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Reduction of Fluoride Deposition in the Vicinity of a Brown Coal-Fired Power Plant as Indicated by Bone Fluoride Concentrations of Roe Deer *(Capreolus capreolus)*

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The skeletal fluoride content of wild deer is a cumulative measure of net fluoride uptake, integrating ambient exposure over time (Suttie et al. 1987; Kierdorf et al. 1989, 1995, 1996b; Machoy et al. 1995; Vikøren et al. 1996). Various authors analyzed bone fluoride concentrations in deer in order to assess the degree of fluoride pollution of the animals' habitats (Karstad 1967; Kay et al. 1975; Newman and Yu 1976; Newman and Murphy 1979; Suttie et al. 1987; Kierdorf 1988; Walton and Ackroyd 1988; Machoy et al. 1991; Samujlo et al. 1994; Hell et al. 1995; Kierdorf et al. 1996a,b, 1997; Vikøren et al. 1996). Variation in skeletal fluoride levels of deer taken in different years from a single population can also be used to record temporal variation of fluoride deposition into an area.

The present paper reports bone fluoride concentrations in roe deer collected over a 26 yr period from the same population in order to assess changes of fluoride deposition into a wildlife habitat adjacent to a huge brown coal-fired power plant.

MATERIALS AND METHODS

In total, thirty macerated skull caps of roe bucks were available, that originated from a forest area of roughly 5 km^2, located about 2 to 4 km SW of a huge brown coal-fired power plant in the opencast lignite mining area W of Cologne, federal state of North Rhine-Westphalia, Germany. The estimated ages (based on dental wear) of the individuals, that had been taken during normal hunting operations in the period 1973-1998, ranged between 1 and 7 yr.

Bone samples were obtained by drilling holes from the inner surface (facies interna) of the frontals into the pedicle bone, using a hand-held electric drill fitted with a tungsten-carbide cutter, and collecting the bone powder. Precisely weighed samples of the bone powder (8 to 12 mg) were dissolved (24 h, constant shaking) in 4 mL of 0.1 *M* perchloric acid, using sealed plastic tubes. Afterwards, the solution was buffered with 4 mL of a total ionic strength adjustment buffer (TISAB II, Orion Research, Beverly, MA, USA) and measured for fluoride using a ion-selective combination electrode (Orion Research model 96-09) connected to an Orion 920A-meter. Fluoride concentrations in the bone sample solutions were

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within the range of linear relationship between log fluoride content and electrode potential. Results are expressed as ppm F^- (= mg F^-/kg) on a dry weight basis. Mean (± SEM) coefficient of variation of multiple determinations was 5.1 (± 1.0) %.

Based on the year of death, the specimens were assigned to one of two time periods (1973-1986 and 1987-1998, respectively). Differences in age and bone fluoride content between these two groups were tested for significance using the Mann-Whitney U-test. The probability level determining significance was $p < 0.05$.

RESULTS AND DISCUSSION

Fluoride concentrations in the bone samples ranged between 141 and 2447 ppm F^- dry wt (table 1). The relationship among the variables age, bone fluoride concentration and year of death for the studied roe deer is illustrated in figure 1, and the data for age and bone fluoride levels in the subsamples from the two time periods are given in table 1. Whereas age did not differ significantly between the two groups, bone fluoride content in the specimens from the early period (1973-1986) significantly exceeded that of the animals from the later one (1987-1998). Our results thus demonstrate a marked reduction of fluoride exposure of the analyzed roe deer population over the study period. This is indicative of a substantial decrease in fluoride emissions from the nearby power plant and a resulting reduced atmospheric fluoride deposition into the roe deer habitat.

Table 1. Age (yr) and bone fluoride content (ppm F^- dry wt) of roe deer from the two sampling periods

Period (sample n)	Age (yr) mean ± SD	Age (yr) median	Age (yr) range	ppm F^- mean ± SD	ppm F^- median	ppm F^- range
1973-86 (13)	3.8 ± 1.3	4	1 – 7	1529 ± 536	1394	914 - 2447
1987-98 (17)	3.9 ± 1.3	3	3 – 7	452 ± 203	547	141 - 910

Significance levels of differences between periods (Mann-Whitney U-tests); age: $p = 0.53$, not significant, fluoride content: $p < 0.00001$

In Germany, reductions of fluoride discharges from industrial sources followed the implementation of emission control measures caused by the enactment of threshold values for atmospheric fluoride concentrations by the federal government in 1974 (2 µg F/m^3 for annual means, 4 µg F/m^3 for peak concentrations) and the subsequent lowering of these values to 1 and 3 µg F/m^3, respectively, in 1979. A further marked decrease of emissions (including F) from power plants was triggered by the enactment of federal regulations restricting permissible discharges from large combustion plants ("Grossfeuerungsanlagen-Verordnung") in 1983 and the subsequent lowering of these emission threshold values in 1986 (Umweltbundesamt 1997). Our data indicate a pronounced effect

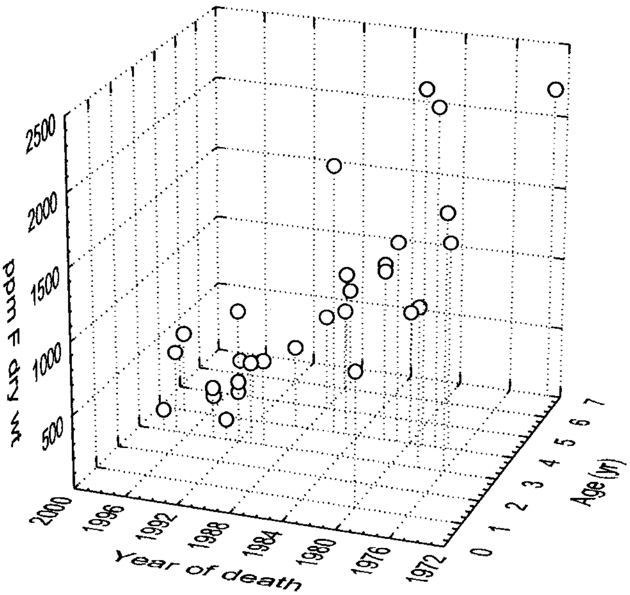

Figure 1. Relationship among the variables age, year of death and bone fluoride content in the roe deer sample (n = 30).

especially of these later measures on the level of fluoride contamination of the study area.

High bone fluoride concentrations have previously been reported for deer living near fluoride emission sources (Karstad 1967, Kay et al. 1975, Newman and Yu 1976, Newman and Murphy 1979, Suttie et al. 1987, Machoy et al. 1991, Samujlo et al. 1994, Hell et al. 1995, Kierdorf et al. 1996a,b, 1997, Vikøren et al. 1996). Most of the fluoride data in these studies are for mandibular bone. In red deer (mean age ± SD: 7.3 ± 3.0 yr) and roe deer (3.3 ± 1.9 yr) samples from habitats severely affected by fluoride emissions from brown coal-fired power plants in N-Bohemia (Czech Republic), mean mandibular bone fluoride concentrations of 2960 and 3078 ppm F^- dry wt, respectively, were recorded, whereas control samples from unpolluted regions exhibited means of 540 (red deer: 7.7 ± 4.3 yr) and 466 (roe deer: 4.8 ± 2.6 yr) ppm F^- (Kierdorf et al. 1996a). For roe deer samples (mean age ≥ 3 yr) from the vicinity of a Slovakian aluminum smelter, Hell et al. (1995) reported mean mandibular bone fluoride concentrations between 4341 and 6122 ppm F^- dry wt. In samples of red deer (individual age > 5.5 yr) living near aluminum smelters in Norway, Vikøren et al. (1996) measured mean mandibular fluoride values between 484 and 4641 ppm F^- ash wt.

Mean fluoride content in pedicle bone of yearling red deer stags from the fluoride-polluted N-Bohemian region was 1448 ppm F^- ash wt, compared to 322 ppm for a control sample from unpolluted areas in West-Germany (Kierdorf et al., 1997). In pedicle bone of roe deer from different regions in England and Scotland, Walton and Ackroyd (1988) recorded fluoride concentrations between 214 and 393 ppm F^- dry wt. A further specimen exhibited a value of 1720 ppm F^-, which was considered anomalous by Walton and Ackroyd (1988). For “skull bones” of a red deer sample (mean age ± SD: 5.6 ± 2.2 yr) from the vicinity of a chemical fertilizer plant in West-Poland, Samujlo et al. (1994) reported a mean fluoride concentration of 698 ppm F^- dry wt, whereas the corresponding values for samples (mean ages: 3.7 ± 2.0 to 5.3 ± 2.0 yr) from control areas ranged between 357 and 444 ppm F^-.

Comparison of the above data with the results of the present investigation indicates that the roe from our study area experienced a considerably increased fluoride exposure in the period 1973-1986 and that in the following period (1987-1998) the fluoride burden on the deer population was markedly reduced to levels only slightly exceeding those from European control regions. The present study underscores the value of a retrospective monitoring of bone fluoride concentrations in deer for assessing temporal variation of environmental contamination by fluorides. This is of special importance, since in Germany fluoride depositions are not continuously recorded by the federal or state environmental protection agencies. Due to its widespread distribution, its abundance (in Germany more than 1 million individuals are killed each year) and its narrow homerange (of normally less than 100 hectares in forest areas), the roe deer is a species particularly suited for use in biomonitoring studies.

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REFERENCES

Hell P, Stanovsky M, Zilinec M (1995) Dentalfluorose des Rehwildes in der Region einer slowakischen Aluminiumfabrik. Z Jagdwiss 41:117-125

Karstad L (1967) Fluorosis in deer *(Odocoileus virginianus).* Bull Wild1 Dis Assoc 3 :42-46

Kay CE, Tourangeau C, Gordon CC (1975) Industrial fluorosis in wild mule and whitetail deer from western Montana. Fluoride 8: 182-191

Kierdorf H, Kierdorf U, Sedlacek F (1996a) Biomonitoring der Fluoridbelastung des Lebensraumes von Wildwiederkäuern aus Immissionsgebieten Nord-böhmens (Tschechische Republik). Z Jagdwiss 42:41-52

Kierdorf H, Kierdorf U, Sedlacek F, Erdelen M (1996b) Mandibular bone fluoride levels and occurrence of fluoride induced dental lesions in populations of wild red deer *(Cervus elaphus)* from Central Europe. Environ Pollut 93:75-81

Kierdorf U (1988) Untersuchungen zum Nachweis immissionsbedingter chronischer Fluoridintoxikation beim Reh *(Capreolus capreolus)*. Z Jagdwiss 34:192-204

Kierdorf U, Kierdorf H, Erdelen M, Korsch JP (1989) Mandibular fluoride concentration and its relation to age in roe deer *(Capreolus capreolus)*. Comp Biochem Physiol 94A:783-785

Kierdorf U, Kierdorf H, Erdelen M, Machoy Z (1995) Mandibular bone fluoride accumulation in wild red deer *(Cervus elaphus* L.) of known age. Comp Biochem Physiol 110A:299-302

Kierdorf U, Richards A, Sedlacek F, Kierdorf H (1997) Fluoride content and mineralization of red deer *(Cervus elaphus)* antlers and pedicles from fluoride polluted and uncontaminated regions. Arch Environ Contam Toxicol 32:222-227.

Machoy Z, Dabkowska E, Nowicka W (1991) Increased fluoride content in mandibular bones of deer living in industrialised regions of Poland. Environ Geochem Health 13:161-163

Machoy Z, Dabkowska E, Samujlo D, Ogonski T, Raczynski J, Gebczynska Z (1995) Relationship between fluoride content in bones and the age in European elk *(Alces alces* L.). Comp Biochem Physiol 111C:117-120

Newman JR, Murphy JJ (1979) Effects of industrial fluoride on black-tailed deer (preliminary report). Fluoride 12:129-135

Newman JR, Yu MH (1976) Fluorosis in black-tailed deer. J Wild1 Dis 12:39-41

Samujlo D, Machoy-Mokrzynska A, Dabkowska E, Nowicka W, Paterkowski W (1994) Fluoride accumulation in European deer antlers. Environ Sci 2:189-194

Suttie JS, Dickie R, Clay AB, Nielsen P, Mahan WE, Baumann DP, Hamilton RJ (1987) Effects of fluoride emissions from a modem primary aluminum smelter on a local population of white-tailed deer *(Odocoileus virginianus)*. J Wild1 Dis 23:135-143

Umweltbundesamt (1997) Daten zur Umwelt. E Schmidt Verlag, Berlin

Vikøren T, Stuve G, Frøslie A (1996) Fluoride exposure in cervids inhabiting areas adjacent to aluminum smelters in Norway. I. Residue levels. J Wild1 Dis 32:169-180.

Walton KC, Ackroyd S (1988) Fluoride in mandibles and antlers of roe and red deer from different areas of England and Scotland. Environ Pollut 54:17-27