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Distribution and concentration of fluorides in native vegetation and small mammal species in the vicinity of the St. Regis Indian Reserve, 1977

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THE DISTRIBUTION AND CONCENTRATION OF FLUORIDES IN NATIVE VEGETATION AND SMALL MAMMAL SPECIES IN THE VICINITY OF THE ST. REGIS INDIAN RESERVE - 1977

By

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The St. Regis Indian Reserve, located along the Canadian - U.S. international boundary by Massena, New York, lies immediately downwind of two primary aluminum smelters (Reynolds Metals and Alcoa) sited along the St. Lawrence Seaway in New York State. Almost three-quarters of the 5000 tribal residents live on Cornwall Island, which lies within the Ontario, Canada portion of the seaway.

Since 1969, Cornwall Island residents have asserted that excessive fluoride emissions from the two smelters adversely impact the Island. Although emission control programs undertaken by Reynolds and Alcoa in the early 1970s have satisfied applicable New York State standards, ambient monitoring programs undertaken by the Ontario Ministry of the Environment demonstrate that existing emission levels chronically exceed both the Ontario criteria and proposed Canadian standards for gaseous fluorides. Despite the completion of these pollution reduction programs, the Indians contend that problems continue and, in 1977, initiated extensive independent studies to assess the extent and degree of emission levels impacting the Reserve area.

The data presented herein are the results of fluoride analyses of more than 1000 samples of native vegetation and small mammal species collected from the St. Regis environs during the summer and fall of 1977 at the request of the tribal leaders. The fluoride levels found in the plant and animal life, particularly on Cornwall Island, support the St. Regis Indians' contention that excessive fluoride emissions, primarily from Reynolds Metals, are accumulating in the Reserve area, with levels up to 200 times greater than those observed in samples from areas not subject to fluoride pollution. Analysis of ambient air and meteorological data and trends during this time period provides evidence that fluctuations in meteorological factors such as rainfall or wind patterns cannot be relied upon to alleviate the continuing fluoride contamination in the area.

THE DISTRIBUTION AND CONCENTRATION OF FLUORIDES IN NATIVE VEGETATION AND SMALL MAMMAL SPECIES IN THE VICINITY OF THE ST. REGIS INDIAN RESERVE - 1977

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Chapter 1

INTRODUCTION AND DESCRIPTION OF ACTIVITIES

The St. Regis Indian Reserve, located in and along the St. Lawrence Seaway, encompasses sections of the United States and Canada, including two Canadian provinces (Ontario and Quebec) and the state of New York. More than 5,000 Mohawk Indians inhabit the Reserve; approximately 3,600 people live on Cornwall Island in Ontario, 1,000 in New York State and 1,000 in the province of Quebec.

Currently there are two major sources of atmospheric fluoride pollution in the St. Regis Reserve area, both of which are located in New York State and are under the jurisdiction of the New York State Department of Environmental Conservation. In 1903, the Aluminum Company of America (Alcoa) established an aluminum reduction facility in Massena, New York, approximately eight miles west-southwest of Cornwall Island. In 1959, the Reynolds Metals Company built an aluminum reduction facility approximately one mile southwest of Cornwall Island. A phosphate fertilizer plant in the city of Cornwall, Ontario, was a third major source of atmospheric fluoride, but this facility was converted to a dry blending operation in 1975 and is no longer considered a significant source of fluoride emissions (Pruner, 1977).

The St. Regis Indians, particularly those people residing on Cornwall Island, have complained to the Ontario Ministry of the

Environment since 1969 about the fluoride pollution in the area. Many people felt that fluorides, particularly those emitted by the nearby Reynolds facility, were adversely affecting such things as forage grasses, plants, livestock or window glass (Cornwall Island residents, personal communication, 1977). During the early 1970s, both New York State and the Ontario Ministry of the Environment conducted ambient air and vegetation monitoring in the areas surrounding the Reynolds facility, which is the closest to the Reserve. Based on the results of these investigations, an order was issued by New York State requiring Reynolds to install pollution abatement equipment in order to meet New York State ambient air standards. In response to this order, $17.8 million worth of pollution control equipment was installed by Reynolds which reportedly reduced potroom emissions from 307.2 pounds of fluoride per hour in 1968 to 112 pounds of fluoride per hour in 1973 and finally to 73.3 pounds per hour by 1977 (Delisle, 1977).

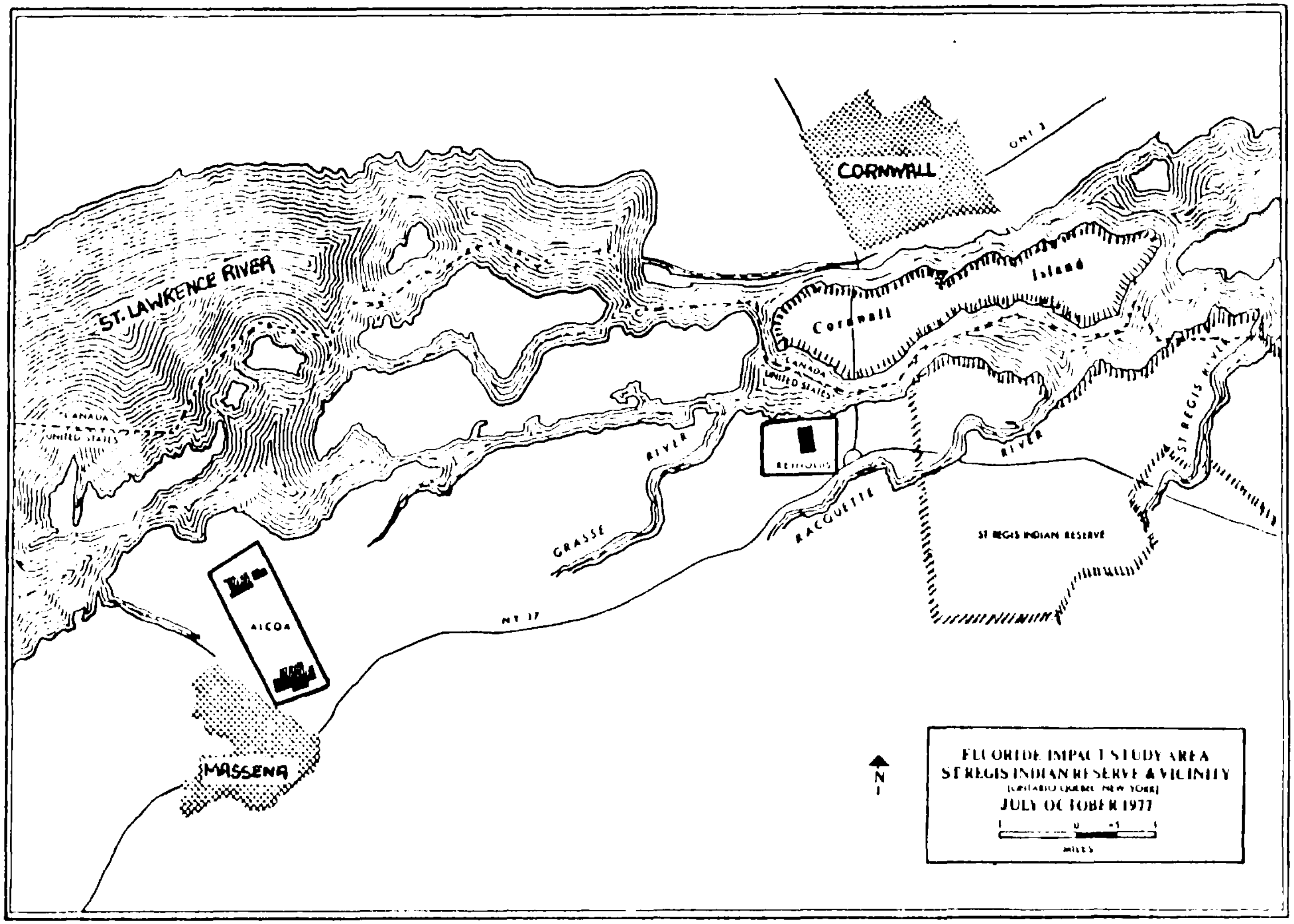
Following the original 1973 compliance deadline, both Canadian and New York State agencies continued monitoring for fluorides in the area. Despite Reynolds' compliance with New York State's emission reduction order, these subsequent studies indicated that by 1977 fluoride pollution was still impacting the Cornwall Island area (St. Regis Band Council, 1977). As a result, the St. Regis Indians initiated extensive independent studies in order to assess the extent and degree of pollution impacting the Reserve area, particularly Cornwall Island.

The data presented in this report are the results of fluoride analyses of various types of native vegetation and small mammal species collected from the St. Regis Reserve environs during the summer and fall of 1977 at the request of the St. Regis Mohawk Band Council. Figure 1 shows the general study area, which included sites around both the Reynolds and Alcoa facilities and several non-Indian owned lands. More than 1,000 vegetation and small mammal species were collected and chemically analyzed for fluoride content. Sampling was conducted at two different time periods during the 1977 growing season to observe patterns of fluoride uptake over time.

During these same time periods, samples of forage grasses, garden vegetable foliage, and coniferous trees were also collected from the Reserve and analyzed for fluoride content. The results of these analyses were previously reported in "Fluoride Levels in Vegetation from the Vicinity of the St. Regis Reserve - 1977," by Joan Miles, September 9, 1981, University of Montana Environmental Studies Laboratory, Missoula, Montana.

The objectives of the study reported herein are: 1) to locate the most impacted areas of the St. Regis Reserve Vicinity; 2) to confirm that Reynolds Metals Inc. and Alcoa are sources of fluoride pollution affecting the area; 3) to establish a record of geographical distribution and concentration of fluoride uptake within various native vegetation species at variable distances and directions from the sources; and 4) to present historical information and perspectives on an international air pollution case challenged by the unique political entity of the Mohawk Indians of the St. Regis Reserve. The summarized findings of other investigations carried out in this area on fluoride uptake in native vegetation, meteorological data, and ambient air levels of gaseous and particulate fluorides have been included as part of this fourth objective.

Figure 1



Fluoride Impact Study Area of St. Regis Indian Reserve and Vicinity

July-October, 1977

Chapter 2

LITERATURE REVIEW OF THE BIOLOGICAL EFFECTS OF FLUORIDE POLLUTION

Comprehensive reviews of the nature and biological effects of fluoride pollution have been published by the World Health Organization (WHO, 1970), the U. S. National Academy of Sciences (NAS, 1971 and 1974), the National Research Council of Canada (Rose and Marier, 1977), and the U. S. Environmental Protection Agency (EPA, 1980). Rose and Marier (1977) note that "plants are affected by fluorides in soil, water and air; animals by fluorides in their forages, feed supplements and water; and man by fluorides in his food, beverages, drugs, cigarettes, and air." Emphasized in each of the review documents, however, is the need for further field research on environmental fluorides as opposed to laboratory research. Weinstein (1977) explains that the long-term effects of low-level, recurrent fumigations or the effects of short-term, high-level fumigations of one or more pollutants on the environment are difficult to assess because laboratory conditions seldom mimic the variability of natural field conditions.

Atmospheric fluorides are absorbed primarily by foliar tissue and then move to the marginal portions of the leaves, causing necrotic lesions and death of the tissue. Gaseous forms of fluoride are more effectively absorbed than particulate forms, and lead to more significant plant injury (EPA, 1980). In general, tissue fluorides increase with increased length of exposure and increased atmospheric concentrations.

Fluoride accumulated in one leaf or section of a plant is generally immobile and does not move to other leaves or organs (NAS, 1971). While there is some translocation from root to shoot (presumably through the transpiration stream), once fluorides localize in leaf tissue, little significant reverse downward translocation of fluorides have been observed (EPA, 1980).

It has been suggested that fluoride accumulation causes inhibition, modification or disruption of metabolic pathways or may alter the production of secondary metabolites (NAS, 1971). Existing data, however, are inadequate to determine subcellular sites of localization, and the metabolism of organic fluorides by plants is not well understood.

Fluoride is found in virtually all plants, in concentrations ranging from 2 to 20 ppm of dried plant matter. Abnormally high concentrations of fluoride are generally documented as detrimental. Excessive fluoride can cause growth inhibition, tip and marginal foliar necrosis, chlorosis, wilting, and eventual death of the plant. Fruit quality and yield can be impaired, as well as inhibition of seed germination (EPA, 1980). Weinstein (1977) reported that when the assimilatory capabilities of a single leaf are reduced by abscission, necrosis, or reduction in mass, it can be assumed that the entire plant is being affected in the same way. Consequently, effects on the organism may be manifested as altered growth, reduced reproduction, decreased resistance, or death.

While the effects of toxic gaseous fluorides are more apparent in vegetation, the deposition of particulate fluoride on vegetation and the accumulation within the plants can seriously affect foraging animals (NAS, 1971). When ingested by cattle, fluorides can induce either acute toxicosis or a debilitating chronic condition referred to as chronic toxicity or fluorosis (NAS, 1974). Although relatively rare, acute toxicosis may cause symptoms such as reduced milk production, nausea, vomiting, incontinence, convulsions, necrosis of mucosa of the digestive tract, weakness, and cardiac failure. Chronic fluorosis is the response most often observed in livestock and wildlife (Shupe et al., 1972). The development of this disease is often very subtle, and symptoms vary. It is difficult to define the precise point at which fluoride ingestion is harmful because of the many factors involved, such as solubility of the fluorides, amount and duration of ingestion, species, general level of nutrition, and individual response.

Fluorides found naturally in soils or added to the soil in fertilizers are inconsequential in fluoride uptake in vegetation (EPA, 1980). Prince et al. (1949) reported that experimental evidence shows conclusively that the amount of fluoride accumulated from the soil is small and that there is little correlation between fluoride concentrations in the plant itself and the soil. Data strongly indicate that after relatively soluble fluoride compounds are added to soil at high rates of application, there is little or no effect on vegetative uptake (NAS, 1971). MacIntire and his co-workers have suggested that the amount of fluorides added to soils by industrial air pollution is also insufficient to affect uptake (MacIntire et al., 1949; MacIntire et al., 1951; MacIntire et al., 1955; MacIntire et al., 1955a).

Chapter 3

FLUORIDE POLLUTION SOURCES NEAR THE ST. REGIS RESERVE

The Aluminum Reduction Process

The two major sources of atmospheric fluoride pollution in the St. Regis Reserve area are the Reynolds Metals and Alcoa Aluminum Reduction facilities. All primary aluminum in the United States is produced by the electrolytic reduction of alumina (Al_2O_3), known as the Hall-Heroult process (EPA, 1978).

Alumina, obtained from bauxite ore, which is found in many parts of the world, including Europe, Africa, North and South America, and Australia, is dissolved in a molten cryolite solution in order to undergo electrolytic reduction. The cryolite, a double fluoride salt of sodium and aluminum (Na_3AlF_6) serves as an electrolyte and solvent for the alumina and is the source of fluoride emissions in the aluminum smelting process.

In the Hall-Heroult process, the electrolytic cell, or "pot," contains a carbon lining, which serves as both the cathode and the container for the melt. A large number of cells are then linked together electrically in series to form a "pot-line," the basic production unit of the reduction plant. In one type of cell (a prebake cell), carbon and anode blocks are suspended in the melt and individually replaced as they are consumed. In another type (known as Soderberg cell), the anode is formed by a paste made of carbon and pitch, which is fed continuously, either vertically or horizontally, from an external source and baked to a solid as it enters the cell. The aluminum, formed by reduction at the cathode, floats as a molten layer above the electrolyte and is in turn covered by a surface layer of solid alumina. Oxygen liberated at the anode by electrolysis reacts with the anode and forms a mixture of carbon monoxide and carbon dioxide, which escapes from the cell (NAS, 1971).

Several other gases and particulate emissions are released from the cell and those containing fluoride include the following: gases of silicon tetrafluoride and hydrogen fluoride; particulate cryolite, aluminum fluoride, calcium fluoride and chiolite, $Na_5Al_3F_{14}$ (EPA, 1980).

The dusts and gases from aluminum plants are largely collected by filters, centrifugal collectors, electrostatic precipitators, and various types of wet-scrubbing systems (NAS, 1971).

Reynolds Metals Company

The Reynolds Metals Plant at Massena, New York began operating in 1959 and utilizes a horizontal Soderberg anode in the reduction process. The alumina used for smelting is shipped primarily from Texas and Arkansas. Reynolds reported that $8 million were spent on environmental controls (wet scrubbers) during the first ten years of the plant's operation to remove particulate and gaseous fluorides from emissions. By 1973, Reynolds had installed $17.5 million worth of pollution abatement equipment to meet New York state standards for fluorides and to meet federal standards for particulate matter. The current pollution controls are floating-bed wet scrubbers. The New York State Department of Environmental Conservation (DEC) estimates the efficiency of these scrubbers at 80% particulate removal and 95% gaseous fluoride removal (Shen et al., 1976).

Emission reports submitted to the New York State DEC in 1977 by Reynolds indicate that Reynolds emits 461 lbs/hour of total particulates, 52 lbs/hour of particulate fluorides and 21 lbs/hour of gaseous fluorides, totalling 1,752 lbs/day of total fluorides or 320 tons/year (Table 1). On the basis of Reynolds' estimated 1977 aluminum production rate (126,000 tons/year), the fluoride output would then average approximately 4.97 pounds of fluoride per ton of aluminum. Based on the above emission estimates, therefore, one pound of gaseous fluorides is emitted for each 2.44 pounds of particulate fluorides.

Alcoa

Alcoa's Massena operations began in 1903 using power from the company's own hydroelectric dam. The plant has been expanded and now uses energy from the Moses-Saunders Dam in Massena. The alumina utilized by Alcoa is primarily from Alabama, and the smelting method is a pre-bake anode procedure. Alcoa began operating with three pot-lines; in the 1950s the company installed two additional pot-lines (P-75 systems), and a large single pot-line (P-225) was installed in 1975, totalling more than 650 pots or electrolytic cells. The P-225 system utilizes a patented environmental control system ("A398") and the older systems utilize floating-bed scrubbers. In the A398 process, flue gases are dispersed into a fluidized bed of aluminum oxide which traps the gases. Particulates are removed in a dry baghouse. This system is rated 98% efficient in capturing gaseous emissions and $>$95% in

Table 1

PRODUCTION CAPACITIES AND EMISSION ESTIMATES OF THE REYNOLDS AND ALCOA FACILITIES

	Reynolds Metals Co.	Alcoa
On-line date	July 1959	1903
Smelting method	Horizontal stud Soderberg	Pre-baked anodes
Plant capacity[1]	126,000 tons aluminum/year, 504 pots	210,000 tons aluminum/year, >650 pots
Daily aluminum output[2]	350 tons/day	583 tons/day
Pollution controls	Floating bed scrubbers	A398 fluidized bed and dry baghouse
Emissions[3]	461 lbs/hr total particulate	197+ lbs/hr total particulate
	52 lbs/hr particulate fluoride	
	21 lbs/hr gaseous fluorides	68 lbs/hr gaseous fluorides
	73 lbs/hr total fluoride	
	1,752 lbs/day total fluoride	1,632+ lbs/day total fluoride
	320 tons fluoride/year	297+ tons fluoride/yr

[1]Personal communication, Alcoa representatives, and informational pamphlets from Alcoa and Reynolds.

[2]Calculated on the basis of estimated yearly output, assuming facilities in operation 365 days per year, 24 hours per day.

[3]Emission estimates submitted by Reynolds and Alcoa to New York State in December 1977, and subsequent calculations by Miles based on these estimates.

capturing particulate fluoride emissions (J. Cagwin and E. Knapp, 1977, personal communication).

Alcoa submitted information to New York State in 1977 indicating that its facility released 68 pounds of gaseous fluorides per hour. Data on particulate emissions are incomplete and do not distinguish particulate fluorides from other particulates. At least 197 pounds/hour of total particulates are released into the atmosphere, and this does not include particulates from roof monitors on the P-75 systems. Total fluoride emissions are 1,632 pounds/day gaseous fluorides and a portion of the daily 4,758+ pounds of particulates. Other estimates indicate that Alcoa emits roughly 2.4 pounds of fluoride per ton of aluminum produced (Brown, August 1977).

Chapter 4

HISTORICAL AND BACKGROUND INFORMATION ON FLUORIDE POLLUTION IN THE ST. REGIS RESERVE AREA

The St. Regis Indians, particularly those people residing on Cornwall Island, have complained to the Ontario Ministry of the Environment (OME) since 1969 about the damaging effects of fluoride pollution. The Island residents and others residing in the area alleged that fluorides were adversely affecting forage grasses, plants, livestock, and window glass. In 1967, a local farmer contended that his dairy cattle were suffering from mottled teeth caused by ingestion of forage grasses high in fluoride content; area residents noted that the Eastern white pines on sections of Cornwall Island nearest to the Reynolds facility were dead or dying; in 1965 and 1966 employees of Reynolds complained that their automobile windows were etched while their automobiles were parked on plant property (Davis and Neighmond, 1970). People have also complained that bee, grasshopper and partridge populations have decreased noticeably since 1969 (Cornwall Island residents, 1977, personal communication).

Ambient air and vegetation sampling conducted in the area in the early 1970s by OME established the fact that "fluoride emissions from Reynolds Metals Company were adversely affecting air quality and had injured and killed some vegetation on Cornwall Island" (Pruner, 1977). Concurrently, New York State was monitoring fluoride levels in the air and vegetation, also establishing records of high fluoride concentrations (Prosser, 1977). On the basis of these findings, an order was issued by the State of New York in 1970 requiring Reynolds to install fume abatement equipment to meet New York State ambient air standards by June 1, 1973. Although Alcoa was recognized as a major fluoride source in the area, no studies had been conducted to establish what responsibility, if any, that company had for the problems on Cornwall Island.

A year later (in 1970) OME received increasing numbers of complaints from Cornwall Island residents about injury to crops and cattle. Cases of cattle fluorosis were confirmed by the Veterinary Services Branch of the Ontario Ministry of Agriculture and Forestry (OMAF) on the E. Benedict premises, Cornwall Island, and negotiations for damage were initiated. Monetary settlements were made by Reynolds, presumably to Mr. Benedict's satisfaction. Any satisfaction, however, was short-lived as area residents began to realize that monetary settlements did nothing to alleviate the cause of the problem.

In 1973, Reynolds was still in the process of installing pollution abatement equipment and failed to meet the deadline under the New York State order. In July of that year Dr. S. R. Silverborg from the State University of New York, College of Environmental Sciences and Forestry in Syracuse, New York, examined Eastern white pines at sites on Cornwall Island less than one mile from the Reynolds plant. He reported that ". . . the prevailing winds are from the direction of this (Reynolds) plant (and) I am convinced that the damage to the pines is the result of gases emitted from this plant" (Silverborg, 1973). Silverborg told island residents to sell remaining living pines and discouraged court action on the damage because the cost of any lawsuits would exceed the value of the trees. The $17.8 million pollution control equipment installed at this time by Reynolds, in accordance with the New York State order, was reported to have reduced that facility's potroom emissions from 307.2 pounds of fluoride per hour in 1968 to 112 pounds per hour in 1973. Subsequent refinements of Reynolds' operating procedures and control equipment reduced potroom fluoride emissions to 73.3 pounds per hour in 1977 (Delisle, 1977).

Continued complaints and evidence of continuing high fluoride levels in the area have resulted in the initiation of extensive studies by Reynolds, New York State and Canadian federal provincial agencies since 1975. These studies indicated that by 1977 fluoride pollution was still significantly impacting the Cornwall Island area (St. Regis Band Council, 1977). As a result, the St. Regis Indians initiated extensive independent studies in order to assess the extent and degree of pollution exposure impacting the Reserve area (particularly Cornwall Island).

In early 1977, the St. Regis tribal council and the National Indian Brotherhood of Canada also contacted the International Joint Commission (IJC) to express concern about the Cornwall Island fluoride problem and requested the IJC's Air Pollution Advisory Board to investigate the problems on Cornwall Island.

The International Joint Commission (IJC) was established pursuant to Article VII of the 1909 Boundary Waters Treaty. It functions as an investigative and study commission on a variety of matters of common interest to both the U.S. and Canada. The IJC has the power to make recommendations to the Secretaries of State of both governments on matters referred to them, but these recommendations are not legally binding. The Commission has six members, three appointed by the President of the United States and three by the Prime Minister of Canada (Bureau of National Affairs, Inc., 1976).

In 1966, an air pollution problem in the Detroit/Windsor area was referred to the IJC. Following investigation of this matter, an International Air Pollution Advisory Board was appointed in 1967 by the Commission to study air pollution problems along boundary areas and submit reports and recommendations to the IJC (IJC Annual Report, 1977). In May 1977, the IJC authorized the Air Pollution Advisory Board to hold a public meeting to discuss fluorides in the vicinity of Cornwall Island with all interested parties. The meeting was held on June 28 and 29, 1977 in Massena, New York.

In January 1978, six months after the Massena meeting, the International Air Pollution Advisory Board submitted a report to the IJC entitled "Transboundary Flow of Fluoride Air Pollution Affecting Cornwall Island." This report summarized the positions of the involved parties and included a one-page list of comments compiled by the Board. The Board advised the IJC that, in its opinion, there is no existing human health problem on Cornwall Island and that forage fluoride levels are in excess of New York and Ontario standards in a limited area which requires further study for accurate delineation.

As of March 1983, more than five years after the Advisory Board report was submitted, no resolutions or findings have been released by the IJC. Because the IJC was set up as an apolitical fact-finding body, it remains questionable whether any decisions or solutions can be expected.

The Reynolds/Cornwall Island situation is unique in several aspects. It is not merely a situation in which one federal government demands that another national government enforce its own regulations. Currently, neither the Canadian nor the U.S. federal governments have established laws and standards to regulate fluoride levels in the ambient air or in plant foliage. The Canadian government has proposed standards for gaseous fluorides, but these have not yet been enacted, so Canada essentially has no legal jurisdiction. The U.S. Environmental Protection Agency also has no jurisdiction, although sections of the St. Regis Reserve are in the United States. The province of Ontario has established criteria for ambient fluorides and fluoride in forage (Table 2). Despite repeated contraventions of these objectives in the past, the provincial government has no enforcement powers because neither the polluter nor the Reserve is in its legal jurisdiction.

The State of New York, however, has established laws and standards regulating fluoride levels in ambient air and forage (Chapter III, 257-8 to 257-8.4, Air Resources, Title 6, Official Compilation of Codes, Rules and Regulations of the State of New York). According to testimony from the New York State Department of Environmental Conservation at the June 1977 meetings in Massena, New York, there had been no violations of state standards observed since 1975 (Prosser, 1977). The Department of Environmental Conservation is, in fact, the only government agency with jurisdiction over Reynolds, and the Department contends that all New York State ambient and emission standards are being met.

The St. Regis Indians, however, contend that the New York laws neither protect the vegetation nor the animal life (cattle) of the

Table 2

ESTABLISHED AND PROPOSED FLUORIDE STANDARDS

Fluorides in Ambient Air

Agency	Averaging Time	Fluoride Type	Allowable Levels* (in ug/m^3)	
Ontario	24 hours	Gaseous	.82	
	24 hours	Total (particulate and gaseous)	1.63	
	30 days	Gaseous	.32	
	30 days	Total	.66	
Canada (proposed)	24 hours	Gaseous	.85	
	7 days	Gaseous	.55	
	30 days	Gaseous	.35	
	70 days	Gaseous	.20	
New York State	12 hours	Gaseous	3.7	(>4.5 ppb)
	24 hours	Gaseous	2.85	(>3.5 ppb)
	7 days	Gaseous	1.65	(>2.0 ppb)
	30 days	Gaseous	.80	(>1.0 ppb)

*Corrected to Standard Temperature of 25^{o}C and Pressure of 760 mm Hg.

Fluorides in Forage

Agency	Averaging Time	Allowable Levels
Ontario	--	35 ppm
New York State	Growing Season (not to exceed 6 consecutive months)	40 ppm
	Any 60-day period	60 ppm
	Any 30-day period	80 ppm

St. Regis Reserve. This position is based on results of extensive studies conducted in the area since 1975 until the present, including the results presented herein which discuss some of the findings observed in 1977 at the request of the St. Regis Indians in relation to distribution and concentration of fluorides in various native vegetation and small mammal species.

Other studies which have been undertaken since 1977 include continued assessments of fluoride uptake in cattle from Cornwall Island, fluoride uptake in vegetation from Cornwall Island and vicinity, ambient air monitoring, and a comprehensive health impact study which is in process, conducted by Dr. Irving Selikoff and associates of the Mt. Sinai Medical Center in New York. Of the reports that have been published, results indicate that even though there has been some improvement, high levels of fluorides are continuing to impact the St. Regis Reserve:

A. Fluoride-induced damage to cattle, which was documented in 1977 on Cornwall Island, is continuing with current levels of emission control at Reynolds (Krook and Maylin, 1979; Maylin and Krook, 1982).

B. Excessive fluoride contamination is still impacting vegetation species in the areas that were most severely affected in 1977. The Ontario Ministry of the Environment noted in its 1981 study that no "major changes" have been observed during the past four years, but did conclude that the overall contamination of vegetation was probably more severe prior to 1978 (Pearson, 1982). However, even the levels observed since 1978 are most pronounced immediately downwind of Reynolds, often exceed the Ontario forage contamination criteria, and are far in excess of normal controls. In its 1980 study, OME attributed this decreased contamination in the years 1978-1980 partly to the lower frequency of winds emanating from the direction of the fluoride sources (Pearson, 1981).

C. As of 1980, ambient air studies reveal that gaseous fluoride emission levels appear to be unchanged since 1977, while particulate levels have decreased somewhat (Bumbaco and Shelton, 1980). No 1981 or 1982 ambient air data is available at this time.

D. In other vegetation studies conducted by the University of Montana Environmental Studies Laboratory in 1981, fluoride levels in forage and hay samples were reported as "far in excess of normal controls," with current emission levels from Reynolds manifesting contamination of cattle forage and visible leaf injury to certain vegetative species (Rice, 1982). Rice noted that injury isopols prepared by OME in 1981 are typical of patterns documented since 1977.

Because the St. Regis Reserve Indians have not noticed the relief anticipated by Reynolds' pollution control program, tribal leaders sought, and were granted, legal standing in the U.S. District Court for the Northern District of New York (Civil Action #80-CV-135). The Tribe asserts that Reynolds and Alcoa are responsible for chronic fluorosis affecting their cattle, and that Island residents are experiencing adverse health effects from the fluoride emissions. Before hearing and trial dates will be set on this now-pending case, the ongoing health study, conducted by Dr. Selikoff in conjunction with the Canadian Health and Welfare Agency, must be completed.

Ultimately it will be up to the U.S. court system, after considering past and current monitoring data and associated impact studies, to determine the effectiveness of applicable fluoride standards and the legal responsibilities of the various bodies involved in an Indian and international air pollution issue.

Chapter 5

SUMMARY AND REVIEW OF METEOROLOGICAL AND AMBIENT AIR DATA

In order to assess the observed distribution and concentration of fluorides in the St. Regis Reserve area in 1977 as compared to other years and to explain observed trends, meteorological records and current ambient air data for gaseous and particulate fluorides were compiled and reviewed.

Precipitation and wind data for the St. Regis Reserve and vicinity for the years 1974 through 1977 were obtained from Climatological Station Reports from the Department of Transport, Canada (Cornwall, Ontario Station) by personnel from the St. Regis Band Environmental Division.

Precipitation Data

Table 3 shows monthly precipitation levels from 1974 through 1977, and Table 4 illustrates precipitation levels during 45-day "growing season intervals" (as utilized by the Ontario Ministry of the Environment in its reports). Figure 2 depicts the monthly precipitation levels given in Table 3. The growing season months are illustrated in black and the total precipitation for the year and growing season are presented below each year's graph.

Table 3

Monthly Precipitation Levels - St. Regis Reserve Vicinity 1974-1977

1974

Month	Precipitation in Inches
January	2.24
February	.91
March	4.44
April	4.70
May	4.97
June	2.69
July	3.93
August	1.13
September	2.26
October	1.77
November	3.74
December	3.51
Total for year	36.29
Growing season total	14.98

1976

Month	Precipitation in Inches
January	4.72
February	3.99
March	4.23
April	1.99
May	4.49
June	3.46
July	2.23
August	3.91
September	2.70
October	4.38
November	1.43
December	2.09
Total for year	39.62
Growing season total	16.79

1975

Month	Precipitation in Inches
January	1.55
February	2.13
March	2.65
April	2.70
May	2.13
June	2.55
July	2.14
August	1.00
September	5.98
October	5.03
November	4.74
December	2.60
Total for year	35.20
Growing season total	13.80

1977

Month	Precipitation in Inches
January	5.35
February	2.28
March	3.60
April	2.34
May	1.29
June	3.29
July	4.82
August	6.03
September	5.02
October	4.57
November	4.54
December	4.24
Total for year	47.57
Growing season total	20.45

Totals (inches)	1974	1975	1976	1977
Total for year	36.29	35.20	39.62	47.57
5-Month growing season (May-Sept.)	14.98	13.80	16.79	20.45

From: Meteorological Branch, Department of Transport, Canada: Cimatological Station Reports - Cornwall, Ontario Station

Table 4

PRECIPITATION LEVELS FOR 45-DAY GROWING

SEASON INTERVALS 1974-1977

ST. REGIS RESERVE VICINITY

	Precipitation in Inches, by Year			
	1974	1975	1976	1977
May 1 - June 15	5.81	4.68	5.15	2.00
June 15 - July 30	5.78	2.14	5.00	7.39
Aug. 1 - Sept. 15	2.36	2.84	5.04	8.28*

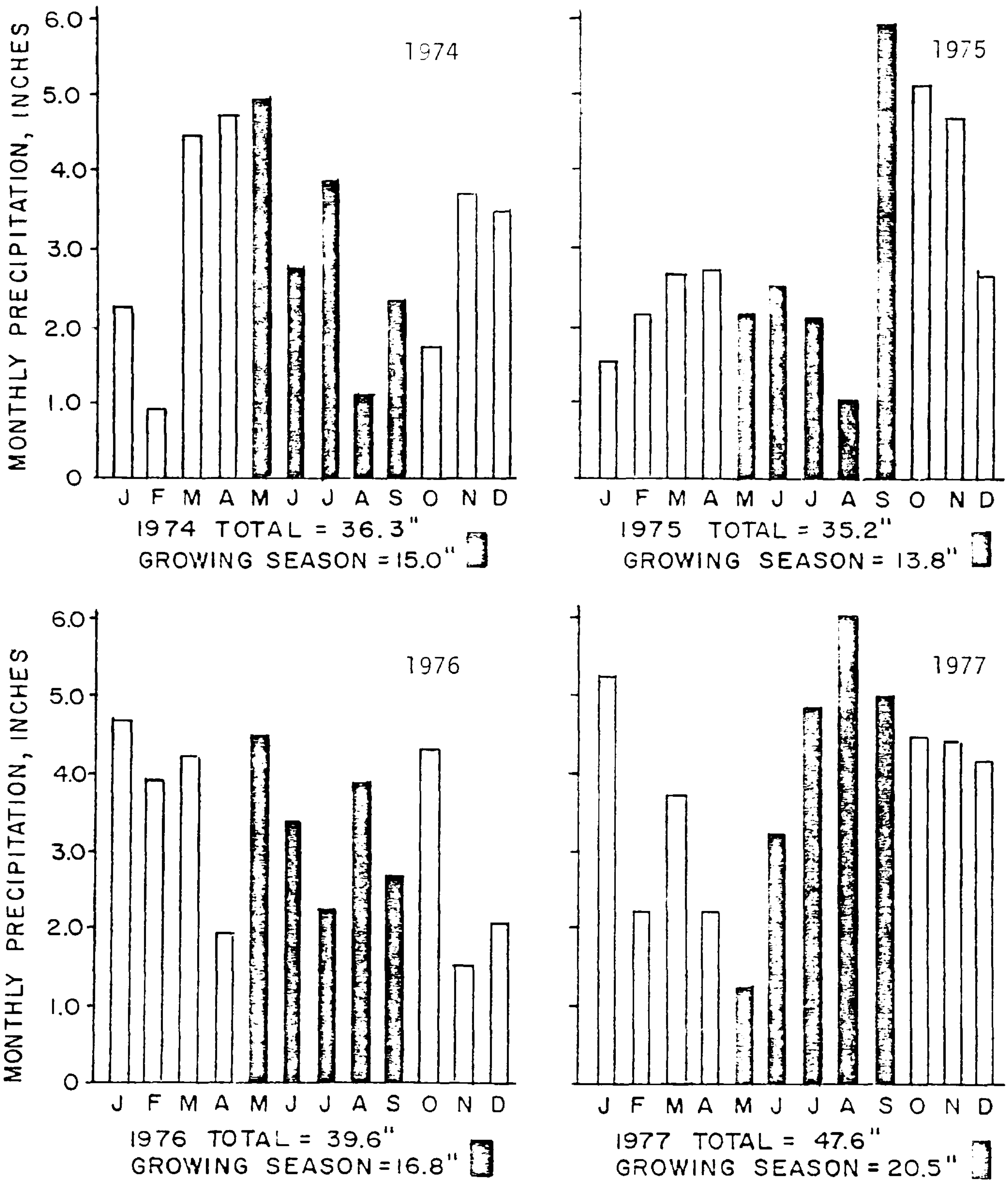
*Estimated from September total rainfall (no daily breakdown available).

Rainfall during the 1977 growing season (May - September) was at least 21.8% higher than in any of the previous years, amounting to 20.45 inches. The wettest months of 1977 were August and September, with 6.03 and 5.02 inches of rain, respectively.

The 1975 growing season rainfall and yearly total were lower than any of the four years, amounting to 13.80 and 32.50 inches, respectively. Of the 13.80 inches of rain recorded during the 1975 growing season, almost half occurred during September. Studies by the Ontario Ministry of the Environment (Pearson, 1975) placed considerable emphasis on the fact that this "dry" year was responsible for elevated vegetation fluoride levels observed in its studies of the Cornwall Island area; the relatively low amount of rainfall was also noted in a memo prepared for Reynolds by MacLean and McCune of the Boyce Thompson Institute in 1977 entitled "Review of Environmental Monitoring Programs in the

Figure 2

TOTAL PRECIPITATION – ST. REGIS RESERVE & VICINITY
1974 — 1977



Cornwall Area" (MacLean, McCune, 1977). According to this memo: "(The) explanation offered in the (Ministry of the Environment) report for the (vegetative fluoride) increases is a valid one: the reduction in both the total amount of precipitation and in the number of rain events did not remove as much surface-borne fluoride as in years of 'normal' precipitation."

The Ontario Ministry of the Environment (OME) calculated that in 1975 and 1976, the amount and frequency of rainfall in the Cornwall Island area was negatively correlated at least at the 95% level of probability with the unwashed fluoride content of vegetation (Pearson, 1976 and 1977). Emission data from Reynolds, submitted to the New York State DEC in 1977 indicate that the ratio of particulate fluoride emissions to gaseous emissions is approximately 2.44 to 1. OME's basic conclusion was that the percentage of water-soluble particulate compounds in areas close to Reynolds, that are probably removed from the vegetation in years of normal rainfall was not observed in 1975. Thus, the elevated 1975 vegetative data was primarily a reflection of environmental factors rather than emission factors, in its opinion.

In 1977, however, OME noted that "the usually strong negative correlation between the amount of rainfall and the fluoride content of maple foliage was not significant" (Pearson, 1978). In other words, the increased rainfall in 1977 "did not have the usual effect of reducing the severity of fluoride contamination of vegetation located close to the plant."

Although the amount of rainfall in a certain year may influence the total fluoride content of vegetation near Reynolds, it appears from this conclusion that rainfall does not necessarily remove excess fluoride from vegetation. It is not clear how much, if any, emissions were reduced between 1975 and 1977 but OME researchers concluded that since 1972 fluoride contamination of forage fluctuated in severity depending on meteorological factors (wind and rainfall). The exception to this finding, however, was in 1977 when increased foliar contamination was noted despite increased rainfall.

The 1977 growing season in the St. Regis Reserve area can be characterized as one of increased total rainfall, particularly during August and September. A total of approximately 8.28 inches of rain fell during the interval between the collection periods undertaken in this study (approximately August 1 September 15, 1977). This is much higher than observed in any of the previous three years (see Table 4).

Wind Data

Analysis of 1974 through 1977 wind data from the Cornwall Climatological station reveals that the predominant winds on Cornwall Island are from the southwest quadrant in which Reynolds Metals is located. Earlier studies in the Cornwall Island area mention that this phenomenon has been responsible for the deposition of fluorides to the northeast of the plant (Pearson, 1975, 1976, 1977, 1978).

Wind roses for 1974-1977 were based on the monthly summaries of the average wind speeds and frequencies (in percents) of the eight major wind directions: north, northeast, east, southeast, south, southwest, west, and northwest (Appendix A). The frequency of calm was negligible in the area.

Table 5 lists the monthly wind frequencies (1974 through 1977) of the predominant wind sources (southwest, west, and northwest) as observed in those years. The total wind frequencies of the remaining five sources are combined and listed as "other." Table 6 summarizes the yearly average frequencies of the same predominant sources, and Table 7 summarizes average frequencies during the growing season (May through September).

On a yearly basis, winds originating from the southwest, west, and northwest are responsible for greater than 50% of the wind frequencies. Because both fluoride sources (Reynolds and Alcoa) are southwest of the St. Regis Reserve and Cornwall Island, it appears that effluents from both plants move toward the Reserve a significant portion of the time.

With the exception of 1977, the predominant winds during the growing season months have been from the southwest quadrant. In 1977, winds from the southwest quadrant showed the lowest frequency in both the yearly and growing season periods than in the three previous years.

The Ontario Ministry of the Environment has also noted the effect of wind frequencies relative to fluoride deposition. In its 1976 growing season study (Pearson, 1977), it was noted that there was a "decrease" in the frequency of winds blowing from the Reynolds Metals Company Plant, and an increase in rainfall. The report noted lower foliar fluoride levels than in the preceding years and stated that the overall reduction in fluoride contamination observed in 1976 could be attributed to those two phenomena. OME also found a highly significant ($p < .01$) month-by-month statistical correlation between the frequency

Table 5

Wind Data: Mean Wind Frequencies by Month, 1974-1977 - St. Regis Reserve Vicinity

1974

Month	Source of Wind: Southwest	West	Northwest	Other
	Mean Wind Frequencies (in percents)			
January	29.8%	15.9%	12.6%	41.7%
February	23.4	23.1	20.0	33.5
March	21.9	14.2	29.0	34.9
April	39.6	10.0	18.1	32.2
May	34.9	8.4	14.0	42.7
June (no data)	-	-	-	-
July	53.7	7.1	18.0	21.2
August	44.1	11.4	9.9	34.6
September	35.7	13.1	14.4	36.8
October	35.1	11.6	26.5	26.8
November	25.4	12.4	15.0	47.2
December	27.4	17.3	11.0	44.3
Mean frequency for year	33.7	13.1	17.1	36.0
Mean frequency May-September	42.1	10.0	14.1	33.8

1976

Month	Source of Wind: Southwest	West	Northwest	Other
	Mean Wind Frequencies (in percents)			
January	24.2%	21.6%	17.9%	36.3%
February	27.1	14.8	22.8	35.3
March (no data)	-	-	-	-
April	15.4	23.9	28.3	32.4
May	27.5	22.1	26.0	24.4
June	31.5	28.1	7.5	32.9
July	31.4	21.7	16.6	30.3
August	32.7	23.8	14.9	28.6
September (no data)	-	-	-	-
October	16.9	9.2	34.1	39.8
November	23.7	25.5	33.6	17.2
December	17.8	15.3	33.0	33.9
Mean Frequency for year	24.8	20.6	23.5	31.1
Mean Frequency May-September	30.8	23.9	16.3	29.1

1975

Month	Source of Wind: Southwest	West	Northwest	Other
	Mean Wind Frequencies (in percents)			
January	25.0%	13.9%	21.2%	39.9%
February	39.3	19.5	10.8	30.4
March	23.8	1.5	27.3	47.4
April	19.9	11.1	42.0	27.0
May (no data)	-	-	-	-
June	39.4	8.8	8.2	43.6
July	60.6	14.5	7.4	17.5
August (no data)	-	-	-	-
September (no data)	-	-	-	-
October (no data)	-	-	-	-
November	33.2	13.9	13.9	39.0
December	13.2	17.9	21.9	47.0
Mean frequency for year	31.8	12.6	19.1	36.5
Mean frequency May-September	50.0	11.7	7.8	30.6

1977

Month	Source of Wind: Southwest	West	Northwest	Other
	Mean Wind Frequencies (in percents)			
January	13.0%	39.9%	21.6%	25.5%
February	17.7	34.6	20.1	27.6
March	15.8	24.9	22.4	36.9
April	16.9	20.7	25.7	36.7
May	18.4	28.8	15.5	37.3
June (no data)	-	-	-	-
July	35.1	19.5	16.2	29.2
August	32.8	26.7	23.2	17.3
September	14.8	13.4	16.2	55.6
October	15.2	18.6	17.1	49.1
November	16.9	10.4	18.4	54.3
December	18.4	16.0	18.7	46.9
Mean Frequency for year	19.5	23.0	19.6	37.9
Mean Frequency May-September	25.2	22.1	17.8	34.9

Table 6

MEAN WIND FREQUENCY BY YEAR 1974-1977

ST. REGIS RESERVE VICINITY

Wind Source	1974 (11)*	1975 (8)	1976 (10)	1977 (11)
SW	33.7%	31.8%	24.8%	19.5%
W	13.1	12.6	20.6	23.0
NW	17.1	19.1	23.5	19.6
All other directions	36.0	36.5	31.1	37.9

Table 7

MEAN WIND FREQUENCIES FOR GROWING SEASONS 1974-1977

ST. REGIS RESERVE VICINITY

Wind Source	1974 (4)*	1975 (2)	1976 (4)	1977 (4)
SW	42.1%	50.0%	30.8%	25.2%
W	10.0	11.7	23.9	22.1
NW	14.1	7.8	16.3	17.8
All other directions	33.8	30.6	29.1	34.9

*Numbers in parentheses indicate the available data in months that these mean frequencies are based on.

of wind direction and the fluoride contents at the various vegetation collection sites.

Consistent with this report, a review of the data shows that in 1976 there were fewer winds from the southwest affecting Cornwall Island than in 1974 or 1975; in 1977, however, the frequency of winds from the southwest was even less. Because of the significance that can be attributed to increased rainfall and decreases in the frequency of winds from the southwest during 1976, the 1977 data in the study reported herein prove to be particularly interesting. There was more rain during the 1977 growing season (3.66 inches more than in 1976) and less wind from the direction of Reynolds. It would be expected that these two factors would contribute significantly to relatively low fluoride levels being observed on the St. Regis Reserve, particularly in the fall of 1977. The data discussion section, however, reveals that this was not the case.

Ambient Air Monitoring Data

While meteorological factors will influence the deposition of airborne pollutants, the major factor influencing degree of contamination of plant or animal matter is the actual amount of pollutant in the air. A summary of the monitoring data for atmospheric fluorides is therefore included here in order to help assess and explain phenomenon observed in the St. Regis area with respect to location and degree of fluoride contamination.

The most common method utilized in the Cornwall Island/St. Regis area during the past decade has been "lime candle" monitoring, consisting of filter paper cylinders treated with calcium oxide or other fluoride-absorbing agents. An estimate of fluoride contamination is determined after exposing the candles to air for stated time intervals (Jacobson and Weinstein, 1977). Fluoride accumulation, or "fluoridation rate," usually is expressed as micrograms (ug) of fluoride per 100 cm^2 per 30 days. Although lime candles cannot provide time-specific information about actual ambient concentrations, lime candles do provide a good estimate of overall fluoride concentrations over time.

The only agency currently utilizing lime candles in the Cornwall Island area is the Ontario Ministry of the Environment. Environment Canada, however, conducts ambient air monitoring at two stations on Cornwall Island, and the New York State Department of Environmental Conservation operates a station just northeast of the Reynolds' property boundary.

Environment Canada (EC) measures both gaseous and particulate fluorides with a sequential sampler. New York State utilizes coated glass tubes to measure gaseous fluorides and a hi-volume sampler to measure total suspended particulates. The New York State DEC does not measure particulate fluoride levels.

Lime Candle Monitoring Data

1969 New York State. The Ontario Air Pollution Control Act established a standard for lime candle fluoride accumulation of 40 ugF/100 cm^2/30 days. In 1969, New York State, in cooperation with Ontario, operated lime candle stations at seven areas around Reynolds and Alcoa. State researchers, in their 1970 study, reported that "all lime candle stations exceeded (the Ontario) standard, although forage at some sites did not exceed the forage standard" (Davis and Neighmond, 1970). The highest ambient fluoride levels reported in this study were recorded at a station one-quarter mile northeast of Reynolds. At this site, lime candle data ranged from 2,870 ug F/100 cm^2 from May 22 to June 18 and 1,830 ug F/100 cm^2 from September 17 to October 16. The minimum reading at this station was 46 times higher than the Ontario standard. At the sites around Alcoa, lime candle data ranges from 26 ug F/100 cm^2/30 days to 95 ug F/100 cm^2/30 days.

In addition to the lime candle station just northeast of Reynolds, a station was placed one-half mile east of the plant at the U.S. Customs House. The lowest reading at the station east of Reynolds was recorded when the highest reading was recorded at the station to the northeast; conversely, the highest reading at the east station was recorded when the levels recorded to the northeast were lowest. This illustrates the responsiveness of ambient fluorides to changes in wind patterns. It is probable that when the predominant winds were to the northeast, the lowest ambient readings were noted to the east and vice versa.

<u>1969 Ontario Department of Energy and Resources Management</u>. While New York State was monitoring fluoride concentrations around Reynolds and Alcoa, S. N. Linzon (of the Ontario Department of Energy and Resources Management) was studying the effects of fluoride on vegegation in Ontario (Linzon, 1971). He reported lime candle accumulations for the growing season as follows:

1 mile NE of Reynolds	4550 ug F/100 cm^2
1.25 miles ENE of Reynolds	932 ug F/100 cm^2
1.75 miles W of Reynolds	124 ug F/100 cm^2

Accumulation in areas northwest and north-northwest of Reynolds was considerably lower. During this time, Linzon noted severe fluoride injury to Eastern white pine, Scots pine, trembling aspen, and choke-cherry in the area. Linzon's vegetation and lime candle data from the 1969 growing season indicate that the areas northeast of Reynolds on Cornwall Island were the most severely impacted.

1973-1977 Ontario Ministry of the Environment. OME maintained lime candles on Cornwall Island and in the vicinity of Reynolds from 1973 through 1977. No monitoring was conducted around Alcoa. Table 8 presents the mean fluoridation rates observed during the growing season period. This data suggests that ambient fluoride levels decreased steadily from 1973 to 1976 but that there was no improvement between 1976 and 1977. In fact, at the two sites closest to Reynolds, the mean fluoridation rates increased over 1976. All three sites were still in excess of the 40 ug standard.

Gaseous and Particulate Fluoride Monitoring Data

1973-1977 New York State. New York, in conjunction with Reynolds, has operated monitors utilizing coated glass tubes around Reynolds Metals Company since 1973 to measure ambient concentrations of gaseous fluorides. The agency also maintains high-volume samplers to measure concentrations of total suspended particulates; there is no separate measurement of particulate fluorides. Before 1974, New York operated monitors at stations west and northeast of Reynolds. At the present

1 mile NE of Reynolds	4550 ug $F/100\ cm^2$
1.25 miles ENE of Reynolds	932 ug $F/100\ cm^2$
1.75 miles W of Reynolds	124 ug $F/100\ cm^2$

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Table 8

MEAN FLUORIDATION RATES 1973-1977

ST. REGIS RESERVE VICINITY

May-September - 5-month average reported as ug F/100 cm^2/30 days
(As reported by the Ontario Ministry of the Environment)

Sampling Station Distance & Direction from Reynolds	1973	1974	1975	1976	1977
0.8 mi NE	98	84*	75	43	53
2.0 mi NE	181	174*	138	92	96
3.8 mi NE	137	72*	84	59	43
2.4 mi NNE	85	71+	54	39	--
2.5 mi NNE	85	49*	37	36	--
0.8 mi N	88	81*	66	--	--
1.7 mi N	91	100*	53	39	--
3.5 mi N	87	65*	58	56	--

* Based on 4 months' average
+ 3 months' average

time, however, monitors are maintained only at a site 0.6 miles northeast of the plant at the Reynolds boundary.

At the June 1977 meeting of the International Joint Commission, David Prosser of the New York DEC presented the following summary of contraventions of New York State standards around Reynolds (IJC Hearings Record, 1977):

--No violations of total particulate standards 1973-1977

--1973: 5 violations of 12-hour gaseous standard (4.5 ppb)
3 violations of 24-hour gaseous standard (3.5 ppb)
1 violation of 7-day gaseous standard (2.0 ppb)

--1974: 12 violations of 12-hour standard
12 violations of 24-hour standard
3 violations of 7-day standard
2 violations of 1-month standard (1.0 ppb)

--No violations of gaseous fluoride standards since 1974.

A complete set of New York State's monitoring data, obtained under the Freedom of Information Act, reveals that levels as high as 17.6 ppb fluoride were observed during a 12-hour period; the New York standard is 4.5 ppb.

It should be noted, however, that the data obtained from the New York State DEC indicate that on June 30, 1977 (one day after the IJC meeting) a reading of 5.27 ppb fluorides, which would represent a violation of the NYS 12-hour standard in 1977, was recorded at the northeast station.

The data also reveal that the proposed Canadian and Ontario 24-hour standard of 0.8 ug/m^3, if applicable, would have been exceeded consistently in 1973 and 1974 and approximately 5-10% of the recorded times in 1975, 1976, and 1977. The recorded exceedances of New York's ambient standards, the Ontario criteria, and proposed Canadian standards for 1977, are presented in Table 9.

<u>1976-1977 Environment Canada</u>. From April 15 through October 15 of both 1976 and 1977, Environment Canada (EC) maintained two sequential air samplers on Cornwall Island which separate gaseous and particulate fractions of ambient fluorides. Station "A" was located

Table 9

EXCEEDANCES OF AMBIENT AIR STANDARDS FOR GASEOUS FLUORIDES

(Recorded at the Northeast Reynolds Station, 1977 (New York State)

	Standard	No. of Violations
New York State	12-hour (4.5 ppb)	1
	24-hour (3.5 ppb)	None
	7-day (2.0 ppb)	None
	30-day (1.0 ppb)	None
Ontario criteria	24-hour (.82 ug/m^3, 1.0 ppb)	5
	30-day (.32 ug/m^3, 0.4 ppb)	1
Proposed Canadian	24-hour (.85 ug/m^3, 1.1 ppb)	5
	7-day (.55 ug/m^3, 0.7 ppb)	1
	30-day (.35 ug/m^3, 0.4 ppb)	1

at the Canadian Customs House, 1.1 miles northeast of Reynolds, and Station "B", 2.5 miles northeast. In 1976, EC monitored 12-hour averages of gaseous fluorides at both sites; in 1977, only 24-hour averages were recorded.

Based on the data in its 1976 and 1977 reports, Table 10 lists Environment Canada's seasonal averages measured for gaseous and particulate fluorides at both stations, and Table 11 lists the frequencies of winds impacting the ambient air monitors on Cornwall Island during both monitoring periods (Bumbaco, 1976, 1978). Table 12 presents Environment Canada's comparison of both year's data with existing and proposed New York and Canadian ambient standards.

In 1976, Bumbaco concluded that the proposed Canadian standards were "exceeded a significant amount of times" while the New York standards (which are approximately three times more lenient) were virtually

Table 10

SEASONAL AVERAGES OF GASEOUS AND PARTICULATE

FLUORIDE LEVELS IN AMBIENT AIR

(Environment Canada Monitoring Data, 1976-1977)

	1976		1977	
	Gaseous	Particulate	Gaseous	Particulate
Station A	.36 ug/m^3 (.45 ppb)	.35 ug/m^3 (.44 ppb)	.28 ug/m^3 (.35 ppb)	.61 ug/m^3 (.76 ppb)
Station B	.28 ug/m^3 (.35 ppb)	.15 ug/m^3 (.18 ppb)	.28 ug/m^3 (.35 ppb)	.37 ug/m^3 (.46 ppb)

Table 11

FREQUENCY OF WINDS IMPACTING AMBIENT AIR MONITORS

ON CORNWALL ISLAND

(Environment Canada Monitoring Data, Apr 15-Oct 15, 1976 and 1977)

	Frequency of Winds	
Source	1976	1977
Southwest	32%	26%
West	28%	20%

Table 12

TABULATION OF FLUORIDE DATA RELATED TO AMBIENT AIR STANDARDS

(Environment Canada Ambient Air Data, 1976-1977)

				Percent Time Allowable Level Exceeded			
			Allowable Fluoride	Station A		Station B	
Agency	Averaging Time	Fluoride Type	(ug/m^3 as HF*)	1976	1977	1976	1977
Ontario (criteria)	24-hr (Apr 15-Oct 15)	Gaseous	0.82	9	4	4	3
	24-hr (Apr 15-Oct 15)	Total (Gas & Particulate)	1.63	10	15	3	7
	30-day (Apr 15-Oct 15)	Gaseous	0.32	64	31	26	30
	30-day (Apr 15-Oct 15)	Total (Gas & Particulate)	0.66	62	69	2	48
New York State (standard)	24-hour	Gaseous	<2.99	<1	<1	<1	<1
	1-week	Gaseous	<1.73	<1	<1	<1	<1
	1-month	Gaseous	<0.84	<1	<1	<1	<1
Canada (objective)	24-hour	Gaseous	0.85	8	4	4	2
	7-day	Gaseous	0.55	15	8	5	3
	30-day	Gaseous	0.35	52	28	10	21
	70-day	Gaseous	0.20	>99	86	>99	>99

*Corrected to standard temperature of $25^{o}C$ and pressure of 760 mm Hg.

FROM: Bumbaco, M.J. and J.H. Shelton. January 1978. Ambient Air Levels of Fluorides at Cornwall Island, Ontario (April 15-October 15, 1977). Air Surveillance Division, Environment Canada.

never exceeded. The highest concentrations of both gaseous and particulate fluorides at each station were measured when the winds were blowing from the direction of Reynolds (southwest). At Station A (1.1 miles northeast), the breakdown of fluorides was 50% gaseous and 50% particulate, while at Station B (2.5 miles northeast), the figures were 60% gaseous and 40% particulate.

In 1977, Bumbaco noted that the Ontario criteria and proposed Canadian standards for gaseous fluorides were exceeded less often in 1977 than in 1976 at Station A, while the number of violations of gaseous standards at Station B remained relatively unchanged.

The raw ambient air data correlates well with the relative numbers of violations: levels of gaseous fluorides were lower at Station A in 1977 but remained unchanged at Station B. However, particulate levels at both stations doubled in 1977.

The 24-hour standards for gaseous fluorides were exceeded only when the winds were blowing predominantly from the southwest. Twenty-four of 29 violations of the Ontario 24-hour criteria for total fluorides occurred when the winds were from the southwest (direction of Reynolds). No violations of any New York standards were noted in 1976 or 1977.

The 1976 ambient data demonstrated that levels of both particulate and gaseous fluorides were higher at the station closest to Reynolds. In 1977, however, Station A had higher levels of particulates than Station B, but gaseous levels at both stations were the same. This suggests that the gaseous portions of Reynolds' emissions are remaining airborne longer than the particulate fractions, thus impacting a larger area.

Chapter 6

METHODS AND MATERIALS

Description of Collection Sites

Twenty-nine areas were sampled for fluoride concentrations in native vegetation and small mammal species.

Identical vegetation and small mammal species types were collected from control sites in similar geographical areas, approximately 75 miles southwest of the St. Regis Reserve at the Wellesley Island Nature Center, located in the Thousand Islands State Park, New York. Because the entire northeast United States is a heavily industrialized region of the country, this control site probably did not represent a completely "unpolluted" area. However, observed levels do represent minimum fluoride concentrations found in vegetation and small mammals in a non-industrialized area of the northeastern United States.

Description of Native Vegetation Species Sampled

Geographically, the St. Regis Reserve is located in a temperate, wet climate and supports a wide diversity of native hardwoods, conifers, shrubs, forbs, and grasses. Table 13 indicates the relative fluoride sensitivities of some of the species common to the vicinity.

The samples chosen at each site were the common, widely distributed species found in the area. Manitoba maple was sampled as frequently as possible because several earlier studies in the area used this as the

Table 13

RELATIVE SUSCEPTIBILITIES OF PLANTS

TO ATMOSPHERIC FLUORIDE

(Based on occurrence of foliar injury)*

Susceptible	Intermediate	Tolerant
Manitoba Maple	Green Ash	Red-Osier Dogwood
Wild Grape	Quaking Aspen	Elm
	White Sweet Clover	Oak Species
	Maples (Red, Silver, Sugar)	Willow sp.
	Sumac	Basswood

*Species listed were collected from the St. Regis Reserve - Cornwall Island study area in 1977.

From: Weinstein, Leonard H. "Fluoride and Plant Life." Journal of Occupational Medicine 19(1):49-78, January 1978; and Environmental Protection Agency. "Reviews of the Environmental Effects of Pollutants: IX. Fluoride." EPA-600/1-78-050, September 1980.

primary indicator species for fluoride uptake. Table 14 lists the common and scientific names of the species sampled in 1977 for fluoride analysis.

Because this study was completed during only one growing season and because the preponderance of vegetation in this area is deciduous, two collections were made in 1977 to evaluate fluoride accumulation in vegetation over time. The first collection (summer) took place in July and August, and the second collection (fall) was made in late September and early October. During the second collection, vegetation sampling was limited to ash species, dogwood, and manitoba maple because of the wide distribution of these species in the areas around Reynolds and Alcoa.

Field Sampling Procedures: Native Vegetation

Sites for the collection of mixed, native vegetation samples were chosen on the basis of geographical and direct exposures to either Reynolds or Alcoa, types of vegetation present, distances and directions from the smelters, and accessibility. Sites were established on Cornwall Island, the United States sections of the St. Regis Reserve, and at various locations within a five-mile radius of Reynolds and Alcoa. Sites were also sampled up to 20 miles northeast of Reynolds.

Several species of grasses and forbs, shrubs, and hardwood trees were sampled and analyzed as described in "Sample Preparation and Chemical Analysis."

Grasses and forbs were clipped 2 to 3 inches above the ground to avoid contamination by soils containing fluorides. Leaf samples only

Table 14

SPECIES COLLECTED FROM ST. REGIS RESERVE AND VICINITY

COMMON AND SCIENTIFIC NAMES*

Understory

Common Name	Scientific Name
Grasses and Forbs	
Quack grass	*Agropyron repens* (L.) Beaux.
Reed canary grass	*Phalaris arundinaceae* L.
Timothy	*Phleum pratensis* L.
Smooth brome	*Bromus inermis* Leyss.
Kentucky bluegrass	*Poa pratensis* L.
Redtop	*Agrostis alba* L.
Milkweed	*Asclepias syriaca* L.
White sweet clover	*Melilotus alba* Desr.
Shrubs	
Sumac (Staghorn sumac)	*Rhus typhina* L.
Dogwood (Red-osier)	*Cornus stolonifera* Michx.
Wild grape (Frost grape)[1]	*Vitis riparia* Michx.

Overstory

Common Name	Scientific Name
Cottonwood	*Populus deltoides* Marsh.
Willow[2]	*Salix* sp.
Quaking aspen	*Populus tremuloides* Michx.
Basswood	*Tilia americana* L.
Elm (American)	*Ulmus americana* L.
Red maple	*Acer rubrum* L.
Sugar maple	*A. saccharum* Marsh.
Silver maple	*A. saccharinum* L.
Manitoba maple (Boxelder)	*A. negundo* L.
White ash[3]	*Fraxinus americana* L.
Green ash[3]	*F. pennsylvanicus* Marsh.
Black ash[3]	*F. nigra* Marsh.
White oak	*Quercus alba* L.
Red oak	*Q. borealis* Michx.
Bur oak	*Q. macrocarpa* Michx.
Bitternut hickory	*Carya cordiformis* Wang.

*From Gleason, Henry A. and Arthur Cronquist. Manual of Vascular Plants of Northeastern United States and Adjacent Canada. D. Van Nostrand Co., N.Y. 1963.

(continued)

Table 14 Footnotes

[1]Climbing vine found predominantly at shrub height (3-5 feet from ground).

[2]Because of the difficulty in separating Willows, identifications were made only to the genus level. However, since characteristics differed quite noticeably at several collection areas, species were grouped into four general categories as follows. These sub-categories are noted in the raw data tables.

Willow Type A:	Leaves 3-4 in., very small bud scars, fine serrations
Type B:	Leaves 3-4 in., large brownish bud scars, conspicuous serrations
Type C:	Leaves 1-2½ in., dark prominent bud scars, conspicuous serrations
Type D:	Leaves 2-3 in., small bud scars, blunt serrations

Note: Gleason and Cronquist identify 31 *Salix* sp. but note that "the species are difficult to define and probable hybrids abound."

[3]Ash species were sometimes difficult to distinguish. The following characteristics were primarily used for identification along with fruit characteristics as described by Gleason and Cronquist:

White ash:	Leaflets 5-9, twigs and leaves mostly glabrous
Green ash:	(Including Red ash) leaflets 5-9, twigs and leaves densely pubescent to less often glabrous
Black ash:	Leaflets 7-11, sessile, conspicuously serrate

were taken from quack grass, reed canary grass, timothy, and smooth brome; stems and flower heads were discarded. The leaves, stems, and flower heads of Kentucky bluegrass, red top, and white sweet clover were collected. Milkweed samples consisted of leaf material.

Samples were collected from exposed sides of shrubs, and the leaf material (with petioles) were saved for analysis. Pole pruners were used to sample the exposed foliage of hardwoods at an average height of 20 to 25 feet above the ground. At least four separate branches were clipped from each tree. Leaves and petioles were saved for analysis; only leaflets and petiolules were saved from ash species and bitternut hickory (the "woody" petioles and rachis were discarded).

Laboratory Preparation and Chemical Analysis: Native Vegetation Species

Laboratory preparation. Four samples of each species were collected at each site when possible. Each sample was placed in a brown paper bag listing species name, site number, individual sample number (1, 2, 3, or 4), and date of collection. Small tags with the same information were placed in the bag with the sample.

Sample bags were taken to a temporary field lab at the St. Regis Village School and air-dried for 3 to 4 days. They were then sent to the University of Montana Environmental Studies Laboratory (EVST) in Missoula for analysis. Once in the EVST Lab, the dried, unwashed samples were placed in forced air ovens (70°-90°F) for 72 to 96 hours to complete the drying process. Dried samples were ground through a 20-mesh wire screen, placed in labeled 13-dram vials with the original identification tag, and stored until analysis.

Chemical analysis for fluorides (specific ion electrode method). The method of fluoride (F^-) analysis employed in this study was developed in 1970 at the University of Montana Environmental Studies Laboratory. This method incorporates the Orion fluoride specific ion electrode as the fluoride sensor. It is precise and rapid, and comparative results using other techniques are in agreement.

For each sample, 0.50 g of dried, ground plant material was placed in a 35-ml nickel crucible with 0.05 g of low fluorine calcium oxide and slurried with distilled water. The slurry was first dried and then charred under infrared, transferred to a muffle furnace, and ashed overnight at 600^{o}C. The crucibles were covered during ashing.

When the crucibles were cool, the ash was moistened with distilled water, dissolved in a minimum of 30 percent perchloric acid, made to 100 ml with 50 percent TISAB, transferred to a plastic beaker, placed on a magnetic stirrer, and the electrodes inserted into the stirred solution. The solution was insulated from the heat of the stirrer with a half inch of sponge, and the millivolt (MVO) reading was recorded after the electrodes had equilibrated.

Immediately prior to sample analysis, the electrodes were calibrated with standard solutions of the following fluoride concentrations: 0.05, 0.10, 0.50, 1.0, 5.0, 10.0, and 19.0 ppm. The preparation of the calibration curve and the calculation of the fluoride concentration of unknown samples were computerized.

Samples with fluoride concentrations falling below the useful range of the calibration curve were treated by adding sufficient fluoride to bring them into the millivolt range. For most plant materials, 1 ml of a 5.0 ppm F solution was sufficient. Reagent blanks (CaO) were carried through the entire procedure with each series of samples in order to determine any fluoride contamination in the procedure.

Samples of known fluoride concentrations were run daily to insure precision and accuracy of the method.

Collaborative analyses. After chemical analysis of samples from the first collection (summer), 14 samples were chosen at random (fluoride values ranging from 0.9 to 482 ppm), split and stored in duplicate vials, and sent to the WARF Institute in Madison, Wisconsin for collaborative fluoride analysis.* These results are presented in Table 15.

Field Sampling Procedures: Small Mammals

Small mammals were trapped on three sites on the St. Regis Reserve and one control site. At each site, one hundred standard snap-traps were baited with string soaked in bacon grease, and trap-lines were set in the designated fields; the traps were set out in the evening and checked in the morning. All specimens were labeled with the date of collection and site number, stored in plastic whirl-paks and frozen for transport back to the EVST Lab. Once in the lab, the animals were identified by species, weighed, and the femurs (with epiphyses) were removed.

Composite forage and grass samples for comparative purposes were also collected at each site and analyzed for fluoride using the same method as described for native vegetation species.

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*WARF Institute, now called "Raltech Scientific Services, Inc." P. O. Box 7545, Madison, Wisconsin 53707.

Table 15

RESULTS OF COLLABORATIVE ANALYSES FOR

FLUORIDE LEVELS IN VEGETATION

	Ppm Fluoride	
Sample Description	Environmental Studies Missoula, Montana	WARF Institute Madison, Wisconsin
Green ash leaves SRC-4	1.2	0.8
Elm leaves SRC-4	2.2	1.4
Basswood leaves SR9-4	11.7	6.6
Manitoba maple leaves SR6-1	18.0	19.3
Milkweed leaves SR5-4	25.3	19.8
Timothy leaves F2-8	30.0	34.1
Dogwood leaves SR3-3	39.7	41.6
Sumac leaves SR8-4	51.4	63.8
Dogwood leaves SR8-4	80.5	78.6
Milkweed leaves SR8-3	117.1	141
Wild grape SR10-1	123.4	148
Red maple SR7A-1	150.2	133
B. hickory SR6-1	214.7	294
B. hickory SR8-2	482 (R=453)*	712 (R=655)*

*R = repeat analysis

The purpose of the trapping effort was to determine levels of fluoride accumulations in small mammals from areas various distances from Reynolds. Sample sizes varied widely, and no attempt was made to determine population sizes, distributions, age of animals, or variations of fluoride content between males and females.

Laboratory Preparation and Chemical Analysis: Small Mammals

Laboratory preparation. The whole femur bones were defleshed by hand as thoroughly as possible and then boiled in Alconox for 2 hours, or until all remaining flesh was removed from the surface of the bone.

After drying the bones in a vacuum oven, they were defatted by boiling with at least six changes of petroleum ether and again dried.

Chemical analysis of fluorides. One of each pair of dry femurs was weighed, placed in a nickel crucible and ashed at 600^{o}C overnight. The crucibles were removed from the muffle furnace, cooled in a dessicator and the ash was dissolved in 2 ml of 20% hydrochloric acid.

The dissolved ash was diluted to 100 ml with 50% TISAB in distilled water and the fluoride activity was determined with a specific ion electrode.

The specific ion electrode was calibrated daily with standard solutions containing fluoride concentrations ranging from 0.01 ppm to 19.0 ppm. The electrode response in millivolts of the standard solutions and unknown solutions, and the weight of each sample were entered into a computer program to calculate concentrations of fluoride in the unknown samples. Fluoride concentrations were reported on a dry, fat-free basis.

Chapter 7

RESULTS - NATIVE VEGETATION SPECIES

The native vegetation sites sampled in the St. Regis Reserve vicinity during the summer and fall of 1977 are listed in Table 16 and graphically illustrated in Figures 3 and 4. Parts per million (ppm) mean fluoride concentrations, sample standard errors (SE) and sample size (n) are presented by species in this chapter. Appendix B contains a master list of all data collected within each site. This master list includes a few miscellaneous species of vegetation which were collected only rarely, and for which little comparative data was generated (quack grass, reed canary grass, and smooth brome grass; willow species and a variety of oaks).

Results - Grasses

The mean ppm fluoride content and the sample standard errors of the predominant grass species from the vicinities of Reynolds and Alcoa are presented in Table 17. Figure 5 graphically illustrates the mean fluoride content in timothy leaves (*Phleum pratensis* L.), the most consistently available grass species, from the summer collection. Timothy leaves were only sampled from three of the sites during the fall (second collection) and no other grass species were collected at that time.

Table 16

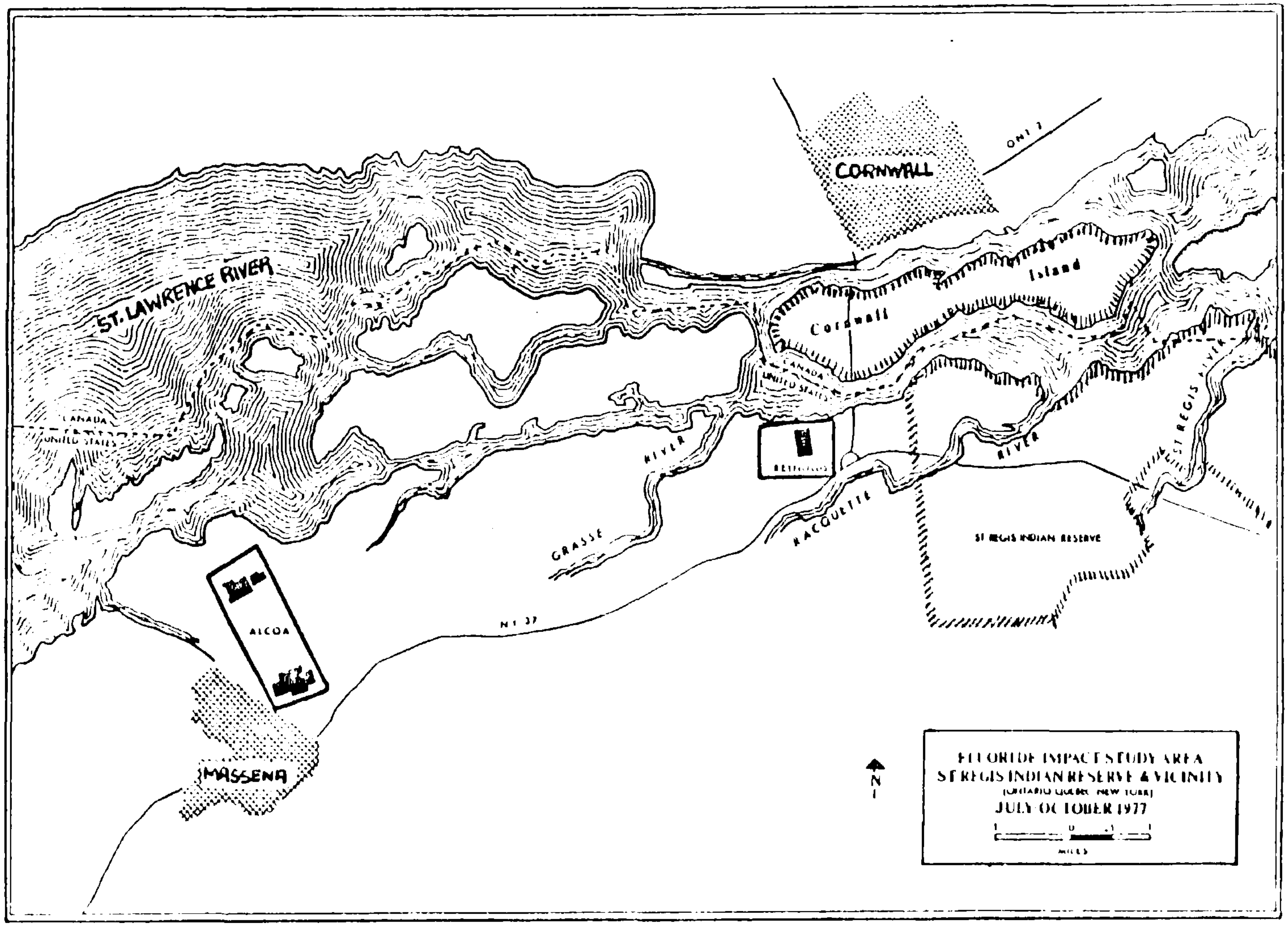
NATIVE VEGETATION COLLECTION SITES AROUND REYNOLDS AND ALCOA

Site Number	Distance/Direction From Reynolds[1]	Household	Location
Native Vegetation Sites			
*SR 1	1.0 mi, 45° NE		Cornwall Is., Ontario
*SR 2	1.5 mi, 30° NE		Cornwall Is., Ontario
*SR 3	4.8 mi, 70° ENE		St. Regis Village, Quebec
*SR 4	1.7 mi, 60° ENE		Franklin Co., NY
*SR 5	3.6 mi, 70° ENE		Cornwall Is., Ontario
*SR 6	2.3 mi, 70° ENE	W. White	Franklin Co., NY
SR 7	0.7 mi, 105° SE		St. Lawrence Co., NY
SR 9	1.4 mi, 300° NW		Barnhart Island, NY
*SR 10	0.9 mi, 20° NNE		Cornwall Is., Ontario
*SR 11	2.5 mi, 50° NE	E. Benedict	Cornwall Is., Ontario
SR 17	10 mi, NE		Ontario, Canada
SR 18	20 mi, NE		Ontario, Canada
SR 19	2.0 mi, 270° W		St. Lawrence Co., NY
*SR 21	1.4 mi, 40° NE	N. Point	Cornwall Is., Ontario
SR 23	0.6 mi, 65° NE		New York State
SR Control	75 mi, SW		New York State
Manitoba Maple			
*BX 1	1.3 mi, 15° NNE	F. Mitchell	Cornwall Is., Ontario
*BX 2	1.2 mi, N	J. Thompson	Cornwall Is., Ontario
*BX 3	1.6 mi, N	R. Seymour	Cornwall Is., Ontario
*BX 4	3.2 mi, 55° NE	A. Lazore	Cornwall Is., Ontario
*BX 5	3.6 mi, 115° SE	I. Papineau	Franklin Co., NY
BX 6	1.3 mi, 210° SW		St. Lawrence Co., NY
Control	75 mi SW		New York State

Site Number	Distance/Direction From Alcoa[1]	Household	Location
Native Vegetation Sites			
SR 8	2.0 mi, 35° NE		Barnhart Island, NY
SR 12	1.9 mi, 75° ENE		St. Lawrence Co., NY
SR 13	1.8 mi, 90° E		St. Lawrence Co., NY
SR 14	2.3 mi, 255° SW		St. Lawrence Co., NY
SR 15	0.7 mi, 50° NE		St. Lawrence Co., NY
SR 16	3.4 mi, 140° SE	Osmun	St. Lawrence Co., NY
*SR 22	4.0 mi, 30° NE		Sheek Island, Ontario
SR Control	75 mi SW		New York State

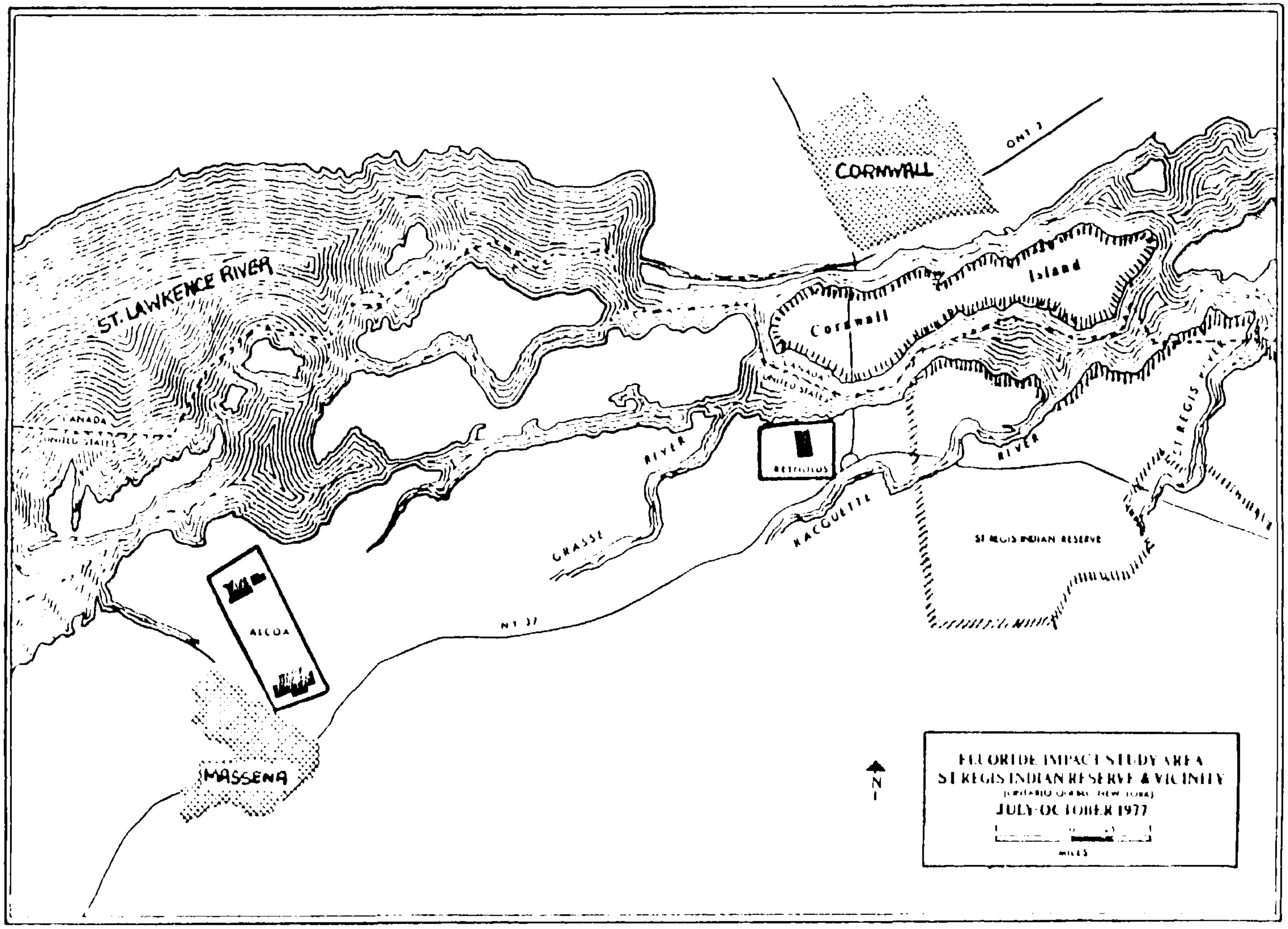
*Situated on the St. Regis Reserve
[1] Bearings from Reynolds (or Alcoa) to the collection area

Figure 3



Native Vegetation Collection Sites

Figure 4



Manitoba Maple Collection Sites

Table 17

MEAN FLUORIDE CONTENT (ppm) OF SELECTED GRASSES
SUMMER AND FALL COLLECTIONS 1977
SITES AROUND REYNOLDS METALS COMPANY

		Timothy		Kentucky Blue Grass	Red-Top	White Sweet Clover
		Summer	Fall	Summer	Summer	Summer
SR 10	0.9 mi NNE	100 *n=4 SE=19.6*		66 *n=4 SE=16.9*	52 *n=4 SE=16.3*	99 *n=4 SE=26.1*
SR 1	1.0 mi NE	132 *n=4 SE=3.9*		68 *n=4 SE=8.9*	49 *n=4 SE=5.2*	125 *n=4 SE=23.2*
SR 2	1.5 mi NE	52 *n=4 SE=9.3*	28 *n=1*		21 *n=4 SE=5.8*	
SR 4	1.7 mi ENE	25 *n=3 SE=1.3*		24 *n=4 SE=4.9*	17 *n=4 SE=1.6*	18 *n=4 SE=4.1*
SR 6	2.3 mi ENE	19 *n=2 SE=5.0*		5 *n=2 SE=1.8*		
SR 5	3.6 mi ENE	10 *n=4 SE=1.0*	2 *n=1*	7 *n=4 SE=0.3*	6 *n=4 SE=1.2*	15 *n=4 SE=1.4*
SR 3	4.8 mi ENE	8 *n=4 SE=0.4*		6 *n=4 SE=1.6*	5 *n=4 SE=0.6*	
SR 9	1.4 mi NW			7 *n=4 SE=1.5*	5 *n=4 SE=0.9*	8 *n=4 SE=1.3*

SE = sample standard error
n = number of samples

Table 17 (continued)

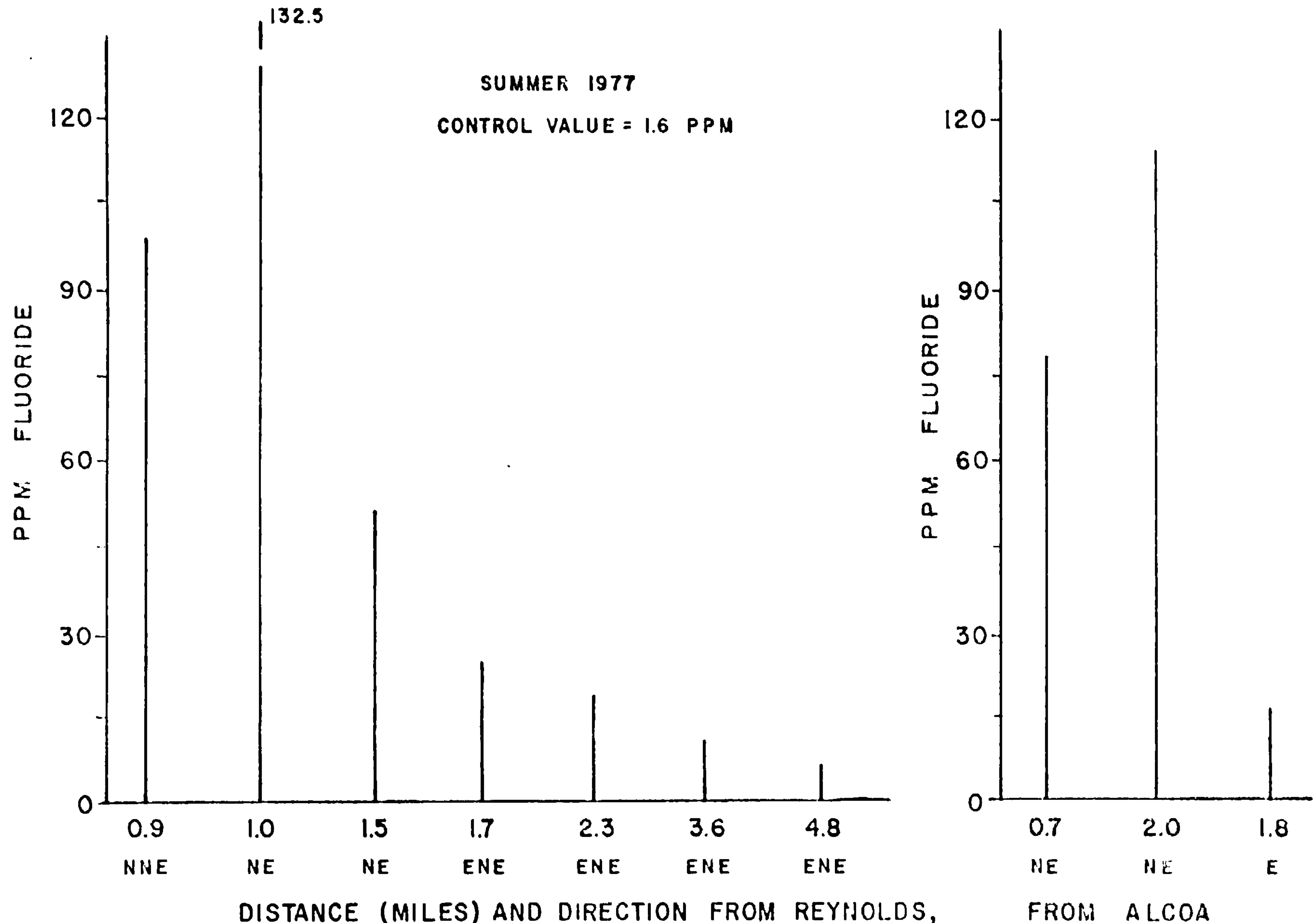
MEAN FLUORIDE CONTENT (ppm) OF SELECTED GRASSES
SUMMER AND FALL COLLECTIONS 1977
SITES AROUND ALCOA

		Timothy		Kentucky Blue Grass	Red-Top	White Sweet Clover
		Summer	Fall	Summer	Summer	Summer
SR 15	0.7 mi NE	79 *n=1*		16 *n=4 SE=2.6*	33 *n=4 SE=4.7*	
SR 8	2.0 mi NE	113 *n=4 SE=8.8*			45 *n=4 SE=5.3*	58 *n=4 SE=6.4*
SR 13	1.8 mi E	16 *n=4 SE=1.4*	24 *n=2 SE=8.0*	6 *n=4 SE=0.7*	6 *n=4 SE=0.7*	15 *n=4 SE=3.9*
SR 14	2.3 mi SW			4 *n=4 SE=0.7*		4 *n=4 SE=0.6*
Controls		1.6 *n=4 SE=0.1*	2.1 *n=2 SE=0.6*	1.3 *n=4 SE=0.[illegible]*	1.1 *n=4 SE=0.5*	0.9 *n=4 SE=0.[illegible]*

SE = sample standard error
n = number of samples

Figure 5

MEAN FLUORIDE CONTENT, UNWASHED TIMOTHY LEAVES, VICINITY OF REYNOLDS, ALCOA 1977



Grasses from sites northeast of both Reynolds and Alcoa had extremely high fluoride levels, 50 to 80 times higher than grasses from control areas.

In general, as the distance from Reynolds increased and directions deviated from the northeast quadrant, fluoride levels decreased at the sites around the facility. At the two sites northeast of Alcoa, however, this differed somewhat. Grass samples, and samples of several other native vegetation species were consistently lower at the site less than one mile northeast of Alcoa (SR 15) than samples from 2.0 miles northeast (SR 8) of the plant. In the Alcoa vicinity, due to the predominantly gaseous emissions, maximum fluoride deposition was noted at points farther away rather than adjacent to the source (as observed around Reynolds).

The various grass species within each site also varied considerably, illustrating differences in fluoride uptake rates. Fluoride levels in grasses from the sites around both Reynolds and Alcoa are comparable and can be considered elevated at all sites.

Very few timothy samples were able to be collected in the fall, but fluoride levels in the leaves of this species followed a pattern evident in several other species. The limited sampling of timothy at the sites around Reynolds showed the fluoride content to be considerably lower in the fall than in the summer, but at the one site 1.8 miles east of Alcoa (SR 13), means and standard errors showed that the fluoride content of timothy increased over the summer months.

Fluoride levels found in other grass species are listed in Table 17. Results show that levels in grasses from around both sources are comparable and can be considered elevated at all sites.

Results - Shrubs and Forbs

Table 18 presents the mean fluoride content and sample standard errors found in various species within the shrub and forb canopy layer during both sampling periods from sites around Reynolds and Alcoa. For comparative purposes, results of dogwood (Cornus stolonifera Michx.) samples taken in the fall from only the sites northeast of Alcoa and Reynolds (in the predominant wind path) are summarized in Table 19. Figure 6 illustrates the mean fluoride content of dogwood leaves (summer and fall collections) and Figure 7 the mean fluoride content of wild grape leaves.

Leaves of the common forb milkweed were sampled because it was found virtually everywhere on the Island and because some of the traditional Indian families still consider it of medicinal value, and therefore of importance to the Tribe (St. Regis Indians, personal communication, 1977). Milkweed from site SR 1, 1.0 miles northeast of Reynolds, contained a mean of 219 ppm (SE=50.7), and milkweed from the site 0.9 miles north-northeast of the plant (SR 10) contained 130 ppm (SE=5.7). Samples from the sites 1.5 miles northeast and 1.7 miles east-northeast contained a mean of 42 (SE=10.8) and 48 (SE=4.4) ppm fluoride, respectively, and samples from sites farther away or in other directions contained considerably less. However, it should be noted that milkweed samples from up to 20 miles northeast of Reynolds still

Table 18

MEAN FLUORIDE CONTENT (ppm) OF FORBS AND SHRUBS
SUMMER AND FALL COLLECTIONS 1977
SITES AROUND REYNOLDS METALS COMPANY

		Milkweed	Sumac	Wild Grape	Dogwood	
		Summer	Summer	Summer	Summer	Fall
SR 10	0.9 mi NNE	130 n=3 SE=5.7	249 n=4 SE=32.4	196 n=4 SE=47.8	168 n=2 SE=72.8	174 n=4 SE=36.2
SR 23	0.6 mi NE					389 n=1
SR 1	1.0 mi NE	219 n=4 SE=50.7	106 n=4 SE=11.3	142 n=3 SE=12.8	182 n=4 SE=16.7	118 n=4 SE=14.8
SR 21	1.4 mi NE					122 n=4 SE=3.9
SR 2	1.5 mi NE	52 n=4 SE=10.8	47 n=4 SE=9.2	40 n=4 SE=5.1		51 n=4 SE=5.9
SR 11	2.5 mi NE		47 n=4 SE=6.4			
SR 17	10 mi NE	14 n=1	8 n=4 SE=2.3	7 n=2 SE=0.7	26 n=4 SE=0.8	
SR 18	20 mi NE	16 n=3 SE=3.8		9 n=3 SE=1.7	18 n=4 SE=1.9	
SR 4	1.7 mi ENE	48 n=4 SE=1.4	24 n=4 SE=1.9	25 n=4 SE=4.3	42 n=2 SE=0.7	36 n=4 SE=3.3
SR 6	2.3 mi ENE	18 n=4 SE=1.4	15 n=4 SE=1.3	4 n=4 SE=0.8		21 n=4 SE=1.5
SR 5	3.6 mi ENE	25 n=3 SE=0.4	16 n=3 SE=0.3	23 n=2 SE=0.4	35 n=4 SE=5.2	16 n=4 SE=2.8
SR 3	4.8 mi ENE	10 n=4 SE=1.3	5 n=4 SE=0.7	4 n=4 SE=0.6	23 n=4 SE=5.7	14 n=4 SE=1.9
SR 7	0.7 mi SE					315 n=2 SE=26.5

(continued)

Table 18 (continued)

MEAN FLUORIDE CONTENT (ppm) OF FORBS AND SHRUBS
SUMMER AND FALL COLLECTIONS 1977

Sites Around Reynolds Metals Company

		Milkweed	Sumac	Wild Grape	Dogwood	
		Summer	Summer	Summer	Summer	Fall
BX 5	3.6 mi SE					10 n=2 SE=1.8
BX 6	1.3 mi SW					22 n=3 SE=1.6
SR 9	1.4 mi NW	10 n=4 SE=1.9	6 n=4 SE=0.9	6 n=4 SE=0.9	16 n=2 SE=3.5	27 n=4 SE=[illegible].3
Sites Around Alcoa						
SR 15	0.7 mi NE	68 n=4 SE=10.8	61 n=4 SE=8.5	11 n=4 SE=[illegible].8	117 n=4 SE=13.0	143 n=4 SE=[illegible]2.8
SR 8	2.0 mi NE	115 n=4 SE=18.9	71 n=4 SE=9.[illegible]	64 n=4 SE=1[illegible].3	119 n=4 SE=[illegible].3	127 n=4 SE=1[illegible].3
SR 22	4.0 mi NE					46 n=4 SE=4.5
SR 12	1.9 mi ENE		25 n=4 SE=4.7			
SR 13	1.8 mi E	13 n=4 SE=2.9		8 n=4 SE=1.9	13 n=4 SE=2.5	34 n=4 SE=4.1
SR 16	3.4 mi SE	5 n=4 SE=0.4		3 n=4 SE=0.7	12 n=4 SE=1.9	
SR 14	2.3 mi SW	10 n=4 SE=2.5	3 n=4 SE=0.7	3 n=4 SE=0.5		31 n=4 SE=3.4
Control		0.9 n=1	0.9 n=4 SE=0.3	1.6 n=4 SE=0.3	2.1 n=4 SE=0.5	3.3 n=4 SE=0.5

SE = sample standard error
n = number of samples

Table 19

MEAN FLUORIDE CONTENT (ppm) OF DOGWOOD LEAVES
COMPARISON OF SITES NORTHEAST OF ALCOA AND REYNOLDS
FALL COLLECTION 1977

Northeast of Alcoa		Northeast of Reynolds	
0.7 mi NE	143.2 *(SE=22.8)*	0.6 mi NE	389.5 *(n=1)*
2.0 mi NE	127.4 *(SE=12.3)*	1.0 mi NE	117.7 *(SE=14.8)*
4.0 mi NE	45.8 *(SE=4.5)*	1.4 mi NE	121.9 *(SE=5.2)*
		1.5 mi NE	51.5 *(SE=5.3)*

SE = sample standard error
n = number of samples

Figure 6

MEAN FLUORIDE CONTENT, UNWASHED DOGWOOD FOLIAGE, VICINITY OF REYNOLDS, ALCOA 1977

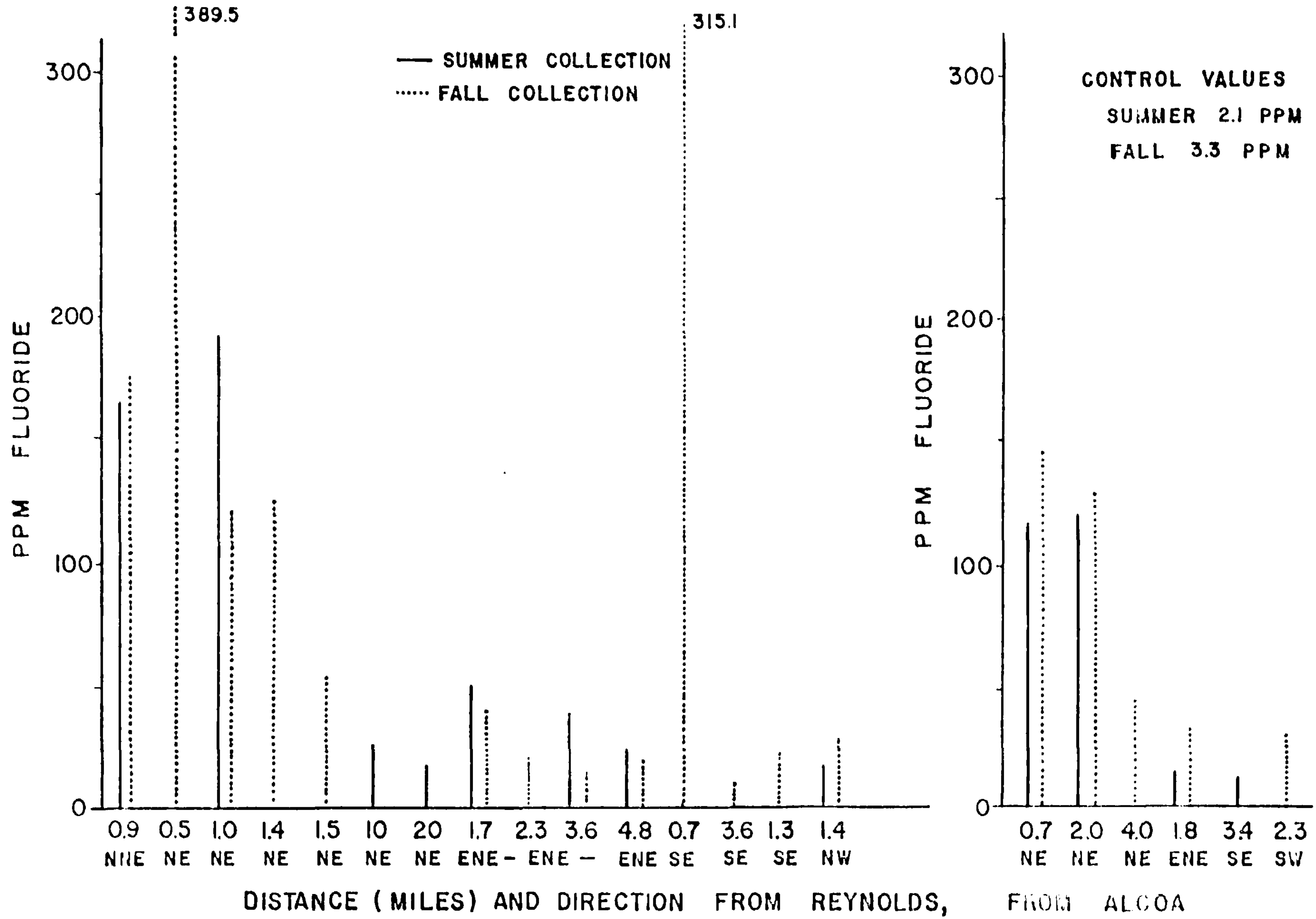
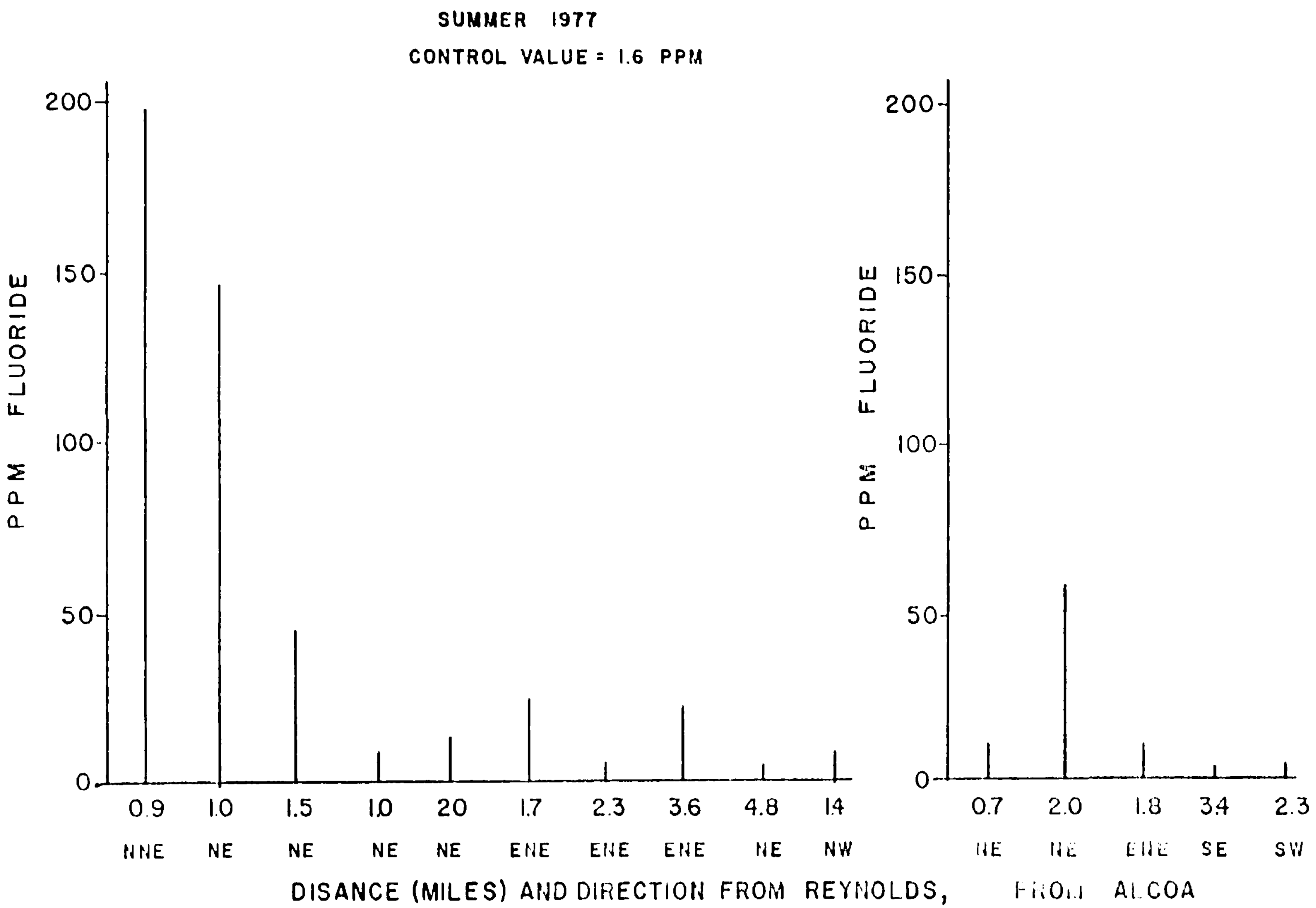


Figure 7

MEAN FLUORIDE CONTENT, UNWASHED WILD GRAPE FOLIAGE, VICINITY OF REYNOLDS, ALCOA 1977



contained more than 15 times the fluoride concentrations found within the control site.

Milkweed from areas around Alcoa average 68 ppm (SE=10.8), 0.7 miles northeast, and 115 ppm (SE=18.9), 2.0 miles northeast. As noted with the grasses, the highest levels were found at the site 2.0 miles from Alcoa rather than adjacent to the source, in contrast to the maximum values noted adjacent to Reynolds.

Accumulation of fluorides in sumac leaves followed a pattern similar to milkweed. The highest concentrations were found 0.9 miles north-northeast of Reynolds, where the average fluoride content was 249 ppm (SE=32.4). As the distance of site locations increased from the sources and directions deviated from the northeast quadrant, mean fluoride contents decreased. Concentrations within the sites northeast of Alcoa were 60 to 70 times higher than those noted within the control area.

Wild grape (*Vitis riparea* Michx.) leaves, found at the shrub level in natural habitats, are extremely sensitive to fluoride damage and have often been used by the Ontario Ministry of the Environment personnel as visual indicators of fluoride injury in the St. Regis area (Pearson, 1978). As seen from the graph illustrating mean concentrations (Figure 7), samples from within the sites northeast of each source had the highest fluoride accumulations and levels decreased as distances from the source increased.

Red-osier dogwood leaves proved to be very efficient scavengers of fluorides and this was the only species from the shrub and forb canopy layer collected during the fall. Results of these collections from the sites around Reynolds showed that fluoride levels were generally lower in the fall than summer, as noted from the means and sample standard errors. The exceptions were at the site 0.9 mile north-northeast of the plant, where levels were comparable, and at the site 1.4 miles northwest of Reynolds where levels increased. Also, several new sites were added around Reynolds in the fall. Dogwood leaves sampled in the fall from the northeast corner of Reynolds' property (less than 1 mile from the facility) averaged 389 ppm (n=1), 315 ppm (SE=26.5) from 0.7 mile southeast of Reynolds, and 122 ppm (SE=3.2) from 1.4 miles northeast of Reynolds.

Concentrations in dogwood samples from around Alcoa collected in the fall were comparable to the levels noted in the summer despite heavy rains during the interval. Table 19 illustrates that dogwood sampled in the fall from the two sites within two miles northeast of Alcoa had comparable, elevated fluoride concentrations with levels decreasing significantly at 4.0 miles northeast.

Results - Deciduous Trees

Mean fluoride contents of several species of maples (*Acer rubrum* L., *Acer saccharum* Marsh., and *Acer saccharinum* L.) sampled from areas around Reynolds and Alcoa during the summer collection period are shown in Table 20. Samples from the most impacted areas (SR 1 and SR 10, within 1 mile of Reynolds, on Cornwall Island) and SR 7 (0.7 mile southeast of Reynolds) had exceedingly high fluoride levels. The highest concentration observed was found at site SR 10 (0.9 mile northeast), where leaves of red maple had a mean fluoride content of 251 ppm (n=1). As noted with other native vegetation species, fluoride levels in the

Table 20

MEAN FLUORIDE CONTENT (ppm) UNWASHED RED, SILVER,
SUGAR MAPLE FOLIAGE
SUMMER COLLECTION 1977

Sites Around Reynolds

		Red Maple	Sugar Maple	Silver Maple
SR 10	0.9 mi NNE	251 *n*=1		
SR 1	1.0 mi NE	91 *n*=2 *SE*=12.9		
SR 2	1.5 mi NE		21 *n*=2 *SE*=7.7	60 *n*=2 *SE*=25.1
SR 11	2.5 mi NE		22 *n*=4 *SE*=2.8	
SR 17	10 mi NE			13 *n*=3 *SE*=0.4
SR 5	3.6 mi ENE			21 *n*=3 *SE*=0.7
SR 7	0.7 mi SE	106 *n*=5 *SE*=13.6	104 *n*=3 *SE*=22.7	
SR 19	2.0 mi W	11 *n*=4 *SE*=2.2		

Sites Around Alcoa

		Red Maple	Sugar Maple	Silver Maple
SR 8	2.0 mi NE		29 *n*=4 *SE*=1.3	
SR 13	1.8 mi E		4 *n*=4 *SE*=1.3	
Control		2 *n*=4 *SE*=0.4	0.9 *n*=4 *SE*=0.4	0.8 *n*=2 *SE*=0.3

SE = sample standard error
n = number of samples

maples generally decreased with increasing distances and deviations from the northeast quadrant around each facility.

Results of analysis of basswood foliage (Tilia americana L.) from sites around Reynolds and Alcoa during the summer and fall are presented in Table 21. Excessively high values were found at sites less than one mile from Reynolds on Cornwall Island and at the site 0.7 mile southeast of Reynolds. The mean fluoride content of basswood leaves from the control areas was 0.6 ppm (SE=0.2); values found around Reynolds represent concentrations from 200 to 400 times higher and around Alcoa 100 times higher than these observed values.

Results of fluoride analysis of cottonwood (Populus deltoides Marsh.), quaking aspen (Populus tremuloides Michx.), and elm (Ulmus americana L.) foliage from the summer collection are presented in Tables 22 and 23. Highest concentrations were again found to the northeast of Reynolds and Alcoa, and southeast of Reynolds. Fluoride content of foliage from the most impacted areas around Reynolds were higher than in areas around Alcoa. Comparison of the sites 0.7 mile northeast of Alcoa and two miles northeast of Alcoa again demonstrate the pattern of higher concentrations noted farther away, rather than in adjacent areas, as was consistently noted in the samples collected around Reynolds.

The results of fluoride analysis of foliage from Bitternut hickory (Carya cordiformis Wang.) leaves from both the summer and fall collections are presented in Table 24. This species was found to contain excessively high levels of fluorides at all sites, but it was also noted that samples from the control area contained an average of

Table 21

MEAN FLUORIDE CONTENT (ppm) UNWASHED BASSWOOD FOLIAGE
SUMMER COLLECTION 1977

Sites Around Reynolds

Location	ppm F
SR 10 0.9 mi NNE	160 *n=4 SE=16.3*
SR 1 1.0 mi NE	259 *n=2 SE=30.6*
SR 2 1.5 mi NE	37 *n=4 SE=2.6*
SR 17 10 mi NE	15 *n=5 SE=2.2*
SR 6 2.3 mi ENE	13 *n=8 SE=1.5*
SR 5 3.6 mi ENE	11 *n=1*
SR 7 0.7 mi SE	153 *n=8 SE=17.7*
SR 9 1.4 mi NW	12 *n=2 SE=0.8*
Control	0.6 *n=4 SE=0.2*

Sites Around Alcoa

Location	ppm F
SR 8 2.0 mi NE	65 *n=4 SE=13.0*
SR 12 1.9 ENE	16 *n=4 SE=2.1*
Control	0.6 *n=4 SE=0.2*

SE = sample standard error
n = number of samples

Table 22

MEAN FLUORIDE CONTENT (ppm) UNWASHED COTTONWOOD
AND QUAKING ASPEN FOLIAGE
SUMMER COLLECTION 1977

Sites Around Reynolds: Location	Cottonwood	Aspen	Sites Around Alcoa: Location	Cottonwood	Aspen
SR 10 0.9 mi NNE	65 *n=4 SE=9.1*				
SR 1 1.0 mi NE	161 *n=4 SE=24.8*	157 *n=4 SE=37.4*	SR 8 2.0 mi NE	33 *n=2 SE=9.4*	80 *n=8 SE=22.8*
SR 4 1.7 mi ENE	25 *n=4 SE=2.4*	28 *n=4 SE=0.4*	SR 12 1.9 mi ENE	14 *n=1*	
SR 6 2.3 mi ENE		18 *n=4 SE=1.1*			
SR 5 3.6 mi ENE	19 *n=4 SE=0.5*	19 *n=4 SE=1.5*			
SR 7 0.7 mi SE		106 *n=2 SE=8.6*			
SR 9 1.4 mi NW	12 *n=4 SE=1.4*				
Control	1.4 *n=3 SE=0.5*	1.1 *n=3 SE=0.6*	Control	1.4 *n=3 SE=0.5*	1.1 *n=3 SE=0.6*

SE = sample standard error
n = number of samples

Table 23

MEAN FLUORIDE CONTENT (ppm) UNWASHED ELM FOLIAGE
SUMMER COLLECTION 1977

Sites Around Reynolds

Location		ppm F	n	SE
SR 1	1.0 mi NE	158	1	
SR 2	1.5 mi NE	53	4	7.8
SR 17	10 mi NE	11	4	0.9
SR 18	20 mi NE	7	4	1.6
SR 4	1.7 mi ENE	62	1	
SR 6	2.3 mi ENE	19	1	
SR 7	0.7 mi SE	85	2	34.5
SR 19	2.0 mi W	17	2	3.5
Control		1.9	4	0.4

Sites Around Alcoa

Location		ppm F	n	SE
SR 15	0.7 mi NE	52	4	5.1
SR 8	2.0 mi NE	71	4	16.8
SR 12	1.9 mi ENE	11	4	1.5
SR 13	1.8 mi E	8	3	2.2
SR 16	3.4 mi SE	2	4	0.4
SR 14	2.3 mi SW	5	4	0.5
Control		1.9	4	0.4

SE = sample standard error
n = number of samples

Table 24

MEAN FLUORIDE CONTENT (ppm) UNWASHED BITTERNUT HICKORY FOLIAGE
SUMMER AND FALL COLLECTIONS 1977

Sites Around Reynolds			Sites Around Alcoa		
Location	Summer	Fall	Location	Summer	Fall
SR 21 1.4 mi NE		705 *n=2 SE=199.8*	SR 8 2.0 mi NE	500 *n=3 SE=17.3*	650 *n=2 SE=150.8*
SR 6 2.3 mi ENE	207 *n=2 SE=7.4*	279 *n=2 SE=49.6*	SR 12 1.9 mi ENE	398 *n=1*	648 *n=2 SE=64.5*
SR 9 1.4 mi NW	246 *n=1*	291 *n=3 SE=23.5*			
Control		161 *n=4 SE=16.7*	Control		161 *n=4 SE=16.7*

SE = sample standard error
n = number of samples

161 ppm (SE=16.7) fluoride, which is much higher than any other species from an unpolluted habitat. Because this species apparently contains high levels of naturally-occurring fluorides, it is probably not an accurate indicator of ambient fluoride contamination.

Other high accumulators of ambient fluorides in the St. Regis area were the three ash species (genus Fraxinus), two of which ("green" and "white" ash) were particularly abundant in the area. These results are presented in Table 25. Extensive sampling of ash species during the fall collection yielded fluoride values ranging from 3 ppm (SE=0.3) at the site 1.4 miles northwest of Reynolds, out of the predominant plume path, to exceedingly high (greater than 300 ppm) at the more impacted site just southeast of the source. Because only one species of ash usually was found at each site, it was difficult to delineate which, if any, of the species accumulated the greatest amount of fluorides. Based on the available data and the sample standard errors from sites SR 21 (1.4 miles northeast of Reynolds) and SR 7 (0.7 mile southeast of Reynolds), where more than one species was sampled, it appears that black ash may be the highest accumulator. Black ash is used by the Indians in basket-making,and because this wood is in great demand, very little black ash was found in the area.

During the summer, the highest fluoride levels in ashes were found within sites northeast of each fluoride source and within the site just southeast of Reynolds. Because of the very limited data within each species, however, no consistent pattern was noted in the fluoride accumulations between the summer and fall collections.

Table 25

MEAN FLUORIDE CONTENT (ppm) UNWASHED FOLIAGE - ASH SPECIES
SUMMER AND FALL COLLECTIONS 1977

Sites Around Reynolds

Location		White Ash		Green Ash		Black Ash	
		Summer	Fall	Summer	Fall	Summer	Fall
BX 3	1.6 mi N		7 *n=3 SE=0.9*				
SR 10	0.9 mi NNE		111 *n=1*			128 *n=1*	
BX 1	1.3 mi NNE		20 *n=4 SE=2.4*				
SR 23	0.6 mi NE				270 *n=3 SE=41.8*		
SR 1	1.0 mi NE		367 *n=1*				
SR 21	1.4 mi NE		106 *n=2 SE=16.4*		153 *n=4 SE=12.8*		176 *n=3 SE=7.4*
SR 2	1.5 mi NE		26 *n=4 SE=5.5*		25 *n=1*		
SR 18	20 mi NE			6 *n=4 SE=0.3*			
SR 6	2.3 mi ENE		30 *n=2 SE=1.4*				
SR 5	3.6 mi ENE			21 *n=1*	12 *n=4 SE=4.6*		
SR 7	0.7 mi SE			78 *n=8 SE=14.3*	294 *n=4 SE=32.6*	63 *n=1*	355 *n=1 SE=46.4*
BX 6	1.3 mi SW				13 *n=4 SE=3.4*		
SR 9	1.4 mi NW	3 *n=2 SE=0.3*	8 *n=2 SE=1.1*		10 *n=4 SE=2.0*		

(continued)

Table 25 (continued)

MEAN FLUORIDE CONTENT (ppm) UNWASHED FOLIAGE - ASH SPECIES
SUMMER AND FALL COLLECTIONS 1977

Sites Around Alcoa

Location		White Ash		Green Ash		Black Ash	
		Summer	Fall	Summer	Fall	Summer	Fall
SR 15	0.7 mi NE			45 *n=4 SE=5.3*	102 *n=4 SE=14.9*		
SR 8	2.0 mi NE		149 *n=5 SE=31.4*				
SR 22	4.0 mi NE		23 *n=4 SE=2.0*		22 *n=4 SE=0.8*		
SR 12	1.9 mi ENE	29 *n=1*	20 *n=1*				
SR 16	3.4 mi SE			3 *n=4 SE=0.5*			
SR 14	2.3 mi SW				25 *n=4 SE=5.0*		
Control			1.5 *n=4 SE=0.3*	0.9 *n=4 SE=0.2*	2.6 *n=4 SE=0.1*	1.9 *n=1*	4.8 *n=4 SE=0.6*

SE = sample standard error
n = number of samples

Figure 8 graphically illustrates the mean fluoride content of ash species from the fall collection. Elevated levels were found southwest of both plants, where the fluoride content of green ash 2.3 miles from Alcoa was almost twice as high as green ash 1.3 miles from Reynolds. Again, the highest levels were found within one mile northeast and southeast of Reynolds and two miles northeast of Alcoa.

Site SR 22, a new collection area during the fall, was located on Sheek Island, four miles northeast of Alcoa. Because elevated fluoride levels were observed two miles northeast of Alcoa on Barnhart Island during the summer within several species, an attempt was made to determine the extent of emissions in the same wind path at areas even further away. Results of ash sampling on Sheek Island indicated that there were still elevated levels of fluoride in the vegetation, but the means and sample standard errors within each site support the fact that at four miles northeast of Alcoa ambient concentrations were significantly lower than those observed at the impacted Barnhart Island site two miles from Alcoa.

The species most commonly sampled by other agencies in the Cornwall Island - St. Regis Reserve area has been Manitoba maple, or "boxelder" (<u>Acer negundo</u> L.). Fifteen sites were sampled in this study for Manitoba maple in the fall, several of which had been sampled by the Ontario Ministry of the Environment (OME) also during 1977. Results of the fluoride analysis of this species in the fall (representative of the total growing season accumulation) are presented in Table 26 along with results of OME's analyses of fluoride content at the same or comparable sites (Pearson, 1978). OME presents average fluoride

Figure 8

MEAN FLUORIDE CONTENT, UNWASHED ASH SPECIES FOLIAGE, VICINITY OF REYNOLDS, ALCOA 1977

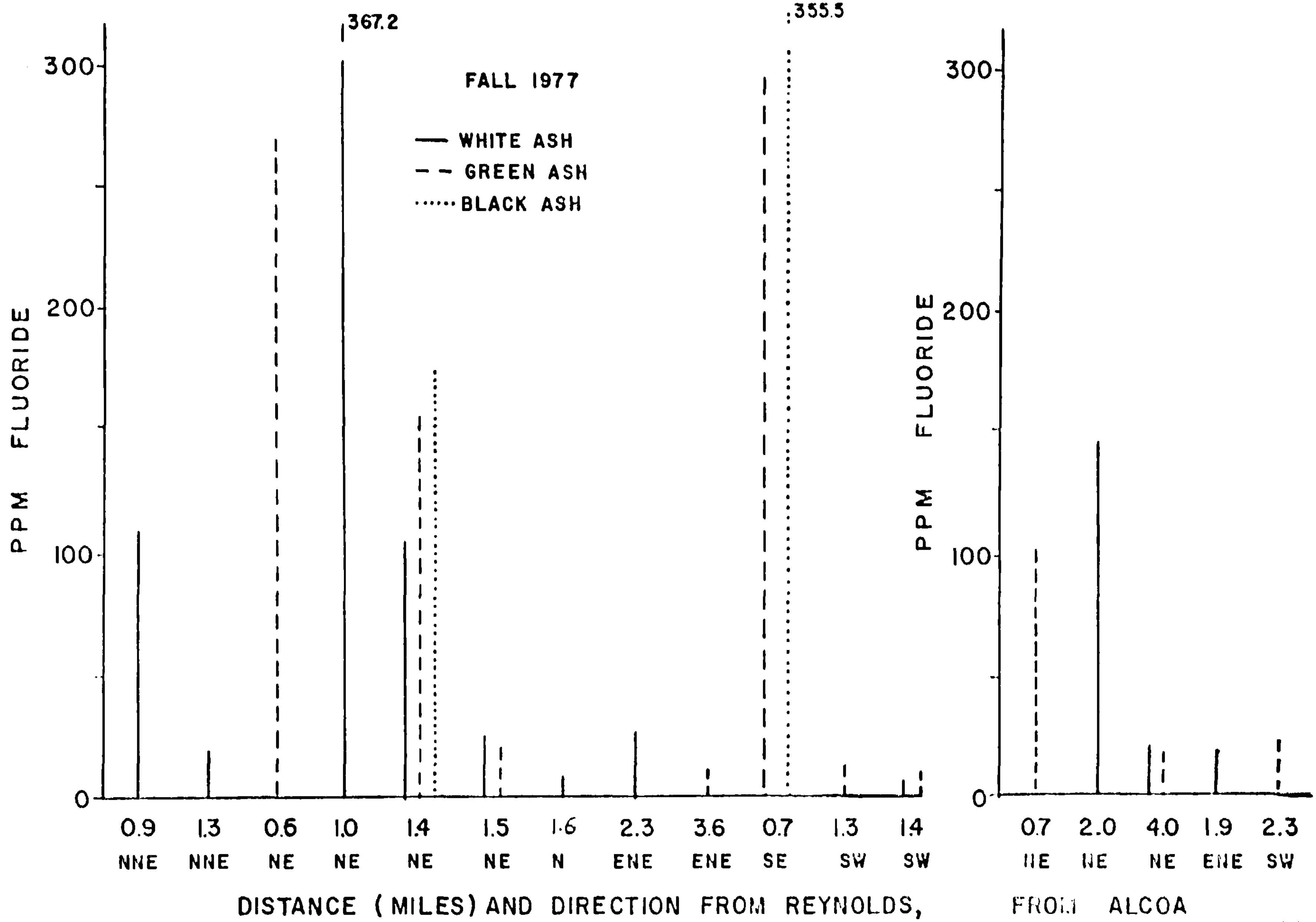


Table 26

MEAN FLUORIDE CONTENT (ppm) UNWASHED MANITOBA MAPLE FOLIAGE
SUMMER AND FALL COLLECTIONS 1977

Sites Around Reynolds

Location		Miles' Data: Summer (July-Aug)	Miles' Data: Fall (Sept-Oct)	OME Data: Approximate Location	OME Data: June	OME Data: July	OME Data: August	OME Data: Seasonal Mean
BX 2	1.2 mi N		28 *n=4 SE=2.2*	(1.2 mi N)	8	12	45	22
BX 3	1.6 mi N		21 *n=7 SE=2.5*	(1.6 mi N)	12	10	14	12
BX 1	1.3 mi NNE		47 *n=4 SE=2.4*					
SR 1	1.0 mi NE		265 *n=1*	(0.9 mi NE)	103	506	750	453
SR 2	1.5 mi NE		49 *n=3 SE=6.7*	(1.9 mi NE)	19	93	217	110
SR 11	2.5 mi NE		59 *n=4 SE=14.1*	(2.6 mi NE)	23	38	56	39
BX 4	3.2 mi NE		25 *n=4 SE=2.6*	(4.3 mi NE)	39	59	37	45
SR 17	10 mi NE	9 *n=4 SE=0.9*						
SR 4	1.7 mi ENE		73 *n=4 SE=3.8*					
SR 6	2.3 mi ENE	20 *n=3 SE=0.8*	35 *n=3 SE=2.3*					
SR 5	3.6 mi ENE		18 *n=4 SE=1.8*					
SR 3	4.8 mi ENE		11 *n=4 SE=1.8*					
BX 5	3.6 mi SE		12 *n=2 SE=0.1*					
BX 6	1.3 mi SW		24 *n=4 SE=3.2*					
Control		2.0 *n=4 SE=0.2*	2.9 *n=4 SE=0.3*					
Sites Around Alcoa								
SR 14	2.3 mi SW	5 *n=4 SE=0.6*	39 *n=4 SE=3.2*					

SE = sample standard error
n = number of samples

levels by month (June, July, August) and the growing season mean based on these three analyses; no sample sizes or standard errors were reported. Figure 9 graphically illustrates this author's results and OME's growing season averages of these observed fluoride accumulations.

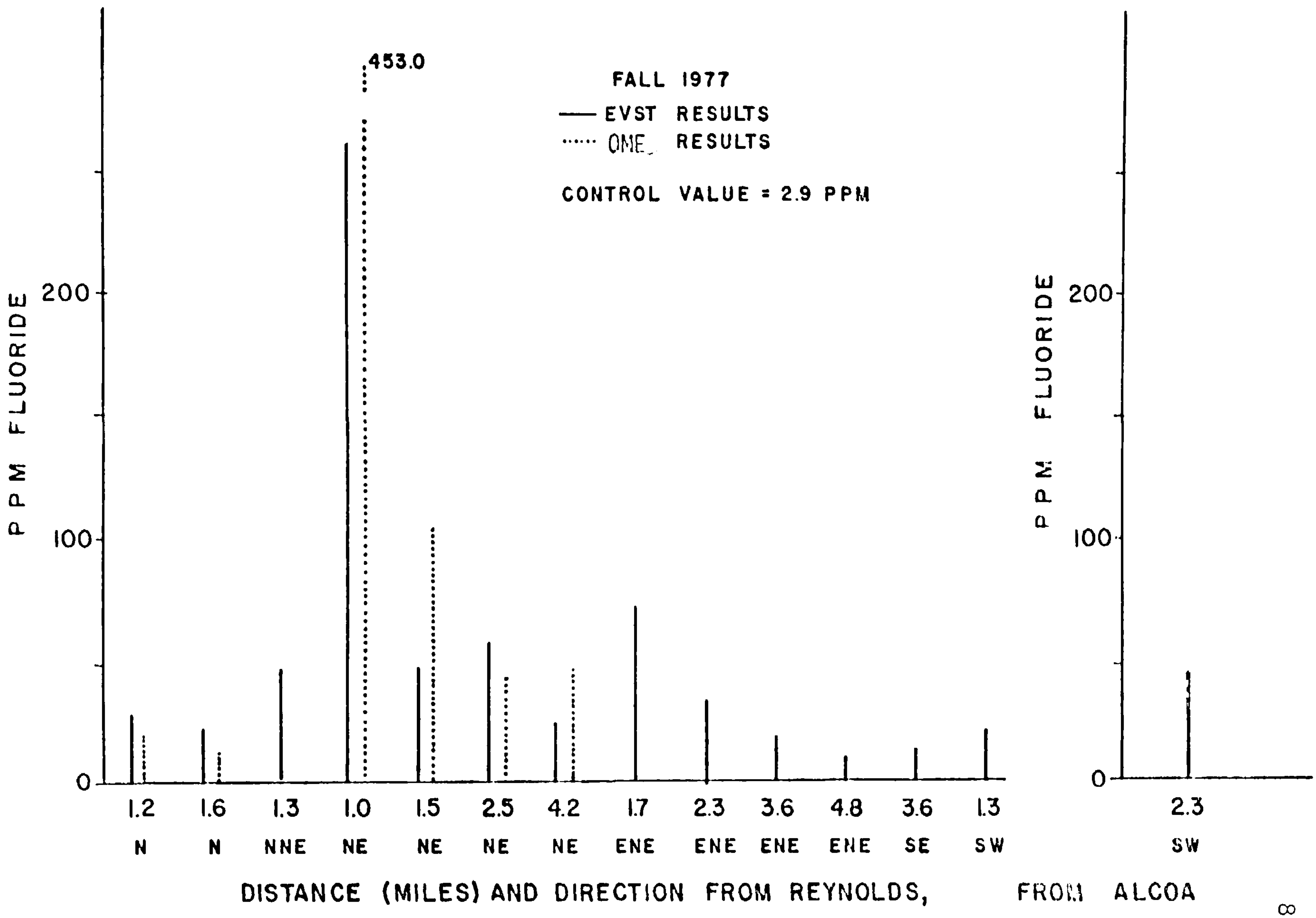
Manitoba maple was found at only one site around Alcoa, 2.3 miles southwest of the plant. The fluoride concentration of Manitoba maple at this site was 39 ppm (SE=3.2). These results are comparable with Manitoba maple at the site half the distance southwest of Reynolds (BX 6, 1.3 miles southwest), where the mean fluoride content of Manitoba maple was 24 ppm (SE=3.2). This follows the general pattern noticed within other species: the more elevated concentrations often appear at greater distances from, rather than adjacent to, Alcoa.

Only one site around each fluoride source was sampled both in the summer and fall. Manitoba maple within both sites accumulated considerable amounts of fluoride during the interval. At the Reynolds site, SR 6 (2.3 miles northeast) the summer mean was 20 ppm (SE=0.8) and the fall mean was 35 ppm (SE=2.3). At site SR 14 (2.3 miles southwest of Alcoa), the summer mean was 5 ppm (SE=0.6) and the fall mean was 39 ppm (SE=3.2). This is a reversal of the usual pattern noted around Reynolds, where the fall collections fluoride values were comparable or lower within most sites.

The results reported by OME at comparable sites generally showed increases in fluoride content each successive month. The most noticeable increases were observed at the sites 0.9 mile and 1.9 miles northeast of Reynolds. At these impacted areas, OME's seasonal averages were considerably higher than the values found in the study

Figure 9

MEAN FLUORIDE CONTENT, UNWASHED MANITOBA MAPLE FOLIAGE, VICINITY OF RENOLDS, ALCOA 1977



reported herein. This could be due to the fact that a large percentage of the ambient fluoride at the sites close to Reynolds may have been caused by particulate fluoride deposition, a theory supported by the 1977 ambient monitoring data. Because higher than normal amounts of rain fell during September and October, the values found in October in this study may have been lower because of particulate removal by this rainfall (OME sampled only through August).

Chapter 8

RESULTS - SMALL MAMMAL SPECIES

Results

Because of the limited numbers of wild animals inhabiting Cornwall Island and time and equipment constraints, collections were limited to small mammal species (mice, moles, and shrews). These mammals were collected from two sites on Cornwall Island and from one site on the New York State section of the Reserve, at varying distances and directions from Reynolds. No small mammals were sampled from the Alcoa vicinity. The predominant species trapped were meadow voles (*Microtus pennsylvanicus*) and short-tail shrews (*Blarina brevicada*). Occasionally, deer mice (*Peromyscus maniculatus*) and jumping mice (*Zapus hudsonius*) were trapped, with one house mouse (*Mus musculus*) and one masked shrew (*Sorex cinereus*) also collected. Control samples of each of these species were trapped from the same area where vegetation controls were sampled (Wellesley Island Nature Center) and from one additional area in the state of Pennsylvania.

The voles and shrews were the most plentiful and since they normally do not range far for their food, it was assumed they would reflect the amount of fluoride contamination in the area in which they were found.

Table 27 lists mean femur fluoride values ($\bar{x}$) in ppm dry fat-free weight, sample size (n), sample standard errors (SE) and high and low femur fluoride concentrations of the species collected from each site. Also included are mean body weights ($\bar{x}$) in grams, sample standard errors of the animals, and the mean fluoride content of a composite grass sample from each site.

Femur fluoride values for meadow voles from site #2, 1.1 miles northeast of Reynolds (the most impacted area), ranged from 1310 to 5599 ppm, with a mean of 3775 ppm (SE=355.2). These values were 9 to 40 times higher than those found in control animals. As an indication of the overall magnitude of contamination at this site, fluoride concentrations in a composite grass sample averaged 88 ppm. Femur fluoride levels in short-tail shrews from this area ranged from 5284 to 8678 ppm, with a mean of 6557 ppm (SE=514.9). This represents values 7 to 12 times higher than those found in control animals. At sites #1 and #3 (2.3 miles northeast and 1.3 miles east-northeast of Reynolds, respectively), femur levels of both species were slightly lower, ranging from 2 to 10 times higher than controls. Fluoride concentrations in composite grass samples at both of these sites were 25 and 13 ppm fluoride, respectively. In general, site #2 (1.1 miles northeast), site #1 (2.3 miles northeast), and site #3 (1.3 miles east-northeast) represent areas of high, intermediate, and moderate fluoride contamination, respectively, based on fluorides found in the composite grass samples. The same trend is consistently seen in femur fluoride values from each site. As noted with native vegetation collections around Reynolds, the most impacted area is the closest to Reynolds in a northeasterly direction, with fluoride levels decreasing as distances from the source increase and directions deviate from the predominant plume path.

Table 27

SMALL MAMMAL DATA - ST. REGIS RESERVE 1977 (REYNOLDS VICINITY)

Ppm Fluoride in Femurs

Species	Trapping Site #2 1.1 miles NE	Trapping Site #1 2.3 miles NE	Trapping Site #3 1.3 miles ENE	Control
Microtus pennsylvanicus (Meadow vole)	Ppm F $\bar{x}$ 3775 SE = 355.2 High 5599 Low = 1310 Body Wt $\bar{x}$ 26.4 g SE = 1.4 $n = 14$	Ppm F $\bar{x}$ - 1153 SE = 86.2 High - 1696 Low 515 Body Wt $\bar{x}$ - 23.9 g SE 1.4 $n = 15$	Ppm F $\bar{x}$ 1531 SE = 238.3 High 2692 Low = 634 Body Wt $\bar{x}$ = 22.4 g SE - 0.8 n 8	Ppm F $\bar{x}$ 153 SE 17.2 High = 203 Low = 126 Body Wt $\bar{x}$ 22.7 g SE = 3.3 $n = 4$
Blarina brevicada (Short-tail shrew)	Ppm F $\bar{x}$ = 6557 SE = 514.9 High = 8678 Low = 5284 Body Wt $\bar{x}$ 19.3 g SE 0.8 $n = 8$	Ppm F $\bar{x}$ - 3317 SE 457.5 High = 3774 Low 2859 Body Wt $\bar{x}$ 19.8 g SE = 0.8 $n = 2$	Ppm F $\bar{x}$ - 1786 SE = 226.4 High - 3168 Low = 877 Body Wt $\bar{x}$ 20.5 g SE 0.7 $n = 13$	Ppm F $\bar{x}$ = 722 SE = 69.1 High = 1083 Low = 428 Body Wt $\bar{x}$ 17.1 g SE 0.6 $n = 11$
Peromyscus maniculatus (Deer mouse)	Ppm F $\bar{x}$ = 1497 SE = 325.3 High = 2142 Low - 1101 Body Wt $\bar{x}$ = 13.2 g SE = 0.9 $n = 3$		Ppm F $\bar{x}$ = 534 Body Wt 14.0 g $n = 1$	Ppm F $\bar{x}$ = 257 SE - 33.5 High = 398 Low 151 Body Wt $\bar{x}$ 22.2 g SE 1.2 $n = 6$
Zapus hudsonius (Jumping mouse)		Ppm F $\bar{x}$ - 619 SE = 287.0 High 1193 Low = 330 Body Wt $\bar{x}$ - 14.5 g SE 2.0 $n = 3$		
Mus musculus (House mouse)	Ppm F = 1253 Body Wt = 10.7 $n = 1$			
Sorex cenerius (Masked shrew)			Ppm F = 3223 Body Wt = -- $n = 1$	
Mean ppm Fluoride in composite grass sample from trapping site	88	25	13	1.2 (New York State) 2.0 (Pennsylvania)

Chapter 9

DISCUSSION

Native Vegetation Species

The results of fluoride analysis of native vegetation species show a substantial degree of variability within sites, within species and within the individuals of each species. This phenomenon is not unusual, however, in natural ecosystems because of the number of factors influencing plume dispersion and subsequent vegetative uptake of pollutants.

The variability in the response within certain plant species or individuals to fluoride accumulation is the result of both biologic and environmental factors. Researchers have identified that plants differ inherently in the biologic characteristics which influence the susceptibility or tolerance to fluorides (Weinstein, 1977). Environmental factors such as temperature, light quality, relative humidity, precipitation, soil conditions, position of the leaf or leaves on the plant and presence of insects or parasites all influence the response of a specific plant to fluoride uptake and injury (EPA, 1980). Also, pollutant characteristics such as particle size or type, gas to particulate ratio, dispersion rates, concentration and duration of exposure and rates of settling all contribute to the plants' exposure.

While these factors determine variability within the species and sites, the fluoride results generally indicate that distance and direction from the source were most responsible in determining the dosage delivered to a site. Stands of vegetation with open, unobstructed exposure, and in locations immediately downwind of either source accumulated the largest quantities of fluorides, and acted as natural shields for the areas and vegetation beyond. An example can be seen in a comparison of the results of vegetation analyses from sites SR 21 and SR 2: SR 21 was located 1.4 miles northeast of Reynolds on a downhill slope, exposed to and immediately downwind of Reynolds. Site SR 2, the Cornwall Island School area, however, was located only 0.1 mile farther away but in an area without open exposure to Reynolds. The fluoride content of vegetation at SR 21 was three to six times higher than levels in vegetation from SR 2. This natural shielding phenomenon can explain why similar species of vegetation from areas within close proximity of each other exhibited tremendous differences in fluoride uptake.

Based on the data generated in this study, the areas most consistently impacted by fluorides in the Reynolds vicinity during the summer and fall of 1977 were within 1.5 miles northeast of Reynolds on Cornwall Island and less than one mile southeast of the source in New York State.

Sites on Cornwall Island with unobstructed exposure to Reynolds were most extensively affected. In these areas excessive levels of fluorides accumulated in the grasses, forbs, shrubs, deciduous trees, and small mammals, evidence of fluorides moving up the food chain.

Sites farther away, or in more sheltered areas from Reynolds, accumulated lower fluoride levels, but concentrations at all sampling sites on Cornwall Island and the St. Regis Reserve can be considered elevated.

Because a large portion of the atmospheric pollutants from Reynolds were particulates and the effluents in general were visibly dense, the most severe contamination occurred within a mile of the plant; the particulates apparently fall out in a relatively short distance, while the airborne gaseous emissions apparently affect the areas farther away.

Because of the predominant wind patterns in the area, Cornwall Island is subjected to the most consistent, high-level fumigations from Reynolds. However, as noted in this study and in studies carried out by the Ontario Ministry of the Environment from 1977 through 1981, when the directional wind patterns are significantly altered (as was the case in August through October 1977), fumigations of ambient fluoride pollutants shifted to the areas affected by these altered wind patterns.

Fluoride levels observed in vegetation in the Alcoa vicinity indicate that high levels of atmospheric fluorides affected areas around Alcoa, particularly to the northeast, at even greater distances from the source than noted around the Reynolds vicinity. Whereas the most impacted areas around Reynolds were adjacent to the plant with levels decreasing as distances increased, the most impacted area sampled around Alcoa was two miles northeast of the plant. Mean fluoride values and sample standard errors at site SR 15, less than half the distance northeast of Alcoa, were significantly lower for many of the vegetation species. At the site four miles northeast of the plant (SR 22), fluoride levels again decreased markedly as compared to the site two miles away, although concentrations in dogwood and ash were still at least 10 times higher than control samples.

The variable air currents caused by the large masses of cold water bodies (the St. Lawrence Seaway) in the area probably influence the pattern of fluoride deposition in certain areas. In addition, the nature of the emissions themselves affects patterns of contamination.

As of 1977, Reynolds' emissions were approximately 32 tons of total fluorides per year, of which at least 92 tons/year were gaseous. Alcoa's emissions were approximately 297 tons of gaseous fluorides alone per year; this was more than three times as much gaseous fluorides as that given off by Reynolds, with no estimate available on Alcoa's particulate fluoride emissions. However, particulates emitted from Alcoa visibly appeared to be very small and probably did not settle out as quickly as particulates from Reynolds. This theory is supported by the vegetation data from both areas, which suggested that vegetation uptake was the result of the more gaseous and small particulate ambient emissions from Alcoa being carried farther than the heavier particulate emissions from Reynolds. Because Alcoa's emissions were dispersed at these substantial distances from the source and because the St. Regis Reserve lands are less than eight miles downwind of Alcoa, it is possible that Reynolds may not be the sole source of fluoride contamination on Reserve lands. The elevated levels observed four miles northeast of Alcoa would suggest that Alcoa's emissions may contribute to the overall exposure of Reserve lands to ambient fluoride pollution.

The data obtained during the fall 1977 collection period versus the summer collection around Alcoa further support the contention that the emissions from Alcoa were predominantly gaseous and affected a wider area. While most of the fluoride levels in vegetation collected during the fall around Reynolds remained relatively stable or decreased (with the exception of site SR 7, 0.7 mile southeast), the means of samples collected from the Alcoa area during this second collection period consistently increased, differing by at least one standard error as compared to the summer values.

Because of the extremely heavy rains that occurred between the collection periods, particulate fluoride deposits from Reynolds may have been washed off or leached from vegetation. It has been reported that the bioelimination of fluorides can occur not only through the loss of leaves or twigs, but through leaching by rain (EPA, 1980). However, the observations suggest that the vegetation affected by the more gaseous emissions around Alcoa steadily accumulated fluorides throughout the growing season.

The most notable exception to this trend was observed at site SR 7, 0.7 mile southeast of Reynolds. Mean fluoride concentrations in green ash samples from this site were significantly higher in the fall than in the summer. The trend of decreasing values noted at most of the other sites around Reynolds was probably attributable to the heavy rainfall (as mentioned above) and the decrease in the winds affecting the usually impacted areas during that time (Chapter 5). Fumigations at SR 7 in late August and September, however, may have been so concentrated that rain did not affect fluoride levels significantly or these fumigations may have contained relatively large amounts of gaseous fluoride, not as easily removed by moisture. Also, winds influencing this area to the southeast increased during the interim period. The combination of these factors probably caused the significant increases noted at this site.

Another factor that may have contributed to the decrease in fluoride values noted around Reynolds in the fall is premature loss of affected leaves. Weinstein (1977) suggested that trees may eliminate fluorides by premature shedding of leaves. The net effect of this is a limited or decreased total fluoride concentration found in the foliar tissues.

While such factors as rainfall, wind directions, sampling period, and pollutant characteristics were influential in determining fluoride dosages to the specific sites, distance and direction from each source were the most consistent factors affecting fluoride uptake. Around the Reynolds facility, the means and standard errors of samples from the three sites within one mile (SR 1, 1 mile northeast; SR 10, 0.9 mile north-northeast; and SR 7, 0.7 mile southeast) were significantly higher than samples from the other sites around Reynolds. Around Alcoa, the sample means and standard errors were significantly higher within the sites up to 2 miles northeast of the facility. Within certain species (red-top grass, milkweed, wild grape, and elm) the sample means were significantly higher (differing by at least one standard error of the sample) at the site 2.0 miles northeast (SR 8), as compared to the site 0.7 mile northeast (SR 15) of Alcoa.

There were no significant differences noted between contamination from Reynolds or Alcoa within these most impacted sites. Likewise, no significant differences in fluoride uptake can be assigned to positions in the canopy layer (grass, shrub, or overstory layers) nor to any given species (with the exception of bitternut hickory , where both samples and control were higher than other species due to naturally-occurring fluorides). Differences between summer and fall values were variable and depended on the species, site location, and pollution source.

Of the more than 30 species of native vegetation and small mammals sampled in and around the St. Regis Reserve, all can be considered contaminated with fluorides, at times reaching levels more than 200 times those observed in samples from unpolluted areas.

Small Mammal Species

The purpose of sampling small mammal species from the St. Regis Reserve area was to extend the investigation of fluoride distribution in the area and possible transference between a higher trophic level of the terrestrial ecosystem. No attempt was made to differentiate fluoride uptake relative to sex, age, or specific diet of the small mammals. Only femur tissues were analyzed rather than soft tissues because it has been estimated that 96% to 98% of fluorides that are ingested by animals are deposited in the bones and teeth (EPA, 1980).

The composite grass samples from each site were analyzed in order to demonstrate overall fluoride status of each trapping area.

With regard to both meadow voles and short-tail shrews, femur fluoride values and sample standard errors were significantly different at each site: the highest levels were noted at the site closest to Reynolds (1.1 miles northeast) where the composite grass sample also showed greatest contamination. The intermediate values for each species were found at the site 2.3 miles northeast of Reynolds, and the lowest values were found 1.3 miles from Reynolds in a more easterly direction out of the predominant plume path. This site was also the least contaminated with regard to fluoride in the composite grass sample. Even at this least contaminated site, femur fluoride values were significantly higher (two to ten times) than those found in control animals.

Within each site, the femur fluoride values of the short-tail shrew species were consistently higher than those of the meadow voles, even though the shrew body weights were considerably lower. There is no immediate explanation for these differences, but this trend probably results from dietary differences between the primarily herbivorous voles versus the more insectivorous shrews. Also, the differing body weights would suggest differences in metabolic rates and perhaps greater food requirements (therefore greater fluoride intake) on the part of the shrews. A useful extension of this work would be to determine the influence of type of diet, metabolic requirements and age of the animals with regard to fluoride accumulation in the bones.

At the two sites where deer mice were found, femur fluoride values were lower than either the shrews or voles. In a similar study from a fluoride-contaminated environment, Wright, Davison and Johnson (1978) found femur fluoride values of long-tail field mice were significantly higher than values found in field voles. No hypothesis is presented for the consistently higher tissue fluorides found in the mice versus the voles, but these authors also suggest that physiological differences, diet and/or sex distribution may be responsible. There is no apparent explanation for differences in Wright et al.'s findings which contradict the findings in this study in regard to levels found in mice versus voles. Wright et al. also tested vegetation and soils from each site and concluded that there was a strong positive relationship between fluoride values and observed environmental fluoride levels.

The results of the study presented herein demonstrate clearly that excessive levels of fluorides are accumulating in the small mammals of the St. Regis Reserve where environmental contamination is the highest. This contamination is not just limited to vegetation; fluorides are accumulating at higher trophic levels.

Chapter 10

CONCLUSIONS

1. Excessive fluoride levels are accumulating in the native vegetation species in and around the St. Regis Reserve area with the highest levels noted within 1 mile northeast of the Reynolds facility on Cornwall Island, and within 2 miles northeast of Alcoa. All vegetation types sampled in this study can be considered contaminated with fluorides, with levels reaching up to 200 times those observed in samples from areas not subject to fluoride pollution.

2. The fluoride levels observed in the femurs of small mammals demonstrate that fluorides are being transferred to higher trophic levels in the St. Regis Reserve environment.

3. The pattern of vegetative and small mammal contamination showing highest levels of fluorides immediately downwind of both Alcoa and Reynolds, with levels decreasing as distances from each source increase, confirm the assumption that Reynolds and Alcoa are the most significant sources of fluoride pollution in the St. Regis Reserve area. These observations are consistent with emission inventory records quantifying ambient fluoride emissions from each facility.

4. While there was no conclusive evidence concerning actual amounts of ambient pollution from Alcoa which may be affecting the St. Regis lands, the high levels of fluoride contamination observed at proportionately greater distances from the Alcoa smelter would suggest that Alcoa's emissions may, at times, depending on meteorological conditions, contribute to the total fluoride deposition on the St. Regis Reserve lands.

5. Based on data available since 1974, ambient air quality in the St. Regis Reserve - Cornwall Island area has not significantly improved since 1974. In fact, air quality measurements on Cornwall Island deteriorated slightly in 1977, despite the fact that the frequency of winds affecting the island in 1977 decreased. The data also revealed that Reynolds' particulate fluoride emissions reaching Cornwall Island doubled from 1976 to 1977.

6. The proposed Canadian standards and Ontario criteria for fluorides in ambient air were exceeded a significant portion of the time in 1977 on Cornwall Island, although there were no contraventions of New York standards.

7. The 1977 vegetative data demonstrated that meteorological factors such as decreased rainfall and directional wind changes cannot be relied upon to mitigate or alleviate fluoride contamination in the area. The only way to prevent fluoride accumulations in vegetation and the small mammal populations in the area is to decrease fluoride emissions at the respective sources.

8. Assessments of fluoride uptake in vegetation and cattle on St. Regis Reserve lands since 1977 indicate that high levels of fluorides are continuing to impact the area, even after completion of emission control programs.

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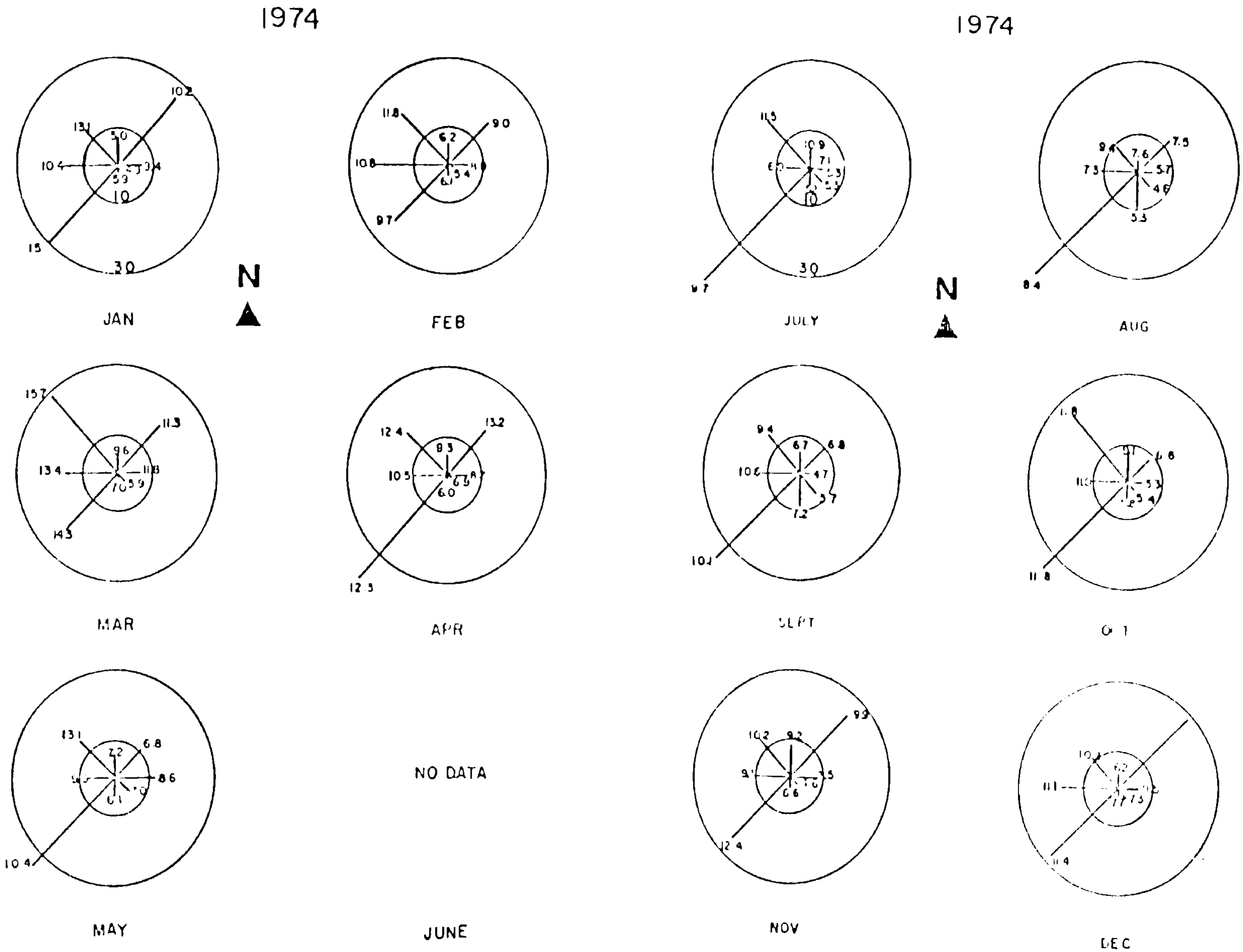
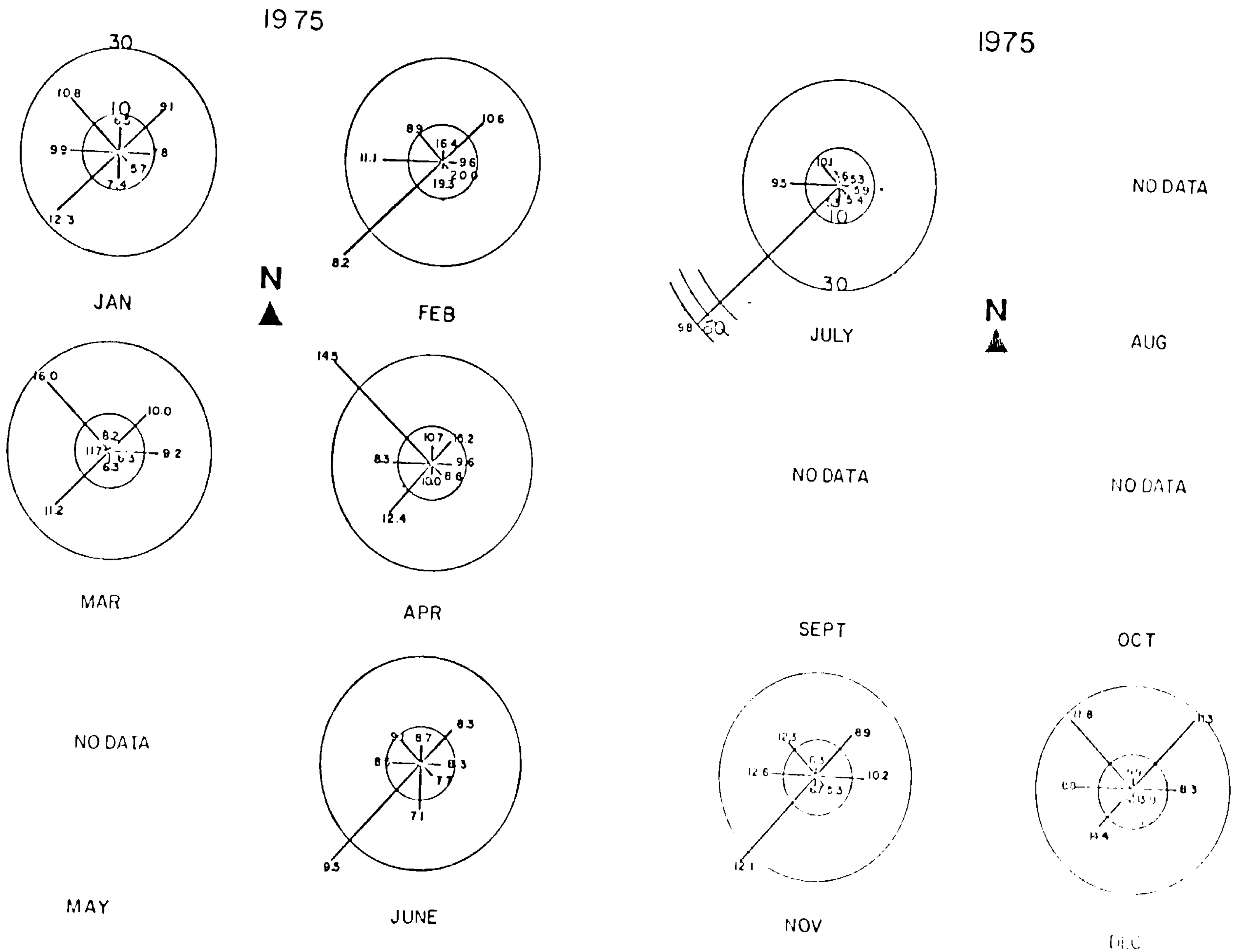
APPENDIX A

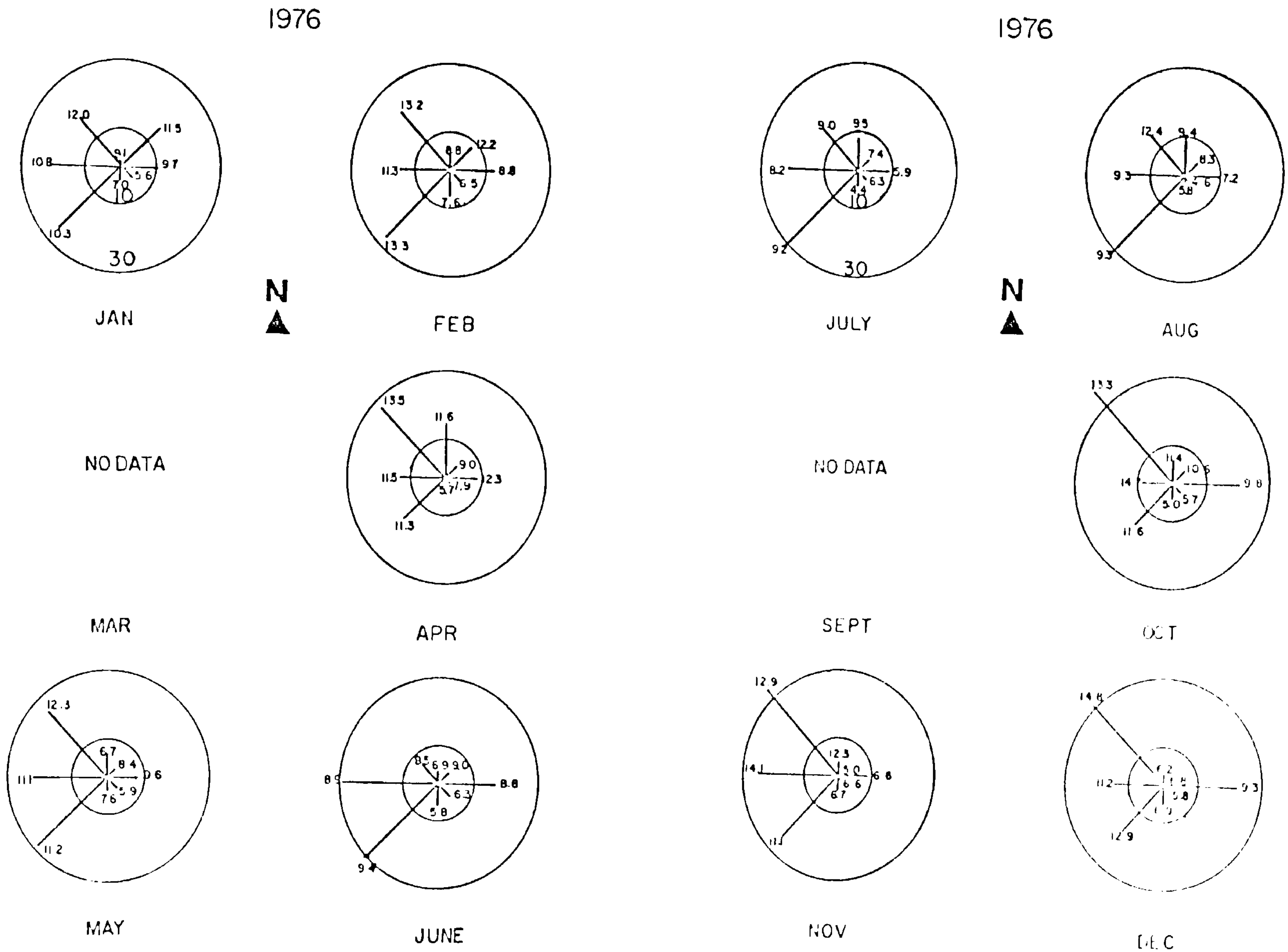
Wind Roses, Cornwall Island Vicinity 1974-1977



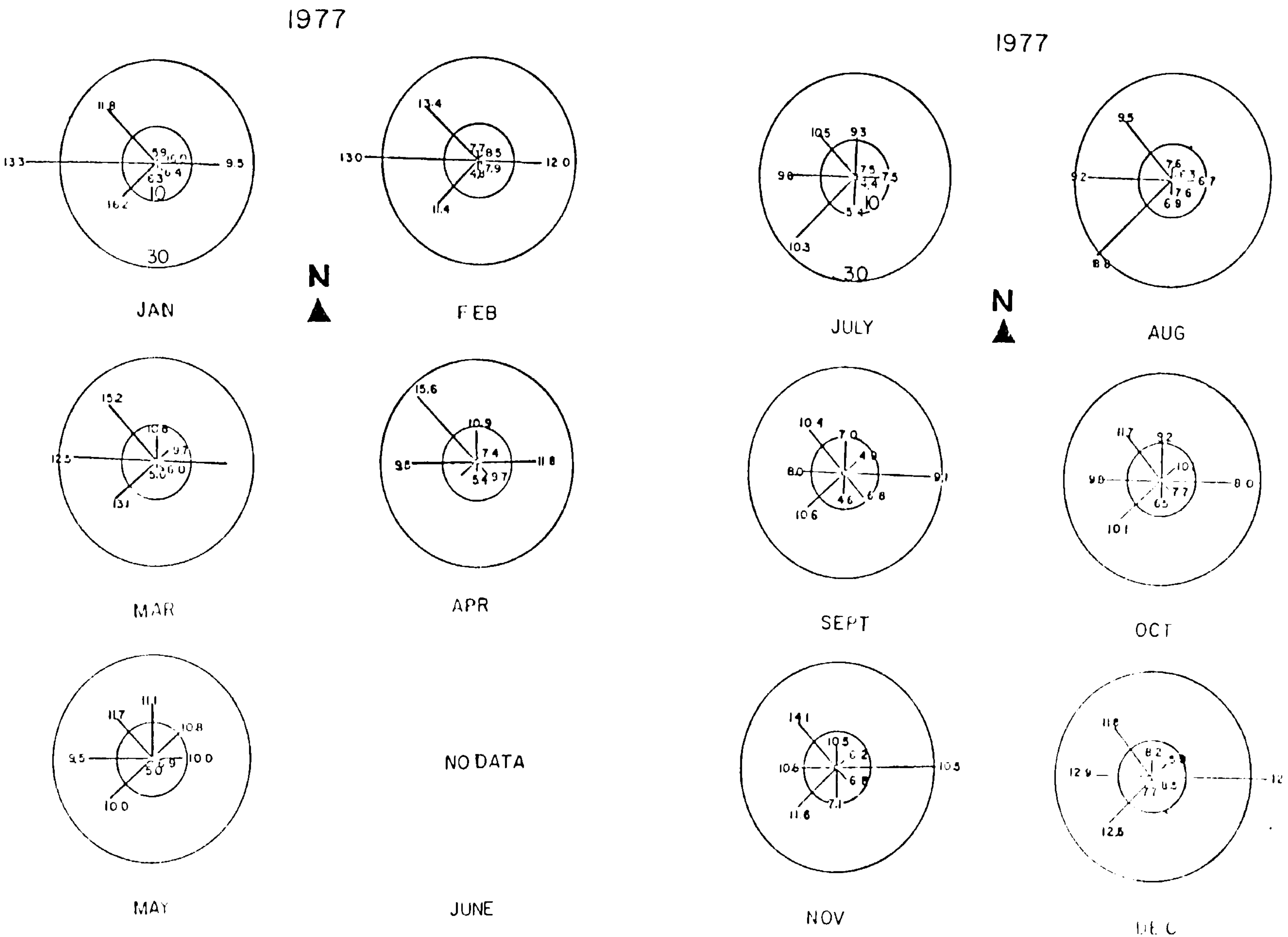
1974 wind roses--Cornwall Island area.



1975 wind roses--Cornwall Island area.



1976 wind roses--Cornwall Island area.



1977 wind roses--Cornwall Island area.

APPENDIX B

Master List of Data from the St. Regis Reserve Vicinity

1977

COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Grass Species

SITE #	Quack grass (leaves)	Reed canary grass (leaves)	Timothy (leaves)	Kentucky blue grass (whole grass)	Red-top grass (whole grass)	Smooth brome grass (leaves)		
SR1	107 SE=17.9 n=3	171 n=1	132 SE= 3.9 n=4	68 SE= 8.9 n=4	49 SE= 5.2 n=4	--		
SR2	--	55 SE=12.2 n=4	52 SE= 9.3 n=4	--	21 SE= 5.8 n=4	--		
SR3	8 SE= 1.4 n=4	8 SE= 2.2 n=4	8 SE= 0.4 n=4	6 SE= 1.6 n=4	5 SE= 0.6 n=4	--		
SR4	34 SE= 9.9 n=2	--	25 SE= 1.3 n=3	24 SE= 4.9 n=4	17 SE= 1.6 n=4			
SR5	--	32 SE= 4.1 n=4	10 SE= 1.0 n=4	7 SE= 0.3 n=4	6 SE= 1.2 n=4	--		
SR6	2 SE= 2.3 n=4	6 SE= 1.4 n=4	19 SE= 5.0 n=2	5 SE= 1.8 n=2	13 SE= 4.5 n=2	--		
SR8	108 SE=25.9 n=4	53 n=1	113 SE= 8.8 n=4	--	45 SE= 5.3 n=4	30 SE= 8.5 n=3		
SR9	--	10 SE= 2.6 n=4	--	7 SE= 1.5 n=4	5 SE= 0.9 n=4	10 SE= 2.5 n=4		
SR10	--	105 SE=28.4 n=4	100 SE=19.6 n=4	66 SE=16.9 n=4	52 SE=16.3 n=4	--		
SR13	--	--	16 SE= 1.4 n=4	6 SE= 0.7 n=4	6 SE= 0.7 n=4	--		
SR14	11 SE= 1.5 n=4	7 SE= 1.0 n=4	--	4 SE= 0.7 n=4	--	--		
SR15	--	--	79 n=1	16 SE= 2.6 n=4	33 SE= 4.7 n=4	--		
Controls	--	1.8 SE= 0.2 n=4	1.6 SE= 0.4 n=4	1.3 SE= 0.2 n=4	1.1 SE= 0.3 n=4	1.4 SE= 0.2 n=4		

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

APPENDIX B : page 2 of 8

COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Forb and Shrub Species; Willows

SITE #	White sweet clover	Milkweed	Wild grape	Sumac	Dogwood	Willows	Willows	Willows
SR1	125 SE=23.2 n=4	219 SE=50.7 n=4	142 SE=42.8 n=3	106 SE=11.3 n=4	132 SE=16.7 n=4	164 SE=37.4 n=4 (type "A")		
SR2	--	52 SE=10.8 n=4	40 SE= 5.1 n=4	47 SE= 9.2 n=4	--	--		
SR3	--	10 SE= 1.3 n=4	4 SE= 0.6 n=4	5 SE= 0.7 n=4	23 SE= 5.7 n=4	11 SE= 1.0 n=4 (type "B")		
SR4	18 SE= 4.1 n=4	48 SE= 4.4 n=4	25 SE= 4.3 n=4	24 SE= 1.9 n=4	42 SE= 0.7 n=2	--		
SR5	15 SE= 1.4 n=4	25 SE= 0.4 n=3	23 SE= 0.4 n=2	16 SE= 0.3 n=3	35 SE= 5.2 n=4	28 SE= 3.3 n=2 (type "A")	21 n=1 (type "C")	16 n=1 (type "D")
SR6	--	18 SE= 1.4 n=4	4 SE= 0.8 n=4	15 SE= 1.3 n=4	--	18 SE= 2.3 n=4 (type "B")		
SR8	58 SE= 6.4 n=4	115 SE=18.9 n=4	64 SE=16.3 n=4	71 SE= 9.3 n=4	119 SE=25.3 n=4	144 SE=12.1 n=3 (type "B")	68 SE=22.7 n=2 (type "C")	
SR9	8 SE= 1.3 n=4	10 SE= 1.9 n=4	6 SE= 0.9 n=4	6 SE= 0.9 n=4	16 SE= 3.5 n=2	12 SE= 1.4 n=4 (type "B")		
SR10	99 SE=26.1 n=4	130 SE= 5.7 n=3	196 SE=47.8 n=4	249 SE=32.4 n=4	168 SE=72.8 n=2	114 SE= 1.5 n=2 (type "A")	121 SE= 4.3 n=2 (type "B")	
SR11	--	--	--	47 SE= 6.4 n=4	--	--		
SR12	--	--	--	25 SE= 4.7 n=4	--	--		
SR13	15 SE= 2.9 n=4	13 SE= 2.9 n=4	8 SE= 1.9 n=4	--	13 SE= 2.5 n=4	--		
SR14	4 SE= 0.6 n=4	10 SE= 2.5 n=4	3 SE= 0.5 n=4	3 SE= 0.7 n=4	--	7 SE= 2.0 n=4 (type "B")		

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Forb and Shrub Species; Willows (cont'd)

SITE #	White sweet clover	Milkweed	Wild grape	Sumac	Dogwood	Willows	Willows	Willows
SR15	--	68 SE=10.8 n=4	11 SE= 2.8 n=4	51 SE= 8.5 n=4	117 SE=13.0 n=4	--		
SR16	--	5 SE= 0.4 n=4	3 SE= 0.7 n=4	--	12 SE= 1.9 n=4	--		
SR17	--	14 n=1	7 SE= 0.7 n=2	8 SE= 2.3 n=4	26 SE= 0.8 n=4	--		
SR18	--	16 SE= 3.8 n=3	9 SE= 1.7 n=3	--	18 SE= 1.9 n=4	--		
Controls	0.9 SE= 0.2 n=4	0.9 n=1	1.6 SE= 0.3 n=4	0.9 SE= 0.3 n=4	2.1 SE= 0.5 n=4	2.1 SE= 0.3 n=2 (type "A")	2.0 SE= 0.5 n=4 (type "B")	2.4 n=1 (type "D")

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Overstory Species

SITE #	Cotton-wood	Aspen	Basswood	Elm	Red Maple	Sugar Maple	Silver Maple	White Ash	Green Ash	Black Ash	White Oak	Red Oak	Manitoba Maple	Bitternut Hickory
SR1	161 SE=24.8 n=4	156 SE=37.4 n=4	259 SE=30.6 n=2	158 n=1	91 SE=12.9 n=2	--	--	--	--	--	--	--	--	--
SR2	--	--	37 SE= 2.6 n=4	53 SE= 7.8 n=4	--	21 SE= 7.7 n=2	60 SE=25.1 n=2	--	--	--	--	--	--	--
SR3	--	--	--	--	--	--	--	--	--	--	--	--	--	--
SR4	25 SE= 2.4 n=4	28 SE= 0.4 n=4	--	62 n=1	--	--	--	--	--	--	--	--	--	--
SR5	19 SE= 0.5 n=4	19 SE=1.5 n=4	11 n=1	--	--	--	21 SE= 0.7 n=3	--	21 n=1	--	--	--	--	--
SR6	--	18 SE= 1.1 n=4	13 SE=1.5 n=8	19 n=1	--	--	--	--	--	--	--	--	20 SE=0.8 n=3	207 SE= 7.4 n=2
SR7	--	106 SE=8.6 n=2	153 SE=17.7 n=8	85 SE=34.5 n=6	106 SE=13.8 n=5	104 SE=22.7 n=3	--	--	78 SE=14.3 n=8	63 n=1	65 SE= 6.6 n=6	73 SE=34.8 n=4	--	--

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

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COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Overstory Species (cont'd)

SITE #	Cotton-wood	Aspen	Basswood	Elm	Red Maple	Sugar Maple	Silver Maple	White Ash	Green Ash	Black Ash	White Oak	Red Oak	Manitoba Maple	Bitternut Hickory
SR8	33 SE= 9.4 n=2	80 SE=22.8 n=8	65 SE=19.0 n=4	71 SE=16.8 n=4	:	29 SE= 1.3 n=4	:	:	:	:	:	:	:	500 SE=17.3 n=2
SR9	12 SE= 1.4 n=4	:	12 SE=0.8 n=2	:	:	:	:	3 SE= 0.3 n=2	:	:	:	:	:	246 n=1
SR10	65 SE= 9.1 n=4	:	160 SE=16.3 n=4	:	251 n=1	:	:	:	:	128 n=1	:	78 n=1	:	:
SR11	:	:	:	:	:	22 SE=2.8 n=4	:	:	:	:	11 n=1	:	:	:
SR12	14 n=1	:	16 SE=2.1 n=4	11 SE=1.5 n=4	:	:	:	29 n=1	:	:	:	:	:	398 n=1
SR13	:	:	:	8 SE=2.2 n=3	:	4 SE=1.3 n=4	:	:	:	:	:	:	:	:
SR14	:	:	:	5 SE=0.5 n=4	:	:	:	:	:	:	:	:	5 SE=0.6 n=4	:

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

COLLECTION PERIOD: Summer 1977
NATIVE VEGETATION TYPE: Overstory Species (cont'd)

SITE #	Cotton-wood	Aspen	Basswood	Elm	Red Maple	Sugar Maple	Silver Maple	White Ash	Green Ash	Black Ash	White Oak	Red Oak	Manitoba Maple	Bitternut Hickory
SR15	--	--	--	52 SE= 5.1 n=4	--	--	--	--	45 SE=5.2 n=4	--	--	--	--	--
SR16	--	--	--	2 SE= 0.4 n=4	--	--	--	--	3 SE= 0.3 n=4	--	--	--	--	--
SR17	--	--	15 SE=2.2 n=3	11 SE=0.9 n=4	--	--	13 SE=0.4 n=3	--	--	--	--	--	9 SE=0.9 n=4	--
SR18	--	--	--	7 SE=1.6 n=4	--	--	--	--	6 SE=0.3 n=4	--	--	--	--	--
SR19	--	--	--	17 SE=3.5 n=2	11 SE=2.2 n=4	--	--	--	--	--	--	--	--	--
Control	1.4 SE=0.5 n=3	1.1 SE=0.6 n=3	0.6 SE=0.2 n=4	1.9 SE=0.4 n=4	2.0 SE=0.4 n=4	0.9 SE=0.4 n=4	0.8 SE=0.3 n=2	--	0.9 SE=0.2 n=4	1.9 n=1	1.1 SE=0.4 n=4	1.0 SE=0.4 n=4	2.0 SE=0.2 n=4	--

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

APPENDIX B: page 7 of 8

COLLECTION PERIOD: Fall 1977
NATIVE VEGETATION TYPE: Mixed species

SITE #	Timothy leaves	Dogwood	White Ash	Green Ash	Black Ash	Manitoba Maple	Bitternut Hickory
SR1	--	118 SE=14.8 n=4	367 n=1	--	--	265 n=1	--
SR2	28 n=1	51 SE= 5.9 n=4	26 SE= 5.5 n=4	25 n=1	--	43 SE=3.7 n=3	--
SR3	--	14 SE=1.9 n=4	--	--	--	11 SE=1.8 n=4	--
SR4	--	36 SE=3.3 n=4	--	--	--	73 SE=3.8 n=4	--
SR5	2 n=1	16 SE=2.8 n=4	--	12 SE=4.6 n=4	--	13 SE=1.3 n=4	
SR6	--	21 SE=1.5 n=4	30 SE=1.4 n=2	--	--	35 SE=2.3 n=3	279 SE=19.6 n=2
SR7	--	315 SE=26.5 n=2	--	234 SE=32.6 n=4	355 SE=45.4 n=4	--	--
SR8	--	127 SE=12.3 n=4	149 SE=31.4 n=5	--	--	--	653 SE=150.8 n=2
SR9	--	27 SE=2.3 n=4	8 SE=1.1 n=2	10 SE=2.0 n=4	--	--	291 SE=23.5 n=3
SR10	--	174 SE=36.2 n=4	111 n=1	--	--	--	--
SR11	--	--	--	--	--	59 SE=14.1 n=4	--
SR12	--	--	20 n=1	--	--	--	648 SE=64.3 n=2
SR13	24 SE=8.6 n=2	34 SE=4.1 n=4	--	--	--	--	--
SR14	--	31 SE=3.4 n=4	--	25 SE=5.0 n=4	--	39 SE=3.2 n=4	--

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)

APPENDIX B: page 3 of 3

COLLECTION PERIOD: Fall 1977
NATIVE VEGETATION TYPE: Mixed species (cont'd)

SITE #	Timothy leaves	Dogwood	White Ash	Green Ash	Black Ash	Manitoba Maple	Bitternut Hickory	
SR15	--	143 SE=22.8 n=4	--	102 SE=14.9 n=4	--	--	--	
SR21	--	122 SE=3.2 n=4	106 SE=16.4 n=2	153 SE=12.8 n=4	176 SE=7.4 n=2	--	705 SE=194.8 n=2	
SR22	--	46 SE=4.5 n=4	23 SE=2.0 n=4	22 SE=0.8 n=4	--	--	--	
SR23	--	389 n=1	--	270 SE=41.8 n=2	--	--	--	
BX1	--	--	20 SE=2.4 n=4	--	--	47 SE=2.4 n=4	--	
BX2	--	--	--	--	--	28 SE=2.2 n=4	--	
BX3	--	--	7 SE=0.9 n=3	--	--	21 SE=2.5 n=7	--	
BX4	--	--	--	--	--	25 SE=2.6 n=4	--	
BX5	--	10 SE=1.8 n=2	--	--	--	12 SE=0.1 n=2	--	
BX6	--	21 SE=1.6 n=3	--	13 SE=3.4 n=4	--	24 SE=3.2 n=4	--	
Control	2.1 SE=0.6 n=2	3.3 SE=0.5 n=4	1.5 SE=0.3 n=4	2.6 SE=0.1 n=4	4.8 SE=0.6 n=4	2.9 SE=0.3 n=4	161 SE=16.7 n=4	

Results for each site presented as: Mean ppm fluoride
Sample standard error (SE)
Sample size (n)