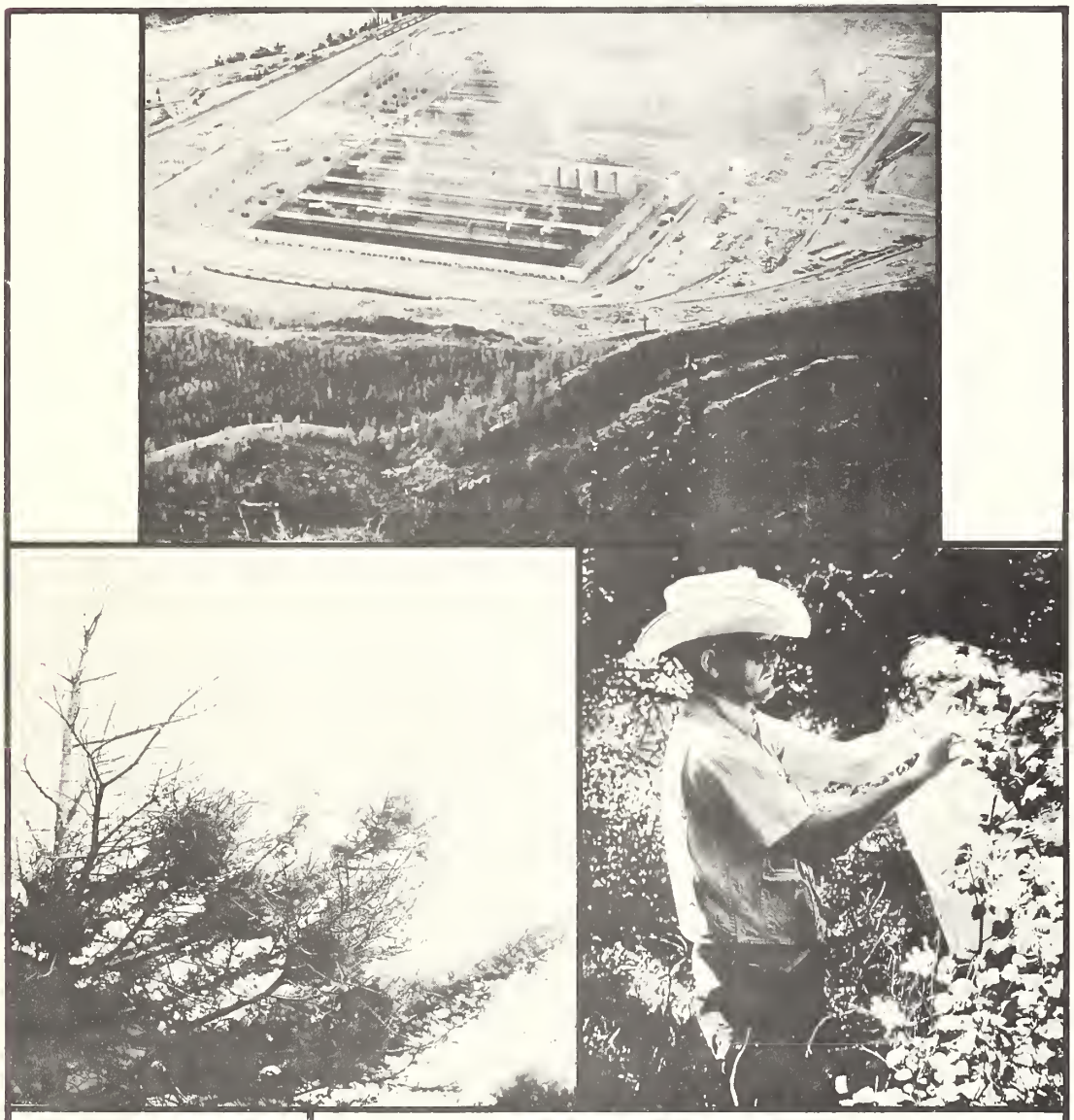
MONITORING FLUORIDE POLLUTION in Flathead National Forest and Glacier National Park



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DIVISION OF STATE AND PRIVATE FORESTRY
FOREST SERVICE - REGION 1
Missoula, Montana

The Anaconda Aluminum Company reduction works at Columbia Falls, Montana, is shown in the top photo. Note fluoride-killed trees in foreground. The lower-left photo shows top dieback and brooming in a Douglas-fir caused by fluorides from the Anaconda Company plant. Biologist Don Berg is sampling forest vegetation for fluoride analyses in the lower-right photo.

MONITORING FLUORIDE POLLUTION IN FLATHEAD NATIONAL FOREST AND GLACIER NATIONAL PARK

by

Clinton E. Carlson[1/]

SUMMARY

During August 1971, we conducted a study to monitor fluoride pollution in Flathead National Forest and Glacier National Park, near Columbia Falls, Montana. The study was done as a followup to our 1970 fluoride study (Carlson and Dewey 1971) of pollution caused by fluoride emissions from the Anaconda Aluminum Company at Columbia Falls. During the 1970 study fluoride emissions by the company were reduced from 7,500 to 2,500 pounds per day.

Fifteen plots, 20 percent of the 77 permanent radial plots established in 1970, were resampled in 1971. Chemical analysis of vegetation indicated average plot fluoride concentration was from 4 percent less in Glacier National Park to 77 percent less close to the aluminum plant when compared to 1970 data. Injury indexes dropped an average of 45.8 percent.

Analysis of conifer, shrub, and forb stem tissues indicated fluorides are accumulated by stem tissue. However, the exact location of accumulation was not determined.

Isopols, illustrated in the 1970 study, were recomputed based on the 1971 monitoring data. The total area polluted was 59 square miles (34,560 acres) less than in 1970, and injury was found on 84 square miles (53,920 acres) less than in 1970.

Analysis of insect tissue in 1971 indicated insects are still accumulating excessive fluorides. Even though the Anaconda Company reduced fluoride emissions at the aluminum plant in 1970, above-normal fluorides are still accumulating in vegetation up to 12 miles distant in Glacier National Park. This represents an area of 179,200 acres. Fluoride injury to vegetation was found on 15,200 acres, indicating the fluoride pollution problem at Columbia Falls has not been alleviated.

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INTRODUCTION

During 1970 the U.S. Forest Service intensively studied fluoride pollution in and adjacent to Flathead National Forest, near Columbia Falls, Montana (Carlson and Dewey 1971). Abnormally high concentrations of fluorides were found in vegetation on 214,000 acres of public and private lands. The Anaconda Aluminum Company at Columbia Falls was found to be the source of the airborne fluorides. Visible fluoride injury to plants, including tree mortality, branch dieback of trees and shrubs, and chlorosis and necrosis of foliage, was found on 69,120 acres.

Insects, including pollinators, foliage feeders, and predators, had abnormally high quantities of fluorides.

The Environmental Protection Agency (EPA) also studied fluorides in 1970 at Columbia Falls (personal communication; report not yet published). Their extensive meteorological data confirmed the Forest Service data on fluoride distribution in the area. EPA also found considerable fluoride-induced injury to indigenous vegetation throughout the area.

Dr. Clarence Gordon, University of Montana, contracted by EPA to study fluorides in Glacier National Park in 1970, found basically the same distribution of fluorides as did the Forest Service (Gordon 1972). Also, he found fluoride-caused injury to vegetation over the same area. Gordon's studies included fauna of the area; he correlated high fluoride concentrations in animal teeth and bones with the high fluorides in plant tissue.

Concurrent with these three independent studies, the Anaconda Company began reducing fluoride emissions at the aluminum plant. Early in 1970 the plant was emitting 7,500 pounds of fluorides per day; by September emissions had been reduced to 5,000 pounds per day; and by summer of 1971, emissions were down to 2,500 pounds per day.[2/] The reductions were attained by installing Venturi scrubbers and limiting aluminum production.

Reductions in fluoride emissions implied that subsequent accumulations of fluorides by vegetation may have been reduced, and that resultant injury may also have been lessened. Therefore, we felt it necessary in 1971 to monitor for possible continuing accumulation and effects of fluorides on vegetation as a followup to the 1970 study. Specifically the objectives of this study were:

2/ Reported by Anaconda Aluminum Company in various issues of Hungry Horse News, Columbia Falls, Montana, from 1970 to 1971.

1. Determine current fluoride concentrations in vegetation in the 1970 study area.

2. Determine current vegetational injury attributable to fluorides.

3. Evaluate current fluoride concentrations in insects.

MATERIALS AND METHODS

Selection of Study Plots

Fifteen of the 77 study plots established in 1970 were selected for this evaluation. They were:

R2-P2[3/]	R5-P4	R5-P6
R3-P3	R5-P5	R8-P5
R3-P4	R6-P3	Columbia Mountain special plot
R4-P2	R6-P4	R4-P6
R5-P3	R7-P3	R5-P9

Control data was obtained by sampling control plot No. 3.

The plots were arbitrarily selected on the basis of 1970 fluoride distribution in the area. We believed it would be desirable to sample plots on the west face of Teakettle Mountain where fluorides were high, plots on the east side where fluorides were moderate, and plots in Glacier National Park where fluorides were low. The 15 plots listed above satisfied these conditions.

Collection of Vegetation

All collections were made in late August of 1971 to correspond with the second sampling of 1970. Two pounds of foliage were collected from each of two conifer species, one or two shrub species, one forb, and one grass species on each plot. Conifer branches were cut to include 3 years' foliage from each tree sampled. Tree species were not held constant from plot to plot. Basically, species were the same as those collected in 1970. Each sample was placed in a clean plastic bag and brought to the Forest Service laboratory in Missoula for observation and analysis.

3/ R = radius; P = plot.

Laboratory Analysis

Specific ion chemical analysis

We were in need of a simple, fast, accurate, and relatively cheap method of fluoride analysis for our work. The cost of contracting analyses to WARF[4/] was prohibitive in 1971. Also, studies at Boyce Thompson Institute (Jacobson and McCune 1969), indicated the colorimetric method used by WARF produced highly variable results between different laboratories. The specific ion method (Durst 1969) showed promise of meeting our criteria. Therefore, we used a Beckman research potentiometer and Orion fluoride and reference electrodes to do the chemical analyses.

To test the method, we obtained 110 samples analyzed by WARF in 1970 for available fluoride. These samples were analyzed with our specific ion equipment.

We also exchanged a small number of samples with Dr. Clarence Gordon. Mr. Phil Tourangeau, Dr. Gordon's laboratory director, analyzed these samples on an Orion specific ion apparatus.

During our analyses, we checked repeatability of the method by twice analyzing 56 of the samples collected in 1971.

Foliar analysis

Conifer foliage was sorted by year of origin: 1969, 1970, or 1971. Injury index (I.I.), an estimate of the proportion of tissue thought killed by fluorides for foliage of a given year, was measured separately for each year's foliage (Carlson and Dewey 1971). Foliage of each year was then dried, ground, and analyzed chemically for available[5/] fluoride. The method is outlined in Appendix I.

Current (1971) foliage of shrubs, herbs, and grasses was chemically analyzed for available fluoride, but injury index was not measured.

4/ Wisconsin Alumni Research Foundation, Madison, Wisconsin.

5/ All samples collected in 1971 were chemically analyzed in the Forest Service laboratory in Missoula, Montana, using the specific ion method. Samples were not washed; thus fluoride determinations reflected particulate and gaseous states, or "available" fluoride.

Stem analysis

Because analyses of bark beetles in 1970 indicated fluorides may be translocated in the woody tissue of trees, we decided to analyze stem tissue in 1971. Conifer, shrub, and forb stems were sorted by year of wood--1969, 1970, and 1971--and chemically analyzed for available fluoride.

Histological analysis

Needles from 30 different conifer samples showing visible fluoride necrosis were prepared for histological analysis and examined as in the 1970 study (Carlson and Dewey 1971).

Collection and Analysis of Insects

Nine insect samples, all from within one-half mile of the aluminum plant, were collected in mid-August 1971. There were pollinators, including mixed Hymenoptera, wood nymph butterflies (*Cercyonis* sp.), skipper butterflies (*Erynnis* sp.), and mixed Syrphildae; predators, including robberflies (mixed Asilidae) and dragonflies (mixed Anisoptera); and foliage feeders, including Arctridae larvae and Notodontidae larvae. Each was chemically analyzed, unwashed, for available fluoride using the same procedure as used for vegetation. At least 5 grams fresh weight were required for each species. If less than 5 grams was collected, the sample was combined with another sample from the same family.

RESULTS AND DISCUSSION

Specific ion method

Comparative analysis of our results to those of WARF is detailed in Appendix II. Linear regression analysis of the paired data gave a correlation coefficient of 0.98. Slope of the regression line was significant at the 99 percent level. This showed our method gave results comparable to those found by WARF in 1970. Samples checked by the University of Montana were also in close agreement with our determinations.

Data on repeatability is given in Appendix III. The average difference was 5.31 p.p.m. $\pm$ 1.99 p.p.m. at the 95 percent level. Again, this variation was well within the limits we were willing to accept. Thus, we accepted the specific ion method as a reliable way to determine fluoride concentration in biological material. This indicated results obtained in 1971 by the specific ion method could be directly comparable to those of 1970.

Foliar analyses

Results of foliar analyses are listed in Appendix IV. The table is designed to compare 1971 data to 1970 data and, for the plots sampled in 1971, includes:

1. 1970 second sampling: average fluoride value and injury indexes by vegetation class.

2. 1971 average fluoride values and injury indexes. (Grand average here does not include 3-year-old tissue.)

3. Difference (+ or -) in fluoride concentration and I.I. between 1970 and 1971 data, using 1970 as base year.

4. Percent difference of (3).

5. One-year change, from 1970 to 1971, in average fluoride concentration for conifer tissue originating in 1969 and 1970.

6. Total 1970 values, total 1971 values, total differences, average differences, and average percent difference, using 1970 as base year. These are averages of average plot values.

Control data

Control vegetation had basically the same amount of fluoride as in 1970. Only the herbaceous plants, at 11.2 p.p.m., exceeded the 1970 established control concentration of 10 p.p.m. The grand average was 8.91 p.p.m., up 0.57 p.p.m. from 1970. Therefore, we accepted 10 p.p.m. as the normal background or control level of fluoride in foliar tissue.

Needles of 1969 origin increased 2.44 p.p.m. from 1970 to 1971. Needles of 1970 origin increased 3.28 p.p.m.

Injury index remained the same at 0.0. The conservative value of 0.006 is still accepted as control I.I.

Radial data

Vegetation from all plots sampled in 1971 contained above-normal fluoride condentrations. A maximum of 24 times the normal complement of fluoride was found at R4-P2 and a minimum of 1.6 times the control value was found at R4-P6 in Glacier National Park.

Injury index ranged from 21 times control at R3-P3 near the aluminum plant to control levels in Glacier Park. The data indicates visible fluoride injury to vegetation occurred up to 6 air miles from the aluminum plant in 1971.

We felt the most important way to look at the data was to evaluate the change in fluoride accumulation and I.I. from 1970 to 1971. This was done by comparing current and 1-year-old conifer tissue and current shrub, forb, and grass tissue collected in 1971 with the same types and ages of tissue collected in the same plots in 1970. By averaging all plots by vegetational type, we found:

1. Average fluoride in 1971 shrubs was 95.1 p.p.m. lower, down 45.6 percent from 1970.

2. Average fluoride in 1-year-old conifer needles was 63.3 p.p.m. lower, down 45.1 percent from 1970.

3. Average fluoride in current conifer needles was 10.1 p.p.m. lower, down 28.6 percent from 1970.

4. Average fluoride in current herbaceous foliage was 59.4 p.p.m. lower, down 41.5 percent from 1970.

5. Average fluoride in current grass foliage was 46.8 p.p.m. lower, down 38.7 percent from 1970.

Considering all data, not stratifying by vegetational type or plot, the average fluoride in 1971 was down 76.1 p.p.m. (49.3 percent) from 1970. Injury index dropped 0.036 (45.8 percent) from 1970.

We also looked at the 1-year change in fluoride concentration of conifer needles. A 1969 needle sampled in 1970 had a given amount of fluoride; how much additional fluoride did that needle accumulate from 1970 to 1971? Obviously it was not possible to sample the same needle because of the destructive nature of the sampling. Therefore, in 1971 we compared different needles but from the same tree sampled in 1970.

Overall, the data indicates that needles originating in 1969 accumulated, on the average, an additional 3.01 p.p.m. fluoride; while those originating in 1970 accumulated an additional 41.53 p.p.m. This is compared with control data which showed 1969 needles increased 2.44 p.p.m. and 1970 needles 3.28 p.p.m.

Stem analyses

Results of stem analyses are in Appendix V.

Control data.--The range in control tissue was 5.8 p.p.m. in 1971 herbaceous tissue to 11.3 in 1970 conifer tissue. The grand average was 8.4 p.p.m. We accepted 10 p.p.m. as a reasonable control concentration.

Radial data.--Based on vegetation type, the fluoride concentration averaged 11.57 p.p.m. in 1970 shrubs to 19.74 p.p.m. in 1970 conifers. On a plot basis, regardless of vegetation type, the range was from 29.5 p.p.m. at R4-P2 near the company to 9.6 p.p.m. at R5-P9 in Glacier National Park.

Histological analyses

Histological analysis of necrotic conifer needles indicated microscopically the same disease syndrome occurred in 1971 and in 1970. Expanded parenchyma and occluded resin canals were common, causing crushing and collapse of adjacent cells.

Insect analyses

Comparative results between 1970 and 1971 are given in Appendix VI. No controls were analyzed in 1971, so data was compared to 1970 controls.

Accumulations were similar to those found in 1970. All were greater than accumulations in the control samples. Pollinators contained the most fluoride, ranging from 81.3 p.p.m. in *Erynnis* sp. to 585.0 in mixed Hymenoptera. Predators, i.e., Asilidae, dragonflies, and damselflies, accumulated from 21.7 to 82.9 p.p.m., and foliage feeders had from 168 to 255 p.p.m. fluoride.

As mentioned earlier, the Anaconda Company reported it had reduced fluoride emissions at its Columbia Falls, Montana, plant from 7,500 pounds per day in 1970 to 2,500 pounds per day in 1971. This is a 67 percent reduction. Our data indicated an average reduction of fluoride in plant tissue of about 50 percent and an average I.I. reduction of about 46 percent. However, this does not imply the pollution problem is solved.

In our 1971 report (Carlson and Dewey 1971), we presented an isopol map depicting lines of equal average plant tissue fluoride concentration. This map is shown in Figure 1. In Appendix III-A of that report we listed calculated acreages within isopols. The 1971 data was used to recompute and adjust the isopols. The isopol map constructed from this data is shown in Figure 2.

The fluoride grand averages, listed in the 1971 report, were grouped and adjusted downward by the following factors, based on data collected in 1971:

1. Plots 1, 2, and 3 reduced 33.8 percent.

2. Plots 4 and 5 reduced 42.7 percent.

3. Plots 6 through 10 reduced 14.3 percent.

FIGURE 1

Isopols of fluoride pollution at Columbia Falls, Montana. 69,120 acres are included within the 30 isopol.

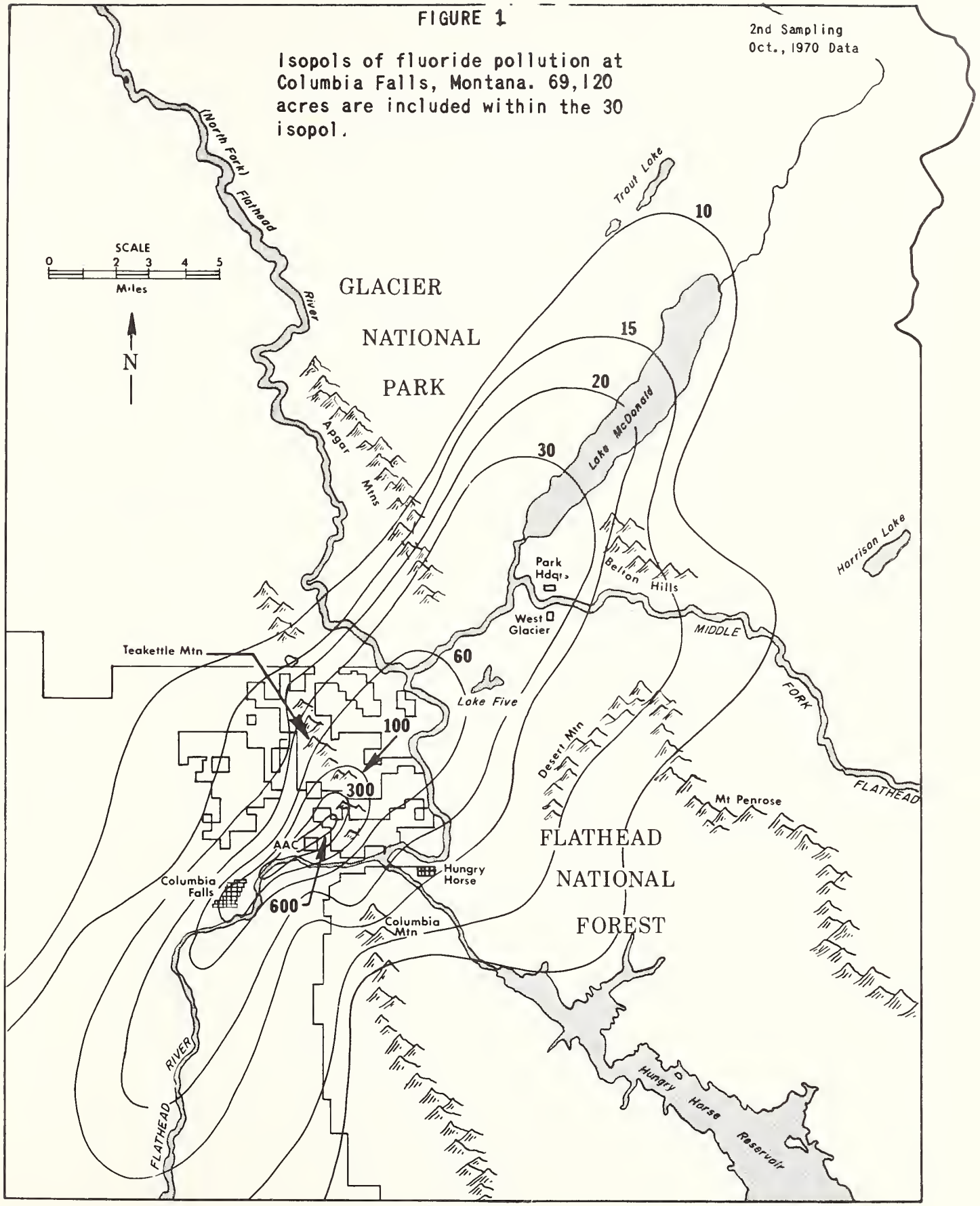
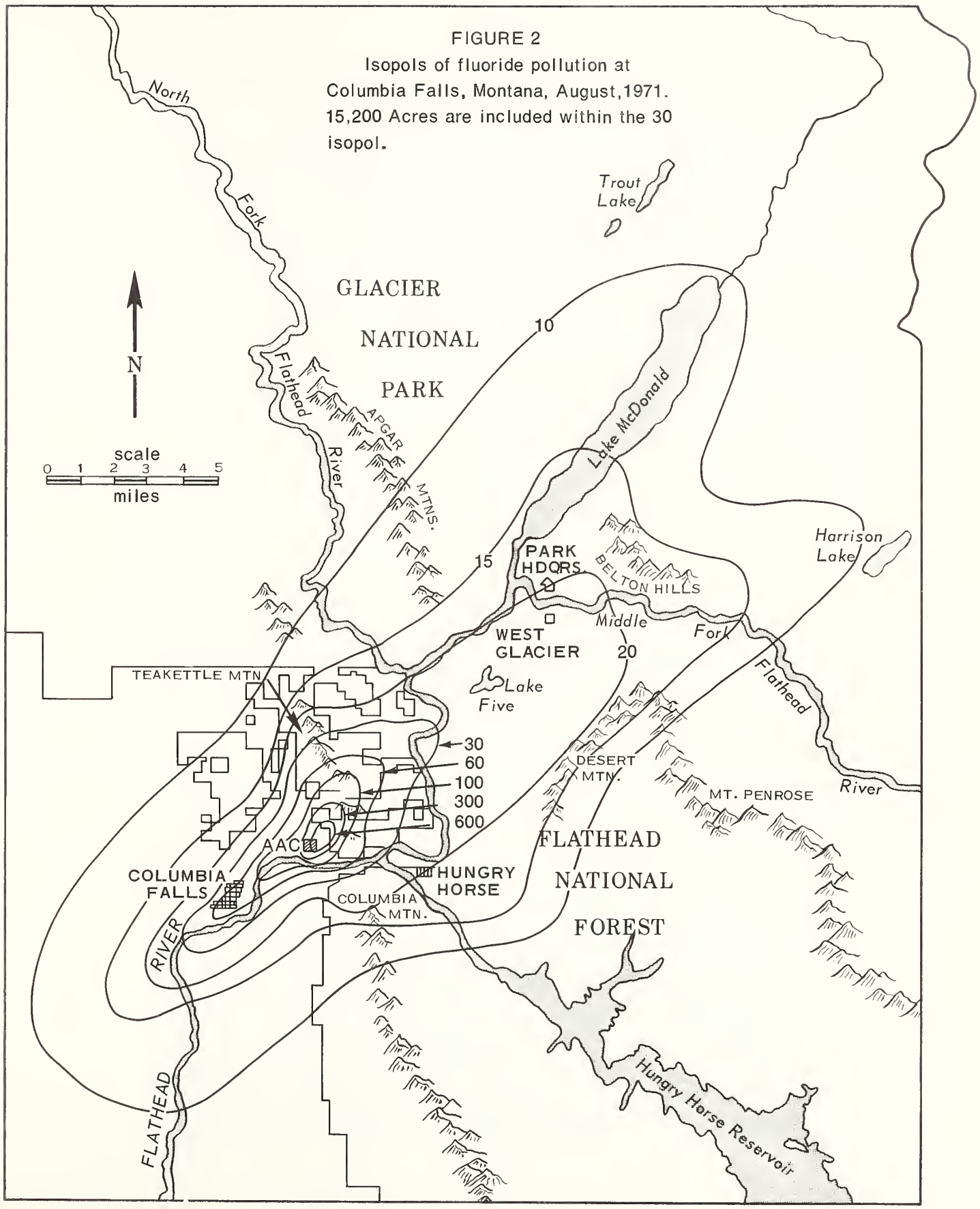


FIGURE 2

Isopols of fluoride pollution at Columbia Falls, Montana, August, 1971. 15,200 Acres are included within the 30 isopol.



Isopols for 1971 were then developed by the same procedure used for 1970 data, using the adjusted 1970 data.

Table 1 shows the area polluted by fluorides as estimated from the new isopol map. The total area polluted (area within the 10 isopol) was 280 square miles (179,200 acres), down 54 square miles (34,560 acres) from 1970. The area sustaining most of the fluoride-induced injury to vegetation (within the 30 isopol) was 23.75 square miles or 15,200 acres. This is down 84.25 square miles or 53,920 acres. The data indicate a critical air pollution problem still exists in the Columbia Falls area.

Table 1.--Area polluted by fluorides

Isopol	All lands		Glacier National Park	
	Square miles	Acres	Square miles	Acres
10	280.00	179,200	100.0	64,000
15	140.00	89,600	20.0	12,480
20	62.00	39,680	1.5	960
30	23.75	15,200		
60	12.00	7,680		
100	5.25	3,360		
300	1.50	960		
600	.75	480		

The area polluted by greater than 10 p.p.m. in Glacier National Park was 100 square miles, down 12 square miles (7,680 acres) from 1970. The 30 and 60 isopols did not extend into the Park as in 1970. The 20 p.p.m. isopol included 1.5 square miles (960 acres) and the 15 isopol included 20 square miles (12,480 acres).

Although the I.I. grand averages in Glacier National Park indicate no injury was found, in R4-P6 fluoride markings were found on 1970 ponderosa pine needles, but were not severe enough to bring the I.I. up to 0.006 or greater.

CONCLUSIONS

In our 1971 report, we indicated that significant reductions in emissions likely would not eliminate fluoride accumulation by vegetation at distant plots, including those in Glacier Park. Current data support that hypothesis. Fluorides in plots R4-P6 and R5-P6 were reduced only 8.0 percent and 4.3 percent, respectively. Vegetation there still is accumulating considerable fluorides, indicated by the 16.1 p.p.m. and 30.3 p.p.m. plot averages computed in this report. It is not unreasonable to hypothesize that even if the aluminum plant reduced fluoride emissions to the State of Montana standard of 864 pounds per day, fluorides will continue to be accumulated by vegetation in Glacier Park.

Stem analyses showed definitely that excessive fluorides do occur either on or within the woody tissue. The exact distribution could be determined by analyzing separately the outer and inner bark and xylem. However, the presence of fluorides either on or in the tissue presents a potential hazard to wildlife browsing on that tissue. Bark beetles may be accumulating these fluorides if they (fluorides) are carried within the vascular system of the tree.

It is not known what effect fluorides are having on insect populations in the study area. We believe that fluorides are being accumulated by insects and passed along the food chain in the area.

In conclusion, it has been determined:

1. Chemical analysis of vegetation in the Columbia Falls area indicates the Anaconda Company aluminum plant likely has reduced its fluoride emissions.

2. The reduction was not enough to prevent vegetation as far away as Glacier Park, up to 12 miles from the Anaconda Aluminum Company plant, from accumulating above-normal amounts of fluoride.

3. Injury to vegetation can still be observed over an extensive area of 15,000 acres.

4. Insects continue to accumulate fluorides.

ACKNOWLEDGEMENTS

I express my appreciation to Mrs. Carma Gilligan for her careful and accurate work in preparing tissues for chemical and histological analysis. Also, a special word of thanks is due Jerald E. Dewey, entomologist in the Northern Region heardquarters, Division of State and Private Forestry, for making the field collections of insects.

Mention of trade names and commercial products in this publication is for information only. No endorsement of the U.S. Department of Agriculture is implied.

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APPENDIX I

CHEMICAL ANALYSIS OF PLANT AND INSECT TISSUE[1/]

Determination of fluoride content in vegetation and insect tissue was done exactly the same. The following procedure was used:

1. Field samples were dried for 2 days at 100° F. in a forced-draft oven.

2. Dried samples were ground in a Wiley mill to pass a 40-mesh screen.

3. 0.5000 gram of dried tissue was weighed into a nickle crucible. 0.0500 gram of low-fluorine calcium oxide was then added.

4. Sample and CaO were slurried with distilled water.

5. Slurry was dried and charred under infrared oven.

6. Charred material was ashed for 16 hours at 600° C. in a muffle furnace.

7. After ashing, crucibles were allowed to cool in a dessicator.

8. Ashed samples were moistened with distilled water, then 3 ml. of 30 percent perchloric acid were added to dissolve the ash.

9. Dissolved ash was then brought to 100 ml. volume by adding TISAB[2/] diluted 50 percent by distilled water.

10. Fluoride activity was determined using a Beckman research pH meter and Orion fluoride and reference electrodes.

1/ From Dr. C. C. Gordon, University of Montana.

2/ Trade name of Orion's product which adjusts pH and total ionic strength of sample solution.

APPENDIX II

COMPARATIVE FLUORIDE ANALYSES WITH WARF[1/]

Data pair	WARF result	Our result	Data pair	WARF result	Our result
1	306.0	147.0	39	19.5	31.0
2	600.0	455.0	40	97.5	113.0
3	469.0	329.0	41	23.0	26.4
4	825.0	294.0	42	78.5	79.0
5	2,750.0	1,440.0	43	10.0	20.8
6	10.5	25.6	44	21.0	26.2
7	1,288.0	595.0	45	31.5	27.6
8	113.0	127.9	46	32.0	38.6
9	198.0	120.0	47	11.5	33.6
10	37.5	44.4	48	104.0	87.2
11	11.5	65.0	49	115.0	118.0
12	3.0	42.0	50	1,425.0	830.0
13	21.0	70.8	51	32.5	40.8
14	99.0	132.0	52	385.0	172.0
15	38.5	79.0	53	293.0	224.0
16	13.5	34.0	54	12.5	10.0
17	19.0	29.6	55	29.0	35.6
18	12.0	39.6	56	22.0	29.8
19	31.0	112.0	57	30.5	30.4
20	91.5	136.0	58	32.5	29.6
21	23.0	52.0	59	30.5	39.6
22	29.0	47.4	60	12.5	34.0
23	113.0	112.0	61	44.5	45.8
24	139.0	162.0	62	16.0	28.4
25	15.0	73.0	63	33.0	30.5
26	16.0	28.0	64	9.0	26.4
27	275.0	236.0	65	508.0	256.0
28	14.0	23.6	66	73.0	75.0
29	198.0	124.0	67	750.0	352.0
30	200.0	164.0	68	16.5	28.0
31	45.0	54.0	69	28.5	23.6
32	59.5	64.4	70	27.0	40.8
33	63.5	50.0	71	76.0	52.4
34	178.0	114.5	72	10.0	16.4
35	8.0	27.8	73	325.0	290.0
36	14.5	23.6	74	168.0	152.0
37	295.0	264.0	75	55.0	65.6
38	293.0	250.0	76	8.0	22.4

1/ P.p.m. fluoride, dry weight basis.

APPENDIX II

COMPARATIVE FLUORIDE ANALYSES WITH WARF[1/] (con.)

Data pair	WARF result	Our result
77	75.0	78.0
78	135.0	112.0
79	14.0	20.4
80	30.5	33.4
81	11.5	39.0
82	10.0	16.4
83	52.5	55.6
84	39.0	62.4
85	25.5	70.0
86	18.5	50.4
87	1,375.0	780.0
88	218.0	152.0
89	190.0	188.0
90	37.0	31.6
91	60.0	69.0
92	130.0	158.0
93	32.5	41.0
94	26.5	52.0
95	6.3	30.2
96	72.5	66.0
97	319.0	134.0
98	8.5	58.2
99	25.5	35.8
100	35.0	44.4
101	15.5	12.4
102	27.5	41.5
103	32.0	51.8
104	96.5	78.4
105	5.5	14.0
106	28.5	34.2
107	16.5	34.0
108	16.0	24.6
109	7.5	10.1
110	293.0	264.0

F ratio for slope = 2685.97

Correlation (r) = 0.98

1/ P.p.m. fluoride, dry weight basis.

APPENDIX III

REPEATABILITY OF SPECIFIC ION FLUORIDE ANALYSES[1/]

Crucible number	Check crucible number	Original analysis	Check analysis	Difference	Percent difference
3	57	20.0	20.0	0.0	0.0
41	56	126.0	136.4	10.4	8.3
12	55	16.4	15.2	1.2	7.3
4	53	28.4	22.4	6.0	21.1
6	52	47.6	39.8	7.8	16.4
13	51	61.8	74.2	12.4	20.0
54	92	74.2	66.4	7.8	10.5
93	98	48.6	48.0	0.6	1.2
84	99	210.0	214.0	4.0	1.9
74	104	10.8	13.4	2.6	24.0
115	122	23.6	27.8	4.2	17.8
128	143	72.2	71.4	0.8	1.1
144	154	15.2	14.9	0.3	2.0
153	161	9.4	11.6	2.2	23.4
151	170	15.2	18.0	2.8	18.4
168	178	12.8	15.2	2.4	18.7
182	187	63.0	60.8	2.2	3.5
199	203	23.2	22.6	0.6	2.6
223	229	14.0	12.6	1.4	10.0
202	230	19.0	18.4	0.6	3.2
232	244	10.2	14.1	3.9	38.2
251	263	46.0	46.4	0.4	0.9
265	274	8.6	8.6	0.0	0.0
252	285	72.2	87.0	14.8	20.5
264	297	71.0	64.0	7.0	9.9
272	309	64.2	51.0	13.2	20.6
318	322	124.0	152.0	28.0	22.6
292	352	240.0	284.0	44.0	18.3
348	350	30.4	18.8	11.6	38.2
362	365	9.6	8.2	1.4	14.6
377	382	21.0	21.6	0.6	2.9
378	384	43.0	49.0	6.0	14.0
386	400	23.6	27.0	3.4	14.4
394	415	14.6	5.0	9.6	61.9
388	424	9.4	16.2	6.8	72.3
382	434	21.6	35.8	14.2	65.7

1/ P.p.m. fluoride, dry weight basis.

APPENDIX III

REPEATABILITY OF SPECIFIC ION FLUORIDE ANALYSES[1/] (con.)

Crucible number	Check Crucible number	Original analysis	Check analysis	Difference	Percent difference
437	450	5.8	6.6	0.8	13.8
440	451	13.8	18.2	4.8	34.8
438	452	12.0	17.0	5.0	41.7
2		30.8	29.2	1.6	5.6
5		20.0	19.0	1.0	5.0
7		49.2	47.8	1.4	2.8
9		20.8	21.8	1.0	4.8
10		21.5	16.0	5.5	34.4
11		292.0	306.0	14.0	4.6
14		10.4	11.6	1.2	10.3
15		10.1	10.4	0.3	2.9
16		100.4	102.4	2.0	2.0
17		11.8	14.4	2.6	18.0
18		19.6	24.4	4.8	19.7
19		16.4	21.2	4.8	22.6
20		64.2	62.6	1.6	2.6
21		17.2	17.8	0.6	3.4
22		32.6	38.4	5.8	15.1
24		27.2	24.2	3.0	12.4
31		16.8	17.0	0.2	1.2
TOTAL				297.2	884.1
AVERAGE				5.31	15.8

S x = .9932
t 56 = 2.00
.05

Confidence interval
at 95 percent level = 5.31 $\pm$ 1.9864

1/ P.p.m. fluoride, dry weight basis.

APPENDIX IV

SUMMARY OF 1971 DATA[1/]

	Shrubs	Conifers Age of foliage 2-Year	Conifers Age of foliage 1-Year	Conifers Age of foliage Current	Herbs	Grass	Grand average	I.I. average	High I.I.	1 yr. change[2/] in average F^- concentration Yr. tissue orig. 1969	1970
Control											
1970	8.13		6.61	5.95	9.50	10.56	8.34	.000	.000		
1971	8.45	9.05	9.23	8.55	11.20	8.00	8.91	.000	.000	+2.44	+3.28
Diff.	+.32		+2.62	+2.60	+1.70	-2.56	+.57	.000	.000		
%	+3.90		+39.60	+43.70	+17.90	-24.20	+.07				
R2-P2											
1970	147.50		100.80	26.80	146.00	44.50	92.6	.211	.279	+33.30	+62.70
1971	111.00	134.10	89.50	22.90	121.00	54.40	75.0	.022	.029		
Diff.	-36.50		-11.30	-3.90	-25.00	+9.90	-17.6	-.189	-.250		
%	-24.70		-11.20	-14.50	-17.10	+22.20	-19.0	-89.600	-89.600		
R3-P3											
1970	281.50		294.50	70.00	137.00	168.00	199.6	.156	.313	-54.50	+68.00
1971	192.00	240.00	138.00	58.00	71.80	72.20	102.1	.125	.258		
Diff.	-89.50		-156.50	-12.00	-65.20	-95.80	-97.5	-.031	-.055		
%	-31.80		-53.10	-17.10	-47.60	-57.00	-48.8	-19.900	-17.600		
R3-P4											
1970	130.00		85.50	23.00	107.00	68.00	90.6	.034	.043	+3.50	+42.00
1971	87.00	89.00	65.00	21.20	83.50	72.00	62.3	.035	.066		
Diff.	-43.00		-20.50	-1.80	-23.50	+4.00	-28.3	+.001	+.023		
%	-33.10		-24.00	-7.80	-22.00	+5.90	-31.2	+2.900	+53.500		

1/ All values are p.p.m. fluoride.

2/ Conifers only.

APPENDIX IV

SUMMARY OF 1971 DATA[1/] (con.)

	Shrubs	Conifers Age of foliage 2-Year	Conifers Age of foliage 1-Year	Conifers Age of foliage Current	Herbs	Grass	Grand average	I.I. average	High I.I.	1 yr. change[2/] in average F^- concentration Yr. tissue orig. 1969	1970
R4-P2											
1970	1244.00				638.0	205.0	832.80				
1971	324.00	700.0	340.0	111.1	240.0	144.0	241.80	.066	.133		
Diff.	-920.0				-398.0	-61.0	-591.00				
%	-74.0				-62.4	-29.8	-71.00				
R5-P3											
1970	294.5		537.5	80.3		508.0	332.20	.132	.250	+32.5	+157.2
1971	284.0	570.0	237.5	50.6	126.0	220.0	172.30	.048	.066		
Diff.	-10.5		-300.0	-29.7		-288.0	-160.00	-.084	-.184		
%	-3.6		-55.8	-37.0		-56.7	-48.20	-63.600	-73.600		
R5-P4											
1970	202.2		228.7	55.0	111.0	128.0	160.00	.063	.114	-116.1	+15.8
1971	53.0	112.6	70.8	16.4	12.4	13.4	36.20	.122	.158		
Diff.	-149.2		-157.9	-38.6	-98.6	-114.6	-123.80	+.059	+.044		
%	-73.7		-69.0	-70.2	-88.8	-89.5	-77.40	+93.700	+38.600		
R5-P5											
1970	59.5		56.5	23.3	270.0	51.0	66.91	.014	.014	+9.6	+11.9
1971	64.6	66.1	35.2	12.8	32.6	41.8	33.60	.025	.031		
Diff.	+5.1		-21.3	-10.5	-237.4	-9.2	-33.30	+.011	+.017		
%	+8.6		-37.8	-45.1	-87.9	-18.0	-49.80	+78.600	+121.000		

1/ All values are p.p.m. fluoride.

2/ Conifers only.

APPENDIX IV

SUMMARY OF 1971 DATA[1/] (con.)

	Shrubs	Conifers Age of foliage 2-Year	Conifers Age of foliage 1-Year	Conifers Age of foliage Current	Herbs	Grass	Grand average	I.I. average	High I.I.	1 yr. change[2/] in average F^- concentration Yr. tissue orig. 1969	1970
R5-P6											
1970	38.0		35.0	11.7	59.5	32.0	30.3	.003	.003	-0.9	+6.4
1971	49.2	34.1	18.1	15.8	56.2	30.0	29.0	.036	.036		
Diff.	+11.2		-16.9	+4.1	-3.3	-2.0	-1.3	+.033	+.033		
%	+29.5		-48.3	+35.0	-5.5	-6.2	-4.3				
R6-P3											
1970	169.5		239.6	44.7	171.0	117.0	148.0	.114	.144	+88.4	+127.3
1971	112.0	328.0	172.0	43.9	124.0	74.2	106.0	.100	.234		
Diff.	-57.5		-67.6	-.8	-47.0	-42.8	-42.0	-.014	+.090		
%	-33.9		-28.2	-2.8	-27.5	-36.6	-28.4	-12.300	-62.500		
R6-P4											
1970	64.2		140.0	76.3	113.0	53.0	83.8	.182	.291	-56.4	-31.0
1971	48.0	83.6	45.3	20.9	46.0	37.2	37.7	.029	.033		
Diff.	-16.2		-94.7	-55.4	-67.0	-15.8	-46.1	-.153	-.258		
%	-25.2		-67.6	-72.6	-59.3	-29.8	-55.0	-84.100	-88.700		
R7-P3											
1970	154.5		142.5	31.3	143.0	160.0	120.0	.105	.154	+77.5	+90.6
1971	200.0	220.0	121.9	41.4	234.0	184.0	134.9	.034	.068		
Diff.	+45.5		-20.6	+10.1	+91.0	+24.0	+14.9	-.071	-.086		
%	+29.4		-14.5	+32.3	+63.6	+15.0	+12.4	-67.600	-55.800		

1/ All values are p.p.m. fluoride.

2/ Conifers only.

APPENDIX IV

SUMMARY OF 1971 DATA[1/] (con.)

	Shrubs	Conifers: Age of foliage 2-Year	Conifers: Age of foliage 1-Year	Conifers: Age of foliage Current	Herbs	Grass	Grand average	I.I. average	High I.I.	1 yr. change[2/] in average F^- concentration Yr. tissue orig. 1969	1970
R8-P5											
1970	33.5		28.1	14.5	31.5	14.5	24.1	0.000	0.000	+1.1	+3.7
1971	25.8	29.2	18.2	10.8	17.8	9.0	15.8	.005	.005		
Diff.	-7.7		-9.9	-3.7	-13.7	-5.5	-8.3	+.005	+.005		
%	-23.0		-35.2	-25.5	-43.5	-37.9	-34.4				
Col. Mt.											
1970			38.2	15.3	25.5		26.6	.081	.160	+11.0	+7.0
1971	44.0	49.2	22.3	19.8	72.2	12.2	28.3	.014	.023		
Diff.			-15.9	+4.5	+46.7		+1.7	-.067	-.137		
%			-41.6	+29.4	183.0		+6.4	-82.700	-85.600		
R4-P6											
1970	32.5		11.7	9.3	15.5		17.5	0.000	0.000	+14.5	+10.0
1971	18.1	26.2	19.3	7.9	28.8	11.8	16.1	.006	.009		
Diff.	-14.4		+7.6	-1.4	+13.3		-1.4	+.006	+.009		
%	-44.3		+65.0	-15.1	+85.8		-8.0				
R5-P9											
1970	69.0		24.8	14.2	32.5	22.0	26.9	.014	.019	-1.4	+9.9
1971	20.4	23.4	24.1	11.5	29.0	10.1	18.7	0.000	0.000		
Diff.	-48.6		-0.7	-2.7	-3.5	-11.9	-8.2	-.014	-.019		
%	-70.4		0.0	-19.0	-10.8	-54.1	-30.5	-100.000	-100.000		

1/ All values are p.p.m. fluoride.

2/ Conifers only.

APPENDIX IV

SUMMARY OF 1971 DATA[1/] (con.)

	Shrubs	Conifers: Age of foliage 2-Year	Conifers: Age of foliage 1-Year	Conifers: Age of foliage Current	Herbs	Grass	Grand average	I.I. average	High I.I.	1 yr. change[2/] in average F^- concentration: Yr. tissue orig. 1969	1970
Total 1970	2920.40		1963.40	495.7	2000.5	1571.00	2251.91	1.109	1.784	42.10	581.50
Total Diff.	1331.30		886.20	-141.8	-831.2	-608.70	-1142.11	-.508	-.768		
Total 1971	1589.10	2705.5	1077.20	353.9	1169.3	962.30	1109.80	.601	1.016		
N	14.00		14	14	14	13	15	14	14	14	14
Average Diff.	-95.10		-63.30	-10.1	-59.4	-46.80	-76.10	-.036	-.055	3.01	41.53
% Diff.	-45.60		-45.10	-28.6	-41.5	-38.70	-49.30	-45.800	-43.000		

APPENDIX V

SUMMARY OF STEM ANALYSES[1/]

Plot	Shrubs Year of tissue 1969	Shrubs Year of tissue 1970	Shrubs Year of tissue 1971	Conifers Year of tissue 1969	Conifers Year of tissue 1970	Conifers Year of tissue 1971	Herbs 1969	Grand average
Control	7.8	9.5	6.9	9.3	11.3	5.9	5.8	8.4
R2-P2	11.6	19.8	12.8	25.5	25.7	24.7	23.8	22.0
R3-P3	10.2	10.4	12.0	24.2	22.7	27.3	16.4	19.7
R3-P4	7.2	7.6	10.8	10.6	16.8	15.8	11.6	13.4
R4-P2	16.6	15.1	37.6	35.0	37.4	41.1	22.6	29.9
R5-P3	19.8	15.2	20.0	19.3	35.5	35.4	--	26.2
R5-P4	9.4	13.0	19.8	16.6	12.8	17.6	10.8	14.7
R5-P5	11.4	8.4	7.8	13.6	14.6	14.2	11.6	12.2
R5-P6	11.8	9.8	15.1	11.3	10.1	11.2	--	11.3
R6-P3	15.2	22.8	16.2	20.4	21.0	17.7	21.0	19.3
R6-P4	14.0	11.6	16.6	21.5	14.6	25.5	14.0	18.9
R7-P3	17.0	20.2	29.8	20.2	19.0	18.3	10.2	19.2
R8-P5	14.8	12.6	9.8	13.2	14.2	10.4	11.2	12.4
Col. Mt.	11.3	9.0	9.8	28.4	21.5	13.8	17.2	15.0
R5-P9	8.5	7.6	6.4	8.4	13.9	7.4	10.4	9.2
R4-P6	10.6	6.8	14.8	17.8	8.7	10.4	11.6	11.1
Veg. type average	12.69	11.57	16.85	19.46	19.74	19.39	14.80	17.10

1/ All values are p.p.m. fluoride, dry weight basis.

APPENDIX VI

FLUORIDE ACCUMULATIONS BY INSECTS[1/]

	Date collected	P.p.m. fluoride
Pollinators		
Bumblebees	8/12/70	406.0[1/]
Mixed Hymenoptera	8/16/71	585.0
Wood nymph butterfly - *Cercyonis* sp.	8/12/70	58.0
Wood nymph butterfly - *Cercyonis* sp.	8/16/71	144.0
Skipper butterfly - *Erynnis* sp.	8/12/70	146.0
Skipper butterfly - *Erynnis* sp.	8/16/71	81.3
Mixed Syraphildae	8/16/71	140.0
Mixed Syraphildae	None collected in 1970	
Predators		
Robberflies - mixed Asilidae	8/16/71	82.9
Robberflies - mixed Asilidae	None collected in 1970	
Dragonflies - mixed Anisoptera	8/16/71	24.8
Damselflies - *Argia* sp.	6/1/70	21.7
Foliage Feeders		
Arctiidae (larvae)	8/16/71	255.0
Notodontidae (larvae)	8/16/71	168.0

1/ P.p.m. fluoride, dry weight basis. Where possible, 1970 results are given for comparative purposes.