THE INFLUENCE OF SMELTER FUMES UPON THE CHEMICAL COMPOSITION OF LAKE WATERS NEAR SUDBURY, ONTARIO, AND UPON THE SURROUNDING VEGETATION[1]

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Abstract

Analyses for sulphate, calcium, and pH have been made on surface waters from 102 lakes and ponds in the Sudbury metal-smelting district, and data are presented for 35 of these. Sulphur pollution is frequently high within about 5 miles of the three smelters, many ponds exhibiting more than three times the sulphate concentration normal for this area, and three waters more than 10 times this level. Outside about 15 miles distance the influence of smelter pollution upon sulphate concentrations in surface waters is negligible. As expected, many of the most polluted waters are strongly acid, with pH values going as low as 3.3. Sulphuric acid from air pollution has also led to increased weathering of calcium from soils and rocks, so that this ion tends to rise in concentration not only in waters above pH 6 (as expected) but also in those below pH 5.

Damage to terrestrial vegetation is frequently marked within about 5 miles of the smelters, while it is seldom obvious to the untrained eye beyond this distance. Severe damage occurs chiefly within about 2 miles of the smelters.

Introduction

In the vicinity of Sudbury, nickel and copper smelting at Copper Cliff, Coniston, and Falconbridge results in emission to the atmosphere of roughly one million tons of sulphur per annum, chiefly as sulphur dioxide (1). Damage to vegetation is severe close by the smelters, and the occasional fumigations which occur under special weather conditions may affect sensitive plants such as white pine at least as far as 25 miles away (8).

Although emission of sulphur dioxide is monitored by a series of automatic recorders in nine widely separated locations (2), the fall-out pattern is not known in any detail. The present investigation was undertaken to provide information upon this point, to ascertain something of the influence of smelter pollution on lake waters in the area, and to examine the effects upon vegetation near the waters surveyed. A general helicopter survey of damage to terrestrial vegetation was also attempted.

Methods

During June 1959, 102 surface water samples were collected by helicopter and automobile from a wide variety of sites in the Sudbury area, in most cases from small and relatively undisturbed bodies of water with restricted local drainage. The samples were taken in pint polyethylene bottles, which were transported to Toronto for analysis within the next month.

Since sulphur dioxide from the smelters is eventually oxidized to sulphate, this anion was chosen as an index of fall-out from such sources, and was measured by the technique of Mackereth (9). (Nitrate is included with sulphate by this method, but is likely to be present in these waters only in extremely

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TABLE I

Representative analyses of waters from the Sudbury area

	Sample No.	Miles from nearest smelter*	Direction from smelter	SO_4, meq per l.	pH		Ca, meq per l.
					Before aeration	After aeration	
(*a*) Lake Wanapitei	42	ca. 12 F	NNE.	0.34	7.23	7.47	0.43
(*b*) Waters more than 15 miles from smelters	95	19.4 F	NNE.	0.37	4.40	4.39	0.17
	29	16.4 CC	S.	0.36	4.60	4.62	0.21
	92	16.6 CC	N.	0.30	4.79	4.90	0.13
	101	39 F	ENE.	0.31	5.00	5.05	0.14
	96	15.6 F	NNE.	0.47	5.34	5.59	0.26
	94	19.8 CC	WNW.	0.24	5.76	6.00	0.18
	102	35 F	NE.	0.29	5.73	6.04	0.17
	93	15.4 CC	NNW.	0.31	6.45	6.61	0.25
	97	15.5 F	NW.	0.31	6.20	6.70	0.22
	26	19.2 CC	SW.	0.24	6.70	7.18	0.27
	28	24.2 CC	SW.	0.39	7.11	7.39	0.41
	27	23.4 CC	SW.	0.29	7.28	7.41	0.34
Arithmetic average		21.6		0.32	5.78	5.99	0.23
(*c*) Dark-brown waters with heavy ferric hydroxide deposit (see also Nos. 41 and 87)	76	10.8 C	SE.	0.34	6.12	6.62	0.22
	80	3.3 F	ENE.	0.28	6.26	7.25	0.45
	85	2.2 F	WNW.	0.28	6.10	6.94	0.37
(*d*) Pond on silt-clay plain	87	6.4 F	WNW.	1.78	6.15	6.72	1.51
(*e*) Three most alkaline waters from kettle-holes in sandy outwash	17	4.0 F	NNE.	0.60	7.67	7.90	0.98
	18	4.1 F	NNE.	0.60	7.43	7.93	1.42
	40	5.2 F	NNE.	0.66	7.34	7.90	1.12
(*f*) Pond receiving much eroded soil	65	1.8 C	SSW.	1.88	5.76	6.10	1.04
(*g*) Examples of severe pollution by atmospheric sulphur compounds	70	1.5 C	NNE.	6.92	3.40	3.40	0.84
	46	1.8 CC	NE.	6.18	3.46	3.45	1.20
	57	1.3 CC	S.	4.35	3.61	3.61	0.58
	7	0.7 F	S.	3.53	3.49	3.50	0.68
	73	4.5 CC	ENE.	2.84	3.51	3.51	0.69
	8	0.7 F	S.	2.75	3.23	3.26	0.70
(*h*) Examples of other human interference							
Sewage	58	1.7 CC	S.	7.07	4.55	4.56	3.55
Railway fill, possible mine waste too	52	2.0 CC	NNW.	10.00	3.56	3.56	4.36
Pond near Garson School, mine waste	5	3.2 F	WSW.	5.00	7.30	7.91	4.78
Some drainage from smelter sludge dump	59	3.4 CC	WSW.	3.42	4.16	4.16	1.19
Clay pit	67	2.2 C	E.	1.12	5.82	6.46	0.65
Silty sand pit	38	4.8 F	WSW.	0.69	7.60	7.98	1.33
Gravel pit	91	11.2 F	NW.	0.44	6.41	6.64	0.83
Pond banked with sawdust	41	7.4 F	NNE.	0.66	5.86	7.00	0.53

*CC—Smelter at Copper Cliff, C—Smelter at Coniston, F—Smelter at Falconbridge.

low concentrations as compared with sulphate.) Because oxidation of sulphur dioxide commonly produces sulphuric acid, the pH values of the water samples were estimated by glass electrode, both before and after aeration to drive off excess carbon dioxide. As the lime status of the soil is also likely to affect the pH of water draining it, calcium concentrations were determined by EDTA titration, with ammonium purpurate as indicator (7). Potassium cyanide was employed in the routine to complex heavy metals which interfered with calcium analyses in waters collected near the smelters. Qualitative visual estimations of water color, and of ferric hydroxide deposition on standing, were also made.

Damage to terrestrial vegetation in the vicinity of the lakes was noted subjectively as very severe, severe, considerable, moderate, or not obvious, on brief examination from some distance away. The categories are defined in Table II. Estimates of the boundaries between the various categories were later made from the air on a series of helicopter transects, and these were used to check the general validity of the lakeside observations. It should be remarked that the estimates of damage refer only to vegetation on thin soils in relatively exposed situations; on the deeper soils in protected valleys damage is not nearly so great.

Results of the Water Analyses

Data for 35 representative water samples are presented in Table I. In addition all the sulphate analyses are shown in Fig. 1. Some waters which have been subjected to human interference apart from smelter pollution are indicated separately as crosses on this figure.

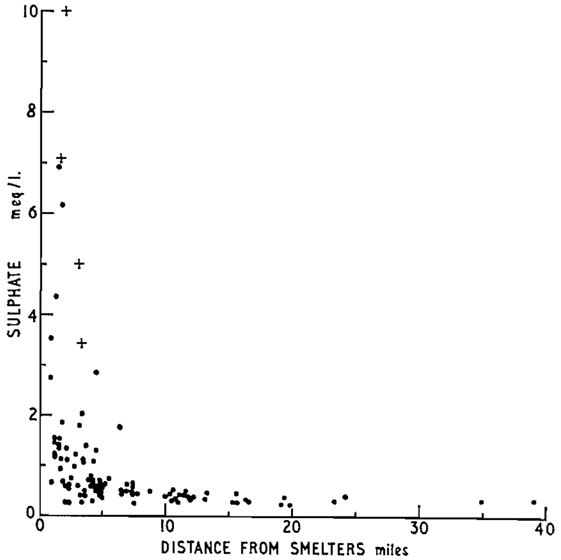

FIG. 1. Sulphate concentrations in lakes and ponds near Sudbury. Crosses represent ponds subject to other than aerial sulphur pollution.

Sulphate

Figure 1 shows very clearly the influence of air pollution in raising concentrations of sulphate in pond and lake waters near the smelter stacks. The maximum level reached in any water not otherwise subject to human interference was 6.92 meq/l., recorded for a small shallow pool on a rocky knoll 1.5 miles NNE. of the Coniston smelter (Table I (*g*), No. 70). The water had probably been concentrated a good deal by evaporation. At this site there is no living higher vegetation whatever, although a few red maple and red oak bushes are present with a small number of associated species in less exposed situations about 100 yards away. A second sample, 1.8 miles NE. of Copper Cliff (Table I (*g*), No. 46), had almost as much sulphate, 6.18 meq/l. The vegetation here is again sparse, with red maple bushes predominating. The next highest of such values was 4.35 meq/l., from another similar pond (Table I (*g*), No. 57) about 1.3 miles S. of the Copper Cliff smelter, with only a few red maple bushes growing around the site.

Of the three other water samples highest in sulphate, all of which contained 5 meq/l. or more, one (Table I (*h*), No. 5) was taken from a pond which drains large amounts of mine waste rich in sulphur. A second (Table I (*h*), No. 52) has had abundant railway fill dumped in it, quite possibly from a mine nearby. The third such sample came from Kelley Lake, about 1.7 miles S. of Copper Cliff (Table I (*h*), No. 58). This lake receives very large amounts of sewage from the Sudbury area.

All but one of the 32 waters with sulphate levels above 1 meq/l. were collected within 5 miles of a smelter stack, the exception being a small pond (Table I (*d*)) about 6.4 miles WNW. of Falconbridge, with water containing 1.78 meq/l. This pond was the only one observed on the extensive silt–clay plain north of Sudbury; presumably these heavy and low-lying agricultural soils are comparatively rich in sulphur compounds. One other sample (Table I (*g*), No. 73), not quite so far from the smelters as the preceding one, also exhibited unusually high sulphate (2.84 meq/l.), without any apparent possibility of other than aerial supply. The site is, however, very centrally located with regard to all three smelters, being 4.5 miles ENE. of Copper Cliff, and no doubt owes its high sulphate levels to air pollution from all of them.

Neglecting the waters receiving other than air-borne pollution, the 49 samples collected within 5 miles of the smelter stacks yielded a logarithmic average sulphate concentration of 0.94 meq/l., logarithms being employed to normalize the frequency distribution. This value may be compared with the concentration of 0.34 meq/l. recorded for Wanapitei Lake (Table I (*a*)), which is a large body of water draining very extensive areas NE. of Sudbury, and with the average of 0.32 meq/l. for the 12 waters collected more than 15 miles away from the smelters (Table I (*b*)).

It should also be remarked that only four of the 49 waters taken within 5 miles of the smelters gave sulphate concentrations of less than 0.40 meq/l.

Three of these samples (for two of them see Table I (*c*), Nos. 80 and 85) were strongly colored by brown organic compounds, and also deposited considerable reddish-brown ferric hydroxide on standing; microbiological reduction may be presumed to account for the low levels of sulphate in these waters.

In general it may be said that Fig. 1 shows a pronounced fall-out of sulphur within 5 miles of the smelters, and only slight fall-out farther away, the smelter effects becoming negligible beyond about 15 miles. It must of course be remembered that occasional severe fumigations occur beyond this distance, and may have considerable biological significance even though they are insufficient to affect lake water concentrations of sulphate.

Acidity

The pH range in unaerated water was 3.23–7.67; after aeration to remove excess carbon dioxide the range became 3.26–8.03. Wanapitei Lake (Table I (*a*)) exhibited values of 7.23 (original) and 7.47 (aerated). Below pH 4.5 the increase following aeration was probably within the range of error, while between 4.5 and 5.0 it was seldom more than 0.1 units. Above this limit the influence of dissolved carbon dioxide increased markedly, and throughout the original pH range of 5.4–7.7, aeration resulted in a rise of about 0.15–0.85 units. Two exceptional samples (Table I (*c*), No. 80, and (*h*), No. 41) showed even greater increases of pH upon aeration, amounting to 0.99 and 1.14 units respectively. In both these pond waters there was a strong brown humus coloration, with in addition a heavy deposit of ferric hydroxide on standing; indeed, many of the waters exhibiting the most marked pH increases upon aeration showed such iron deposits. Presumably most of the iron derived from ferrous bicarbonate, which upon oxidation would release considerable carbon dioxide into the water, and thus lower pH below the levels characteristic of the other waters.

While carbonic acid was present in most of these waters, the strongly acid samples undoubtedly owed their low pH values to free sulphuric acid, with all nine samples of pH less than 3.7 showing sulphate concentrations above 1.3 meq/l. Apart from some of these obviously polluted waters, the highest equivalent ratio of hydrogen ions to sulphate ions (0.36) came from a small pool set in an extensive area of drained bog peat 4.9 miles N. of Copper Cliff. This pool contained water with a sulphate level of 0.55 meq/l. and a pH of 3.70, the ionic ratio of hydrogen to sulphate being not unusual for bog waters.

The general trend in these Sudbury waters was for sulphate to be relatively low, about 0.25–0.75 meq/l., above pH 5; with a gradual rise to about 0.5–1.5 meq/l. as pH fell to 4, and a very sharp rise below pH 4 (see Table I (*g*) for the most acid samples). However, in five circumneutral waters between pH 6 and 7, sulphate concentrations of 1–2 meq/l. were observed. With the exception of the sample from the silt–clay plain these waters were collected from ponds fairly near the smelters and in sites exhibiting considerable soil erosion, the most extreme example being a water collected 1.8 miles SSW.

of Coniston (Table I (*f*)). Such erosion, brought about mainly by pollution damage to the vegetation, doubtless accounts for the low acidity of these waters, since it enriches them in lime. For instance, the water mentioned above (Table I (*f*)) had a calcium concentration of 1.04 meq/l.

One single pond, near Garson School and about 3.2 miles WSW. of Falconbridge (Table I (*h*), No. 5), combined high sulphate with an alkaline reaction; this pond drains spoil heaps near the Garson mine. In contrast, another pond which contains a great deal of railroad fill combined high sulphate with strong acidity (see Table I (*h*), No. 52). Relative to its high sulphate concentration, Kelley Lake (Table I (*h*), No. 58) was not very acid, perhaps owing to the inflow of large amounts of sewage.

It is noteworthy that even far from the smelters some pond waters were moderately acid, probably owing to the presence of sulphuric acid from natural sources (3, 4, 5). For example three waters at 10, 12, and 19 miles distance (for one of these see Table I (*b*), No. 95) exhibited pH values of 4.3–4.4, with about 0.4 meq of sulphate per liter. All these waters were low in calcium, which only amounted to 0.15–0.17 meq/l.

Calcium

The waters richest in calcium were the pond near Garson School (Table I (*h*), No. 5) with 4.78 meq/l., a pond 2 miles NNW. of Copper Cliff (Table I (*h*), No. 52) with 4.36 meq/l., and Kelley Lake (Table I (*h*), No. 58) with 3.55 meq/l. The first drains mine waste, and the second contains abundant railway fill, possibly from a mine nearby. Kelley Lake, on the other hand, is a major receiver of Sudbury sewage. Of the less disturbed ponds, that on the silt–clay plain (Table I (*d*)) had water containing 1.51 meq of calcium per liter, while several rather alkaline waters from among the ponds located in sandy outwash north and east of Falconbridge were also fairly rich in calcium, with up to 1.42 meq/l. (for the three most alkaline samples see Table I (*e*)). Of the ponds with waters between pH 6 and 7, those into which there has been considerable erosion were relatively high in calcium, and reached a maximum of 1.04 meq/l. (see Table I (*f*)). Between pH 5 and 6 calcium levels were less than 0.4 meq/l., while below pH 5 they tended to rise again, probably because the sulphuric acid produced from smelter fumes weathers additional lime from the soil. Among the acid waters affected only by the smelter fumes and not by fill or waste of any kind, the maximum calcium concentration observed was 1.2 meq/l. (Table I (*g*), No. 46). It was noticeable that among such acid waters, those from the more peaty drainage areas tended to exhibit higher calcium levels at a given pH, possibly because a lesser concentration of acid is required to bring about replacement of calcium adsorbed by peat than to weather it from inorganic soil minerals. The lowest calcium value found was 0.06 meq/l. in a sample of pH 3.9; apart from this several samples between pH 3.7 and 5.4 yielded concentrations of about 0.15 meq/l. Water from Wanapitei Lake (Table I (*a*)), collecting drainage over a wide area to the north and east of Sudbury, contained 0.43 meq/l.

TABLE II

Approximate categories of pollution damage used in the survey of terrestrial vegetation

	Damage				
	Not obvious	Moderate	Considerable	Severe	Very severe
Vegetation cover*	Complete	Up to $\frac{1}{3}$ knolls exposed	$\frac{1}{3}$ to $\frac{2}{3}$ of rock and shallow soil exposed	Over $\frac{2}{3}$ of surface exposed	Whole surface exposed
Crown damage†	Not evident except for chlorosis on white pine	On $\frac{1}{3}$ trees, esp. birch; some spp. chlorotic	Slight to moderate on all trees, or considerable on more than $\frac{1}{3}$ trees	Extensive on most and apparent on nearly all trees. Suckering from base common	Extensive on few remaining trees. Suckering from base prevalent.
Species present					
Red maple	+	+	+	+	+
Red oak	+	+	+	+	+
White birch	+	+	+	+	
Red pine	+	+	+		
Jack pine	+	+	+		
White spruce	+	+			
Trembling aspen	+	+			
Large-toothed aspen	+	+			
White pine	+				
Soil erosion	None	Not well marked	Evident	Strongly evident	Organic horizon almost wholly gone

*May be more or less evenly distributed, or in mosaic patterns of variable damage.
†Leads from chlorosis to browning of foliage, death of whole twigs, and in severe cases to extensive stem dieback.

Survey of Pollution Damage to Terrestrial Vegetation

The notes on vegetation damage near the ponds were usually made from the helicopter while taking water samples at a short distance from shore. Although qualitative (see Table II for definition of categories, and Fig. 2 for illustrations of them), their value is increased by the fact that a great number of sites was examined in all directions from the sources of pollution, and it may be added that local impressions were largely verified later by aerial survey during a special series of helicopter flights.

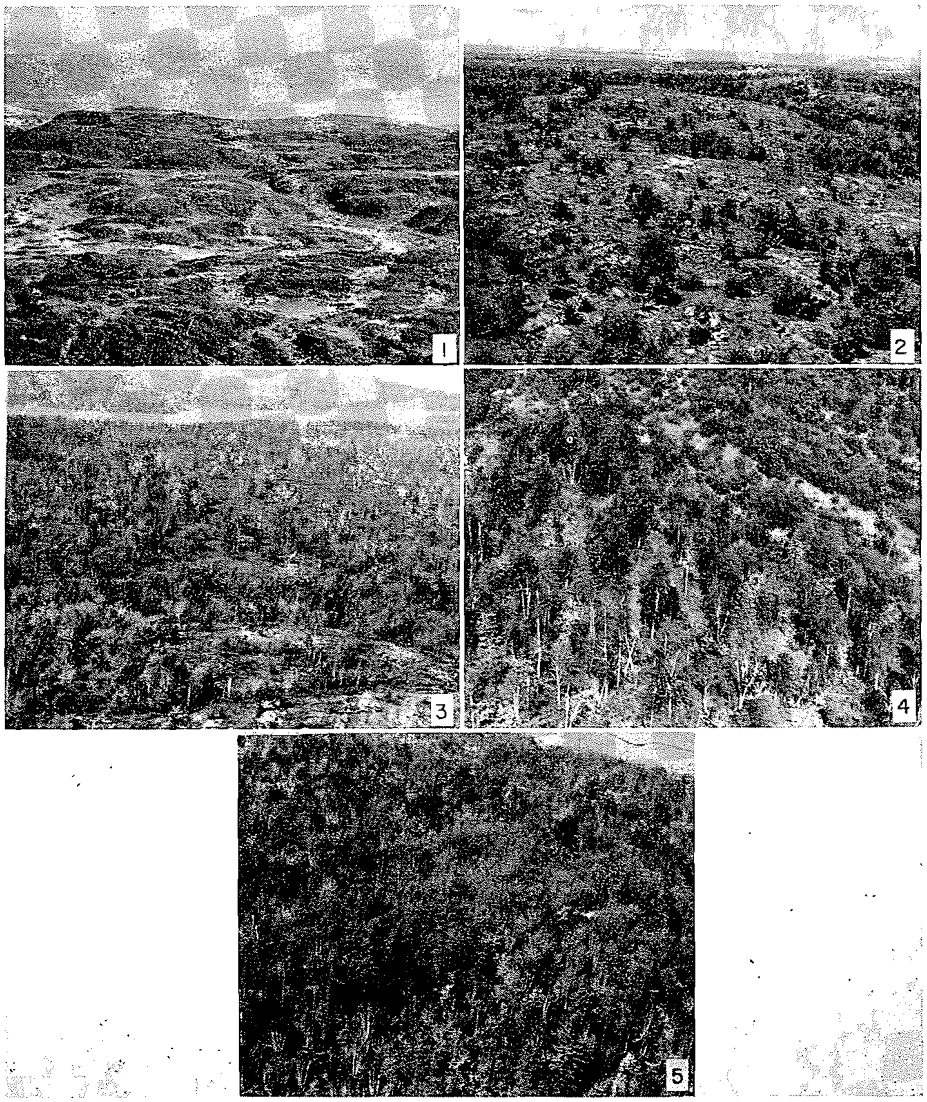

FIG. 2. Examples of the different categories of vegetation damage used in this survey: (1) very severe, (2) severe, (3) considerable, (4) moderate, (5) not obvious.

Table III summarizes the local notes, which refer only to thin soils in sites well exposed to pollution. It indicates that extensive damage is generally restricted to the area within 5 miles of the smelters, that is, the same area within which extremely high levels of sulphur fall-out were observed. Moreover, the very severely damaged sites were all located within 2 miles of the smelters. Beyond 5 miles damage to terrestrial vegetation was observed only twice, although some species such as white pine were absent beyond this distance owing to their especial sensitivity (cf. refs. 6, 8).

Regarding the later aerial observations, these suggested that the boundary between very severe and severe damage lies at an average of 2 miles from the smelters, the boundary between severe and considerable damage at about 4 miles, and between considerable and moderate damage at about 4.5 miles. The outer limit for moderate damage seemed to lie at about 6 miles from the smelters. Thus it appears that the aerial survey agrees reasonably well with the local lakeside notes, when one considers that a good deal of overlap occurs in both sets of records.

TABLE III

Estimates of vegetation damage in relation to distance from the sources of aerial smelter pollution

	Miles from smelter stacks																												
Damage	0.5		1.0		1.5		2		3		4		5		6		7		8		10		12		15		20		40
Very severe		1		3		3																							
Severe		2		2		5		5		3		2																	
Considerable				1		2		5		7		10				1													
Moderate										3		5						1											
Not obvious										1		1		2		4		5		2		10		3		8		4	

While close and expert examination of the flora might well have revealed injury from sulphur dioxide fumes at distances much greater than 5 miles from the smelters (cf. refs. 10, 11), the present records provide the kind of estimate that might be made by untrained observers, and so may be useful in judging the influence of smelter pollution upon the amenity value of the vegetation. A more detailed study of vegetation NNE. of Falconbridge (6) showed a generally comparable picture of damage, the numbers of macrophyte species declining sharply within about 4–7 miles of the smelter, and the proportion of open ground increasing in inverse ratio.

It may be remarked that no assessment is given for the effect of wind direction upon the distance to which pollution effects extend. Although one might expect the effects to be most marked in the NE. direction of the prevailing wind, variations in terrain, fire damage, and human interference render any such assessment extremely difficult, and it has not been attempted.

The effects of sulphur pollution upon vegetation may be examined in yet another way, by correlating the qualitative estimates of local damage with the sulphate concentrations in the nearby lake waters. This evident correlation is demonstrated in Table IV. It may also be of interest to give the logarithmic averages of sulphate concentration for lakes in areas suffering different degrees of pollution damage. Where damage is very severe, sulphate averages nearly 3 meq/l., as against 1.2 meq in severe cases and 0.7 meq where damage is recorded as considerable. In moderately damaged sites, sulphate levels average 0.5 meq/l., while in sites showing no obvious damage the average concentrations are only about 0.4 meq/l.

TABLE IV

Lake water sulphate in relation to damage of surrounding vegetation by smelter pollution

Sulphate, meq/l.	Vegetation damage				
	Very severe	Severe	Considerable	Moderate	Not obvious
0.21–0.40		1	4	1	19
0.41–0.60		1	4	6	16
0.61–0.80		2	9	1	4
0.81–1.00		1	1		
1.01–1.25		5	3		
1.26–1.50	2	3	2		
1.51–2.00	1	2	1		
2.01–4.00	1	2	1		
>4.00	3				

Concluding Remarks

The present study indicates the extent of damage in the vicinity of the Sudbury group of smelters. However, its potential application in connection with the location of future smelters or similar industrial installations elsewhere may also be of interest. For example, should a new smelter be contemplated then its location should be sufficiently far from residential and other community areas so that their amenities may be preserved from pollution and damage. For smelters of capacity within the range of those at Sudbury, a distance of more than 5 miles should be recommended. In addition a preliminary survey might reveal situations in which pollution damage could be restricted as far as possible to the less productive land types, leaving those areas with a greater proportion of productive land types for forest or agricultural use.

Acknowledgments

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