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EFFECTS OF FLUORIDE EMISSIONS FROM A MODERN PRIMARY ALUMINUM SMELTER ON A LOCAL POPULATION OF WHITE-TAILED DEER (*ODOCOILEUS VIRGINIANUS*)

J. S. Suttie,[1] R. Dickie,[2] A. B. Clay,[1] Per Nielsen,[1] W. E. Mahan,[3] D. P. Baumann,[3] and R. J. Hamilton[3]

ABSTRACT: The influence of fluoride emissions from a modern aluminum smelter on concentrations of skeletal fluoride and dental fluorosis in a resident population of white-tailed deer was studied. The smelter was located on Mount Holly Plantation in South Carolina, and concentrations of skeletal fluoride in the deer collected at Mount Holly increased approximately five-fold 3 yr after the operation began. Increases in skeletal fluoride of less than two-fold were observed in deer obtained from Medway Plantation which has its nearest boundary 1.6 km from the smelter site. No dental fluorosis was observed in deer collected at Medway Plantation, but mild dental fluorosis was observed in a significant number of deer collected at Mount Holly Plantation. The dental fluorosis that was observed was not associated with incisor wear or with fluoride-induced molar wear. Osteofluorosis of mandibles or metacarpals was not observed in any of the deer obtained from either plantation. The data obtained from this study indicated that the presence of a modern aluminum smelter caused a detectable increase in concentration of skeletal fluoride in the resident population of white-tailed deer, but that no adverse health effects were seen.

INTRODUCTION

Fluoride emissions from industrial sources constitute a known hazard to grazing domestic animals (National Research Council, 1971, 1974). These emissions also represent a potential threat to the health of wild ungulates. Signs of excessive fluoride ingestion have been observed in various species of deer obtained in areas surrounding primary aluminum smelters or phosphate fertilizer plants or in areas with naturally occurring high-fluoride water (Robinette et al., 1957; Karstad, 1967; Kay et al., 1975; Newman and Yu, 1976; Newman and Murphy, 1979; Shupe et al., 1984). These reports represented the chance accumulation of necropsy material, and it has not been possible to assess the relationship between the degree of dental fluorosis or osteofluorosis observed and fluoride intake from a particular source. It has also not always been possible to determine if reported concentrations of skeletal fluoride were the result of consumption of vegetation containing high concentrations of fluoride or if water with naturally occurring high concentrations had been consumed. The industrial facilities implicated in the available reports have not represented the most modern technology in air pollution control, and the potential hazard to wild animals in the vicinity of a more recently constructed facility has not been assessed. This report describes the results of a systematic study of the influence of a modern primary aluminum smelter on alterations of concentrations of skeletal fluoride and appearance of dental fluorosis in a local population of white-tailed deer.

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MATERIALS AND METHODS

A modern pre-bake primary aluminum reduction plant was constructed near Charleston, South Carolina, by Alumax of South Carolina in 1978–1980. It consists of two lines of Alcoa 697 reduction cells with a production capacity of about 182,000 metric tons of aluminum per

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yr. The plant began operation in June 1980, reached full capacity in December 1980, and ran at 100% capacity until the last specimens of deer were collected in the Fall of 1983. The plant was the only completely new smelter built in the U.S. since the passage of the 1977 amendments to the clean air act and meets Best Available Control Technology and New Source Performance Standard (1976) requirements. Fluoride emissions from the smelting operation are controlled by a dry scrub system, and fluoride emissions over the study period were in the range of 0.4–0.5 kg F/ton of aluminum produced.

Deer were monitored for fluoride effects on two plantations, Mount Holly and Medway. The Alumax Mount Holly Plantation is 2,530 ha in size with 120 ha being used for the aluminum smelter site. The remaining acreage is unfenced and operated as a tree farm and wildlife refuge used for environmental research. The property consists mainly of timbered lands with bottomland hardwoods covering 485 ha, upland pine covering 1,420 ha, and mixed pine hardwood stands covering 200 ha. The remainder of the land consists of fields, roads, buildings, ponds, and power line rights of way. The forest is managed by controlled burning, thinning, and harvesting. During 1978, 1979, and 1980, about 120 ha of soybeans were planted. In 1981 and subsequent years this was reduced to 40 ha in cowpeas, soybeans, corn, and wheat rotations planted solely for wildlife consumption. Medway Plantation is located due east of Mount Holly Plantation and its closest border is about 1.6 km from the smelter site. It is unfenced and for the most part covered with pine stands that are generally more mature than those at Mount Holly. Managed timber harvest is practiced. Approximately 25 ha of rotating crops are planted for wildlife feed. Under a regulated hunting program, about 150 deer are harvested annually by archers. The majority of the deer at both plantations were harvested from August through October. Estimates of deer densities during the period of the study ranged from 4 to 8 ha/deer at Mount Holly, and about 4 ha/deer at Medway.

Prevalence of dental fluorosis and changes in the concentration of skeletal fluoride in deer from both plantations were monitored from 1979 (pre start-up) through 1983. This study period included 3 yr during which the smelter was operated at full capacity. Mandibles, including the incisor teeth, and a metacarpal bone were obtained from at least three deer of each sex of ages <1 yr (fawns), 1½ yr, and 2½ or greater yr at each plantation annually. Age of mature deer was assessed by tooth development and wear (Severinghaus, 1949). These samples were provided by the manager of Medway Plantation from deer killed by archers or were collected on Mount Holly Plantation by representatives of the South Carolina Wildlife and Marine Resources Department. The Wildlife and Marine Resources Department also provided material from a limited number of deer obtained on the Waterhorn Hunt Unit of the Francis Marion National Forest. This area is about 130 km from the Alumax Mount Holly smelter. Mandibles and metacarpals were sampled as described by Suttie et al. (1985), and fluoride analyses were carried out with a specific fluoride ion electrode as described by Singer and Armstrong (1968) and are expressed as ppm F ash wt. Incisors were inspected for fluorotic lesions of the enamel utilizing a light transmitted from behind the tooth. The crowns of the right second incisor were cut from the jaw, ground to pass a 250 micron screen, and enamel and dentin separated as described by Manly and Hodge (1939). Enamel flouride concentration was determined with a specific ion electrode (Singer and Armstrong, 1968) and expressed as ppm F dry wt. A vegetative fluoride monitoring program of the area was conducted by Environmental Strategies Inc., 309 The Parkway, Ithaca, New York 14850, USA. Statistical analysis of data was performed by analysis of variance on log transformations utilizing Duncan's new multiple range test as described by Steel and Torrie (1960).

RESULTS AND DISCUSSION

Data on the fluoride content of mandible and metacarpal bones obtained from both plantations for all 5 yr of the study are shown in Table 1. Statistical analysis of the data indicated that there was no difference due to sex, and data from males and females have been pooled in Table 1. For most age groups and most time periods, the fluoride content of the mandible was slightly but not statistically significantly higher than that of the metacarpal, and the coefficient of variation within groups was slightly higher for the mandible samples. These differences were not large, and either bone can serve as a satisfactory sample to monitor skeletal fluoride accumulation. Skeletal fluoride content of samples obtained in 1979

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TABLE 1. Concentration[a] of fluoride (ppm F, ash wt) in bones from white-tailed deer collected at three locations in South Carolina.

		Mt. Holly			Medway			Waterhorn		
Year	Age	*n*	Metacarpal	Mandible	*n*	Metacarpal	Mandible	*n*	Metacarpal	Mandible
1979	Fawn	7	74 ± 24[b]	52 ± 31[b]	6	53 ± 13	38 ± 13[b]			
	1½	7	169 ± 71[b]	182 ± 69[b]	6	111 ± 36[d]	134 ± 51[d]	4	120 ± 25	118 ± 46
	2½+	6	260 ± 51[c]	286 ± 100[c]	6	168 ± 74[b]	225 ± 109[e]			
1980	Fawn	7	261 ± 172	205 ± 132	6	88 ± 36	79 ± 50	2	61 ± 7	65 ± 10
	1½	6	376 ± 183[e]	570 ± 269	6	166 ± 83	195 ± 73	1	94	137
	2½+	6	465 ± 299[f]	485 ± 206[c]	6	309 ± 186	383 ± 250	2	147 ± 12	200 ± 70
1981	Fawn	10	407 ± 605	473 ± 765	5	76 ± 43	88 ± 33	1	47	68
	1½	7	700 ± 510	868 ± 639	7	208 ± 52	270 ± 51	2	211 ± 98	268 ± 123
	2½+	7	630 ± 223	923 ± 328	7	280 ± 66	391 ± 148	2	198 ± 20	278 ± 30
1982	Fawn	8	270 ± 130	276 ± 145	6	81 ± 79	86 ± 74	1	29	27
	1½	7	801 ± 414	811 ± 407	6	264 ± 93	286 ± 109			
	2½+	8	1,263 ± 949	1,510 ± 1,058	6	408 ± 186	461 ± 219	5	213 ± 48	226 ± 47
1983	Fawn	14	340 ± 233	337 ± 221	6	105 ± 32	96 ± 22			
	1½	7	508 ± 130	586 ± 215	6	164 ± 19	138 ± 49	1	171	231
	2½+	9	1,132 ± 650	1,275 ± 624	6	265 ± 70	369 ± 233	6	276 ± 86	325 ± 129

[a] Values are mean ± SD for the number of deer indicated.
[b] Concentration is less than that of the same age group at this plantation in all other years ($P < 0.05$).
[c] Concentration is less than that of the same age group at this plantation in 1981, 1982, and 1983 ($P < 0.05$).
[d] Concentration is less than that of the same age group at this plantation in 1981 and 1982 ($P < 0.05$).
[e] Concentration is less than that of the same age group at this plantation in 1982 ($P < 0.05$).
[f] Concentration is less than that of the same age group at this plantation in 1982 and 1983 ($P < 0.05$).

represented fluoride exposure in this area before the smelter was in operation. These values were low and ranged from about 50 ppm F in fawns to 200–300 ppm F in animals 2½ yr or older. Concentrations of skeletal fluoride in all age groups increased at Mount Holly in 1980, and further increases were seen in the older animals during 1981. There was a large variation in concentration of skeletal fluoride within each age group, and there was no statistically significant increase beyond 1981. Small increases in skeletal fluoride above that observed in 1979 were seen also at Medway following the start-up of the smelter. The concentrations of skeletal fluoride in the metatarsal are illustrated in Figure 1.

The most sensitive criterion of fluoride ingestion by young animals is the appearance of characteristic fluoride-induced mottling of the developing permanent dentition (National Research Council, 1971, 1974). The incisor teeth of all deer collected after the smelter operation was started were inspected. The data in Table 2 indicated that discernible mottling was present in a significant number of the animals obtained from the Mount Holly Plantation. The mottling observed was not severe, and with one exception the teeth would only have been scored as a no. 2 (slight effect) by the scoring system used to assess dental fluorosis in cattle (National Research Council, 1974). The two deer from Medway Plantation and three of the 12 deer from Mount Holly Plantation had dental lesions that were discernible, but did not appear to be typical of fluoride-induced mottling. These teeth were given a score of 1 (questionable effect). The most severe fluoride-induced dental lesion observed in a deer obtained at Mount Holly Plantation and an example of the lack of any discernible effect seen at Medway Plantation are illustrated in Figure 2. In addition to causing lesions of the developing incisors, fluoride ingestion can in-

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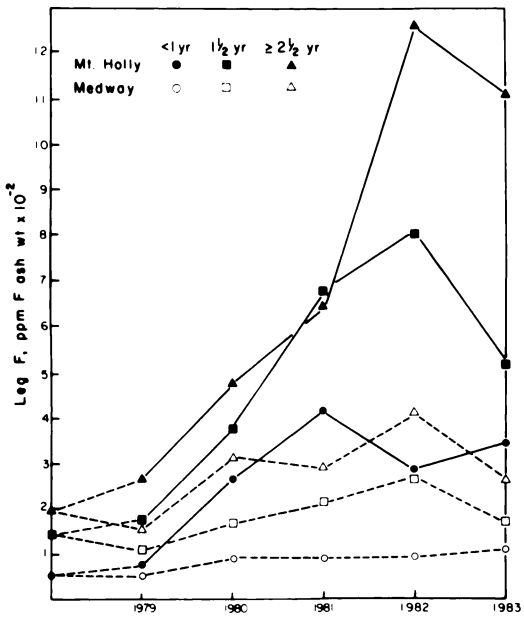

FIGURE 1. Changes of metacarpal fluoride content of white-tailed deer at two plantations near the Alumax, South Carolina aluminum smelter with time. The values plotted are averages of bones from both male and female deer, and the values plotted on the ordinate are the means of values obtained from the Waterhorn Hunt Unit for all 5 yr.

fluence the rate of wear of the molar teeth, leading to an extremely uneven attrition line (Robinette et al., 1957; Karstad, 1967; Shupe et al., 1984). Fluoride-induced effects on molar wear were not observed in any of the deer collected from either plantation.

Dental fluorosis scores are rather subjective, and it has been shown (Mortenson et al., 1964; Suttie and Faltin, 1971, 1973; Suttie et al., 1972) that the enamel fluoride concentration and the amount of enamel in the tooth are more objective measures that are correlated with these subjective scores. Mature enamel is an avascular tissue and enamel fluoride content, therefore, is related to the animal's fluoride intake at the time the tooth was forming. Both enamel hypoplasia and an increased rate of wear will decrease the ratio of enamel to dentin in the tooth

TABLE 2. Prevalence of dental mottling in white-tailed deer* at Mount Holly and Medway Plantations, South Carolina.

Area	Deer with mottling/total deer			
	1980	1981	1982	1983
Medway	0/8	1/11	1/11	0/10
Mount Holly	0/11	3/13	2/15	7/13

* Values are for the number of deer 1½ yr or more older that had incisors that would be scored no. 1 (questionable effect), no. 2 (slight fluorosis) or no. 3 (moderate effect) by the scoring system used to assess bovine dental fluorosis (National Research Council, 1974). Only one tooth with a fluorosis score more severe than no. 2 was observed. Incisors were not saved on all mandibles collected, and less than 12 deer were, therefore, available for inspection in some years.

crown. The fluoride content of the enamel and the percent of the 2nd incisor crown that was enamel for all deer that formed teeth after the smelter was started are shown in Figure 3. It can be seen that the percent of the crown that is enamel is somewhat lower in older deer and that fluoride-induced mottling was seen only in those teeth where the incisor enamel F concentration exceeded 300 ppm F. In general, the incisor crowns of animals with observable dental fluorosis also had a decreased amount of enamel. This was particularly noticeable in those teeth that had the higher concentrations of fluoride in the enamel.

Concentrations of fluoride within the avascular enamel are much lower than skeletal tissue or dentin but do give an indication of what the fluoride intake was at a given time in the animal's history. The data in Table 3 show a slight but statistically significant increase in enamel fluoride of the 2nd incisor at Medway Plantation following the start-up of the smelter, and a three-fold increase at Mount Holly. The measured increase in enamel fluoride at Medway Plantation indicates the sensitivity of this biological monitor of increased fluoride intake. The vegetation monitoring program carried out by Environmental Strategies for Alumax of South Carolina included samples of some

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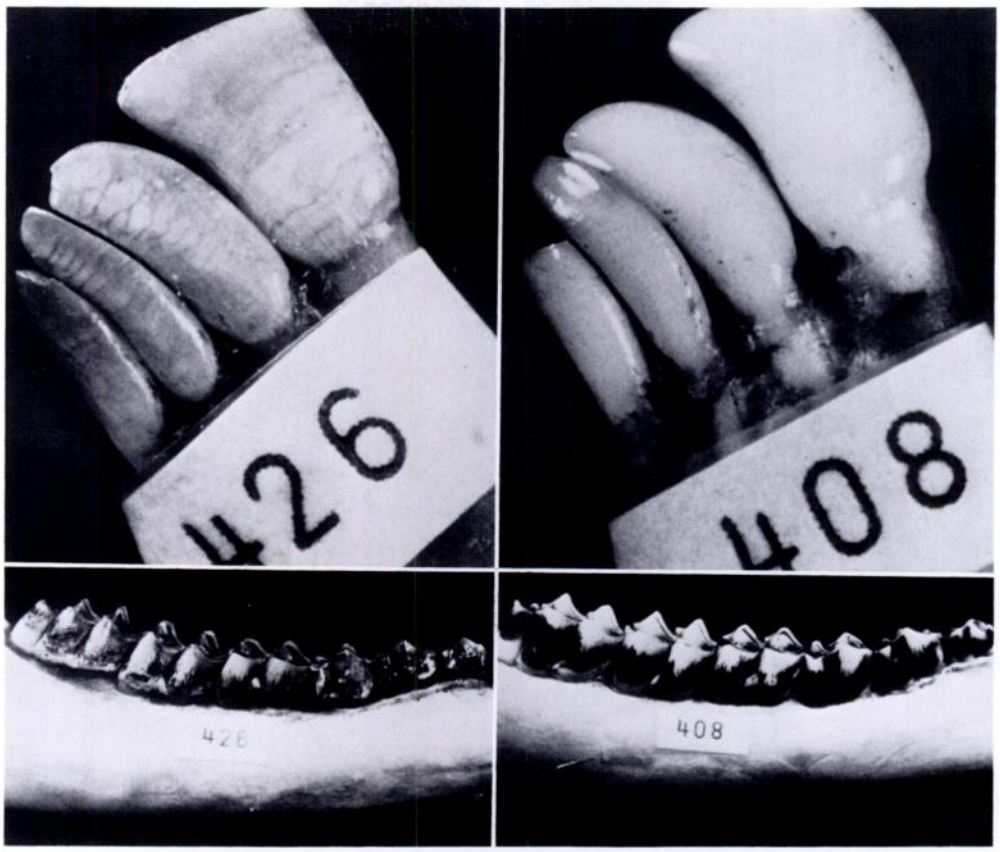

FIGURE 2. Dentition of two white-tailed deer obtained on two plantations near the Alumax, South Carolina aluminum smelter. Upper Left: Incisor teeth of a 3½-yr-old deer (no. 426) showing extensive generalized mottling obtained at Mount Holly Plantation in 1983. The 2nd incisor enamel F content was 1,538 ppm F which was the highest value seen in any animal, and the degree of dental fluorosis scored as no. 3 (moderate effect) and was the most severe observed. Lower Left: Mandibular premolars and molars from the same animal. Upper Right: Incisor teeth of a 2½-yr-old deer (no. 408) obtained at the Medway Plantation in 1983. The 2nd incisor enamel F content was 126 ppm F, and there was no evidence of dental fluorosis. Lower Right: Mandibular molars from the same animal.

of the more common species of deer browse, and data on some of the more extensively sampled species of vegetation are shown in Table 4. Plant operations began in 1980 and, although the concentration of fluoride in vegetation sampled at the Mount Holly Plantation was higher than at Medway Plantation, concentrations of fluoride in the vegetation at both locations were low.

Most deer utilize a relatively small range (Severinghaus and Cheatum, 1956), and Sweeney (1970) has estimated a home range of 170 ha for white-tailed deer in South Carolina. Fluoride concentrations in vegetation at Mount Holly are known to decrease rapidly as the distance from the smelter increases (Dickie et al., 1984). Thus, fluoride exposure of individual deer was undoubtedly extremely variable over the 2,530 ha plantation. A large fraction of the total number of deer collected in the study was, however, obtained in two general areas: one located about 1 km to the north, and the other about 1 km to the southwest of the plant site. The data in

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TABLE 3. Concentration of fluoride in incisor enamel of white-tailed deer at three locations in South Carolina.

Location	Smelter start-up	No. of animals examined	2nd incisor F (ppm F, dry wt)
Waterhorn	Before	13	117 ± 65[a]
	After	9	173 ± 117
Medway	Before	29	110 ± 55
	After	24	178 ± 56[b]
Mount Holly	Before	33	149 ± 73
	After	28	457 ± 333[b]

[a] Values are means ± SD for the right 2nd incisor of deer of various ages in which the 2nd incisor was formed before or after smelter operations were started. The Waterhorn Hunt Unit is about 130 km north of the smelter.
[b] "After" concentration differs from "before"; $P < 0.001$.

Figure 4 indicate that skeletal fluoride content of the deer obtained was very much dependent on the area in which they were harvested. The predominant wind direction is from the southwest, and the deer collected northeast of the smelter had about twice the skeletal fluoride content of those obtained in the other area.

Antlers have been shown (Suttie et al., 1985) to accumulate fluoride very rapidly over the short time period that this tissue is vascularized. Antlers were obtained from a number of deer collected at Mount Holly Plantation in 1982 and 1983, and

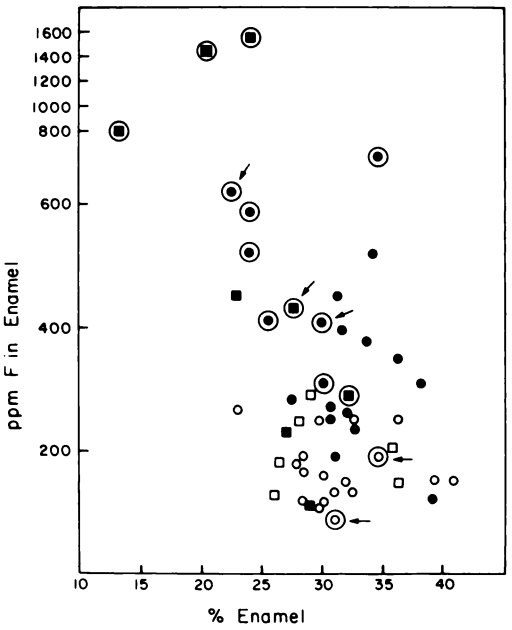

FIGURE 3. Enamel fluoride and percent enamel in crown of white-tailed deer obtained on two plantations near the Alumax, South Carolina aluminum smelter. The values plotted are the concentration of fluoride and percent of incisor crown which was enamel (see Materials and Methods) of all 2nd incisor teeth formed after the start-up of the smelter in 1980. ● = Mount Holly Plantation, 1½-yr-old deer; ■ = Mount Holly Plantation, ≥2½-yr-old deer; ○ = Medway Plantation, 1½-yr-old deer; □ = Medway Plantation, ≥2½-yr-old deer. The symbols which have been circled represent those teeth which were judged to have dental fluorosis by visual inspection. The small arrow indicates the teeth that were thought to have a non-fluoride-induced mottling (score no. 1) by visual inspection.

TABLE 4. Fluoride content of vegetation sampled[a] at Mount Holly and Medway Plantations, South Carolina.

	Fluoride content (ppm F, dry wt)									
	Vaccinium myrsinites (blueberry)		Mixed grasses		*Quercus virginiana* (live oak)		*Quercus nigra* (water oak)		*Acer rubrum* (red maple)	
Year	MH[b]	M[c]	MH	M	MH	M	MH	M	MH	M
1979	1.4	3.9	3.3	4.0	N.S.[d]	1.0	2.6	0.7	2.9	N.S.
1980	9.3	2.2	3.9	2.9	0.8	1.2	4.3	1.1	29.8	N.S.
1981	4.0	4.2	7.4	2.7	9.6	0.5	2.8	0.4	32.7	4.2
1982	2.9	1.4	5.7	1.6	3.1	1.8	2.7	0.1	27.0	1.2
1983	N.S.	N.S.	7.4	0.9	9.6	0.5	4.1	N.S.	4.3	N.S.

[a] The number of samples of each species obtained at each location varied from year to year but averaged four at Medway and nine at Mount Holly.
[b] MH = Mount Holly Plantation.
[c] M = Medway Plantation.
[d] N.S. = Not sampled.

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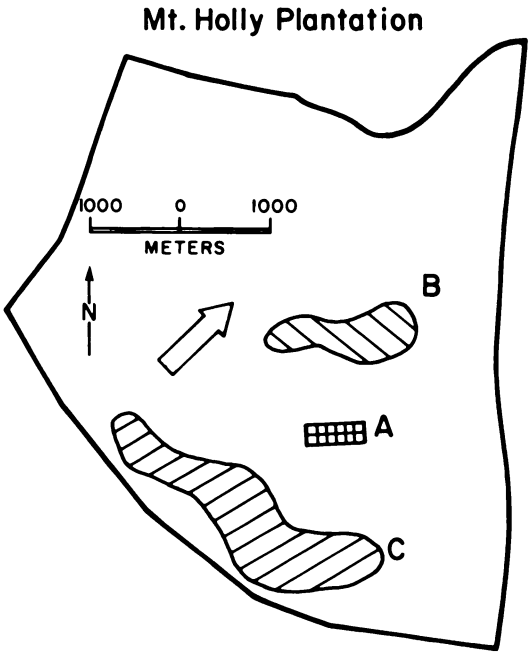

FIGURE 4. Location of the Alumax smelter site within Mount Holly Plantation, South Carolina. Some white-tailed deer were obtained from almost all areas of the plantation, but a majority was obtained from either site B to the north or site C to the southwest of the smelter site A. The large arrow indicates the predominant wind direction. The mandibular fluoride content (ppm, ash wt) of all deer ≥1½ yr old obtained in area B in 1981–1983 was 1,252 ± 716 (SD, n = 22) and in area C was 698 ± 483 (SD, n = 14). Fluoride concentrations of the mandibles in area B were greater ($P < 0.025$) than in area A.

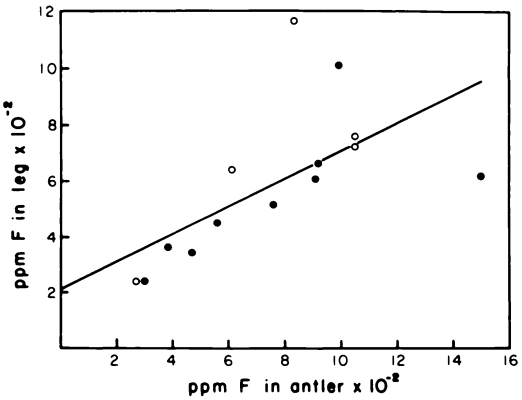

FIGURE 5. Relationship between concentration of fluoride in the ash of the metacarpal bone or antlers of white-tailed deer obtained at Mount Holly Plantation, South Carolina. Samples of antler were analyzed from deer obtained at Mount Holly in 1982 (○) and 1983 (●) and compared to the fluoride content of the metacarpal bone. The line represents metacarpal F = 0.49 × antler F + 223, $r^2 = 0.40$.

the data shown in Figure 5 illustrate that concentrations of fluoride in antlers are reasonably well correlated with concentrations of fluoride in skeletal tissue. Analyses of antlers would, therefore, be a convenient and simple way to monitor an increase in concentration of fluoride in the vegetation within an area.

This study has demonstrated that the impact of fluoride emissions from a modern aluminum smelter on the resident wild deer population of an area was minimal. None of the examples of severe dental fluorosis of incisors or irregular and abnormal wear of the molars which have been documented (Robinette et al., 1957; Karstad, 1967; Shupe et al., 1984) in wild ungulates from various locations was seen in deer inhabiting the area adjacent to this modern 182,000 metric tons/yr smelter. Fluoride emissions from this facility were in the range of 200–250 kg F/day. The observations made in this study should aid in estimates of potential impact from other fluoride emitting industries.

This study also points out the extreme sensitivity and usefulness of wild herbivores as indicators of increased concentrations of fluoride in the environment. Although no adverse impact on the deer population was evident, up to five-fold increases in fluoride concentrations of bones of various aged deer were seen at Mount Holly Plantation following smelter start-up. Detectable increases in skeletal fluoride were seen also in deer at Medway Plantation even though demonstrable increases in fluoride content of vegetation were not observed. Slight but discernible fluoride-induced mottling also was seen at Mount Holly but not at Medway. The increases in skeletal fluoride seen in this study can be compared to values reported for skeletal fluoride concentrations of deer

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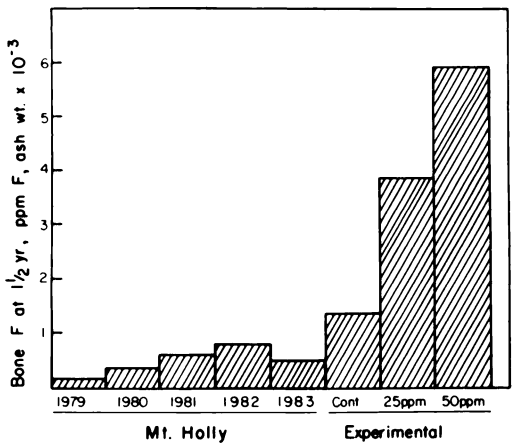

FIGURE 6. Comparison of skeletal fluoride in white-tailed deer obtained at Mount Holly Plantation, South Carolina and deer experimentally fed known amounts of fluoride. Data plotted are the mean metacarpal ash fluoride content from 1½-yr-old deer obtained at Mount Holly Plantation and similar analyses of metacarpal ash of white-tailed deer fed known amounts of F for 2 yr (Suttie et al., 1985).

collected in the vicinity of some other industries. Mandibular fluoride concentrations of the adult deer during the last 3 yr of this study were about 1,000 ppm F at Mount Holly Plantation, 300 ppm F at Medway Plantation, and 250 ppm F at the Waterhorn Hunt Unit control area. Kay et al. (1975) have reported mandibular fluoride values of 160–190 ppm F in deer obtained from nonindustrial areas of Montana, and an average of 4,300 ppm F for 18 deer obtained near a primary aluminum smelter utilizing older and less efficient pollution control technology. Karstad (1967) found concentrations of mandibular fluoride averaging 3,300 ppm F in 14 deer drinking fluoride-contaminated water from an unspecified industrial source, and 360 ppm F in six presumably normal deer. The fluoride values in these two studies were expressed on a dry fat-free basis and are only about 70% of what they would have been on an ash weight basis. The degree of fluoride exposure of the resident population of deer near the Mount Holly smelter was, therefore, much less than has been reported from older, less well controlled facilities.

The extent and significance of the increased fluoride concentration in the environment surrounding the Alumax smelter can be assessed by a comparison to a recent controlled feeding study (Suttie et al., 1985). In the controlled study, penned white-tailed deer were fed 25 or 50 ppm F as NaF added to a commercial horse ration (Omolene 100, Ralston Purina). The data in Figure 6 indicate that the concentration of fluoride in the metacarpal ash of deer obtained from the Mount Holly Plantation contained less fluoride than deer maintained on a commonly fed commercial horse ration. The intake of fluoride by deer in this area was therefore lower than that of many domestic animals. These data again point out that although a detectable effect was seen, the Alumax Mount Holly aluminum smelter caused a level of fluoride related effects which was well within acceptable limits.

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