ACIDITY OF OPEN AND INTERCEPTED PRECIPITATION IN FORESTS

AND EFFECTS ON FOREST SOILS IN ALBERTA, CANADA

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ABSTRACT

Emissions of sulphur dioxide appear to have an acidifying effect
on grossfall (open rainfall), throughfall, stemflow and soil
solution at sites near major sources. Resulting effects on
soil chemistry include elevated extractable acidity and
aluminum and depressed exchangeable bases, especially calcium
and magnesium.

These changes are mostly in the incipient phases in the study
area.

INTRODUCTION

Approximately half a million tons of SO_2 are emitted annually into
the atmosphere throughout Alberta, mostly as a result of residual in-
efficiency of sulfur recovery from sour (H_2S containing) natural gas.
An earlier paper in these proceedings describes the regional effects of
these emissions on acidity of precipitation (Nyborg, Crepin, Hocking,
and Baker 1976).

Several large point sources of SO_2 are located in forested areas
where they have strong local influences (Baker, Hocking and Nyborg 1973).
Preliminary laboratory studies showed that solution and transformation
of sulfur compounds in the soil is likely to greatly influence the
solubility, mobility and distribution of soil minerals and nutrients
(Baker, in press).

This article reports early field data on chemistry of rainfall
from two localities in Alberta affected by SO_2 emissions, and effects
on forest soils from one of those localities.

METHODS

One locality surrounded two major sulfur-extraction natural gas treatment plants near Rocky Mountain House, Alberta (lat. 52° 20' N, long. 115° 20' W), referred to as RMH, and is an area where soil studies are progressing. The other locality surrounded a processing plant on the Athabasca Oil Sands of northeastern Alberta near Fort McMurray (lat. 56° 50' N, long. 112° 5' W), and will be referred to as the GCOS area.

Experimental sites were located at varying distances from the SO_2 sources. Each site was equipped with plastic rainfall traps that included units to collect grossfall (uninterrupted rain), stemflow, and three throughfall (dripping off foliage) collection units. Collections were made throughout the growing season at weekly intervals or after each major rain event. Fallen snow was collected once from each site during the winter. All samples were analyzed for pH and sulfur content after storage at 1°C for varying periods.

At the RMH sites, soil was sampled from the litter surface (L-F-H), and at 0-5 cm, 5-15 cm and the 15-30 cm depths in the autumn when the soils were very wet. Samples were stored in plastic bags in cold rooms (1°C) until analyzed (for four months).

Field-moist samples were used in all analyses. Eight grams of moist L-F-H and 16 grams of moist mineral soils (approximately equivalent to 4 and 12 grams on an oven dry basis) were first extracted alternately with 25 ml each of warm (40°C) water and 1 n KCl, until 250 ml of leachate was collected. Each addition of water or salt was allowed to completely drain (one hour) before the addition of the next. Extraction was carried out using a V-shaped funnel equipped with a quantitative filter paper and a hardened #50 Whatman filter disc on the surface of the sample. These water-KCl extracts were analyzed for pH, exchangeable acidity, Al, Ca, Mg, Mn, Fe, N, and P.

Following the KCl extraction, selected samples from sites near to and distant from the SO_2 source were sequentially further leached with 100 ml each of (40°C) 0.5 n NaF, 0.1 n NaOH and 0.5 n H_2SO_4 (after Jackson, 1958), for P and N determinations. Aliquots were digested with concentrated H_2SO_4 and a Cu catalyst. No attempt was made to differentiate between organic and inorganic phosphorus and nitrogen. Evaluations were made of the total amounts removed by the various extractants.

Measurements for pH and KCl-extractable acidity were carried out on an automatic Metrohm E436 potentiograph. Titrations were done with 0.04 n Ca(OH)$_2$. The normal and a differentiated titration curve was carried out for each extract. Measurements of Al, Ca, Mg, Mn and Fe in KCl were made using a P.E. Atomic Absorption equipped with a digital concentration readout. Total phosphorus content in each of the four extracts was determined by the molybdate method using ascorbic acid as a reducing agent. Nitrogen evaluations were obtained by distillation

of the digested extracts into boric acid and titrated to pH 4.7 with standard H_2SO_4 using a dead-stop Radiometer titrator.

Where sulfur was determined, the Dean modification (Dean 1966) of the method of Johnson and Nishita (1952) was used.

RESULTS

The pH values of grossfall, throughfall and stemflow for each major rainfall event of the 1974 growing season are shown in Fig. 1 for 3
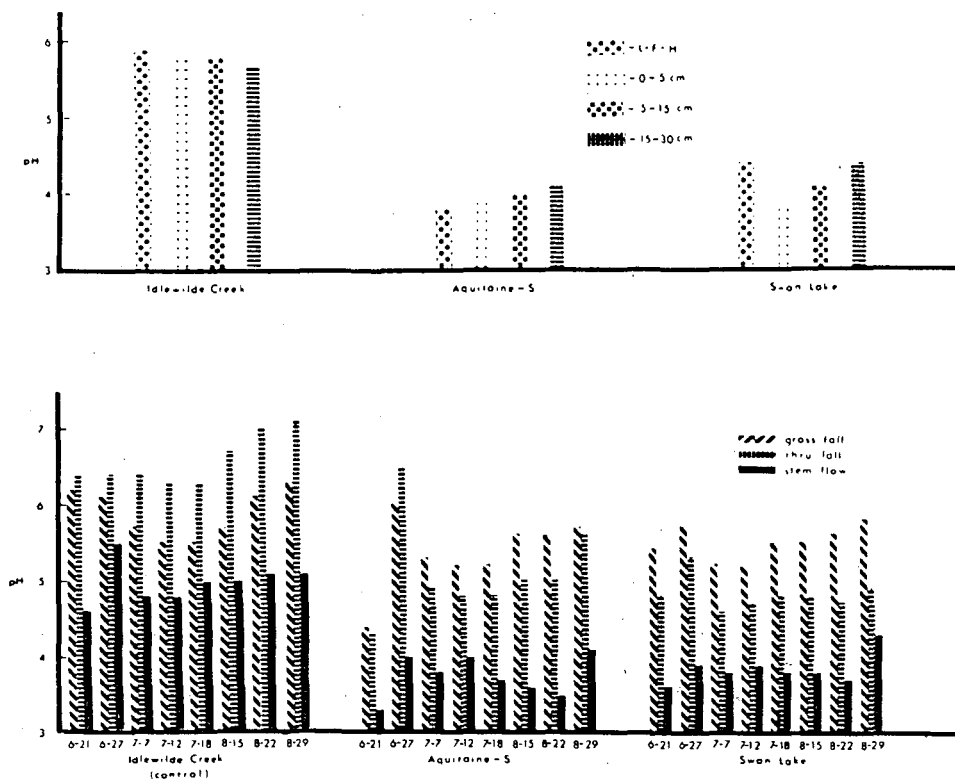

Figure 1. Change in pH of gross fall, thru' fall and stem flow solutions from control and S-contaminated sites during the 1974 sampling period; soil pH values (KCL extracts) for corresponding sites are also given.

representative sites in the RMH area, one distant and two relatively near the SO_2 source. Included in Fig. 1 are pH values of KCl extracts from various soil layers of the given test sites. Average pH values of throughfall and stemflow for the entire season are shown in Fig. 2 for sites near the SO_2 source in the RMH area, and are given in Table 1

5.3, 4.1
•

↓
⊕→N
Scale
1 cm = 1.3 Km

4.9, 4.2
•

5.7, 3.9   5.0, 3.9
•   •

GAS PLANT
▰

"Controls"(40 Km W)
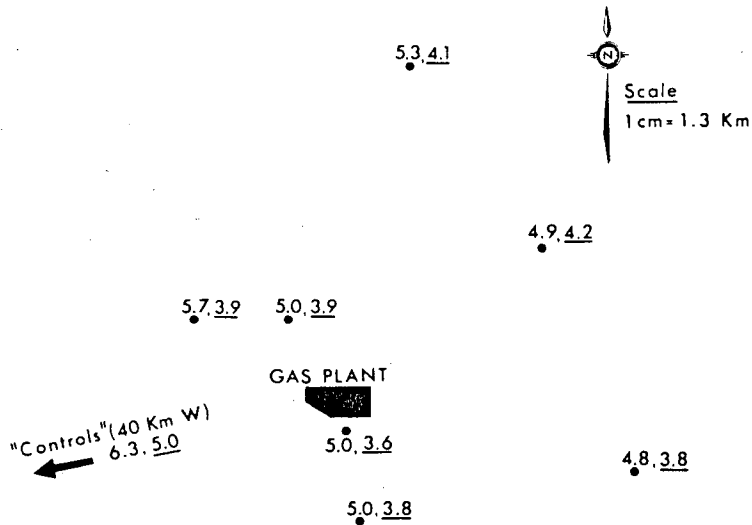6.3, 5.0
←

5.0, 3.6
•

4.8, 3.8
•

5.0, 3.8
•

Figure 2. Average throughfall and stemflow
(underlined) pH, June 15 - September 5, 1974,
of selected sites in the RMH area.

Table 1. Average pH of Crossfall and Spruce Stemflow During
July - September, 1974; for sites in the GCOS area.

Site	Distance (direction) From SO₂ source (km)	pH	
		Crossfall	Stemflow
Controls (2)	24,56 (SW)	6.4	6.4
GCOS - 1	3.2 (NW)	6.5	4.7
GCOS - 2	8 (NW)	6.5	4.7
GCOS - 4	2.2 (E)	6.0	5.3
GCOS - 5	3.8 (E)	6.0	4.8
GCOS - 6	9 (E)	6.2	4.5

for sites in the GCOS area. Table 2 gives the sulfur content of the
season's rains in the GCOS area, converted to S deposited in Kg/ha/
month. The pH and the sulfur content of snow from the two areas, are
shown in Table 3.

Table 2. Sulfur Deposited by Grossfall and by Spruce Throughfall*,
GCOS area, during the 1974 growing season (July – September)

Site	Distance (direction) From SO₂ Source (km)	S Deposited (kg/ha/month) Grossfall	Throughfall
Control – 1	56 (SW)	0.11	0.22
Control – 2	24 (SW)	0.11	0.33
GCOS – 1	3.2 (NW)	0.66	1.3
GCOS – 2	8 (NW)	0.55	3.2
GCOS – 3	8 (S)	1.2	4.1
GCOS – 3A*2	8 (S)	1.2	0.88

*1 Data for stemflow not given owing to lack of a quantitative
factor for conversion.

*2 Aspen

Notable in the rainfall data are the consistently lower pH values
of intercepted rain than open rain, and the seasonal trend (Fig. 1)
towards slightly higher values from an initial minimum.

Data for the constituents of the soil leachates are presented in
Tables 4 and 5 for individual sites in the RMH area. Averaged for sites
distant from the main SO₂ source, the most prominent features of the
data are that for KCl extractable and exchangeable Al which are much
higher in high exposure/impingement sites than in distant ones (Fig. 3
& 4).

Values for the ratio between exchangeable Al : extractable bases
are plotted against soil pH, for samples of mineral soil, in Figure 4;
showing a strong trend of reduced pH for greater Al : base ratio.

DISCUSSION

Notable in the rainfall data (Fig. 1) is the trend towards in-
creasing pH in successive events through the season. Also, although
not as acidic as near some heavily industrial areas, the pH of inter-
cepted precipitation, especially stemflow, is consistently much lower
than open grossfall. The contained sulfur also is significantly greater

TABLE 3. Soluble Sulfur Content and pH of Snow

Direction and distance (km) of site from source	Sampling Date (1974)	S in Snow Kg/ha	pH of Snow
GCOS area			
S, 4.8	February 22	0.17	5.2
S, 9.6	February 22	0.53	4.8
NW, 3.2 (on river ice)	February 22	0.66	5.2
SW, 56	February 22	0.36	5.2
E, 2.4	April 10	0.22	5.0
E, 4	April 10	0.23	5.4
SW, 24	April 10	0.28	5.2
RMH area			
N, 1.0	April 4	0.34	5.0
N, 12.8	April 4	0.22	5.2
NW, 6.4	April 4	0.30	5.0
S, 1.0	April 4	0.83	4.8
S, 4.0	April 4	0.23	5.4
SE, 11.7	April 4	0.23	5.4
SE, 20	April 4	0.15	5.2
E, 1	April 4	0.50	4.9
NE, 9.6	April 4	0.50	4.9
NE, 24	April 4	0.25	5.2
Northern Saskatchewan ("Control")	February 9	0.11	5.0

Included in Fig. 1 are pH values of KCl extracts from various soil layers of the given test site. Comparing soil extract pH values with those of stemflow, grossfall and throughfall, suggest that the acidic nature of these solutions, particularly near the SO_2 source, is having an acidifying effect on the soil, especially in the two surface layers of soil.

The minimum pH value of rain the first-sampled event of the season, perhaps is due to a greater portion of surface-absorbed SO_2 being collected in the first sampling period. Within any sampling date, there seems to be a positive relationship between grossfall and throughfall, i.e. an increase or decrease in the pH of grossfall is usually accompanied by an increase or decrease in throughfall. This relationship is

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Table 4 - Phosphorus and nitrogen, in mg/kg, removed sequentially by various extractants from soils sampled at sites near to and distant from the SO₂ source, RMH area.

		Phosphorus					Nitrogen				
		KCl	NaF	NaOH	H₂SO₄	Total	KCl	NaF	NaOH	H₂SO₄	Total
Idlewilde Ck.	L-F-H	60	150	881	110	1201	969	1584	1565	391	4509
(distant from SO₂ source, 32 Km W.)	0-5 cm	70	22	59	28	179	25	189	253	66	533
	5-15 cm	58	3	14	14	89	47	41	100	50	238
	15-30 cm	1	8	26	66	101	23	59	112	43	237
	Total	189	183	980	218	1570	1064	1873	2030	550	5517
Radiant Ck.	L-F-H	47	164	225	138	574	2020	1186	1899	319	5424
(distant from SO₂ source 38 Km W.)	0-5 cm	3	97	194	62	356	230	551	950	76	1867
	5-15 cm	5	56	84	57	202	221	353	409	64	1047
	15-30 cm	3	42	61	34	160	152	188	294	41	675
	Total	58	359	574	291	1292	2623	2278	3552	500	9013
Average for distant sites	L-F-H	54	157	553	124	888	1495	1385	1732	355	4967
	0-5 cm	37	60	127	45	269	128	370	602	71	1171
	5-15 cm	32	30	49	36	147	134	197	255	57	643
	15-30 cm	2	25	44	50	121	88	124	203	42	457
	Total	125	272	773	255	1425	1845	2076	2792	525	7238
Aquitaine-S	L-F-H	13	158	147	29	347	748	928	1336	136	3148
(near SO₂ source 1 Km S.)	0-5 cm	9	26	31	14	80	187	211	216	57	671
	5-15 cm	5	10	23	10	48	139	84	161	43	427
	15-30 cm	5	7	23	12	47	45	25	146	31	247
	Total	32	201	224	65	522	1119	1248	1859	267	4493
Aquitaine-N	L-F-H	272	181	171	92	716	2423	1842	3483	576	8324
(near SO₂ source 8 Km N.)	0-5 cm	7	93	96	63	250	105	331	397	50	883
	5-15 cm	5	38	73	49	165	90	108	176	43	417
	15-30 cm	20	7	15	39	81	23	54	43	27	147
	Total	303	320	355	243	1212	2641	2335	4099	696	9771
Swan Cl.	L-F-H	53	117	112	42	324	906	816	1115	215	3052
(near SO₂ source 14 Km S.)	0-5 cm	1	46	72	40	159	46	254	320	65	685
	5-15 cm	1	27	61	40	129	135	192	155	32	514
	15-30 cm	9	9	14	17	49	22	44	48	32	146
	Total	64	199	259	139	661	1209	1308	1638	344	4499
Swan Lk.	L-F-H	73	146	90	32	341	1575	1208	1422	299	4504
(near SO₂ source 18 Km SE.)	0-5 cm	2	50	82	33	167	116	314	331	66	827
	5-15 cm	1	94	85	51	231	46	316	209	66	637
	15-30 cm	nil	54	74	46	174	nil	240	227	79	546
	Total	76	344	331	162	913	1737	2078	2189	510	6514
Average for near sites	L-F-H	103	151	130	49	433	1413	1199	1839	307	4758
	0-5 cm	5	54	70	38	167	114	278	316	60	768
	5-15 cm	3	42	61	38	144	103	175	175	46	499
	15-20 cm	9	19	32	29	89	23	91	116	42	272
	Total	120	266	293	154	833	1653	1743	2446	455	6297

Table 5. Acidity, exchangeable cations and pH of 1 n KCl extracts for soils sampled at sites near to and distant from the SO_2 source, RM1 area.

Sites		KCl Ext. pH	Acidity - meg/100 g KCl Ext.		Cations - meg/100 g					Total* bases	Al/bases	Ca/Mg
			to pH 5.5	to pH 8.2	Al	Ca	Mg	Fe	Mn			
Idlewilde Ck.	L-F-H	5.9	0.4	2.8	1.3	66.7	9.2	0.5	0.1	76.5	0.017	7.5
(distant from SO₂ source, 32 Km W.)	0-5 cm	5.8	0.2	0.5	0.2	15.0	2.1	nil	nil	17.1	0.012	7.1
	5-15 cm	5.8	0.1	0.5	0.2	6.5	1.9	nil	nil	7.5	0.027	6.5
	15-30 cm	5.7	0.1	0.5	0.2	10.6	1.8	nil	nil	12.4	0.016	5.9
Radiant Ck.	L-F-H	5.9	0.8	3.8	1.4	43.8	6.9	0.5	0.2	51.4	0.027	6.3
(distant from SO₂ source, 38 Km W.)	0-5 cm	4.7	0.8	1.3	0.9	11.8	2.1	nil	0.1	14.0	0.064	5.6
	5-15 cm	4.7	1.0	1.7	1.1	10.2	2.1	nil	nil	12.3	0.089	4.9
	15-30 cm	5.0	0.3	0.5	0.5	14.9	2.9	nil	nil	17.8	0.028	5.1
Ricinus	L-F-H	6.1	nil	1.3	0.4	29.7	5.1	nil	nil	34.3	0.011	5.8
(distant from SO₂ source, 32 Km SE.)	0-5 cm	4.8	0.3	0.7	0.6	6.8	1.5	0.1	nil	8.4	0.071	4.5
	5-15 cm	5.0	0.2	0.5	0.3	10.3	2.0	nil	nil	12.3	0.024	5.2
	15-30 cm	4.9	0.1	0.5	0.4	9.7	2.5	nil	nil	12.2	0.033	3.9
Jack Fish Lk.	L-F-H	6.1	nil	1.9	1.2	47.8	8.6	nil	0.1	56.5	0.021	5.6
(distant from SO₂ source, 78 Km NXW)	0-5 cm	5.5	nil	0.6	0.3	8.9	1.8	nil	nil	10.7	0.028	4.9
	5-15 cm	4.9	0.1	0.6	1.1	3.6	1.0	0.2	nil	4.8	0.229	3.6
	15-30 cm	4.5	0.4	0.8	0.7	2.8	0.8	0.1	nil	3.7	0.189	3.5
Average for four distant sites	L-F-H	6.0	0.3	2.4	1.1	47.0	7.5	0.3	0.1	54.9	0.019	6.3
	0-5 cm	5.2	0.3	0.8	0.5	10.6	1.9	nil	nil	12.5	0.044	5.5
	5-15 cm	5.1	0.4	0.8	0.7	7.7	1.4	nil	nil	9.1	0.092	5.1
	15-30 cm	5.0	0.2	0.6	0.5	9.4	2.0	nil	nil	11.4	0.067	4.6
Aquitaine-S	L-F-H	3.8	3.6	5.8	2.5	22.4	3.1	2.4	1.3	29.2	0.086	7.2
(near SO₂ source, 1 Km S.)	0-5 cm	3.9	3.6	4.6	3.5	6.3	1.4	0.4	0.2	8.3	0.422	4.5
	5-15 cm	4.0	4.6	5.7	4.4	6.6	2.0	0.1	nil	8.7	0.506	3.3
	15-30 cm	4.1	4.8	5.8	4.6	9.7	3.2	nil	nil	12.9	0.357	3.0
Gulf	L-F-H	4.5	1.8	5.5	1.2	33.9	6.4	0.1	0.5	40.9	0.029	5.3
(near SO₂ source, 1 Km SW.)	0-5 cm	4.4	1.7	2.9	2.8	10.6	3.0	0.2	nil	13.8	0.203	3.5
	5-15 cm	4.7	0.6	0.9	0.8	3.1	1.0	0.1	nil	4.2	0.190	3.1
	15-30 cm	4.5	0.5	0.9	0.6	3.3	1.0	nil	nil	4.3	0.140	3.3
Pr. Ck. Camp	L-F-H	5.8	nil	2.3	0.4	51.2	6.5	nil	0.2	57.9	0.007	7.9
(near SO₂ source, 12 Km NW.)	0-5 cm	4.6	0.6	1.1	0.7	17.3	2.6	nil	nil	19.9	0.035	6.7
	5-15 cm	4.4	1.4	2.3	1.7	12.7	2.6	0.1	nil	15.4	0.110	4.9
	15-30 cm	4.7	0.3	0.8	1.3	10.4	2.2	0.3	nil	12.9	0.101	4.7
Pr. Ck. Ran. Sta.	L-F-H	4.0	4.0	6.9	2.3	15.1	3.6	0.1	0.8	19.6	0.117	4.2
(near SO₂ source, 13 Km N.)	0-5 cm	4.0	5.1	6.2	6.0	1.4	0.4	nil	nil	1.8	3.333	3.5
	5-15 cm	4.9	0.3	0.7	0.3	4.3	1.0	nil	nil	5.3	0.057	4.3
	15-30 cm	4.6	0.6	1.1	0.7	13.6	3.4	nil	nil	17.0	0.041	4.0

Table 5. Continued

| Sites		KCl Ext. pH	Acidity - meg/100 g KCl Ext. to pH ÷ 5.5	to pH ÷ 8.2	Al	Ca	Mg	Fe	Mn	Total* bases	Al bases	Ca/Mg
Aquitaine-N	L-F-H	4.7	2.5	5.9	1.5	34.5	7.3	0.6	3.3	45.7	0.033	4.7
(near SO₂ source, 8 Km N.)	0-5 cm	4.2	1.5	2.4	1.4	7.2	1.0	0.1	0.1	8.4	0.167	7.2
	5-15 cm	4.1	2.0	2.9	2.1	4.5	0.7	0.2	nil	5.4	0.389	6.4
	15-30 cm	4.2	2.3	3.4	2.6	9.7	2.6	0.1	nil	12.4	0.210	3.7
Swan Ck.	L-F-H	4.3	1.8	4.7	1.1	23.7	4.2	0.5	1.9	30.3	0.037	5.6
(near SO₂ source, 14 Km S.)	0-5 cm	4.6	0.4	1.2	0.5	13.0	2.5	nil	0.1	15.6	0.032	5.2
	5-15 cm	4.5	2.0	2.6	1.8	7.7	1.5	nil	nil	9.2	0.196	5.1
	15-30 cm	4.6	1.0	1.5	0.9	6.6	1.2	nil	nil	7.8	0.115	5.5
Swan Lk.	L-F-H	4.4	2.3	5.7	1.4	25.1	5.4	0.8	2.1	33.4	0.042	4.6
(near SO₂ source, 18 Km SE.)	0-5 cm	3.8	8.1	10.1	7.5	5.3	1.5	0.2	0.1	7.1	1.056	3.5
	5-15 cm	4.1	4.7	5.8	4.3	7.0	1.5	nil	nil	8.5	0.506	4.7
	15-30 cm	4.4	0.9	1.6	1.0	6.7	1.5	nil	nil	8.2	0.122	4.5
Average for seven near sites	L-F-H	4.5	2.3	5.5	1.5	29.4	5.2	0.6	1.6	36.8	0.050	5.6
	0-5 cm	4.2	3.0	4.1	3.2	8.5	1.8	0.1	0.1	10.5	0.750	4.9
	5-15 cm	4.4	2.2	3.0	2.2	6.4	1.5	0.1	nil	8.0	0.280	4.5
	15-30 cm	4.4	1.5	2.2	1.7	8.6	2.2	nil	nil	10.8	0.155	4.1

* excluding Al
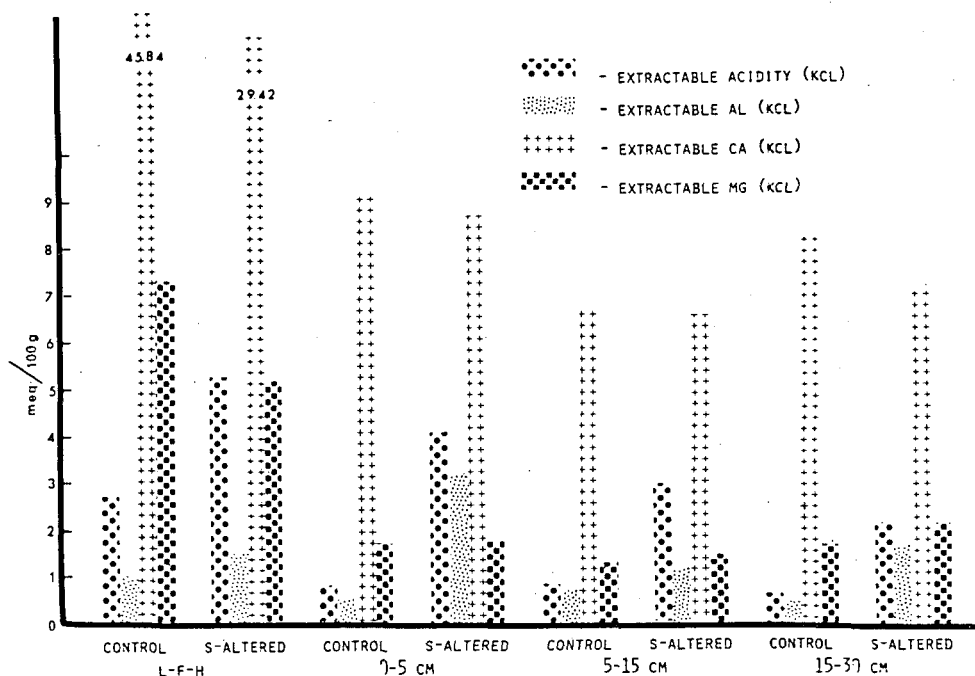
Figure 3. Effect of S-products on certain soil characteristics.
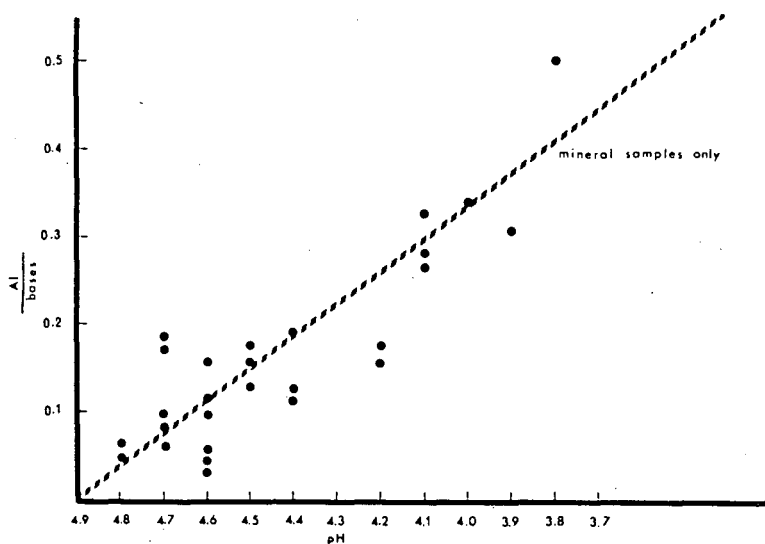
Figure 4. A plot indicating the general re-
lationship between pH and the Al/bases (mainly
Ca + Mg) ratio.

not seen in the pH of stemflow and suggests that the acidic nature of
bark may play as great or greater role in the pH of stemflow as surface
absorbed SO_2. With both throughfall and stemflow, the leaching of or-
ganic acids from vegetation likely contributes to the acidity of these
solutions. Greater acidity of grossfall should create conditions for
greater leaching of organic acids.

There is a general trend for grossfall and throughfall pH values
to increase over the sampling season which suggests that with frequent
summer rains there is little chance for SO_2 accumulation in the atmos-
phere or on the surfaces of foliar material. This feature is not seen
in the pH values of stemflow throughout the sampling season and may
confirm the suggestion that acid groups leached from bark surfaces may
be more responsible for the acidity found in stemflow.

The basis for sequential extraction of P and N lay in the hypo-
thesis that if SO_2 fallout increased the acidity of the soil solution,
there would be greater solubility and mobility of soil constituents,
particularly phosphorus, which is normally considered fairly immobile
(Fried and Broeshart, 1967). In addition, if solubilization of phos-
horus-bearing constituents took place, the released phosphorus should
be found in association initially with aluminum, and, as the soil pH
decreased further, in association with iron. Work presently in pro-
gress in this laboratory indicates that, for forest soils, the
extractants used remove phosphorus fractions which differ somewhat from
aerated cultivated soils, as originally suggested by Jackson (1958).
This sequential leaching approach should be useful in detecting shifts
in distribution of soil phosphorus between different cationic species,
especially Al and Fe.

Current data (Table 4) show that both phosphorus and nitrogen are concentrated in the litter surface (L-F-H horizon) and thus the source of much of these constituents probably is organic. This fact should make any effect of SO_2 on the distribution of soil-P more easily detectable. Generally, NaOH removed more phosphorus and nitrogen than the other extractants, suggesting alkaline hydrolysis and liberation of organically bound phosphorus, particularly from the L-F-H horizon. In mineral soils, phosphorus may be removed by anion exchange, hydroxyl ions liberating phosphates bound to aluminum and iron (Fried and Boeshart, 1967).

Mineral soil samples from sites near the SO_2 source show KCl extracts with much higher exchangeable aluminum values than soils from distant sites. This suggests that the acidifying effect of SO_2 in the soil solution brought about a modification of the cation composition of the soil exchange complex and that Al^{3+} is much more prominent in SO_2 contaminated soils. There is in fact, a consistent relationship between the aluminum content of the KCl extract and acidity, both to pH 5.5 and pH 8.2 (unpublished preliminary results) than between acidity and pH, suggesting that pH alone may not be a reliable parameter to describe changes in soil acidity.

CONCLUSIONS

After two years of studies, data show that SO_2 appears to be having an acidifying effect on grossfall, throughfall, stemflow and soil solution at sites near major sources. Potassium chloride extractable acidity and aluminum are greater in soils suspected of SO_2 contamination, while exchangeable bases (especially calcium and magnesium) are lower. The acidifying effect of SO_2 at present, is not as obvious on soil anionic constituents as that on soil cationic constituents.

Nitrogen and phosphorus cycling are vitally linked with forest growth. Increased soil acidification could ultimately bring about the removal of these from the root zone or create conditions whereby these elements would be chemically or physically fixed so as to be unavailable for stand use.

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