POST-SUPERSTACK SUDBURY SMELTER EMISSIONS AND THEIR FATE IN THE ATMOSPHERE: AN OVERVIEW OF THE SUDBURY ENVIRONMENT STUDY RESULTS

WALTER H. CHAN and MARIS A. LUSIS

Air Resources Branch, Ontario Ministry of the Environment, 880 Bay Street, 4th Floor, Toronto, Ontario, Canada M5S 1Z8

(Received July 20, 1984; revised December 4, 1984)

Abstract. This paper describes investigations into the characteristics and atmospheric fate of emissions from the INCO and Falconbridge Ni smelters at Sudbury, Ontario. The former smelter was of special interest, being one of the largest point sources in North America, with most of its emissions being released from a 381 m 'Superstack'. Emission rates of S compounds, trace metals, and a number of other substances, have been quantified. The subsequent dispersion, transformation and deposition of these pollutants has been studied in a number of intensive field campaigns, as well as by longer-term precipitation and air monitoring programs. The results of these investigations are summarized, and the contribution of Sudbury emissions to wet and dry deposition in the Sudbury Basin assessed.

1. Introduction

Sudbury, the location of one of the world's largest Ni smelters (Figures 1 and 2) and a major source of SO_2 and trace metal emissions in North America, has in recent years been the focus of heated debates, both in the context of local impacts of the smelting activities, and the long-range transport of air pollutants and the acid rain phenomenon.

Several changes in the local smelting practices have taken place since the beginning of the century (Ontario Ministry of the Environment, 1982a), progressing from the crude ignition of open piles consisting of sandwiches of sulfide ore and timber (which caused extensive local damage of soils and vegetation due to trace metal and SO_2 pollution), to the construction of smelters in the 1920's and 1930's with stacks to disperse the emissions. At present, two smelters operate in the Sudbury area-INCO Limited and Falconbridge Limited – having current rated production capacities in the order of 137 and 40×10^6 kg Ni yr^{-1}, respectively. Damage to the ecosystems in the vicinity of Sudbury due to smelter emissions has been well documented (for example, see references Hutchinson and Whitby, 1974a, b, 1977; Whitby and Hutchinson, 1974; Beamish *et al.*, 1975a, b; Beamish and Van Loon, 1977; Freedman and Hutchinson, 1980b; Gorham and Gordon 1960a, b). In August 1972, with the belief that tall stacks would significantly improve local air quality, the world's tallest stack, the INCO 381 m 'Superstack' became operational.

In 1973, MOE initiated a multi-disciplinary study to assess both the atmospheric and aquatic environmental impact aspects in the Sudbury area. The aquatic study has been reported elsewhere (Ontario Ministry of the Environment, 1982b). The early atmos-

Water, Air, and Soil Pollution **26** (1985) 43–58. 0049–6979/85.15.

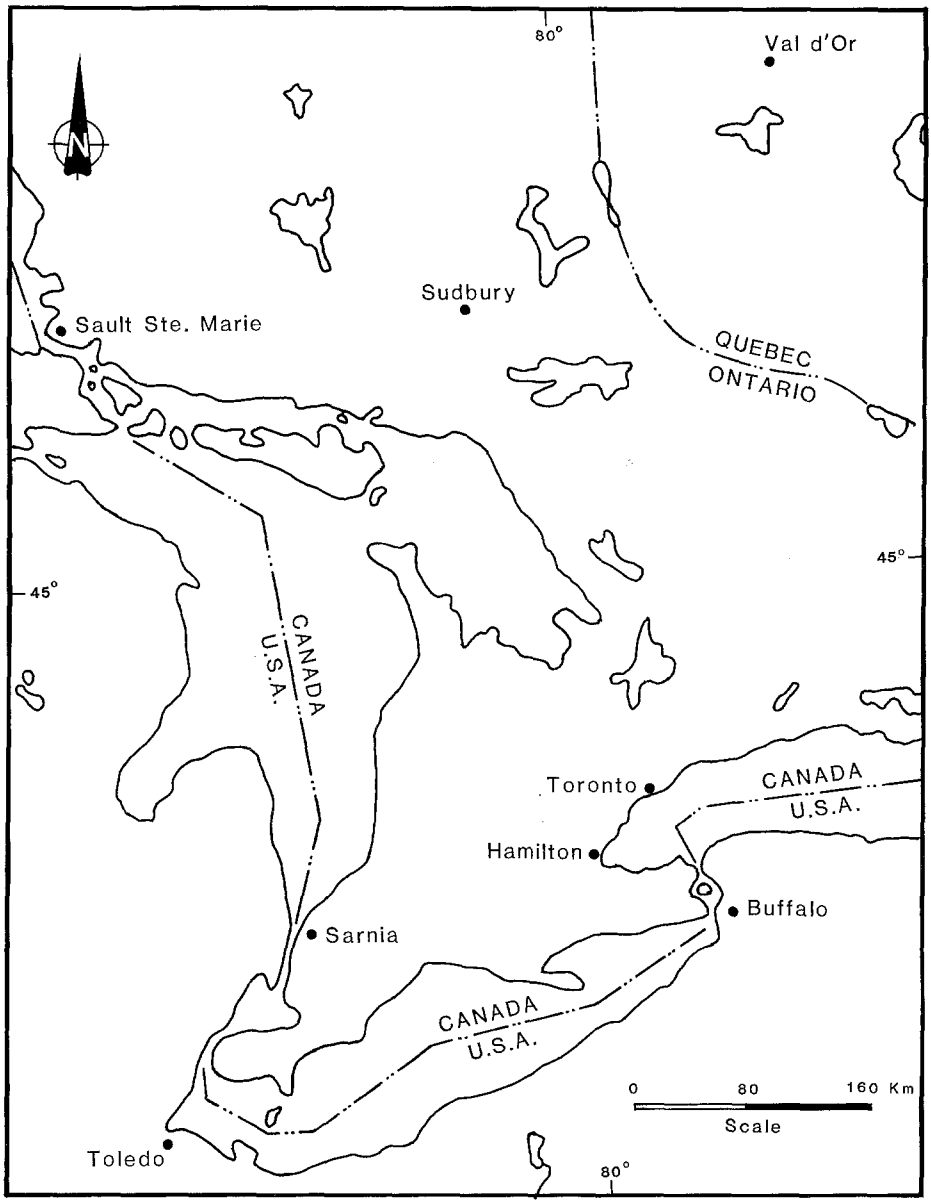

Fig. 1. Location of the Sudbury Basin in Ontario.

pheric studies took the form of projects awared to universities and consulting firms. As a consequence of the findings of the early work, the Air Resources Branch (ARB) of the Ministry began an integrated program in 1977, an overview of which is given by Chan (1982).

In essence, the problem under study was to determine the origin and fate of the pollutants, the nature and quantity of the emissions, their dispersion, chemical trans-

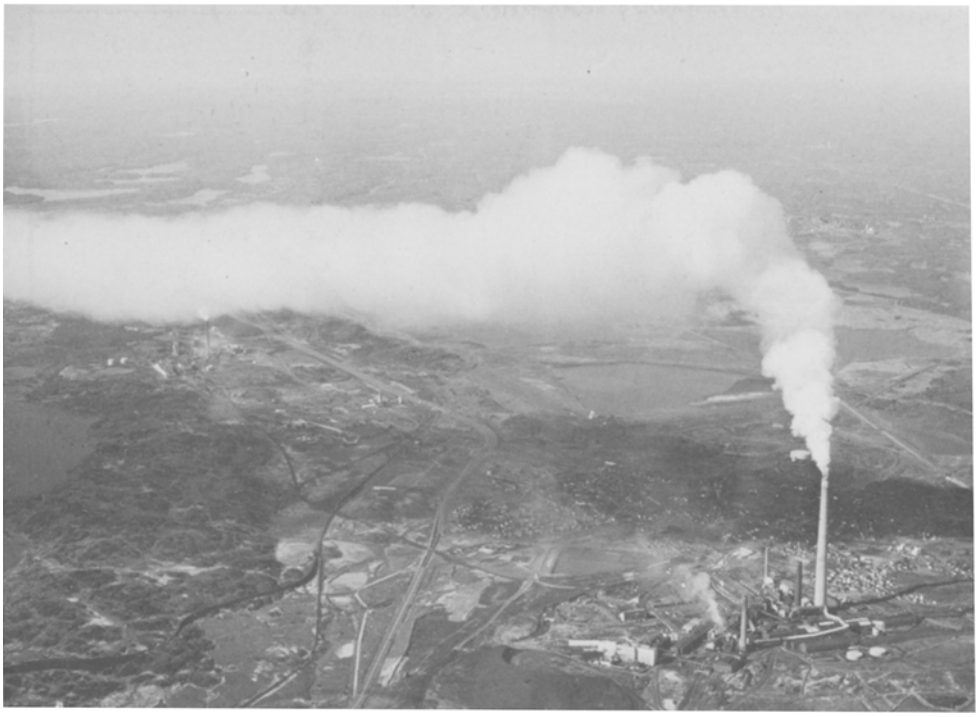

Fig. 2. The INCO 381 m Superstack.

formation, and ultimate deposition as listed in Figure 3. Information on emission rates and characteristics was obtained by stack tests or airborne plume-level sampling near the stack; information on the subsequent fate of the emissions resulted from routine air and precipitation monitoring as well as special field studies. Mathematical models developed for the Sudbury area were used not only to aid in the interpretation of the experimental data, but also to examine various emission control scenarios.

This account summarizes the results of the atmospheric research carried out after the Superstack was erected and operational, with a special emphasis on the comprehensive studies carried out by the Ontario Ministry of the Environment (MOE).

2. Source Testing

There are a number of chimneys associated with the Sudbury smelters which emit gases from pyrometallurgical smelting processes (the 381 m and 194 m stack at INCO, and the 93 m stack at Falconbridge). Appreciable amounts of SO_2 and trace metals also emanate from low-level sources such as roof vents and a pelletizer plant (which has two 45 m stacks). An extensive series of emission tests over the period 1973–1981 (Ozvacic, 1982) has led to the overall annual average values for the major emittants shown in Table I. It should be noted that over the study period, there has been a considerable variation in annual emission rates, largely due to smelter shutdowns or process changes.

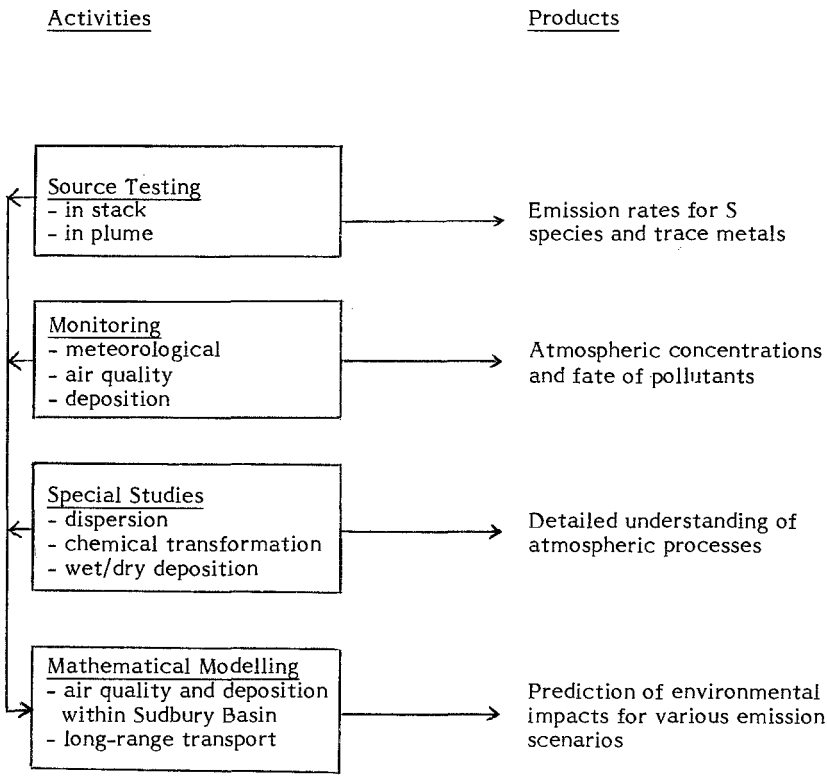

Fig. 3. Schematic representation of the atmospheric component of the Sudbury Environmental Study.

TABLE I

Average yearly emissions (tonne) of major pollutants in the Sudbury Basin for the period 1973–1981[a]

Source	SO_2	H_2SO_4	Total particulate	Fe	Cu	Ni	Pb	As
INCO 381 m stack	885667	7270	11417	990	245	228	184	114
INCO 194 m stack	54568	1664	2380[b]	643	171	226	6	4
INCO smelter (low level)	12000	88	586	70	242	31	0.6	0.1
INCO Two 45 m stacks			4073	2354				
Falconbridge 93 m stack	173000	438	865	98	11	9.6	13.4	6.4
Total	1125235	9460	15248	1801	669	500	204	125

[a] Basis: 365 days × 24 hr day^{-1} production.
[b] Based on tests conducted in 1977 only.

TABLE II

Variation of SO_2 and particulate emission rates (10^3 tonne) at INCO 381 m Superstack

	SO_2	Particulates
1973	1171	12.2
1974	1126	14.1
1975	1141	14.3
1976	1163	14.5
1977	1069	13.8
1978	535	8.1
1979	383	6.5
1980	733	10.1
1981	650	9.2

Table II, for example, shows the annual variation in emission rates of SO_2 and particulates at the INCO 381 m stack, over the period 1973–1981. It may also be noted that measurements of NO_x and HCl have also been made at the 381 m stack, showing emission rates of about 3000 and 500 tonne yr^{-1}, respectively.

Particle size distributions have been measured, both in the two tallest stacks (Ozvacic, 1982) and in the plume near the stacks (Chan *et al.*, 1982a, b, 1983). The in-plume studies, which are perhaps of greater interest in the context of atmospheric deposition (since particle size distributions are modified when the hot gases are mixed with ambient air on exiting from the stack), have shown that in the 381 m stack plume, metals such as Fe, Cu, Ni, Al, Mg and Mn are in particles with mass median diameters greater than 2.5 μm, while Pb, Zn, As, $SO_4^=$, NH_4^+ and Se occur in particles with mass median diameters less than 2.5 μm. Observations were generally similar in the Falconbridge stack plume, while at the 194 m stack plume, particle size distributions appeared to depend on the plant operating conditions. Most of the emitted sulfate is in the form of H_2SO_4 and depending on the source is about 0.4 to 3% of the SO_2 emissions.

3. Monitoring Programs

A number of monitoring programs have been carried out, especially over the period 1978–1980 to determine the ambient concentrations and fate of the pollutants deposited through wet and dry mechanisms, and to provide data for model validation.

3.1. Wet Deposition Monitoring Networks

3.1.1. *Bulk Sampling Network*

The first organized precipitation monitoring program in the Sudbury area in the early seventies was made by Kramer (1973, 1975, 1976) who operated a comprehensive bulk deposition network in central and northern Ontario. Using data obtained from this precipitation network with a varying number of stations from 1970 to 1974, Muller and Kramer (1977) concluded that with the commissioning of the 381 m stack, while there

was less deposition of $SO_4^=$, Fe, Cu and Ni near the source, the affected area had increased. However, deposition rates were still found to be highest in the Sudbury area. Deposition rates (in decreasing order) were found to be as follows: Fe, Ni, Cu, and S. The fraction of emitted pollutant deposited within an area of 3200 km^2 varied from over 100% for Fe, 69% for Ni, 42% for Cu, to 0.6% of total S.

Qualitatively similar results from bulk sampler measurements, suggesting that the smelter-emitted trace metals are mainly locally deposited whereas most of the SO_2 undergoes long range transport, have been reported by other investigators (Freedman and Hutchinson, 1978; Scheider *et al.*, 1980; Jeffries and Snyder, 1981; Kramer and Snyder, 1977).

3.1.2. *Cumulative Monthly Network*

The Air Resources Branch of MOE completed the installation of a wet only monthly deposition monitoring network in mid-1978, to measure the long term wet deposition field out to a radius of 150 km from Sudbury (see Figure 4). This network and the daily network described in Section 3.1.3 were operational until the end of the Sudbury Environmental Study field program in May 1980 (Chan *et al.*, 1982c, d).

Concentration and deposition fields of the pollutants measured in the cumulative monthly network were assessed in the light of concurrent smelter operations (Chan *et al.*, 1982c, 1984b). It is of special interest that INCO was not operating, either due to maintenance shutdowns or labor disputes during most of the first study year (Falconbridge shutdown – July 1 to August 21, 1978; INCO shutdown – July 17 to August 27, 1978; and INCO strike – September 16, 1978 to June 7, 1979). Thus comparison of the two periods June 1978 – May 1979 and June 1979 – May 1980 yielded information on the long-term smelter impact on wet deposition in the Sudbury Basin.

The overall network geometric mean precipitation concentrations (for all cumulative network stations) of the 78/79 and 79/80 periods referred to above were: pH, 4.1 (78/79) and 4.2 (79/80); and in mg L^{-1} sulfate, 3.4 and 3.4; N-nitrate, 0.6 and 0.6; N-ammonium, 0.3 and 0.4; Fe, 0.06 and 0.07; Cu, 0.003 and 0.004; Ni, 0.001 and 0.002; Pb, 0.013 and 0.009; and Zn, 0.011 and 0.009. Results of a Student – t test analysis indicate that the mean concentrations for the two periods are not statistically different at the 95% confidence level. Bearing in mind of the significant difference in the emission rates for the two study years, it is clear that other factors, such as the contribution of species by long range transport from sources outside of the immediate area, and year-to-year meteorological variability, have a significant effect on Sudbury area precipitation chemistry.

The precipitation concentration and wet deposition fields for the 1978/79 and 1979/80 periods showed the greatest impact of smelting activities to be for Cu and Ni (i.e., there were clearly elevated long-term concentrations in the vicinity of Copper Cliff, when the INCO smelter was operating). A second group of trace metals (Pb and Cd) showed only a minor smelter impact. Results for the other substances examined (H^+, $SO_4^=$, NO_3^-, NH_4^+, Fe, Al, Mg^{++}, Ca^{++}, K^+, Na^+, and Cl^-) apparently were largely governed

by non-smelter-related phenomena, such as long range transport into the study area, and contributions of local windblown dust or vehicular traffic.

3.1.3. *Daily Network*

In June 1978, a second deposition monitoring network was also installed, which measured daily bulk deposition on days of precipitation. This daily network was established to assess the impact of the INCO sources within about 50 km of Sudbury. The results for Falconbridge were based on relatively few data, and will not be discussed further here. The two year daily network data were stratified according to the type of frontal passage, and whether or not the sampler was downwind of the smelter during each storm. Based on the analysis (Chan *et al.*, 1982d, 1984a) of the stratified data, the following conclusions were made:

Background precipitation concentrations (as observed in the Sudbury area) associated with cold fronts were usually lower compared to those accompanying warm frontal passages. Concentrations of species such as H^+, $SO_4^=$, NO_3^-, and trace metals which are found in sub-micron particles and have a contribution due to long range transport, were in general higher with back-trajectories from the S and SW sectors, where the major emission areas in Canada and the United States are located. Note that warm frontal storms are generally associated with southerly back-trajectories at this location.

Table III summarizes the results obtained from the daily network. With the exception of Cu and Ni, most of the wet deposition in the Sudbury area (about 80% or more) can

TABLE III

Average wet deposition of INCO emissions within a 40 km radius of the smelter per precipitation event

Parameter	Emitted (kg)	Deposited (kg)	Emissions removed (%)	INCO contribution to total wet deposition (%)
H^+	4.5×10^2	1.4×10^2	32	6
S	5.2×10^5	3.4×10^3	0.7	8
Fe	7.1×10^2	4.8×10^2	68	20
Cu	2.6×10^2	2.7×10^2	100	69
Ni	3.5×10^2	1.3×10^2	36	67
Pb	2.1×10^2	0.7×10^2	33	14
Zn	0.55×10^2	0.5×10^2	93	12
Al	1.8×10^2	1.4×10^2	80	8
Cd	0.16×10^2	0.04×10^2	23	28
Cr	0.52×10^2	0.01×10^2	2.3	8

be attributed to sources other than the INCO smelter. Long range transport from sources to the south is implicated for many parameters, notably acidity and the acid-precursors, sulfate and nitrate. Nevertheless, careful meteorological analysis of the data showed a definite influence of the smelters on the local downwind precipitation quality can be detected for almost all of the substances known to be emitted. For

example, average downwind precipitation Cu and Ni concentrations were found to be an order of magnitude higher than upwind values in INCO's case. In many cases, this smelter influence can be detected to a distance of more than 40 km from the source.

It was found that most particulate constituents (acids, sulfates, trace metals) are scavenged quite efficiently from the smelter emissions during both rain and snow storms, and almost total removal occurs within the Sudbury area for some trace metals (e.g. Cu). The percentage of the total emitted S thus removed is much lower, mainly because this S is largely in the form of SO_2, which is subject to a low precipitation scavenging efficiency.

3.2. Dry deposition monitoring network

In order to assess the ambient concentration and dry deposition rate of various pollutants, an ambient particulate matter (APM) monitoring network was set up using high volume (HiVol) samplers, and occasionally Andersen impactors (see Figure 4). This air sampling program operated from July 1978 to May 1980 (Chan *et al.*, 1982e, 1984a). Ambient SO_2 concentration data were obtained from the routine monitoring network operated in the area by the Ministry's regional office.

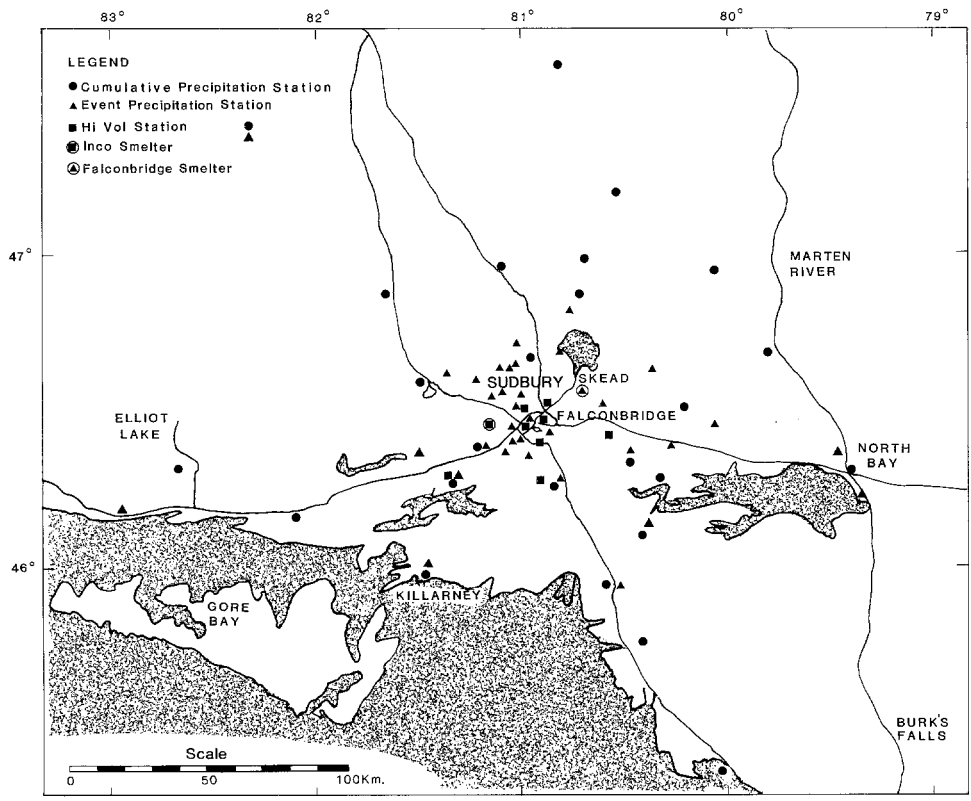

Fig. 4. Locations of Monitoring Sites around Sudbury.

Typical geometric mean air concentrations ($\mu g\, m^{-3}$) over the 2 yr of network operation were found to be: SO_2 = 12.7, $SO_4^=$ = 2.6, $N\text{-}NO_3^-$ = 0.16, $N\text{-}NH_4^+$ = 0.54, Fe = 0.29, Ni = 0.005, Pb = 0.04, Zn = 0.02, Cd = 0.001, and Al = 0.10.

Direct measurement of atmospheric dry deposition is extremely difficult, and no existing method is suitable for routine field use. The approach taken in this study was to determine the ambient concentration of the particular substance of interest (be it in the gaseous or particulate form), and then to estimate the deposition flux from the product of concentration and a 'deposition velocity' (the magnitude of which can be estimated, although for some substances with large uncertainty; Hicks *et al.*, 1980).

Ambient concentration data for SO_2 and a number of particulate consituents (including sulfates, nitrates and trace metals), together with meteorological data, were used to distinguish HiVol samples impacted by smelter emissions from those exposed to background air. From these data, dry deposition rates for various substances were inferred, both within the plume sector and in 'background' areas. Particle sizing measurements (Chan *et al.*, 1982a, b, 1983) were used to estimate deposition velocities for the particulate constituents. Long term average values used in the present calculations were within the following range (in $cm\, s^{-1}$): SO_2, 0.3; $SO_4^=$, 0.09–0.14; Fe, 1.1–1.8; Ni, 1.4–1.8; Pb, 0.23–0.34; Zn, 0.25–0.38; Al, 0.87–1.2; and Cd, 0.25–0.49. The SO_2 deposition velocity was calculated based on the method of Sheih *et al.* (1979) which attempts to account for factors such as land use, biological status of the vegetation and atmospheric stability. Particulate deposition velocities were estimated from McMahon and Dennison (1979). More details are given in Chan *et al.* (1982e, 1984a).

Table IV summarizes the results of the present investigation. These estimates showed dry deposition to be a relatively inefficient removal mechanism for SO_2, and substances predominantly in sub-micron particles (i.e., $SO_4^=$, Pb, Zn, Cd), less than 2% of the emissions being deposited within 40 km of the smelter sources. On the other hand, a

TABLE IV

Dry deposition of INCO emissions (r = 40 km)

Parameter	Daily emissions (kg)	Estimated plume dry deposition (kg)	Deposited (%)	Background deposition (kg)	Total[a] (%)
SO_2	2333548	3975	0.17	10427	28
$SO_4^=$	49067	58.49	0.12	2074.07	2.7
S	1183130	2006.84	0.17	5904.86	25
Fe	1626	247.88	15.2	1090.12	19
Ni	798	20.72	2.6	11.68	64
Pb	480	2.57	0.53	33.68	7.1
Zn	126	2.22	1.67	26.04	7.9
Cd	37	0.01	0.03	1.07	0.9
Al	406	36.28	8.9	484.05	7.0

[a] INCO deposition/(INCO deposition + background deposition).

higher proportion of the coarse particles (containing most of the airborne Fe, Al and Ni) – up to about 15% or more (long term average) – can be deposited within the same area.

During the study period, the smelter contribution to the total dry deposition, within 40 km of the sources was greatest for S (primarily due to SO_2) and Ni, making up 25% and 64% of the total, respectively. The smelter contribution of the other metals examined (Fe, Pb, Zn, Cd, and Al) was generally less than 20% of the total. Due to sampling problems, the corresponding figures could not be derived for Cu, but results simular to those for Ni are expected, based on the available emission rate and particle size information.

3.3. Relative importance of wet and dry deposition and contribution of INCO emissions to total deposition

Data reported in Tables III and IV were combined to obtain information regarding (i) the relative importance of smelter wet and dry deposition in the Sudbury area defined by a radius of 40 km, and (ii) the contribution of INCO emissions to the total deposition. In this calculation, two assumptions were made: (i) precipitation occurs on every third day and (ii) Cu dry deposition is similar to that of Ni. The results are summarized in Table V. No similar comparison for the Falconbridge source was made

TABLE V

A comparison of wet and dry deposition and relative contribution due to INCO and background[a]

Parameter	$\frac{(\text{Wet})_{\text{INCO}}}{(\text{Dry})_{\text{INCO}}}$	$\frac{(\text{Wet + Dry})_{\text{smelter}}}{(\text{Wet + Dry})_{\text{total}}}$
S	0.39	0.18
Fe	0.45	0.23
Cu	3.0[b]	0.7[b]
Ni	1.39	0.69
Zn	5.26	0.12
Pb	6.24	0.13
Al	0.90	0.09
Cd	6.17	0.23

[a] Assuming precipitation occurs, on average, every third day. Results are average values for an area within 40 km of the source.

[b] Assuming similar dry deposition to that for Ni.

because of uncertainties associated with the wet deposition results. Note that whereas for S and trace metals in large particles (Fe, Ni, Al) wet and dry deposition is similar, particles in the sub-micron size range (Zn, Pb, Cd) are primarily deposited by precipitation.

As far as total deposition is concerned, the major smelter impact is seen to be from Cu and Ni. For most of the other substances examined, INCO contributes about 25% or less of the total atmospheric deposition. These results agree qualitatively with the work of Freedman and Hutchinson (1980a) who made total dustfall-rainfall collections in the 1976 and 1977 growing seasons in the area.

4. Supporting Studies

4.1. Local meteorology and dispersion studies

Quantitative determination of the fate of Sudbury emittants requires a detailed knowledge of the meteorological characteristics of the area. From September 1976 to September 1979 a program was carried out to measure the wind speed, wind direction and temperature in the vertical using a pilot-balloon minisonde system. The balloon releases were carried out twice daily (once in the early morning and once in the early afternoon) under fair weather conditions. The balloons were tracked using dual theodolites (Vet *et al.*, 1982).

Priority was given to characterizing the meteorology of the Sudbury area during summer months (May to September) because most of the plume impingement episodes occur then, resulting in relatively high ground level concentrations and dry deposition rates. On warm, sunny days, the 381 m stack plume begins looping (due to large scale convective turbulence) typically between 1000 and 1100 hr and stops roughly 1 to 2 hr before sunset. The initial impingement of individual plume loops usually occurs at a distance of 3 to 10 km from the stack. During such looping conditions, SO_2 concentrations within the mixing layer, and on the ground at various distances from the 381 m stack, have been measured by instrumented aircraft and ground vehicles during two intensive field programs conducted in June 1978 and August 1979 (Vet and Chan, 1982). Ground-level concentrations typically in the range of 100 to 300 ppb SO_2 (half-hourly average) were observed.

The meteorological program yielded a great deal of data, which were useful for interpreting the behavior of the 381 m stack plume, and for mathematical model development. The results indicate that the effectiveness of a tall stack for eliminating ground level concentrations is inadequate during convective periods (because of plume looping) which frequently occur during the summer. This is not the case however, during the night and during winter months, when inversions are generally below the plume level and tend to keep the plume aloft thus preventing it from impinging at the ground level. On the other hand, at these times low level smelter emissions are trapped in stable air near the ground and can result in relatively high smelter produced ground-level concentrations.

4.2. Plume oxidation rate studies

A number of investigations have been carried out into the oxidation rate of SO_2 to $SO_4^=$ in the INCO Superstack plume (Lusis and Wiebe, 1976; Chan *et al.* 1979, 1980).

These studies, carried out at different times of the year, indicated that: 1) the SO_2 oxidation rate is low, typically 1% hr^{-1} or less; 2) as far as 100 km from the source, 10% or less of the S is in the form of $SO_4^=$; and 3) the particulate S is predominantly in the form of H_2SO_4.

It is interesting to note that the oxidation rate under 'wet' conditions (the above conclusions were based on aircraft studies when there was no precipitation) has been estimated using data from the daily precipitation chemistry network (Chan *et al.*, 1982d), and found to be comparable to the 'dry' value above.

4.3 Plume washout study

In 1980 and 1981, a series of intensive field experiments was carried out, involving the use of remote sensors to locate the 381 m stack plume, and direct sampling of precipitation underneath the plume at different downwind distances (Millan *et al.*, 1982; Lusis *et al.*, 1983). These studies clearly showed the impact on precipitation quality of a large point source of S and trace metal emissions, and the results were used to obtain scavenging coefficients for use in the mathematical models. For sulfates, the scavenging coefficient was found to be about $10^{-4}\,s^{-1}$ and for trace metals, in the (1 to 2) $\times\ 10^{-4}\,s^{-1}$ range. These results are in qualitative agreement with the earlier work of Wiebe and Whelpdale (1977) who estimated, from intensive under-plume sampling, that less than 1% of the emitted S was scavenged within 50 km of INCO, whereas the fraction of trace metals was much higher.

5. Mathematical Modelling

As part of the Sudbury Environmental Study, mathematical models were developed to account for long and short range, as well as long and short term, air quality and atmospheric deposition.

5.1. Long range, long term model

A simple statistical model was developed for estimating long term concentrations of S pollutants associated with long range transport (Venkatram *et al.*, 1981, 1982). The model consists of three components; transport and dispersion, scavenging and chemistry. Dispersion and wet scavenging are treated statistically. Dry deposition is treated by assuming loss at the ground through a dry deposition velocity. Sulfur dioxide oxidation is described by a first-order process. The model was used to estimate wet deposition of S over a grid covering northeastern United States and Canada. The model predictions compared well to corresponding measurements of annual deposition, and the model has been applied to estimate the relative contribution of INCO and Falconbridge to the total S deposition in Ontario and Quebec (Venkatram *et al.*, 1981, 1982).

The model predicted that, for typical mid-1970's SO_2 emission levels, Sudbury sources contributed up to 30% of the total S deposited during precipitation in most of central Ontario, with INCO's emissions dominating, but Falconbridge also contributing a significant amount (up to 5% of the total). The results from this model are not directly

comparable to the measurements made within the Sudbury Basin (Table III), but they do suggest a comparable impact of the smelters within the Basin itself and farther afield, such as in the ecologically sensitive Muskoka–Haliburton area of central Ontario.

5.2. Short Range, Short Term Fumigation Model

Conventional 'Gaussian type' dispersion models cannot predict ground level concentrations (glc) of pollutants associated with plumes emitted into the convective boundary layer. Most high glc's at Sudbury are associated with convective conditions. A model was therefore developed for this purpose (Venkatram, 1982). The required data inputs to the model are the climatology of the mixed layer height, solar radiation, mixed layer winds and the temperature structure above the mixed layer. The model is only applicable to daytime convective conditions. It was evaluated with ground level concentrations obtained in the dispersion field studies described earlier, as well as data sets obtained elsewhere. In about 80% of the cases, model predictions were found to be within a factor-of-two of the observations.

5.3. Short Range, Long Term Model

The short range, long term model uses long term meteorological data in the simulation of transport and dispersion of pollutants (Ellenton, 1984). Ambient ground level concentration, as an annual or seasonal average for locations within a distance of about 100 km from the source of emission, as well as annual wet and dry deposition, can be calculated. This model is still under development, and details will not be given here except to mention that a preliminary comparison of the calculated seasonal SO_2 concentration with the observed values has given encouraging results. Furthermore, it is interesting to note that the model predicts that the Sudbury sources' contribution to S dry deposition is highly direction-oriented (due to local prevailing wind patterns) and quite significant near the smelters, making up as much as 70% of the total (note that the estimates in Table IV, based on air measurements, are regional average estimates).

6. Summary

As part of the Sudbury Environmental Study, a concerted effort has been made to characterize the atmospheric emissions of the Sudbury smelters, and determine their atmospheric fate.

Emission rates of the major pollutants (SO_2, H_2SO_4, and particulate Fe, Cu, Ni, Pb, and As), as well as a number of other substances have been quantified. The subsequent atmospheric dispersion, transformation and deposition of these pollutants has been studied in a number of intensive field campaigns, as well as by longer-term precipitation and air monitoring programs, with emphasis on the largest single source in the Sudbury area, viz, the 381 m INCO Superstack. These studies, and concurrent mathematical modelling of air quality and atmospheric deposition both in the Sudbury Basin and farther afield, have yielded the following major findings:

- The contribution of the smelter emissions to the total wet deposition in the Sudbury

Basin was generally small, less than 20% for S and a number of other substances with the exception of Cu and Ni, where the contribution was about 70%. The above figures refer to smelter emission rates representative of the late 1970's. For S, mathematical modelling suggested that the smelter contribution could be of a similar magnitude farther afield as well, such as in the ecologically sensitive Muskoka–Haliburton area of central Ontario.

– The results were generally similar with regard to dry deposition, which was estimated from ambient concentrations and particle size distribution information, using a deposition velocity. For S, the INCO smelter was found to contribute a large proportion of the total atmospheric dry deposition. Mathematical modelling predicted the S dry deposition to be very direction-oriented, and quite high near the smelters (making up as much as 70% of the total).

– On warm, sunny days during the summer, the plume from the largest source in the area, the 381 m Superstack, begins looping in the late morning due to large-scale convective activity leading to ground impingement at a distance of about 3 to 10 km, and resulting in relatively high local ground level concentrations thus limiting the effectiveness of the tall stack. This condition has been successfully simulated with a short range short term fumigation model. On the other hand, during the winter months, and at night, when the atmosphere is stable below plume level, the plume from this source is kept aloft and can travel for large distances in a coherent form. An extensive study of the local meteorology has been used to determine the frequency of occurrence of various atmospheric dispersion conditions.

– The chemical transformation rate of SO_2 to sulfates in the smelter plume is relatively low, typically less than 1% hr^{-1}. Although the above figure comes from experiments carried out largely under fair-weather conditions, estimates based on precipitation chemistry measurements suggest that it may also be representative of 'wet' conditions.

– Precipitation scavenging of the smelter emissions is very efficient (with the exception of SO_2). Typical scavenging coefficients for sulfates were found to be about 30 to 40% hr^{-1}, and for trace metals in the 60 to 80% hr^{-1} range.

– Nevertheless, on a long-term basis, a relatively small portion of most of the total smelter emissions is deposited in the Sudbury Basin by wet and dry processes. Thus, just as pollutants imported into the area due to long-range transport are a significant contributor to atmospheric deposition in the Sudbury Basin as a whole, most of the local emissions leave the Basin and in turn contribute to ecological impacts farther afield.

Acknowledgments

This account is based on results published in many reports. Appreciation is extended to all those authors and scientists whose devoted efforts have made this possible. Special thanks are due to Messrs R. J. Vet and E. W. Piche for their support throughout the Sudbury Environmental Study, and Dr B. M. McCormac and Mr. R. Potvin for their critical review and comments of this article.

References

Beamish, R. J.: 1974, *Water Res.* **8**, 85.

Beamish, R. J., Lockhart, W. L., Van Loon, J. C., and Harvey, H. H.: 1975a, *Ambio* **4**, 98.

Beamish, R. J. McFarlane, G. A., Van Loon, J. C., and Lichwa, J.: 1975b, 'An Examination of the Possible Effects of Sudbury Nickel Mining and Smelting Operations on Fishes and Water Chemistry of Lakes within the Whitefish Lake Indian Reserve', Fish, Mar. Serv. Res. Dev. Tech. Rep. 579, 52 p.

Beamish, R. J. and Van Loon, J. C.: 1977, *J. Fish. Res. Board Can.* **34**, 649.

Chan, W. H.: 1982, 'Sudbury Environmental Study Atmospheric Research Program: A Synopsis', Report ARB-27-82-ARSP, Air Resources Branch Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., and Lusis, M. A.: 1979, 'Airborne Investigations of the INCO and Falconbridge Stack Plumes, 1976-1977', Report ARB-TDA-60-79, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Lusis, M. A., Hunt, J. E., and Stevens, R. D. S.: 1980, *Atmospheric Environ.* **14**, 1159.

Chan, W. H., Vet, R. J., Skelton, G. B., and Lusis, M. A.: 1982a, 'Size Distribution and Emission Rate Measurements of Particulates in the 93 m Falconbridge Smelter Stack Plume, 1979', Report ARB-TDA-57-80, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Lusis, M. A., and Skelton, G. B.: 1982b 'Size Distribution and Emission Rate Measurements of Particulates in the INCO 381 m Chimney and Iron Ore Recovery Plant Plumes, 1979-80', Report ARB-TDA-62-80, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Ro, C. U., Tang, A. J. S., and Lusis, M. A.: 1982c, 'Precipitation Quality and Wet Deposition in the Sudbury Basin: Sudbury Environmental Study Cumulative Precipitation Network Results', Report ARB-04-82-ARSP, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Ro, C. U., Tang, A. J. S., and Lusis, M. A.: 1982d, 'An Analysis of the Impact of Smelter Emissions on Precipitation Quality and Wet Deposition in the Sudbury Area: Sudbury Environmental Study Event Precipitation Network Results', Report ARB-05-82-ARSP, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Ro, C. U., Tang, A. J. S., and Lusis, M. A.: 1982e 'An Analysis of the Impact of Smelter Emissions on Atmospheric Dry Deposition in the Sudbury Area: Sudbury Environmental Study Event Airborne Particulate Matter Network Results', Report ARB-12-81-ARSP, Air Resources Branch, Ontario Ministry of the Environment.

Chan, W. H., Vet, R. J., Lusis, M. A., and Skelton, G. B.: 1983., *Atmospheric Environ.* **17**, 1173.

Chan, W. H., Vet, R. J., Ro, C. U., Tang, A. J. S., and Lusis, M. A.: 1984a, *Atmospheric Environ.* **18**, 1001.

Chan, W. H., Vet, R. J., Ro, C. U., Tang, A. J. S., and Lusis. M. A.: 1984b, *Atmospheric Environ.* **18**, 1175.

Ellenton, G.: 1984, unpublished results.

Freedman, W. and Hutchinson, T. C.: 1978, 'Smelter Pollution Near Sudbury, Ontario, Canada and Effects on Forest Litter Decomposition', presented at the NATO Advanced Research Institute: Effects of Acid Precipitation on Terrestrial Ecosystems, Toronto, May 22-26.

Freedman, W. and Hutchinson, T. C.: 1980a, *Can J. Botany* **58**, 108.

Freedman, W. and Hutchinson, T. C.: 1980b, *Can J. Botany* **58**, 1722.

Gorham, E. and Gordon, A. G.: 1960a, *Can. J. Botany* **38**, 477.

Gorham, E. and Gordon, A. G.: 1960b, *Can. J. Botany* **38**, 307.

Hicks, B. B., Wesely, M. L., and Durham, J. L.: 1980, 'Critique of Methods to Measure Dry Deposition. Workshop Summary', Environmental Sciences Research Report, Research Triangle Park, North Carolina.

Hutchinson, T. C. and Whitby, L. M.: 1974a, *Environmental Conservation* **1**, 123.

Hutchinson, T. C. and Whitby, L. M.: 1974b, 'A Study of Airborne Contamination of Vegetation and Soil by Heavy Metals from the Sudbury Ontario Copper-Nickel Smelters', Symposium Proceeding, pp. 175-178, Trace Substances in Environmental Health - VII, D. D. Hemphill Ed.

Hutchinson, T. C. and Whitby, L. M.: 1977, *Water, Air, and Soil Pollution* **7**, 421.

Jeffries, D. S., and Snyder, W. R.: 1981, *Water, Air, and Soil Pollution* **15**, 127.

Kramer, J. R.: 1973, 'Fate of Atmospheric Sulphur Dioxide and Related Substances as Indicated by Chemistry of Precipitation', McMaster University, Department of Geology Report.

Kramer, J. R.: 1975, 'Fate of Atmospheric Sulphur Dioxide and Related Substances as Indicated by Chemistry of Precipitation', McMaster University, Department of Geology Report.

Kramer, J. R.: 1976, 'Fate of Atmospheric Sulphur Dioxide and Related Substances as Indicated by Chemistry of Precipitation', McMaster University, Department of Geology Report.

Kramer, J. R. and Snyder, W. R.: 1977, 'Precipitation Scavenging of Sulfur Isotopes', pp. 129–136, Proceedings, Precipitation Scavenging Symposium, Champaign, Illinois, 14–18 October, 1974.

Lusis, M. A. and Wiebe, H. A.: 1976, *Atmospheric Environ.* **10**, 793.

Lusis, M. A., Chan, W. H., Tang, A. J. S. and Johnson, N. D.: 1983, 'Scavenging Rates of Sulfur and Trace Metals from a Smelter Plume', Proc. 4th Int. Conf. on Precipitation Scavenging, Dry Deposition and Resuspension, Santa Monica, November 29 – December 3, 1982, (Edited by Pruppacher, H., Semonin, R. G. and Slinn, W. G. N.), Elsevier Publishing Co. Inc., Amsterdam.

McMahon, D. J. and Dennison, P. J., 1979, *Atmospheric Environ.* **13**, 571.

Millan, M. M., Barton, S. C., Johnson, N. D., Weisman, B., Lusis, M., Chan, W., and Vet, R.: 1982, *Atmospheric Environ.* **16**, 2709.

Muller, E. F. and Kramer, J. R.: 1977, 'Precipitation Scavenging in Central and Northern Ontario', pp. 590–601, Proceedings, Precipitation Scavenging Symposium, Champaign, Illinois, 14–18 October, 1974.

Ontario Ministry of the Environment: 1982a, 'Sudbury Environmental Study: Synopsis 1973–1980', Acidic Precipitation in Ontario Study Co-Ordination Office.

Ontario Ministry of Environment: 1982b, 'Studies of Lakes and Watersheds near Sudbury Ontario: Final Limnological Report', Water Resources Branch, Report 009/82 SES.
Resources Branch, Report 009/82 SES.

Ozvacic, V.: 1982, 'Emissions of Sulfur Oxides, Particulates and Trace Elements in Sudbury Basin', Report ARB-ERTD-08-82. Air Resources Branch, Ontario Ministry of the Environment.

Scheider, W. A., Jeffries, D. S., and Dillon, P. J.: 1980, *Atmospheric Environ.* **15**, 945.

Sheih, C. M., Wesely, M. L., and Hicks, B. B.: 1979, 'A Guide for Estimating Dry Deposition Velocities of Sulfur over the Eastern United States and Surrounding Regions', Argonne National Laboratory, Report ANL, RER 79-2.

Vet, R. J. and Chan, W. H.: 1982, 'Two Intensive Field Studies of Dispersion of the INCO 381 m Stack Plume in the Convective Boundary Layer', Report ARB-003-81-ARSP, Air Resources Branch, Ontario Ministry of the Environment.

Vet, R. J., Linauskas, S., Bardswick, W., and Chan, W. H.: 1982, 'Sudbury Environmental Study Meteorology Program', Report ARB-09-81-ARSP, Air Resources Branch, Ontario Ministry of the Environment.

Venkatram, A.: 1982, 'Short Range Short-Term Fumigation Model for the INCO Superstack', Report AQM-AM8 1. Air Resources Branch, Ontario Ministry of the Environment.

Venkatram, A., Ley, B. E., and Wong, S. Y.: 1981, *Atmospheric Environ.* **16**, 249.

Venkatram A., Ley, B. E., and Wong, S. Y.: 1982, 'A Statistical Model to Estimate Long-Term Concentrations of Pollutants Associated with Long Range Transport and its Application to Emissions from the Sudbury Region', Report ARB-36-81-SES, Air Resources Branch, Ontario Ministry of the Environment.

Wiebe, H. A. and Whelpdale, D. M.: 1977, 'Precipitation Scavenging from a Tall-Stack Plume', pp. 118–126, Proceedings, Precipitation Scavenging Symposium, Champaign, Illinois, 14–18 October, 1974.

Whitby, L. M. and Hutchinson, T. C.: 1974, *Environmental Conservation* **1**, 181.