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FLUORIDE EXPOSURE IN CERVIDS INHABITING AREAS ADJACENT TO ALUMINUM SMELTERS IN NORWAY. I. RESIDUE LEVELS

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ABSTRACT: Mandibular fluorine concentrations were determined in 1,425 red deer (*Cervus elaphus*), 240 moose (*Alces alces*), and 424 roe deer (*Capreolus capreolus*) collected in Norway from 1990 to 1993 in seven municipalities in which aluminum smelters are located, in eight neighboring municipalities, and in eight reference areas representing background levels. Background fluorine concentration was significantly correlated with age in all three species. Roe deer had the highest mean background fluorine level in each age group, followed by red deer. Due to differences in fluoride exposure, large variations in bone fluorine residues were evident between locations. In Årdal, the district most severely exposed to fluoride contamination, nine of ten cervids had fluorine concentrations exceeding background levels. The proportions of red deer with fluorine residues exceeding background levels also were high in neighboring municipalities to Årdal. We propose that roe deer are a better biomonitor of local fluoride exposure than red deer and moose, due to their more sedentary behavior.

Key words: Fluoride emission, bone fluorine, cervids, *Cervus elaphus*, *Alces alces*, *Capreolus capreolus*.

INTRODUCTION

Fluorine-containing compounds are used in the aluminum producing process. The resulting emissions from electrolysis cells include both gaseous and particulate fluoride. In Norway, the first primary aluminum reduction plants began operating in 1908. Prior to 1960, aluminum smelters were located on fjords to enable transport of raw materials and processed aluminum by sea, and in valley bottoms to use hydroelectric power. The topographical conditions hindered rapid dispersal of airborne emissions from the smelters. Four of these smelters are still operating: in Høyanger, Årdal, Sunndal, and Vefsn. Fluorosis in farm animals in Norway due to fluoride emissions first was reported in the early 1930's by Slagsvold (1934). Experimental studies in which sheep were fed grass grown in the vicinity of an aluminum smelter enabled Slagsvold (1934) to document the development of fluorotic lesions.

The Norwegian aluminum industry expanded rapidly during the 1950's. This resulted in industrial fluorosis becoming a severe problem in livestock locally in emission-exposed areas. In Norway, the extent of adverse effects on animals and vegetation is influenced not only by levels and composition of the emissions, but also very largely by local topographical and meteorological conditions (Flatla and Ender, 1967). Reduction plants established in Norway after 1960, in Kvinnherad, Karmøy, and Farsund, were located more favorably with respect to rapid dispersal of fumes.

The Norwegian Smoke Control Council initiated a program, running from 1967 to 1992, to summarize data concerning emissions from all Norwegian aluminum plants, and assess detrimental effects on vegetation and livestock (Hansen, 1994). Wild animals were not included in this monitoring program. However, sporadic cases of fluorosis in cervids have been recorded, and red deer (*Cervus elaphus*) collected in Årdal, Norway, in 1970, had considerable bone fluorine loads (Holt, 1978). On the initiative of the Norwegian aluminum manufacturers, a comprehensive study for monitoring the effects of fluoride emissions on the local environment was started in 1990. As part of this study, a systematic

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survey of the effects of fluoride exposure on red deer, roe deer (*Capreolus capreolus*), and moose (*Alces alces*) was carried out. Our objective was to evaluate the extent of exposure on local cervid populations; we used mandibular fluorine concentrations as an index of fluoride exposure. Fluorotic lesions associated with fluoride exposure are described separately (Vikøren and Stuve, 1996).

MATERIALS AND METHODS

Between 1990 and 1993, mandibles from a total of 1425 red deer, 240 moose, and 424 roe deer were sampled in Norway. The collecting sites included seven municipalities of variable area in which primary aluminum smelters are located, eight neighboring municipalities to these, and eight reference areas (Fig. 1).

Municipalities with aluminum smelters and their respective sample numbers were distributed as follows: Farsund (58°05′N, 06°50′E), 165 roe deer; Karmøy (59°18′N, 05°19′E), 23 red deer; Kvinnherad (59°53′N, 05°45′E), 259 red deer; Årdal (61°19′N, 07°49′E), 95 red deer and 30 moose; Høyanger (61°13′N, 06°08′E), 130 red deer; Sunndal (62°41′N, 08°33′E), 313 red deer and 162 roe deer; and Vefsn (65°51′N, 13°11′E), 117 moose. Samples from the eight neighboring municipalities included: Bokn (59°13′N, 05°25′E), 25 red deer; Lærdal (61°06′N, 07°30′E), 62 red deer; Sogndal (61°14′N, 07°05′E), 138 red deer; Leikanger (61°11′N, 06°53′E), 34 red deer; Solund (61°04′N, 04°50′E), 18 red deer; Hyllestad (61°10′N, 05°17′E), 47 red deer; Balestrand (61°12′N, 06°32′E), 44 red deer; and Luster (61°25′N, 07°17′E), 36 red deer. The reference material included 201 red deer mandibles from Masfjorden (60°48′N, 05°17′E), Surnadal (62°58′N, 08°43′E), Meldal (63°04′N, 09°40′E), and Hemne (63°19′N, 09°06′E), 93 moose mandibles from Åsnes (60°37′N, 12°02′E) and Verdal (63°48′N, 11°30′E), and 97 roe deer mandibles from Østfold (59°00′ to 59°40′N, 10°45′ to 11°30′E) and Fosen (63°30′ to 63°50′N, 09°50′ to 10°50′E). Additionally, 25 sheep mandibles from Sogndal were collected at slaughter in 1992.

Hunters collected mandibles from cervids shot during regular hunting seasons, from the end of September until the middle of November for red deer and moose, and from the middle of August until the middle of December for roe deer. The samples were stored at −20 C. Data recorded including location, date, estimated age, sex, general condition, and carcass weight, were submitted to the Central Veterinary Laboratory in Oslo at the end of each hunting season.

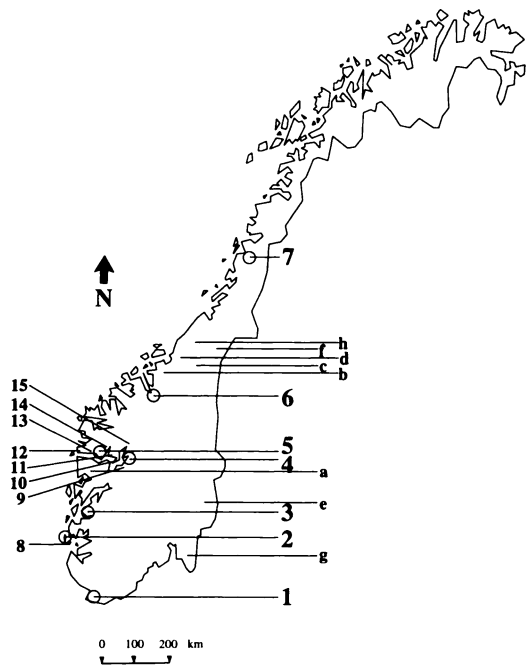

FIGURE 1. Map of Norway with locations where mandibles of cervids were collected from 1990 to 1993. Sites 1 to 7 are municipalities of variable area in which aluminum smelters are located; (1) Farsund, 268 km^2, (2) Karmøy, 228 km^2, (3) Kvinnherad, 1136 km^2, (4) Årdal, 982 km^2, (5) Høyanger, 906 km^2, (6) Sunndal, 1712 km^2, and (7) Vefsn, 1700 km^2. Sites 8 to 15 are neighboring municipalities to some of these; (8) Bokn, (9) Lærdal, (10) Sogndal, (11) Leikanger, (12) Solund, (13) Hyllestad, (14) Balestrand, and (15) Luster. Reference materials were collected in (a) Masfjorden, (b) Surnadal, (c) Meldal, and (d) Hemne for red deer, in (e) Åsnes and (f) Verdal for moose, and in the districts (g) Østfold and (h) Fosen for roe deer.

Bone samples for fluorine analyses from moose, adult red deer, and sheep were obtained 15 mm upwards from the *margo ventralis* of the right *corpus mandibulae,* between the fourth premolar and the second molar. In roe deer and fawns of red deer, the ventral half of the *pars molaris* on the right *corpus mandibulae* between the first premolar and the last molar was sampled, including the ventral part of the *angulus mandibulae* in roe deer fawns. The bone specimens were dipped in boiling water to facilitate removal of soft tissues.

The fluorine analyses were performed at the laboratory of Hydro Aluminium in Årdal, Norway. Bone samples were converted to ash at 700 C for a minimum of 8 hr. The fluorine (F)

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concentration in bone ash was determined using a solid-state fluoride-selective electrode (Sintalyzer-system) as described by Nagy and Keul (1978). Two parallel ash samples from each specimen were analyzed, and the mean recorded as the fluorine concentration. Results were expressed as ppm F (mg F/kg) in bone ash. For quality control, fluorine analysis of 15 random samples each of bone and ash specimens was performed at the Foundation for Scientific and Industrial Research (SINTEF) in Trondheim, Norway, using the same method as at the laboratory in Årdal. No discrepancy was noted in results of the two laboratories.

Ages of sheep were given by the owners. Ages of red deer (≤2.5 yr), and moose and roe deer (≤1.5 yr), were determined by tooth replacement (Reimers, 1981). Older animals were aged by examination of annuli in incisor tooth cementum after decalcification and staining of tooth sections (Reimers and Nordby, 1968). Younger cervids were grouped as 0.5, 1.5, and 2.5 yr, whereas older animals were grouped as 3.5 to 5.5 yr and >5.5 yr, to get sufficiently large groups for statistical analyses.

The Student procedure was used for calculating confidence intervals for the means (Altman, 1991). Species differences in background fluorine levels were examined for each age group using the Kruskall Wallis test and the Wilcoxon two-sample test (Altman, 1991). The latter also was used to test differences in fluorine levels between males and females. Correlation analysis was carried out using the Pearson model (Altman, 1991). Statistical significance in this study refers to $P < 0.05$.

In order to distinguish between cervids exposed to fluoride emissions and animals that were not, we estimated a limit called the upper normal limit (UNL). This limit was estimated for each species and age group using the sample 95th percentiles of mandibular fluorine concentrations in cervids collected at reference sites. Also we wanted to graduate the fluorine-load and chose the upper normal limit × three (UNL × 3) to represent the threshold for which the mandibular fluorine concentration had to exceed for cervids to be termed definitely fluorine-loaded. Thus, the extent of fluoride exposure in each municipality was described by the percentage of cervids having mandibular fluorine concentrations exceeding the UNL and UNL × 3.

RESULTS

In cervids from the reference sites, large individual variations in mandibular fluorine level occurred within age groups (Fig. 2). Fluorine concentration was significantly correlated with age ($r = 0.74$, $r = 0.67$, and $r = 0.54$, in moose, red deer, and roe deer, respectively). No significant difference by sex was observed in fluorine level.

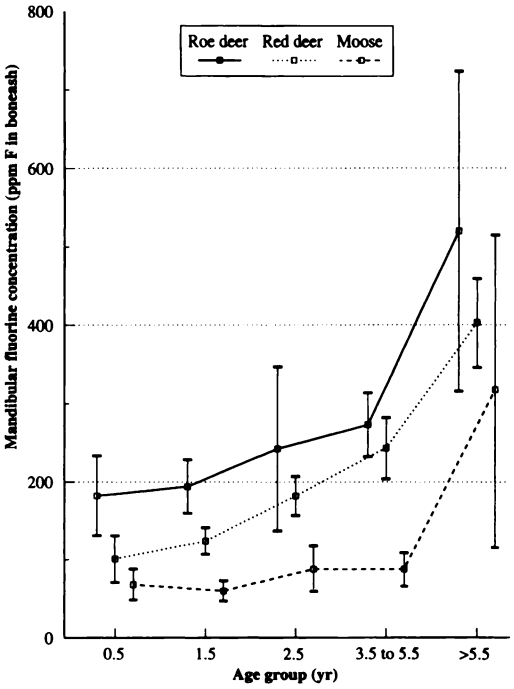

FIGURE 2. Mandibular fluorine (F) concentrations (ppm F in bone ash) in red deer, moose, and roe deer from reference sites not exposed to industrial fluoride emissions, given as means with confidence intervals for each age group.

When comparing species, significant ($P < 0.01$) differences in background bone fluorine content were found in all age groups, except >5.5 yr. When testing species two by two, significant differences were evident, except between fawns of red deer and moose, and between red deer and roe deer yearlings (1.5 yr) and 2.5-yr-olds. Generally, roe deer had the highest mean background fluorine concentrations, followed by red deer.

The UNL for mandibular fluorine concentrations in unexposed cervids had interspecific variation within age groups (Table 1). Roe deer had the highest UNL in all age groups. In all three species, the UNL was higher in the 0.5-yr-olds than in the 1.5-yr-olds. Between 1.5-yr-old and

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TABLE 1. Upper normal limit (UNL) for the mandibular fluorine concentration (ppm F in bone ash) in various age groups of roe deer, red deer and moose, based on the 95th percentiles of mandibular fluorine levels in cervids collected from reference sites, 1990 to 1993.

Age group (yr)	Roe deer		Red deer		Moose	
	Number sampled	UNL[a] (ppm F)	Number sampled	UNL[a] (ppm F)	Number sampled	UNL[a] (ppm F)
0.5	27	530	31	310	22	150
1.5	17	310	66	270	22	110
2.5	8	490	52	350	20	230
3.5 to 5.5	36	530	24	350	22	160
>5.5	9	880	28	610	7	750

[a] The figures have been rounded off to the nearest 10.

>5.5-yr-old, the UNL increased with age, except for moose between the two age groups of 2.5-yr-old and 3.5–5.5-yr-old.

The highest mean mandibular fluorine level in all age groups of red deer was found in Årdal, followed by Sunndal (Table 2). Moreover, the mean fluorine residues in red deer from Sogndal and Bokn, neighboring municipalities to Årdal and Karmøy, respectively, exceeded the corresponding levels in animals from Høyanger, Karmøy, and in part Kvinnherad, all with smelters within the municipality (Table 2).

Moose from Årdal had considerably elevated fluorine concentrations compared with the reference group, whereas the population in Vefsn was moderately fluorine-loaded (Table 3). The mean fluorine levels in roe deer from the fluoride emission-exposed municipalities of Sunndal and Farsund clearly exceeded those of the reference group (Table 4).

In sheep sampled from Sogndal, the mean mandibular fluorine level was 1006 ppm F and the average age 6.3 yr.

Årdal had the highest proportion of red deer with bone fluorine levels above UNL and UNL × 3 (Table 5). The percentage of red deer exceeding UNL was in the same order of magnitude in Sogndal and Sunndal. However, regarding UNL × 3, a three-fold higher percentage was revealed in Sunndal (Table 5). Of red deer sampled in Lærdal and Luster, municipalities located southwest and northwest of Årdal, 68% and 36% had fluorine levels above UNL, respectively, whereas less than 8% exceeded UNL × 3. Bone fluorine concentrations in red deer collected in other municipalities located on the Sognefjord, namely Leikanger, Solund, Hyllestad, and Balestrand, did not exceed UNL.

On the basis of local geography and topography, the red deer material from the municipality of Sunndal was split into three groups; Sunndalen (n = 107), Ålvundeid (n = 121), and Øksendalen (n = 79). The proportions of red deer with fluorine levels exceeding UNL in these three location groups, were 79%, 60%, and 34%, respectively, whereas 39%, 12%, and 1%, respectively, exceeded UNL × 3.

When regarding the proportions of moose and roe deer exceeding UNL and UNL × 3, distinct differences between the two fluoride-exposed moose populations were evident, whereas in roe deer only small differences existed between Sunndal and Farsund (Table 5).

DISCUSSION

Accumulation of fluoride with age was a prominent feature in the present study. In roe deer, the correlation coefficient of the relation between background fluorine level and age, corresponded well with that reported by Kierdorf (1988). However, better correlations were documented in moose and red deer. This may reflect the age determination method used, which is more precise in these two species than in roe deer. Kierdorf et al. (1995) found a

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TABLE 2. Parts per million (ppm) of mandibular fluorine (F) concentrations in bone ash in different age groups of red deer collected from five fluoride-exposed sites: Årdal, Sunndal, Kvinnherad, Høyanger, and Karmøy; from Sogndal and Bokn, neighboring municipalities to Årdal and Karmøy, respectively; and from reference sites not exposed to industrial fluoride emissions, 1990 to 1993.

Age group (yr)		Reference sites	Fluoride exposed sites					Neighboring sites	
			Årdal	Sunndal	Kvinnherad	Høyanger	Karmøy	Sogndal	Bokn
0.5	Number sampled	31	13	57	32	10	0	14	6
	Mean	101	657	317	161	104		194	171
	SD	83	1,096	291	224	77		133	75
	95% Confidence interval	71–131	0–1,319	240–394	81–242	49–159		117–270	91–251
	Range	18–310	76–4,165	41–1,479	31–767	47–302		76–525	77–237
1.5	Number sampled	66	34	87	54	37	2	43	7
	Mean	124	1,283	446	262	194	199	351	556
	SD	69	1,185	391	253	147	—[a]	163	132
	95% Confidence interval	107–141	870–1,697	362–529	193–331	145–243	—	301–401	434–678
	Range	44-358	252–5,436	80–1,755	51–1,158	43–721	124–223	132–845	391–730
2.5	Number sampled	52	25	66	38	43	8	23	4
	Mean	182	1,374	637	400	252	315	387	587
	SD	90	569	527	359	177	68	142	222
	95% Confidence interval	157–207	1,139–1,608	508–767	282–518	197–306	258–372	325–448	234–940
	Range	77-546	524–2,859	113–2,528	119–1,762	66–916	249–440	164–742	350–885
3.5 to 5.5	Number sampled	24	13	74	79	22	10	33	3
	Mean	243	2,512	995	716	398	348	554	727
	SD	92	2,163	876	1,380	217	72	299	371
	95% Confidence interval	204–282	1,205–3,819	793–1,198	407–1,025	301–494	297–400	448–660	~0–1,649
	Range	29–471	522–6,820	125–4,035	154–8,433	130–863	258–445	252–1,787	451–1,148
>5.5	Number sampled	28	10	29	56	18	3	25	5
	Mean	403	4,641	1,690	907	949	484	1,216	1,355
	SD	146	4,013	1,053	897	608	199	607	609
	95% Confidence interval	346–459	1,770–7,512	1,290–2,091	667–1,147	646–1,252	~0–978	965–1,466	599–2,111
	Range	198–844	914–12,707	529–5,457	138–5,761	285–2,187	285–683	396–2,431	685–2,044

[a] —, not calculated.

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TABLE 3. Mandibular fluorine (F) concentrations (ppm F in bone ash) in different age groups of moose collected from two fluoride-exposed sites (Årdal and Vefsn) and from two reference sites not exposed to industrial fluoride emissions, 1990 to 1993.

Age group (yr)		Reference sites	Vefsn	Årdal
0.5	Number sampled	22	20	2
	Mean	68	141	178
	SD	45	260	—[a]
	95% Confidence interval	49–88	19–263	—
	Range	28–176	26–1,219	152–203
1.5	Number sampled	22	42	6
	Mean	60	159	487
	SD	28	163	186
	95% Confidence interval	48–73	108–210	291–682
	Range	22–107	36–890	206–713
2.5	Number sampled	20	19	9
	Mean	88	194	531
	SD	64	230	790
	95% Confidence interval	58–118	83–305	~0–1,138
	Range	22–273	35–775	141–2,591
3.5 to 5.5	Number sampled	22	30	7
	Mean	88	171	1,302
	SD	47	125	1,233
	95% Confidence interval	67–109	124–218	162–2,442
	Range	40–166	50–514	175–3,407
>5.5	Number sampled	7	6	6
	Mean	318	432	3,042
	SD	213	319	1,493
	95% Confidence interval	121–515	97–767	1,476–4,609
	Range	123–748	172–1,043	1,805–5,406

[a] —, not calculated.

higher correlation coefficient ($r = 0.81$) in red deer than we did, probably due mainly to the fact that their animals were marked (ear tagged) as postnatal fawns, thus the ages at death were exactly known. Suttie et al. (1987) and Kierdorf et al. (1989) found no difference in fluoride accumulation between males and females, which agrees well with our findings.

In general, background fluorine levels reported in the literature cannot be directly compared due to methodological differences, especially those regarding bone sampling and age determination. Fluorine concentrations vary both between bones and between different parts of a bone (Kay, 1975). Nevertheless, the background fluorine levels found in Norwegian roe deer in the present study seem to be among the lowest reported for this species. Kierdorf (1988) reported a mean mandibular fluorine level of 470 ppm (dry, fat-free weight) in 83 roe deer of an average age of 4.7 yr, a figure comparable to 670 ppm F on an ashweight basis. The corresponding figure in our roe deer material was 254 ppm F at a mean age of 2.9 yr. Taking the lower average age into account, the background fluorine level observed in Norway seems somewhat lower than that reported from Germany by Kierdorf (1988). The mean fluorine levels demonstrated by Walton and Ackroyd (1988) in roe deer from different localities in England and Scotland were similar to or higher than that found in our study.

Only a few data exist on background fluorine concentrations in red deer (Machoy et al., 1991; Kierdorf et al., 1995) and in moose (Kay et al., 1975a). In summary, it

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TABLE 4. Mandibular fluorine (F) concentrations (ppm F in bone ash) in different age groups of roe deer collected from two fluoride-exposed sites: Farsund and Sunndal, and from various reference sites not exposed to industrial fluoride emissions, 1990 to 1993.

Age group (yr)		Reference sites	Farsund	Sunndal
0.5	Number sampled	27	51	58
	Mean	182	304	396
	SD	129	448	436
	95% Confidence interval	131–233	178–430	281–511
	Range	73–594	38–2,759	66–2,759
1.5	Number sampled	17	24	12
	Mean	194	563	618
	SD	66	466	568
	95% Confidence interval	160–228	366–760	257–979
	Range	90–307	125–2,034	170–2,172
2.5	Number sampled	8	20	23
	Mean	242	935	807
	SD	126	1,611	1,145
	95% Confidence interval	137–347	181–1,689	312–1,302
	Range	129–487	141–7,430	168–4,861
3.5 to 5.5	Number sampled	36	62	56
	Mean	273	833	1,192
	SD	121	1,176	1,599
	95% Confidence interval	232–314	534–1,132	764–1,620
	Range	114–568	135–6,830	81–8,696
>5.5	Number sampled	9	8	13
	Mean	520	1,004	1,855
	SD	265	1,478	2,304
	95% Confidence interval	316–724	~0–2,240	463–3,247
	Range	131–883	251–4,644	164–8,042

TABLE 5. Proportion (%) of red deer, moose, and roe deer with mandibular fluorine level exceeding the upper normal limit (> UNL) and upper normal limit × 3 (> UNL × 3), in each of the fluoride-exposed sites; Årdal, Sunndal, Karmøy, Kvinnherad, Høyanger, Vefsn, and Farsund, and in Sogndal and Bokn, neighboring municipalities to Årdal and to Karmøy, respectively, 1990 to 1993.

Species	Site	> UNL (%)	> UNL × 3 (%)
Red deer	Årdal	89	53
	Sunndal	60	19
	Karmøy	39	0
	Kvinnherad	38	7
	Høyanger	23	2
	Sogndal	58	7
	Bokn	72	12
Moose	Årdal	87	47
	Vefsn	35	5
Roe deer	Sunndal	38	14
	Farsund	31	11

seems that minor geographical differences in background fluorine levels exist within species, as demonstrated by Kay et al. (1976), and Walton and Ackroyd (1988). This partly may be due to geological differences and varying degrees of anthropogenic addition of fluoride. Norwegian lakes had significant differences in median fluorine concentrations in water between different geological provinces, ranging from 13 to 80 mg F/l (Skjelkvåle, 1993). Comparisons between the various reference sites was not carried out in our study due to small sample size and heterogeneous age composition.

No interspecies difference in background fluorine level was found in the age group >5.5 yr, most probably due to the heterogeneous age composition resulting from disparity in life expectancy of the species, and the small sample size. In all

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the other age groups, roe deer had significantly higher background fluorine levels than moose. As discussed by Kay et al. (1976), specific differences in fluorine level are difficult to explain, especially in closely related species with similar dietary preferences.

The UNL was higher in the 0.5-yr-olds than in the 1.5-yr-olds, due to a few high individual fluorine levels among the calves, most prominent in roe deer. This was probably a result of variable individual biological response to fluoride.

In Norway, the aluminum producing process is the only fluoride-emitting industry of concern (Flatla and Ender, 1967). From 1990 to 1993, the average total fluoride emissions (includes both gaseous and particulate fluoride) from the primary aluminum smelters in Årdal, Sunndal, Karmøy, Vefsn, Kvinnherad, Høyanger, and Farsund, were 18, 14, 11, 8, 7, 6, and 6 kg F/hr, respectively (Anonymous, 1994). Animals may be exposed to fluoride by consumption of polluted feed, intake of polluted drinking water, or through inhalation of airborne fluoride. Inhalation of fluoride may be regarded as negligible in cervids (National Research Council, 1971). In Årdal and Sunndal, water from streams taken within 5 km of the smelters contained only slightly raised fluorine levels (Skjelkvåle, 1993). Thus, we believe that contaminated vegetation was the main fluoride source for cervids inhabiting areas adjacent to Norwegian aluminum smelters. This assumption was supported by studies of fluoride contamination of vegetation in these areas (Horntvedt and Øyen, 1994).

We observed varying degree of elevated mandibular fluorine levels in cervids living in the vicinity of different aluminum smelters. This was in agreement with the results of previous studies (Kay et al., 1975b; Newman and Yu, 1976; Holt, 1978; Newman and Murphy, 1979; Suttie et al., 1987). Moreover, cervids from some neighboring municipalities also had skeletal fluorine residues exceeding background levels. Mandibular fluorine concentration varied considerably between individuals of the same age and population. This may reflect variable individual biological response, variations in home range and browsing preferences, and different extent and pattern of migration. Individuals with sedentary home range in highly fluoride contaminated areas are exposed to higher fluoride doses than less sedentary animals migrating between areas with variable fluoride contamination, as well as sedentary animals living in areas with low fluoride contamination. Roe deer are generally more sedentary and utilize smaller home ranges than red deer and moose (Semb-Johansson, 1990). In Farsund and Sunndal, the highest bone fluorine concentrations were found in roe deer shot within 1 to 5 km from the smelters. It is likely that these animals had their home range in these areas, in which the vegetation was contaminated with high levels of fluoride (Horntvedt and Øyen, 1994). High bone fluorine levels in the less sedentary red deer and moose, may reflect long residence time in highly polluted areas.

Cervids from Årdal had the highest mandibular fluorine levels, and also the largest proportion fluorine-loaded cervids was found in this municipality. Cervids were collected as far as 23 km from the smelter. Animals with mandibular fluorine residues exceeding UNL were collected in all parts of the municipality. Both red deer and moose migrate locally, and individuals with large bone fluorine residues must have periodically grazed in heavily contaminated locations. The fluoride tolerance level given for dairy cattle, 30 mg F/kg dry matter in ration (Shupe and Olson, 1971; Hansen, 1994), was exceeded in vegetation collected up to 23 km from the smelter (Horntvedt and Øyen, 1994). Thus, vegetation in large parts of the municipality was fluoride contaminated, and fluorine levels up to 365 mg F/kg dry weight were found in rowan (*Sorbus aucuparia*) within 1 km from the smelter (Horntvedt and Øyen, 1994). The alumi-

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num smelter in Årdal is located in a narrow valley 10 km from the innermost end of the Sognefjord. The valley branches off in several directions at this site. Lateral diffusion of smelter emissions is restricted by high mountains exceeding 1,000 m above sea level, and formation of inversion layers prevents vertical dispersion of fumes (Thrane, 1988). The climate is dry. Taking the extent of the fluoride emission from the Årdal smelter into account, the local topography may have a contributory effect on the degree of environmental fluoride contamination.

The mean fluorine concentration in 1.5-yr-old red deer from Årdal (Table 2) was less than that found in white-tailed deer (*Odocoileus virginianus*) fed a diet containing 35 ppm of soluble fluoride for 2 yr (Suttie et al., 1985). However, the most severely affected red deer yearlings in Årdal had fluorine residues of the same order of magnitude as the white-tailed deer, thus these individuals must at least have been exposed to fluorine levels comparable to a daily intake of 35 ppm F in the ration.

In Sogndal, Luster, and Lærdal, all municipalities west of Årdal, the proportion of red deer with fluorine residues above UNL was surprisingly large. Fluorine concentrations in vegetation collected from eastern parts of Sogndal and Luster did not exceed background levels (Horntvedt and Øyen, 1994), nor did mandibular fluorine residues in sheep from Sogndal. Thus, these neighboring municipalities seemed to not be within the fallout area affected by fluoride emissions from the smelter in Årdal. Local hunters allege red deer having traditional migration routes between these areas. Considering all information available, we believe that elevated bone fluorine levels in red deer from the municipalities neighboring Årdal, were attributable to local and regional migration of animals leading to variable residence time in contaminated areas in Årdal.

In Sunndal, areas affected by emissions were geographically limited, mainly due to the local topography. The Sunndal smelter is located in Sunndalsøra at the inner end of the Sunndalsfjord. A long, narrow valley, Sunndalen, extends about 35 km in south-easterly direction from Sunndalsøra. In Sunndalen, high mountains and formation of inversion layers prevent dispersion of fumes from the smelter (Thrane, 1988). The Ålvundeid area is topographically separated from Sunndalen to the north, as is Øksendalen in the southwesterly direction. The main fallout area for fluoride emissions, as indicated by residues in vegetation exceeding 30 mg F/kg dry matter, included Sunndalen up to 30 km from the smelter, and the innermost part of the Sunndalsfjord (Horntvedt and Øyen, 1994). This corresponds well with the findings in our study, where the proportion of red deer with bone fluorine levels exceeding UNL and UNL × 3 was higher for animals collected in Sunndalen than for those shot in the two other parts of the municipality. The higher fluorine residues in red deer from the Ålvundeid area than from Øksendalen most probably were due to the migration-pattern of the local red deer population. In roe deer, the geographical difference in fluorine-load between Sunndalen and other parts of the municipality was even more distinct (data not shown). We believe this was due to the more sedentary behavior of this species.

The topographical and meteorological conditions in Farsund, Karmøy, and Kvinnherad were more favorable as regards the rapid dilution of airborne fluoride pollution. In Farsund, roe deer were collected as far as 16 km from the aluminum smelter. Except for two animals, roe deer with fluorine levels exceeding the UNL × 3 were collected within 2 km of the smelter. Particulate fluoride constituted the main part of the emission from the Farsund smelter (Anonymous, 1994), thus limiting contamination to the immediate surrounding areas, as demonstrated in the vegetation study (Horntvedt and Øyen, 1994).

Mean skeletal fluorine concentrations in fawns and yearlings of white-tailed deer living in the vicinity of the Alumax alumi-

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num plant in South Carolina (USA) (Suttie et al., 1987), were slightly higher than the corresponding values in roe deer from Farsund. However, most of the white-tailed deer were obtained within 1 km from the Alumax plant, whereas only 18% of the roe deer in Farsund were shot within 2 km from the smelter.

The significance of meteorological conditions for fluoride exposure was exemplified in Karmøy, a windy, flat island with a wet climate. The aluminum smelter is located in the northeast of the island and had medium large emissions by Norwegian standards. Red deer from Vestre Bokn, an island 9 km southeast of the smelter, had higher mean bone fluorine levels and were to a larger extent fluorine-loaded than animals from Karmøy itself, which mainly live in the south part of the island. During the growing season, northwesterly winds predominated. Fluorine levels in plants exceeded 30 mg F/kg dry matter as far as 4 km southeast of the smelter (Horntvedt and Øyen, 1994). Vegetation studies were not carried out on Vestre Bokn but, based on our findings, we propose that this island was subject to fallout originating from fluoride emissions from the Karmøy smelter. On the other hand, local hunters allege that red deer migrate back and forth between the mainland and several islands, including Karmøy and Vestre Bokn. Periodic residence in fluoride-exposed areas closer to the smelter thus might have caused the moderate fluorine load demonstrated in red deer collected from Vestre Bokn.

Several studies have been performed using deer as indicator of industrially-emitted fluoride (Karstad, 1967; Kay et al., 1975b; Newman and Yu, 1976; Newman and Murphy, 1979; Suttie et al., 1987; Kierdorf, 1988; Machoy et al., 1991). Generally, data of these kind are influenced by numerous factors and are therefore less suited for direct comparison. These authors found that different species of deer were suitable bioindicators of fluoride exposure but, as stressed by Kay et al. (1975b), the use of deer as quantitative biomonitors is subject to inherent variability due to the deer's seasonal movements. Thus, sedentary home range behavior is an important criterion for the choice of appropriate species for monitoring local fluoride exposure. Roe deer are more sedentary than moose and red deer (Semb-Johansson, 1990), and seem therefore to be a better monitoring tool for local fluoride exposure, as indicated by our results from Farsund and Sunndal.

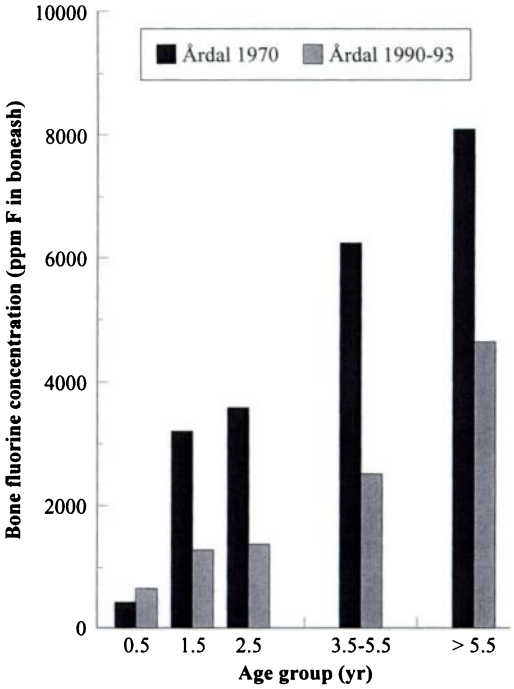

FIGURE 3. Mean mandibular fluorine (F) concentrations (ppm F in bone ash) in various age groups of red deer collected from Årdal in 1970 (Holt, 1978) and in 1990 to 1993. Number sampled in each age group in 1970 were: 0.5 yr, n = 3; 1.5 yr, n = 3; 2.5 yr, n = 4; 3.5–5.5 yr, n = 3; and >5.5 yr, n = 3. The numbers sampled in 1990 to 93 are given in Table 2.

Our results support our belief on the great importance of the topographical and meteorological conditions in relation to the local effect of fluoride emissions. Over the last 10 to 15 yr, emissions from Norwegian aluminum smelters have been considerably reduced (Anonymous, 1994). Nevertheless, we found that cervid popu-

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lations were still exposed to fluoride to varying degrees, particularly in Årdal and Sunndal, where topographical features are extremely unfavorable in relation to adequate dispersion of the emissions. However, the mean fluorine levels in various age groups of red deer from Årdal found in our study were lower than those found in 1970 by Holt (1978), except for fawns (Fig. 3). Although the study of Holt was based on few animals, comparison of the results from the two studies clearly demonstrates a considerable reduction in bone fluorine residues in red deer from Årdal during these 20 yr. This corresponds well with the reduction of total fluoride emission from 59 kg F/hr to 18 kg F/hr (Anonymous, 1994) during these 20 yr.

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