

ECOLOGICAL CONSEQUENCE OF ACIDIC AND HEAVY-METAL DISCHARGES FROM THE SUDBURY SMELTERS

L. M. WHITBY, P. M. STOKES, T. C. HUTCHINSON AND G. MYSLIK
*Institute for Environmental Studies and Department of Botany, University of Toronto,
Toronto, Ontario.*

ABSTRACT

Studies have been made of the effects on terrestrial and aquatic ecosystems, of heavy-metal emissions, sulfur dioxide, and acidic rain-induced problems, caused by smelting activities in the Sudbury region. The effects have been profound. Phytoplankton populations have been reduced from 2,000 cells/ml to 'negligible' in many of the lakes close to the Coniston smelter, and around both Falconbridge and Copper Cliff. Levels of nickel (up to 6.5 ppm) and copper (up to 0.2 ppm) occur in these lakes, and these, combined with increased acidity, are major obstacles to survival of primary producers. The whole food chain has suffered parallel and often even greater extinctions in these lakes and especially in those to the northwest.

For terrestrial systems, emissions of sulfur dioxide have directly damaged very extensive areas of forest, with soil acidification and subsequent major soil erosion. In addition to this, the smelted heavy metals have accumulated to very high concentrations over a wide area, especially in surface soils. These levels were, even in the absence of sulfur dioxide, toxic to a wide range of species tested in extracts or in single-metal solutions. Lime amendments to soil had dramatic effects on growth and survival of plants. Metal uptake was found to be correspondingly reduced compared with uptake on unamended soils.

RÉSUMÉ

Des études ont été effectuées sur les effets des émissions de métal lourd, de dioxyde sulfurique et des problèmes d'acides provoqués par la pluie et causés par des activités de fusion dans la région de Sudbury, sur des écosystèmes terrestres et aquatiques. Les effets ont été importants. Les populations de phytoplancton ont été réduites de 2,000 cellules/ml à des proportions négligeables dans plusieurs lacs près de la fonderie Coniston, et autour de Falconbridge et de Copper Cliff. Des niveaux de nickel (jusqu'à 6.5 ppm) et de cuivre (jusqu'à 0.2 ppm) apparaissent dans ces lacs et ceux-ci combinés avec une acidité accrue, sont des obstacles majeurs à la survie des producteurs premiers. Toute la chaîne de nourriture a connu des extinctions parallèles et même souvent plus grandes dans ces lacs et particulièrement dans ceux du nord-ouest.

Quant aux systèmes terrestres, les émissions de dioxyde sulfurique ont endommagé directement de

très vastes régions forestières, par l'acidification du sol et les érosions terrestres majeures subséquentes. De plus, les métaux lourds fondus se sont accumulés jusqu'à de très fortes concentrations sur une grande étendue, et surtout dans les sols superficiels. Ces niveaux étaient toxiques, même en l'absence de dioxyde sulfurique, à une grande variété d'espèces étudiées dans des extraits ou dans des solutions à métal unique. Les améliorations du sol avec la chaux ont eu des effets dramatiques sur la croissance et la survie des plantes. La proportion de métal s'est trouvée réduite, par la même occasion, à comparer à celle des sols non-améliorés.

(Traduit par le journal)

INTRODUCTION

Aquatic and terrestrial studies of possible heavy-metal effects were commenced around Sudbury in 1968 to determine whether particulate emissions had caused changes in the chemical and physical properties of the aquatic and terrestrial ecosystems in the region, and to determine possible long-term consequences of such emissions compared with sulfur dioxide.

The very high accumulation of Ni and Cu especially in soils and their patterns of distribution relative to soil depth and distance from the smelter have already been reported (e.g. Costescu & Hutchinson 1972; Hutchinson & Whitby 1974; Whitby & Hutchinson 1974). The toxicity of soils over an area of many square miles has been emphasized and the long-term problems described. Studies have focused on transects from the Coniston smelter, which was closed in 1972 but previously had run continuously since 1913.

The water bodies in the Sudbury area are mainly small Shield lakes and mine streams. In their natural state these lakes are typically oligotrophic, soft-water, shallow and generally with a low conductivity, a low organic content and with a poor buffering capacity. The alteration in the chemical status of the lakes close to Sudbury due to mining and smelting activities can be estimated by selecting a range of lakes at different distances from a point source such as a single smelter and determining correlations

between distance and chemical parameters. Similarly, the biological changes which might be occurring can be determined by field studies (e.g. primary productivity, standing crop, algal cell numbers, species diversity) and by laboratory bioassay of lake waters with standard test organisms.

Such data have been acquired over the past six years. The lakes selected range from 1.6 km to 12.0 km from the Coniston smelter.

MATERIALS AND METHODS

Choice of terrestrial sampling areas

The Coniston smelter was selected as the focus for the terrestrial studies (Fig. 1). Coniston is off the main ore bodies, and away from dykes; no tailings are nearby, and only a small area around it is covered by slag heaps.

Sites were chosen at distances from 0.8 to 50 km from the Coniston smelter, selected to traverse Dreisinger's 1970 isopleths (Dreisinger 1970). Soil samples from 10cm-depth profiles

were collected in June 1969 and 1970 at sites at least 100 m from roadways. All sites were on hill tops, where the plumes from smelter stacks impinge most often.

In 1969, natural vegetation was collected twice (June and August), at nine of the sites. In many instances, a species present at one site was absent at several others, especially at sites close to the smelter where plant cover and species composition were poor.

Analytical methods

Methods for the analysis and determination of pH, conductivity, loss on ignition, total heavy-metal concentrations, water-soluble metals, water-soluble sulfate *etc.* have been described previously (Hutchinson & Costescu 1972; Hutchinson & Whitby 1974, 1975; Whitby 1974).

Samples of natural vegetation were washed to remove all surface contamination, rinsed in 5 changes of deionized water, then dried for 48 hours at 60°C. 0.2g plant material was predigested for 16 hours in 3 ml concentrated nitric-

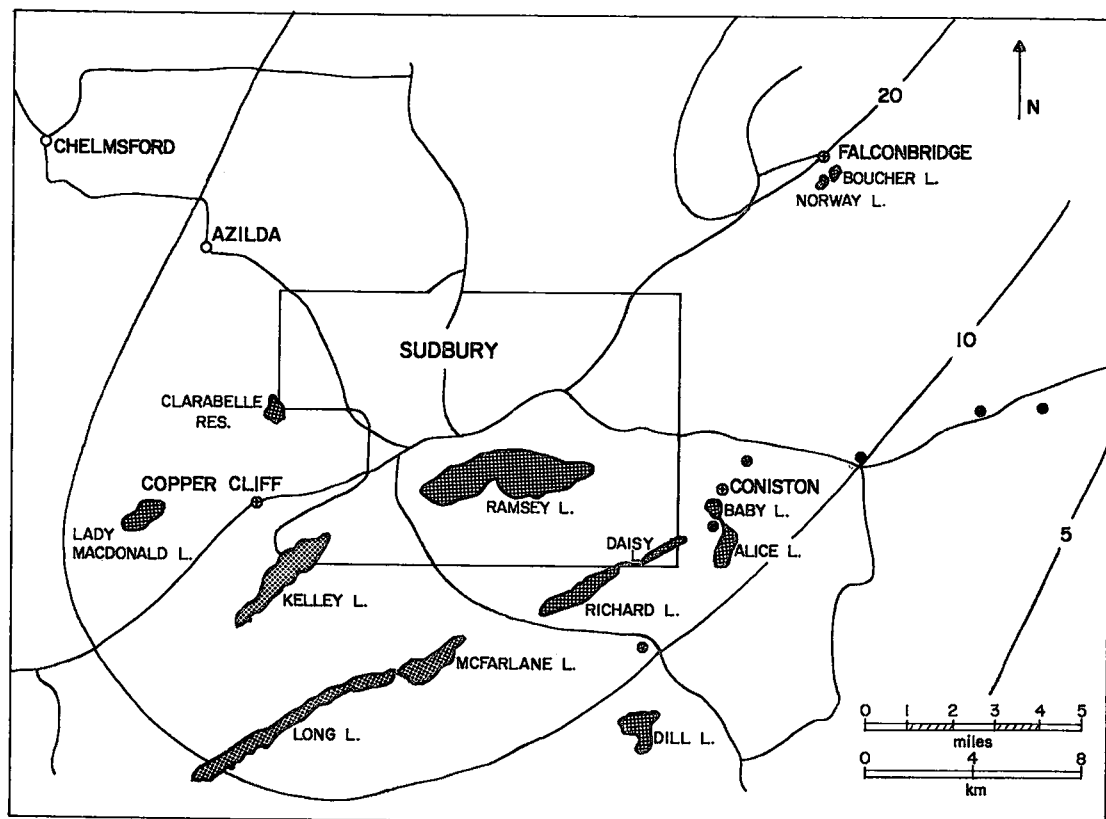


FIG. 1. Sketch map of the Sudbury region, with sites marked for soil and vegetation samples and lakes in which bioassays were carried out. The lines marked 20, 10 and 5 join points indicating number of damaging fumigations noted in 1971 (after Dreisinger 1970).

perchloric acid (ratio 1:1), then heated to 75°C in a sandbath for 5 hours. Each sample was diluted to 10 ml with deionized water without filtering.

Rainfall-dustfall samples were collected throughout the sampling region during 1970 and 1971 (Hutchinson & Whitby 1975).

All samples were analyzed for Ni, Cu, Co, Al, Zn, Mn, and Fe by atomic-absorption spectrophotometry.

Bioassays for seedling growth using root elongation as an index

The radicle elongation of germinated seedlings was used as a method for assaying the toxicity of Sudbury soils, with root growth acting as an index of soil response. A soil-water extract was obtained from each of the sites at three depths, by shaking 33.3 g soil in 100 ml deionized water for 4 hours.

A series of bioassays was also performed to determine the effects of single-metal salts in nutrient solution (Whitby & Hutchinson 1974; Whitby 1974).

Field and greenhouse experiments

Field experiments were conducted during the summers of 1969-1971. Experiments were conducted at nine sites — 1.5, 1.7, 1.9, 3.8, 7.4, 10.4, 13.5, 19.3 and 49.8 km from the Coniston smelter. Four 1m-square plots were set out at each site. One plot was left untreated, one plot fertilized with 100 g So-Green 7-7-7 fertilizer, one plot was limed to pH 7, and the fourth plot both limed to pH 7 and fertilized with 100g So-Green 7-7-7 fertilizer.

Seeds of a variety of naturally-occurring and horticultural species were planted in the plots in June and harvested after 75 days. All plants were carefully removed from the soil to preserve root systems. The plants were washed to remove soil particles, rinsed five times in deionized water and dried at 60°C for 48 hours. Dry weights for each plant were obtained after the roots were removed, since complete root systems could not be preserved. The plants were digested and analyzed for heavy-metal contents as described previously.

Tests were run on soils collected from the field under controlled greenhouse conditions during 1969-71. The soil samples from each site were divided into 1 kg sections — one was left untreated, a second was treated with 14 g So-Green 7-7-7 fertilizer, a third limed to pH 7, and a fourth limed to pH 7 and fertilized with 14 g So-Green 7-7-7 fertilizer. The soil from each treatment was then placed in 20 × 7.5cm plastic pots. Four test species were used and

germinated before planting. After 28 days, the plants were harvested and treated in a similar manner to the field-experiment plants.

Aquatic studies

Most of the methods used in the algal bioassays of lake waters from the Sudbury region have been described in detail (Hutchinson & Stokes 1975). Lake waters were collected from a range of water bodies arranged along a transect south of the Coniston smelter (Fig. 1). Algal counts were expressed as cells per ml of original lake-water sample. Algal cells were preserved in the field using Lugol's iodine.

Water for laboratory bioassays was filtered immediately after collection, to remove major debris. Four laboratory test organisms were used for the study of the potential ability of lake waters to support algal growth. The effect of pH adjustment and addition of nutrients on these lake-water samples was also tested by increasing the pH of the acidic lake waters to 6.8 and by addition of a one-tenth strength solution of Bolds Basal Medium (BBM), an inorganic algal nutrient solution.

Chemical methods were standard atomic-absorption spectrophotometry for heavy metals and a modified barium-chloride turbidimetric method for sulfate.

RESULTS

Terrestrial studies

The physical and chemical properties of the soils have been extensively reported (Hutchinson & Costescu 1972; Hutchinson & Whitby 1974, 1975) and therefore are briefly summarized.

The pH of the soils is naturally acidic, ranging from 3.8 to 4.8 in areas unaffected by smelting operations. The lowest pH values occurred in surface soils close to the smelter. Surface pH values were below 3.0 to a distance of 7.4 km from the smelter. pH values increased through the soil profile.

The conductivity values were high in soils close to the smelter (552 μ mhos at 1.6 km) and decreased to normal levels as distance from Coniston increased (58 μ mhos at 49.8 km). The highest conductivity was in the surface soils and decreased through the soil profile.

Concentrations of Cu and Ni, the major metals smelted in the region, were also highest in surface soils closest to the smelter. Ni showed the highest concentrations, e.g. 5104 ppm at 1.1 km, 282 ppm at 10.4 km and 35 ppm at 49.8 km. Both Ni and Cu occurred at concentrations greater than 1,000 ppm in all

soils within 7.4 km of the Coniston smelter. The patterns for the other elements smelted at Coniston, such as Co, Fe and Ag, were similar, with the highest concentrations close to the smelter, and a decreasing concentration with increasing distance from the smelter. Metals not smelted at Coniston showed no patterns related to distance from the smelter or depth in the soil profile.

Two species, the blueberry, *Vaccinium angustifolium* and the red maple, *Acer rubrum*, were collected at eight sites. Four species were collected in at least six sites — trembling aspen *Populus tremuloides*, sweet fern *Comptonia peregrina*, paper birch *Betula papyrifera*, and wavy hair grass *Deschampsia flexuosa*.

Concentrations of Ni, Cu, Co, Al, Fe, and

TABLE 1. METAL CONTENT IN FOLIAGE OF NATURALLY-OCCURRING PLANT SPECIES ALONG A TRANSECT FROM THE CONISTON SMELTER.

Species	Distance (km)	Mean ppm of two homogenized samples					
		Ni	Cu	Al	Fe	Mn	Co
<i>Vaccinium angustifolium</i> (Low Sweet Blueberry)	1.6	92	75	100	618	223	10
	2.1	55	30	60	383	455	8
	7.4	45	35	240	245	133	7
	13.5	37	22	120	223	1120	5
	19.3	36	19	70	135	1260	5
	28.9	16	19	100	123	1130	5
<i>Acer rubrum</i> (Red Maple)	49.8	14	14	35	88	465	5
	1.6	98	37	125	698	197	8
	2.1	88	26	170	455	135	8
	7.4	45	31	125	173	180	8
	13.5	33	20	20	170	690	5
	19.3	57	28	80	208	220	8
	28.9	14	16	22	275	1055	6

Samples were collected in June 1969.

TABLE 2. CONCENTRATIONS, IN SUDBURY SOILS, OF HEAVY METALS EXTRACTED BY DISTILLED WATER

Distance miles	km	Depth (cm)	pH	(ppm-field wet soil condition)				
				Ni	Cu	Co	Al	Zn
0.5	0.8	0	3.08	381.0	126.0	13.8	151.8	2.9
		5	3.36	42.8	22.8	0.6	20.7	0.2
		10	4.48	31.8	18.3	nd	17.4	0.1
0.95	1.5	0	3.40	321.9	178.5	10.2	231.3	2.7
		5	3.59	168.0	59.7	4.5	155.4	1.6
		10	3.69	57.6	29.7	1.2	69.0	0.4
1.2	1.9	0	3.25	426.0	178.5	13.2	296.7	4.1
		5	3.48	108.9	29.7	3.0	93.3	1.0
		10	3.86	50.1	18.0	0.6	41.4	0.2
2.4	3.8	0	3.59	43.2	24.3	0.6	31.2	0.6
		5	3.57	36.9	19.2	0.3	31.2	0.5
		10	3.67	41.1	21.6	0.3	31.2	0.5
4.6	7.4	0	3.41	25.8	9.0	nd	19.5	0.3
		5	3.54	12.6	4.5	nd	19.5	0.2
		10	3.65	6.3	2.4	nd	13.2	0.2
6.5	10.4	0	3.86	19.5	0.9	nd	45.9	1.2
		5	4.11	3.9	nd	nd	16.5	0.1
		10	4.22	3.6	nd	nd	16.5	0.1
8.4	13.5	0	3.93	3.0	0.6	nd	6.6	nd
		5	4.32	nd	nd	nd	nd	nd
		10	4.69	nd	nd	nd	nd	nd
12.0	19.3	0	4.39	nd	nd	nd	nd	nd
		5	4.46	nd	nd	nd	nd	nd
		10	4.70	nd	nd	nd	nd	nd
31.0	49.8	0	4.79	0.3	nd	nd	1.5	nd
		5	4.69	nd	nd	nd	1.5	nd
		10	4.44	nd	nd	nd	1.5	nd

nd = below limits of detection by this technique.

Mn in *Vaccinium angustifolium* and *Acer rubrum* are given in Table I. In all species tested, Ni, Cu, Co, and Fe were elevated in foliage collected close to the smelter. For example, *Vaccinium angustifolium* contained 92 ppm Ni, 75 ppm Cu, 10 ppm Co, and 618 ppm Fe at 1.6 km which decreased to 14 ppm Ni, 14 ppm Cu, 5 ppm Co and 88 ppm Fe at 49.8 km. Al, which was not smelted at Coniston, was also elevated in vegetation growing close to the smelter and decreased with increasing distance. Foliage of *V. angustifolium* contained 100 ppm Al at 1.6 km and 35 ppm at 49.8 km.

There was no consistent pattern of metal elevation in foliage when samples collected in August were compared with those from June. For example, *Vaccinium angustifolium* had higher concentrations of Ni in August than in June, whereas *Acer rubrum* had higher concentrations of Ni in June than in August.

Soil-water extracts

The water-soluble concentrations of heavy metals from three soil depths at different distances from Coniston and soil pH values are given in Table 2. For all soils, except that obtained from 49.8 km from the smelter, the pH was lowest at the surface and increased with depth through the soil profile. In addition, pH increased with distance from the smelter.

In the water extracts, Ni concentrations were generally higher than Al and Cu. The highest metal concentrations again occurred in the surface soils and decreased through the profile. Also, the highest concentrations generally occurred closest to the smelter. Beyond a distance of 13.5 km, the concentrations of most of the metals were below the limits of detection of the technique and were recorded as non-detectable.

It must be emphasized that all metal concentrations recorded were for water extracts and are therefore probably an underestimate of the concentrations actually available to plants for growth.

Root elongation bioassays

A soil-water extract of 1:3 was used for bioassays. When seeds were germinated in these extracts, at all sites closer than 3.8 km from the smelter there was almost complete inhibition of radicle elongation. At distances greater than 3.8 km, the relative inhibition of root elongation as compared with controls, related to distance from the smelter. In addition, the surface soils were generally most toxic, except at distances greater than 13.5 km.

Investigations using single-salt metal solu-

tions of Cu, Ni, Co or Al confirmed that the concentrations of these metals in the soil-water extracts were inhibitory to root elongation (Whitby & Hutchinson 1974; Whitby 1974).

Although we attempted to establish vegetation on the Coniston soils in the field for three successive years (1969-71), only the results for 1970 are reported. Seedlings of several species, such as sugar maple *Acer saccharum* and red maple *Acer rubrum*, did not emerge at any site although the seeds were pre-treated and found to germinate well in the laboratory. Most seedlings which grew, especially those close to the smelter, showed marked sulfur-dioxide damage symptoms.

Germination at the field sites was very poor. The number of survivors and the mean weight (above ground growth) of seedlings of buckwheat *Fagopyrum sagittatum* are given in Table 3. The metal concentrations in the roots and leaves of *F. sagittatum* grown in the field experiments are given in Figure 2.

Fertilizer additions increased the above-ground biomass over that obtained with no treatment. This increased biomass became more significant as distance from the smelter increased. Growth increased dramatically when lime was added (with or without fertilizer). For example, at 7.4 km *F. sagittatum* grown in unamended soil had a mean weight of 13 mg, in fertilizer-treated soil 92 mg, in lime-treated soil 155 mg, and in fertilizer and limed soils 310 mg. Thus, raising the soil pH to 7.0 very markedly increased plant growth. Plants that survived in untreated or fertilizer-only additions failed to produce flowers. Plants grown in lime or lime and fertilizer produced flowers and abundant fruits.

After harvest, the plants were divided into roots, stems, leaves, flowers and seeds, for metal analysis. In most instances, the highest concentrations of metals were found in the roots. The lowest levels were found in the stems, followed by flowers and seeds. Leaves gave higher metal concentrations than did stems. For example, *Fagopyrum sagittatum* grown in fertilized soils at 1.6 km from Coniston, contained 594 ppm Ni and 909 ppm Cu in the roots, but only 299 ppm Ni and 175 ppm Cu in the shoots (Fig. 2).

The highest metal uptake occurred on unamended soils and those treated only with fertilizer. The application of lime decreased the concentration of metals found in the leaves and roots. For example, at 7.4 km, the roots of *Fagopyrum sagittatum* contained 882 ppm Ni and 1098 ppm Cu when grown in unamended soils, 292 ppm Ni and 183 ppm Cu when grown

TABLE 3. NUMBER OF SURVIVORS AND MEAN WEIGHT (mg) OF THE ABOVE-GROUND PORTIONS OF *Fagopyrum sagittatum* (BUCKWHEAT) GROWN IN THE FIELD DURING 1970.

Site Km		Treatments			
		None	Fertilizer	Lime	Lime & Fertilizer
1.5	* # survivors	--	---	8	10
	mean weight (mg)	--	---	188	443
1.7	* # survivors	--	4	1	49
	mean weight (mg)	--	74	178	442
7.4	* # survivors	8	48	27	10
	mean weight (mg)	13	92	155	310
10.4	* # survivors	1	18	30	25
	mean weight (mg)	21	174	227	257
19.3	* # survivors	3	2	3	1
	mean weight (mg)	33	85	343	273
49.8	* # survivors	1	3	1	---
	mean weight (mg)	21	27	134	---

At 1.9, 3.8 and 13.5 km, no seedlings emerged.

*Orig. # of seeds 50 per treatment.

TABLE 4. MEAN SHOOT WEIGHT OF *Raphanus sativus* GROWN UNDER GREENHOUSE CONDITIONS ON THE SOILS COLLECTED AT VARIOUS DISTANCES FROM THE CONISTON SMELTER.*

Distance (km)	Treatment			
	None	Fertilizer (mg./seedling)	Lime	Lime & Fertilizer
7.4	5.6±1.8	4.6±1.8	41.7±27.0	23.0±14.0
10.4	16.5±4.4	43.0±35.9	60.8±11.8	123.1±38.6
13.5	15.7±5.8	78.5±55.8	42.8±13.2	87.0±25.9
19.3	8.7±1.1	7.8±3.3	59.5±15.3	121.0±46.3
49.8	67.8±17.9	76.4±39.3	101.8±28.1	62.9±34.3

*Each weight is the mean of 10 seedlings that survived the one-month growing period. Seedlings grown on soils collected within 7.4 km of Coniston did not survive the one-month growing period.

in fertilizer-treated soils, 213 ppm Ni and 209 ppm Cu when grown in limed soils, and 88 ppm Ni and 158 ppm Cu when grown in limed and fertilized soils.

Metal concentration in the plants decreased with increasing distance from the Coniston smelter. For example, *Fagopyrum sagittatum* roots in limed and fertilized soils at 19.3 km contained 226 ppm Ni and 271 ppm Cu, compared with 335 ppm Ni and 478 ppm Cu in limed and fertilized soils from 1.5 km.

Species differed in the quantities of metals they accumulated (Whitby 1974). The cereals, oats and barley, and the grass *Deschampsia flexuosa* contained higher concentrations than did other species such as buckwheat and jack pine.

Greenhouse experiments

Dry weights for *Raphanus sativus* grown under greenhouse conditions on treated and untreated soils are given in Table 4. Seedlings

grown on soils collected within 7.4 km of the smelter died within the 28-day period.

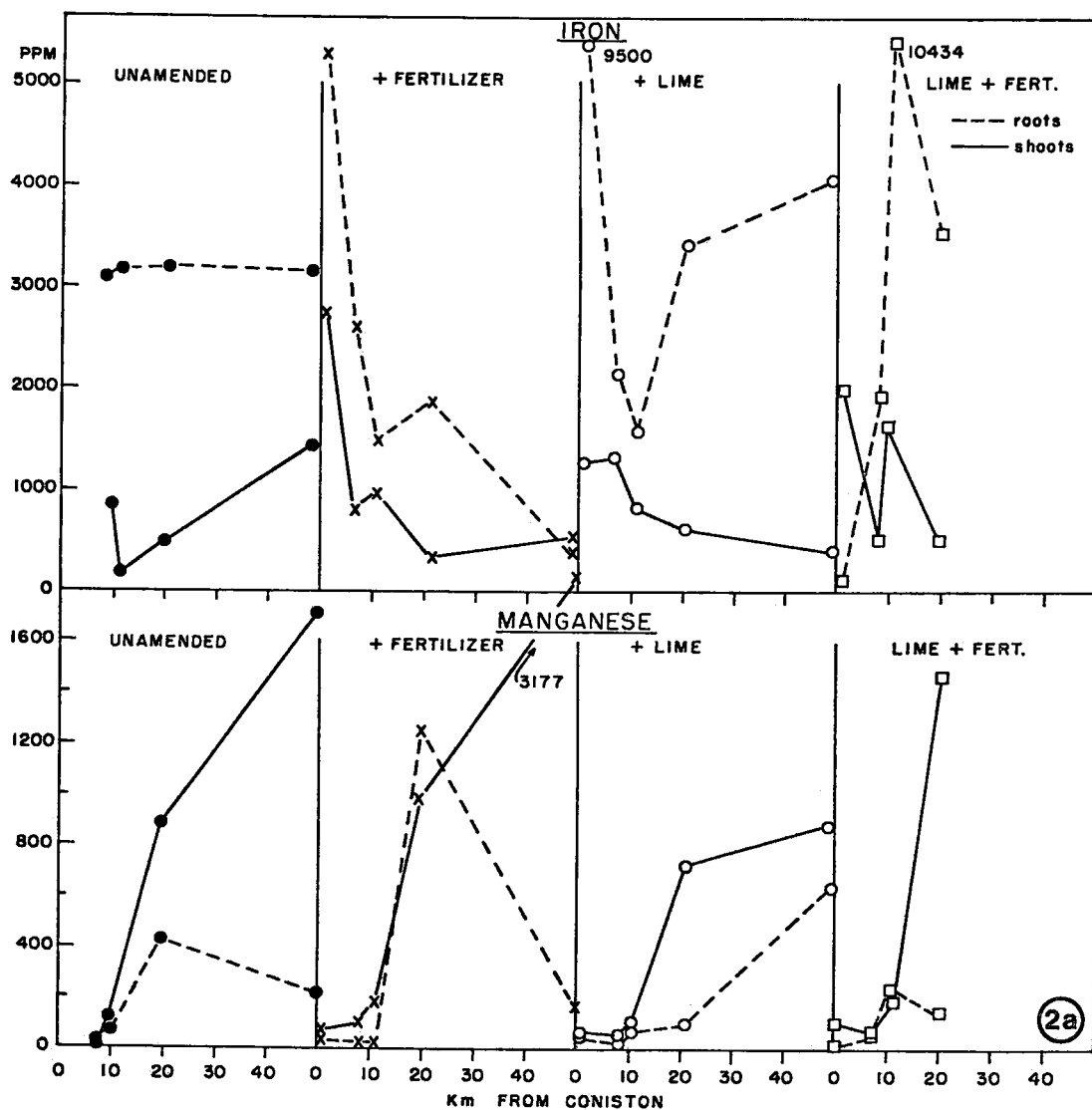
On the soil collected at 7.4 km from Coniston all species survived, but growth was extremely poor on the unamended soil. Fertilizer treatments alone did not increase growth. Plants grew best on the limed soils. Lime plus fertilizer treatments increased plant growth compared with controls (Table 4). Similarly on soil collected from 10.4 km and beyond, the growth was poorest in unamended soils. With the exception of the 19.3 km site, however, there was a marked response to fertilizer. Indeed, in some cases, the response was as high or higher than that to lime. For example, at 13.5 km the mean

weight of *Raphanus sativus* was 15.7 mg in unamended soils, 78.5 mg in fertilized soil, 42.8 mg in limed soil and 87.0 in limed and fertilized soil.

The concentrations of Ni, Cu, Fe, and Mn in roots and shoots from greenhouse experiments are given for *Raphanus sativus* in Table 5.

Plants grown in unamended soils contained the highest concentration of metals. These concentrations were lower in fertilizer applications. Concentrations were lowest in the lime plus fertilizer treatment.

Metal concentrations were higher in the roots than in the shoots for Ni, Cu, and Fe. Mn distributions between shoot and root did not follow



this pattern. The highest Mn concentrations were often found in the shoots rather than in the roots. Mn in the seedlings also increased with distance from the smelter.

Aquatic studies

The location of the test lakes are shown in Figure 1. The standing crop of phytoplankton in the lakes was measured in 1969 and 1970 and data for five of the lakes are given in Table 6. Lakes close to the smelter had markedly-reduced algal populations, with only a very few individual cells. A count of the order of 2,000 cells per ml is considered normal for the oligotrophic Shield lakes. Dill Lake (11.6 km from the smelter) alone had such values.

Species diversity (not shown in Table 6) increased with distance from the smelter, i.e. 3 species in Baby Lake and more than 50 species in Dill Lake. The Baby Lake species were metal-tolerant (Stokes *et al.* 1973). The algal groups in the polluted lakes were unicellular

green algae, whereas diatoms were present as dominants in Dill, and also present in Richard. Blue-green algae and Chrysophyceae were present in Daisy Lake. This increase in diversity and apparent sensitivity of certain groups is paralleled in the higher plants at terrestrial sites along transects from the Sudbury smelters.

Fish populations are now absent from Alice and Baby Lakes and invertebrates very rare. Elevated Cu and/or Ni levels occur in Alice, Baby and Daisy Lakes. Algal bioassays of some lake waters (Fig. 3) are shown for the unicellular green alga *Chlamydomonas eugametos*, both for unaltered lake waters and for additions of nutrients (BBM) or pH adjustment to 6.8. Long Lake had low Ni and Cu, whereas Kelley showed good growth despite elevated Ni and Cu levels. Boucher and Norway lakes are also high in Ni and Cu. They are located near Falconbridge (Fig. 1), whereas Lady Macdonald Lake is approximately 1 km west of Copper Cliff. Long Lake, the furthest of those shown

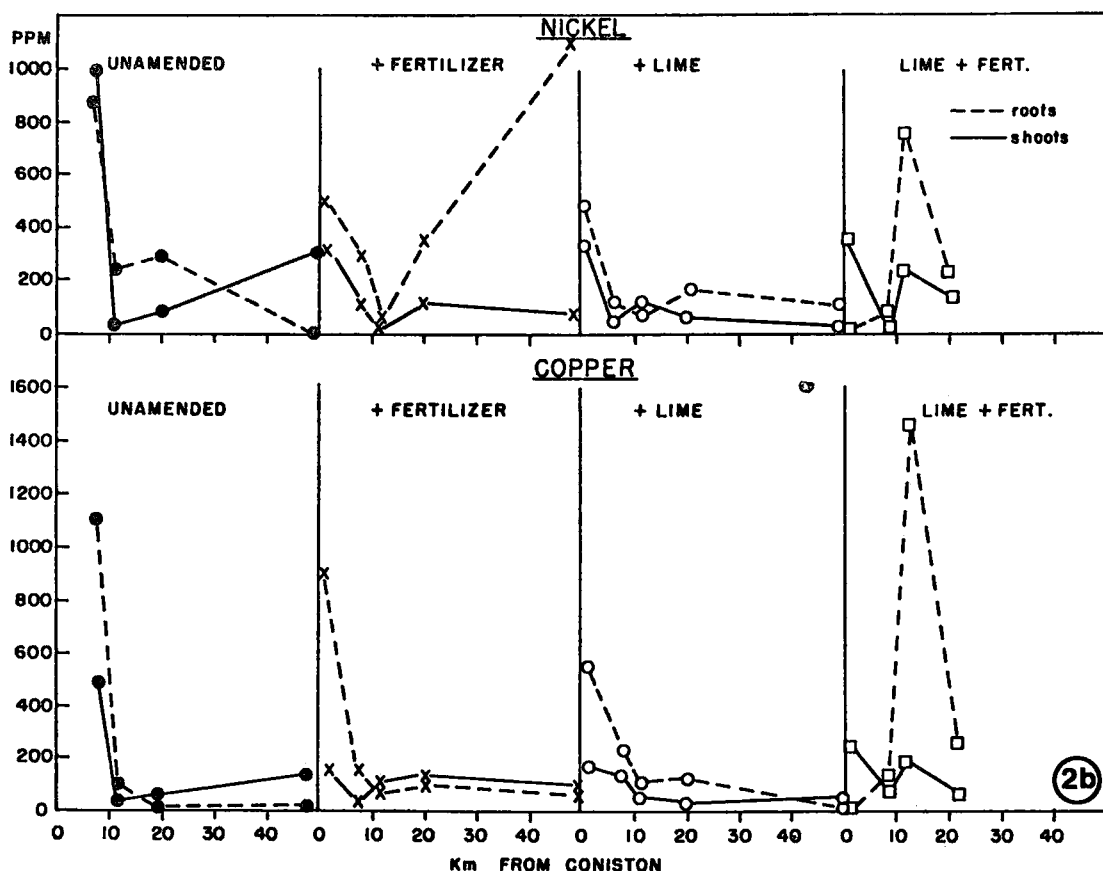


FIG. 2. Metal concentrations in leaves and roots of *Fagopyrum sagittatum* (buckwheat), grown in field experiments, 1970: (a) Fe and Mn; (b) Ni and Cu.

TABLE 5. CONCENTRATIONS OF METALS IN *Raphanus sativus* GROWN IN GREENHOUSE, ON SOILS COLLECTED IN CONISTON AREA.

Distance from Coniston smelter	Treatment	Plant Part	Ni	Cu	Fe	Mn
7.4 km	N	Shoot	433	305	347	81
		Root	1,150	926	4,920	76
	F	Shoot	94	74	204	43
		Root	669	513	3,790	39
	L	Shoot	51	25	226	26
		Root	534	406	1,472	32
	L & F	Shoot	101	49	102	37
		Root	300	420	1,800	20
14.3 km	N	Shoot	220	24	1,296	164
		Root	429	136	1,857	29
	F	Shoot	28	8	143	39
		Root	206	126	853	52
	L	Shoot	16	12	115	38
		Root	192	153	552	34
	L & F	Shoot	7	6	183	22
		Root	83	97	1,187	47
49.8 km	N	Shoot	14	4	84	358
		Root	67	46	700	947
	F	Shoot	4	3	95	317
		Root	20	32	574	495
	L	Shoot	3	2	58	50
		Root	20	60	883	151
	L & F	Shoot	100	26	2,005	283
		Root	163	138	2,875	213

*Treatment - N = no amendment, F = fertilizer added, L = lime added, L & F = lime plus fertilizer.

TABLE 6. PHYTOPLANKTON STANDING IN SOME SUDBURY LAKES (CELLS PER ml)

Lake	Distance from Coniston smelter km	Date				
		July 1969	Aug 1969	May 1970	June 1970	July 1970
Alice	2.0			negligible		
Baby	2.4			negligible		
Daisy	4.0	11	195	191	523	144
Richard	6.4	1040	1496	427	1311	740
Dill	11.6	2121	1504	1503	3288	1154

Alice:	deep glacial till on one side, blackened eroded rocks on the other three, largely devoid of vegetation. <i>Typha</i> on glacial till at lake edge.
Baby:	steep, eroding rocky sides and slopes, lacking vegetation.
Daisy:	sparse vegetation, a few deciduous SO ₂ -resistant trees in protected areas, no ground vegetation.
Richard:	deciduous trees, good ground cover, glacial till over bedrock, complete ground cover.
Dill:	jack pine in with deciduous trees, complete ground cover.

TABLE 7. HEAVY METALS (ppm) AND pH VALUES FOR SOME LAKES IN THE CONISTON AREA OF SUDBURY, SEPTEMBER 1970

Lake	Distance (km) from nearest smelter	pH	Ca	Cu	Ni	Ag	Cd	Fe	Sulfate
Alice	2.0 km	6.3	11.2	0.06	6.4	0.001	0.006	0.23	275
Baby	2.4	4.1	3.6	0.52	2.7	0.003	0.003	0.28	72
Daisy	4.0	6.1	5.8	0.20	0.40	nd	0.002	0.10	48
Richard	6.4	6.4	12.8	0.005	0.15	nd	0.002	0.25	40
Dill	11.6	6.7	8.2	0.003	nd	nd	nd	0.30	25
Long	7.0	7.1	5.0	0.03	0.12	0.003	nd	0.30	-
Kelley	3.2	6.5	60.8	0.16	0.95	0.008	0.008	0.73	-
Lady Macdonald	3.0	5.0	14.6	0.49	3.02	0.03	0.006	0.37	-

nd = below detection limits of method.

in Figure 2, is approximately 15 km SSW of Coniston and 7 km SW of Copper Cliff.

The stippled histograms for unamended waters clearly indicate the deleterious nature of the lake waters to algal growth. Lakes such as Norway, Boucher, Alice, Baby and Lady Macdonald are quite toxic to this organism. Growth in Long Lake was the best in unamended waters. The acidic nature of lakes such as Baby and Lady Macdonald (pH 3.7) was probably a contributory factor in this poor response. The adjustment of all pH's to 6.8 and the addition of nutrients (added since this may be a limiting factor in oligotrophic and/or acidic Lakes) evoked marked responses in some instances. Norway Lake became a better medium for algal growth and large responses occurred in Kelley, Ramsey and Long Lake flasks. However, the inhibitory factor(s) in Alice, Baby and Lady Macdonald Lakes were hardly touched by these adjustments.

Sulfate levels declined with distance with excessive levels in those lakes close to the stacks (Table 7). Bioassays on single-salt solutions of sulfate (up to 250 ppm), of Ni (up to 1.5 ppm), and of Cu (to 1.0 ppm) indicated a non-toxic effect of sulfate even at very high levels, a high toxicity of Cu even at 0.1-0.2 ppm, and lesser toxicity of Ni, with death of cells at 0.5-1.0 ppm (Stokes & Hutchinson 1975).

The very high Ni levels of Alice Lake (6.4 ppm) and Cu levels in Baby Lake support the contention that metals were the main source of toxicity to algae. Lady Macdonald was found to have 1.5 ppm Cu and up to 7.0 ppm Ni in 1971. Boucher Lake in 1970 had a pH of 7.5, Cu levels of 0.10 ppm but Ni of 2.5 ppm.

DISCUSSION

A number of studies have emphasized the deleterious ecological consequences of sulfur-dioxide emissions from the Sudbury-area smelters on the forests of the area (Linzon 1958; Gorham & Gordon 1960; Dreisinger 1970), and on the quality of lake waters (Gordon & Gorham 1963; Gorham & Gordon 1960b). Leblanc & Rao (1966) correlated the decline of lichens and mosses in the Sudbury area with their sensitivity to sulfur-dioxide fumigation.

In the present studies emphasis has been placed on heavy-metal emissions and their accumulation in soils as potential toxic factors in seedling establishment of the forest species (Hutchinson & Whitby 1974; Whitby & Hutchinson 1974). Widespread contamination of the soils of the area has been shown for the smelted metals nickel, copper and iron. Co, which is

produced in lesser quantities, has also accumulated in surface soils around the Coniston smelter. Ni and Cu especially are currently present to levels of 1000 ppm and more for a distance of 7.4 km from the Coniston stack. Concentrations decline with distance from the smelters and with depth through the soil profile. Elevated levels of Ni, Cu, and Fe occur in the foliage of natural vegetation. For example, Bowen (1966) quotes 'normal' levels for land plants as 3 ppm for Ni, and 14 ppm for Cu. Stone (1968) found, for *Acer rubrum*, that foliar Cu levels in a non-polluted area were 3.6 to 9.0 ppm. In the present study, *Acer rubrum* Ni levels were in excess of 30 ppm to 20 km from Coniston, and Cu in excess of 20 ppm also to 20 km (Table 1). Al and Fe concentrations showed similar patterns of elevation. The increased acidity of soils close to the smelters is

believed to be a contributing factor in increasing heavy-metal uptake, and in converting these metals to soluble (and therefore 'available') forms.

In the field experiments a large number of species were sown as seed at sites on transects out from Coniston. Most species failed to germinate and/or survive for even a single growing season. Buckwheat (*Fagopyrum sagittatum*) gave the best over-all performance. Many seedlings of several surviving species showed sulfur-fumes damage to their foliage. Analysis of field-grown plants disclosed very high levels of Ni and Cu within the foliage of the seedlings (Fig. 2). These levels were markedly reduced by a soil amendment of lime, which also increased the size of the plants and their survival (Table 3).

Rather similar results were obtained when

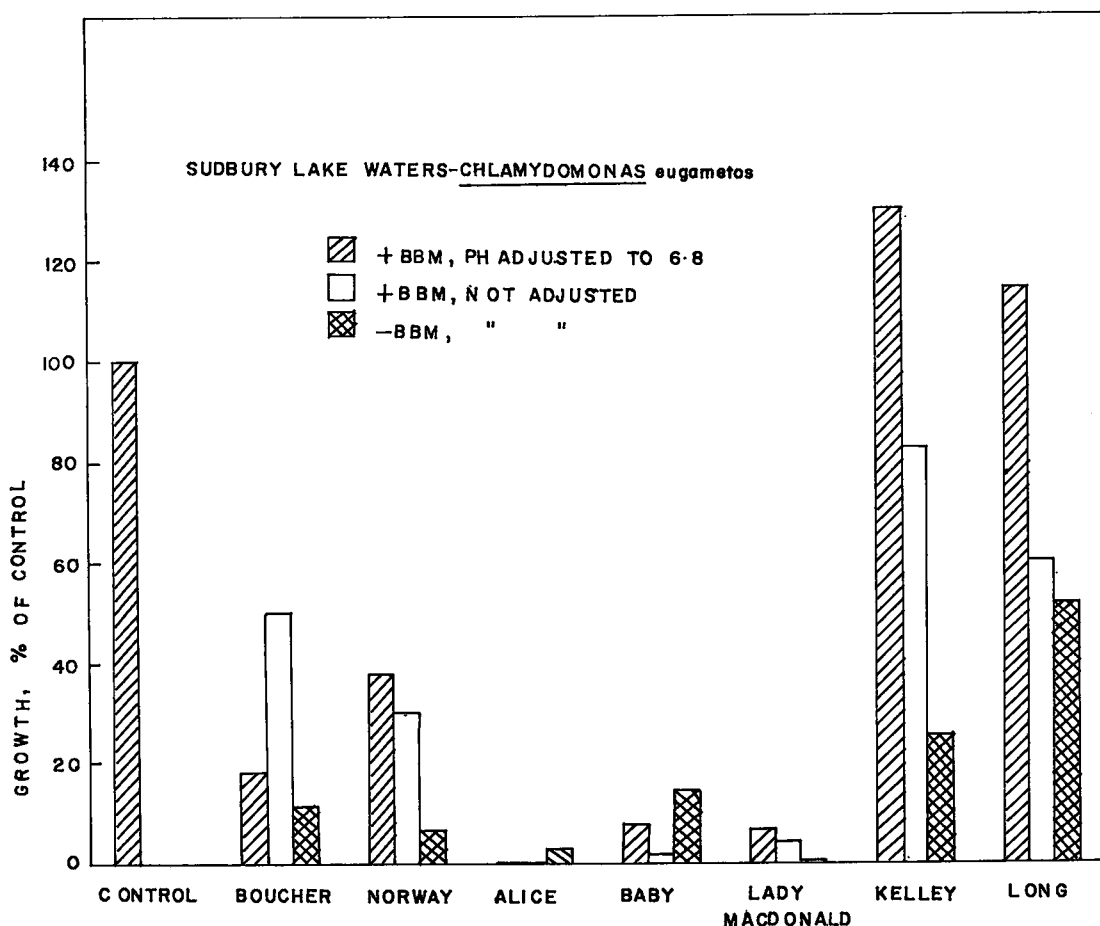


FIG. 3. Growth, expressed as cell counts after 10 days in cultures, of cells of *Chlamydomonas eugametos* in controlled laboratory conditions when grown in lake waters from the Sudbury region, with or without pH adjustment and nutrient (BBM) additions.

soils were removed from the field and seedlings grown on them under controlled greenhouse conditions. These conditions excluded the complicating field factors of sulfur-dioxide fumigations, droughts and micro-meteorological differences between sites. None of the seedlings of the species tested survived the 28-day experimental period on soils obtained within 7.4 km from the Coniston smelter (Table 4). Clearly these soils were toxic to the seedlings and the similar responses in the laboratory and at the field sites are strongly suggestive that heavy-metal accumulations have exceeded seedling tolerances over a wide area. Forest regeneration, without soil amendment, is thus seriously in doubt. Indeed, the greenhouse-grown plants showed symptoms typical of heavy-metal toxicity. Lime and lime plus fertilizer amendments were again successful in a) improving growth and survival and b) decreasing heavy-metal uptake to the tops (Table 5).

Water extracts of the field-collected soils confirmed the highly soluble nature of a significant percentage of the Ni, Cu, and Al in soils within 10 km of the smelter. Water-extractable Ni at concentrations to 20 ppm (and as high as 420 ppm) was found in surface soils. A similar pattern was found for Cu and for Al (Table 2). All three of these metals have been found to reduce root growth of a number of test species by as much as 50% at solution concentrations of 2 ppm or less (Whitby & Hutchinson 1974). The potential toxicity to vegetation of these soils in the field is clear. The situation is not unique however, as Buchauer (1973) reported similar vegetational effects near a Pennsylvania smelter. The scale is much greater at Sudbury.

The loss of the forest cover and the consequent erosional loss of the soil has also been a factor in contamination of the many lakes in the area. Soluble and insoluble heavy metals have been entering water bodies for the past 40-50 years, and the metal loss of the waters has been accentuated by the acidifying nature of the precipitation, which yields sulfur-laden acid rains over an extensive area.

The phytoplankton studies have indicated problems regarding both increased lake acidity and increased Ni, Cu, and sulfate contents of lakes close to the smelters (Stokes *et al.* 1973; Hutchinson & Stokes 1975). The decline in fish populations which correlate with increased acidification has already been referred to in the literature (Beamish & Harvey 1972).

Algal standing crops were measured in 1969-71 and the depauperate nature of lakes close

to Coniston was revealed. Both standing crops and species diversity were found to be markedly reduced (Table 6). The chemical analyses revealed several lakes to have pH as low as 4.0 and Cu and Ni concentrations to 1 and 6 ppm respectively. The presence of a very few surviving cells in some of the lakes strongly polluted with heavy metals is perhaps surprising in view of the high toxicity of Cu to algae at levels as low as 0.1 ppm and Ni at levels as low as 0.5 ppm (Hutchinson 1973). However, having found some cells still surviving, it is then not surprising to find them to be both nickel- and copper-tolerant (Stokes *et al.* 1973). An additional factor is that for many algae, Ni and Cu mutually enhance the toxicity of each other, in an example of heavy-metal synergism (Hutchinson 1973; Stokes 1974).

Although pH adjustments and nutrient additions had marked effects in ameliorating the toxicity of some waters, for the worst-contaminated lakes even this was not enough to allow algal growth, *e.g.* Alice Lake. The case of Kelley Lake is a special one — perhaps suggesting an approach to acid lake restoration other than simple ant-acid additions via lime. Kelley Lake should have a poor phytoplankton population and be highly toxic to algae if its heavy-metal content alone is considered. However, the presence of a high organic content via sewage inputs appears to play a major role in reducing the "expected" toxicity. Organic ligands complexing potentially-toxic heavy metals seem to be part of the answer, although the increased divalent-cation levels, especially of calcium, may be contributing factors. This aspect is currently being studied at the University of Toronto (*e.g.* Stokes & Hutchinson 1975).

The possibility of the high sulfate levels being controlling toxic factors was largely ruled out by bioassays using sulfate gradients as high as 250-300 ppm sulfate. These high levels did not inhibit algal growth.

Since the acidity of lakes over a wide area around Sudbury, and especially to the southwest, has increased over the past 15 years, one can anticipate a continuing erosion of life in the affected lakes. Heavy-metal and acidity interactions are likely to lead to reduction and loss of the primary producers. This is paralleled and often preceded by loss of zooplankton and fish populations. Control of sulfur-dioxide emissions appears to be an essential first step. Heavy-metal controls on emissions also need to be instituted, although so much is now available from soil run-off that it is apparent that very long-term damage is already done.

ACKNOWLEDGEMENTS

The work was partly supported by National Research Council operating grants to two of the authors (P. M. Stokes and T. C. Hutchinson).

REFERENCES

- BEAMISH, R. J. & HARVEY, H. H. (1972): Acidification of the La Cloche Mountain lakes, Ontario and resulting fish mortalities. *J. Fish. Res. Bd. Canada* 29, 1131-1143.
- BOWEN, H. J. M. (1966): *Trace Elements in Biochemistry*. Academic Press, New York 241 p.
- BUCHAUER, M. J. (1973): Contamination of soil and vegetation near a zinc smelter by zinc, cadmium, copper and lead. *Environ. Sci. Tech.* 7, 131-135.
- COSTESCU, L. M. & HUTCHINSON, T. C. (1972): The ecological consequences of soil pollution by metallic dust from the Sudbury smelters. *Proc. 18th Ann. Mtg. Inst. Environmental Sci.*, New York, 540-545.
- DREISINGER, B. R. (1970): SO₂ levels and vegetation injury in the Sudbury area during the 1969 season. *Dept. Energy Res. Management, Province of Ontario, Canada*, publ. April 1970, 45 pp.
- GORHAM, E. & GORDON, A. G. (1960a): Some effects of smelter pollution northeast of Falconbridge, Ontario. *Can. J. Bot.* 38, 307-312.
- & ——— (1960b): The influence of smelter fumes upon the chemical composition of lakes waters near Sudbury, Ontario and upon the surrounding vegetation. *Can. J. Bot.* 38, 477-487.
- & ——— (1963): Some effects of smelter pollution upon the aquatic vegetation near Sudbury, Ontario. *Can. J. Bot.* 41, 371-378.
- HUTCHINSON, T. C. (1973): Comparative studies of the phytotoxicity of heavy metals to phytoplankton and their synergistic interactions. *Water Pollution Res. Can., Proc. 8th Can. Symp.* (in press).
- & STOKES, P. M. (1975): Heavy metal toxicity and algal bioassays. In *Water Quality Parameters*. Amer. Soc. Testing Mat. Spec. Tech. Pub. 573, Philadelphia.
- & WHITBY, L. M. (1974): Heavy metal pollution in the Sudbury mining and smelting region of Canada. 1. Soil and vegetation contamination by nickel, copper, and other metals. *Environmental Conservation* 1, 123-132.
- & ——— (1975): The effects of acid rainfall and heavy metal particulates on a boreal forest ecosystem near the Sudbury smelting region. Presented at *First Internat. Symposium on Acid Precipitation and the Forest Ecosystem*.
- LEBLANC, F. & RAO, D. N. (1966): Réactions de quelques lichens et mousses épiphytiques à l'anhydride sulfureux dans la région de Sudbury, Ontario. *Bryologist* 69, 338-346.
- LINZON, S. N. (1958): The influence of smelter fumes on the growth of White Pine in the Sudbury region. *Ontario Dept. Lands Forests and Ontario Dept. Mines*. 45 pp.
- STOKES, P. M. (1974): Uptake and accumulation of copper and nickel by metal tolerant strains of *Scenedesmus*. *Proc. XIX Congress Internat. Assoc. Limnology*.
- & HUTCHINSON, T. C. (1975): Copper toxicity to phytoplankton, as affected by organic ligands, other cations and inherent tolerance of algae to copper. *Symp. Internat. Joint Commission Toxicity of Biota to Metal Forms in Natural Waters*. Duluth Oct. 1975 (in press).
- , ——— & KRAUTER, K. (1973): Heavy metal tolerance in algae isolated from contaminated lakes near Sudbury, Ontario. *Can. J. Botany* 51, 2155-2168.
- STONE, E. L. (1968): Microelement nutrition of forest trees; a review. In *Forest Fertilization: Theory and practice*. Tennessee Valley Authority USA. 306 pp.
- WHITBY, L. M. & HUTCHINSON, T. C. (1974): Heavy metal pollution in the Sudbury mining and smelting region of Canada. II Soil toxicity tests. *Environmental Conservation* 1, 191-200.
- WHITBY, Leslie (Costescu) (1974): *The Ecological Consequences of Airborne Metallic Contaminants from the Sudbury Smelters*. Ph.D. thesis, Univ. Toronto, Dept. Botany.

Manuscript received July 1975, emended November 1975.