the center (Fig. 1). The 31 mature lodgepole pine sites included 24 of the 26 sites established in 1981 (sites 1–26; Addison et al. 1984), 1 relocated site, and 6 new sites established in 1991. One site (15), established in 1981, was destroyed in an  $S^0$  block fire in 1984, and site 6 was relocated across Highway 752 to avoid grazing pressure from domestic animals. Site 17 proved unsuitable for a full sampling regime in 1991 because of its small size and proximity to disturbance; however, it was retained to preserve continuity and sampled on a more limited basis.

Six new, mature, and even-aged lodgepole pine sites (sites 30–35) were established in 1991 (numbers 27 to 29 were not used). They were added so that the study would more fully reflect the range of  $S^0$  and  $SO_2$  deposition levels in the forests around

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## MATERIALS AND METHODS

the two plants (see Addison et al. 1984 for selection criteria of original sites). Their locations were chosen on the basis of sulfation levels as measured by static sulfation stations and as predicted by stack plume dispersion models. Site 30 is located on a plateau about 4 km east of the Ram River plant and was selected because it falls within an area shown to have high levels of sulfation. Site 31, about halfway between the Ram River and Strachan plants, and site 33, east of Baseline Mountain, were both selected to represent a gradient in sulfation levels. Site 32 is located halfway up Baseline Mountain where the stack plume dispersion models predict periodic high levels of  $SO_2$  during inversions (Concord Scientific Corporation 1989). Site 34, located 200 m south of site 2 near the Strachan plant, was selected to represent an area with elevated  $S^0$  deposition and an intact understory stratum. Site 35, located above Highway 752, was selected due to the previously discussed limitations of site 17.

The six young lodgepole pine sites were established in stands <25 years old. The strategy was to select stands that became established within a few years of the commencement of emissions from the two gas plants and stands that also

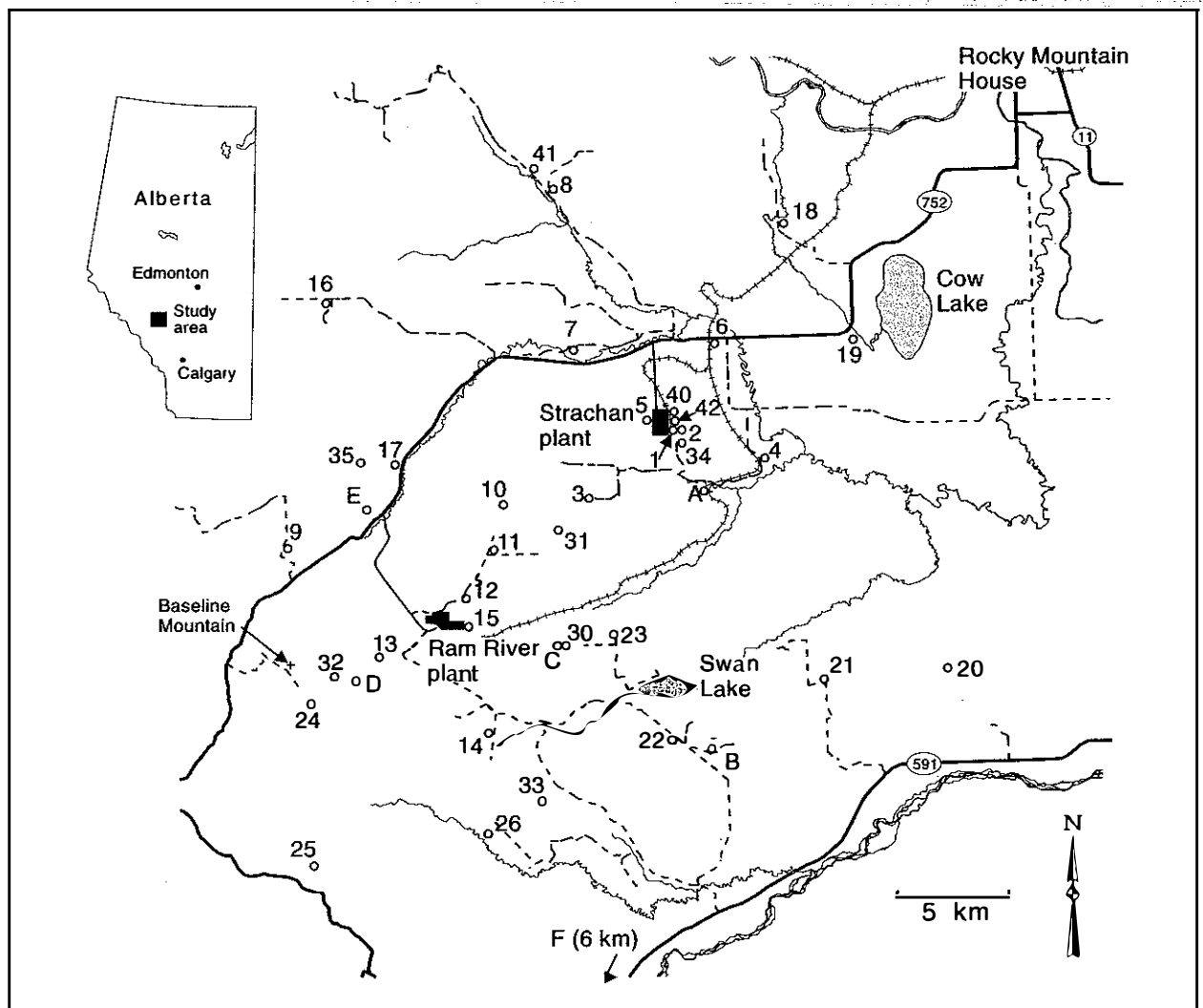


Figure 1. Location of biomonitoring plots (numbered and lettered) and sour gas processing plants in west-central Alberta.

represented a range of deposition levels. The sites were named A-F (Fig. 1).

Three trembling aspen sites (sites 40-42) were selected on the basis of  $S^0$  deposition levels. Only three sites were established due to the paucity of trembling aspen dominated stands in the study area. Site 42, located next to the Strachan sour gas plant and downwind of the  $S^0$  block, had a highly impacted understory, visible  $S^0$  in the surface organic horizon (LFH), and  $SO_2$  deposition  $>0.070$  equivalent (eq.)  $mg SO_3 100 cm^{-2} d^{-1}$ , and was thus selected as a high  $S^0$  impacted aspen stand. Site 40, located only 500 m north of site 42 adjacent to the Strachan  $S^0$  block, also had  $SO_2$  deposition  $>0.070$  eq.  $mg SO_3 100 cm^{-2} d^{-1}$ ; however it had an intact

understory stratum and lower  $S^0$  deposition. Site 41, located 12.5 km northwest of the Strachan gas plant, had no measurable  $S^0$  deposition and background levels of  $SO_2$  deposition.

Ten ( $50 \times 2$  m) quadrats were set up in the 30 mature pine-dominated sites (except site 17) and the three trembling aspen sites. Where possible the quadrats were placed 50 m apart and were at least 50 m from cut lines and other forest margins in order to maximize stand representation and minimize edge effects. Ten ( $10 \times 4$  m) quadrats were set up in the 6 young pine sites. The smaller quadrat size in the six young pine stands was used because of the much greater tree density in young pine stands relative to mature stands. The quadrats were



marked using tree paint and all trees within the quadrats were tagged.

## Climate

The study area is located in the transition zone between the Lower Boreal Cordilleran and Upper Boreal Cordilleran ecoregions (Strong and Leggat 1992) with cool, wet summers and mild winters relative to ecoregions in the rest of Alberta. The climate diagram for the Prairie Creek Ranger Station (Fig. 2) shows that over 70% of yearly precipitation falls during the growing season months,

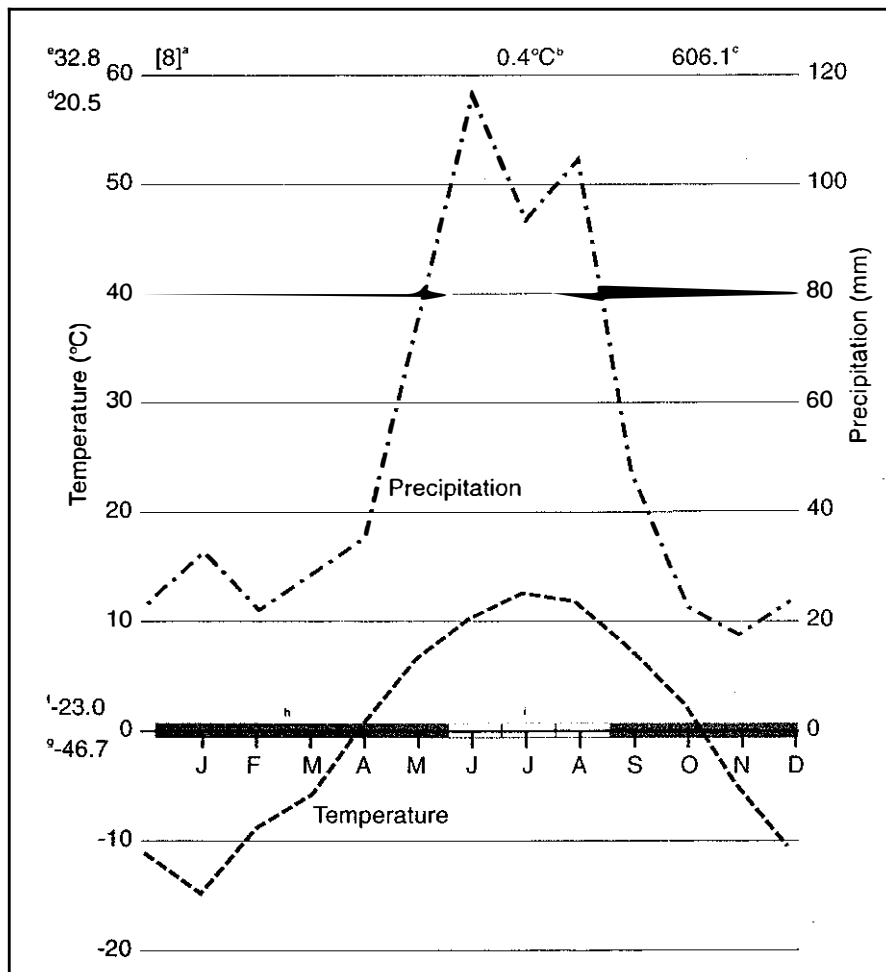
which, according to the temperature curve, is between May and September. Temperature and precipitation conditions during these months is a major factor affecting growth of trees and understory vegetation. A 25-year record of mean temperature and total precipitation for the growing season is shown in Figure 3. The Baseline Mountain lookout record is used because continuous meteorological record keeping was sporadic at the Prairie Creek Ranger Station. For the purposes of this study it is important to note the weather conditions of the 1981, 1985, and 1991 growing seasons and the growing season prior to each of these sampling years.

The years 1980 and 1981 had the wettest growing seasons of the last 25 years, and 1984 and 1985 had two of the four driest growing seasons over the same period. The year 1990 had the second wettest growing season in this period, and 1991 had about average precipitation. Mean growing season temperatures of the three sampling years were not markedly different.

## Soil and Foliar Chemical Analysis

### 1981, 1985, and 1991 Comparison

Soil chemical analyses were compared from the 25 sites sampled in 1981, 1985, and 1991. Soil samples were collected using the same method in all 3 years. Five replicates from the LFH and the top three mineral horizons were sampled. Four pits were placed at the corners of the 20×20-m permanent plots originally established in 1981. A fifth pit was located on the side facing the nearest gas plant. Samples were collected in plastic bags and stored at -20°C before processing.



**Figure 2. Climate diagram (Walter 1984) for Prairie Creek Ranger Station (1173 m).** a = years of observation, b = mean annual temperature, c = mean annual precipitation (mm), d = mean daily maximum temperature of warmest month, e = absolute minimum temperature, f = mean daily minimum temperature of coldest month, g = absolute minimum temperature, h = months with mean daily minimum temperature <0°C, i = months with absolute minimum temperature <0°C.

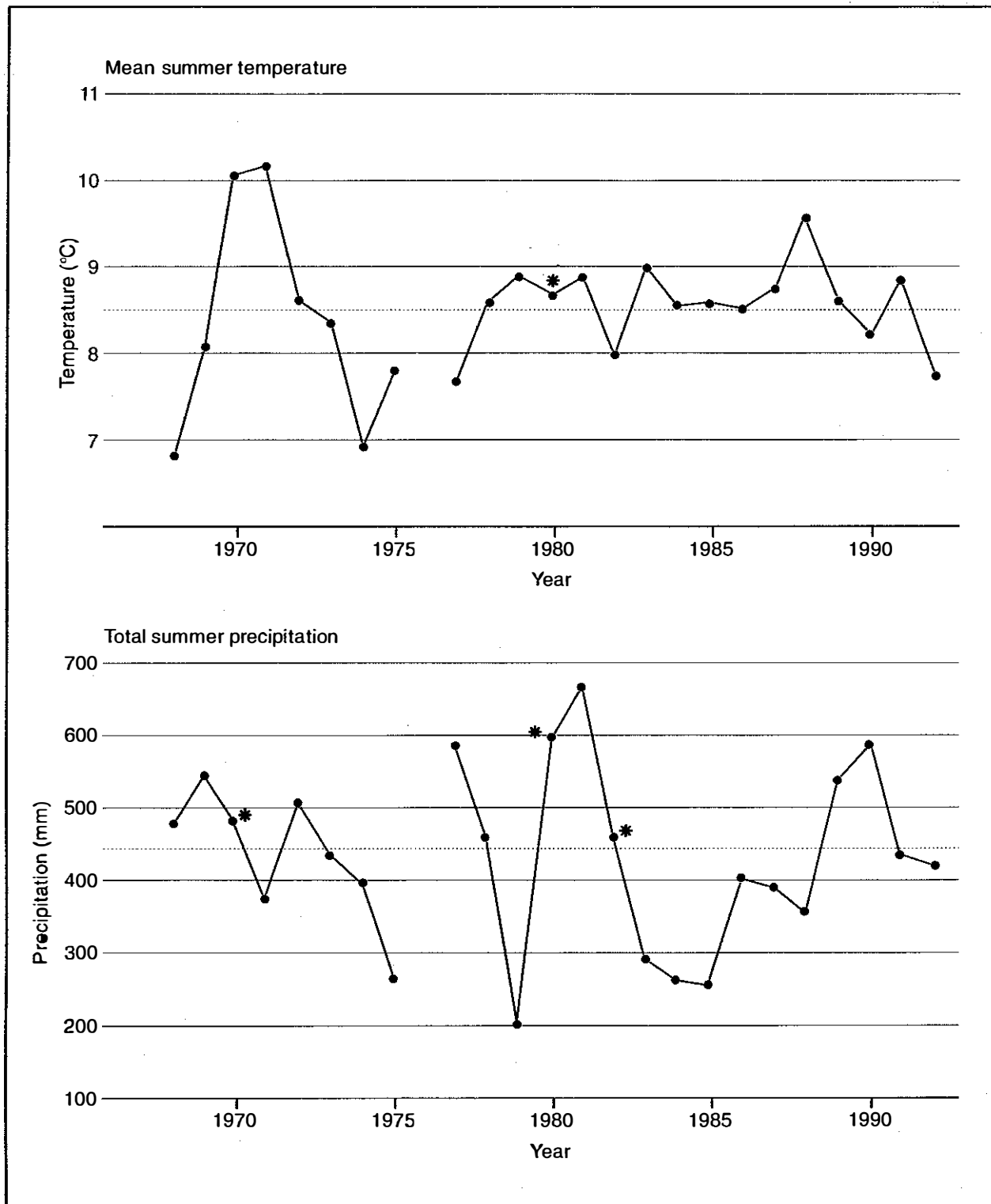


Figure 3. May to August precipitation and average temperature at the Baseline Mountain Lookout between 1968 and 1992. The dashed lines indicate precipitation and temperature normals based on 30 years of records for May to August conditions. Asterisk indicates an estimate of a year with missing records.

Elemental S and pH were measured using the same methods for all 3 sampling years. Elemental S and pH were determined on field-moist samples. Elemental S was measured only on LFH samples and was determined colorimetrically as a ferric thiocyanate complex following extraction with 95% acetone (Maynard and Addison 1985). The pH of LFH was determined in a 2:1 water to soil ratio (vol wt<sup>-1</sup>) after the soil was brought to saturated paste with water. The pH of mineral soils was determined in a 2:1 water to soil ratio (vol wt<sup>-1</sup>) (Kalra and Maynard 1991). In 1985 and 1991, pH was also measured using 0.01 M CaCl<sub>2</sub> in place of water.

Exchangeable cations were determined using different methods in 1981 and 1985. In 1991, air-dried soils were extracted with 1.0 M unbuffered ammonium chloride (NH<sub>4</sub>Cl) using a mechanical vacuum extractor. The extracts were analyzed for Ca, Mg, potassium (K), Al, iron (Fe), manganese (Mn), and S by inductively coupled plasma-atomic emission spectrometry (ICP-AES) (Kalra and Maynard 1991). Archived samples from 1981 and 1985, (kept at room temperature) were analyzed for exchangeable cations using the 1.0 M unbuffered NH<sub>4</sub>Cl method. Cation exchange capacity (CEC) was also determined on all soil samples from the 3 years except the 1985 LFH using the same NH<sub>4</sub>Cl method.

Total elemental analysis of LFH was determined on air-dried subsamples ground in a Wiley mill and passed through a 60-mesh sieve (0.25 mm). The 1981 and 1985 samples were digested with nitric acid-perchloric acid-hydrochloric acid (HNO<sub>3</sub>-HClO<sub>4</sub>-HCl) in an aluminum block digester (Hogan and Maynard 1984). The acid digestions were analyzed for S, P, Ca, Mg, K, Mn, Al, and Fe by ICP-AES. Only the 1985 samples were analyzed for Al by ICP-AES. The 1991 samples were digested with nitric acid-hydrogen peroxide-hydrochloric acid (HNO<sub>3</sub>-H<sub>2</sub>O<sub>2</sub>-HCl) in a microwave oven (Kalra et al. 1989) and analyzed for all the previously listed elements including Al by ICP-AES. A comparison of the two methods and analysis of certified National Institute of Standards and Technology (NIST) plant samples found the two digestion methods gave similar results within the certified values for the NIST standards (Kalra et al. 1989).

Total elemental analysis of the mineral horizons were determined on air-dried subsamples ground in a Spex mixer/mill (Spex Industries, Scotch Plains, New Jersey) and passed through a

60-mesh sieve. The same method was used for all years. Soils were digested with nitric acid-perchloric acid-hydrofluoric acid-hydrochloric acid (HNO<sub>3</sub>-HClO<sub>4</sub>-HF-HCl) followed by analysis by ICP-AES (McQuaker et al. 1979).

Total nitrogen (N), ammonium nitrogen (NH<sub>4</sub>-N), and nitrate nitrogen (NO<sub>3</sub>-N) were measured on the LFH samples in 1991 only. Total N was determined using a modified Kjeldahl digestion technique (Kalra and Maynard 1991). Samples were digested in an aluminum block digester using a sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and potassium sulfate-copper sulfate (K<sub>2</sub>SO<sub>4</sub>-CuSO<sub>4</sub>) catalyst mixture (Kjeltec). Total N was determined by distillation with an automated system (Tecator Kjeltec 1030).

Ammonium and nitrate were determined by an automated spectrophotometric method following extraction with 2.0 M potassium chloride (KCl) (Maynard and Kalra 1993). Samples were extracted moist in a 10:1 KCl to soil ratio (vol wt<sup>-1</sup>). No detectable NO<sub>3</sub>-N was found.

Ten replicate samples from 1-year-old lodgepole pine needles, 1-year-old white and black spruce needles (*Picea glauca* [Moench] Voss and *Picea mariana* [Mill.] B.S.P.), twin-flower (*Linnaea borealis* L.), and feather moss (*Pleurozium schreberi* [Brid.] Mitt.) were collected in 1981, 1985 and 1991. Green alder (*Alnus crispa* [Ait.] Pursh) leaves were collected in 1981 and 1991 only.

The lodgepole pine samples in 1981 were taken from the leaders of trees harvested for growth measurements. In 1985 and 1991 the samples were taken from the lower one-third of the crown. In 1981 and 1985 the trees were randomly sampled from the edge of the permanent 20 × 20-m plots. In 1991, one sample was taken from each of the 10 quadrats within the same stand. Pine foliage samples were collected between August and October for all 3 sampling years.

The spruce samples collected in 1981 and 1985 were collected from the leader of permanently marked trees. In 1991, samples from one tree per quadrat were collected. The permanently marked trees were too difficult to locate in 1991; consequently, the same sequence for sampling the lodgepole pine was done for the spruce. The spruce was sampled at the same time as the pine.

Ten replicates of the understory species were collected from a 15-m band surrounding each plot

in 1981 and 1985. Each replicate consisted of foliage from several individuals. In 1991, one replicate was collected from each quadrat.

Foliage samples were dried at 60°C, ground to pass through a 20-mesh (0.8-mm) screen in a Wiley mill. Total elemental analysis in the foliage followed the same procedures (microwave digestion and total N) as outlined for the LFH. The 1981 foliage samples were not analyzed for Al by ICP-AES.

### 1991 Mature Pine Sites

In 1991, six sites were added to the original network of 25 sites. A permanent 20×20-m plot was established at each of these new sites and soil samples were collected in the same way as outlined in the 1981, 1985, and 1991 comparison section.

Two LFH samples were collected from each of the ten (50 × 2 m) quadrats at 30 of the 31 mature pine sites (site 17 was excluded). The 20 additional LFH samples were collected to provide a more representative measure of nutrient concentrations for comparison with foliar chemical analysis, the forest health survey, and tree growth. These data were used to classify the sites by deposition class and to compare the total and extractable concentrations in the LFH among deposition classes. The soil chemical analysis followed the same methods described for the original plots.

In 1991, current foliage of lodgepole pine and spruce were collected at all 30 plots. The collection, sample preparation and analysis were completed the same as for the 1-year-old foliage collected in 1991. Two additional understory species, bunchberry (*Cornus canadensis* L.) and bog cranberry (*Vaccinium vitis-idaea* L.) were collected in 1991 at all 30 plots. Bunchberry and bog cranberry are common understory species of these forest ecosystems that disappeared rapidly from sites acidified by the deposition and oxidation of S<sup>0</sup> (Kennedy et al. 1988). Sample collection, preparation and analysis was the same as for the other understory species collected in 1991.

### 1992 Young Pine and Mature Aspen Sites

In 1992, soils of the six young lodgepole pine sites and three mature aspen sites were sampled for chemical analysis. The sampling and analysis of these sites followed the same procedures as outlined for the 1991 mature pine sites.

In 1992, current and 1-year-old lodgepole pine needles were collected from 10 young trees (<25 years old). Current and 1-year-old needles were analyzed separately. One replicate was collected from the edge of each 4 × 10-m plots established in the six stands. Five of the most dominant understory species, bunchberry, prickly rose (*Rosa acicularis* Lindl.), fireweed (*Epilobium angustifolium* L.), twin-flower, and feather moss were collected from each stand. Ten replicates (each replicate made up of foliage from several individuals), one from each of the ten (4 × 10 m) plots were collected (except prickly rose at site C was not present in one quadrat).

Leaves of trembling aspen were collected from ten randomly selected trees. One replicate was collected from each of the 10 quadrats at the three aspen sites. Up to 10 samples, one from each quadrat, of two understory species, bunchberry and fireweed were collected at all three sites. Bunchberry was present in only 4 of the 10 quadrats at site 42, the most heavily impacted site. Twin-flower and wild strawberry (*Fragaria virginiana* Duchesne) were collected at the background (site 40) and moderately S<sup>0</sup> contaminated site (site 41). These species were not present at site 42. Other understory species sampled at one or two of the aspen sites were green alder (sites 40 and 42), wild sarsaparilla (*Aralia nudicaulis* L.) (site 42), and willows (*Salix* L. spp.) (sites 41 and 42). These species were not present in every quadrat at site 42. The foliage samples collected in 1992 were prepared and analyzed using the same method as in 1991.

## Forest Health Survey

A forest health survey was conducted on the same sites in June and July, 1991, at the ten (50 × 2 m [100 m<sup>2</sup>]) quadrats. Forest health surveys of young lodgepole pine stands and the three trembling aspen sites were conducted in July of 1992.

Trees that were in the plots were examined individually for health condition and classified as either healthy, declining, recently dead or dead more than one year. This was based on the condition of the foliage in the crown. If there was no foliage in the crown the tree was considered dead more than 1 year. Trees with present but entirely red or brown foliage were considered recently dead. Trees with a full complement of green foliage were considered to be healthy. Trees with ≥50% foliage missing were considered to be declining.

Upper crowns and stems of trees were examined for evidence of insects and disease or mechanical injury. Crowns, stems, and butts of trees were examined for wounds, symptoms, and signs of disease causing agents or insects. The information was recorded and a diagnosis of the responsible agent was made wherever possible. The root collar region of trees that were recently dead was examined for evidence of root collar weevil (*Hylobius warreni* Wood) or root rotting fungi. Healthy and declining trees were not destructively sampled. Symptoms and disease or insect diagnosis were coded and recorded in a database.

## Pine Growth

### Measures of Tree Productivity

Tree growth was assessed by stem analysis. By counting and measuring annual rings on sections taken at different intervals along tree stems, tree growth through time can be reconstructed. Several characteristics of tree growth can be measured from these observations. Each characteristic reacts to internal and external conditions affecting the tree. Three measurements derived from these observations are particularly useful in evaluating external influences on trees: an index of tree growth based on radial increments at breast height, an index based on the annual volume increment, and an index based on the annual specific volume increment.

The first measurement is the radial growth of the trees at some fixed point along its stem (called a horizontal sequence at a specified height from the base of the tree). This sequence is most easily obtained by extracting an increment core from the stem. Stump height (30 cm from the ground) or breast height (1.4 m from the base) is used to make this assessment. This is the most inexpensive and non-destructive sampling method; however, it is not without disadvantage. The sensitivity of these measurements decreases with increasing tree age. To control for the effects of age, Fritts' (1976) procedure was adapted for the present study. The effects of tree age were discounted by fitting a negative exponential curve to the original data thus tree age was not controlled in the analysis of data obtained from increment cores. The mean index of tree growth based on increment cores taken from trees in each plot was assigned a value of 1.00 for the decade prior to plant openings. This mean index was divided into the mean for each of the two decades following the opening of the plants.

The other two indexes were based on stem analyses and used to evaluate the effects of S deposition on tree growth. Measurements of annual volume increment and specific volume increment were both calculated from the same trees and the differences obtained reflect the variability of these measurements in detecting the impacts of environmental conditions on tree growth. As in the treatment of the increment core data, the mean growth of the tree prior to the opening of the gas plants was taken as the basis for comparison. The mean growth of trees for each of the two decades following the opening of the plants divided by the mean for the decade prior to plant opening provides an index of performance. In this case, however, the individual tree growth was used as the observation unit, rather than the mean for the whole plot. The analysis of variance used, therefore, partitioned variation observed in the index analyzed respectively among tree age, to control for age effects; deposition class; the site, nested within deposition class; the decade in which growth occurred, relative to the gas plant openings; and the interaction between decade and deposition class. The annual volume increment is simply the volume of wood applied to the stem in a single year. It has characteristics similar to specific volume increment, which is discussed next.

The third of these measures is the specific volume increment, the average volume growth per unit area of cambium. The cambium area is calculated as the surface area of the tree at the beginning of the year in question. (Dimensional analysis of this measure reveals that it is also the radial increment averaged over the whole stem: it is reported in units of length.) The specific volume growth is also the average radial growth rate of the tree. This measure is costly to obtain but more accurately reflects environmental changes than radial increments taken from the boles of trees. Specific volume increment estimates also appeal because they integrate the effects of factors over the whole stem.

The value used for comparisons between newly established stands and stands established prior to the gas plant opening was the cumulative volume growth to age 11 years. Age eleven was used because it is the age of the youngest newly established stand. The analysis of variance examined the influence of the following on cumulative volume growth to age eleven: deposition class in which the stems now grow; the era relative to the operation of the gas plants in which the tree grew in the juvenile state; and the interaction between deposition class and the era of early growth.

## Field and Laboratory Procedures

Two co-dominant trees were selected for stem analysis from each of the mature lodgepole pine study sites. The trees were adjacent to each of two randomly selected plots (the 50 × 2-m quadrats) within the study stand. The trees were selected to be free of defect and representative of stand conditions near the plot. In addition, an increment core at breast height was obtained from one co-dominant tree from each of the remaining eight plots within the stand.

Trees selected for stem analysis were felled, heights and dbh measured, and disks removed from the base and from every metre along the stem. These disks were returned to the lab and stored in a freezer until prepared for measurement. The disks were prepared for measurement by sanding the surface and determining the average diameter of the disk with a diameter tape. Annual increments along an average radius were measured from the bark to the pith using a Digimicrometer Model MK-1B (DIGIMIC) machine. These data were inspected to detect missing rings. An inputted value of 0 was inserted at the appropriate location in the tree ring sequence using cross dating to account for missing rings.

Various measures of tree growth were calculated from the tree ring data. The horizontal sequence on a radius at 1 m was obtained from the rationalized data obtained from that disk. Height growth at specific ages was obtained by linear interpolation between adjacent disks of known heights and ages. Volumes were calculated as the sum of the volumes of a cone (at the apex) and the frusta of cones 1 m in height corresponding to each sample interval between disks. Specific volume increments were obtained by dividing the calculated volume increment by the surface area of the tree (also calculated as the sum of areas of an apical cone and the associated frusta of cones). The cumulative volume was obtained by summation over the years of tree growth.

The results of the analyses on each tree were plotted to display the horizontal sequence at 1 m, the specific volume growth, the annual volume growth, the oblique sequences for 5-year periods starting in 1991 and going back for five periods, the cumulative height growth, and the cumulative volume growth.

Annual rings in the increment cores obtained from the other eight plots in each stand were measured using the DIGIMIC. The data from each

core was standardized using Fritts' (1976) procedure. The oldest five increments were deleted and a negative exponential curve was fitted to the remaining ring width measurements. An index of tree growth was obtained by dividing the fitted curve value to the observed value for that year. The mean index derived from the index of each of the eight cores from the site provides a chronology for the site.

## Analysis

Data on tree growth from all sites were grouped according to S deposition classes derived from the chemical analysis of soils and SO<sub>2</sub> deposition. The trend in tree growth of all trees in the decade prior to the opening of the gas plants was assumed to be indicative of pristine growth rates. Trees at unaffected sites provided an indication of annual growth, controlled for tree age and location, spanning the 20 years of sour gas processing in the region. These trends were compared to those of trees growing on the affected sites before and after the beginning of sour gas processing.

This analytical procedure provides a means for overcoming many of the local and temporal trends that would tend to obfuscate the effects of sulfur deposition on tree growth. The growth in the decade prior to plant openings provides the basis for comparison, within sites, of tree performance following the opening of the plants. Tree age was used as a covariate in the analysis. The ratio of growth before and after plant openings, controlled for tree age, should be the same in all deposition classes if there were no impact.

## Statistics

The 1981, 1985, and 1991 soil and foliar chemical data were analyzed by year and site with a two-way analysis of variance (ANOVA) using the general linear model (GLM) procedure of the Statistical Analysis System (SAS) Institute, Inc (1990). Least square means (LS-means) were used for comparisons where significant effects were detected by the two-way ANOVAs. Data from the 1991 samples taken from mature pine stands were analyzed by deposition class and site (deposition class) using GLM. Data from 1991 samples of young pine stands and aspen stands were analyzed separately by a one-way ANOVA, mean separations were calculated and the significance of these differences assessed by the Tukey test (Zar 1984).

Chi-square analyses were used to test the independence of tree species frequencies in the

various health classes and the deposition classes. Spearman's ranked correlation tests were used to determine correlations in the forest health survey data because the Spearman's test is non-parametric and requires no assumptions regarding the distribution of the data (Zar 1984).

The complete data summary set is presented in a separate Appendix available from the senior author. Only results directly cited and relevant to the discussion are presented in the main body of this report.

## RESULTS AND DISCUSSION

### Pollution Distribution

The Ram River and Strachan plants were, respectively, the largest and tenth largest sour gas processing plants in Alberta during the period of this study (1981-92). Together they process and recover about 3000 t d<sup>-1</sup> of S<sup>0</sup>. In 1981, despite S recovery rates of 98% at Ram River and 97% at Strachan, the stack emissions from both plants totalled about 70 t d<sup>-1</sup> of S. By 1985 S emissions were reduced to 56 t d<sup>-1</sup> and by 1991, with declining throughput and improved S recovery, the S emissions declined to 25 t d<sup>-1</sup> (Fig. 4).

### Sulfation

Sulfur dioxide is the largest component of the S emissions and is the major emission from an acid deposition perspective. Hydrogen sulfide (H<sub>2</sub>S), carbon dioxide (CO<sub>2</sub>), water, nitrous oxides (NO<sub>x</sub>), trace hydrocarbons, metals, and other S gases are also emitted. The incinerator stacks at both plants disperse SO<sub>2</sub> over a wide area. Wind patterns and topography strongly influence the deposition pattern of SO<sub>2</sub> (Concord Scientific Corporation 1989) (Fig. 5). Both Husky Oil and Gulf Canada Resources maintain networks of static sulfation

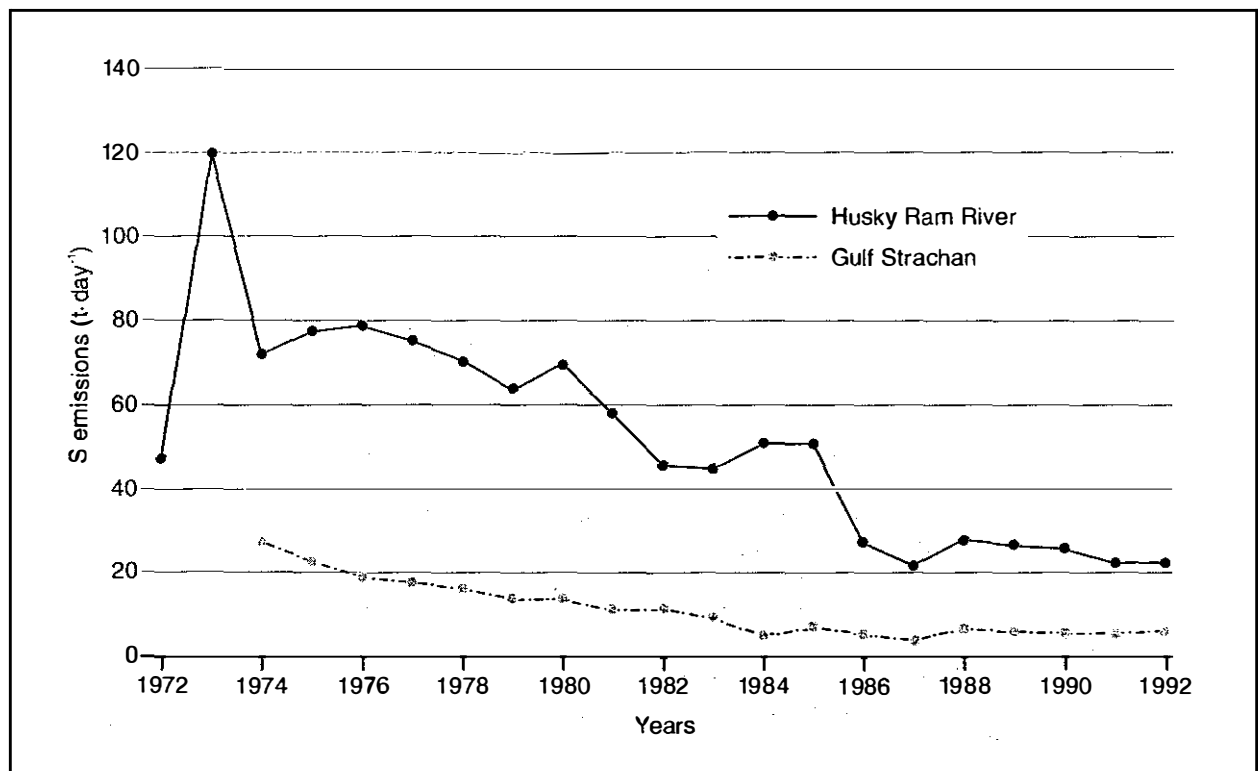


Figure 4. Sulfur dioxide emissions from the incinerator stacks of the Ram River and Strachan sour gas processing plants (1972-92).

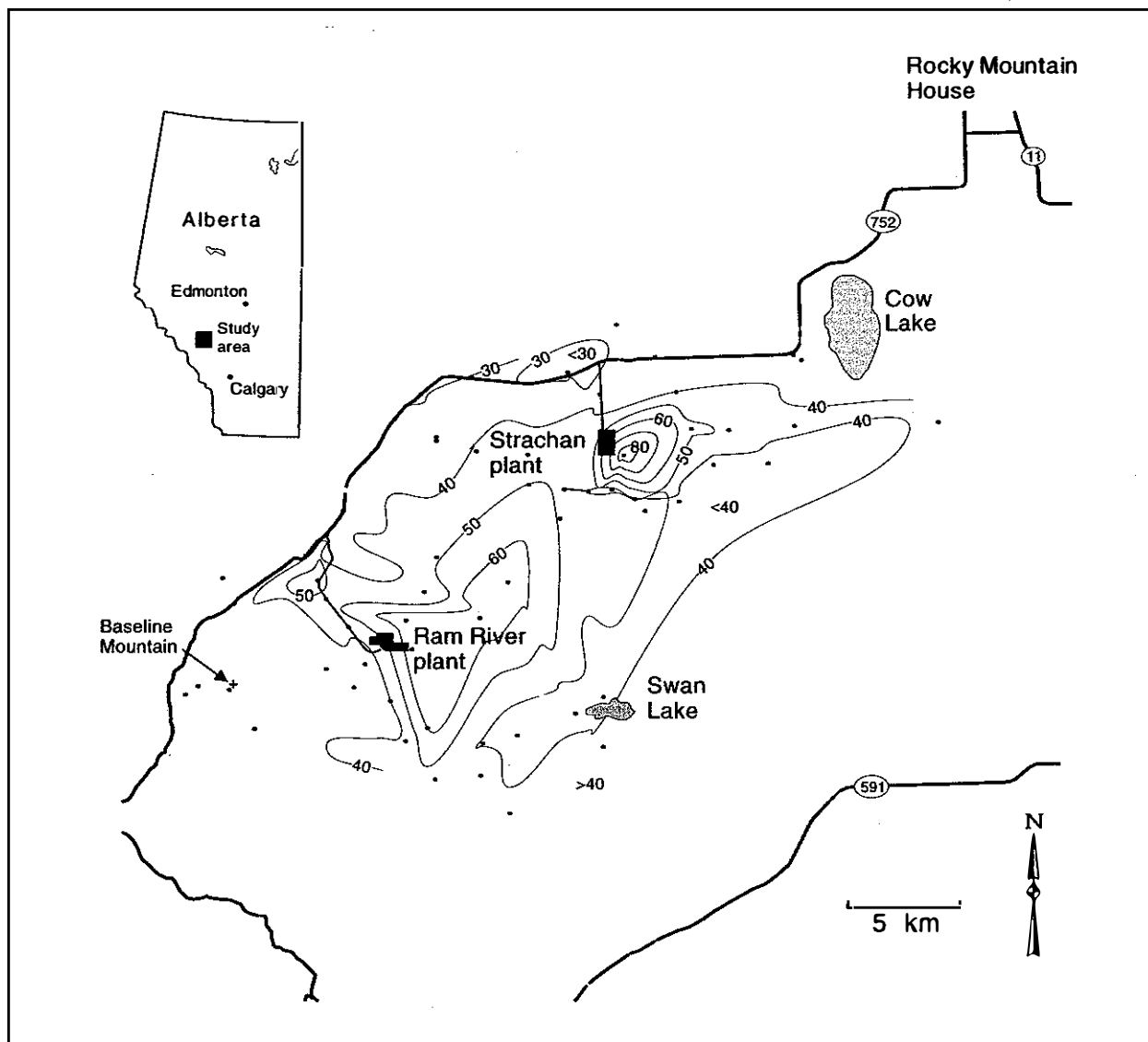


Figure 5. Isopleths of average sulfation rates near sour gas processing plants in west-central Alberta, 1975–86. Contour units are equivalent  $\text{mg SO}_3 \text{ 100 cm}^{-2}\text{d}^{-1}$  ( $\times 1000$ ).

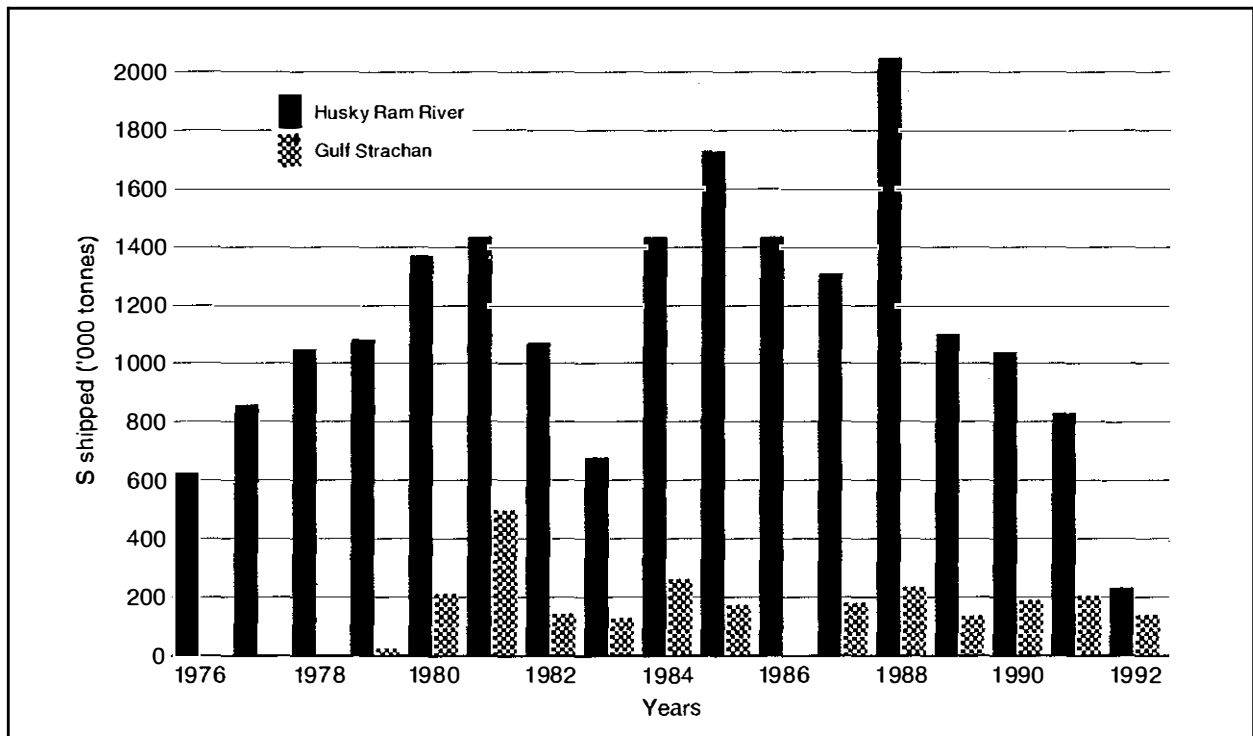
stations surrounding their gas plants, and  $\text{SO}_2$  and  $\text{H}_2\text{S}$  deposition was measured on a monthly basis between 1975 and 1986. Measurements from static sulfation stations taken between 1975 and 1986 indicated high  $\text{SO}_2$  deposition rates east of the Strachan plant and over an area ranging from northeast to southeast of the Ram River plant. The  $\text{SO}_2$  deposition data after 1986 could not be used in calculating mean deposition rates due to a change in static sulfation station methodology in 1987 for the Gulf sulfation stations. Plume dispersion models predict high episodic  $\text{SO}_2$  deposition on the northeast slopes of Baseline Mountain southwest of the Ram River plant (Concord Scientific Corporation

1989). Fugitive emissions from the two plants and emissions from the flare stacks may contribute significantly to overall gaseous emissions; however, the level of these emissions have not been quantified.

### Elemental Sulfur Particulate

The recovered  $\text{S}^0$  is shipped by rail as either liquid or formed product; however, when production exceeds demand,  $\text{S}^0$  is stored in S blocks. Mechanical breakup of these  $\text{S}^0$  blocks for shipping resulted in extensive dusting of nearby downwind forested areas. This occurred at the Strachan plant in 1980 and 1981 (shipping began in 1979; Fig. 6)





**Figure 6. Formed sulfur shipped from Ram River and Strachan sour gas processing plants (1976–92).**  
 Note: Amount of sulfur shipped from Gulf Strachan in 1986 is unavailable.

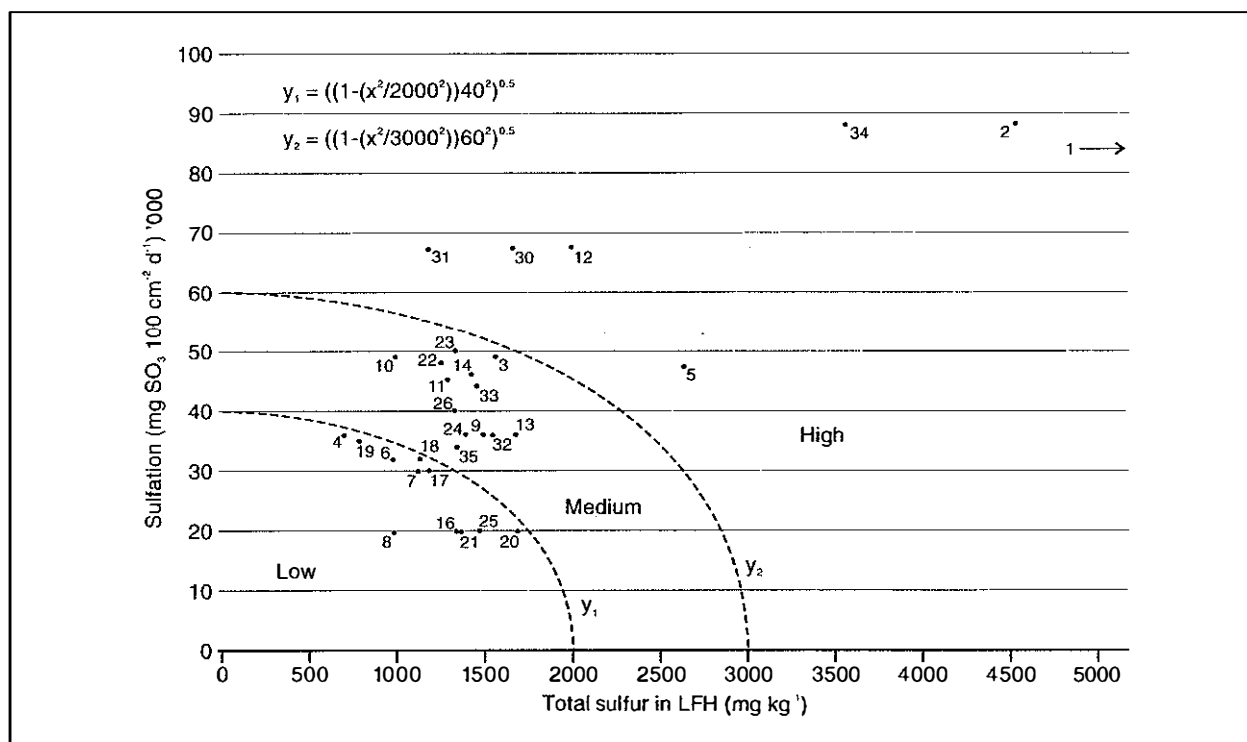
and resulted in heavy  $S^0$  dusting of the forest stand downwind (east) of the S block. The majority of the  $S^0$  (accumulated  $S^0$  deposition was in the  $t\ ha^{-1}$  range) was deposited within 1 km of the plant. The removal of this block by the mid 1980s resulted in a reduction in  $S^0$  dusting. Similar dusting also occurred near the Ram River  $S^0$  block; however, the study of this impact was prevented by a block fire that destroyed the adjacent stand and study plot in 1984. Large-scale shipment of stored S from Ram River began in 1986 and ended in 1989. Both companies conduct a yearly soil sampling program to document S deposition within a 1- to 2-km radius of the two gas plants. The results of the Ram River program (HBT AGRA 1993) indicate that the area of  $S^0$  deposition peaked in 1988 when the greatest amount of formed  $S^0$  was shipped. The introduction of alternate S handling methods, such as Ellithorpe remelters, has resulted in reductions of dustings at both gas plants. These methods, however, have not completely replaced mechanical breakup. Despite reductions in dusting,  $S^0$  may have been dispersed over a wider area through the introduction of prilling towers and S vapor off gases from the Ellithorpe remelters. Rail loading areas are additional sources of  $S^0$  dust at each plant.

### Liming

Both companies have liming programs involving the aerial application of lime ( $CaCO_3$ ) in the areas immediately surrounding the two plants. The study sites affected by liming were 1, 2, 5, and 34, near the Strachan plant, and site 12, near the Ram River plant. From 1981 to 1984 sites 1, 2, and 5 were excluded from the regular liming programs for the purposes of the original study (Addison et al. 1984; Maynard 1990). Since 1985 these sites have been included again in the regular annual liming programs.

### Classification of Stands

All mature pine sites were assigned to deposition classes based on  $SO_2$  deposition (estimated from the nearest static sulfation station[s]) and total S in the LFH (Fig. 7). The  $SO_2$  deposition rates were based on mean deposition between 1975 and 1986, and total S in the LFH was based on quadrat samples taken in 1991. The high deposition class included seven sites, including six sites with detectable  $S^0$  ( $148\text{--}7490\ mg\ S^0\ kg^{-1}$ ) in the LFH (sites 1, 2, 5, 12, 30, and 34). Site 31 had no measurable  $S^0$  but was located in an area of high sulfation ( $0.070\ eq.$



**Figure 7. Classification of mature lodgepole pine study sites on the basis of sulfation and total sulfur in the LFH.** The  $y_1$  function curve separates the low and medium deposition classes and the  $y_2$  curve separates the medium and high impact classes.

mg  $\text{SO}_3$  100  $\text{cm}^{-2}$   $\text{d}^{-1}$ ) and was thus included in the high deposition class. The medium deposition class was represented by a cluster of 13 sites with lower total S in the LFH and sulfation levels between 0.035 and 0.055 eq. mg  $\text{SO}_3$  100  $\text{cm}^{-2}$   $\text{d}^{-1}$ . This class represented sites not heavily affected by particulate  $\text{S}^0$  deposition but located in areas where gaseous  $\text{SO}_2$  deposition was higher than background levels. The remaining 11 sites fall within the low deposition class where sulfation and total LFH S most closely approximated background levels (sulfation  $<0.035$  eq. mg  $\text{SO}_3$  100  $\text{cm}^{-2}$   $\text{d}^{-1}$ ). Sulfation rates at sites 4 and 19 slightly exceeded the 0.035 eq. mg  $\text{SO}_3$  100  $\text{cm}^{-2}$   $\text{d}^{-1}$  threshold; however, these sites were still included in the low deposition class because of their very low concentrations of total S in the LFH.

Static sulfation stations using lead dioxide cylinders have a higher affinity for  $\text{SO}_2$  and other S gases than vegetation and soil surfaces (Noël et al. 1989). Dry sulfate ( $\text{SO}_4$ ) deposition onto soil and vegetation would be about 30% of the measured  $\text{SO}_4$  deposition onto lead dioxide cylinders in static sulfation stations (Leahey and Schroeder 1985). Thus the 0.035 eq. mg  $\text{SO}_3$  100  $\text{cm}^{-2}$   $\text{d}^{-1}$  threshold

deposition rate between the medium and low deposition classes is equivalent to 4.4 kg  $\text{SO}_4$   $\text{ha}^{-1}$   $\text{yr}^{-1}$ .

The six young lodgepole pine stands were classified using the previously described criteria into high, medium, and low deposition classes for the purposes of the forest health survey. Site C was classified as a high deposition site; sites B, D, and E were classified as medium deposition sites; and sites A and F were classified as low deposition sites.

## Description and Composition of Stands

### Mature Stand Survey

A total of 5715 trees were examined on the 31 mature pine sites. Lodgepole pine was the dominant species comprising 83% of all the trees on all the sites (Table 1). Black spruce, white spruce, and trembling aspen comprised 7, 5, and 5%, respectively of all the trees on all the sites. Balsam fir (*Abies balsamea* [L.] Mill.) and balsam poplar (*Populus balsamifera* L.) made up a very small proportion of the

**Table 1. Number and proportion of trees by major tree species in the three deposition classes of the mature pine stands, in the young pine stands, and in the mature aspen stands**

Stand type	No. of sites	Lodgepole pine		White spruce		Black spruce		Trembling aspen		Total trees <sup>a</sup>
		No. of trees	% of all trees	No. of trees	% of all trees	No. of trees	% of all trees	No. of trees	% of all trees	
Mature pine	31	4 742	83.0	300	5.0	385	7.0	271	5.0	5 715
High	7	1 073	85.0	49	4.0	3	0.2	133	11.0	1 259
Medium	13	2 347	90.0	100	4.0	120	5.0	21	1.0	2 595
Low	11	1 322	71.0	151	8.0	262	14.0	117	6.0	1 861
Young pine	6	1 906	96.0	1	0.1	3	0.2	71	4.0	1 983
Aspen	3	39	10.0	20	5.0	0	0.0	306	79.0	385

<sup>a</sup> Total trees includes tree species not shown in the table.

total tree population, 0.23 and 0.07%, respectively. Species such as willow and green alder were noted but not included in the health assessment. There were 1259 trees on the high S sites, 2595 trees on the medium S sites, and 1861 on the low S sites. Lodgepole pine was the dominant species on all three deposition classes: high (85%), medium (91%), and low (71%). Black spruce was relatively more common in the low S sites (14%, versus 0.2% and 5%, respectively at the high and medium S sites); however, the bulk of this black spruce was concentrated at site 18, which was surrounded by wetlands. Trembling aspen was relatively more common in the high sites (11%, versus 1 and 6%, respectively in the medium and low S sites); however, again the bulk of this trembling aspen was concentrated in the 3 sites immediately east of the Strachan plant (sites 1, 2, and 34).

Species composition of these sites was most likely related to stand origin, which on the eastern slope of the Rocky Mountains is primarily fire. Lodgepole pine is a fire species (one that requires fire or intense heat to regenerate) and thus it was the predominant species in the study area. Black spruce was found on wetter sites in the study area. Trembling aspen was not found frequently. This may be related to the age of the stands as aspen is not a long-lived species. The low frequency of white spruce was most likely related to its late successional nature. Analyses and discussion of health condition and damage was confined to the major species present at the study sites: lodgepole pine, white and black spruce, and trembling aspen.

The average ages of lodgepole pine trees (determined by stem analysis) growing on low S sites

were 92.9 ( $\pm 27.4$ ) years, medium S sites 109.0 ( $\pm 18.6$ ) years, high S sites 82.8 ( $\pm 21.7$ ) years. An analysis of variance was performed on tree age by sulfur class. There was a statistically significant difference ( $p < 0.05$ ) in ages of lodgepole pine trees growing on the different sulfur class sites. A least-square-means multiple comparison test showed that there was a statistically significant difference ( $p < 0.05$ ) between the ages of lodgepole pine trees growing on the high S sites and the medium S sites. There was no statistically significant difference between the ages of trees on the high S sites and those on the low S sites, nor was there any significant difference in the ages of trees on the medium S sites and the low S sites. Ages of other tree species growing on these sites were not determined.

### Aspen Stand Survey

Three mature aspen sites were surveyed. Trembling aspen was dominant in all three stands comprising 80, 62, and 91% of all trees in the high, moderate, and non-S<sup>0</sup> impacted sites, respectively (sites 42, 40, and 41). Balsam poplar was also present in the moderate S<sup>0</sup> site, comprising 18% of all trees. Lodgepole pine comprised 14, 13, and 5% of all trees in the high, moderate, and non-S<sup>0</sup> sites, respectively. White spruce comprised 5, 8, and 3% of all trees in the high, moderate, and non-S<sup>0</sup> sites, respectively. Ages of aspen in these stands were not determined; however, the stands were considered to be mature.

### Young Stand Survey

Six young lodgepole pine stands were surveyed for health conditions. Lodgepole pine was

the major species observed in all of these stands (Table 1). Trembling aspen made up a small proportion of these stands and white spruce, black spruce and balsam fir were rarely found. The average age of lodgepole pine was 14.5 years old on the low S sites (sites A and F), 14 years old on the medium S sites (sites B, D, and E), and 10 years old on the high S site (site C).

## Soil and Foliar Chemical Analysis

### Sulfur

Elemental S concentrations in the LFH at sites 1 and 2 in 1991 were <20% of the 1981 concentrations (Table 2). A similar decrease in total S was also observed (Table 3). Elemental S deposition at sites 1 and 2 has decreased between 1981 and 1991; however, S concentrations at these sites remain the highest of any site. Improved methods of S<sup>0</sup> handling and S<sup>0</sup> block removal have reduced S<sup>0</sup> deposition at these sites. In addition, the southern S<sup>0</sup> block next to sites 1 and 2 was removed in the mid- to late-1980s, and the majority of S<sup>0</sup> handling activity was at the northern S<sup>0</sup> block and the load-out area. This also contributed to a reduction in S<sup>0</sup> deposition at sites 1 and 2.

The S<sup>0</sup> concentrations at site 5 and 12 decreased between 1981 and 1991 but not to the same extent as at sites 1 and 2. Total S concentrations increased and extractable S concentrations did not change. The changes in S concentrations observed in 1991 indicated the S<sup>0</sup> oxidation rate exceeded the S<sup>0</sup> deposition rate with a portion of the oxidation products of S<sup>0</sup> being retained in the LFH in an organic form. The differences in S<sup>0</sup> deposition of sites 1 and 2 versus sites 5 and 12 reflect the variable distribution of S<sup>0</sup> as a result of changes in the location and methods of S<sup>0</sup> handling activity and amounts of S<sup>0</sup> shipped.

Sites 3 and 4 in 1985 and site 14 in 1991 had S<sup>0</sup> in the LFH near the detection limit (100 mg kg<sup>-1</sup>; Table 2). One of the five LFH samples at site 13 also had measurable S<sup>0</sup>. In 1991, no measurable S<sup>0</sup> was found at sites 3 and 4. The measurable S<sup>0</sup> found in 1985 at sites 3 and 4 could have been an artifact of the methodology; however, the deposition rate between 1985 and 1991 could also have been below the S<sup>0</sup> oxidation rate and thus, S<sup>0</sup> in the LFH would not be detectable.

Two of the six sites (sites 30 and 34) established in 1991 had measurable S<sup>0</sup> in the LFH and higher than expected total S (Tables 2 and 3). Site 34 was

**Table 2. Elemental sulfur concentrations of the LFH horizon at selected mature pine sites in 1981, 1985, and 1991. Values are means ±95% confidence limits (n = 5).**

Site <sup>a</sup>	Deposition class	Elemental S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	43 500 ± 12 700	51 300 ± 19 100	7 490 ± 6 480
2	High	14 600 ± 14 300	7 860 ± 7 440	870 ± 620
5	High	581 ± 452	3 360 ± 1 350	3 260 ± 317
12	High	B.D. <sup>b</sup>	361 ± 55	148 ± 59
30	High	N.D. <sup>c</sup>	N.D.	176 ± 25
34	High	N.D.	N.D.	819 ± 234
3	Medium	B.D.	128 ± 50	B.D.
13	Medium	B.D.	B.D.	B.D. <sup>d</sup>
14	Medium	B.D.	B.D.	100 ± 11
23	Medium	B.D.	B.D.	B.D.
4	Low	B.D.	155 ± 35	B.D.

<sup>a</sup> All other sites had elemental sulfur concentrations below the detection limit of the method.

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

<sup>c</sup> N.D. = not determined (sites established in 1991).

<sup>d</sup> One of five replicates had detectable S<sup>0</sup> in the LFH (138 mg kg<sup>-1</sup>).

**Table 3. Total sulfur concentrations of the LFH horizon at selected mature pine sites in 1981, 1985, and 1991.** Values are means  $\pm$  95% confidence limits (n = 5).

Site	Deposition class	Total S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	58 600 $\pm$ 12 900a	48 000 $\pm$ 12 400b	12 550 $\pm$ 6 750c
2	High	16 030 $\pm$ 11 600a	11 000 $\pm$ 5 750b	2 900 $\pm$ 1 170c
5	High	2 340 $\pm$ 624c	3 810 $\pm$ 1 200b	5 430 $\pm$ 2 440a
12	High	1 010 $\pm$ 366b	1 440 $\pm$ 243ab	1 710 $\pm$ 171a
30	High	N.D. <sup>a</sup>	N.D.	3 550 $\pm$ 633
34	High	N.D.	N.D.	1 650 $\pm$ 100
3	Medium	1 150 $\pm$ 240a	945 $\pm$ 195a	1 210 $\pm$ 298a
13	Medium	920 $\pm$ 503b	1 044 $\pm$ 174b	1 666 $\pm$ 104a
14	Medium	1 080 $\pm$ 390b	1 040 $\pm$ 184b	1 500 $\pm$ 288a
23	Medium	1 265 $\pm$ 239b	952 $\pm$ 112c	1 590 $\pm$ 172a
20	Low	865 $\pm$ 232b	1 050 $\pm$ 110b	1 340 $\pm$ 389a
25	Low	1 040 $\pm$ 190ab	970 $\pm$ 273b	1 320 $\pm$ 288a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

<sup>a</sup> N.D. = not determined (sites established in 1991).

located about 200 m south of site 2. There was no obvious impact on the understory. This is consistent with the earlier observation that visible impact to the understory was limited to areas with  $>5000$  mg kg<sup>-1</sup> of S<sup>0</sup> after 5–10 years of exposure in the LFH horizon (Maynard 1990). Site 30 is about 4 km east of the Ram River load-out area. Between 1985 and 1989 the S<sup>0</sup> block at the Ram River plant was broken up and the prilling tower was used extensively (D. McCoy, personal communication, telephone interview, March 7, 1994). This could have resulted in a wider distribution of S<sup>0</sup> at deposition rates greater than the oxidation rate of S<sup>0</sup> in the LFH.

The total S concentrations in the LFH of 16 of the 21 non-S<sup>0</sup> sites did not change between 1981 and 1991. The total S concentrations in the LFH of sites 13, 14, and 23 (medium S deposition class) increased by 50% in 1991 (Table 3) suggesting the deposition and retention of considerable amounts of S. Total S concentrations at these sites were 30–40% above the mean S concentrations for the rest of the sites in 1991. Sites 20 and 25, located in the low deposition class, also had increased S concentrations in the LFH in 1991 compared to 1981 but were only slightly higher than the mean S concentrations in the LFH for all non-S<sup>0</sup> sites. These observed increases between 1981 and 1991 occurred during a period when SO<sub>2</sub> emissions and, hence, predicted S deposition had declined substantially.

Large decreases in extractable S in the LFH of sites 1 and 2 were found between 1981 and 1991 (Table 4). The decreases were similar to those found for total S. Extractable S concentrations at sites 5 and 12 did not change between years. For the 25 non-S<sup>0</sup> sites, extractable S concentrations in the LFH decreased 20–35% between 1981 and 1985 and did not change from 1985 to 1991.

Changes in extractable S concentrations of the mineral horizons among years were observed only at sites 1 and 2 (Table 5). The highest S concentrations in the mineral soils at sites 1 and 2 were found in 1985. This peak in S concentrations resulted from a downward pulse of soluble S following the heavy deposition of S<sup>0</sup> in the late 1970s and the subsequent oxidation of S<sup>0</sup> to SO<sub>4</sub>-S. The amount of S retained in the mineral horizons decreased between 1985 and 1991 probably because the S<sup>0</sup> deposition rate had also decreased during the same time at sites 1 and 2.

Total and extractable S concentrations in the 1991 LFH samples differed among the three deposition classes (Table 6). Sulfur concentrations were the highest at sites contaminated with S<sup>0</sup> and in areas of the highest sulfation (high deposition class), which to a large degree are coincident. The medium deposition class sites had mean total and extractable S concentrations higher than the mean

**Table 4. Extractable sulfur (1.0 M NH<sub>4</sub>Cl) concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991. Values are means ±95% confidence limits (n = 5).**

Site	Deposition class	Extractable S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	12 200 ±6 130a	4 330 ±952b	3 220 ±1 480c
2	High	2 280 ±1 260a	1 150 ±259b	322 ± 104c
5	High	796 ± 428a	320 ± 61a	490 ± 140a
12	High	284 ± 104a	192 ± 85a	224 ± 42a
3	Medium	219 ± 60a	116 ± 24b	154 ± 52b
13	Medium	222 ± 34a	69 ± 15c	163 ± 38b
14	Medium	187 ± 42a	114 ± 22b	154 ± 46ab
23	Medium	219 ± 59a	113 ± 57b	146 ± 42b

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 5. Extractable sulfur (1.0 M NH<sub>4</sub>Cl) concentrations of the mineral horizons at sites 1 and 2 in 1981, 1985, and 1991. Values are means ±95% confidence limits (n = 5).**

Site and horizon	Deposition class	Extractable S (mg kg <sup>-1</sup> )		
		1981	1985	1991
Site 1	High			
IAe		204 ±99b	341 ± 50a	184 ± 80b
Bm		183 ±82c	953 ±372a	635 ±314b
IIAe		46 ±30c	239 ±202a	157 ± 41b
Site 2	High			
IAe		45 ± 9b	130 ± 44a	44 ± 15b
Bm		56 ±31c	408 ±268a	282 ±182b
IIAe		22 ±15c	92 ± 44a	54 ± 24b

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

S concentrations of the low deposition class sites. Elemental S contributed to the S concentrations at six of the seven sites in the high deposition class. Site 31 had no measurable S<sup>0</sup> but was located in an area of high sulfation. The total S concentrations in the LFH at site 31, however, were ranked in the lower one-third of all sites.

The medium deposition class sites had increased S concentrations in the LFH as a result of S deposition either as gaseous SO<sub>2</sub> or particulate S<sup>0</sup> during the last 20 years. The increases were about 6–10% (when compared to the mean S concentration

of the low deposition class sites) in extractable S of the LFH and 20% in total S concentrations of the LFH. The larger increase in total S in the LFH, could have resulted from the incorporation of SO<sub>4</sub>-S (oxidized from S<sup>0</sup> or from adsorbed SO<sub>2</sub>) into soil organic matter (Robarge and Johnson 1992). On a site by site basis, increases in the S concentrations in the LFH between 1985 and 1991 were found in 3 of the 13 medium deposition class sites (13, 14, and 23). These sites were within 6–8 km from the Ram River gas plant and close enough to the plant that low level S<sup>0</sup> deposition or S<sup>0</sup> vapor could be the source. This does not, however, exclude SO<sub>2</sub>

**Table 6. Total and extractable or exchangeable concentrations of the LFH in 1991 by deposition class. Values are means  $\pm$ 95% confidence limits.**

Element	Deposition class		
	High	Medium	Low
pH	5.3 $\pm$ 0.1a	4.8 $\pm$ 0.1c	5.1 $\pm$ 0.0b
----- (mg kg <sup>-1</sup> ) -----			
Sulfur			
Total	5 020 $\pm$ 159a	1 370 $\pm$ 117b	1 140 $\pm$ 133c
Extractable	748 $\pm$ 37a	158 $\pm$ 27b	145 $\pm$ 31c
Calcium			
Total	16 400 $\pm$ 509a	5 640 $\pm$ 374c	6 420 $\pm$ 426b
Exchangeable	10 400 $\pm$ 267a	5 110 $\pm$ 196c	5 410 $\pm$ 223b
Magnesium			
Total	1 030 $\pm$ 28b	862 $\pm$ 21c	1 050 $\pm$ 23a
Exchangeable	501 $\pm$ 20c	653 $\pm$ 14b	716 $\pm$ 16a
Potassium			
Total	842 $\pm$ 20b	1 080 $\pm$ 15a	1 110 $\pm$ 17a
Exchangeable	596 $\pm$ 18c	848 $\pm$ 13a	772 $\pm$ 15b
Manganese			
Total	955 $\pm$ 53b	864 $\pm$ 39b	1 130 $\pm$ 44a
Exchangeable	455 $\pm$ 23c	579 $\pm$ 17b	663 $\pm$ 20a
Aluminum			
Total	2 560 $\pm$ 96b	2 350 $\pm$ 71b	2 880 $\pm$ 81a
Exchangeable	94 $\pm$ 6b	172 $\pm$ 5a	109 $\pm$ 5b
Iron			
Total	2 890 $\pm$ 127b	2 560 $\pm$ 93c	3 360 $\pm$ 106a
Exchangeable	23 $\pm$ 3b	38 $\pm$ 2a	27 $\pm$ 2b
Phosphorus			
Total	993 $\pm$ 17b	1 120 $\pm$ 13a	1 020 $\pm$ 14b
Extractable	147 $\pm$ 5c	207 $\pm$ 4a	182 $\pm$ 4b

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

deposition, but SO<sub>2</sub> emissions from both plants decreased by >50% between 1985 and 1991.

The S concentrations in 1-year-old pine and spruce needles at selected sites are given in Tables 7 and 8. Total S concentrations at the four S<sup>0</sup> contaminated sites (1, 2, 5, and 12) were higher than at any other site. Total S concentrations in the pine and spruce foliage from 1991 were similar to the

concentrations found in 1981. Changes in the total S concentrations of the understory species between 1981 and 1991 were more variable (Tables 9–11). Sulfur concentrations in the feather moss of the 25 non-S<sup>0</sup> sites remained relatively constant with small increases (about 100 mg kg<sup>-1</sup>) at about one-half the sites. Sulfur concentrations in the feather moss of three sites in the medium deposition class (3, 13, and 14) were about 40% higher in 1991 than 1981 or

**Table 7. Total sulfur concentrations in 1-year-old pine needles at selected sites in 1981, 1985, and 1991. Values are means  $\pm$  95% confidence limits (n = 10).**

Site	Deposition class	Total S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	2 460 $\pm$ 355a	2 070 $\pm$ 228b	2 420 $\pm$ 186a
2	High	2 030 $\pm$ 229a	1 770 $\pm$ 223b	1 810 $\pm$ 215b
5	High	1 460 $\pm$ 143b	1 460 $\pm$ 144b	1 690 $\pm$ 153a
12	High	1 510 $\pm$ 247a	1 160 $\pm$ 81b	1 420 $\pm$ 114a
3	Medium	1 320 $\pm$ 149a	839 $\pm$ 87b	1 280 $\pm$ 103a
13	Medium	1 380 $\pm$ 119a	882 $\pm$ 91c	1 210 $\pm$ 75b
14	Medium	1 400 $\pm$ 167a	1 080 $\pm$ 81b	1 200 $\pm$ 72b
23	Medium	1 040 $\pm$ 106b	972 $\pm$ 93b	1 280 $\pm$ 92a

Note: Means in each row followed by the same letter do not differ significantly (p  $\geq$  0.05).

**Table 8. Total sulfur concentrations in 1-year-old spruce needles at selected sites in 1981, 1985, and 1991. Values are means  $\pm$  95% confidence limits (n = 10).**

Site	Deposition class	Total S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	2 890 $\pm$ 235a	2 000 $\pm$ 310c	2 510 $\pm$ 468b
2	High	1 770 $\pm$ 203a	1 200 $\pm$ 170b	1 830 $\pm$ 349a
5	High	1 280 $\pm$ 108a	918 $\pm$ 103b	1 250 $\pm$ 118a
12	High	1 040 $\pm$ 89a	712 $\pm$ 57b	1 140 $\pm$ 132a
3	Medium	1 160 $\pm$ 53a	619 $\pm$ 41b	1 050 $\pm$ 35a
13	Medium	857 $\pm$ 61a	652 $\pm$ 75b	994 $\pm$ 95a
14	Medium	1 180 $\pm$ 109a	704 $\pm$ 84c	980 $\pm$ 100b
23	Medium	993 $\pm$ 110a	693 $\pm$ 84b	1 010 $\pm$ 109a

Note: Means in each row followed by the same letter do not differ significantly (p  $\geq$  0.05).

1985. The increase in S concentration of the feather moss at sites 13 and 14 coincided with a similar increase in total S concentrations of the LFH.

Sulfur concentrations in twin-flower (Table 10) decreased between 1981 and 1991, however, the reasons for this are not known. With the exception of site 13, in which S concentrations increased by about 20%, the S concentrations in 1991 were about 15% lower than 1981. Sulfur concentrations in the alders of the 25 non-S<sup>0</sup> sites increased from 1981 to 1991 (Table 11) probably because of the time of sampling. Alder leaves were sampled in late

September and early October in 1981, and sampling was done in July and August in 1991. Sulfur concentrations generally decrease during the growing season and are the lowest in the fall (van den Driessche 1974).

Sulfur concentrations in the foliage of the tree and understory species sampled in 1985 (alder was not sampled in 1985) were 20–40% lower than in the foliage sampled in 1981 or 1991. The decrease in S concentrations of the foliage coincided with two dry summer years, 1984 and 1985. The summers of 1980 and 1981 were the wettest of any years since



**Table 9. Total sulfur concentrations in feather moss at selected sites in 1981, 1985, and 1991.** Values are means  $\pm$ 95% confidence limits (n = 10 except where indicated).

Site	Deposition class	Total S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	79 100 $\pm$ 29 900	N.D. <sup>a</sup>	17 500 $\pm$ 468
2	High	20 700 $\pm$ 5 040a	4 690 $\pm$ 712c	9 490 $\pm$ 2 730b
5	High	9 880 $\pm$ 1 070a	2 110 $\pm$ 138c	4 600 $\pm$ 1 060b
12	High	2 040 $\pm$ 82ab	1 800 $\pm$ 53b	2 580 $\pm$ 194a
3	Medium	1 260 $\pm$ 94b <sup>b</sup>	1 170 $\pm$ 65b	1 650 $\pm$ 335a
13	Medium	1 110 $\pm$ 67b	1 160 $\pm$ 54b	1 580 $\pm$ 196a
14	Medium	1 200 $\pm$ 108b	1 230 $\pm$ 72b	1 400 $\pm$ 71a
23	Medium	1 250 $\pm$ 44ab	1 160 $\pm$ 30b	1 300 $\pm$ 46a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

<sup>a</sup> N.D. = Not determined. There was no sample in 1985 since the feather moss had died at the plot. The 1991 samples were collected from the quadrats (n = 3) where the feather moss was still alive.

<sup>b</sup> n = 9.

**Table 10. Total sulfur concentrations in twin-flower at selected sites in 1981, 1985, and 1991.** Values are means  $\pm$ 95% confidence limits (n = 10 except where indicated).

Site	Deposition class	Total S (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	16 400 $\pm$ 3 460	5 220 $\pm$ 1 280	3 320 <sup>a</sup>
2	High	6 400 $\pm$ 1 100a	2 670 $\pm$ 162b	2 260 $\pm$ 369b <sup>b</sup>
5	High	3 310 $\pm$ 330a	1 710 $\pm$ 104b	1 240 $\pm$ 119b
12	High	1 670 $\pm$ 81a	1 490 $\pm$ 91a	1 280 $\pm$ 121a
3	Medium	1 450 $\pm$ 67a	788 $\pm$ 55c	1 120 $\pm$ 75b <sup>c</sup>
13	Medium	1 030 $\pm$ 50b	734 $\pm$ 30c	1 280 $\pm$ 106a
14	Medium	1 550 $\pm$ 115a	900 $\pm$ 69c	1 070 $\pm$ 101b
23	Medium	1 150 $\pm$ 39a	689 $\pm$ 45c	1 040 $\pm$ 78b

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

<sup>a</sup> Only two samples were collected in 1991. Eight of the quadrats had no twin-flower present.

<sup>b</sup> n = 9.

<sup>c</sup> n = 6.

the gas plants startups, and the summers of 1990 and 1991 were also wetter than normal (Fig. 3). Moist conditions provide good conditions for microbial activity. In dry years, microbial activity may be limited by moisture. The majority of S in the LFH is retained in an organic form and is made available for plant uptake by microorganisms. If conditions limited microbial activity, the amount of S oxidized to sulfate would be reduced, therefore, less S would be available for uptake by vegetation.

Moisture conditions may also have a direct effect on foliar nutrient concentrations. Leaf et al. (1970) found that precipitation in the year prior to and the year of sampling will affect the nutrient concentrations of tree foliage. The majority of foliage characteristics studied such as N and K concentrations showed lower values in drier years, however, P, Ca, and Mg concentrations showed the reverse trend. No data for S were presented. van den Driessche (1974) indicated that nutrients

**Table 11. Total sulfur concentrations in alder at selected sites in 1981 and 1991.**  
Values are means  $\pm$ 95% confidence limits (n = 10 except where indicated).

Site	Deposition class	Total S (mg kg <sup>-1</sup> )	
		1981 <sup>a</sup>	1991
1	High	10 600 $\pm$ 1 080a	3 860 $\pm$ 874b <sup>b</sup>
2	High	3 980 $\pm$ 397a	2 820 $\pm$ 306b
5	High	2 750 $\pm$ 442a	2 400 $\pm$ 183a
12	High	2 410 $\pm$ 308a	2 600 $\pm$ 262a
3	Medium	2 160 $\pm$ 299a	2 000 $\pm$ 224a
13	Medium	1 730 $\pm$ 159b	2 500 $\pm$ 215a
14	Medium	2 150 $\pm$ 273b	2 450 $\pm$ 275a
23	Medium	1 480 $\pm$ 229	2 230 <sup>c</sup>

Note: Mean in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

<sup>a</sup> No alder was collected in 1985.

<sup>b</sup> n = 3.

<sup>c</sup> Only one alder sample collected at site 23. No alder at nine of the quadrats.

respond differently in terms of foliar concentrations to climatic and moisture conditions.

The total S concentrations in the 1991 foliage by deposition class are given in Table 12. The S concentrations by deposition class followed the same pattern as total and extractable S in the LFH. The S concentrations were the highest in the high deposition class sites and decreased significantly from the medium to the low deposition class sites for all species sampled except bunchberry, for which there was no difference between concentrations in medium and low deposition classes.

Elemental S was present at site 14 and possibly site 13 (only one of five samples contained measurable S<sup>0</sup>) in 1991. No S<sup>0</sup> was detectable at site 23 and detectable amounts of S<sup>0</sup> were measured at site 3 in 1985. The increases (about 50%) in total S in the LFH (sites 13, 14, and 23) and the feather moss (sites 3, 13, and 14) between 1985 and 1991 were more typical of S<sup>0</sup> deposition than gaseous S deposition. Large amounts of S<sup>0</sup> were shipped between 1985 and 1989 (peaked in 1988, Fig. 6). The breakup of the S<sup>0</sup> block and the extensive use of the prilling tower may have resulted in S<sup>0</sup> being deposited further away from the gas plants than was previously found (Addison et al. 1984; Maynard 1990). Elemental S deposited during the latter part of the 1980s could have been oxidized in the LFH to levels below the detection limit. Since 1988, the amount of S<sup>0</sup> shipped has decreased considerably. Therefore,

the increased total S concentrations in the LFH at these sites could have been from the oxidation products of S<sup>0</sup> being retained in the surface organic horizon.

Sulfur dioxide emissions from both gas plants have decreased considerably since 1972 with a parallel decrease in sulfation rates at static sulfation stations near sites 3, 13, 14, and 23 (accurate sulfation data near site 3 only exist up to 1986). Elemental sulfur or S<sup>0</sup> vapor not adsorbed by the static sulfation stations could have caused the higher S concentrations in certain vegetation and the LFH at these four sites (3, 13, 14, and 23). Deposition of SO<sub>2</sub>, however, cannot be completely ruled out, and it is possible SO<sub>2</sub> was responsible for a portion of the elevated S concentrations in certain vegetation.

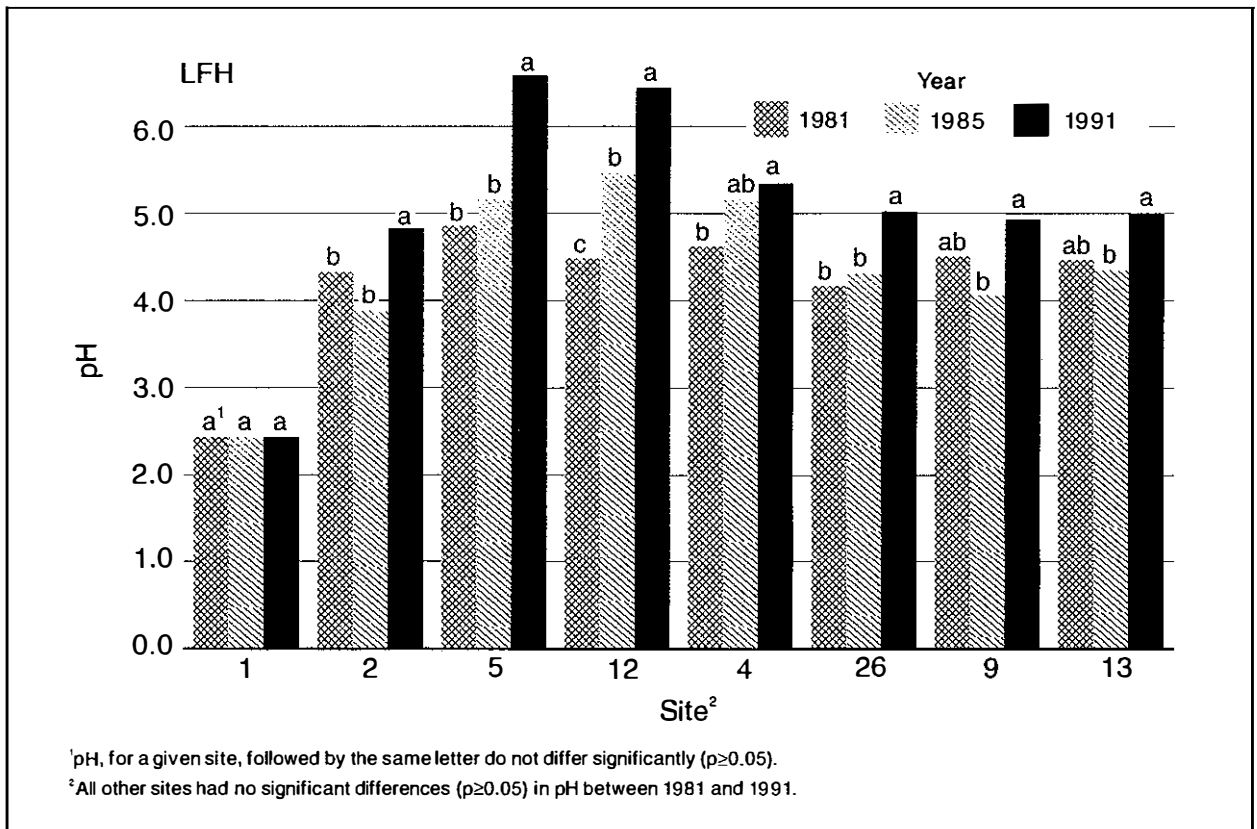
### pH, Calcium and Magnesium

The pH of the LFH between 1981 and 1991 did not change except for three sites that were contaminated with S<sup>0</sup> and limed and two sites that had been disturbed (Fig. 8). Sites 2, 5, and 12 were contaminated with S<sup>0</sup> and were also limed. Site 26 had a pipeline cut through a corner of the plot, and the plot was relocated. Site 4 was located within a grazing lease and had been grazed between 1981 and 1991. For all 5 sites, the pH of the LFH increased from 1981 to 1991. Site 1 was the most heavily impacted site in 1981 with a pH of 2.5. The pH at site 1 did not change between 1981 and 1991, even

**Table 12. Total sulfur concentrations in 1991 foliage by deposition class.**  
 Values are means  $\pm$  95% confidence limits.

Species	S deposition class ( $\text{mg kg}^{-1}$ )		
	High	Medium	Low
<b>Pine</b>			
Current-year	1 380 $\pm$ 16a	1 170 $\pm$ 12b	1 100 $\pm$ 13c
1-year-old	1 670 $\pm$ 19a	1 230 $\pm$ 14b	1 150 $\pm$ 16c
<b>Spruce</b>			
Current-year	1 220 $\pm$ 14a	963 $\pm$ 10b	908 $\pm$ 12c
1-year-old	1 440 $\pm$ 23a	979 $\pm$ 17b	919 $\pm$ 19c
Feather moss	6 320 $\pm$ 132a	1 320 $\pm$ 88b	1 200 $\pm$ 100c
Twin-flower	1 450 $\pm$ 21a	1 060 $\pm$ 14b	1 000 $\pm$ 16c
Alder	2 690 $\pm$ 55a	2 320 $\pm$ 38b	2 120 $\pm$ 57c
Bunchberry	3 150 $\pm$ 52a	2 530 $\pm$ 34b	2 590 $\pm$ 38b
Bog cranberry	1 540 $\pm$ 25a	1 270 $\pm$ 23b	1 190 $\pm$ 16c

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).



**Figure 8. The pH of the surface organic horizon at selected sites in 1981, 1985, and 1991.**

though the site had been limed, and  $S^0$  was still present in the LFH.

The pH of the mineral soils are given in Figures 9–11. The pH in the three mineral horizons of sites 1 and 2 (the most heavily  $S^0$ -impacted sites) decreased between 1981 and 1991. The decreases in the pH of the mineral horizons sampled at site 1 were >1 pH unit. At site 2, the decreases were >1 pH unit only in the IAe horizon. Liming at site 5 resulted in increased pH of the IAe horizon in 1991. No increases in pH because of liming were observed below the IAe horizon. At all other sites, pH changes in the mineral horizons were <0.5 pH unit with a few exceptions. Both increases and decreases in pH were observed. In mineral horizons where the pH increased or decreased by >0.5 pH there was no consistent pattern of pH change throughout the sampled soil profile. The changes were limited to one horizon at any given site. Overall, the only decreases in pH of the mineral soils that could be attributed to S deposition were found at the  $S^0$  sites 1 and 2.

Sites 13 and 16 showed a decrease in the pH of all three mineral horizons between 1981 and 1985.

Maynard (1990) indicated the pH decreases at site 13 in 1985 were probably related to road construction along two sides of the plot. There was no known reason for the decreases observed at site 16. The pH of the mineral soils of both sites in 1991 were similar to 1981 levels. Thus, the pH observed at sites 13 and 16 in 1985 appear to be anomalies and related to edaphic or other disturbances such as road construction.

Exchangeable Ca concentrations in the LFH are given in Table 13. There were no differences among years for any sites except the four sites next to the gas plants that had been contaminated with large amounts of  $S^0$  and limed. The exchangeable Ca concentrations in the LFH of sites 5 and 12 in 1991 were double the concentrations in 1985. Even with liming, Ca concentrations remained constant at site 2 and decreased at site 1 between 1981 and 1991. Exchangeable Ca concentrations in the mineral horizons did not change among years.

Exchangeable Mg concentrations in the LFH are given in Table 14. Sites 1 and 2 had the lowest Mg concentration in the LFH of any site. The Mg concentration at site 1 increased from 1981 to 1985

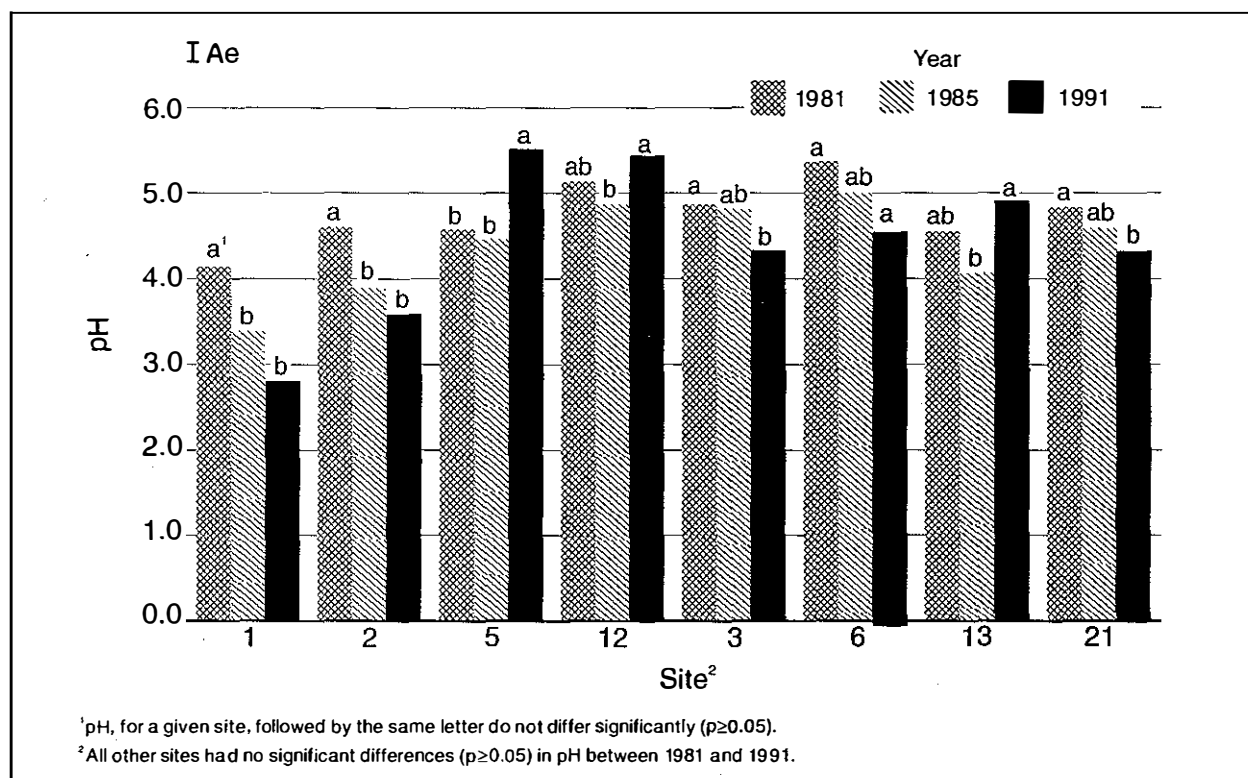


Figure 9. The pH of the IAe horizon at selected sites in 1981, 1985, and 1991.

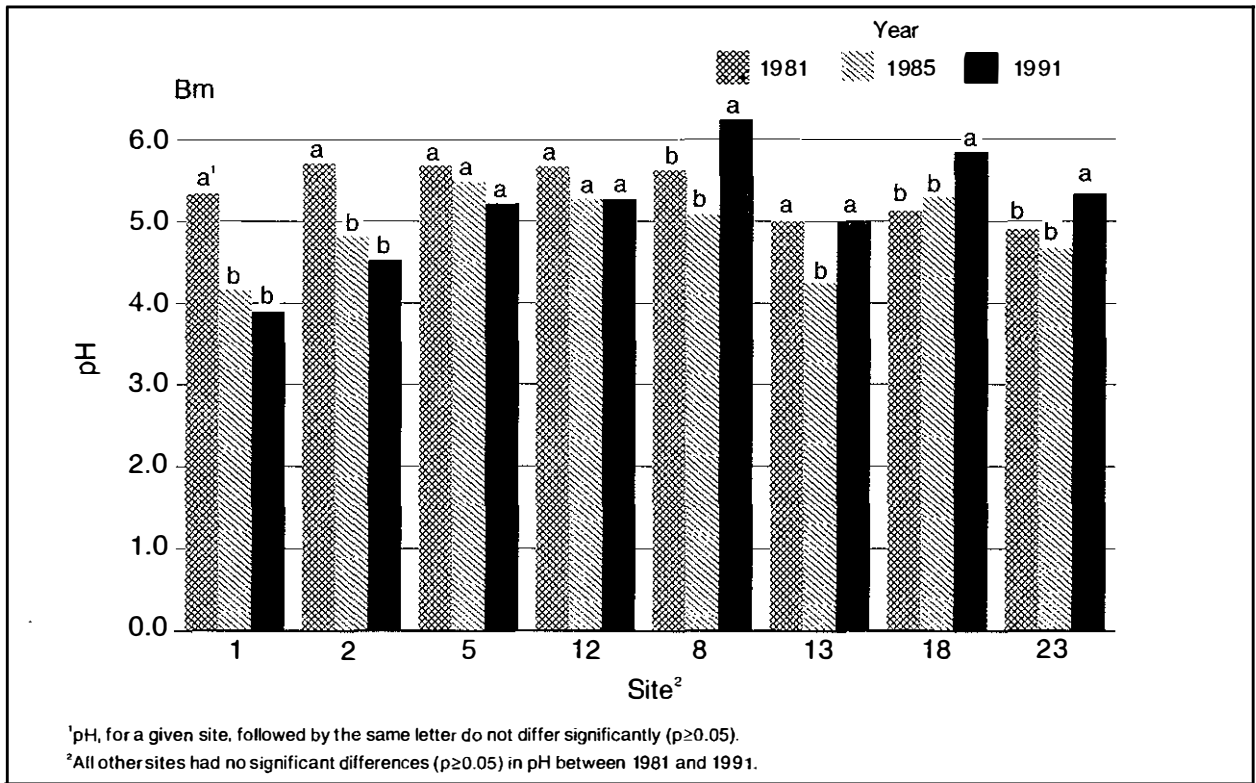


Figure 10. The pH of the Bm horizon at selected sites in 1981, 1985, and 1991.

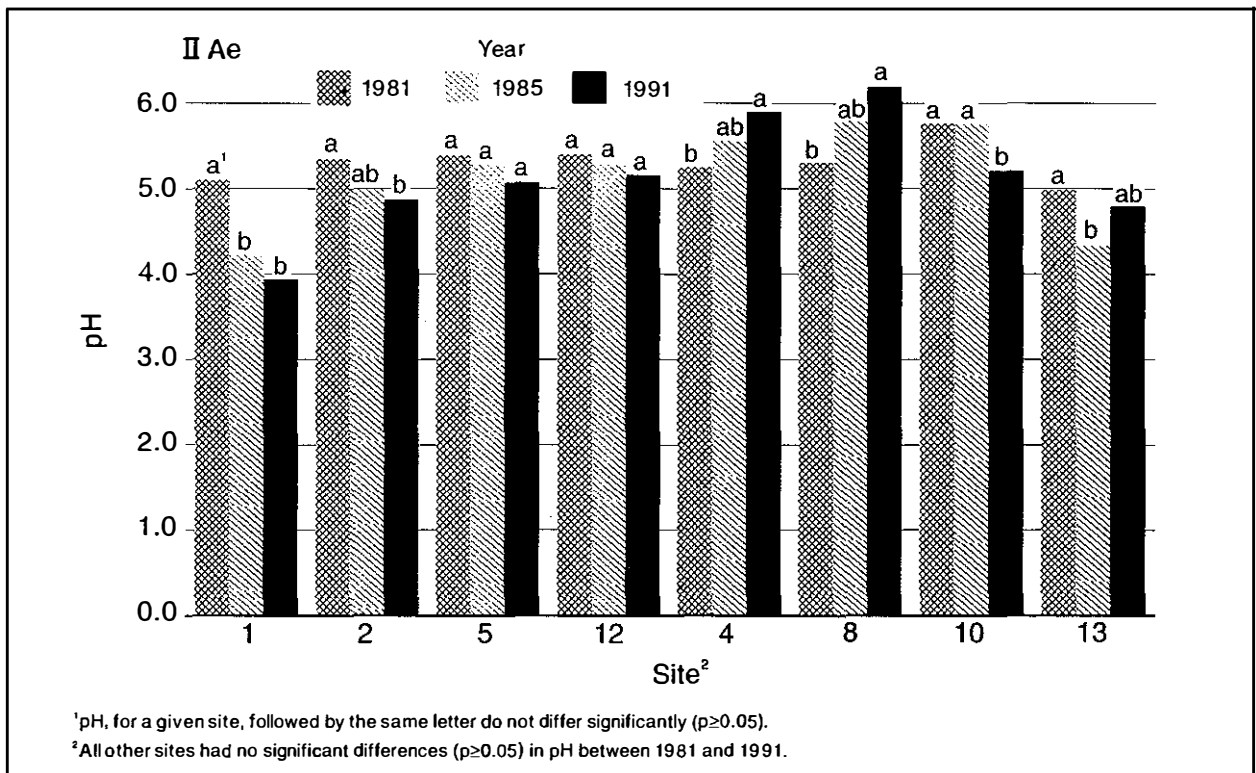


Figure 11. The pH of the II Ae horizon at selected sites in 1981, 1985, and 1991.

**Table 13. The exchangeable calcium (1.0 M NH<sub>4</sub>Cl) concentrations of the LFH at selected sites in 1981, 1985, and 1991. Values are means  $\pm$ 95% confidence limits (n = 5).**

Site	Deposition class	Exchangeable Ca (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	10 500 $\pm$ 5 470a	6 600 $\pm$ 3 390b	3 500 $\pm$ 2 440c
2	High	6 820 $\pm$ 3 990a	6 820 $\pm$ 3 260a	8 490 $\pm$ 5 710a
5	High	8 010 $\pm$ 4 860b	7 440 $\pm$ 2 720b	18 200 $\pm$ 4 380a
12	High	4 010 $\pm$ 2 000c	8 630 $\pm$ 1 020b	16 000 $\pm$ 5 410a
3	Medium	4 120 $\pm$ 1 630a	4 010 $\pm$ 1 160a	3 560 $\pm$ 1 170a
13	Medium	4 460 $\pm$ 1 170a	3 930 $\pm$ 1 520a	6 180 $\pm$ 2 010a
14	Medium	4 420 $\pm$ 979a	3 980 $\pm$ 1 150a	4 570 $\pm$ 1 240a
23	Medium	2 780 $\pm$ 632a	2 850 $\pm$ 733a	3 010 $\pm$ 138a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 14. The exchangeable magnesium (1.0 M NH<sub>4</sub>Cl) concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991. Values are means  $\pm$ 95% confidence limits (n = 5).**

Site	Deposition class	Exchangeable Mg (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	237 $\pm$ 90b	520 $\pm$ 299a	136 $\pm$ 77b
2	High	333 $\pm$ 182a	353 $\pm$ 81a	298 $\pm$ 150b
5	High	411 $\pm$ 51ab	293 $\pm$ 91b	526 $\pm$ 156a
12	High	487 $\pm$ 133a	454 $\pm$ 164a	556 $\pm$ 95a
3	Medium	625 $\pm$ 130ab	703 $\pm$ 156a	499 $\pm$ 145b
13	Medium	626 $\pm$ 217a	625 $\pm$ 307a	724 $\pm$ 205a
14	Medium	600 $\pm$ 52a	771 $\pm$ 127a	600 $\pm$ 137a
23	Medium	431 $\pm$ 108a	590 $\pm$ 160a	453 $\pm$ 24a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

and then decreased to a level slightly lower than originally measured in 1981. The increase from 1981 to 1985 was attributed to a dolomitic liming source that was applied by Gulf Canada in the fall of 1984 or spring of 1985 (Maynard 1990). The Mg added as a result of the lime application had been leached from the LFH by 1991. Changes in the Mg concentration of the LFH of other sites were small and not related to S deposition.

The Ca and Mg concentrations of the lodgepole pine and spruce 1-year-old needles sampled in 1991 were higher than in the 1-year-old needles sampled in 1981 or 1985. The 1991 concentrations of Ca and

Mg at all sites were within the ranges reported for 1-year-old needles of mature pine and spruce (Morrison 1974; Kimmins et al. 1985; Weetman et al. 1985). The critical range used to assess the Ca status of lodgepole pine and white spruce is 600–1000 mg kg<sup>-1</sup> and 1000–1500 mg kg<sup>-1</sup>, respectively. The Ca concentrations exceeded these values for both species at all sites in 1991 (Ca concentrations in tree foliage were  $>2400$  mg kg<sup>-1</sup>). The critical range used to assess the Mg status of lodgepole pine and white spruce is 600–1000 mg kg<sup>-1</sup>. The Mg concentrations for both species at all sites in 1991 were at the upper range or exceeded these values (Mg concentrations in tree foliage were  $\geq 900$  mg kg<sup>-1</sup>).

The pH in the LFH and Ca and Mg in soils and foliage (Tables 6, 15, and 16) were also assessed by deposition class. The pH of the LFH and Ca concentrations of the LFH and the foliage (except alder) at the high deposition class sites were greater than in either of the other deposition class sites because of liming at five of the seven sites in the high deposition class. Magnesium concentrations of the LFH and foliage in the high deposition class sites tended to be less than in the low deposition class sites. The Mg content of spruce was the exception.

The pH of the LFH and Ca concentrations in the LFH and foliage of the medium deposition class sites were less than for the low deposition class sites. The decrease in Ca concentrations ranged from 3–17% with the largest difference in feather moss Ca and the smallest difference in bunchberry and twin-flower. Magnesium concentrations also tended to be lower in medium deposition class sites compared to the low deposition class sites. Pine needles, feather moss, twin-flower and bog cranberry had about 10% lower Mg in the medium

**Table 15. Total calcium concentrations in 1991 foliage by deposition class. Values are means  $\pm$  95% confidence limits.**

Species	Ca deposition class ( $\text{mg kg}^{-1}$ )		
	High	Medium	Low
Pine			
Current year	1 960 $\pm$ 59a	1 820 $\pm$ 44a	1 980 $\pm$ 49a
1-year-old	3 480 $\pm$ 115a	3 260 $\pm$ 85b	3 580 $\pm$ 96a
Spruce			
Current year	5 630 $\pm$ 157a	4 710 $\pm$ 115c	5 140 $\pm$ 132b
1-year-old	9 070 $\pm$ 241a	7 410 $\pm$ 176c	8 340 $\pm$ 202b
Feather moss	11 100 $\pm$ 352a	4 650 $\pm$ 234c	5 570 $\pm$ 267b
Twin-flower	11 400 $\pm$ 185a	10 400 $\pm$ 128c	10 800 $\pm$ 143b
Alder	9 780 $\pm$ 219c	10 400 $\pm$ 193b	11 500 $\pm$ 326a
Bunchberry	22 100 $\pm$ 259a	20 100 $\pm$ 167c	20 800 $\pm$ 188b
Bog cranberry	5 360 $\pm$ 120a	4 700 $\pm$ 114b	5 320 $\pm$ 79a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 16. Total magnesium concentrations in 1991 foliage by deposition class. Values are means  $\pm$  95% confidence limits.**

Species	Mg deposition class ( $\text{mg kg}^{-1}$ )		
	High	Medium	Low
Pine			
Current year	1 100 $\pm$ 20b	1 150 $\pm$ 15b	1 250 $\pm$ 17a
1-year-old	1 310 $\pm$ 29b	1 280 $\pm$ 22b	1 450 $\pm$ 25a
Spruce			
Current year	1 130 $\pm$ 18a	1 110 $\pm$ 13a	1 160 $\pm$ 15a
1-year-old	1 160 $\pm$ 24a	1 110 $\pm$ 17a	1 130 $\pm$ 20a
Feather moss	1 020 $\pm$ 28b	965 $\pm$ 18b	1 100 $\pm$ 21a
Twin-flower	2 460 $\pm$ 48c	2 980 $\pm$ 33b	3 120 $\pm$ 37a
Alder	2 030 $\pm$ 49b	2 430 $\pm$ 43a	2 570 $\pm$ 73a
Bunchberry	4 730 $\pm$ 96b	5 650 $\pm$ 62a	5 490 $\pm$ 70a
Bog cranberry	1 010 $\pm$ 25c	1 070 $\pm$ 24b	1 150 $\pm$ 16a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

deposition class sites compared to the low deposition class sites.

The Ca and Mg concentrations in the LFH and foliage were about 10% less in the medium deposition class sites compared to the low deposition class sites. This corresponded to lower pH and greater S concentrations in the LFH in the medium versus the low deposition class sites. It was not possible to determine a cause and effect relationship between higher S in the medium deposition class sites and lower pH, Ca, and Mg. There were no decreases in pH, Ca, or Mg concentrations among years for a given medium deposition class site (e.g., sites 3, 13, 14, and 23) in the comparison of the 1981, 1985, and 1991 soils data. The Ca and Mg concentrations in the LFH and foliage at the medium deposition class sites were at the upper end of the range of concentrations reported in the literature (Morrison 1974; Stark 1983; Kimmins et al. 1985; Maynard and Fairbarns 1994). Therefore, the decreases observed in Ca and Mg concentrations (whether because of S loading or not) would have little or no impact on the nutrient status of the trees at the medium deposition class sites.

### Other Elements

Exchangeable K concentrations in the LFH are given in Table 17. Potassium concentrations decreased between 1981 and 1991. Four sites (11, 12, 18, and 19) had decreases of about 50%; however, from 1981 to 1991, K concentrations in the LFH at site 1 and 2 decreased 70 and 88%, respectively. Potassium concentrations in the mineral horizons

remained constant among sampling years. The exchangeable K concentrations in the mineral horizons and the foliage of the trees and understory species did not change among years with a few exceptions (data not presented). Potassium concentrations in feather moss at site 2 were 15% lower than at any other site. Previous studies showed that as the moss was damaged at S<sup>0</sup> contaminated sites (sites 1 and 15), plant tissue nutrient concentrations generally decreased (Addison et al. 1984).

Total and exchangeable K were lower in the LFH of the high deposition class sites (Table 6); however, the reduction in K of the LFH was not associated with decreased foliar K (Table 18). The K concentrations of most species sampled were greater in the high deposition class sites compared to the low and medium deposition classes (K in bog cranberry and alder did not change). Potassium in the LFH at sites 1 and 2 decreased from 1981 to 1991, but no changes were found in the mineral soils. The reductions in exchangeable and total K were probably related to increased inputs of SO<sub>4</sub>-S that resulted in increased K solubility. The increased foliar K concentrations in 1991 at sites 1 and 2 were an indication that K uptake by vegetation increased as a result of increased K solubilization in the LFH. There was no difference in K concentrations of the soil and foliage in the medium versus the low deposition class sites.

The exchangeable Al concentrations in the LFH are given in Table 19. Exchangeable Al at the 21 non-S<sup>0</sup> sites generally decreased from 1981 to 1985 and then increased in 1991 to levels similar to those

**Table 17. Exchangeable potassium (1.0 M NH<sub>4</sub>Cl) concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991. Values are means ± 95% confidence limits (n = 5).**

Site	Deposition class	Exchangeable K (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	879 ± 452a	173 ± 34b	107 ± 14b
2	High	965 ± 319a	348 ± 246b	286 ± 86b
5	High	883 ± 344a	872 ± 187a	568 ± 217a
12	High	978 ± 431a	1 190 ± 651a	490 ± 91b
3	Medium	864 ± 288b	1 360 ± 356a	879 ± 411b
13	Medium	992 ± 375a	1 020 ± 451a	830 ± 310a
14	Medium	963 ± 355a	1 300 ± 394a	878 ± 331a
23	Medium	976 ± 345a	1 110 ± 562a	886 ± 118a

Note: Means in each row followed by the same letter do not differ significantly (p ≥ 0.05).



**Table 18. Total potassium concentrations in 1991 foliage by deposition class. Values are means  $\pm$ 95% confidence limits.**

Species	K deposition class ( $\text{mg kg}^{-1}$ )		
	High	Medium	Low
Pine			
Current year	6 260 $\pm$ 128a	5 790 $\pm$ 94b	5 740 $\pm$ 107b
1-year-old	5 060 $\pm$ 96a	4 640 $\pm$ 71b	4 760 $\pm$ 80b
Spruce			
Current year	9 150 $\pm$ 128a	8 410 $\pm$ 94b	8 260 $\pm$ 107b
1-year-old	6 180 $\pm$ 116a	5 560 $\pm$ 85b	5 430 $\pm$ 97b
Feather moss	3 780 $\pm$ 108a	3 900 $\pm$ 72a	3 790 $\pm$ 82a
Twin-flower	14 000 $\pm$ 198a	14 400 $\pm$ 136a	13 900 $\pm$ 153a
Alder	8 690 $\pm$ 207b	9 620 $\pm$ 182a	9 130 $\pm$ 308ab
Bunchberry	13 700 $\pm$ 199a	13 000 $\pm$ 129b	12 900 $\pm$ 145b
Bog cranberry	4 070 $\pm$ 62a	3 940 $\pm$ 90a	4 180 $\pm$ 62a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 19. Exchangeable aluminum (1.0 M  $\text{NH}_4\text{Cl}$ ) concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991. Values are means  $\pm$ 95% confidence limits ( $n = 5$ ).**

Site	Deposition class	Exchangeable Al ( $\text{mg kg}^{-1}$ )		
		1981	1985	1991
1	High	859 $\pm$ 493a	115 $\pm$ 74b	133 $\pm$ 78b
2	High	267 $\pm$ 242a	50 $\pm$ 65b	75 $\pm$ 88b
5	High	160 $\pm$ 124a	25 $\pm$ 29a	15 $\pm$ 9b
12	High	256 $\pm$ 104a	18 $\pm$ 7b	15 $\pm$ 5b
3	Medium	240 $\pm$ 201a	77 $\pm$ 62a	246 $\pm$ 73a
13	Medium	208 $\pm$ 64a	82 $\pm$ 47b	127 $\pm$ 98ab
14	Medium	151 $\pm$ 61a	61 $\pm$ 58a	108 $\pm$ 80a
23	Medium	293 $\pm$ 72a	135 $\pm$ 89b	253 $\pm$ 46ab

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

found in 1981. The Al concentrations in the four most heavily impacted  $\text{S}^0$  sites (1, 2, 5, and 12), however, decreased between 1981 and 1991, and 3 of the 4 sites had the lowest Al concentrations of any site in 1991. The decreased exchangeable Al at the four sites may be the result of different processes. At sites 5 and 12, the lower exchangeable Al concentrations probably resulted from the increase in LFH pH due to liming. The lower Al concentrations at site 1 and probably site 2 resulted from the increased solubilization of Al with the lowering of the pH. The solubilities of Al in water increase rapidly below pH 4.7 and accelerate to high concentrations

below pH 4.0 (McLean 1976). Thus at site 1 where the pH of the LFH has been at 2.5 for at least 10 years the high extractable Al in 1981 followed by decreased concentrations in 1985 and 1991 could be explained by the increased solubilization of Al and its subsequent leaching. The critical soil pH at which Al becomes available in toxic concentrations depends on many factors including organic matter levels, plant species, and concentrations of exchangeable ions (Foy 1984; Cronan et al. 1989).

Exchangeable Al concentrations changed significantly for all three mineral horizons only at

site 1 (Table 20). In the IAe horizon of site 1, Al concentrations increased from 1981 to 1985 and then decreased by 3.5 times in 1991. The Al concentrations increased four to six fold in the Bm and IIAe horizons of site 1 between 1981 and 1991. The Al concentration in the IAe horizon of site 2 increased twofold between 1981 and 1991. These changes in the Al concentration of sites 1 and 2 (IAe horizon) coincided with decreasing pH and increasing S concentrations in the horizons. Johnson and Taylor (1989) indicated the introduction of mobile, mineral acid anions (such as  $\text{SO}_4\text{-S}$ ) to soil solution will increase Al solubility. The changes in Al concentrations observed in the mineral horizons of sites 1 and 2 are consistent with this hypothesis. Other changes in Al concentrations of the mineral horizons at other sites were of a lesser magnitude and were generally decreases.

Aluminum concentrations were only measured in foliage samples in 1985 and 1991 (data not presented). There was a large difference in the Al concentration between pine and spruce. Aluminum concentrations in spruce were  $<100 \text{ mg kg}^{-1}$  and ranged from 400 to  $900 \text{ mg kg}^{-1}$  in pine. There was no apparent pattern of Al concentration in either tree species that was related to S deposition class. Similar observations pertain to the understory species sampled. The soils at site 1, and to some extent at site 2, had increased Al solubility, but it did not result in increased Al concentrations in the aboveground foliage. That does not rule out the possibility that increased Al has affected the roots.

Cronan et al. (1989) in a review of studies on Al toxicity on several tree species found nutritional effects (i.e., decreased foliar concentrations of Ca and Mg) were typically more sensitive to Al than the various growth indexes. The Ca and Mg concentrations of the tree and understory foliage in this study, however, do not indicate any impact of Al toxicity.

The exchangeable Mn concentrations of the LFH are given in Table 21. In 1991 the Mn concentrations in the LFH of sites 1 and 2 were an order of magnitude lower than at any other site. The greatest decrease in Mn occurred at these sites between 1981 and 1985. Manganese concentrations in the soil, however, were generally the most variable of any element measured in this study and there was no statistical difference in the Mn concentrations among years. Exchangeable Mn concentrations in the mineral horizons were also extremely variable. The exchangeable Mn in the IAe horizon at sites 1 and 2 were among the lowest of any site and decreased by at least 10 times between 1981 and 1991. There was no measurable differences in Mn concentrations below the IAe horizon.

Manganese concentrations in the various plant species were extremely variable among sites and years. There was no consistent pattern in Mn concentrations that could be attributed to S deposition except possibly higher Mn concentrations in pine and alder at the highest  $\text{S}^0$ -contaminated sites (sites 1 and 2, data not presented).

**Table 20. Exchangeable aluminum (1.0 M  $\text{NH}_4\text{Cl}$ ) concentrations of the mineral horizons of sites 1 and 2 in 1981, 1985, and 1991. Values are means  $\pm$  95% confidence limits (n = 5).**

Site and horizon	Deposition class	Exchangeable Al ( $\text{mg kg}^{-1}$ )		
		1981	1985	1991
Site 1	High			
IAe		414 $\pm$ 125b	634 $\pm$ 166a	181 $\pm$ 86c
Bm		191 $\pm$ 95b	620 $\pm$ 39a	796 $\pm$ 518a
IIAe		98 $\pm$ 48c	420 $\pm$ 141b	667 $\pm$ 264a
Site 2	High			
IAe		300 $\pm$ 229b	503 $\pm$ 177a	627 $\pm$ 180a
Bm		187 $\pm$ 221a	373 $\pm$ 326a	306 $\pm$ 179a
IIAe		80 $\pm$ 69a	80 $\pm$ 50a	160 $\pm$ 32a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 21. Exchangeable manganese (1.0 M NH<sub>4</sub>Cl) concentrations of the LFH horizon at selected sites in 1981, 1985, and 1991. Values are means ±95% confidence limits (n = 5).**

Site	Deposition class	Exchangeable Mn (mg kg <sup>-1</sup> )		
		1981	1985	1991
1	High	569 ± 806 <sup>a</sup>	41 ± 19	20 ± 9
2	High	611 ± 590	137 ± 199	52 ± 20
5	High	447 ± 318	195 ± 259	373 ± 150
12	High	1 070 ± 838	237 ± 145	405 ± 187
3	Medium	987 ± 870	422 ± 101	704 ± 541
13	Medium	430 ± 279	284 ± 139	790 ± 540
14	Medium	700 ± 344	455 ± 143	688 ± 591
23	Medium	887 ± 803	620 ± 142	756 ± 817

<sup>a</sup> There was no significant ( $p \geq 0.05$ ) year × site interaction for exchangeable Mn.

Increased Fe solubility occurs at lower pH than Al, thus the only site with a pH low enough to increase Fe solubility in the LFH was site 1. Concentrations at site 1 were 2 to 4 times those at the site with the next highest Fe concentrations. Increases in exchangeable Fe of the mineral horizons were observed in the IAe horizon of sites 1 and 2. No changes were observed below the IAe horizon. Increases in Fe solubility in the IAe of sites 1 and 2 coincided with decreases in pH below 3.8. Exchangeable Fe concentrations in the soil and total Fe concentrations in the foliage of the vascular plants were low (generally in the range of 100 mg kg<sup>-1</sup>) compared to Al and Mn and did not vary among years or sites. There were some differences in Al, Fe, and Mn concentrations among the deposition classes; however, the differences were small and showed no pattern related to the S deposition class.

### 1992 Young Pine Sites

The total and extractable S concentrations in the LFH of the quadrats are given in Table 22. Site C had the highest S concentrations of any young stand; however, the S concentrations at site C were not significantly different ( $p \geq 0.05$ ) than the S concentrations at sites D and F. The S concentrations of the LFH at site C were similar to the mean S concentrations found in the medium deposition sites of the mature lodgepole pine.

The S in the pine needles followed a similar pattern to the LFH S concentrations. Site C had the highest concentrations of any site (Table 23). The S content of the current foliage of the young pine was

generally higher than in the current foliage of mature pine. The opposite was true for 1-year-old needles.

Sulfur concentrations were the highest in foliage from site C for all understory species sampled (Table 24). The differences in S concentrations among sites, however, were only significant for twin-flower and feather moss. The S concentration of twin-flower at site C was significantly higher than for any other site.

No measurable S<sup>0</sup> was found in the LFH at site C; however, S<sup>0</sup> was found in the mature stand (site 30) 200 m east of the young stand. Although no S<sup>0</sup> was measured at site C, the presence of S<sup>0</sup> at site 30 indicated that significant amounts of S<sup>0</sup> have been deposited in this area. Thus, the higher S concentrations observed at site C were probably the result of S<sup>0</sup> deposition at a rate lower than the detection limit for S<sup>0</sup>, or deposition occurred at a rate lower than the oxidation rate of S<sup>0</sup> in the LFH.

The pII of the LFH ranged from 5.7 at site D to 4.7 at site A. The pH at site C was in the mid-range of pH for the 6 young pine sites. The total and extractable concentrations of the LFH for the other nutrient elements measured were generally highest at sites C and D and lowest at sites A and E. There was no impact of increased S concentrations in the LFH of site C on the pH or nutrient status of the site. Thus, S deposition (gaseous or S<sup>0</sup>) had no measurable impact on the chemical properties of the young stands except for elevated S concentrations in the LFH and certain plant species at site C.

**Table 22. Total and extractable sulfur in the quadrat LFH of the young lodgepole pine sites. Values are means  $\pm$ 95% confidence limits (n = 20).**

Site	S (mg kg <sup>-1</sup> )	
	Total	Extractable
A	966 $\pm$ 168b	92 $\pm$ 13c
B	892 $\pm$ 114b	135 $\pm$ 18ab
C	1 370 $\pm$ 251a	171 $\pm$ 34a
D	1 200 $\pm$ 172ab	143 $\pm$ 17ab
E	883 $\pm$ 200b	93 $\pm$ 16c
F	1 030 $\pm$ 118ab	125 $\pm$ 19bc

Note: Means in each column followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 23. Total sulfur concentrations in the current and 1-year-old needles of the young lodgepole pine. Values are means  $\pm$ 95% confidence limits.**

Site	Pine (mg kg <sup>-1</sup> )	
	Current-year	1-year-old
A	1 220 $\pm$ 65b	942 $\pm$ 61d
B	1 400 $\pm$ 52bc	1 140 $\pm$ 62ab
C	1 680 $\pm$ 86a	1 170 $\pm$ 120a
D	1 620 $\pm$ 102a	1 060 $\pm$ 78bc
E	1 310 $\pm$ 99cd	969 $\pm$ 58cd
F	1 430 $\pm$ 107b	1 090 $\pm$ 64ab

Note: Means in each column followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

**Table 24. Total sulfur concentrations in the foliage of understory species of the young lodgepole pine stands. Values are means  $\pm$ 95% confidence limits.**

Site	Understory species (mg kg <sup>-1</sup> )				
	Bunchberry	Fireweed	Twin-flower	Feather moss	Prickly rose
A	2 590 $\pm$ 202a	1 960 $\pm$ 152a	1 180 $\pm$ 50b	1 540 $\pm$ 54a	2 550 $\pm$ 346a
B	2 340 $\pm$ 175a	1 910 $\pm$ 133a	997 $\pm$ 41c	1 150 $\pm$ 81b	2 780 $\pm$ 226a
C	2 680 $\pm$ 284a	2 040 $\pm$ 269a	1 340 $\pm$ 92a	1 610 $\pm$ 76a	2 740 $\pm$ 282a
D	2 610 $\pm$ 205a	1 690 $\pm$ 104a	1 060 $\pm$ 60bc	1 570 $\pm$ 172a	2 430 $\pm$ 260a
E	2 680 $\pm$ 208a	1 950 $\pm$ 192a	1 090 $\pm$ 73bc	1 490 $\pm$ 113a	2 590 $\pm$ 144a
F	2 580 $\pm$ 279a	1 930 $\pm$ 193a	1 000 $\pm$ 64c	1 250 $\pm$ 113b	2 480 $\pm$ 146a

Note: Means in each column followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

## 1992 Aspen Sites

The pH, and total and extractable concentrations of various elements in the LFH from the three aspen sites are given in Table 25. Site 40 (moderate S<sup>0</sup> site) and 42 (high S<sup>0</sup> site) had measurable S<sup>0</sup> in the LFH (1860 ± 1040 and 7020 ± 7150 mg kg<sup>-1</sup>, respectively), and both were limed. Site 42 is next to the mature pine (site 1) and was the most heavily impacted. Total and extractable S concentrations in the LFH of

site 42 were significantly higher than either sites 40 or 41 (non-S<sup>0</sup> site). There was no significant difference in S<sup>0</sup> concentration between site 40 and 41. This is largely because of the high variability associated with the S<sup>0</sup> contaminated sites. There was very little understory vegetation remaining at site 42 near the edge of the stand next to the Strachan gas plant. The quadrats furthest from the edge of the stand had some vegetation remaining. Site 40, although impacted with S<sup>0</sup> still had an intact understory.

**Table 25. The pH, total, and extractable or exchangeable concentrations in the LFH of the trembling aspen stands. Values are means ± 95% confidence limits (n = 20).**

Element	High S <sup>0</sup> site (site 42)	Moderate S <sup>0</sup> site (site 40)	Non S <sup>0</sup> site (site 41)
pH	5.6 ± 0.7b	7.0 ± 0.1a	6.6 ± 0.2a
----- (mg kg <sup>-1</sup> ) -----			
<b>Sulfur</b>			
Total	28 300 ± 6 310a	7 540 ± 2 080b	1 330 ± 294b
Extractable	3 860 ± 869a	600 ± 110b	136 ± 132b
<b>Calcium</b>			
Total	42 300 ± 9 150a	44 900 ± 8 520a	12 000 ± 2 260b
Exchangeable	16 400 ± 2 530b	49 200 ± 10 900a	8 220 ± 2 110b
<b>Magnesium</b>			
Total	3 260 ± 877ab	3 650 ± 944a	2 160 ± 86b
Exchangeable	364 ± 66b	522 ± 52b	1 010 ± 200a
<b>Potassium</b>			
Total	501 ± 80c	1 160 ± 116b	1 520 ± 132a
Exchangeable	323 ± 72b	648 ± 59a	749 ± 178a
<b>Manganese</b>			
Total	267 ± 93b	835 ± 226a	1 080 ± 185a
Exchangeable	102 ± 36b	75 ± 13b	222 ± 80a
<b>Aluminum</b>			
Total	946 ± 153b	2 140 ± 667b	4 700 ± 1 080a
Exchangeable	18 ± 22a	7 ± 1a	11 ± 4a
<b>Iron</b>			
Total	2 110 ± 333b	3 040 ± 805b	6 970 ± 1 540a
Exchangeable	14 ± 16a	4 ± 1a	3 ± 1a
<b>Phosphorus</b>			
Total	893 ± 79b	1 290 ± 79a	1 260 ± 160a
Extractable	117 ± 26a	143 ± 18a	134 ± 52a

Note: Means in each row followed by the same letter do not differ significantly (p ≥ 0.05).

The measurement of the impact of  $S^0$  deposition and its subsequent oxidation on pH and base cations was complicated by liming. Both sites 40 and 42 were heavily limed. As a result Ca concentrations in the LFH at both impacted sites were higher than at site 41. Similarly, but to a lesser extent, Mg concentrations were higher at the  $S^0$ -impacted sites. The pH of site 40 was higher than site 41; however, site 42 had the lowest pH. Thus, even with liming, the pH of the LFH at site 42 was lower than at site 41.

Site 41 had the highest and site 42 the lowest total concentrations of K, Mn, Al, and Fe of any of the three aspen sites. The general depletion of nutrient elements from the LFH of the site 42 is consistent with the results found at site 1 (most heavily impacted pine site). The biggest difference in chemical properties of the LFH between site 42 and site 1 was pH. The pH of site 42 was high compared to the pH of site 1. The large amount of lime (indicated by Ca and Mg concentrations) was a contributing factor. In addition, the surface

organic horizons of aspen stands generally have higher pH than pine stands as a result of the higher demand for base cations in aspen because aspen are nutrient pumps that take up large quantities of base cations from the entire soil profile and then return the nutrients to the soil surface as litter fall (Jones and DeByle 1985).

Differences in the pH and extractable or exchangeable concentrations of S, Ca, and Mg were found in the three mineral horizons sampled (Table 26). Sulfur concentrations in all three mineral horizons of site 42 were higher than the other sites. The S concentrations at site 40 (moderate  $S^0$  site) were intermediate between sites 42 (high  $S^0$  site) and site 41 (non- $S^0$  site). The differences were not all significant because of the large variability associated with sites contaminated with  $S^0$ . The increase in S concentrations coincided with decreasing pH. The pH of the site 42 mineral horizons was about 1.5 pH units lower than sites 40 or 41, even though the site had been limed. The pH of site 41 was the same as site 40 for the three mineral horizons. Thus, the

**Table 26. The pH and extractable or exchangeable concentrations in the mineral horizons of the trembling aspen stands. Values are means  $\pm$  95% confidence limits (n = 5).**

Parameter Horizon	High $S^0$ site (site 42)	Moderate $S^0$ site (site 40)	Non $S^0$ site (site 41)
<b>pH</b>			
IAe	3.9 $\pm$ 0.4b	5.5 $\pm$ 1.1a	5.5 $\pm$ 0.4a
Bm	4.4 $\pm$ 0.1b	5.8 $\pm$ 1.3a	5.8 $\pm$ 0.4a
IIAe	4.4 $\pm$ 0.2b	5.9 $\pm$ 1.3a	6.0 $\pm$ 0.2a
----- (mg kg <sup>-1</sup> ) -----			
<b>Sulfur</b>			
IAe	132 $\pm$ 66a	103 $\pm$ 44a	26 $\pm$ 5b
Bm	624 $\pm$ 486a	92 $\pm$ 29b	20 $\pm$ 7b
IIAe	128 $\pm$ 50a	72 $\pm$ 32b	22 $\pm$ 3c
<b>Calcium</b>			
IAe	926 $\pm$ 271b	4 120 $\pm$ 5 020a	1 510 $\pm$ 386ab
Bm	563 $\pm$ 459b	2 380 $\pm$ 2 520a	1 200 $\pm$ 514ab
IIAe	1 600 $\pm$ 1 190a	2 300 $\pm$ 1 640a	1 530 $\pm$ 466a
<b>Magnesium</b>			
IAe	29 $\pm$ 8b	169 $\pm$ 207ab	250 $\pm$ 63a
Bm	20 $\pm$ 21b	178 $\pm$ 228ab	221 $\pm$ 85a
IIAe	81 $\pm$ 63b	267 $\pm$ 241ab	329 $\pm$ 116a

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

amount of S<sup>0</sup> deposition, and the acidity produced from the oxidation of S<sup>0</sup> appeared to be neutralized by liming at site 40. At site 42, however, the lime applied was not sufficient to counter the acidification of the soil horizons (LFH or mineral).

The mineral horizons of site 42 had lower concentrations of Ca and Mg than site 41. It appeared that Ca was being leached from the mineral horizons even though the site had been limed. The depletion of Mg from the mineral horizons was more evident probably because there was less Mg in the lime material. There were no differences in K concentrations among sites (data not presented). These results are consistent with the mature pine sites. There was evidence for increased leaching of Mg and Ca (complicated by liming), but there was little or no change in the exchangeable K concentrations of the mineral horizons.

The S and Ca concentrations in the aspen foliage and the two understory species present at the three aspen sites are given in Table 27. There was an obvious gradient in S concentrations among sites. The highest S concentrations were found in the foliage at site 42 and the lowest S concentrations at site 41. Calcium concentrations were also highest in the two S<sup>0</sup>-contaminated sites (42 and 40) because of liming at both locations.

## Forest Health Survey

### Health Conditions

The proportion of trees of all species in the four health classes in all mature pine sites is shown in

Figure 12. A Chi-square analysis found that there was a difference among the three S classes in the proportion of trees in the various health classes ( $\chi^2 = 113.93$ ,  $p < 0.0001$ ). Further testing was done to determine which S classes differed. There was no difference in the proportion of trees assigned to the different health classes between the low and medium deposition classes ( $\chi^2 = 6.06$ ,  $p = 0.109$ ); however, there was a difference in the proportion of trees in the various health classes between the low and high deposition classes ( $\chi^2 = 57.57$ ,  $p < 0.0001$ ) and the medium and high deposition classes ( $\chi^2 = 87.86$ ,  $p < 0.0001$ ). In each of these cases there was a greater proportion of dead trees and a smaller proportion of trees dead longer-than-1-year in high deposition class sites than in low and medium deposition class sites. No correlation was found between LFH total S and the percentage of dead trees ( $r_s = 0.328$ ,  $p = 0.111$ ).

The proportion of lodgepole pine trees in the various health classes are shown in Figure 13. A Chi-square analysis found that there was a difference in the proportion of lodgepole pine trees in the various health classes ( $\chi^2 = 16.61$ ,  $p = 0.011$ ). There was no difference in the proportion of lodgepole pine trees assigned to the different health classes between the low and medium deposition classes ( $\chi^2 = 5.25$ ,  $p = 0.154$ ); however, a difference in pine health did exist between the low and high deposition classes ( $\chi^2 = 10.006$ ,  $p = 0.019$ ) and the medium and high deposition classes ( $\chi^2 = 11.296$ ,  $p = 0.010$ ). There was a greater proportion of healthy trees and smaller proportion of trees dead longer-than-1-year in high deposition class sites than both low and

**Table 27. Total sulfur and calcium concentrations of selected species from the three trembling aspen stands. Values are means  $\pm$  95% confidence limits.**

Element	Elemental concentration (mg kg <sup>-1</sup> )		
	High S <sup>0</sup> site (site 42)	Moderate S <sup>0</sup> site (site 40)	Non S <sup>0</sup> site (site 41)
<b>Sulfur</b>			
Trembling aspen	5 200 $\pm$ 605a	4 260 $\pm$ 378b	2 300 $\pm$ 160c
Bunchberry	6 550 $\pm$ 185a	4 160 $\pm$ 491b	2 550 $\pm$ 200c
Fireweed	3 830 $\pm$ 246a	4 130 $\pm$ 456a	2 190 $\pm$ 210b
<b>Calcium</b>			
Trembling aspen	13 800 $\pm$ 1 900ab	16 000 $\pm$ 2 970a	11 600 $\pm$ 784b
Bunchberry	26 200 $\pm$ 748a	24 700 $\pm$ 1 120a	18 700 $\pm$ 913b
Fireweed	13 800 $\pm$ 1 380a	11 100 $\pm$ 790b	8 740 $\pm$ 576c

Note: Means in each row followed by the same letter do not differ significantly ( $p \geq 0.05$ ).

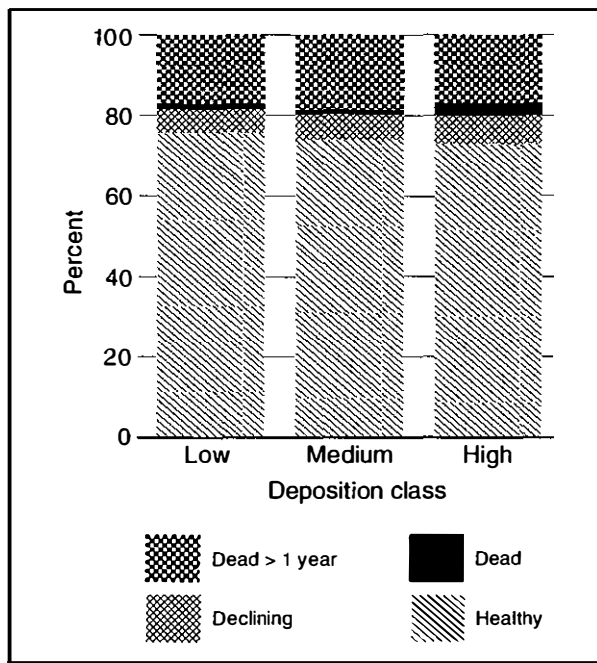


Figure 12. Proportion of all trees from the 31 mature lodgepole pine study sites in the four health classes.

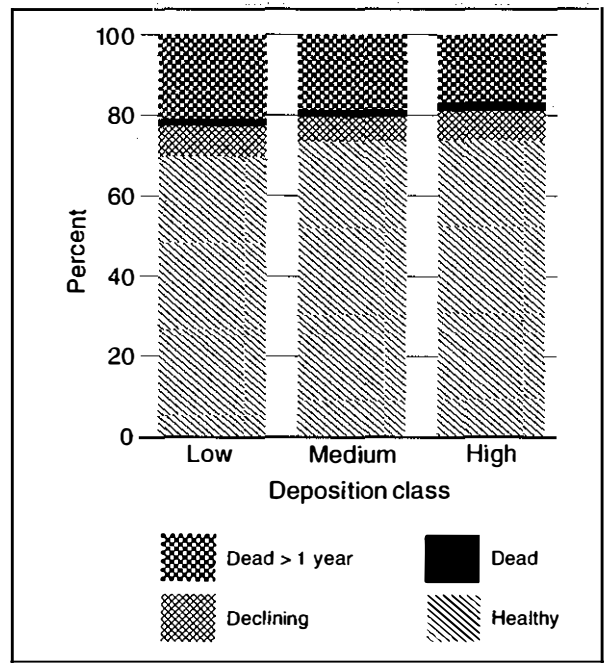


Figure 13. Proportion of lodgepole pine trees from the 31 mature lodgepole pine study sites in the four health classes.

medium deposition class sites. There was no correlation between LFH total S and the percentage of healthy trees ( $r_s = 0.405$ ,  $p = 0.164$ ); however, there was a significant negative correlation between percentage of healthy trees and S in lodgepole pine foliage from the mature pine sites ( $r_s = -0.404$ ,  $p = 0.027$ ). As S concentration decreased in lodgepole pine foliage the percentage of healthy lodgepole pine trees increased.

Figures 14–16 show the diameter classes and density relationships for healthy and dead lodgepole pine in the three deposition classes. Proportions of trees in the various diameter classes did not differ dramatically among deposition classes. This indicates that there has not been a shift in the size of trees that are dying in the medium and high deposition class sites when compared to those in the low S deposition class.

The hypothesis that there was no difference among the three deposition classes in the proportion of black spruce trees in the various health classes was tested using a Chi-square analysis. The observations in the high deposition sites were combined with those from the medium deposition sites because there were too few observations. Likewise, observations in the dead trees and trees dead

longer-than-1-year were combined because there were too few observations in the dead category. There were no differences between S impacted and S non-impacted sites in the proportion of trees in the healthy, declining, and dead classes ( $\chi^2 = 4.436$ ,  $p = 0.109$ ) for black spruce. It was not possible to perform these tests on white spruce and trembling aspen proportions because there weren't enough trees of these species in our samples.

The survey of trembling aspen health in all the mature pine stands showed that there was a trend for a greater number of dead and declining trees in the high deposition class than in the low and medium deposition classes (Fig. 17). Very few trembling aspen trees were growing in the medium deposition sites; therefore, it was necessary to examine the health of trembling aspen in sites 40, 41, and 42. The proportions of aspen trees in the various health classes growing on these sites is shown in Figure 18. There was a large difference in the proportions of healthy aspen trees in the non-S<sup>0</sup> site (96%), the moderate S<sup>0</sup> site (65%), and the high S<sup>0</sup> site (48%). There was a high amount of mortality on the high and moderate S<sup>0</sup> sites as compared to the non-S<sup>0</sup> site, 42, 25, and 2%, respectively. Correlation tests between LFH S and trembling aspen health classes could not be done because there were



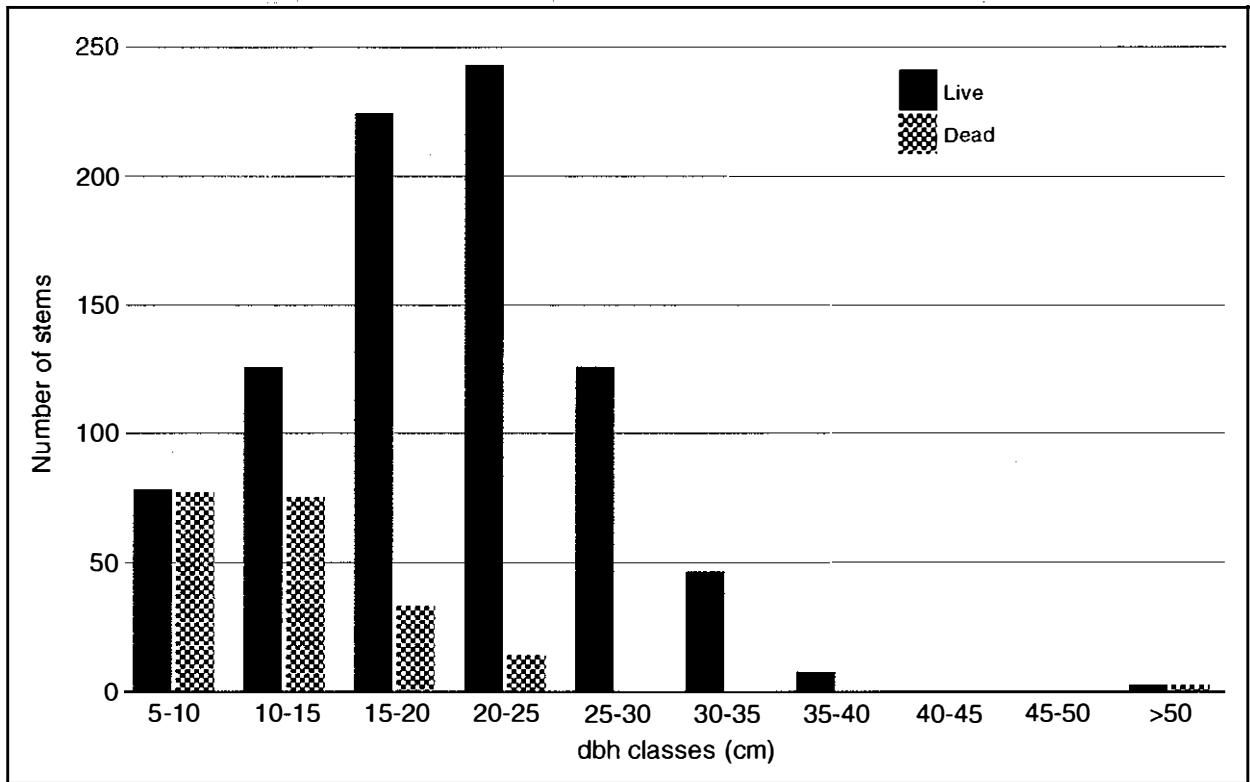


Figure 14. Size class distribution of lodgepole pine in the seven mature high deposition class sites.

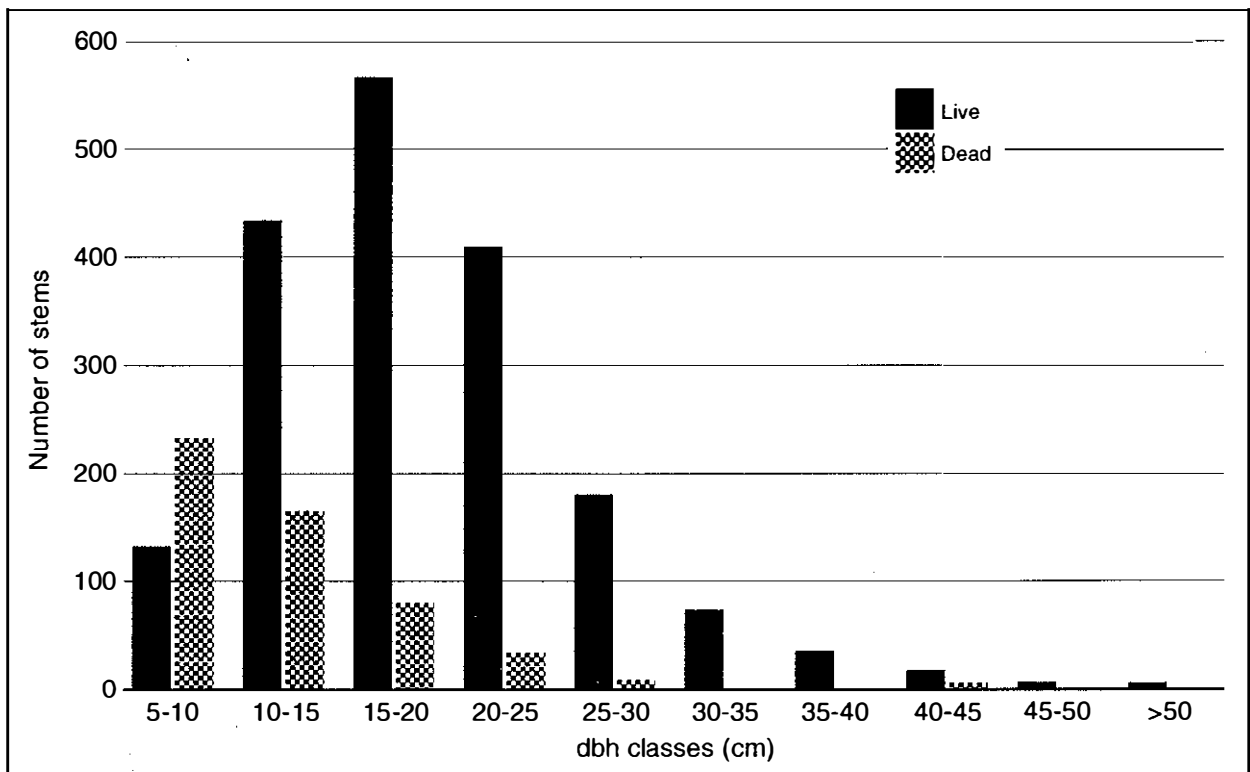


Figure 15. Size class distribution of lodgepole pine in the 13 mature medium deposition class sites.

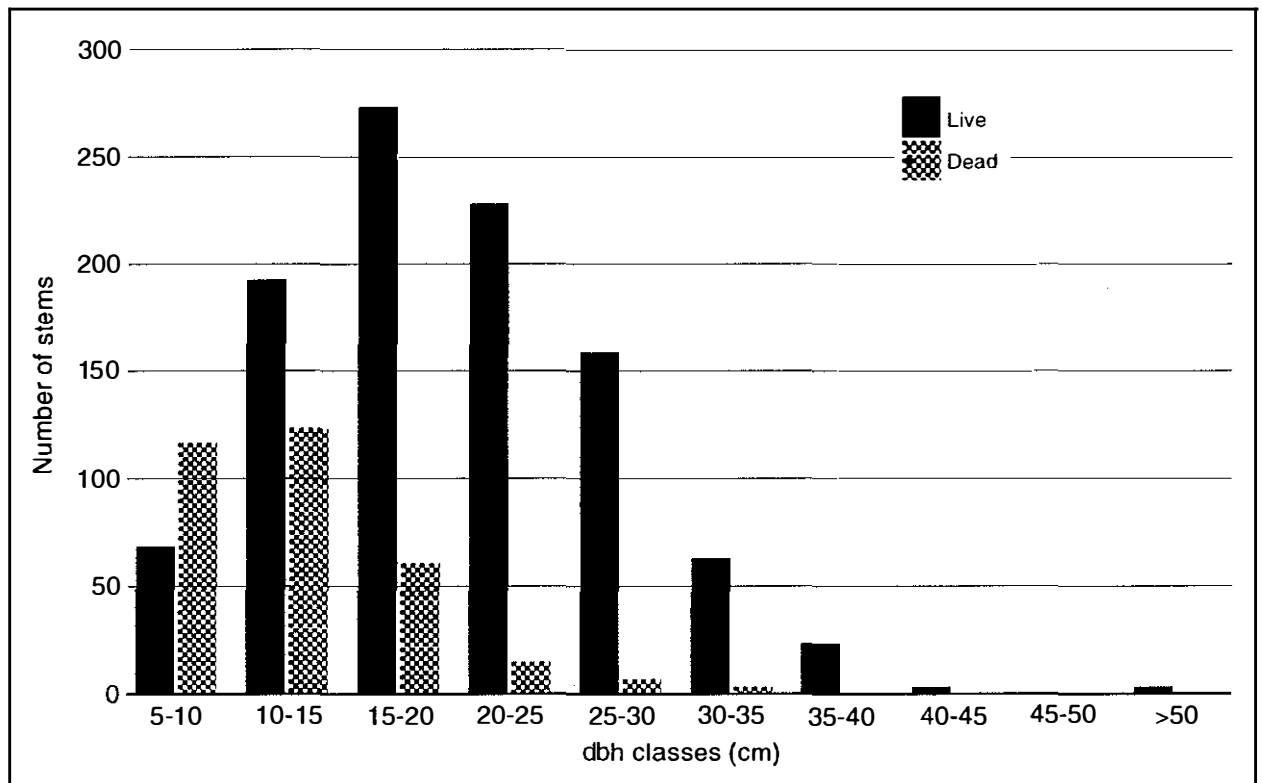


Figure 16. Size class distribution of lodgepole pine in the 11 mature low deposition class sites.

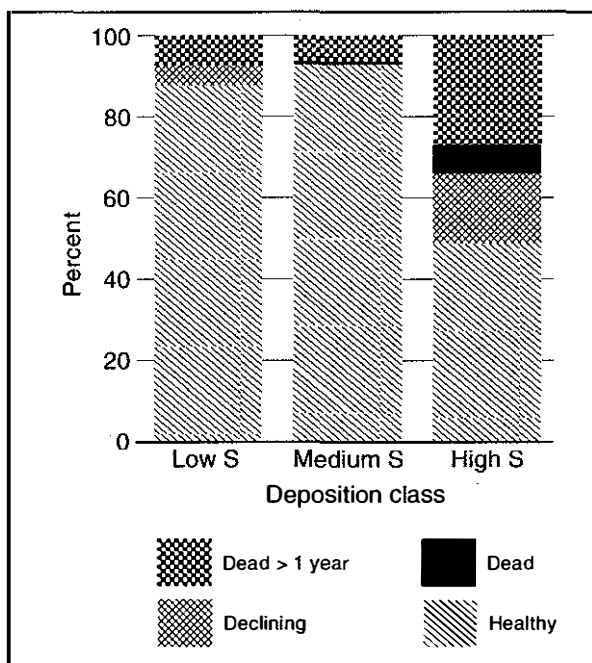


Figure 17. Proportion of trembling aspen trees from the 31 mature lodgepole pine study sites in the four health classes.

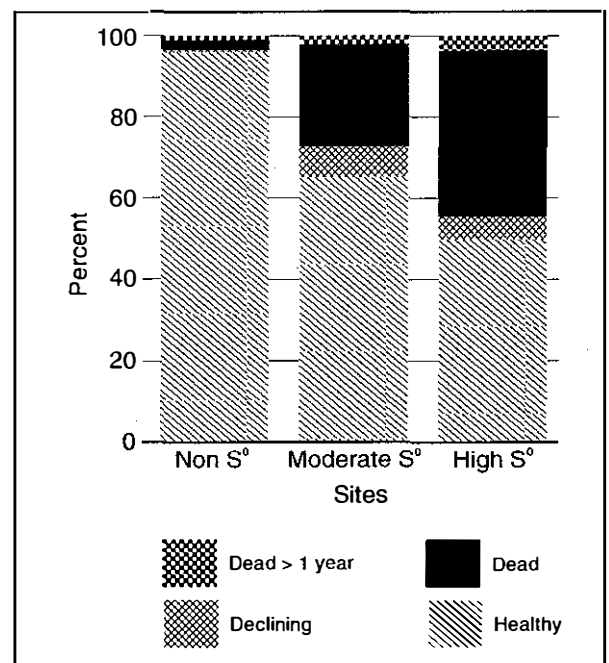


Figure 18. Proportion of trembling aspen trees from the three trembling aspen study sites in the four health classes.

too few sites to compare. Despite the small sample size, however, it appears that aspen tree health is extremely sensitive to  $S^0$  deposition.

Six young lodgepole pine stands were surveyed for health conditions. The proportions of the trees in the different health classes are shown for the three S deposition classes in Figures 19–21. All of the aspen trees examined in the young stands were healthy. The hypothesis that there was no difference in the proportions of healthy, declining, or dead lodgepole pine trees in the three deposition classes was tested using a Chi-square analysis. Declining, dead, and dead longer-than-1-year classes were combined because there were too few observations in each class to analyze separately. The hypothesis was rejected, as there was a difference in the proportions of trees in the two classes ( $\chi^2 = 17.89$ ,  $p < 0.001$ ). The high deposition site had more healthy trees and fewer dead trees than the low or medium deposition sites. Thus, there was no apparent negative impact of S in the young lodgepole pine stands. There was insufficient data to do correlation tests on young lodgepole pine health survey data.

Other factors that may have influenced forest health were examined to determine their effect on the forest health survey results. Spearman rank correlation tests were performed on a percentage of healthy and dead trees versus age, density, growth, pH, and elevation. No significant correlations were found except between the percentage of dead lodgepole pine trees and LFH pH ( $r_s = 0.466$ ,  $p = 0.01$ ) and between the percentage of dead lodgepole pine trees and elevation ( $r_s = -0.423$ ,  $p = 0.021$ ). Neither of these comparisons indicated a relationship between S deposition and lodgepole pine mortality. No correlation was found between the percentage of declining lodgepole pine and percentage of dead lodgepole pine trees ( $r_s = 0.143$ ,  $p = 0.125$ ).

Figures 22–25 show the proportion of living trees in the three deposition classes that had some symptoms of disease, insects, or physical injury. All the tree species except trembling aspen showed the same trend; proportionally more trees on the high deposition sites had some form of damage compared to trees on the medium and low deposition sites. There were insufficient trembling aspen trees in the low and medium deposition sites for a valid comparison of the proportion of aspen trees with symptoms. In the three aspen sites, however, there was a higher percentage of live trees without any symptoms or signs of damage in the high  $S^0$  and

moderate  $S^0$  sites than on the non- $S^0$  site (Fig. 26). This can be explained by the higher mortality on the high and moderate  $S^0$  sites. The trees that had died in the high and moderate  $S^0$  sites were probably those that were diseased or injured as indicated by the higher proportion of dead aspen trees with symptoms in these sites compared to the non- $S^0$  site (Fig. 27). In the non- $S^0$  site these diseased and injured trees may live longer.

### Tree Part Damage

Figure 28 shows the proportion of damage on the different tree parts for lodgepole pine trees for the three deposition classes. Damage involving stems had the highest incidence in sites from all three deposition classes. Stem and terminal damage was greater on high and medium deposition sites as compared to low deposition sites. Incidence of crown damage was similar on high, medium, and low deposition sites. Trees growing in the medium and high deposition class sites had a higher incidence of root damage than trees growing on low deposition sites. Figure 29 shows the proportion of lodgepole pine trees with stem symptoms to trees with other symptoms. Stem symptoms were the most common symptom noted regardless of which deposition class site the trees were growing in; however, there was a greater proportion of stem symptoms to non-stem symptoms in the high deposition sites. This may be a reflection of damage created by trees falling over and wounding standing trees.

There was a higher incidence of stem damage than other tree part damage in the white and black spruce on all sites. There was no difference in the proportions of stem-affected trees between deposition classes nor were there any differences in proportions of other affected tree parts.

The survey results from the three aspen sites indicated that there was a higher proportion of trembling aspen trees with root and stem damage in the high  $S^0$  site than in the non- $S^0$  and moderate  $S^0$  sites. Armillaria root rot was found on 28, 15, and 0.7% of the aspen trees in the high, moderate, and non- $S^0$  sites, respectively. Hypoxylon canker was found on 40, 28, and 2% of the aspen trees in the high, moderate, and non- $S^0$  sites, respectively. In the 1992 trembling aspen site survey *Armillaria ostoyae* (Romagn.) Herink mushrooms were found at the base of affected trees, aiding in diagnosis; however, mushrooms were not found during the 1991 mature stand survey and therefore data from the two surveys cannot be lumped and compared.

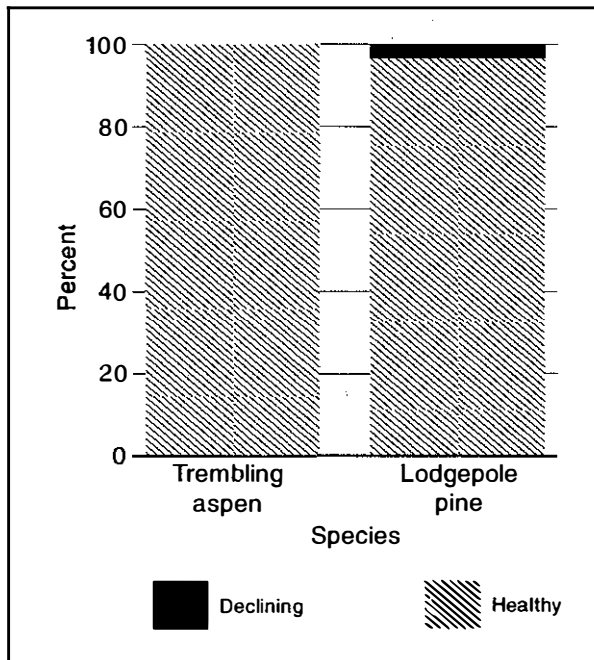


Figure 19. Proportion of all trees from the two low S deposition young lodgepole pine sites in the three health classes.

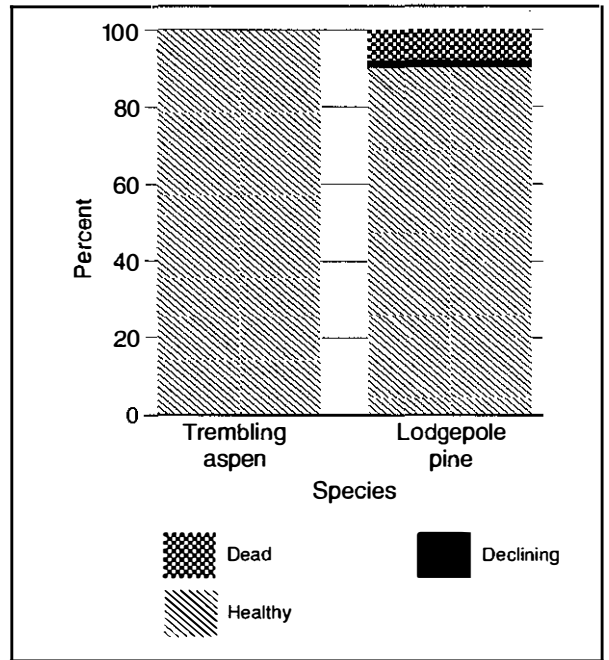


Figure 20. Proportion of all trees from the three medium S deposition young lodgepole pine sites in the three health classes.

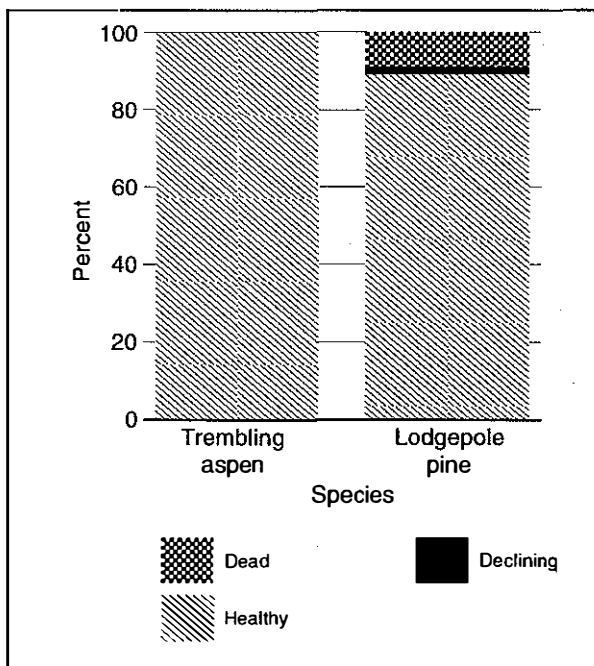


Figure 21. Proportion of all trees from the high S deposition young lodgepole pine site in the three health classes.

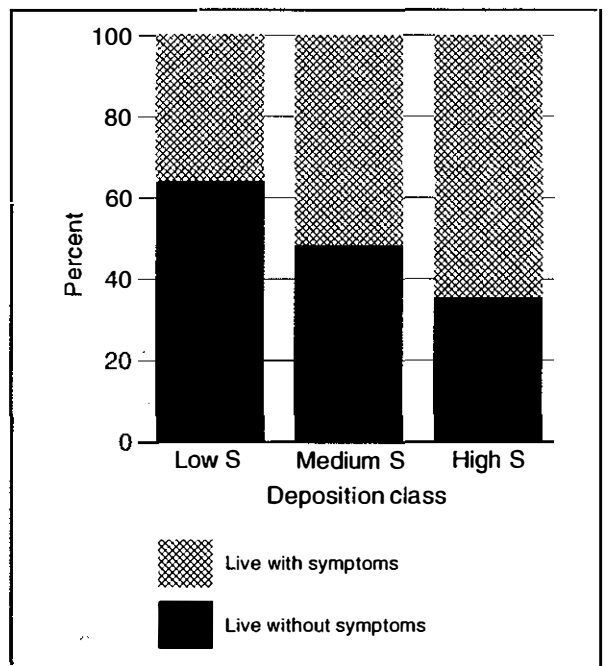


Figure 22. Proportion of all living trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class.

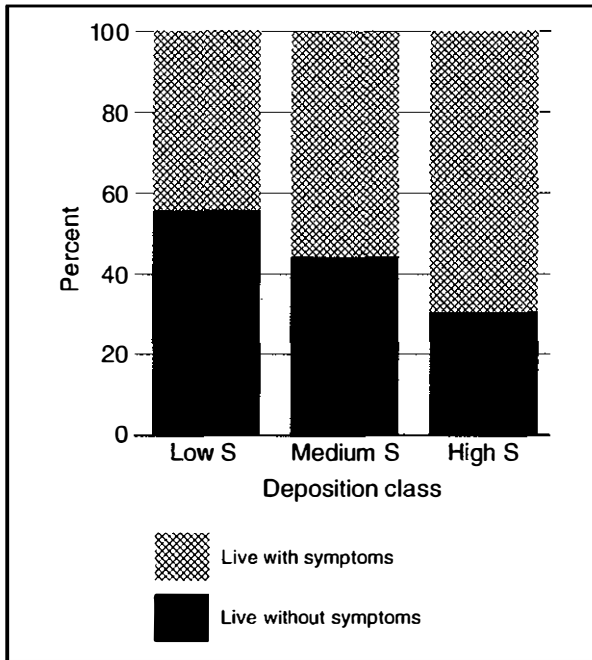


Figure 23. Proportion of living lodgepole pine trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class.

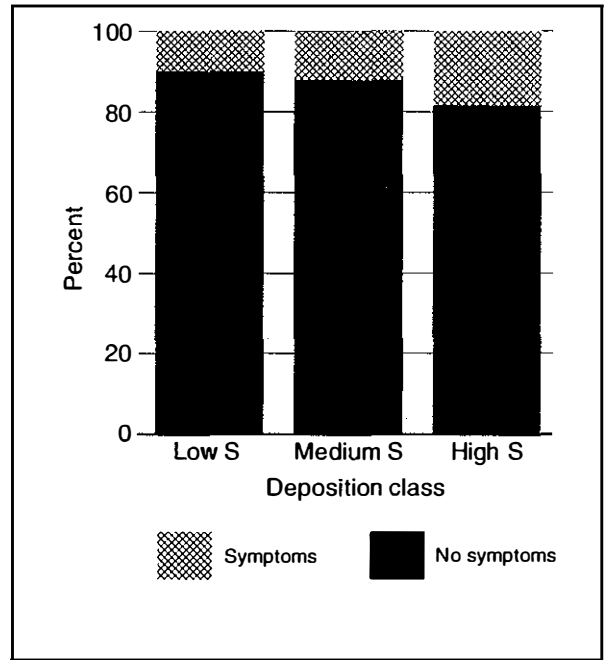


Figure 24. Proportion of living white spruce trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class.

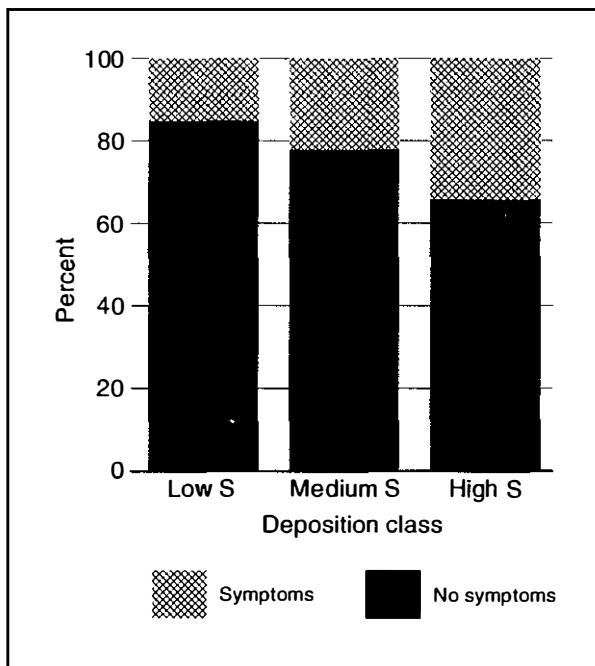


Figure 25. Proportion of living black spruce trees in the mature lodgepole pine sites with symptoms vs. no symptoms by S deposition class.

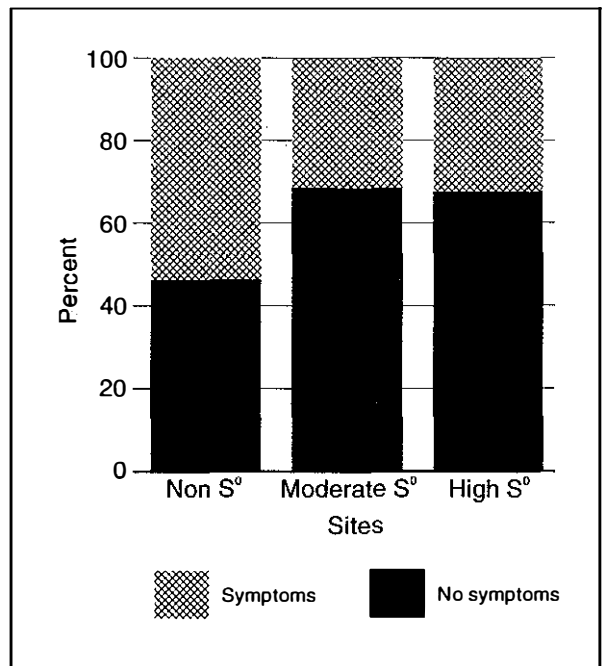


Figure 26. Proportion of living trembling aspen trees in the three trembling aspen sites with symptoms vs. no symptoms by S deposition class.

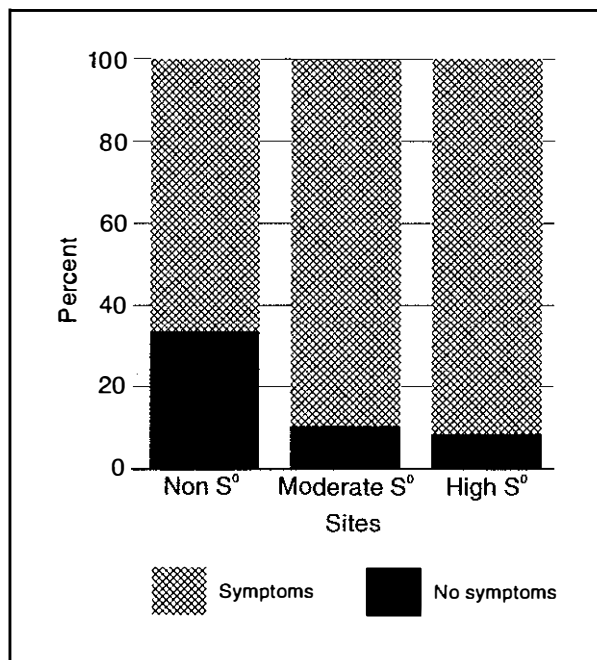


Figure 27. Proportion of dead trembling aspen trees in the three trembling aspen sites with symptoms vs. no symptoms by S deposition class.

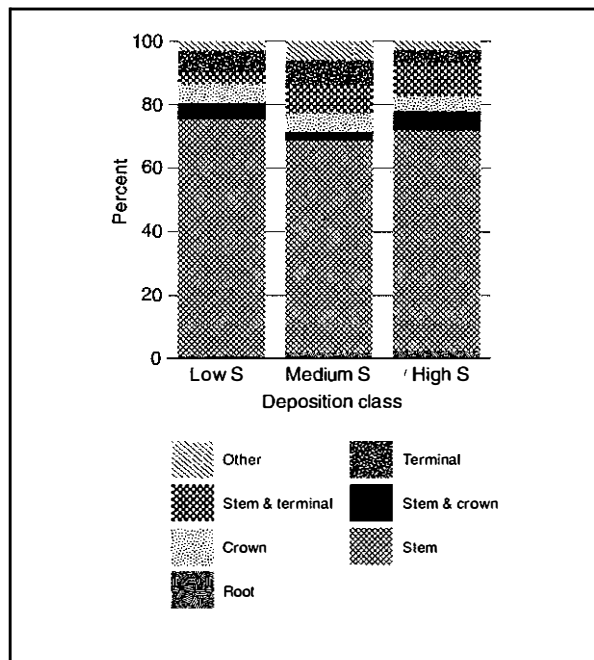


Figure 28. Proportion of total observed symptoms and damage on each tree part of lodgepole pine trees in the mature lodgepole pine sites by S deposition class.

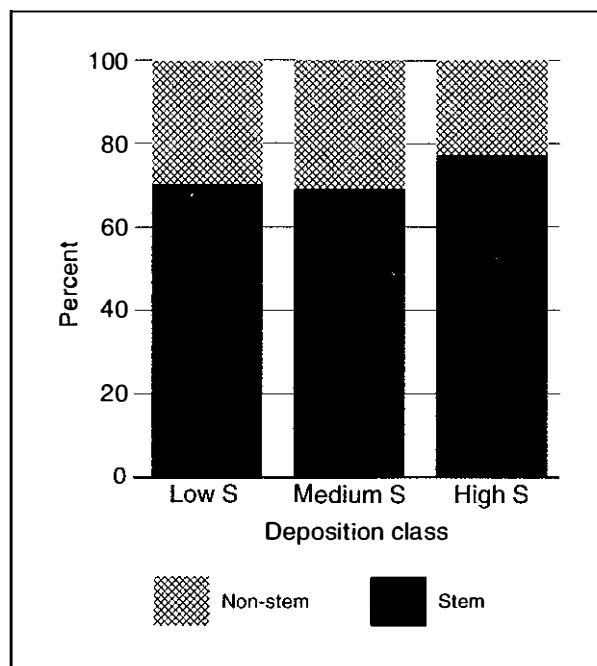


Figure 29. Proportion of stem damage to non-stem damage on lodgepole pine trees in the mature lodgepole pine study sites by S deposition class.

In conclusion, the stem was the most common tree part affected in all three deposition classes and there is evidence that there was a greater incidence of stem damage on some tree species in high deposition sites as compared to medium and low deposition sites.

### Damaging Agents

Damaging agents were broadly classified into two groups: biotic and abiotic. Some trees had multiple agents. Often no agent or symptom could be attributed to the dead-longer-than-1-year health class because these trees had a variety of secondary decomposing organisms associated with them. The agent category associated with these trees was unknown. Table 28 shows the incidence of the agents as a percentage of trees in each deposition class. The proportions of damage from biotic and abiotic agents did not differ significantly between the low, medium, and high deposition classes ( $\chi^2 = 4.27$ ,  $p = 0.118$ ).

There was no correlation between any of the agents and LFH S concentrations, nor were agents correlated with S in the pine foliage.

**Table 28. Incidence (%) of damaging agents in the three sulfur classes of the 31 mature pine stands**

Damaging agent	Deposition sites		
	High	Medium	Low
Pine needle cast	8.3	5.3	5.1
Spruce needle cast	0	5.3	1.3
Spruce needle rust	0	1.0	0.8
Spruce broom rust	2.0	2.2	0.5
Armillaria root rot—all species	1.2	0.7	0.9
Armillaria root rot—lodgepole pine	1.2	0.8	1.4
Armillaria root rot— all species, dead trees only	32.4	35.5	37.7
Atropellis canker	0.4	0.4	0
Western gall rust	1.3	0.3	0.4
White rot of aspen	3.0	9.5	5.1
<i>Peniophora polygonia</i> (stain and decay)	5.1	0	0
Red heart rot	0	0	0.1
<i>Trichaptum abietinus</i>	0	0	0.07
Cytospora canker	1.0	0	0
Hypoxyylon canker	0	0	0.9
Root collar weevil	2.4	2.3	1.4
Unidentified insect damage	0.4	0.4	0.2
Bark beetle damage	0	0	0.1
Cooley spruce gall aphid	0	0	0.3
Poplar stem borer ( <i>Saperda calcarata</i> )	0	0	0.9
Terminal weevil ( <i>Pissodes terminalis</i> )	0	0	0.1
Pitch moth	0.1	0	0
Stem—scar or cause unknown	47.4	36.2	22.4
Crown damage—unknown cause	7.8	8.1	7.5
Terminal damage—unknown cause	6.5	7.5	3.5
Suppressed trees	3.1	0.5	0.5
Mechanical damage	0.5	1.1	1.4
Frost injury	0.2	0	0.05

The incidence of any particular known agent in any of the deposition classes was relatively small. Armillaria root rot and Hypoxyylon canker were associated with dead trees in the aspen sites. The proportion of Armillaria root rot on dead trees was similar in all lodgepole pine sites. Healthy and declining trees were not examined for Armillaria root rot and therefore conclusions about incidence of this disease and S deposition cannot be made.

Stem damage was the most common form of damage on any of the sites. Trees with mechanical scars or damage from an unknown agent were common. It is not known whether these scars or damage were due to a biological agent that could not be

detected with the survey methods employed or whether these scars represented old infections or injuries that had healed over. Stem damage from scars or unknown agents increased from the low (22%), medium (36%), to high (47%) deposition sites. This increase in stem damage was probably related to injuries sustained from falling trees or branches, and thus may be an indirect measure of past mortality.

*Armillaria ostoyae*, causal agent of Armillaria root rot, was the major damage causing agent in the young lodgepole pine stands in all three deposition classes. Aspen leaf blight was found on young trembling aspen trees in all six of these sites (Table 29).

**Table 29. Incidence of major pests in the young lodgepole pine stands in the three deposition classes**

Agents (%)	High sites	Medium sites			Low sites	
	C	B	D	E	A	F
Armillaria root rot (all trees)	2.7	9.2	7.5	6.4	8.9	3.8
Armillaria root rot (dead trees)	92.0	67.0	95.0	94.0	83.0	57.0
Root collar weevil (all conifers)	0.3	5.0	0.4	0.4	1.1	3.2
Root collar weevil (dead conifers)	8.3	33.0	5.4	5.9	11.0	43.0
Western gall rust (all pine)	0	1.4	1.1	0.5	0.6	39.0
Needle cast (live pine)	0	16.0	0	0.2	0	0
Terminal weevil (live conifers)	0	0	0.6	0.9	0.6	0
Aspen leaf blight (live aspen)	100.0	60.0	100.0	100.0	100.0	100.0

The Canadian Forest Service's Insect and Disease Survey unit have monitored insect and disease pest conditions in the area since 1950. The agents that have been found in this study have been recorded as being present in the area before 1970.

## Pine Growth

### Analysis of Increment Cores

An analysis of variance of the indices derived from increment cores indicated that there is no evidence to suggest that growth depends on deposition class (Table 30). The interaction between decade and deposition class was not significant. Only the decade was a significant source of variation, and the behavior of the index is as would be expected: as the trees age the growth index declines. In the decade after 1970, radial growth of all trees declined 3% of the growth rate in the decade starting in 1961. A decline of a further 9% was seen for the decade beginning in 1981. These changes reflect the changing growth capacity of the trees and the variation in growing conditions in the period covered by the analysis.

### Analysis of Annual Volume Increments

All factors examined were significant sources of variation in the index of annual volume increment (Table 31). Tree age is significant because the volume increment of trees increases exponentially as they grow. Although this value eventually declines, the age at which this occurs was not attained in any of the trees sampled. Site variation in the rate of tree growth is a random variable over which there is little control, but it is useful in the analysis because it isolates site effects in the

response variable. That leaves the deposition class, the decade, and the interaction between them for interpretation. An examination of the least squares mean annual volume increment index indicates that the trees increased their growth 35% for the first decade of gas plant operation and a further 24% for the next decade (Table 32). The growth index over the three decades was not statistically different for trees growing in the low and medium deposition class sites. There was, however, a depression in this index for trees growing in the high deposition class sites. The volume growth appears to be about 25% below that of the expected value.

There is considerable variation in the index of annual volume growth among sites within deposition classes. Thus, whereas the sites in the low deposition class increased growth consistently in the three decades studied, the sites in the medium deposition class also increased growth consistently but this change was not significantly different in the last two decades. The same pattern of change is evident in the high deposition class sites. This result may be due in part to the anomalous reaction of certain sites within the medium deposition class. Sites 3 and 13 grew at substantially lower rates (60 and 53%, respectively) than the class mean. Similarly, site 30 grew at a substantially higher rate (51%) than the mean for high deposition class stands.

Over time the stands have increased their annual volume growth, even in the high deposition class stands; however, this rate is not as great in the stands of the highest deposition class as it is in stands in the other two deposition classes. Certain sites, notably 3, 13, and 30 contribute substantially to variation within the index of annual volume increment.



**Table 30. Analysis of variance of aggregated standardized radial increments for the decades after 1960**

Source	DF	Mean sq.	F value	Pr > F
Deposition class (D)	2	0.01370	0.71	0.4934
Decade (Y)	2	0.10226	5.32	0.0068
Interaction (D × Y)	4	0.00571	0.30	0.8792
Error	81	0.01923	–	–

**Table 31. Analysis of variance of annual volume increment index for decades after 1960**

Source	DF	Mean sq.	F value	Pr > F
Tree age	1	2.8152	24.24	0.0001
Deposition class	2	1.2217	10.52	0.0001
Site (deposition class)	27	0.4960	4.27	0.0001
Decade	2	2.6861	23.13	0.0001
Deposition class × decade	4	0.4698	4.05	0.0039
Error	179	0.1161	–	–

### Analysis of Specific Volume Increments

The specific volume increment index is a measure of how efficiently the tree accumulates stem wood. It is less subject to endogenous factors influencing tree growth and more sensitive to environmental perturbation. As a reflection of this, tree age and the decade in which the growth ring was laid down were not significant sources of variation in the index (Table 33). There is, however, a trend suggesting an overall decline in specific volume increment in the three decades considered in this study (Table 32). The local site within deposition class and the deposition class were, however, significant sources of variation (Table 33). The interaction between deposition class and decade in which growth occurred was not significant.

The results of the analysis indicate that there is a significant depression in the efficiency of stem wood accumulation in the medium and high deposition classes of 9 and 12%, respectively. The trend indicated in the sites suggest that trees growing in the medium and high deposition sites were 84 and 81%, respectively, as efficient as trees growing in the low deposition sites two decades after the gas plants started operating. Again there is considerable variation in the pattern of change among the sites

within the deposition classes. Trees in sites 3 and 13 were far less efficient than the others in the medium deposition class and those in site 30 were more efficient than the others in the high deposition class.

In an attempt to determine what might be associated with the change in efficiency and consequently the change in growth rates, the relationship between the deposition of S in the LFH layer and the annual specific volume increment index for sites were compared. The Spearman rank correlation coefficient was  $-0.175$  and was not statistically significant ( $p > t_{30} = 0.353$ ). There is thus insufficient evidence to implicate sulfation, by itself, as the cause of reduced efficiency. Unfortunately a test to isolate the effects of sulfation and  $S^0$  deposition is not possible with the current information unless the interpolated data from the sulfation isopleths in Figure 5 is used. Examination of Figure 7 also suggests that sulfation alone is not likely to be the explanation of the reduced efficiency. The anomalous site 30 has a high sulfation value but is also growing above the norm for the group. It is not presently possible to identify the causes of the association between loss in efficiency and deposition class.

The observed difference in trends of annual volume increment and specific volume increment

**Table 32. Least square means of specific volume and annual volume increment indexes for decades after 1960 and deposition classes**

Deposition class	Decade	Least square mean indexes <sup>a</sup>	
		Annual volume increment	Specific volume increment
Low	All	1.268a	1.007a
Medium	All	1.185a	0.911b
High	All	0.930b	0.883b
All	Pre-1972	0.816a	0.982a
All	Pre-1982	1.161b	0.950ab
All	Pre-1992	1.405c	0.870b
Low	Pre-1972	0.786a	0.979a
Low	Pre-1982	1.345b	1.072a
Low	Pre-1992	1.673c	0.970a
Medium	Pre-1972	1.022a	1.002a
Medium	Pre-1982	1.184ab	0.898ab
Medium	Pre-1992	1.348b	0.835b
High	Pre-1972	0.640a	0.965a
High	Pre-1982	0.956b	0.878a
High	Pre-1992	1.192b	0.807a

<sup>a</sup> Least square means in a group followed by the same letter are not significantly different at  $p \geq 0.01$ .

**Table 33. Specific volume increment index for decades after 1960**

Source	DF	Mean sq.	F value	Pr > F
Tree age	1	0.0264	0.70	0.4041
Deposition class	2	0.2312	6.13	0.0028
Site (deposition class)	27	0.1214	3.22	0.0001
Decade	2	0.1047	2.77	0.0658
Deposition class x decade	4	0.0686	1.82	0.1286
Error	179	0.0377	–	–

**Table 34. Analysis of variance of cumulative volume increment to age 11 years for trees germinating before and after the sour gas plants opened (era)**

Source	DF	Mean sq. $\times 10^6$	F value	Pr > F
Deposition class (D)	2	2.0374	1.95	0.1712
Era (E)	1	21.4848	20.57	0.0003
Interaction (D $\times$ E)	2	2.1870	2.09	0.1523
Error	23	1.0447	–	–

in relation to the three deposition classes requires some explanation. The annual volume increment is less sensitive than specific volume increment in detecting change because of the pattern of tree growth along the stem. Volume increment at a particular segment of a tree is calculated as the product of the radial increment and the surface area of the cambium at that location. The largest surface area is at the base of the tree, but this is also the location where radial growth is smallest. By contrast, the largest radial increments are in the crowns of trees, but because of taper in tree stems, the surface area generating this growth is smaller. Thus the annual volume increment is a balance involving the contributions of the large radial increment at tree tops (reduced by the effects of small stem surface area) and the contributions of smaller increments at the tree base (magnified by the larger stem surface area). The specific volume increment ignores this influence of surface area and is simply the average radial increment over the whole stem without being influenced by stem size at the location at which the growth is generated. Thus a change in the radial growth of the tree in the tree crown (where most growth reductions due to environmental agents occur) will have a proportionally larger influence on the specific volume increment (unweighted by surface area) than it will on the annual volume increment (where the effect of change will be reduced by the influence of a small stem surface area).

There are other measures that may be derived from stem analyses. One measure, the oblique sequence, is the profile of tree growth along the tree stem for a particular year or interval of years. The radial growth of trees is greatest in the upper crown. Factors affecting the efficiency of the crown are most likely to affect the productivity in this portion of the stem. The result is that the general level of the oblique sequence and its shape will be modified. The disadvantages of the method are that the profile is also costly to obtain. These measures are highly correlated with the three measures used in this study and are useful in detection of errors and missing rings in datasets; they were not used in this analysis.

Two other measurements are routinely obtained from the dissection of tree stems. These are the cumulative height curve and the cumulative volume curve. Their utility depends on the interval at which the growth rings are sampled. For work on large trees they are most useful in generating yield curves. They are less sensitive in detecting environmental impacts than the annual increment measures previously described.

## Comparison of Early Growth in Young and Old Stands

In comparing the early growth of trees in stands established long before the gas plants began operation with growth of stands that originated after the plants were opened, the impact of S deposition on site productivity can be evaluated.

The cumulative volume growth of stands originating in several different decades before the gas plants began operation was compared to those originating in the two decades after the plants opened. A simultaneous comparison between those growing in the various deposition areas provide a means of assessing if sulfur has had an impact on site productivity. Analysis of variance indicated that whereas era of stand establishment had a significant influence on the cumulative volume increment to age eleven, neither the deposition class, nor its interaction with era had any statistically significant effect on this tree response (Table 34). It would appear that current growth rates are almost three times what they were in the decades prior to plant operations (Table 35). There is no suggestion of a trend between growth and deposition class. If any statement could be made, one

**Table 35. Least squares means of cumulative volume to age 11 years for periods before and after the opening of the sour gas plants (era) and deposition class**

Deposition class	Era	Cumulative volume increment
All	After	3.190a
All	Before	1.098b
Low	All	2.561
Medium	All	1.655
High	All	2.216
Low	New	3.043
Low	Old	2.080
Medium	New	2.550a
Medium	Old	0.760b
High	New	3.977a
High	Old	0.454b

Note: Least square means in a group followed by the same letter are not significantly different at  $p \geq 0.01$ .

might suggest that the growth improvement is highest in the highest deposition class.

This aspect of the study should be interpreted with caution, however. Only six pairs of (old and new) stands were compared. This sample size is inadequate because there were few young stands available close to the study sites. One of the locations where sulfation and deposition levels were

highest was site 30; this site is, in many respects, anomalous. The interpretation of this result is further complicated by the uncertainty of the early stand development of the old stands studied. The data do not indicate a negative impact with respect to cumulative volume growth; however, the young lodgepole pine sites may now have an improved growing potential.

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## SUMMARY

The environmental consequences of processing sour gas in west-central Alberta, which involves the extraction and shipping of  $S^0$ , has been a concern to industry and the public. The recovery of S from natural gas was about 97% efficient in 1981 and was about 99% efficient by 1991. In absolute terms, the average daily emission of S was 70 t in 1981, but this declined to 25 t by 1991. The physical processing of  $S^0$  for shipment has resulted in  $S^0$  dust being deposited in forest stands up to 6–8 km from the processing plants and handling facilities with high deposition levels ( $>5 \text{ t ha}^{-1}$ ) occurring within 1 km from the source. The dominant vegetation of the landscape is a mosaic of lodgepole pine stands.

A study was initiated in 1981 to assess the impact of emissions of  $\text{SO}_2$  and  $S^0$  on mature lodgepole pine stands near the Ram River and Strachan gas processing plants. Soil chemistry and species richness were assessed at 26 study sites then and remeasured in 1985. The study sites were re-assessed and expanded in 1991 and 1992. Additional sites were added to the study site network to replace sites that had been destroyed and fill in gaps evident in the original coverage. In addition, forest health and tree growth studies were instituted at each study site in 1991. Additional LFH samples (20) were taken to include the forest health plot network established at each study site in 1991. Surveys were initiated in natural lodgepole pine stands, which were established after the gas plants began operations and in stands dominated by trembling aspen. In total, 31 mature lodgepole pine stands, 6 young lodgepole pine stands, and 3 trembling aspen stands were assessed.

Overall, there has been a decline in the concentration of  $S^0$  in LFH between 1985 and 1991. There was a decrease at three sites (1, 2, and 12), and one site (5) remained unchanged. In 1991 only five sites (1, 2, 5, 12 within 500 m of the plants, and 14) had

consistently detectable levels (100 ppm) of  $S^0$ . The inclusion of site 14 and possibly site 13, where one sample had detectable levels of S, indicate that the area of  $S^0$  deposition now extends 4–6 km from the gas processing plants. In addition, the S concentrations in either the LFH or foliage was elevated in sites 3 and 23. All these sites, including site 30 established in 1991, are within 6 km of the plants. Some of the S loading in the LFH and foliage may be due to gaseous emissions; however, gaseous emissions and  $\text{SO}_2$  deposition declined during the study period. Elemental S block breakup, forming, and shipping (sources of  $S^0$  dust) peaked between 1986 and 1988.

A combination of 1991 total S concentration in the LFH and  $\text{SO}_2$  deposition interpolated from a network of sulfation stations was used to objectively assign study sites to S deposition class. Three groupings were designated as high (7 stands), medium (13 stands), and low (11 stands) deposition classes. In 1992, mature aspen stands were chosen to represent a range of S deposition. Six young lodgepole pine stands were established in the low (2), medium (3), and high (1) deposition classes. These groupings were used in the analyses to determine the impact of S deposition on soil and foliar chemistry, tree growth, and forest health.

Analysis of variation among the various indicators of soil and foliar properties indicated that there were no detectable differences among sites in the low and medium S deposition classes on any of their chemistry except for pH and two exchangeable cations: Ca and Mg. Calcium and Mg concentrations were lower in the medium than in the low deposition class. This corresponds to a lower pH in the LFH soil horizon in the medium deposition class. In the decade over which observations were made, however, there have not been consistent changes in the pH or Ca and Mg concentration at the sites in the medium deposition class. It is

therefore reasonable to conclude that the differences observed between the medium and low deposition classes are not associated with elevated S concentrations in the medium deposition class.

Trees were classified as either long-dead, recently dead, declining, or healthy in the forest health survey. There was a greater proportion of recently dead trees in sites located in areas of high S deposition. There was, however, no difference in this proportion between stands in the low and medium deposition classes. Yet, there was a greater proportion of healthy lodgepole pines in high deposition class stands. This may be due to the accelerated demise of declining trees in these stands. This was corroborated by the increased incidence of stem damage in the surviving healthy trees in the high S deposition areas. By assessing more than 5700 trees in the mature lodgepole stands, no evidence was found of an increase in insect and disease activity associated with S loading. There was no correlation between S concentration in the LFH horizon and the proportion of recently dead lodgepole pine trees in the sites surveyed. Nevertheless, there was a significant negative correlation between S concentration in the current foliage of lodgepole pine and the proportion of healthy trees in the stand. Other environmental factors, such as elevation (which was inversely correlated with pH) was also associated with survival.

Although the sample sizes were small, reflecting the small proportion of the area occupied by these stands, there was a greater proportion of healthy lodgepole pine trees in the young stand growing in the high S deposition area than in stands growing in the other deposition classes. Again, there was no indication of increased insect and disease activity associated with S deposition class.

Some impacts were, however, detected in trembling aspen stands. The proportion of declining and recently dead trees in the high S<sup>0</sup> and moderate S<sup>0</sup> sites was higher than in the non-S<sup>0</sup> site. The elevated proportion of affected trees was associated with an increased incidence of stem and root diseases of aspen. There was no association between the

incidence of mortality of young aspens growing in the young lodgepole pine stands surveyed and S deposition class. The number of aspen stands surveyed was small, however, and these conclusions should be viewed with caution. Further study of the effects of S deposition on aspen stands is warranted.

Analysis of tree growth using radial increments at breast height revealed no impact associated with S deposition class. Radial growth on the lower bole of older trees is too insensitive to detect changes associated with subtle environmental impacts.

Annual volume increments in the trees studied have not declined in association with S deposition. The rates of increase in annual volume increment have declined in the highest S deposition class relative to the tree response in the other two classes. The specific volume increment, which is an index of physiological efficiency at which stem wood is accumulated is more sensitive at indicating changes in tree growth than annual volume increment. The specific volume increment was depressed in both the medium and high deposition classes when compared to the mean of trees growing in the areas of low S deposition.

There is no evidence of reduced cumulative volume growth to age 11 years in lodgepole pine trees established since the gas plants began operating relative to stands originating 60 to 100 years ago. The evidence from the six young stands sampled suggest that there is elevated volume growth in the areas of higher S deposition. This must be viewed with caution, however, because it is not possible to reconstruct early densities of the presently mature stands.

The only indicators of extensive S impacts on the major forest community detected to date are the elevated S concentrations in the LFH and foliage, the proportion of healthy lodgepole pines, and a depression in the annual specific volume increment. This study of forest health near two point sources of air pollution has shown that there is no widespread forest decline in the surrounding forest outside of the areas dusted with S<sup>0</sup>.

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