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ACID PRECIPITATION - ITS INFORMATION
DEPOSITION AND EFFECTS ON
LAKE WATER CHEMISTRY

A REVIEW OF THE LITERATURE

JANUARY, 1978

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DEPOSITION AND EFFECTS ON
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A REVIEW OF THE LITERATURE

MINNESOTA ENVIRONMENTAL QUALITY BOARD
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ABSTRACT

Current literature on acid precipitation was reviewed to determine sources, deposition mechanisms and effects on lake water chemistry. Values of precipitation and lake pH, SO_4 content of precipitation, ambient SO_2 and SO_4 , and SO_4 loading were obtained in order to compare northeastern Minnesota with other areas of the world.

Precipitation has become increasingly acidic in regions of North America and Europe during the last two decades. The most highly impacted areas include Norway, the northeastern U.S., and the vicinity of Sudbury, Ontario, where precipitation with pH of four is not unusual. Remote areas of the world have been found to have precipitation pH values near 5.7, the pH of water in equilibrium with carbon dioxide in the atmosphere. Northeastern Minnesota precipitation has an average pH of near five.

Decreasing precipitation pH has occurred simultaneously with increases in sulfur and nitrogen oxide emissions from anthropogenic sources. Sulfur oxides are considered to be the major precursors of acid precipitation. Long distance transport of these pollutants from industrial areas causes precipitation to be acidic in areas with no local polluters.

Lake water pH has been reduced because of acid precipitation. Especially vulnerable to acidification are lakes which are poorly buffered due to their geologic environments. Northeastern Minnesota lakes have an average pH of seven, while many lakes downwind of industrial pollution sources have pH values of five or less, and have experienced loss of aquatic plant and animal populations. Increased solubility of metals, which occurs at low pH levels, causes additional stress to aquatic biota.

INTRODUCTION TO THE REGIONAL COPPER-NICKEL STUDY

The Regional Copper-Nickel Environmental Impact Study is a comprehensive examination of the potential cumulative environmental, social, and economic impacts of copper-nickel mineral development in northeastern Minnesota. This study is being conducted for the Minnesota Legislature and state Executive Branch agencies, under the direction of the Minnesota Environmental Quality Board (MEQB) and with the funding, review, and concurrence of the Legislative Commission on Minnesota Resources.

A region along the surface contact of the Duluth Complex in St. Louis and Lake counties in northeastern Minnesota contains a major domestic resource of copper-nickel sulfide mineralization. This region has been explored by several mineral resource development companies for more than twenty years, and recently two firms, AMAX and International Nickel Company, have considered commercial operations. These exploration and mine planning activities indicate the potential establishment of a new mining and processing industry in Minnesota. In addition, these activities indicate the need for a comprehensive environmental, social, and economic analysis by the state in order to consider the cumulative regional implications of this new industry and to provide adequate information for future state policy review and development. In January, 1976, the MEQB organized and initiated the Regional Copper-Nickel Study.

The major objectives of the Regional Copper-Nickel Study are: 1) to characterize the region in its pre-copper-nickel development state; 2) to identify and describe the probable technologies which may be used to exploit the mineral resource and to convert it into salable commodities; 3) to identify and assess the impacts of primary copper-nickel development and secondary regional growth; 4) to conceptualize alternative degrees of regional copper-nickel development; and 5) to assess the cumulative environmental, social, and economic impacts of such hypothetical developments. The Regional Study is a scientific information gathering and analysis effort and will not present subjective social judgements on whether, where, when, or how copper-nickel development should or should not proceed. In addition, the Study will not make or propose state policy pertaining to copper-nickel development.

The Minnesota Environmental Quality Board is a state agency responsible for the implementation of the Minnesota Environmental Policy Act and promotes cooperation between state agencies on environmental matters. The Regional Copper-Nickel Study is an ad hoc effort of the MEQB and future regulatory and site specific environmental impact studies will most likely be the responsibility of the Minnesota Department of Natural Resources and the Minnesota Pollution Control Agency.

INTRODUCTION

Rain or snow with a pH of less than 5.6 is considered acidic (Likens 1976), since water in equilibrium with carbon dioxide in the atmosphere produces a precipitation pH of about 5.7 (Reuss 1975, Kramer 1976a). Precipitation pH values of close to 5.7 have been recorded in remote areas of the world, long distances from sources of atmospheric pollution (Kramer 1976a)(see Figure 2). Many other regions receive acid precipitation, sometimes with pH levels of three or lower (Likens and Bormann 1974, Likens and Bormann 1975, Cogbill 1975, Likens et al. 1975, Hutchinson and Whitby 1974). Zones of highly acidic precipitation are expanding (Hysing-Dahl et al. 1976, Likens 1976) and precipitation pH is decreasing in many areas (Likens 1976).

Acid precipitation is of concern because of its effects on aquatic and terrestrial ecosystems. Acidification of lakes and streams due to acid precipitation has eliminated or severely reduced populations of aquatic plants and animals (Leivestad et al. 1976, Beamish and Harvey 1972, Schofield 1976, Hendrey et al. 1976, Bolin 1971). Acid rain causes changes in the rate of leaching of elements from soils, which may affect vegetation growth (Abrahamsen et al. 1976, Malmer 1976, Likens and Bormann 1974) and change the composition of runoff that reaches water bodies. Vegetation may also be injured by direct contact with acid precipitation (Abrahamsen et al. 1976). Other potential problems due to acid precipitation include effects on predator-prey relationships, effects on the metabolism of organisms, deterioration of buildings, and effects on human health (Likens and Bormann 1974, Oden 1975, Bolin 1971).

The increased acidity of precipitation during the past 20 years in northern Europe and the northeastern United States correlates with increased anthropogenic emissions of sulfur dioxide and nitrogen oxides (Likens 1976). These two pollutants are the major precursors of acid precipitation (Likens 1976, Dovland et al. 1976, Galloway et al. 1976a, Summers and Whelpdale 1975). A high correlation has been found between acid precipitation episodes and storm tracks that have passed over areas of high density anthropogenic emissions (Junge 1963, Cogbill and Likens 1974, Dovland et al. 1976, Bolin 1971). Concentrations of other pollutants, such as heavy metals and pesticides, have also increased in the eastern U.S., Ontario, and Scandinavia and show the same distribution as acid precipitation (Lodge et al. 1968, Hagen and Langeland 1973, Almer et al. 1974, Conroy et al. 1975). In recent years taller stacks have been constructed in an effort to reduce local pollution, and the areal distribution of acid precipitation has been observed to increase concomitantly. Thus, human activities are believed to be the cause of increasing acidic precipitation.

This report is a review of the current literature concerning acid precipitation, its formation, and its effects on lake water quality. Of particular interest was literature dealing with acid precipitation in areas of the world which have geologic and climatic characteristics similar to those of northeastern Minnesota. Copper-nickel deposits have been discovered in northeastern Minnesota. Smelting of similar ores in Sudbury, Ontario has resulted in acidification of precipitation due to SO_2 emissions, followed by acidification of lakes, elimination of aquatic species, and damage to terrestrial vegetation (Conroy et al. 1975, Gorham and Gordon 1960).

PRECURSORS OF ACID PRECIPITATION

The presence of acids in the atmosphere can cause precipitation to be acidic. Strong acids have been found to be the most important contributors to acid precipitation (Likens 1976, Dovland et al. 1976, Galloway et al. 1976a, Summers and Whelpdale 1975, Gorham 1975, Krupa et al. 1975), although weak acids may also contribute (Galloway et al. 1975). The precursors of acid precipitation are chloride, which forms hydrochloric acid; sulfur dioxide, which is converted to sulfate and then to sulfuric acid; and nitrogen oxides, which form nitric acid (Likens 1976, Dovland et al. 1976, Galloway et al. 1976a). These compounds are released to the atmosphere by various natural and human activities.

Chloride is released to the atmosphere by anthropogenic emissions, and naturally from sea spray and volcanic eruptions. Gorham (1958) observed substantial amounts of chloride in smoke solids from coal burning. Hydrochloric acid also has been detected in precipitation in British urban areas in amounts too great to be accounted for by natural sources (Gorham 1958); coal combustion is the likely source, according to Gorham (1975). Coal-burning Tennessee Valley Authority plants have been found to emit hydrochloric acid in flue gases. Hydrochloric acid is not deemed as important as sulfuric acid and nitric acid as a component of acid precipitation; however, it has been detected in precipitation of the northeastern U.S. (Likens 1976) as well as the Minneapolis-St. Paul area (Krupa et al. 1975).

Weak and Bronsted acids contribute only slightly to the H^+ ion concentration in northeastern U.S. precipitation (Galloway et al. 1975). In an aqueous solution, total acidity is made up of both free and bound protons. Free

protons constitute the measurable pH, while bound protons have no influence on pH and can be determined only by titration (Galloway et al. 1976a). The dissociation of a weak acid such as H_2CO_3 is dependent upon pH. At pH 5 or greater, H_2CO_3 contributes to both free and bound acidity. At pH less than 5, however, it contributes only to bound acidity, and does not affect pH. Bronsted acids are likewise not H^+ ion sources below pH 5 (Galloway et al. 1976a). As Figure 2 indicates, precipitation pH values of less than 5 are not uncommon. Galloway et al. (1975) conclude that although acid precipitation is a mixture of strong and weak acids, weak acids contribute primarily to the total acidity and only negligibly to free acidity.

Sulfuric acid is the predominant strong acid which causes acid precipitation (Gorham 1975). Sulfur compounds are introduced to the atmosphere by three main processes: H_2S from biological decay, SO_4 from sea salt, and SO_2 from anthropogenic sources, such as sulfide ore smelters and industries which burn fossil fuels (Dovland et al. 1976). Electric generating plant emissions account for about 60 percent of the sulfur emitted by human activities in the U.S. (U.S.EPA 1976b). Global sulfur budgets reported in the literature vary greatly, and Likens (1976) is of the opinion that there is not enough data at present to evaluate their reliability. The literature reviewed by Gorham (1975) indicates that sulfur mobilization from natural sources is 133 to 152 $\times 10^6$ metric tons per year compared to 50 to 55 $\times 10^6$ metric tons per year from anthropogenic emissions. Although natural emissions are estimated at over twice anthropogenic emissions, natural emissions are not considered a major factor in producing acid rain, because they are assumed to be in balance with natural sources of neutralizing bases (Gorham 1975). Because SO_2 and SO_4 in the atmosphere are of great

concern as precursors of acid precipitation, their emission sources, ambient concentrations, and concentration trends will be discussed in a later section.

Nitrogen compounds, the precursors of nitric acid, enter the atmosphere as ammonia released by biological decay processes, and as nitrogen oxides from biochemical reactions in soil and combustion processes (Dovland et al. 1976). Estimates of the quantities of nitrogen compounds released from various sources are difficult to make; however, it has been calculated that annual global emissions of nitrogen oxides from natural sources are 150×10^6 metric tons. This compares to 8.2, 6.6, and 0.6×10^6 tons per year from combustion of coal, petroleum, and natural gas, respectively (Anon. 1975).

Anthropogenic nitrogen emissions have been increasing, with a corresponding increase in nitrogen oxide content of precipitation. Likens (1972) blames increased NO_3 content of precipitation since about 1945 on increased use of natural gas and motor fuels. Improved fossil fuel combustion techniques, which use higher flame temperatures, have also caused greater nitrogen oxide emissions and precipitation content (Dovland et al. 1976). The increased use of nitrogen fertilizers since about 1950 may have contributed additionally to the NO_x content of precipitation (Dovland et al. 1976).

Although both sulfur dioxide and nitrogen oxide emissions increased sharply after 1960, nitrogen oxide emissions increased at twice the rate of SO_2 emissions between 1960 and 1970 (Likens 1976). The concentration of nitrogen oxides in precipitation has also increased compared to sulfur dioxide concentration. In the Hubbard Brook Experimental Forest in New Hampshire, the contribution of SO_4 to the acidity of precipitation in

1964-1965 was 83 percent, while in 1973-1974 it was down to 66 percent. The NO_3 contribution, on the other hand, increased from 15 to 30 percent (Likens 1976). Annual input of hydrogen ions in precipitation increased by 1.4-fold during this period, which correlates highly with the annual rate of nitrate input. Sulfate annual input did not increase significantly (Likens et al. 1975). Thus, although sulfuric acid remains the largest contributor to acid precipitation, nitric acid is apparently becoming more important in the northeastern U.S. (Likens et al. 1975) and in northern Europe (Dovland et al. 1976).

Excess sulfate (sulfate from all sources other than sea spray) concentration is considered by Dovland et al. (1976) to be a good measure of the acidifying effect of precipitation in Norway. This is because the correlation coefficient between the concentration of excess sulfate and strong acid in precipitation was 0.7 to 0.9 at most of the stations observed. Sulfate concentration is important in other areas of the world, also, where it is the cause of acid precipitation. Figure 1 and Table 1 show sulfate concentrations in precipitation. Junge and Werby (1958) reported 2.2 mg/l as a good average of excess sulfate in precipitation over land for the whole earth. This value is exceeded by most stations listed in the table.

SULFUR DIOXIDE AND SULFATE EMISSION SOURCES, AMBIENT LEVELS AND TRENDS

Sulfur oxide emissions, mostly in the form of SO_2 , have generally increased in recent years due to human activities. In Europe, sulfur emissions are reported to have continuously increased by two to five percent per year during the last fifteen years (Oden 1975). During the 1960s sulfur oxide emissions in the U.S. increased by 45 percent (Commission on Natural

Figure 1. Sulfate concentration in precipitation.

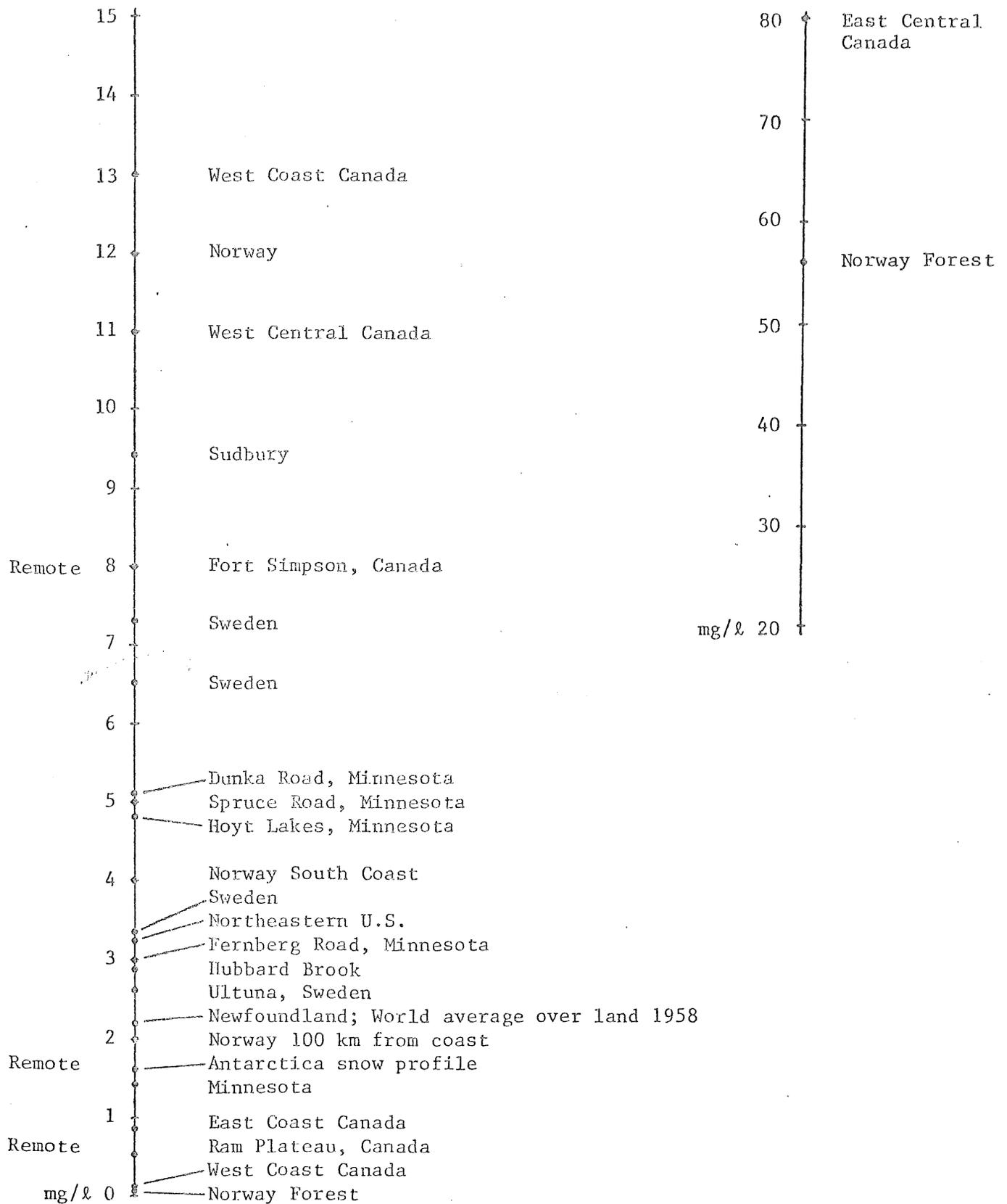


Table 1. Sulfate concentration in precipitation.

Location	SO ₄ (mg/l)	Comments	Reference
Remote background			
Ram Plateau, N.W.T., Canada	<.5	bulk sample, 1 station 6/75-8/75	Kramer 1975
Fort Simpson, N.W.T.	0.1-8	precipitation only	Whelpdale and Summers 1975
Cape Dyer, N.W.T.	0.8	snow event	
Resolute, N.W.T.	<5	rain event	Kramer 1973a
Antarctica	1.6	snow profile	Gjessing and Gjessing 1973
Newfoundland	2.2	single station, 1 year precip	Gorham 1961
Wisconsin	2.9	single station, 1 year precip	
Ojebyn, northern Sweden	2.5	single station, 1 year precip	
Ultuna, central Sweden	2.6	single station, 1 year precip	
Georgetown, British Guiana	1.3	3 year average precip	
Yonkers, N.Y., 24 km north of NYC	<1-20 4.8 mean	precipitation events 1974	Jacobson et al. 1975
Northeastern U.S.	4.3-5.2 3.19-4.96	bulk sample, 18 sites 1965-68 precipitation, 5 sites 1972-73	Pearson and Fisher 1971 Cogbill and Likens 1974
Hubbard Brook Experimental Forest, N. Hamp	2.9	bulk precipitation, weighted annual mean 1964-74	Likens et al. 1975
Canada			
West Coast	0.1-13	bulk precipitation	Whelpdale and Summers 1975
West Central	<0.1-11	bulk precipitation	
East Central	<.3-80	bulk precipitation	
East Coast	.9-3.2	bulk precipitation	
Sudbury, Ontario	<.5-9.4	precipitation events--variation over 11 days 1,2, & 3/76	Kramer 1976b
Central Alberta	2.7	152 rain samples 1967-70, mean	Summers and Hitchon 1973

Table 1. contd.

Location	SO ₄ (mg/l)	Comments	Reference
Norway	.1-12	precipitation events	Scholdager 1973
17 stations	.8-3	precipitation 1974-75	Summers and Hitchon 1973
South Coast	4	mean in precipitation 1972-73	Dovland et al. 1976
100 km from coast	2	mean in precipitation 1972-73	
Western	1-2	mean in precipitation 1972-73	
Birkenes watershed	3.24	mean of 20 precipitation events, 1974	Semb 1975
Forest	.05-56	precipitation	Bjor et al. 1974
Sweden	7-7.3		Hornstrom et al. 1973
	3.3-6.5	bulk precipitation	Andersson 1972
	1.5-2.7	bulk precipitation	Malmer 1974
Northeastern Minnesota	1.4	single station, 1 year precip.	Gorham 1961
Fernberg Road	3.0	mean, 2 bulk precip samples 3&5/77	MEQB 1977
Spruce Road	5	mean, 3 bulk precip samples 3,4&5/77	
Dunka Road	5.1	mean, 2 bulk precip samples 4&5/77	
Hoyt Lakes	4.8	mean, 2 bulk precip samples 4&5/77	
Land areas, whole earth	2.2	average based on precip. SO ₄ concentrations in the literature	Junge and Werby 1958

Resources 1975); however, from 1972 through 1975 emissions appear to have decreased slightly (U.S.EPA 1976a). Certainly in the early 1970s, and possibly during the 1960s, emissions in urban areas decreased as sources began to comply with air pollution control regulations (CNR 1975; U.S.EPA 1976b).

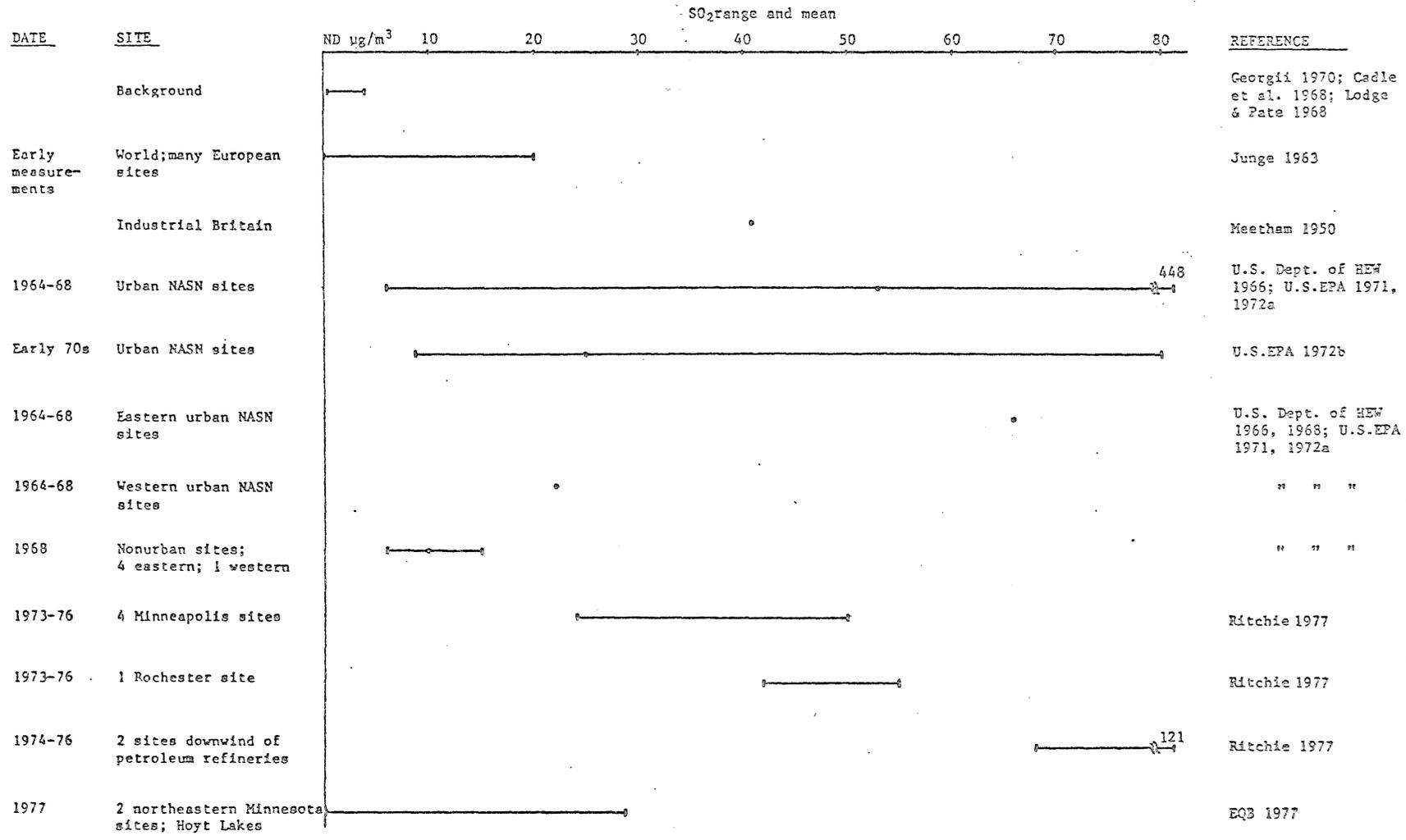
Large point sources located outside of cities were responsible for most of the increased SO_2 emissions in the 1960s, and electric power plants contributed about 88 percent of the total from these sources (CNR 1975). Power plant sulfur oxide emissions continued to increase in the early 1970s, when the overall trend was a slight decrease (U.S.EPA 1976b). At present, large point sources are the emitters most likely to violate federal air quality standards (U.S.EPA 1975).

Sulfur oxide emissions are expected to continue to increase if no further abatement programs are implemented (CNR 1975). The National Academy of Engineering has predicted a 66 percent increase in emissions in the U.S. from 1970 to 1990. Emissions from power plants are expected to double from 1970 to 1980 and triple from 1970 to 1990 (CNR 1975).

About 80 percent of the total electric generating capacity derived from fossil fuels in the U.S. is located in the eastern half of the country; about 50 percent of the total is located in the northeastern states (Shriner et al. 1977). The northeast also contributes about half the total sulfur oxide emissions (CNR 1975), and has the highest ambient SO_2 and SO_4 levels.

Concentrations of sulfur dioxide in the air are shown in Figure 2. The effect of different quantities of sulfur oxide emissions can be seen:

Figure 2. Sulfur dioxide concentration in air.



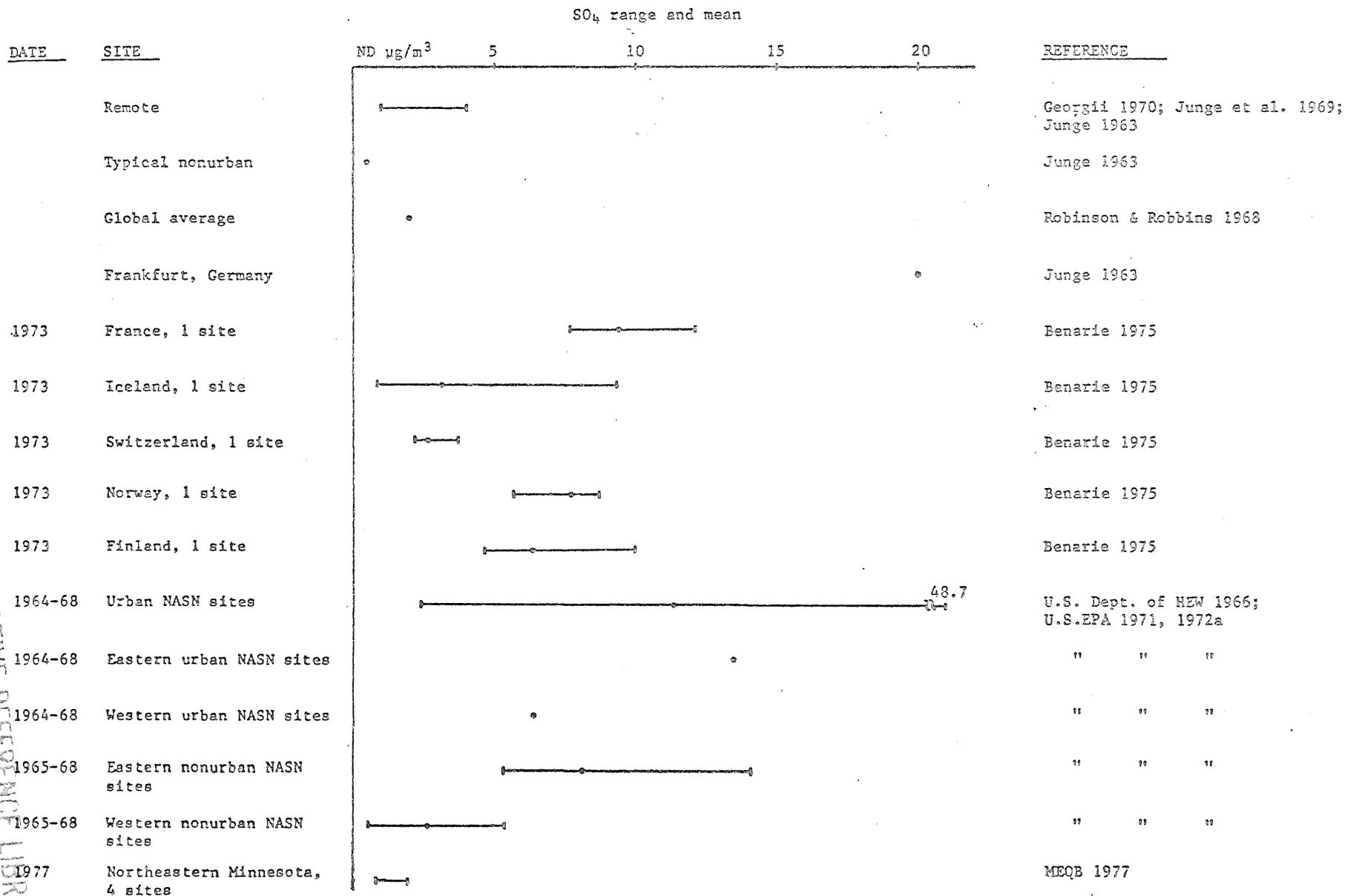
background sites have levels of $4 \mu\text{g}/\text{m}^3$ or less (Georgii 1970, Cadle et al. 1968, Lodge and Pate 1966), whereas industrial and urban areas have much higher levels. Data from the National Air Surveillance Network (NASN) reveal that the average SO_2 concentration at eastern U.S. urban sites was three times the average concentration at western urban sites in 1964 through 1968 (Altshuller 1973). NASN data from 1963 through 1972 showed western sites and midwestern sites west of the Mississippi to have only 10 to 20 percent of the east coast levels (Altshuller 1976). Highest SO_2 levels were found in the Northeast (Middleton 1970).

Five Minnesota sites had SO_2 concentrations within the early 1970 NASN reported values, but above the NASN mean (Ritchie 1977). The two sites downwind of petroleum refineries had much higher values. Only two sites in northeastern Minnesota, which are probably influenced by power plant emissions, had enough SO_2 data above the detection limit to justify inclusion in Figure 2. On one particular date, the SO_2 concentration reached $28.8 \mu\text{g}/\text{m}^3$ at one of the sites (MEQB 1977).

Figure 3 shows ambient sulfate concentrations. As with SO_2 concentrations, eastern and urban values are higher than western and nonurban values.

Background areas throughout the world have reported SO_4 levels of 1 to $4 \mu\text{g}/\text{m}^3$ (Georgii 1970, Junge et al. 1969, Junge 1963), while U.S. urban NASN sites reported levels approaching $50 \mu\text{g}/\text{m}^3$ from 1964 through 1968 (Altshuller 1973). Western urban values are generally one-half eastern urban values (CNR 1975), while western mountain states were found to have only 15 to 25 percent of the SO_4 concentration at eastern sites (Altshuller 1973).

Figure 3. Sulfate concentration in air.



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SO_4 levels in rural areas differ greatly between east and west (Altshuller 1973). West of the Mississippi SO_4 concentrations over forest and farmland ranged from 1.5 to 5 $\mu\text{g}/\text{m}^3$, whereas similar sites in the East and Midwest east of the Mississippi had values of 5 to 10 $\mu\text{g}/\text{m}^3$. Altshuller (1973) attributes this difference to a residue SO_4 level of at least 5 $\mu\text{g}/\text{m}^3$ in the East, caused by long distance transport of SO_2 , and the conversion of SO_2 to SO_4 during transport. This is due to emission patterns; sites west of the Mississippi had, at most, one SO_2 emission source within 160 km (Altshuller 1973), while eastern sites, on the average, were within 30 to 60 km of a source (Shriner et al. 1977). Transport and transformation of SO_2 is responsible for over 50 percent of the SO_4 at eastern sites and midwestern sites east of the Mississippi, and responsible for over 75 percent of the SO_4 at nonurban eastern sites (Altshuller 1976).

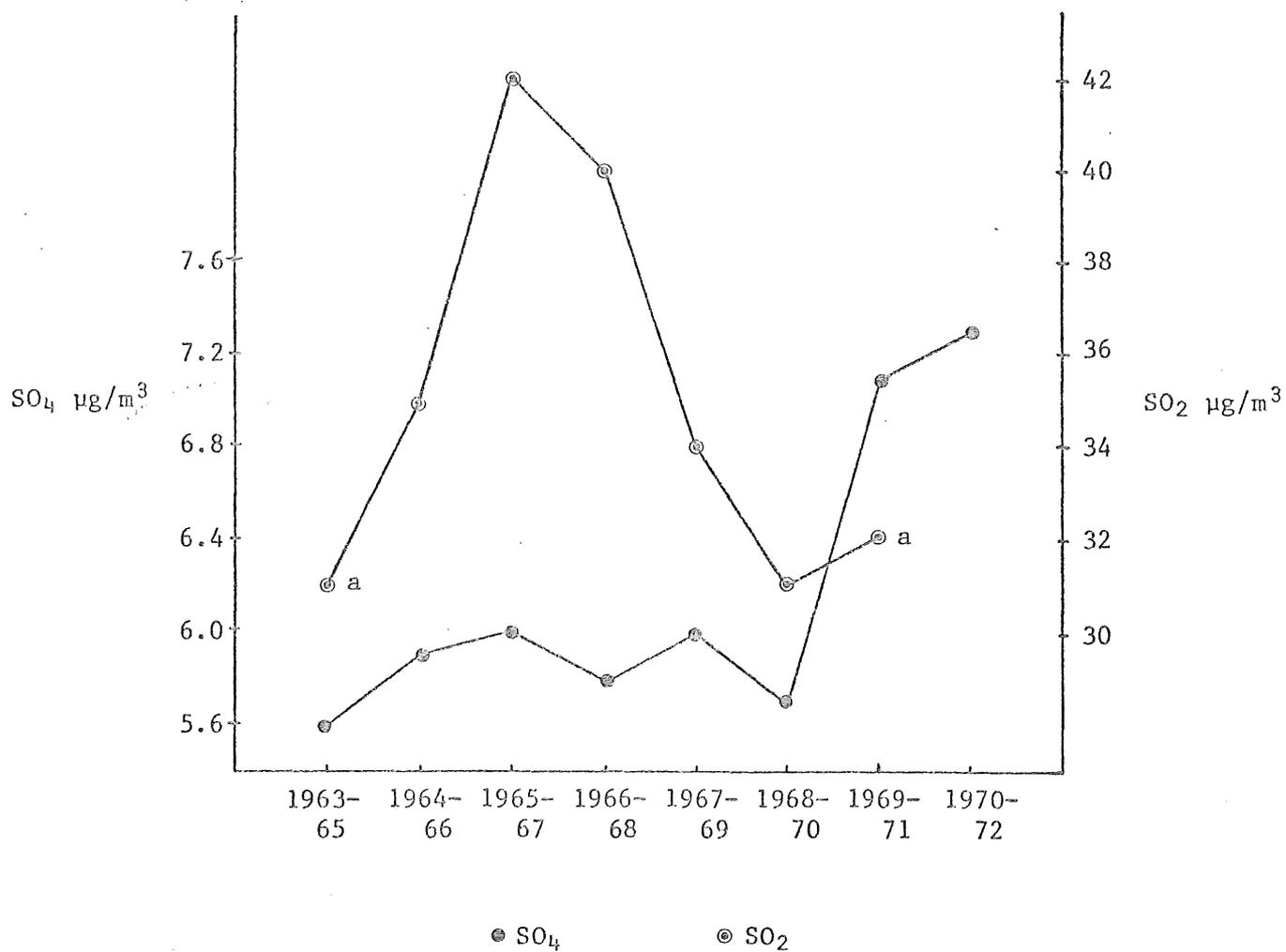
Recent trends in SO_2 and SO_4 concentrations have been determined for the U.S. and Minnesota. In Minnesota, nine sites (seven of which are shown in Figure 2) were monitored by the Pollution Control Agency (PCA) for SO_2 from 1972 through 1976 (Ritchie 1977). The period of record for most sites was not the full five years, however. The sites represented downtown, urban residential and urban commercial-residential areas, and two sites were source oriented downwind of two petroleum refineries. Trend analysis was conducted for these sites. One of the petroleum refinery influenced sites showed a decrease in SO_2 concentration, and the other an increase. Most other sites exhibited no statistically significant changes. Increases in SO_2 concentration occurred at several sites in the fall and winter of 1976. These data must be used with caution, however, because increased heating due to the extreme cold during that time may have caused greater

SO₂ emissions. Insufficient data did not permit regional analysis for SO₂ trends for Minnesota.

Other urban areas outside of Minnesota showed distinct SO₂ concentration trends. The 1969 through 1971 NASN data indicate a decrease of about 50 percent in the average SO₂ level at urban sites (CNR 1975), and from 1971 through 1975 a decrease of about 30 percent (U.S.EPA 1976a). In the nation as a whole, SO₂ concentrations decreased rapidly from 1970 through 1973 and then ceased to decrease after this period. During 1975 the levels remained relatively stable and had possibly begun to increase in some places, apparently because of failure or inability of some sources to use clean fuels (U.S.EPA 1976a). Altshuller (1976) looked at NASN data from 1963 through 1972 and determined that SO₂ levels exhibited a downward trend at east coast sites and midwest sites east of the Mississippi. A smaller downward trend may have occurred in the Southeast and West, but results were too incomplete to be certain.

Sulfate concentration decreased in the U.S. as a whole, but not to the same extent as sulfur dioxide concentration. Sulfur dioxide concentration decreased 55 percent at east coast sites during 1963-1965 and 1969-1971, while SO₄ concentration decreased only 15 percent. In the Midwest, east of the Mississippi, SO₂ decreased 39 percent during 1965-1967 and 1969-1971, while SO₄ concentration first decreased, then increased to slightly above the early 1960 levels (Altshuller 1976). Shriner et al. (1977) reported rather stable urban SO₄ levels from 1957 through 1970. A statistical study of NASN data showed the same for 1964 through 1970 (Frank 1974). Figure 4 shows what may be an increasing SO₄ concentration trend in Minneapolis,

Figure 4. Three-year running averages for sulfur dioxide and sulfate at Minneapolis (1963-1977).



^aMeasurements for only 2 of 3 years

Adapted from Altshuller 1976.

Minnesota, as well as SO_2 concentration changes, from 1963 through 1972. NASN SO_2 data do not, however, correlate with PCA data (Ritchie, personal communication).

It may appear to be a paradox that sulfur oxide emissions in the U.S. increased about 45 percent in the 1960s, whereas urban SO_2 concentrations decreased, and urban SO_4 concentrations remained approximately the same (CNR 1975). However, as mentioned earlier, the major increase in sulfur oxide emissions occurred at large point sources outside of urban areas. Rural areas have not typically been as intensely monitored as urban areas. In 1973, for example, of 213 SO_2 surveillance sites operated by the EPA, 93 percent were in heavily populated areas, and only 5 percent in background areas, and 2 percent source-oriented (U.S.EPA 1974). Because nonurban areas are not adequately reflected in SO_2 and SO_4 trend data bases, the effects of shifting emission patterns may be difficult to ascertain.

Nonurban SO_2 data are too sparse to justify trend analysis (CNR 1975, Altshuller 1976), although a slightly perceptible decrease was seen at 10 background sites from 1970 through 1973 (Shriner et al. 1977).

More extensive data are available for nonurban SO_4 concentrations. Altshuller (1976) found an increase in most regions of the U.S. between 1965 and 1972. A general increase from 1962 through 1970 was also found by CNR (1975). It appears as though man-made sulfate increased by 40 to 100 percent in nonurban areas during the 1960s. This increase is in agreement with sulfur oxide emission trends (CNR 1975). The limited data obtainable suggest that nonurban SO_4 trends may reflect the influence of SO_2 emission charges more closely than urban SO_4 trends (Shriner et al. 1977).

Nonurban Minnesota SO_2 and SO_4 ambient concentrations and trends were not discussed in the literature. From information available, however, it is clear that areas with low SO_2 emissions (such as northeastern Minnesota, the northeastern U.S., and northern Europe) may have SO_4 levels well above background concentrations due to long distance transport and transformation of SO_2 , and therefore, may have acid precipitation problems (CNR 1975, Altshuller 1976).

CURRENT ACIDITY OF PRECIPITATION

Increases in precipitation acidity have occurred simultaneously with SO_2 and NO_x emission increases. Likens (1976) reports that the pH of precipitation in the eastern U.S. dropped significantly between 1930 and 1950. Free acidity (pH) was not measured prior to 1940, so exact pH values are not known; however, methyl orange tests indicated that the pH was above 4.6 (Likens and Bormann 1974). In addition, relatively large amounts of HCO_3 were found in precipitation at Geneva, New York, before 1930 (Collison and Mensching 1932). This probably indicates that pH values were 5.7 or higher (Likens and Bormann 1974).

A marked decrease in mean precipitation pH in Norway occurred throughout the 1955 to 1976 period (Dovland et al. 1976). The rate of pH change of bulk precipitation (both wet and dry deposition) in Sweden and Norway was $-.03$ to $-.08$ pH units per year from 1955 to 1969 (Oden 1975). Other sources indicate a drop of $.05$ to $.09$ pH units per year in Swedish precipitation from 1955 to 1975 (Dickson 1975, Andersson 1972).

Recent precipitation pH values in the literature ranged from 2.1 to 8.6 (see Figure 5 and Table 2). Areas defined by the literature as "remote"

Figure 5. Selected precipitation pH values.

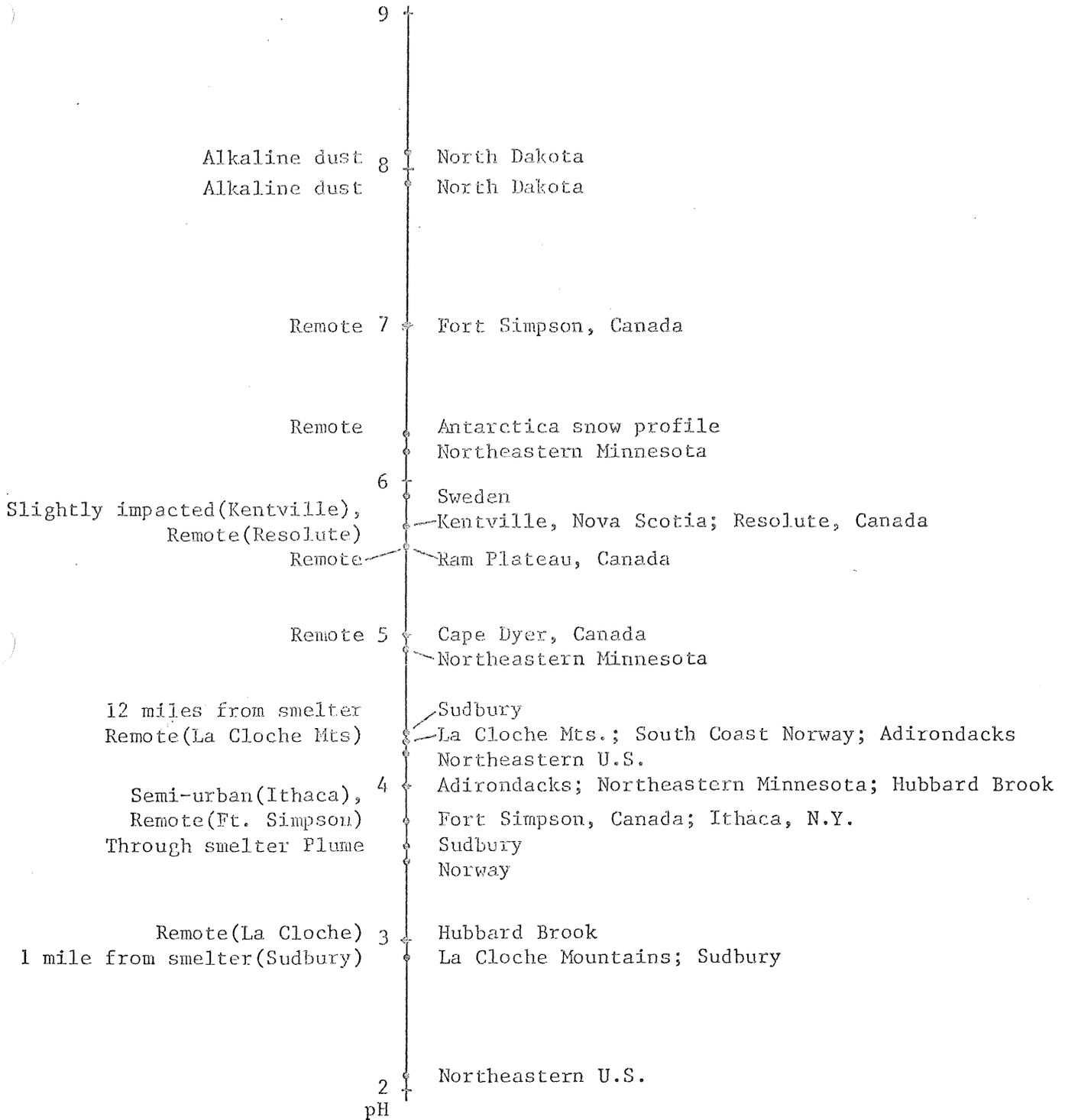


Table 2. Precipitation pH.

Location	pH	Comments	Reference
Remote background			
Ram Plateau, N.W.T, Canada	5.55	bulk sample	Kramer 1975
Fort Simpson, N.W.T.	3.8-7.0	precipitation only	Wheelpdale and Summers 1975
Cape Dyer, N.W.T.	5.0	snow event	
Resolute, N.W.T.	5.7	rain event	Kramer 1973a
Antarctica	6.32	snow profile	Gjessing and Gjessing 1973
North Dakota	7.9,8.1	snow--dustfall from arid lands raises pH	Adomaitis et al. 1967
Northeastern Minnesota			
Ely	4.0,4.5,4.8,4.8, 5.0,5.2	rain events, 1977	MEQB 1977
Kawishiwi Lab	4,3.4.7,4.7,4.8, 6.2	rain events, 1977	
Burntside Lake	5.7	rain event, 1977	
Average 3 stations	4.9	12 rain events, 1977	
Northeastern U.S.	2.1-3.0 4.0-4.2	individual storm minimums annual average, precip	Likens and Bormann 1974, Likens and Borman 1975, Cogbill 1975
Hubbard Brook Experimental Forest, N. Hamp. (no nearby industrial or population centers)	4.03-4.21 3.0	average annual weighted pH 1964-74 event minimum	Likens et al. 1975
Ithaca, N.Y.	3.82-4.18	rain and snow, Feb-June 1975	Galloway et al. 1975
Adirondacks	4.05-4.31	rain and snow	Schofield 1975
Sudbury, Ontario	<4 2.85 4.34	rain and snow average within 10 miles of Sudbury rainfall, dust fall average 1 mile south of Coniston Smelter rainfall, dustfall average 12 miles east of Coniston Smelter	Hutchinson and Whitby 1974

Table 2. (cont.)

Location	pH	Comments	Reference
Sudbury contd.	3.6	rain event falling through plume	Wiebe and Whelpdale 1975
	3.8-5.2	precipitation events, variation over 11 days	Kramer 1976b
LaCloche Mountains, Ontario (remote area southwest of Sudbury)	4.3	average of 18 precipitation events 1972-73	Beamish and Van Loon 1975
	3.6-5.5	5 rainfall events, 1969-71	Beamish and Harvey 1972
	2.9	precipitation event minimum	
Kentville, Nova Scotia (agricultural area, little industrial pollution)	5.7	23 rain and snow samples 1952-54	Herman and Gorham 1957
Norway	4.3	annual mean--south coast, area of most acidic precip	Dovland et al. 1976
	3.5	precipitation minimum several stations	
	3.7-4.9	precipitation events	Scholdager 1973
	3.35-5.8	forest, precipitation	Bjor et al. 1974
Sweden	4.3-4.4		Hornstrom et al. 1973
	3.9-5.9	bulk sample	Andersson 1972

generally have pH values above 5, whereas values near 4 are not unusual in highly impacted areas. In much of the northeastern U.S., precipitation has an average pH of between 4.0 and 4.2 (Likens 1976). The average precipitation pH in the Sudbury area is reported to be about 4.5 (Kramer 1976a). Most precipitation pH values in Figure 2 are below 5.7, indicating acidic precipitation.

REMOVAL MECHANISMS

Pollutants are removed from the atmosphere by several different mechanisms. Wet deposition, in the forms of rain and snow; and dry deposition, composed of dry fallout, impacted aerosols, and adsorbed gases; contribute to the total amount of pollutant which reaches land and water surfaces (Galloway and Likens 1977). Bulk precipitation includes both wet and dry deposition. Types of precipitation collectors are discussed generally in Galloway and Likens (1977), and in more detail in Galloway and Likens (1975).

Rain is a more efficient scavenger of pollutants from the air than snow (Summers and Hitchon 1973, Herman and Gorham 1957). Nova Scotian rain and snow samples collected for 16 months from 1952 through 1954 revealed 40 percent as much sulfur, 33 percent as much ammonia, and 50 percent as much nitrate in snow as in rain (Summers and Hitchon 1973). Average SO_4 concentrations in precipitation in Alberta were 2.0 to 3.0 mg/l in summer and less than 0.5 mg/l in winter (Summers and Hitchon 1973). Kramer (1975) found maximum SO_4 loadings in summer and Likens (1972) found summer rains to be generally more acidic than winter precipitation. In the northeastern U.S. higher hydrogen ion concentrations occur in summer precipitation than in winter precipitation (Hornbeck et al. 1975). Sulfate deposition exhibited a nearly identical seasonal pattern as hydrogen ion deposition.

Several hypotheses have been advanced to explain the different concentrations of pollutants in rain and snow. Herman and Gorham (1957) suggest that either snowflakes have a lower efficiency of removing material from the atmosphere than raindrops, or that air masses from which snow falls have lower concentrations of materials than air masses from which rain falls. Summer convective storms are very efficient in removing SO_2 from the atmosphere, whereas stable air masses from which snow occurs have very little vertical transport of air, causing SO_2 emissions to be trapped in the lowest few thousand feet of the atmosphere and not enter the precipitation mechanism (Summers and Hitchon 1973). Hornbeck et al. (1975) suggest that summer electric power generation produces more acid-forming emissions than winter heating.

Junge (1963) states that rainout efficiency is proportional to the liquid content of the clouds. Rainout is the incorporation of SO_2 or sulfate aerosols into the physical processes of the cloud and subsequent fallout in rain (Summers and Hitchon 1973). In Alberta the liquid content of winter snow-producing clouds is typically one-tenth of the value found in summertime cumulus. In addition, the oxidation rate of SO_2 is lower in the presence of cloud droplets in summertime clouds. These factors account for much of the difference in rain and snow sulfate concentrations (Summers and Hitchon 1973). Work by Bushtueva (1954, 1957) indicates that oxidation of SO_2 to H_2SO_4 occurs more readily in high relative humidity conditions.

Wet deposition of pollutants may be greater than dry deposition. Garland (1974) found from the literature that annual wet deposition of sulfur compounds was 1.5 to 6.4 times greater than dry deposition of SO_2 gas and SO_4 particles. A dispersion model used to estimate the amount of dry deposition in Norway showed that wet deposition of SO_2 is about three times

as large as dry deposition (Dovland et al. 1976). Other scientists believe, however, that dry deposition to lakes is approximately equal to wet deposition (Kramer 1976b).

Dry fallout of oxides is relatively greater near the emission sources, whereas wet deposition is more important hundreds or thousands of kilometers from the sources (Overrein 1975). Thus, Norway, which is a greater distance from the major sources of sulfur dioxide and nitrogen oxide emissions in Europe, receives a greater wet deposition of pollutants than dry deposition.

It should be noted that Figure 1 and Table 1 include both wet and bulk deposition data, as well as some seasonal data. Based on the information in the preceding paragraphs, the SO_4 concentration values in the figure and table may not be directly comparable.

ACID BUFFERING

The acidity of precipitation can be neutralized by various substances in the atmosphere. Additions of even small amounts of particulates to the atmosphere may raise the pH of precipitation because their surfaces have the ability to adsorb H^+ (Kramer 1976a). Norton (1975) reports that most inorganic particulates tend to react with and consume acid in precipitation. In areas where there is abundant windblown dust, pH values for precipitation of 7 to 8 are not uncommon (Kramer 1976a). Gorham (1975) found differences in snow acidity in Minnesota during 1974-1975, presumably related to greater dustfall in the western cultivated area than in the eastern forested area. Western snow was alkaline (pH up to 9.5) with a high particulate content and eastern snow was acid (pH 4.5-5.6) with a low particulate content. Bases in the atmosphere which are capable of neutralizing acid precipitation

include sea spray and ammonia (Gorham 1975). Dairy farms have been suggested as sources of atmospheric ammonia in parts of Denmark and southern Sweden (Barrett and Brodin 1955).

Likens and Bormann (1974) found that the sulfur content of rain and snow in New York State is 70 percent lower now than prior to 1950, while precipitation acidity has increased since 1950. They hypothesize that a conversion of fuel source from wood and coal to natural gas has caused this change. Particulates released when coal was burned neutralized the acid formed by SO_2 emissions. Present use of natural gas produces less SO_2 , but also fewer neutralizing components. Tall stacks, equipped with precipitators to remove particulates, cause SO_2 and SO_4 to be transported to long distances. The authors conclude that local "soot problems" have been transformed into a regional "acid rain problem."

Before acid precipitation reaches a lake or stream, it may be additionally neutralized in the watershed. Henriksen and Wright (1977) found that 75 percent of the incoming acid was neutralized in the watershed of a small acid lake in Norway. When rain fell through the forest canopy in the Hubbard Brook Experimental Forest in New Hampshire, the pH rose from 4.0-4.1 to 4.7-5.0 (Hornbeck et al. 1975). Soils also have a neutralizing capability, especially those derived from carbonate-rich sedimentary rocks (Gorham 1975). Gjessing et al. (1976) found that runoff in Norway has an average weighted pH of 0.2 to 0.9 units higher than precipitation.

Finally, acid precipitation may be neutralized in lakes. Lakes in drainage basins high in easily weathered calcareous material (such as dolomite or limestone) are generally well buffered due to the presence of carbonate and

bicarbonate ions (Wright and Gjessing 1976). OH^- ions formed from the hydrolysis of CO_3^{2-} and HCO_3^- cause the water to be alkaline (Wetzel 1975). Norwegian lakes in areas of this type have pH values above 6.5, regardless of acid loading.

Lakes in drainage basins composed of highly resistant crystalline rocks are poorly buffered, on the other hand, and in Norway have pH levels of 5.5 to 6.0 if they do not receive acid precipitation, and pH levels below 5.0 if they do receive acid precipitation (Wright and Gjessing 1976, Gjessing et al. 1976, Wright and Henriksen 1976).

Cation-Al-silicates and weak organic acids are also H^+ sinks (Kramer 1973b). The cation-Al-silicate buffering system involves a slower reaction than the HCO_3^- system, and will fix the system near pH 6. Weak organic systems generally buffer between pH 4 and 5 (Kramer 1973b).

Trophic state has an effect on buffering capacity. High concentrations of phosphate, silicate, and borate may impart alkalinity. Eutrophic lakes are, therefore, more strongly buffered than mesotrophic and oligotrophic lakes (Lenhart 1976).

EFFECTS OF ACID PRECIPITATION ON LAKES

Poorly buffered lakes will eventually become acidic if they receive continued input of acid precipitation. This has happened in parts of the northeastern U.S. (Likens 1976, Cogbill and Likens 1974), Ontario, Canada (Conroy et al. 1975, Beamish 1975), and Scandinavia (Dovland et al. 1976, Bolin 1971). Long distance transport of pollutants has been found to cause acid precipitation and subsequent acidification of lakes downwind of

industrial areas (Conroy et al. 1975, Likens 1976, Dovland et al. 1976). Figures 6 and 7 are an example of how this type of impact is determined. The direction of the prevailing winds in Sudbury (Figure 6) can be matched with the zone of low pH lakes (Figure 7). Smelters in Sudbury emit 1.5×10^6 metric tons of SO_2 per year, and this pollutant has apparently been the cause of the acid lakes (Conroy et al. 1975).

The rate of lake pH change is slow, often less than seasonal variations or analytical reproducibility (Almer et al. 1974). Because of this, irreversible ecological damage may occur before acidifying trends are established (Kramer 1976a). Rates of lake pH change reported in the literature were -.01 to -.17 pH units per year (Table 3). The remote lakes within and to the east of the La Cloche Mountains in Ontario were among the lakes which experienced the most rapid acidification. These lakes are about 65 km southwest of Sudbury and receive no visible surface effluent of industrial origin (Beamish and Harvey 1972). pH measurements were available for 11 of the 22 lakes from 1961 or earlier. Each of these lakes showed a ten-fold to more than one hundred-fold increase in H^+ ions by 1971 (Beamish and Harvey 1971, Beamish 1974). Input of acid precipitation to a group of lakes in southern Norway caused a 30 to 60-fold increase in acidity from 1941 to 1975 (Gjessing et al. 1976).

Most lakes and streams in southeastern Norway now have pH values below 5 (Gjessing et al. 1976). Some high altitude lakes in the Adirondacks have pH levels of about 4.3 (Sawyer 1977). Figure 8 and Table 4 show lake pH in other areas of the world as reported in the literature. The pH of lakes in the Copper-Nickel Study area ranges from 6.4 to 7.6 (MEQB 1977).

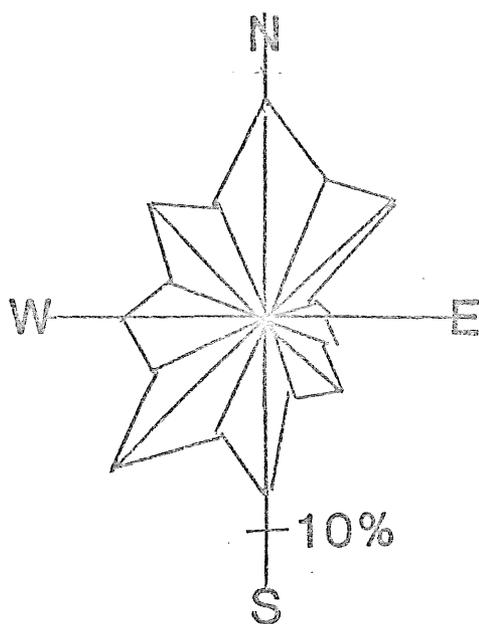


FIGURE 6. WINDROSE DIAGRAM
FOR SUDBURY (1955-1966).

SOURCE: CONROY ET AL. 1975

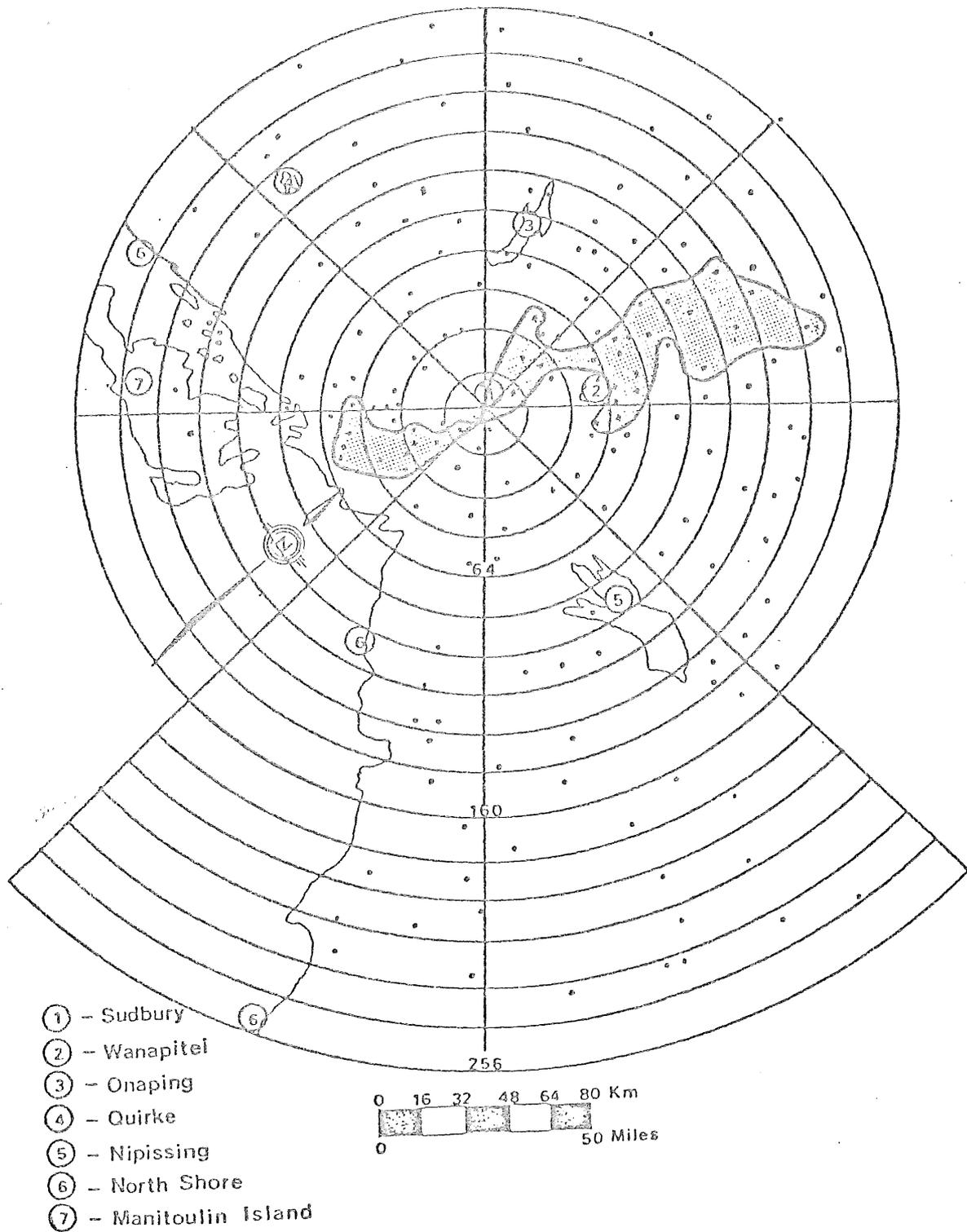


Figure 7. Sketch map of Sudbury area showing a contour for lakes with pH=5.5. For geographical reference the numbers indicated are:

- | | |
|---------------------|-------------------------------|
| 1) Sudbury | 5) Lake Nipissing |
| 2) Lake Wahnapiitae | 6) North Channel - Lake Huron |
| 3) Lake Onaping | 7) Manitoulin Island |
| 4) Quirke Lake | |

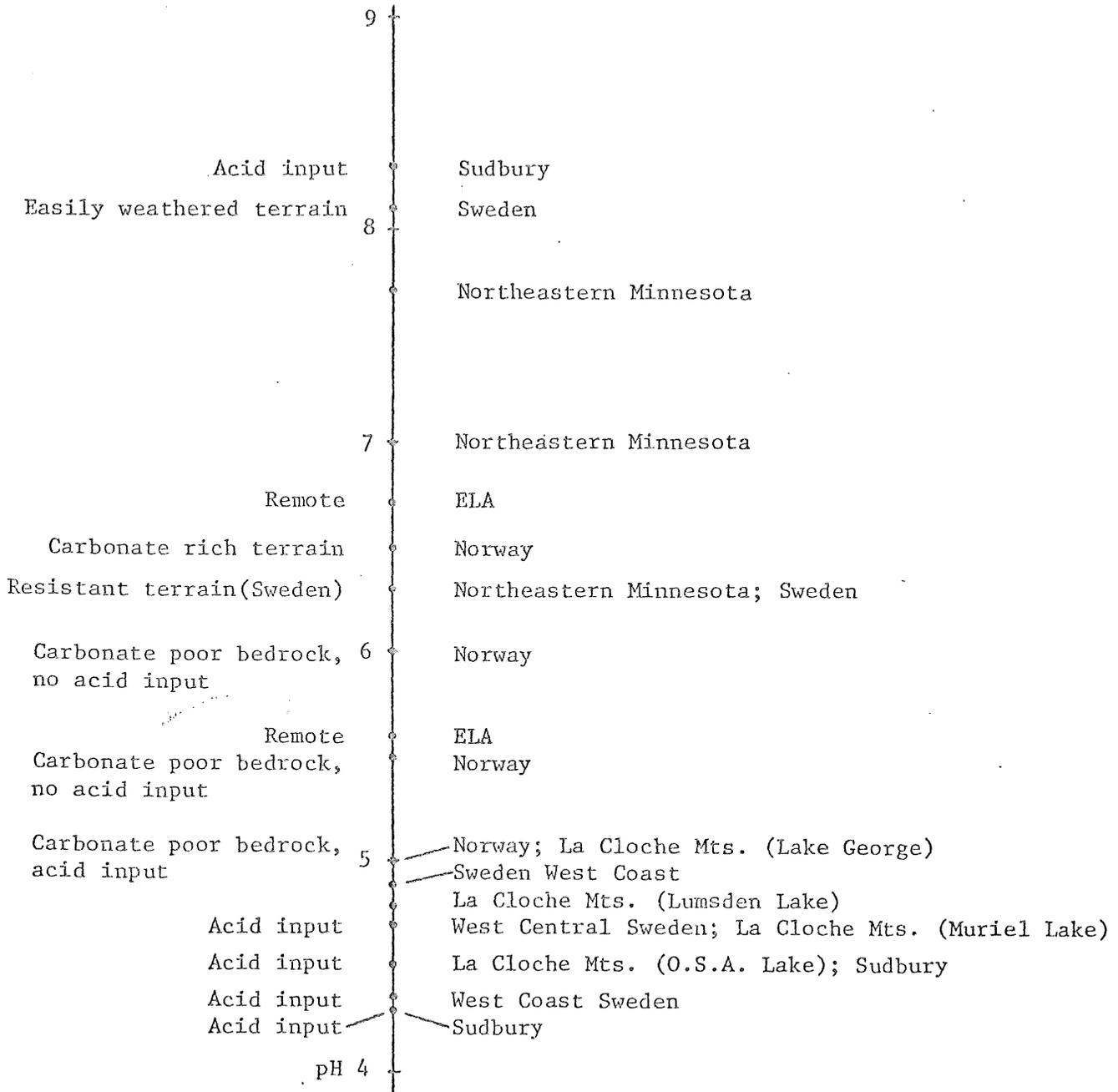
Note the northeast-southwest trend corresponding to the windrose depicted in Figure 6.

SOURCE: Conroy et al. 1975.

Table 3. Rate of lake pH change due to acid precipitation.

Location	Number of Lakes	Years	pH Change/Yr (pH units)	Reference
Sweden		1963-69	-0.01 to -0.16	Oden and Ahl 1970
Southwestern	8	1943-71	-0.04	Dickson et al. 1973
	6	1933-71	-0.03	
Westcentral	5	1937-73	-0.06	Grahn et al. 1974
Southcentral	5	1933-73	-0.03	Dickson et al. 1975
Southernmost	51	1935-71	-0.015	Malmer 1975
Norway	10	1941-75	-0.04	Gjessing et al. 1976
East Central	11	1941-75	-0.05	
Ontario		1953-71	-0.03 to -0.09	Conroy and Keller 1972
In and east of La Cloche Mountains	22	1959-71	-0.16	Beamish and Harvey 1972
North of La Cloche Mountains	26	1970-71	-0.08	

Figure 8. Lake pH.



Lake pH.

	Number or Name of Lakes	pH	Reference
Central Lake Area, Ontario	40	5.6-6.7	Armstrong and Schindler 1971
	102	6.5	Beamish et al. 1975
Central Minnesota	26	6.3-7.7 7.0 ave.	MEQB 1977
Basins of highly resistant to poor bedrock and precipitation		<5.0	Gjessing et al. 1976
Highly resistant to poor bedrock without precipitation		5.5-6.0	
Carbonate rich terrain without acid precipitation		>6.5	
Precipitation areas			
Central Sweden	112	4.37	Hornstrom et al. 1973
Central Sweden	6	4.66	Grahn et al. 1974
Adirondack Mountains, Ontario	4	4.7	
	O.S.A.	4.5	Beamish 1974
	Muriel	4.7	
	George	5.0	Beamish et al. 1975
	Lumsden	4.8	Beamish and Van Loon 1975
	4	4.5	Scheider et al. 1975
	8 within 200 km	4.3-8.3	Conroy et al. 1975
Weathered terrain	6	8.1	Dickson 1975
Unweathered terrain	7	6.3	
Forest area	3	4.9	

Hydrogen ion loading in different areas of the world was not reported in the literature; however, sulfate loading was reported and is shown in Figure 9 and Table 5. Two remote Canadian stations receive loadings of less than $365 \text{ mg/m}^2/\text{year}$. Most rural station values are less than $5000 \text{ mg/m}^2/\text{year}$. Continued input of sulfate via acid precipitation to acidified Norwegian lakes has caused a change from bicarbonate to sulfate as the major lake water anion (Wright et al. 1975).

Fluctuations in Lake pH

Periodically, extremely acid precipitation events occur and large amounts of acid are deposited in lakes and streams over short periods of time. In the Birkenes watershed in Norway 25 percent of the entire excess sulfate deposition of three years occurred in acid precipitation during 15 days (Dovland et al. 1976). After one of these events the H^+ concentration in one stream rose 300 percent, SO_4 concentration rose 20 percent, and NO_3 concentration rose 90 percent (Gjessing et al. 1976). While these increases occurred within a few hours, recovery to pre-episodic concentrations of H^+ and SO_4 took weeks. Nitrate returned rapidly to low concentrations, presumably due to biological immobilization in the forest ecosystem (Gjessing et al. 1976). Acid precipitation episodes in Norway occur most frequently in the fall, when precipitation is greatest (Gjessing et al. 1976).

Spring snowmelt also supplies large amounts of acid and other pollutants to lakes and streams (Air Pollution Across National Boundaries 1971). Lab and field studies by Johannessen et al. (1975) showed that concentrations of H^+ , SO_4 , NO_3 , and heavy metals were two to three times greater in the first meltwater than in snow. Minimum annual pH levels in Finish rivers occur

Figure 9. Sulfate loading from bulk precipitation.

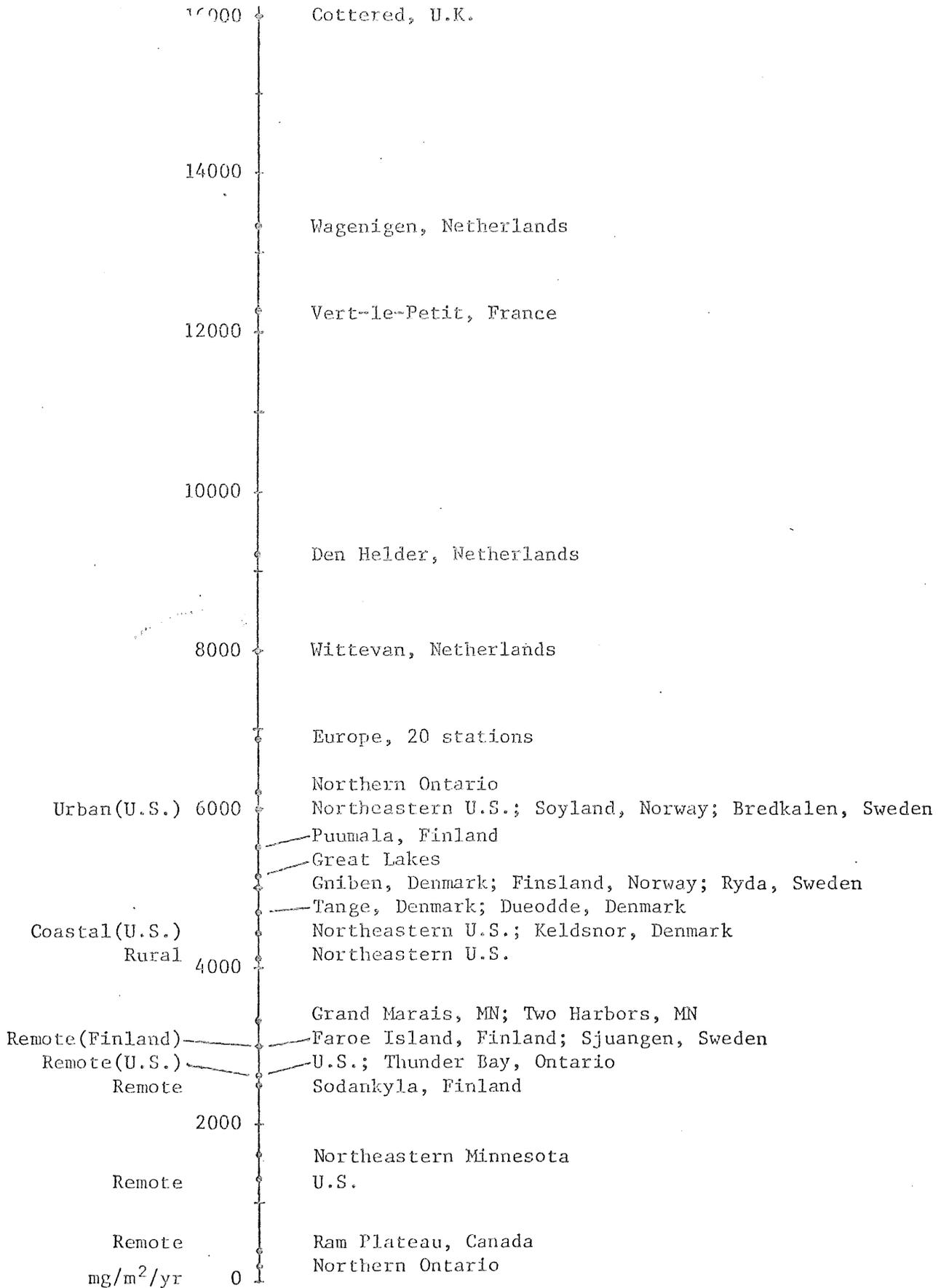


Table 5. Sulfate loading from bulk precipitation.

Location	SO ₄ (mg/m ² /yr)	Comments	Reference
Remote background			
Ram Plateau, N.W.T.	<365		Kramer 1975
Faroe Island, Denmark	3000	anthropogenically uninfluenced	Benarie 1975
Sjüanjen, Sweden	3000	anthropogenically uninfluenced	
Sodankyla, Finland	2500	anthropogenically uninfluenced	
Continental U.S.	1300-2600	anthropogenically uninfluenced	Wolaver and Lieth 1972
Northeastern U.S.			
Rural inland stations	2465-6363	14 stations	Pearson and Fisher 1971
	4140 ave.		
Coastal stations	2921-5612	3 stations	
	4382 ave.		
Urban station	5663-6621	1 station (Albany, NY)	
	6042 ave.		
Canada			
Great Lakes	5110		Shiomi and Kuntz 1973
Northern Ontario	183-6205		Kramer 1975
Europe			
Hanstholm, Denmark	4500		Benarie 1975
Tange, Denmark	4700		
Keldsnor, Danmark	4400		
Griben, Denmark	5000		
Dueodde, Denmark	4700		
Vert-le-Petit, France	12800		
Rjupnahaed, Iceland	4800		
Finsland, Norway	5000		
Soyland, Norway	6000		
Wagenigan, Netherlands	13600		
Wittevan, Netherlands	8000		
Den Helder, Netherlands	9200		
Ryda, Sweden	5000		
Bredkalen, Sweden	6000		
Jomala, Finland	5600		
Jokiainen, Finland	6800		
Puumala, Finland	5500		
Ahtari, Finland	4200		
Cottered, U.K.	16000		
Eskdalenuir, U.K.	6000		
Average 20 stations	6890		

Table 5. contd.

Location	SO ₄ (mg/m ² /yr)	Comments	Reference
Thunder Bay, Ontario	2599	average, 2 samplers, 10/73-6/75	Acres 1975
Grand Marais, Minn.	3285	average, 2 samplers, 5/74-2/75	Acres 1975
Two Harbors, Minn.	3281	average, 2 samplers, 4/74-2/75	Acres 1975
Northeastern Minnesota			
Hoyt Lakes	1600	geometric means of monthly loading rates, 1977	MEQB 1977
Dunka Road	1800		" "
Spruce Road	1300		" "
Fernberg Road	1800		" "
average, 4 stations	1600		" "

during spring melt due to acid meltwater (Haapala et al. 1975). Gjessing et al. (1976) found that large amounts of H^+ are released into south central Norwegian lakes in the first phases of snowmelt. The melt-freeze cycles that occur during snowmelt increase the concentration of pollutants (Kramer 1976a, Gjessing et al. 1976).

Natural, seasonal, and diurnal pH changes do occur in lakes because of photosynthetic activity (Oden and Ahl 1970, Johannessen et al. 1975). pH generally increases during growing periods (daylight, spring bloom, August bloom)(Kramer 1976a). In acidic lakes which have lost their bicarbonate buffering systems, however, large pH fluctuations can occur in response to episodic inputs of acid (Wright and Gjessing 1976, Gjessing et al. 1976).

Additional Effects on Water Chemistry

Acid precipitation may cause changes in lake water chemistry in addition to acidification. Acid precipitation is usually high in trace elements such as Cu, Ni, Pb, Zn, and As from urban and industrial areas (Gorham 1975). Extremely high Cu and Ni concentrations occur in precipitation near Sudbury (Conroy et al. 1975). Concentrations of metals in lakes are, therefore, elevated due to direct deposition by precipitation (Conroy et al. 1975). Organic toxins and nutrients, particularly phosphorus, nitrogen, calcium, and potassium, may also accompany acid precipitation (Gorham 1975).

Runoff of acid precipitation across land surfaces also adds chemical components to lakes (Wright and Gjessing 1976). Acid precipitation may affect soils in various ways, including:

- 1) increase the mobility of most elements,
- 2) increase the loss of clay minerals,

- 3) change the cation exchange capacity-increase or decrease,
- 4) generally increase the rate of removal of all cations from the soil,
- 5) increase flux of nutrients through ecosystems contiguous with the soil and through aquatic ecosystems (Norton 1975).

Unusually high aluminum and magnesium levels have been found in the acidic La Cloche Mountain and Scandinavian lakes (Wright et al. 1975). Since the solubility of Al is negligible at pH above 5, the appearance of Al in lakes is probably due to washout from the soil by acid precipitation (Norton 1975, Wright et al. 1975). Ca and Mg are also leached from soils by acid rain (Overrein 1972, Henriksen 1972).

In acid lakes, a greater portion of trace metals are in the soluble fraction from desorption (Kramer 1976a, Ontario Water Resources Commission 1970). Manganese and zinc levels are high in the La Cloche Mountain lakes and Swedish lakes, apparently as a result of increased solubility (Beamish and Van Loon 1975). Galloway et al. (1976b) found that both Al and Mn are impoverished in sediments of an acid lake in the Adirondack Mountains, indicating that lake sediments may act as a source of these elements to lake water.

Kramer (1976a) suggests that pH should be considered the master variable in lake chemistry because it affects, directly or indirectly, primary and secondary aquatic production and lake production. The residence time of trace metals and nutrients increases in acid lakes because of decreased biological production and decay (Kramer 1976a). pH of less than 5.5 has an adverse effect on the aquatic biological community (Conroy et al. 1975), and sudden pH drops due to spring meltwater or acid precipitation episodes may cause a severe shock to aquatic life at higher pH levels (Wright et al.

1975). The additive effects of heavy metals and acid may cause stress at a higher pH than if metals are not present (Beamish 1975, Dickson 1975).

CONCLUSION

Acid precipitation has caused damage to terrestrial and aquatic ecosystems in several areas of the world, notably Scandinavia, the northeastern U.S., and Ontario, Canada (Likens 1976, Cogbill and Likens 1974, Conroy et al. 1975, Beamish 1975, Dovland et al. 1976, Bolin 1971, Kramer 1976a).

Precipitation has become increasingly acidic over the past two decades and pH values of four or lower are now not unusual in the areas mentioned above (Likens 1976, Dovland et al. 1976, Hutchinson and Whitby 1974). Additional areas can be expected to receive acid precipitation if acid-forming pollutants, particularly sulfur dioxide and nitrogen oxides, continue to be released to the atmosphere in increasing amounts. Especially vulnerable to acid precipitation are lakes which are poorly buffered due to geologic environment and are located downwind of areas of high pollutant emissions (Wright and Gjessing 1976). These lakes may suffer from, or continue to suffer from, acidification and other associated chemical changes, and subsequent loss of fish populations and other aquatic life.

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